Physics – Uspekhi **40** (6) 599 – 622 (1997) ©1997 Uspekhi Fizicheskikh Nauk, Russian Academy of Sciences

REVIEWS OF TOPICAL PROBLEMS

PACS numbers: **72.70.+m**, 73.50.Td

$1/f$ noise and nonlinear effects in thin metal films

G P Zhigal'skiĭ

Contents

1.	**Introduction**	**599**
2.	**Properties of $1/f$ fluctuations in conducting materials**	**600**
	2.1 Hooge's empirical formula; 2.2 Selected experimental data on $1/f$ noise; 2.3 Models and hypotheses of physical mechanisms of $1/f$ noise	
3.	**$1/f$ noise in metal films induced by structural microdefects**	**605**
	3.1 Early ideas, prerequisites, and models; 3.2 Vacancy model of $1/f$ noise; 3.3 Experimental results in support of the vacancy mechanism of $1/f$-like noise in metal films	
4.	**Nonlinear effects in thin metal films and their relationship with flicker fluctuations**	**616**
	4.1 General remarks; 4.2 Mechanisms of cubic nonlinearity in metal films; 4.3 Experimental studies of CVC cubic nonlinearity in metal films; 4.4 Some data on CVC quadratic nonlinearity in metal films	
5.	**$1/f^2$ noise**	**618**
6.	**Conclusions**	**619**
	References	**619**

Abstract. **Work on $1/f$ noise and nonlinear effects in thin metal films is reviewed. The experimental dependences of the $1/f$-noise level and the *I-V* cubic nonlinearity coefficient of films on their thickness, temperature, and internal mechanical stresses are presented. The data on the effect of film microstructure on the $1/f$-noise level are also given. The $1/f$-noise spectral density and the *I-V* nonlinearity coefficient both show an activation temperature dependence and an exponential internal-mechanical-stress dependence, for metal films with elevated mobile-defect concentrations. A physical model of the $1/f$ noise and *I-V* nonlinearity is analyzed which involves the creation and annihilation of quasi-equilibrium vacancies in the bulk of the metal film and enables the observed relationship between the experimental data and the $1/f$ noise and the *I-V* nonlinearity to be explained.**

1. Introduction

Characterization of noise with a $1/f$-like spectrum, also referred to as an excess or flicker (sometimes, current) noise, provides one of the most important problems in modern radiophysics. The reason is that, on the one hand, the nature of these fluctuations remains poorly known although their possible origin has been discussed in scientific literature for many decades. On the other hand, this noise limits the sensitivity and stability of many radioelectronic devices, the requirements to which are enhancing constantly.

G P Zhigal'skiĭ Moscow Institute of Electronic Engineering (Technical University), 103498 Moscow, Russia
Tel. (7-095) 532 99 24
Fax (7-095) 530 22 33
E-mail: zh@orgland.ru

Received 19 July 1996, revised 28 November 1996
Uspekhi Fizicheskikh Nauk **167** (6) 623 – 648 (1997)
Translated by Yu V Morozov; edited by A Radzig

The above names are also applied to noise with $1/f^{\gamma}$ spectra, where the frequency exponent $\gamma > 0$ specifies the shape of the spectrum. The term '$1/f$ noise' usually implies that γ is close to unity. Flicker noise with $0.8 \leqslant \gamma \leqslant 1.2$ is the most common observed.

Recently, there has been sharply increasing interest in $1/f$ noise in thin metal films which can be accounted for by their wide application in different areas of physics and technology, especially in modern microelectronics which makes high demands of thin films of different materials in manufacturing commutation layers, resistors, and contacts for integrated microcircuits (IMC).

The first studies of flicker noise made by Johnson and Shottky date back to 1925 – 1926 [1, 2]. Since then, flicker fluctuations in solids have been investigated by many theorists and experimenters. Current concepts of flicker noise and related issues have been discussed and summarized in a large number of monographs and reviews [3 – 14].

Flicker fluctuations reflect many processes at the electron and atom levels and specific features of solid-state microstructures which makes $1/f$ noise a valuable informative parameter for evaluating the quality of materials and reliability of devices containing semiconductors and IMC [15 – 19]. Also, $1/f$ noise is used to predict the electromigration immunity of thin-film metallization in IMC [20, 21].

Recent studies have yielded a considerable amount of experimental and theoretical data on $1/f$ noise in conducting materials. The objective of the present paper is to review the results of $1/f$-noise studies in continuous metal films beginning from the first work by Hooge in 1969 [22]. $1/f$-noise models and related experimental findings discussed comprehensively in earlier reviews [10 – 14] are not included here. Nor do we examine the Handel quantum-mechanical theory of $1/f$ noise [23 – 26] and its encountered difficulties [27, 28] because this theory does not explain $1/f$ noise in metals despite attempts of certain authors to treat it as a fundamental theory of $1/f$-noise mechanisms in solids [7]. For the same

reason [12, 29], we do not consider here McWhorter's trap model [30, 10].

2. Properties of $1/f$ fluctuations in conducting materials

2.1 Hooge's empirical formula

In 1969, Hooge and Hoppenbrouwers reported an observation of $1/f$ noise in continuous gold films [22]. The noise magnitude was well-described by the empirical formula which Hooge had previously derived for semiconductors [31]. It was suggested to describe the voltage U or resistance R fluctuations over a sample (at a given current I) by the expression

$$\frac{S_U(f)}{U^2}=\frac{S_R(f)}{R^2}=\frac{\alpha_{\mathrm{H}}}{N_{\mathrm{c}}f}=\frac{\alpha_{\mathrm{H}}}{n_{\mathrm{c}}Vf}\,. \tag{2.1}$$

Here, $S_U(f)$ and $S_R(f)$ are the spectral densities (SD) of the voltage and resistance fluctuations over the sample, respectively, N_{c} is the number of carriers in the sample ($N_{\mathrm{c}} = Vn_{\mathrm{c}}$, where V is the sample volume and n_{c} is the electron concentration), and f is the frequency. The dimensionless coefficient $\alpha_{\mathrm{H}} = 2\times10^{-3}$ is referred to as the Hooge constant and expression (2.1) is called the Hooge formula. However, further studies have demonstrated that the coefficient α_{H} is not constant for different materials [10] and should rather be termed a parameter (Hooge himself was of the same opinion, see Ref. [32]).

Also, Hooge suggested [31] that $1/f$ fluctuations in all homogeneous materials are defined by formula (2.1). This expression is correct if the experimentally examined SD of the voltage fluctuations over the sample is directly proportional to the square of the applied voltage or current and inversely proportional to the frequency. Fulfillment of the $S_U \propto U^2$ law makes it possible to pass to the relative fluctuation spectrum S_U/U^2 and interpret the $1/f$ noise as a result of equilibrium resistance fluctuations [8, 10, 11, 31]. When the law of quadratic U-dependence of S_U is not satisfied exactly, then expression [12]

$$S_U(f)=\frac{\alpha U^{2+\theta}}{N_{\mathrm{c}}f^{\gamma}} \tag{2.2}$$

is also called the Hooge formula. Here, parameter α is a dimensional quantity. The frequency exponent γ may differ from $\gamma = 1.0$ while the exponent θ defines the nonlinearity of the current-voltage characteristic (CVC).

According to different authors, the parameter α for various metals is of the order of $10^{-4}-10^{-2}$ [10, 22, 33–36]. The frequency dependence of $1/f$-noise power spectral density (PSD) in metal films in the form (2.2) can be seen in the frequency range 10^{-4} to 10^4 Hz at $0.8 \leqslant \gamma \leqslant 1.2$ [22, 33, 34, 36, 37].

In the case of quadratic dependence of the noise PSD on the applied voltage and γ values close to unity, the noise level is not infrequently estimated from the dimensionless parameter α:

$$\alpha=\frac{S_U(f)n_{\mathrm{c}}Vf}{U^2}\,. \tag{2.3}$$

It is universally accepted that $1/f$ noise can occur in semiconductors and metals due to fluctuations in the number or mobility of carriers, their resistance being dependent on both the concentration and mobility of charge carriers. However, debates on whether fluctuations of mobility or the number of carriers serve as the primary source of $1/f$ noise in metals have ended in favour of the former mechanism [6, 12, 38, 39]. Hooge, Kleinpenning, and Vandamme [11, 32, 38] demonstrated that $1/f$ noise in resistive silicon layers was induced by fluctuations of charge carrier mobility. $1/f$ noise was shown to abate in strongly alloyed semiconductors. The authors attributed this effect to the fact that only a part of the mobility which is due to the charge-carrier scattering from phonons undergoes fluctuations, whereas the remaining part associated with scattering by impurities is not subject to fluctuations. It is worthwhile to note that atoms of an alloying admixture in a crystal may be regarded as stable defects. Results of special experiments [11, 40, 41] also support the mobility fluctuation hypothesis.

Another argument in favour of this mechanism (charge-carrier mobility fluctuations) of $1/f$ noise in metals is the tensor nature of conduction fluctuations [10] shown to occur in carbon, gold, and chromium films [29, 42]. In addition, marked anisotropy of conduction fluctuations was recorded in bismuth films [43]. It has been shown [29, 44] that anisotropic conduction fluctuations may be due to mobility fluctuations, for example, in the case of scattering from static or diffusive microdefects of a structure. At the same time, scalar conduction fluctuations were reported to occur in silicon films [44], in agreement with the model of fluctuations in the number of carriers captured by a trap.

The hypothesis assuming $1/f$ noise to be a sequel of scattering by phonons, gave rise to a modified expression for the Hooge parameter α in the form [11, 32, 45]

$$\alpha=\left(\frac{\mu}{\mu_{\mathrm{L}}}\right)^2\alpha_{\mathrm{H}}\,, \tag{2.4}$$

where μ is the effective mobility of current carriers, and μ_{L} is the lattice mobility. The validity of this expression was verified by measuring α in semiconductors, with the carrier mobility being modulated by varying the temperature and alloying admixture concentration [46, 47].

Moreover, the hypothesis of $1/f$-noise dependence on processes involving phonons is supported by experiments in which the noise abates with the decreasing relative contribution of scattering by phonons to the overall resistance of p- and n-type silicon samples. Parameter α was also shown to decrease with the increasing contribution of carrier scattering from the boundaries of a bismuth film surface (as its thickness reduced) [48]. However, all these data provide no direct evidence of $1/f$ noise being due to lattice scattering; nor do they explain its physical mechanism (see Refs [49, 50] where the parameter α has been computed).

An enhanced noise level ($\alpha \sim 1$) was found to occur in whisker crystals of copper, tin, zinc, and bismuth [51, 52]. According to Voss and Clarke [33], the measured α for bismuth films was approximately 10^3 times lower than $\alpha_{\mathrm{H}} = 2\times10^{-3}$; for manganin, $\alpha < 10^{-4}$ [53, 54]. Platinum [55] and niobium [56] films were also characterized by noise of very low magnitude. The major reason for this discrepancy is probably the difference between the estimated numbers of carriers N_{c} in the sample. The effective concentration of electrons contributing to the conduction in metals is known to be tens or hundreds of times lower than their total concentration in the conduction zone (electrons with energies in the kT range near Fermi energy level) [32, 57, 58]. At the same time, earlier studies estimated the parameter α from

the number of valence electrons. Clarke and Voss [54] noticed that $1/f$-noise PSD in bismuth films was in better agreement with experimental findings if the number of atoms N_a was substituted into formula (2.1) instead of the number of carriers. However, other authors argued that this inference aroused from the erroneous estimation of the number of current carriers in the film [48, 59].

Hooge [32, 60] provided theoretical and experimental considerations in support of the presence of the factor $1/N_c$ in expression (2.1), instead of $1/N_a$. Hooge's opinion was confirmed by the semiconductor experiments reported by Kleinpenning [61]. At the same time, no such confirmation was obtained for metals, and the dependence of the $1/f$-noise magnitude in metals on the number of current carriers, rather than on other factors, awaits a more convincing demonstration.

Fleetwood and Giordano [36] found the dependence of α on the specific resistance of metal films.

Later studies provided experimental evidence of the validity of expression (2.1) for a wide variety of materials (excepting metal films and homogeneous semiconductors), namely, diodes with pn-transitions and Shottky barriers, field-effect transistors [6, 11, 38, 39], point contacts [62, 63], etc.

The above data indicate that (2.1) is not a universal formula for the description of $1/f$ noise in metals and semiconductors. Nevertheless, it is the only known relation which allows estimation (even roughly) of the magnitude of $1/f$ noise in homogeneous conductors, with each measurement accurate to within one – three orders of magnitude.

2.2 Selected experimental data on $1/f$ noise

2.2.1 The problem of low-frequency boundary in the spectrum is a most important one in the studies on the nature of noise with $1/f$-type spectra. The $1/f$ law was many times confirmed for various electronic devices down to very low frequencies ($f_l \simeq 5 \times 10^{-7} - 10^{-4}$ Hz) [12, 64 – 67]. The high-frequency boundary of the spectrum is as a rule masked by both the thermal noise of the sample and the background noise of the measuring unit. In strong-noise systems, e.g., carbon resistors $1/f$ noise prevails over thermal noise up to frequencies of $f_h \sim 10^6$ Hz [68].

The greatest difficulty encountered in the explanation of $1/f$ noise is related to the elucidation of the physical mechanisms of its generation at the lowest frequencies since they must be mechanisms with large correlation times ($\tau_{corr} \sim 1/f_l$).

2.2.2 The problem of stationary fluctuations with $1/f$-like spectra is the key one for the construction of the $1/f$-noise theory and understanding its nature. The nonstationary $1/f$-noise hypothesis was first suggested by Malakhov [69] and has since been extensively discussed in the literature [64, 70 – 72].

Certain authors attempted to address the problem by analyzing statistical noise characteristics. Specifically, Brophy [70, 73, 74] and some others [75, 76] performed statistical experiments and came to the conclusion that $1/f$ noise has nonstationary nature. However, it has been shown in Refs [77, 78] that this inference resulted from the erroneous interpretation of the results, which can be explained without the nonstationary hypothesis.

Potemkin and Stepanov [79, 80] investigated the statistical properties of $1/f$ noise in semiconductor devices and concluded that the assumption of the nonstationary nature of this noise in the frequency range being examined (25 Hz – 10 kHz) is unnecessary. Taken together, the results of many experimental studies do not appear to support the nonstationarity hypothesis of $1/f$ noise (see reviews [10, 12]). The stationary hypothesis was checked up by Tandon and Bilger [64]. They found that the noise intensity for a resistor and a semiconductor stabilitron persisted for 2.5 and 4.5 years, respectively (within experimental uncertainty).

A series of experiments have demonstrated the nonstationary behaviour of macroparameters. For example, Tandon and Bilger [64] observed the resistance drift in their 150-nm bismuth films at a rate of about 0.001% per hour. Noise was measured in these samples with a frequency spectrum of $1/f^{\gamma}$ ($1.3 \leqslant \gamma \leqslant 1.5$). The authors therefore attributed the noise to a nonstationary statistical process manifested in the resistance drift. Dutta, Dimon, and Horn [81] found that changes of resistance in bismuth films depend on the time elapsed after their preparation. Their magnitude was higher in as-manufactured films, whereas no drift was apparent two weeks later. Bismuth films heated up to 150 °C and then abruptly cooled to room temperature also exhibited no resistance drift, and the noise magnitude remained virtually unaltered.

It should be borne in mind that the strict concept of a stationary fluctuation process cannot be applied to real physical systems unless the time of the relaxation process τ_{rel} and observation time τ_{obs} are specified. The parameters of real physical objects remain unaltered only during a certain finite period, and their fluctuation patterns may change with time. The fluctuation process can be regarded as quasi-stationary provided the condition $\tau_{obs} \ll \tau_{rel}$ is satisfied [82, 83]. Real physical systems are lacking an ideal intrinsic equilibrium, rather they tend to be thermodynamically equilibrated, age or remain in a quasi-equilibrium state. The process of the system's transition to a thermodynamic equilibrium is a random nonstationary process known to induce $1/f$ noise.

At present, many investigators of $1/f$ noise agree that in the majority of cases, flicker fluctuations are thermodynamically equilibrium in nature.

2.2.3 An important characteristic of flicker noise is its amplitude distribution which may be determined by a random Gaussian process indicating that the noise originates from a large number of random sources (fluctuators). The Gaussian nature of $1/f$ noise was observed in a variety of samples [70, 77]. Its small deviation from the normal distribution was also reported in Refs [84, 85] and attributed to the influence of burst noise [84].

The probability characteristics of actual $1/f$-noise intensity in graphite and chromium thin-film microresistors were measured in Refs [86, 87]. The authors obtained markedly asymmetric histograms with a long right-hand 'tail' suggesting a strong non-Gaussian nature of the $1/f$ noise being examined, which was especially noticeable in specimens of ultimately small volume but depressed as the size of the sample increased. For example, niobium samples 10^{-11} cm^3 in size as well as silver, bismuth and AgPb alloy samples of 10^{-14} cm^3 exhibited all signs of non-Gaussian statistics suggesting a small amount of fluctuators in these systems [88].

2.2.4 Many authors relate $1/f$ fluctuations in homogeneous materials (resistive layers, semiconductors, metal films, etc.) to conduction fluctuations which are known to occur even in

the absence of a current in the sample. Assuming that fluctuations of voltage $U(t)$ across a sample at a given direct current I_0 arise due to fluctuations of the current-independent resistance $R(t)$, the SD of the voltage fluctuations may be characterized by the dependence quadratic in current [8]:

$$S_U(f) = I_0^2 S_R(f)\,, \tag{2.5}$$

where $S_R(f)$ is the spectral density of resistance fluctuations (SDF).

The law for the quadratic-in-current dependence of noise PSD is not consistently fulfilled in a strict sense because the sample's resistance in the general case is a nonlinear function of current I. The resistance may be represented as a power series with coefficients fluctuating in time t [89, 90]:

$$R(I,t) = \sum_{n=0}^{\infty} R_n(t) I^n(t)\,. \tag{2.6}$$

The current-independent zero term $R_0(t)$ defines the linear part of the CVC for the sample, i.e. Ohm's law. This term reflects the equilibrium fluctuations of film conduction which exist in the absence of current flowing through the sample. Coefficients of terms higher in order than the zero one contribute to the resistance (hence, to its fluctuations as well) only if the current $I > 0$. Therefore, the fluctuations of these coefficients may be related to nonequilibrium (nonlinear) conduction fluctuations which could be responsible for both stationary and nonstationary noise.

Nonequilibrium fluctuations cause deviation from the quadratic dependence in (2.5). They may be due either to different mechanisms of transfer and scattering of current carriers [89, 90] or local overheating of the film by the Joule heat [91]. In case of small currents, only equilibrium $1/f$ noise is apparent.

The so-called $1/\Delta f$ noise [95] is induced by fluctuations of the coefficient $R_0(t)$, whereas fluctuations of the coefficients R_1 and R_2 (the quadratic and cubic terms of the CVC) lead to modulation of the second and third harmonics of the signal-response by $1/f$ noise, which arises as a result of the harmonic influence on the sample [89, 90].

That $1/f$ noise frequently results from fluctuations of the linear part of resistor resistance was many times confirmed in experiments on nonmetal samples. Specifically, $1/\Delta f$ noise was reported to occur when an alternating current with a frequency f_1 flowed along a resistor [95–99], as a result of amplitude modulation of voltage across a sample. If $1/f$ noise is induced therewith by fluctuations of the linear portion of the sample's resistance, then a noise spectrum arises within two side bands placed peripherally to f_1 frequency, which varies in accordance with the same $1/f$ law as at the zero frequency in case of a direct current through the resistor. Jones and Francis [97] let direct and alternating currents simultaneously pass through a resistor [97]. The resulting $1/f$ and $1/\Delta f$ noises were perfectly correlated suggesting an equilibrium nature of the resistance fluctuations.

Voss and Clarke obtained direct experimental evidence of equilibrium resistance fluctuations by measuring PSD fluctuations of thermal noise with a $1/f$ spectrum in InSb films and island niobium films in the absence of current flowing through the samples. These fluctuations were also observed when a direct, sinusoidal or pulse current passed through a sample [33, 53]. All measuring techniques revealed identical $1/f$ spectra. Similar results were reported by Beck and Spruit using carbon films [100]. These findings indicate that neither direct nor alternating current was the cause of $1/f$ noise, which was actually induced by equilibrium resistance fluctuations.

At the same time, in continuous molybdenum films deposited upon oxidized silicon substrates by magnetron sputtering at a relatively low condensation rate ($w_{\text{cond}} = 1$ nm s^{-1}), fluctuations of the quadratic term of the CVC made a considerable contribution to the PSD of $1/f$ noise (up to 60% at 10 Hz) at a rather low density of direct current ($j < 5 \times 10^4$ A cm^{-2}) [89, 90]. Such films had an elevated content of reactive-gas contaminants, and the induction of nonequilibrium $1/f$ noise was apparently due to nonmetallic conduction mechanisms.

2.2.5 The differential effect of substrates on the level of $1/f$ noise in various deposited films was observed by many authors [12, 35, 36, 101]. The $1/f$-noise level in tin and indium films lowered with the increasing heat conduction of the substrate (glass or sapphire) and improving its thermal coupling with the film. The substrate was shown to have the most pronounced effect on $1/f$ noise near the metal transition into the superconducting state [102, 103]. The temperature resistance coefficient being abnormally high in this case, it is supposed that flicker noise is induced by thermal fluctuations [33, 53, 104].

At the same time, certain authors failed to observe the effect of the substrate on the $1/f$-noise magnitude. In fact, it did not depend on the type of substrate (glass, fused quartz, sapphire) underlying silver, copper, gold, lead, and platinum films [34, 36, 101]. Also, the noise level remained unaltered in silver, gold, copper, and nickel films placed in the air, helium or a vacuum and after coating the sample with a layer of silicon monoxide [34].

Cr and Al films deposited under identical conditions onto substrates having significantly different coefficients of thermal conduction, had identical noise levels as well [105, 106] (Cr films on devitrified glass and oxidized silicon [105] or Al films on glass and oxidized silicon [106]). These results indicate that the mechanism of noise induction in these films is associated with bulk processes and excludes the effect of heat exchange between the film and the substrate or the environment [12, 101].

However, it should be borne in mind that the experimentally examined effects of the substrate on $1/f$ noise may just as well be related to its action upon the film microstructure [105] and temperature stress level in the film [107].

2.3 Models and hypotheses of physical mechanisms of $1/f$ noise

Various models have been suggested to account for $1/f$ noise in solids, each designed to be used in a specific case. There are two principal approaches to explaining $1/f$ noise in conducting materials. One considers equilibrium models generating stationary noise, while the other is concerned with degradation models which describe nonstationary processes (i.e. processes resulting in thermodynamic equilibrium or aging). The former approach assumes that the $1/f^\gamma$ spectrum occurs in a limited frequency range between f_l and f_h. The objective is to provide a theoretical substantiation of the frequency range boundaries and temperature dependence of $1/f$ noise.

2.3.1 The model of 'exponentially wide relaxation-time distribution' [6, 10, 12] appeared to be especially popular. It considers $1/f$ noise as a superposition of random relaxation

processes characterized by a relaxation time τ distribution, which remains continuous throughout a certain interval between τ_l and τ_h ($\tau_l \gg \tau_h$) and is described by the distribution function $g(\tau)$.

In the simplest case, when the fluctuation kinetics of a random quantity $x(t)$ is characterized by single relaxation time τ, the fluctuation SD has the form of the Lorentz function [5, 8]:

$$S_x(f) \propto \frac{\tau}{1+\omega^2\tau^2} . \tag{2.7}$$

If the fluctuations of $z(t)$ are defined by a set of relaxation times τ with the continuous distribution function $g(\tau)$, the fluctuation SD of $z(t)$ is determined by integrating (2.7) with the statistical weight $g(\tau)$ [4, 5, 8]:

$$S_z(f) = \int_0^\infty g(\tau) S_x(f)\,d\tau . \tag{2.8}$$

Specifically, if the weight function distribution obeys the statistical law

$$g(\tau) \propto \frac{1}{\tau} \qquad (\tau_h \leqslant \tau \leqslant \tau_l) , \tag{2.9}$$

then (2.8) gives rise to the $1/f$ spectrum in the frequency range $f_l \ll f \ll f_h$, where one has

$$f_{l,h} = \frac{1}{2\pi\tau_{l,h}} . \tag{2.10}$$

The desired distribution (2.9) is obtained if the $1/f$ noise is due to activation processes. It has been shown [108] that the mean duration of the atomic stay in a potential well of depth E_a is $\tau = \tau_0 \exp(E_a/kT)$, where τ_0 is the atomic thermal vibration period ($\tau_0 \simeq 10^{-13}$ s), k is the Boltzmann constant, and T is the absolute temperature.

When the activation energy E_a is uniformly distributed over a sufficiently broad interval, i.e. $G(E_a) = \text{const}$ ($E_1 \leqslant E_a \leqslant E_2$), the following expression holds true for $1/f$-spectrum boundaries:

$$f_{l,h} = f_0 \exp\left(-\frac{E_{2,1}}{kT}\right) . \tag{2.11}$$

Here, $f_0 = \tau_0^{-1}$ is the average frequency of atomic thermal vibrations.

The $1/f^\gamma$ spectrum occurs if the weight function is distributed in accordance with the law $g(\tau) \propto \tau^{\gamma-2}$ [6].

For the $1/f$ law to be strictly fulfilled, the energy distribution function must be continuous even though the $1/f$ spectrum can be simulated with sufficient accuracy by a discrete set of processes (e.g., when there is only one relaxation process per frequency decade) [109, 110]. In the case of the activation energy distribution from E_1 to E_2, the number of frequency decades with a $1/f$ spectrum is derived from the formula [81]

$$\frac{f_h}{f_l} \sim \exp\left(\frac{E_2 - E_1}{kT}\right) . \tag{2.12}$$

The model of an exponentially wide activation-energy distribution was first employed by van der Ziel [111] and Du Pre [112] to explain the existence of $1/f$ noise in semiconductors by a random set of atomic ionization energies and was subsequently applied to the analysis of $1/f$ noise in various types of disordered systems.

Based on the results obtained for metal films in Ref. [34], Dutta, Dimon, and Horn [81] demonstrated that the $G(E_a)$ distribution is nonuniform which accounts for the observed deviation of the frequency exponent γ from unity.

2.3.2 The most universal model for the description of $1/f$ noise is the model of two-level systems (TLS) in which the heights of local energy barriers are random quantities [12, 13]. Analysis of an isolated TLS with two local minima separated by a relatively low potential barrier leads to the $1/f$-noise model suggested by Kogan and Nagaev for the case of particle tunnel transition [10, 113, 114], and Dutta and Horn, for particle activation transition [12]. This model was also applied by Yakimov [115] and Kozub [116] to metal films and tunnel contacts. The spectrum of the process has the $1/f$ form if the heights of local barriers ΔE are uniformly distributed over the range from ΔE_1 to ΔE_2 [115]. The upper and lower boundaries in the spectrum are defined by the relation

$$f_{l,h} = f_0 \exp\left(-\frac{\Delta E_{2,1}}{kT}\right), \qquad \Delta E_1 \leqslant \Delta E \leqslant \Delta E_2 , \tag{2.13}$$

where ΔE_1 and ΔE_2 are the characteristic values of the activation energy, which specify the uniform distribution limits.

The $1/f$-like spectrum can also be obtained for the distribution of activation-energy excess ΔE of the Boltzmann type [115, 117], but the physical mechanisms responsible for such a distribution remain to be elucidated.

2.3.3 The following mathematical models of $1/f$ noise are worth mentioning. The $1/f^\gamma$ spectrum is sometimes obtained by representing a random process as a Poisson pulse sequence. With this approach, pulses are given in different ways: as suggested by Schonfeld [4, 6, 69] or Halford [118]. In the former case, the spectrum at low frequencies is defined by the slowly decaying pulse 'tail' ($\propto t^{-1/2}$). In the latter, the boundaries of the frequency range are given by the pulse length distribution.

Mathematically, the $1/f$ spectrum may be obtained from white noise with the help of the fractional order integration [4, 8]. Specifically, if a white noise source is connected to an infinite RC-chain, then the voltage across the chain will have the $1/f$ spectrum [109, 119]. In the case of a finite RC-chain, there appears a low-frequency boundary of $1/f$ noise [120].

2.3.4 The most widely known hypothesis applicable to metal films is that suggested by Voss and Clarke, which relates $1/f$ noise to equilibrium temperature fluctuations [33, 53, 54]. The temperature of any body in thermal equilibrium with the environment playing the role of a thermostat at an average temperature T_0 undergoes fluctuations caused by heat exchange between the body and the medium. The mean square of these fluctuations takes the form [121]

$$\overline{\delta T^2} = \int_0^\infty S_T(f)\,df = \frac{kT_0^2}{C} , \tag{2.14}$$

where C is the heat capacity, and $S_T(f)$ is the SD of temperature fluctuations.

According to the Voss and Clarke model, the temperature fluctuations lead to sample resistance (R) fluctuations around

an average value and to fluctuations of voltage U across the sample at a given current I ($U = RI$). The relative SD of voltage $S_U(f)/U^2$ or resistance $S_R(f)/R^2$ fluctuations are defined as

$$\frac{S_U(f)}{U^2} = \frac{S_R(f)}{R^2} = \beta^2 S_T(f)\,, \tag{2.15}$$

where β is the temperature coefficient of resistance (TCR) of the film.

Voss and Clarke [33] measured noise in continuous films of gold, silver, copper, tin, bismuth, and a manganin alloy at room temperature. Noise with the $1/f$ spectrum was found to occur in all specimens except manganin. The latter material having a low TCR ($\beta < 10^{-4}\ \mathrm{K}^{-1}$), the authors considered the result to indicate that the noise was induced by temperature fluctuations.

In Ref. [33], the SD of temperature fluctuations $S_T(f)$ was calculated from the solution of the heat conduction equation for temperature fluctuations $\delta T(t)$ with Langevin sources. This yielded the power dependence for $S_T(f)$ characteristic of the diffusion process with $\gamma = 0.5$ and $\gamma = 1.5$ but containing no $1/f$-like spectral region ($\gamma = 1$). In order to explain $1/f$ noise, the authors introduced an empirical $1/f$ region into the $S_T(f)$ spectrum; in this region, $S_T(f) \propto 1/f$ in the frequency range f_1 to f_2 defined by inverse times of heat propagation along the film length L and width b, respectively. Using the normalization condition (2.14), Voss and Clarke [33] obtained

$$S_T(f) = \frac{kT^2}{cV\left[3 + \ln(f_2/f_1)\right]f} \qquad (f_1 \leqslant f \leqslant f_2)\,, \tag{2.16}$$

where c is the specific heat capacity. The characteristic frequencies f_1 and f_2 are defined through the thermal conductivity coefficient D_T of metal:

$$f_1 = \frac{D_T}{\pi L^2}\,, \qquad f_2 = \frac{D_T}{\pi b^2}\,. \tag{2.17}$$

In fact, the $1/f$-like spectrum was postulated. The $1/f$-noise level measured in Ref. [33] turned out to be very close to the PSD value (2.16). This finding was taken as an argument in favour of the hypothesis that $1/f$ noise is caused by equilibrium temperature fluctuations.

Van der Ziel [6] demonstrated that expression (2.16) is formally equivalent to the Hooge formula (2.1) and yields the desired order of magnitude of the parameter α because, in accordance with the Dulong and Petit law, the heat capacity $C = 3N_a k$, where N_a is the number of atoms in the sample which defines the number of current carriers N_c in (2.1). It should always be borne in mind that such comparisons are somewhat arbitrary because there is a risk of incorrect determination of the number of effective carriers in a sample (see Section 2.1).

That temperature fluctuations may serve as a source of $1/f$ noise in metal films has been confirmed by experiments showing the spatial correlation of fluctuations between different parts of bismuth [33] and chromium [122] films at $f \sim 1$ Hz. However, a number of studies [124, 125] failed to reveal a spatial correlation even in case of a good thermal contact between the films [124].

The frequency (f)-dependent correlation length $L_{\mathrm{corr}} \sim 1$ mm in chromium films [122] is rather close to the theoretical one [33, 123]

$$L_{\mathrm{corr}} = \left(\frac{D_T}{\pi f}\right)^{1/2}. \tag{2.18}$$

Ref. [122] reports an increase in the coefficient of spatial correlation (CSC) r for $1/f$ noise with growing specific power dissipated by the film. The analysis of dependences found in Ref. [122] indicates that CSC is extrapolated to $r = 0$ at $P_{\mathrm{sp}} \simeq 0.02\ \mathrm{W\,cm^{-2}}$ (in this case, the film is overheated, with $\Delta T < 2$ K). This result is in agreement with the theoretical value predicted by Yakimov [126], who showed that spatial correlation of fluctuations in a film originated from intrinsic (primary) delta-correlated-in-space fluctuations of film resistance (their true nature remains to be elucidated) rather than from equilibrium temperature fluctuations. Resistance fluctuations may be related, for instance, to fluctuations in the number of vacancies in the sample (see Section 3), which are responsible for temperature fluctuations with finite correlation radius. The effect is heightened by virtue of Joule heating in consequence of thermal feedback. In the case of weak overheating of the film, there must be no spatial correlation of $1/f$ noise. This observation accounts for the absence of correlation in the experiments [124], where overheating of the samples did not exceed 1 K, when the correlation was measured. Similarly, the correlation was absent in case of a small dissipated power in experiments carried out in Ref. [122].

At the same time, the spatial correlation of the noise reported in Ref. [122] had a thermodiffusive nature and was due to heat exchange between different parts of the chromium film. This finding was experimentally confirmed by the absence of correlation in specimens having leads of aluminium film about 1 μm thick deposited upon a chromium film. Being a perfect heat dissipater, the aluminium film precludes the propagation of thermal waves through adjacent parts of the chromium film.

Major difficulties encountered in the thermodiffusion theory of noise [33] are related to the prediction of the $1/f$ spectrum in a relatively narrow frequency range (about 4 decades for film samples of a typical size) and the explanation of the low-frequency spectrum ($f < f_1$), since the theory implies spectrum saturation: $S_T(f) = \mathrm{const}$ or $S_T(f) \propto \ln\omega^{-1}$ at $f < f_1$. At higher frequencies ($f > f_2$), the spectral density of temperature fluctuations in a film is $S_T(f) \simeq f^{-3/2}$ [33].

Ketchen and Clarke [127] observed the noise-spectrum plateau at low frequencies in freely suspended tin films close to the superconducting transition. Refs [122, 128] investigated the dependence of the characteristic frequency f_2 defined by (2.17) on the chromium film width. Theoretical and experimental findings for f_2 turned out to be in good agreement. However, the noise intensity reported in Ref. [128] was five orders of magnitude greater than that predicted by the model of equilibrium temperature fluctuations. It has been shown in Ref. [128] that the observed noise is due to nonequilibrium temperature fluctuations although their mechanism remains to be elucidated. This inference is supported by experimental data demonstrating that spectrum inflections (like the spatial correlation in Ref. [122]) occur when the film dissipates a relatively large power (Fig. 1). This figure shows that spectrum inflections appear and the frequency exponent γ (in the region of $1/f^{\gamma}$ spectrum) increases as the current grows. Simultaneously, the current dependence of the noise

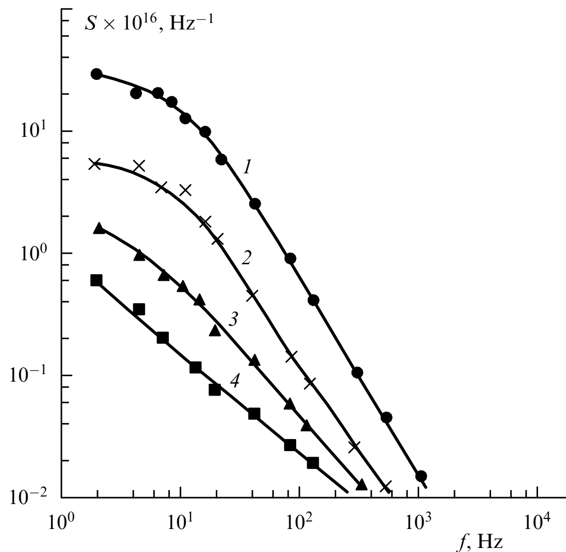


Figure 1. Frequency dependence of flicker-noise PSD in Cr films of thickness $h = 100$ nm and width $b = 0.22$ mm at $\rho = 40$ μΩ cm and different currents flowing along the sample [128]: (*1*) $I = 10$ mA; (*2*) 8 mA; (*3*) 5 mA; (*4*) 2 mA.

PSD deviates from quadratic suggesting a manifestation of nonequilibrium conduction fluctuations [89, 90] probably associated with local film overheatings and generation of excess vacancies [91].

Although the hypothesis suggesting links between $1/f$ noise and equilibrium temperature fluctuations is at variance with many experimental findings concerning $1/f$ noise in metals (specifically, the thermodiffusion theory does not explain the activation temperature dependence of $1/f$ noise [34, 129]), this mechanism appears to be related to a universal source of fluctuations in physical systems [82], and hence a large number of publications on the subject [130–133]. Some authors attempted to derive the $1/f$ spectrum of noise from a thermodiffusion process or diffusion patterns of any particles [134–139]. With certain assumptions and an appropriate choice of boundary conditions, it is possible to obtain the $1/f$-noise spectrum over a limited frequency range.

According to the $1/f$-noise model suggested by Klimontovich [135, 136], 'natural excess noise' occurs in the low-frequency range of any diffusive system with Brownian motion. The noise intensity is inversely proportional to the number of particles in the system, and the shape of the spectrum is unaffected by the body shape. The lower frequency of the spectrum depends on the observation time.

2.3.5 The diffusive rearrangement model suggested by Yakimov [72] considers $1/f$ noise to be a random nonstationary process stemming from the diffusive degradation of the sample. It can be applied to thin conducting films showing structural nonequilibrium. The spectrum of these fluctuations has the form $1/f^{\gamma}$, where $\gamma = 3/2$. However, the structure is stable in the annealed films and $1/f$ noise eludes explanation through the agency of structure rearrangement processes. Ref. [140] explains the mechanism of $1/f$ noise in metal films by physical sorption and desorption of gases from the environment. Although the absorption mechanism of $1/f$ noise may manifest itself under certain conditions [141] and yet one fails to clarify many experimental results using this approach.

2.3.6 It should be emphasized that in all steady-state models, $1/f$ noise occurs in a bounded frequency range from f_l to f_h which suggests thermodynamic limitations on the mechanism of $1/f$ noise [142]. For thermodynamic reasons, noise with the $1/f$ spectrum cannot exist in a boundless frequency range.

3. $1/f$ noise in metal films induced by structural microdefects

3.1 Early ideas, prerequisites, and models

Eberhard and Horn [34, 144] suggested that $1/f$ noise in metal films is due to vacancy diffusion, and the rise in its level with temperature stems from the increased concentration of vacancies. Robinson [145] hypothesized that $1/f$ noise in metals may originate from chaotic motion of defects 'frozen' in the lattice.

Pelz and Clarke [146] obtained experimental evidence of the relationship between the $1/f$-noise magnitude and the concentration of defects in polycrystalline copper films. The defects were induced by bombarding the films with fast 500 keV electrons maintained at $T = 90$ K. It turned out that the change in film resistivity $\Delta\rho_f$ was proportional to the total number of defects n_d. At the same time, the $1/f$-noise PSD increment expressed through the parameter α in accordance with (2.3) was defined by the relation $\Delta\alpha \propto n_d^{0.6} \propto \Delta\rho_f^{0.6}$ (Fig. 2), with n_d including the defects 'frozen' at $T = 90$ K. The annealing process reduced both the resistance and the noise.

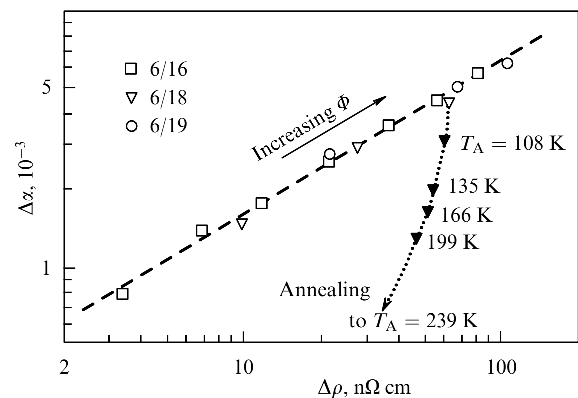
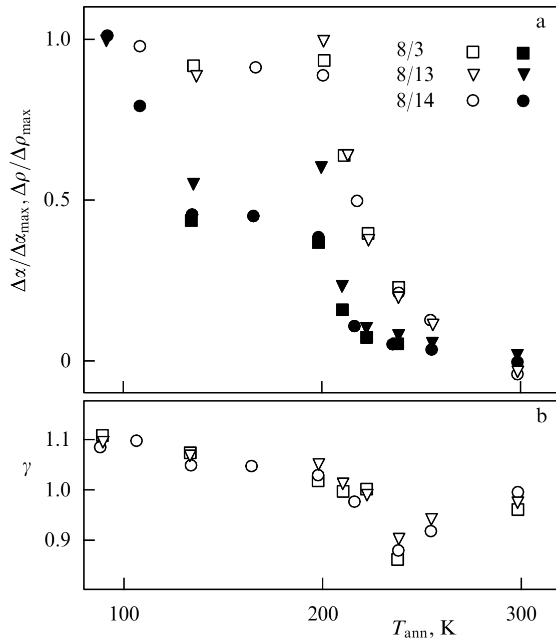


Figure 2. Plots of changes in $1/f$-noise magnitude (α parameter) vs. changes in sample resistivity in Cu films with increasing radiation dose Φ [146]. The dashed line is drawn to show the dependence $\Delta\alpha \propto \Delta\rho^{0.6}$, while solid triangles correspond to annealing at successively higher temperatures T_{ann} (up to T_A). For $T_A = 239$ K, $\Delta\rho \simeq 11.6$ nΩ cm, and $\Delta\alpha \simeq 7 \times 10^{-5}$.

The authors explained these findings by the dependence of excess flicker noise on mobile defects which make up a small fraction of the total number ($\simeq n_d^{0.6}$). This explanation was verified in annealing experiments. (Annealing is usually understood as the holding of a film in a vacuum or inert gas at the elevated temperature for a certain period. 'Annealing of defects' means the removal of defects from a crystal lattice). Mobile defects are more readily annealed even at low temperatures (between 100 and 200 K), which leads to a lowering in the additional noise by as much as an order of magnitude for a temperature rise from 100 to 200 K (Fig. 3a). At the same time, the sample resistance in this temperature range does not vary since it largely depends on other types of



Figire 3. Annealing behaviour of irradiated Cu films showing additional resistivity $\Delta\rho_{\max} \simeq 90$ nΩ cm and excess noise $\Delta\alpha_{\max} \simeq 6 \times 10^{-3}$ after irradiating [146]: (a) recovery of the $1/f$-noise level (dark points) and resistivity (light points) vs. annealing temperature; (b) the frequency exponent γ vs. annealing temperature.

induced defects annealed at higher temperatures (between 200 and 300 K). Therefore, the additional resistance decreases by almost one order of magnitude for a rise in temperature from 200 to 300 K, while the excess noise remains practically unaltered over this temperature range. The authors explain these findings by the fact that $1/f$ noise in metal films arises from mobile defects whose nature awaits elucidation. The annealing of defects also leads to a smaller γ (Fig. 3b).

Furthermore, Pelz and Clarke [147] have demonstrated that excess-noise PSD in Cu films doped by In is proportional to the additional resistivity caused by radiation damage. At the same time, bombardment with krypton, which creates more cluster defects than point ones, causes a smaller noise excess per unit addition of resistivity than electron irradiation. Vacancies are supposed to be the most 'noisy' defects in a crystal. As well as Pelz and Clarke [146], Fleetwood and Giordano [148] found that annealing of amorphous AuPd films reduces noise PSD by approximately one order of magnitude at 300 K and cause a 1.3-fold decrease in resistance.

A study of films of ten different metals [55] showed a close relationship between the root-mean-square fluctuations of resistance and its portion due to scattering from defects, estimated based on the residual resistance at $T = 4.2$ K.

Miller [149] developed a model in which $1/f$ noise in metal films arises from the creation and annihilation of surface defects (e.g., vacancies) as a result of their diffusion through the medium. However, the surface model of $1/f$ noise in metals is in conflict with experimental findings (see above).

Kogan and Nagaev [150] estimated the contribution of relaxation processes responsible for internal friction (IF) in metals to $1/f$ noise. It is known [151] that at low frequencies IF is due to the motion of various defects. The assumption of a wide distribution for defect relaxation times leads to the $1/f$ spectrum of current noise [150]. $1/f$-noise intensities estimated from IF measurements agree fairly well with the Hooge parameter $\alpha \simeq 2 \times 10^{-3}$ observed in metal films. Based on this correlation, the authors speculated that $1/f$ noise in metals is caused by the same processes of movement or reorientation of defects which are responsible for the damping of elastic mechanical oscillations.

To summarize, many authors have demonstrated that $1/f$ noise in metal films is associated with microstructural defects. However, the physical nature of these defects and mechanisms of the wide distribution of their activation energy need clarification if the $1/f$ spectrum in a wide frequency range is to be explained.

3.2 Vacancy model of $1/f$ noise

Vacancies are supposed to be the most important defects in metals because their formation and migration require a relatively small energy. Vacancies are involved in both the movement and the rearrangement of other defects. The contribution of vacancies to the resistivity is defined as [152]

$$\rho_{\rm v}(t) = A n_{\rm v}(t) = A\,\frac{N_{\rm v}(t)}{N_{\rm a}}\,, \tag{3.1}$$

where A is the coefficient of proportionality, $n_{\rm v}$ is the atomic concentration of vacancies, and $N_{\rm v}$ and $N_{\rm a}$ are the numbers of vacancies and atoms in the sample, respectively.

The number density of equilibrium vacancies is [152, 153]

$$n_{\rm v} = A_{\rm v} \exp\left(-\frac{u_{\rm v} - \sigma V_{\rm v}}{kT}\right) = A_{\rm v} \exp\left(-\frac{E_{\rm v}}{kT}\right), \tag{3.2}$$

where $A_{\rm v}$ is the entropy factor, $u_{\rm v}$ is the internal energy of vacancy formation, σ is the mechanical stress, $V_{\rm v}$ is the vacancy formation volume, and $E_{\rm v}$ is the activation energy of vacancy formation.

For bulky metals, $A_{\rm v} \simeq 1$ [152]. For films, $A_{\rm v} \gg 1$ because of their high quasi-equilibrium vacancy concentration which is known to reach 0.1 – 1 at.% at room temperature [154, 155]. This is many orders of magnitude higher than the equilibrium vacancy concentration in massive metals.

Let us assume that $1/f$ noise in metal films is induced by resistance fluctuations due to fluctuations in the number of sample vacancies $N_{\rm v}$ with lifetime $\tau_{\rm v}$, which is a random variable [156]. The appearance of vacancies enhances the resistance by ΔR during the period $\tau_{\rm v}$.

The lifetime of vacancies is determined by the average distance between their sources (sinks). The average number of vacancy jumps η from creation to annihilation instants can be very large. In one study [157], the number of equilibrium vacancies in aluminium amounted to $\eta = 8 \times 10^7$. The total resistance change of the film due to vacancy number fluctuations can be described by a superposition of rectangular pulses of amplitude ΔR and duration $\tau_{\rm v}$. In this case, the fluctuation spectrum of voltage across a sample is defined through the spectrum of vacancy number fluctuations $S_{N{\rm v}}$ [156]:

$$S = \frac{S_U(f)}{U^2} = \frac{S_{\rho\,{\rm f}}(t)}{\rho_{\rm f}^2} = \frac{A^2}{\rho_{\rm f}^2 N_{\rm a}^2}\, S_{N{\rm v}}(f)\,, \tag{3.3}$$

where $S_{\rho\,{\rm f}}(f)$ is the SD of resistivity fluctuations, and $S_{N{\rm v}}(f)$ is the SD of vacancy number fluctuations in the film.

Vacancy sinks in a homogeneous sample (e.g., a massive metal or a film with perfect structure) are uniformly

distributed throughout the bulk. The probability of annihilation of each vacancy in each time interval during its lifetime is constant. The vacancy creation and annihilation events are statistically independent while the average lifetime of each vacancy equals [157]

$$\tau_{v0} = \tau_0 \exp\left(\frac{E_v}{kT}\right) . \tag{3.4}$$

In this case, the power spectrum of the noise arising when a current I_0 is flowing through the specimen containing an average number of vacancies N_v will have the Debye–Lorentz profile [5, 157]

$$S_U(f) = 4\overline{\Delta R^2} I_0^2 N_v \frac{\tau_{v0}}{1+\omega^2\tau_{v0}^2} , \tag{3.5}$$

where $\overline{\Delta R^2}$ is the variance of resistance fluctuations in the film specimen.

A voltage fluctuation spectrum of the form (3.5) associated with thermally activated vacancies was observed by Celasco et al. [157] in experiments with Al films of homogeneous microstructure and mean grain size around 0.5 μm. Figure 4 shows the energy spectra of current noise in Al films at two temperatures (see Ref. [157]). Besides the noise induced by equilibrium vacancies (curves *1*), there is a low-frequency component of $1/f^\gamma$-like spectrum, where $\gamma > 2$ (curves *2*). The measurements having been made at high temperatures, the latter component of the nonequilibrium $1/f^\gamma$ noise is supposed to be due to atomic diffusion along the grain boundaries (see Section 5).

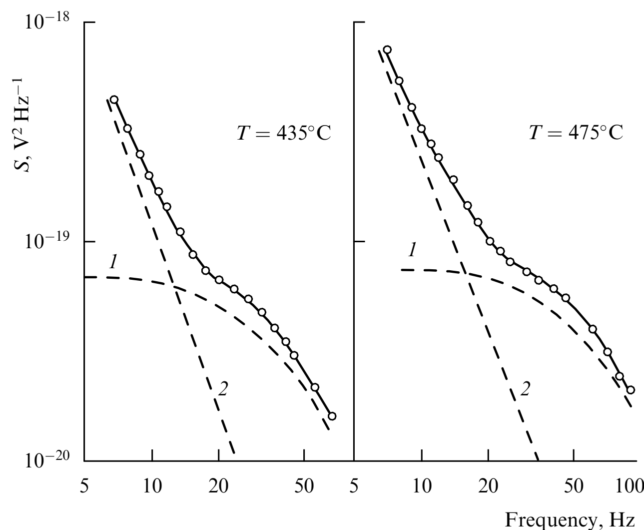


Figure 4. Energy spectra of the current noise in thin Al films taken at two different temperatures with a current density $j = 3.6 \times 10^5$ A cm^{-2} [157]. Dashed curves represent a best-fit decomposition of experimental data into components of equilibrium vacancy noise (curves *1*) and $1/f^\gamma$ noise ($\gamma > 2$) (curves *2*). Points are averages over ten samples.

Thin metal films have an inhomogeneous structure. There are different vacancy sources (sinks) nonuniformly distributed over the film volume. In sinks, vacancies may be created and annihilated. The internal energy of vacancy creation varies from one source to another, because an atom gives rise to vacancies by breaking a different number of bonds, depending on its position at the boundary of a grain, dislocation or step or at the pore surface [158]. Major vacancy sinks in metals are grain boundaries, pores, and dislocations. In films, vacancies are largely created at grain boundaries which was confirmed in an experiment [157], where the mean vacancy diffusion range in Al films turned out to be comparable with the mean grain size.

The crystalline structure of a real film is not in a thermodynamically equilibrium state and its free energy is enhanced due to numerous defects. Being sources and sinks of vacancies, these defects change their position upon emitting or absorbing vacancies, i.e. dislocations can move and pores grow. The concentration of microdefects in the film is reduced by annealing. The situation may be realized after annealing at lower temperatures, when the defects turn out to be fixed. In this case, the defects move very slowly or do not move at all, and each source is characterized by a quasi-equilibrium concentration of vacancies [158]. The emission or absorption of vacancies by a source does not lead to a change in the free energy of the crystal which thus remains in the quasi-equilibrium (local equilibrium) state, while the arising flicker noise may be regarded as quasi-stationary due to vacancy number fluctuations.

Numerous experiments confirmed that external conditions (temperature, current, etc.) may cause a film transition to the nonequilibrium state and give rise to nonstationary $1/f$ fluctuations.

Creation of a vacancy in a perfect crystal requires that about half the bonds in the lattice be broken, on the assumption that an atom which leaves the bulk is bound at the crystal or pore surface. The energy of vacancy creation in metals is $u_v = 0.7-2.5$ eV [158], and the energy per atomic bond in the lattice equals $u_{v1} = 0.1-0.5$ eV.

At thermodynamic equilibrium, the vacancy creation and annihilation rates are identical, and the average vacancy concentration remains constant in time. The lifetime τ_v of vacancies depends on the distance L_v between their sinks and is defined by the relation [158]

$$\tau_v = \frac{L_v^2}{\pi^2 D_v} , \tag{3.6}$$

where D_v is the vacancy diffusion coefficient.

A set of time constants τ_v is related to the distribution of activation energy for vacancy diffusion and the distance L_v between vacancy sinks, which are random variables for the film volume. The randomness of the activation energy is due to a variety of structural imperfections. Owing to the nonuniform distribution of sinks in the bulk of real metal films, they have a large set of relaxation times related to the mechanism of vacancy creation and annihilation, which is responsible for $1/f$ noise over a wide frequency range.

Let us estimate the relaxation-time distribution limits at $T = 350$ K. The minimum relaxation time τ_h appears to correspond to the vacancy lifetime (3.4) provided the atom overcomes the potential barrier equal to the energy needed to break a single bond in the crystal lattice. The assumption of $E_v = 0.1$ eV yields $\tau_h \simeq 10^{-11}$ s; for the highest boundary frequency of the $1/f$ spectrum, one finds $f_h \simeq 10^{10}$ Hz in accordance with (2.10).

The lower boundary frequency f_l of the $1/f$ spectrum may be related to the vacancy lifetime (3.6). A vacancy arising at the boundary of the grain or a micropore can diffuse through the bulk of a crystallite until it reaches a neighbouring sink. If

the distance between the sinks is taken as $L_v = 20$ nm and the lattice diffusion coefficient for Al at $T = 350$ K is $D_v \simeq 10^{-24}$ m^2 s^{-1} [153, 158], then it follows from (3.6) that $\tau_l = 4 \times 10^7$ s, while $f_l \simeq 4 \times 10^{-9}$ Hz in accordance with (2.10). These estimates give evidence for a broad range of relaxation times for equilibrium vacancies.

Assuming that the variance of vacancy number fluctuations in a sample is identical to their mean number N_v [121], the SD of vacancy number fluctuations in the frequency range f_l to f_h can be written as [156]

$$S_{N\text{v}}(f) = \frac{N_v}{f \ln(f_h/f_l)} \ . \tag{3.7}$$

The substitution of (3.7) into (3.3) yields the relative SD of voltage fluctuations in a film:

$$S = \frac{S_U}{U^2} = \frac{A^2 n_v}{\rho_f^2 N_a f \ln(f_h/f_l)} \ . \tag{3.8}$$

According to (3.2) and (3.8), the PSD of $1/f$ noise induced by fluctuations in the number of vacancies exhibits activation temperature dependence and exponential dependence on the mechanical stress.

It follows from (3.8) that the parameter α defined in (2.3) is

$$\alpha = \frac{A^2 N_c n_v}{\rho_f^2 N_a \ln(f_h/f_l)} \ . \tag{3.9}$$

Let us estimate the parameter α for Al films. Assuming $\rho_f = 3$ μΩ cm, $N_c/N_a = 3$, $A = 220$ μΩ cm [152], and $\ln(f_h/f_l) = 40$, expression (3.9) gives $\alpha \simeq 3 \times 10^2 n_v$. Computed and experimental values for films examined under different conditions (see Fig. 3 in Ref. [156]) agree if $n_v \simeq 3 \times 10^{-2} - 6 \times 10^{-6}$ is true of thin films [154, 155], with $n_v \simeq 10^{-5}$ corresponding to $\alpha \simeq 2 \times 10^{-3}$.

3.3 Experimental results in support of the vacancy mechanism of $1/f$-like noise in metal films

3.3.1 Dependence of $1/f$ noise on film thickness. Experimental evaluation of the $1/f$-noise PSD dependence on the film thickness is difficult because the noise magnitude varies in films of a given thickness differing in terms of structure and concentration of microdefects. The level of $1/f$ noise in a film may be tens of times higher or lower than in another film of identical thickness made under the same technological regimes. The difference may even exceed the effect being examined [10, 13]. Films for experiments must have a thickness $h > 10$ nm, since thinner layers are normally discontinuous, having an island or reticular structure [155]. Films with thickness $h > 1$ μm are inconvenient because it is difficult to ensure the current density necessary to detect flicker noise [12].

In order to study the thickness dependence of noise, one must have films of different thicknesses with similar concentration of microdefects and impurities. Such films can be deposited only in one technological circuit, for example, by moving the shutter over the substrate during condensation [128, 159] or reducing the film thickness by anodizing [162, 163].

Numerous studies with metal films have shown that $1/f$ noise arises in the bulk of the film, because the SD of fluctuations is inversely proportional to the volume V and thickness h (the width being unaltered) or to the number of carriers (atoms) in the sample [12, 22, 33, 37, 165]:

$$\frac{S_U(f)}{U^2} \propto \frac{1}{V} \propto \frac{1}{h} \propto \frac{1}{N_c} \ . \tag{3.10}$$

The Hooge formula (2.1) obeys this dependence, which was reported to occur in thin gold [22] and platinum [37] films. The thickness of platinum films was increased eight-fold ($h = 8 - 65$ nm), while the number of atoms varied from 10^8 to 10^{14}.

If $1/f$-noise appearance is associated with the sample surface (e.g., in the Celasco et al. model [166]), then the thickness dependence of the noise PSD is presented as

$$\frac{S_U}{U^2} \propto \frac{1}{h^2} \ . \tag{3.11}$$

According to this model, $1/f$ noise is generated in a thin subsurface layer (film/substrate interface), while the rest of the film is 'silent' and only shunts the subsurface source of the noise. This hypothesis is at variance with experimental findings [22, 37].

It was shown that the surface of Ag films is more 'noisy' near 100 K than the bulk [12]. Gold microconductors were reported to exhibit the (3.11)-like dependence typical of surface noise [167].

Bakshee et al. [162, 163] discovered a surface source of noise in Al films on oxidized silicon, which developed at the film/substrate interface during high-temperature annealing in an atmosphere of hydrogen ($T_{\text{ann}} = 723$ K, $t_{\text{ann}} = 1$ h). The presence of this source was confirmed by the dependence of the noise PSD on the film thickness in conformity with (3.11). The noise intensity in the films on the plate side not subjected to annealing was one or two orders of magnitude lower and varied with the film thickness in accordance with (3.10). The film thickness was changed by anodizing.

The thickness dependence of the noise PSD may have quite a different form from that given by formulas (3.10) and (3.11). Figure 5 shows such a dependence for films on a glass substrate [128]. The film thickness was modulated by shutter movements. Figure 5 also presents the thickness dependence of the through-macropore density n_p (right scale). The lowest

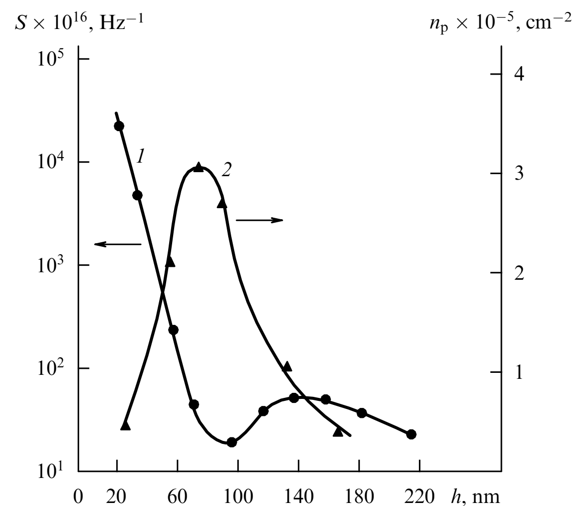


Figure 5. Thickness dependences of $1/f$-noise PSD at 20 Hz (curve *1*) and through-macropore density (curve *2*) for Cr films on glass [128]; condensation rate $w_{\text{cond}} = 0.5$ nm s^{-1}, condensation temperature $T_{\text{cond}} = 470$ K.

noise and the highest macropore density were observed in the thickness range between 80 and 100 nm. The minimum resistivity also occurred within this range [159]. The observed effects can be accounted for by the thermodynamic advantage of vacancy withdrawal from the bulk of the film to macropores at a given thickness [168, 169]. For this reason, films of thickness $h \simeq 100$ nm have the lowest concentration of vacancies. The rapid rise in noise intensity with decreasing thickness (at $h < 40$ nm) may be due to the structural dispersion of thin films. In films thicker than 150 nm, macropore formation is thermodynamically disadvantageous. The generation of $1/f$ noise in such films is a volume effect related to fluctuations of the vacancy number in the sample even though the dependence of the noise PSD on the film thickness (see Fig. 5) disobeys expression (3.10).

Alloy films, unlike metal films, have no minimum in the thickness dependence of the noise PSD. Specifically, flicker noise in films of resistive (metal-silicide) alloys MLT-3M and RS-3001 monotonically abates with increasing thickness from 20 to 200 nm [159, 161, 170]. This phenomenon is due to the absence of vacancy diffusion mechanism in alloys [158], where vacancy movement to the sinks with the formation of macropores is hindered.

The origin of a surface noise source in aluminium films on oxidized silicon following high-temperature annealing [162, 163] may be attributed to the vacancy mechanism of $1/f$ noise. Heating the films results in Al diffusion into silicon oxide film [171]. Simultaneously, a layer with high levels of vacancy concentration and $1/f$ noise is formed at the film/substrate interface which accounts for the appearance of the surface noise source.

Generally speaking, the surface noise source observed in some experiments may have a different nature. For example, it may be related to the drift of alkali metal ions [128] in films on customary glasses or the effect of gas layer absorbed at the substrate [172]; these gases subsequently dissolve in the surface layer of the condensate giving rise to a contaminating atom–vacancy complex [158, 173]. This, in turn, leads to a high vacancy concentration in the film and an elevated noise level. The formation of such complexes was confirmed by measurements of internal mechanical stresses in molybdenum films [174].

A surface noise source can also arise from the nonuniform distribution of mechanical stresses throughout the depth of the film; the stresses were reported to be especially strong at the film/substrate interface [107]. Under certain conditions, the stresses in the subsurface layer can make a substantial contribution to the $1/f$ noise of the film.

3.3.2 Variation of the flicker noise in metal films caused by annealing or aging. The effect of annealing on $1/f$ noise was examined in aluminium [91, 129, 176] and chromium [128] films prepared by thermal evaporation in a vacuum. The samples were annealed in a vacuum chamber to prevent their oxidation.

Aluminium films, deposited at the condensation temperature $T_{\text{cond}} = 370$ K and condensation rate $w_{\text{cond}} = 6$ nm s^{-1}, were used to evaluate the time of defect annealing [91, 176]. The films were first heated up to $T_{\text{ann}} = 410$ K for 5 min, then annealed for another 5 min, and finally cooled for 1 h. This heat cycle was repeated 3 or 4 times following measurement of the noise PSD at room temperature. The cooling rate was chosen to be low (less than 2 K min^{-1}) to exclude quenching of defects. The annealing resulted in a fall of $1/f$-noise level owing to a decreased concentration of nonequilibrium microstructural defects [177]. A few heat cycles were needed to anneal defects, which were completely removed over tens of minutes at $T_{\text{ann}} = 410$ K. The annealing of defects and decrease of $1/f$-noise magnitude were especially pronounced within the first two or three thermocycles of the total duration 10–15 min.

The variation of $1/f$-noise PSD during the course of thermal treatment was investigated in Al films deposited at $T_{\text{cond}} = 400$ K [129]. The films were first heated up to 400–450 K for 20–60 min, hold at this temperature for another 20–60 min, and cooled down to 300–320 K for 60–90 min. The PSD of flicker noise was measured throughout the thermocycle at a fixed frequency and a given current through the sample. Simultaneously, the variations of film resistance were also measured.

Typical temperature dependences of the $1/f$-noise PSD and resistance in Al films during a thermocycle (with regard for current heating) are shown in Fig 6. Here, S_0 is the $1/f$-noise PSD at $f = 120$ Hz and R_0 is the film resistance prior to

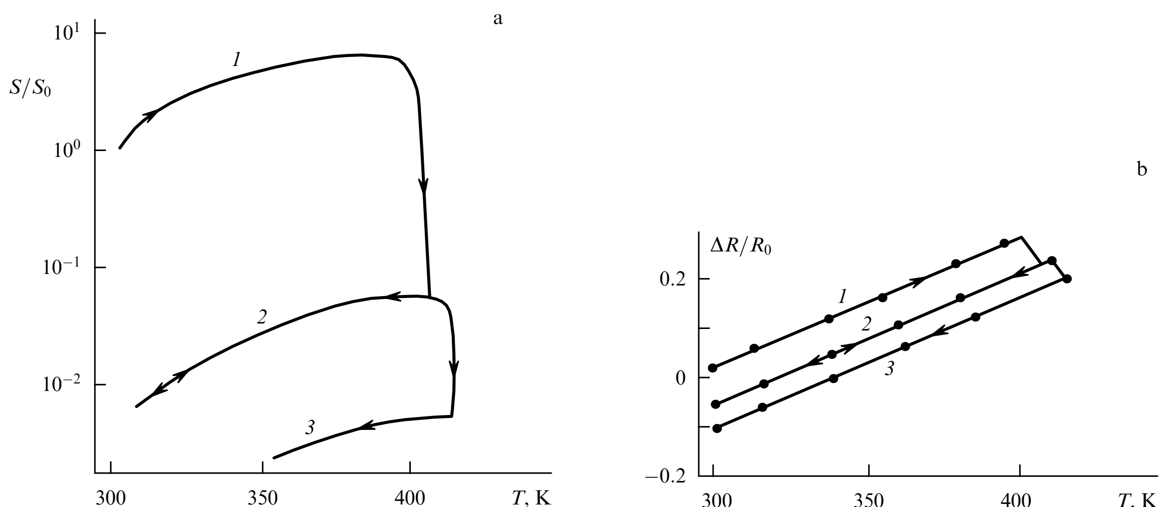


Figure 6. Temperature dependences of $1/f$-noise PSD (a) and resistance (b) for Al films during thermocycling [129]; $h = 100$ nm, $T_{\text{cond}} = 400$ K, $f = 120$ Hz; (*1, 2*) the first cycle; (*2, 3*) the second cycle.

annealing ($\Delta R = R - R_0$). The PSD of $1/f$ noise increases with heating but drops sharply by 1–3 orders of magnitude for 20–30 min at the annealing temperature $T = 400-410$ K. Simultaneously, there is a 2–10% irreversible fall in resistance. During the second (or third) heating cycle (curves *2* and *3* in Fig. 6), both the noise level and the resistance grow reversibly parallel to the cooling curve in the cycle unless the maximum annealing temperature of the previous cycle is exceeded. Exceeding the maximum annealing temperature of the previous cycle results in a further irreversible decrease of noise level and resistance. However, the magnitude of the relaxation-induced abatement of $1/f$ noise during each cycle is much less than in the previous cycle. The available data indicate that films have defects with different annealing energies; those with smaller activation energy are annealed at a lower temperature. This inference is confirmed by the results of a study concerning the effect of annealing temperature on $1/f$-noise level in Cr films [128].

According to the Mattissen rule [155], portions of resistivity ρ_f in a metal film originating from scattering by phonons ρ_L, defects ρ_d, and surface boundaries of the film ρ_s are additive at low concentrations of impurities and defects:

$$\rho_f = \rho_L(T) + \rho_d + \rho_s\,. \tag{3.12}$$

Aluminium films studied in Ref. [129] had an initial (before annealing) resistivity $\rho_f = 3.8-4.2$ μΩ cm (for a massive metal, $\rho_0 = 2.7$ μΩ cm [178]). The linear growth of resistance with temperature (Fig. 6b) was due to electron scattering from phonons [152] [term $\rho_L(T)$ in expression (3.12)]. The decreased concentration of point defects after annealing had no effect on $d\rho_f/dT$, but caused only a parallel shift in the temperature dependence of resistance (Fig. 6b).

Therefore, the irreversible decrease in the resistance and PSD of flicker noise at 400–410 K can be attributed to annealing of defects in the crystal lattice. Assuming $A = 2.2$ μΩ cm as the contribution of one atomic percent of vacancies to the resistivity for Al [152] leads to the conclusion that a 10% fall in the resistivity (caused by annealing) corresponds to a decrease in vacancy concentration Δn_v by approximately 0.9 at.%. This appears to be a realistic estimate for metal films [154, 155].

Studies [94, 179] concerned the effect of induced defects annealing on $1/f$-noise PSD in Al films over the temperature range 10 to 300 K. The defects were induced by irradiating the films with electrons of energy 1 MeV, and fluence of 3.7×10^{23} e m^{-2} at 10 K. The irradiated samples showed a six-fold rise in the noise level and a 25% increase in resistance (at $T = 10$ K). Isochronic annealing of the samples for 600 s, with the temperature gradually rising to 300 K, resulted in the recovery of the initial $1/f$-noise level and resistance at 200 K (Fig. 7). Measurements at 40 K revealed an additional stage in the $1/f$-noise recovery process at $T = 70$ K, which is absent in the recovery of resistance (see inset to Fig. 7). The authors explained this finding by a lower number of mobile defects. It follows from Fig. 7 that the defects differ in terms of annealing temperature. Ref. [179] reports the distribution of noise activation energy $G(E_a)$ derived from the temperature-dependent noise PSD for Al films, based on the model of Dutta et al. [81]. In unirradiated samples, the distribution of $G(E_a)$ was uniform over the range of a few hundredths to 0.2 eV. In irradiated samples, further annealed at $T_{ann} = 105$ K, the uniform distribution could be followed for activation energies of up to 0.15 eV.

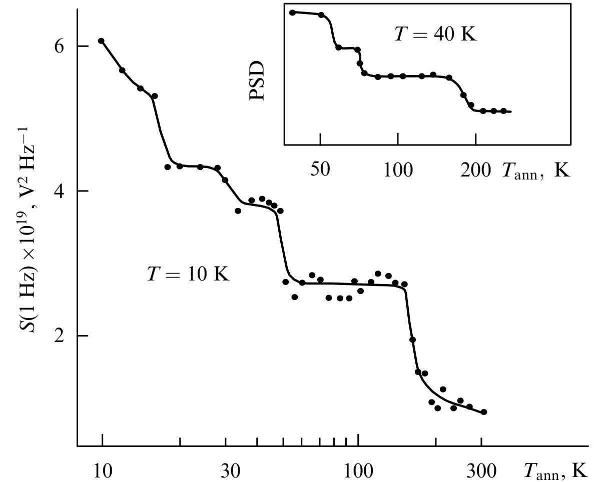


Figure 7. Recovery of $1/f$-noise PSD in irradiated Al films at 10 K (in the inset: at 40 K) on isochronic annealing with a gradual rise in temperature [94]; annealing time ranges 600 s, $f = 1$ Hz.

The effect of γ-radiation on $1/f$ noise in niobium films was evaluated in Ref. [91]. The activation energy of induced microdefects in Nb being higher than in Al [94] or Cu [146], the authors measured $1/f$ noise at room temperature. An increase in $1/f$ noise following the irradiation of Nb films was due to the creation of additional defects in the crystal lattice. Current-induced annealing decreased the noise magnitude to approximately that in unirradiated samples.

Now, let us consider changes in the $1/f$-noise PSD with time in naturally aging films. Figure 8 shows the time dependence of the noise PSD at $f = 120$ Hz and the resistivity for Al films deposited at $T_{cond} = 300$ K [91]. These samples had a highly nonequilibrium structure with a large number of microdefects. Soon after condensation, the defects were 'annealed' at room temperature due to diffusion processes. This resulted in an irreversible reduction of both $1/f$ noise and resistance (time $t = 0$ corresponds to the termination of film condensation). The PSD of $1/f$ noise decreases in compliance with the exponential law (with constant $\tau_0 = 5$ min till time

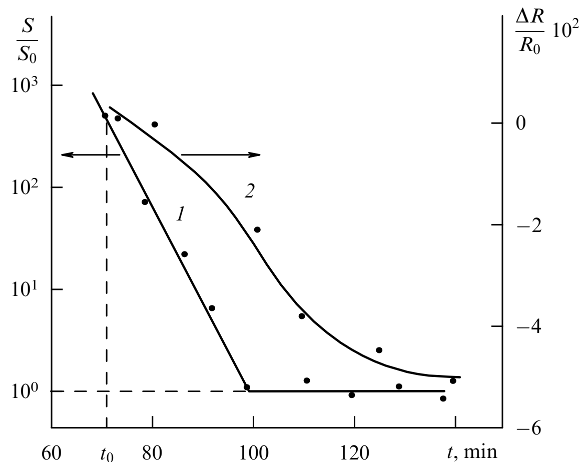


Figure 8. Variation of $1/f$-noise PSD at 120 Hz (curve *1*) and resistance (curve *2*) with time for Al films [91]; $h = 100$ nm, $T_{cond} = 300$ K, R_0 is the film resistance 70 min after condensation, S_0 is the noise PSD more than 100 min after condensation.

$t = 100$ min) characteristic of point microdefects annealing [177]:

$$\Delta S = S - S_0 \propto \exp\left(-\frac{t}{\tau}\right), \qquad (3.13)$$

where S_0 is the steady-state value of the noise PSD settled within time $t \geqslant 100$ min after the termination of condensation.

Figure 8 shows that the relaxation of the noise PSD to a steady-state value requires less time than the relaxation of resistance. This observation taken together with the results for annealed and irradiated Cu films in Fig. 3 indicates that flicker noise at a given frequency is due to mobile defects with small activation energies, which are annealed faster and at lower temperatures than defects contributing only to resistance. Defects with higher activation energies contribute therewith to the PSD of $1/f$ noise at lower frequencies.

Similar effects of the annealing temperature and duration on $1/f$ noise were observed in Cr films [128, 180] deposited at $T_{\text{cond}} = 400$ K. In a film deposited on a heated substrate, the relaxation of $1/f$ noise occurred faster, i.e. for the time necessary for the substrate to cool from condensation to room temperature [180].

Analysis of the dependence of the $1/f$-noise PSD on annealing time in Cr films [128] has demonstrated that the noise PSD decreases in accordance with the law (3.13), with a time constant τ_0 of about 5 min. At $T_{\text{ann}} = 620$ K, almost all defects with low activation energy are annealed over 30 min.

The frequency exponent γ was shown to decrease in aging Cr films [180]. Measurements taken immediately after the fabrication of Cr films gave $\gamma \simeq 2-3$. It decreased to $0.7-1.2$ within $30-45$ min after placing the samples in a vacuum. Higher γ values characterized more nonequilibrium condensates with a higher concentration of excess vacancies. The level of $1/f$ noise in aging films decreased in parallel with a reduction of the intrinsic mechanical stresses and resistivity [180] due to coalescence of vacancies into pores [154].

The effect of annealing on the $1/f$-noise level in thin-film Ti–Al and V–Al contacts was investigated in Ref. [160]. The condensation and annealing temperatures were 390 and 520 K, respectively. Figure 9 shows the $1/f$-noise PSD dependence on the Al-film thickness for a Ti–Al contact with a Ti-film thickness of 30 nm. Curves *1* and *3* were obtained immediately after film contact deposition; curves *2* and *4*, after annealing for 15 min. It is clear that the annealing resulted in a decrease of the noise level and a change in the thickness dependence of the noise PSD for Al films. The minimum corresponding to the lowest vacancy concentration was associated with an Al-film thickness of $h_{\text{Al}} \simeq 80$ nm and occurred concurrently with the transient resistance minimum and the macropore density maximum [160]. These results are similar to those presented in Fig. 5 for continuous Al films. For flicker noise in the contacts before annealing, $\gamma = 2-3$, whereas for Ti and V films it is $1-1.2$. Elevated γ values for contacts which were not subjected to annealing suggest the appearance of a nonequilibrium $1/f^{\gamma}$ noise in these systems.

The results of annealing and aging studies on metal films indicate that they contain mobile defects with different activation energies annealed at different temperatures and during different time periods. Annealing of films at temperatures higher than the condensation temperature causes an abatement of flicker noise due to the decreased concentration of vacancies in the condensate. Flicker noise in as-deposited metal films contains a component of nonstationary $1/f$ noise induced by excess vacancies, the level of which depends on the concentration of these vacancies. The same component was found in metal films irradiated with high-energy particles which influence the concentration of microdefects. Annealing of nonequilibrium microdefects results in their quasi-equilibrium concentration in the films which persists (or changes very slowly) at temperatures below the annealing temperature. The $1/f$ noise in such films may be regarded as stationary or quasi-stationary.

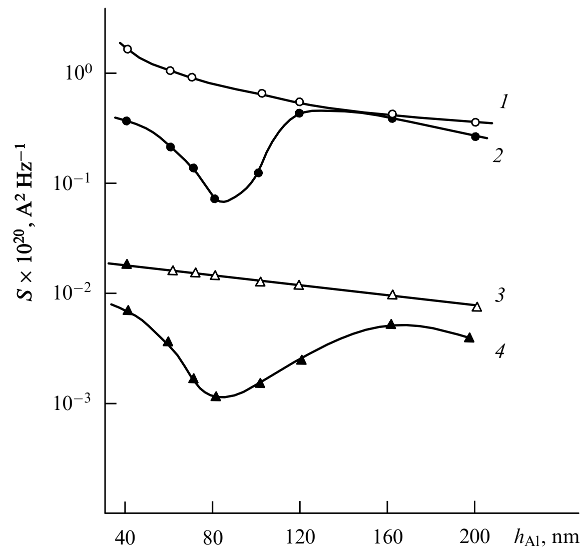


Figure 9. $1/f$-noise PSD of a Ti–Al film contact vs. Al-film thickness ($h_{\text{Ti}} = 30$ nm) [160]: (*1*, *3*) before annealing; (*2*, *4*) after annealing; (*1*, *2*) $f = 30$ Hz; (*3*, *4*) $f = 200$ Hz.

3.3.3 Temperature dependence of $1/f$ noise. Investigations into the temperature dependence of $1/f$ noise in annealed metal films are conducive to the elucidation of its physical nature. The temperature dependence of $1/f$ noise was first examined by Hooge and Hoppenbrouwers in gold films [22], where the dependence of $1/f$ noise was not stronger than predicted by the $\alpha \propto T^{1/2}$ law. Voss and Clarke [33] reported an abatement of $1/f$ noise in metal films with decreasing temperature but failed to specify the type of this dependence.

Eberhard and Horn [34, 144] appear to have been the first to observe the strong temperature dependence of $1/f$ noise in Ag, Cu, Au, and Ni films. Later, it was found in Bi [181] and Al [129] films. In annealed Ag, Au, Cu, and Al films, the temperature dependence of the $1/f$-noise PSD was shown [34, 129] to be of activation nature over a certain range with the activation energy E_{a} (Fig. 10):

$$\frac{S_U}{U^2} \propto \exp\left(-\frac{E_{\text{a}}}{kT}\right). \qquad (3.14)$$

For Ag, Au, and Cu films, this range lies between 220 and 350 K [34], while for Al between 220 and 460 K [129].

In Refs [34, 144], the observed values of $E_{\text{a}} = 0.1-0.2$ eV for Ag, Cu, and Au films are ascribed to vacancy formation energies although they are significantly lower than those in bulky metals. The energy E_{a} was shown to grow slowly with the film thickness, probably due to the bigger grain size [34] (see Section 3.3.6). In all the experiments, the activation

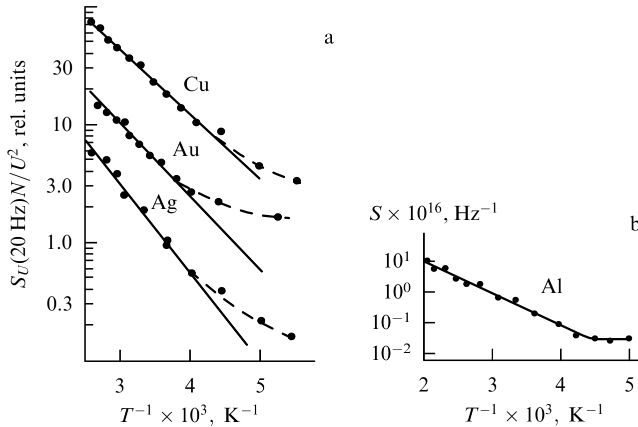


Figure 10. Relative $1/f$-noise PSD for annealed (a) Cu, Au, Ag [34] and (b) Al [129] films vs. inverse temperature: (a) N is the total number of atoms in the film, $h = 80$ nm, $f = 20$ Hz (sapphire substrate), dashed lines show the tendency to flattening at low temperatures; (b) $h = 100$ nm, $f = 120$ Hz (glass substrate).

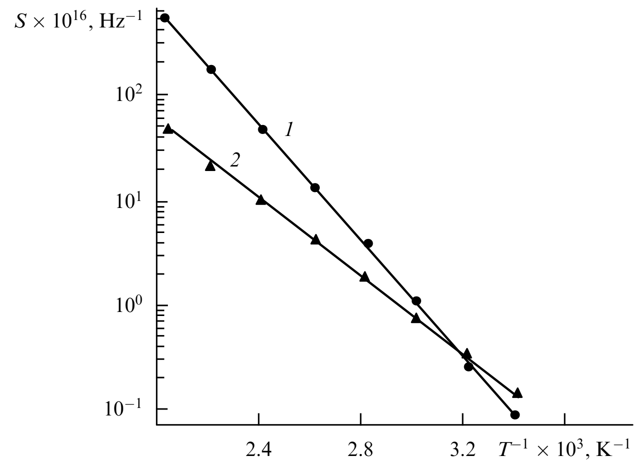


Figure 11. Temperature-dependent $1/f$-noise PSD for Ta (*1*) and Mo (*2*) films: $h = 0.5$ μm; $f = 10^3$ Hz [172].

temperature dependence held true only for $1/f$ noise. The film resistance increased linearly with temperature, in accordance with the dependence $\rho_L(T)$ in (3.12).

Beyond the activation region, the low-temperature dependence of $1/f$ noise tends to flatten [34, 129, 144], probably because mobile defects are 'frozen' at low temperatures and the noise PSD becomes independent of temperature. Observed $1/f$ noise in the plateau region is supposed to be related to the effect of zero-point atomic oscillations in the lattice on the migration of point defects [182]. Alternatively, this noise can be generated by scattering from the lattice [32].

Eberhard and Horn [34, 144] observed maxima in the temperature dependence of $1/f$ noise in Ag and Cu films which were later explained by Yakimov [140], who related the mechanism of $1/f$ noise to the physical sorption and desorption of ambient gases by a metal film. Going over of the noise PSD through the maximum at a certain temperature (see Figs 5 and 6 in Ref. [34]) can also be accounted for by diminished macrostresses in the film due to a decrease in thermal stresses, which become negative at a temperature above the condensation temperature T_{cond} [107] (see Section 3.3.4). Unfortunately, it is impossible to evaluate the effect since Ref. [34] specifies neither the condensation temperature nor the internal stresses.

The effect of structural factors on the temperature dependence of the noise PSD was assessed for Cr [105], Mo and Ta [92, 143, 175] films. The Cr films were prepared by means of thermal evaporation in a vacuum on devitrified glass or oxidized silicon substrates followed by annealing in a vacuum chamber at $T_{\mathrm{ann}} = 620$ K for 30 min. Films of refractory metals were deposited on oxidized silicon wafers by the ion sputtering technique in the temperature range 300 to 500 K. All the films showed an activation temperature dependence of the noise PSD, which was well approximated by expression (3.14). The experimentally found E_a values corresponded to the energy per one or two bonds in the lattice of a bulky metal. Higher E_a values were recorded for metals with higher atomic binding energies in a crystal (Mo, Ta), which supports the hypothesis of the vacancy mechanism of $1/f$ noise in metals.

Figure 11 shows the dependence of the noise PSD on inverse temperature for Mo and Ta films at a frequency of 1 kHz. Activation energies derived from these dependences were $E_a = 0.4-0.45$ eV for Ta, and $E_a = 0.3-0.35$ eV for Mo.

It has been demonstrated [105, 164] that the activation energy E_a is related to the film microstructure, i.e. the mean grain size and the degree of crystallization inside a grain. Higher E_a values are typical of course-grained films. Specifically, an increase of the mean grain size in Cr films from 30–40 to 50 nm led to a rise in the E_a value from 0.2–0.3 to 0.4 eV (for the noise PSD at 1 kHz), with other experimental conditions being identical [164].

The Cr films with a small degree of crystallization inside grains (practically amorphous structures) had low E_a values (0.14 ± 0.02 eV) which increased to 0.33 ± 0.05 eV in films with a moderate degree of crystallization, the mean grain size (30–40 nm) in the two films being almost identical [105].

The activation energy E_a has been shown to depend on the frequency at which the $1/f$-noise PSD is measured [92, 143, 175]. E_a values increased with decreasing frequency (Fig. 12). This observation indicates that the spectrum of $1/f$ noise at different frequencies is formed by defects with different activation energies. Moreover, defects with higher activation energies contribute to the energy spectrum at lower frequencies.

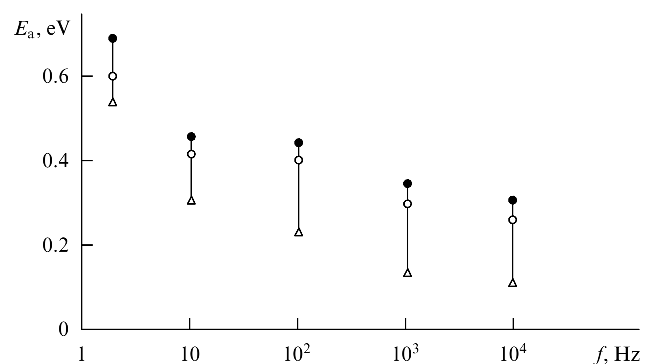


Figure 12. Activation energies for $1/f$-noise PSD in Mo films of varying thickness at different frequencies [143, 175]: $h = 68$ nm(•); $h = 162$ nm (○); $h = 560$ nm (△).

Dutta et al. [144] studied the effect of the substrate material (quartz, sapphire) on the temperature dependence of $1/f$ noise in Cu and Ag films. The noise level in Ag films proved to be unrelated to the type of the substrate over the entire temperature range examined (100 – 600 K). At the same time, Cu films on different substrates were dissimilar in terms of noise magnitude at temperatures below room temperature. It was suggested to account for this finding by the existence of two types of noise in metal films, i.e. 'type A' showing a weak temperature dependence, and 'type B' which is characteristic of a metal and shows activation temperature dependence. Based on this conjecture, the authors explained the weak temperature dependence of $1/f$ noise in gold films [22], and the strong one in Ag and Cu films [144]. At $T > 100$ K, the overall noise in a film was largely due to 'type B'. However, the mechanisms of types A and B noise were not clarified.

At the same time, the results of studies [143, 175, 183] suggest a strong temperature dependence of $1/f$ noise induced by fluctuations in the number of vacancies in the sample. In films with a low concentration of mobile defects, $1/f$ noise is generated by fluctuations of mobility upon scattering from phonons; this noise shows a weak temperature dependence [22].

3.3.4 Effect of internal macrostresses on flicker noise in metal films.

An important argument in support of the vacancy mechanism of flicker noise in metal films is the dependence of its PSD on mechanical stresses σ experimentally examined in Al, Mo, and Ta films [92, 106, 143, 183] and obeying the law

$$S \propto \exp\left(\frac{\sigma V_a}{kT}\right), \tag{3.15}$$

where V_a is the activation volume.

Figure 13 shows the noise PSD dependence on macrostresses constructed in semilogarithmic coordinates for Cr and Mo films [143, 183]. The activation volumes deduced from the slopes of the straight lines were $V_a = (1.4 \pm 0.4) \times 10^{-29}$ m^3 for Cr, and $V_a = (1.1 \pm 0.3) \times 10^{-29}$ m^3 for Mo [92, 143]. These values are close to the atomic volumes of bulky metals ($\Omega_{Cr} = 1.2 \times 10^{-29}$ m^3, $\Omega_{Mo} = 1.56 \times 10^{-29}$ m^3) [57].

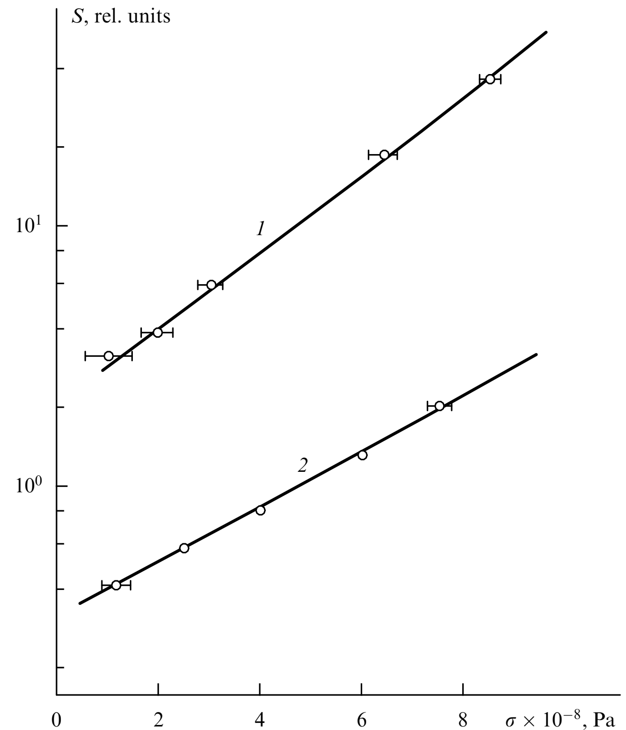


Figure 13. $1/f$-noise PSD vs. internal mechanical stresses in Cr (*1*) and Mo (*2*) films [183]: (*1*) $f = 120$ Hz; (*2*) $f = 480$ Hz.

The dependence $S(\sigma)$ which obeys the (3.15)-law was also found in experiments on Al films obtained by thermal evaporation in vacuum [106] or the atmosphere of argon [92]. However, the activation volumes involved are bigger than the volume per atom, due to peculiarities of the aluminium f.c.c. crystallographic structure. This finding has been discussed at greater length in Ref. [92].

It should be emphasized that the dependence $S(\sigma)$ in Fig. 13 is constructed based on the averaged value for internal macrostresses σ. At the same time, microstresses are irregularly distributed across the local film areas and can be significantly (sometimes, by an order of magnitude) greater than the macroscopic stress at intergrain boundaries. Occasionally, microstrains amount to $\varepsilon_m \simeq 10^{-2}$ [154].

Because microstresses are randomly distributed in the bulk of the film and their magnitudes are locally different, the activation energy of vacancy creation and migration shows 'spreading'. Changes of the activation energy ΔE_a due to microstresses were evaluated in Ref. [92] on the assumption of the validity of Hooke's law, using the formula $\Delta E_a = \sigma_m V_a = \varepsilon_m E_{Cr} V_a$. If the microstrain in a Cr film is $\varepsilon_m = 10^{-2}$, $V_a = 1.2 \times 10^{-29}$ m^3, and Young's modulus $E_{Cr} = 27.3 \times 10^{10}$ Pa [178], then $\Delta E_a \simeq 0.2$ eV. This value is comparable with the energy per bond in a crystal and accounts for the continuous activation energy spectrum of microdefects in films from fractions of to a few electron-volts, which is necessary to obtain the $1/f$ spectrum over a wide frequency range.

Of interest for the analysis of effect of mechanical stresses on flicker noise is the approach developed in Refs [184, 185]. The authors maintain that tensile mechanical stresses facilitate creation and heating of dilatons regarded as negative solid-state density fluctuations. The dilaton region irreversibly receives a heat flux from the adjacent regions of the body which leads to a break of a chemical bond and the appearance of a vacancy [184]. In the process of vibrational relaxation, dilatons play the role of temperature fluctuation sources. They are most effective in the domains of structural inhomogeneity in the crystal, for instance, at grain boundaries, where anharmonism of the interatomic interactions arises and the time of vibration energy relaxation increases.

Based on the dilaton concept, Timashev [185] predicted the relationship between the relative PSD of $1/f$ noise and tensile mechanical stress in solids in the form of a phenomenological expression coincident with (3.15) for the case of $\sigma V_a \leqslant kT$.

Tensile mechanical stresses affect the $1/f$-noise level in metal films through concentration and mobility of microdefects. In turn, a change in the concentration of microdefects during the relaxation processes leads to the alteration of structural macrostresses, which increase the elastic strain energy. But in so doing the energy of nonequilibrium vacancies is diminished. Equilibrium is reached when the thermodynamic potential is minimal. Coalescence of vacancies into the pores is arrested when their motion to the sinks becomes thermodynamically disadvantageous. For this reason, in films with an initially elevated concentration of

vacancies, their final amount after annealing is higher; this accounts for the increased $1/f$ noise in such films.

Internal macrostresses also affect the spectral characteristics of $1/f$ noise. Films with larger stresses have higher γ values [92, 164, 180, 186]. Figure 14 presents the dependence of the frequency exponent γ on macrostresses in Cr and Al films [92, 164, 186]. γ grows with an increase in absolute values of macrostresses. These results indicate that different γ values in films of the same material may be due to variations in the level of internal mechanical stresses.

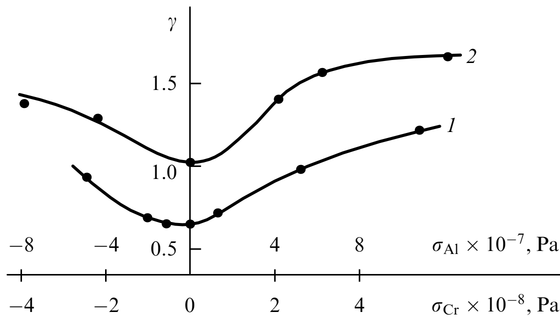


Figure 14. Frequency exponent vs. internal mechanical stresses in Al (curve *1*) and Cr (curve *2*) films [92, 164, 186].

3.3.5 Dependence of $1/f$ noise on mechanical strains. Fleetwood and Giordano [187] observed a rise of about one order of magnitude in $1/f$ noise upon deformation of platinum, gold, silver, lead, and tin films deposited on glass and elastic substrates. Removal of the strain caused noise relaxation to smaller levels for a period of a few hours to several months although the resulting noise magnitude remained higher than initially. The authors explained these changes by the creation and annihilation of structural microdefects. However, the method used to apply mechanical stresses [187] prevented the determination of their level and the nature of the film deformation (elastic or plastic); it also failed to provide quantitative data characterizing the effect of mechanical stresses on $1/f$ noise.

Also, it was shown in Ref. [187] that $1/f$ noise in films on elastic substrates is less stable than in films on glass (the amount of applied mechanical stress in the elastic substrates was higher).

Quantitative studies broaching the effect of externally induced controlled strains and mechanical stresses on $1/f$ noise in Cr films on glass and Al films on the elastic polyimide substrate PM-1 are reported in Refs [180, 188, 189].

Tensile and compressive stresses were generated in Cr films by bending the console-fixed substrate. Tensile stress developed when the external bending force was applied to the free end of the substrate perpendicular to its plane from the side occupied by the deposited film. The same force was applied from the opposite side to raise a compressive stress [180, 188]. Displacement of the unfixed end of the substrate with the film was used as a measure of the relative deformation and for calculating the mechanical stress in the elastic strain region by the formulas presented in Ref. [180].

Bending the substrate (console) by an external force generates an asymmetric plane stress in the film. In the case of a tensile force, the internal stresses σ and stress σ' induced by the external force parallel to the x axis (along the console) are summed to give the total stress in the film

$$\sigma_x = \sigma + \sigma'(x)\,. \tag{3.16}$$

In the case of an external compressive force

$$\sigma_x = \sigma - \sigma'(x)\,. \tag{3.17}$$

When the substrate is fixed with a console, the relative deformation of the film (hence, the mechanical stress induced by an external bending force) is not uniform along the entire film length and depends on the distance x from the console fixation line (it is maximal near the line). Mean values for the strain ε and stress σ in a film ($x = 20$ mm) are presented below [180, 188].

Figure 15 shows the dependence of the noise PSD on mechanical stresses in the elastic strain regions of two Cr films. Points σ_1 and σ_2 on the abscissa axis indicate the internal macrostresses (in the absence of external strains) and the corresponding noise intensities are shown on the ordinate axis. It can be seen that the film with larger internal stresses has a higher $1/f$-noise level. Increasing the tensile stress to $\sigma \leqslant 8 \times 10^8$ Pa causes a reversible rise in the magnitude of $1/f$ noise at $f < 10$ kHz. Film resistance in this strain region also undergoes a reversible growth by 0.5 – 1%. These dependences occur in the elastic strain region. An increase in tensile stress from $\sigma_x = \sigma = 10^8$ to $\sigma_x = 3 \times 10^8$ Pa leads to a rise in frequency exponent γ from 1 to 2.5 [180].

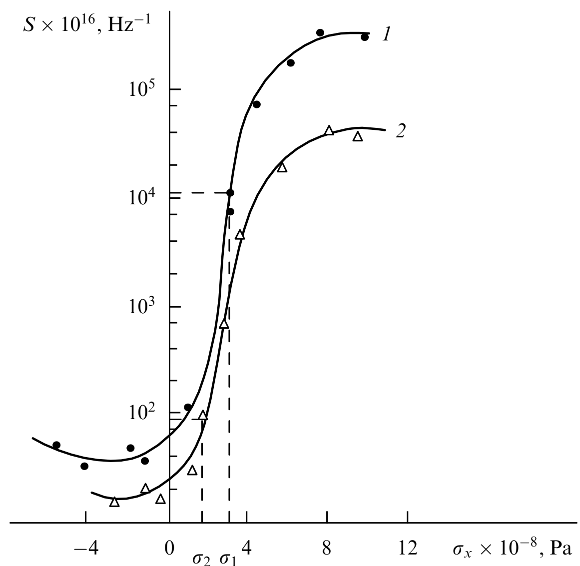


Figure 15. $1/f$-noise PSD at 300 Hz vs. externally induced mechanical stresses in Cr films deposited in an atmosphere of argon ($h = 80$ nm) [180]: (*1*) at argon pressure $P_{\mathrm{Ar}} = 4 \times 10^{-3}$ Pa; (*2*) at $P_{\mathrm{Ar}} = 8 \times 10^{-3}$ Pa (σ_1, σ_2 are the internal macrostresses).

The application of external compressive forces abates the noise which becomes lowest and approximately equal in all films which originally differed in its initial noise level by two or three orders of magnitude and had different mechanical stresses.

Let us discuss possible causes of the stronger $1/f$ noise in the case of elastic strain. The interatomic distance increases as the tensile stresses grow, leading to a decrease in the activation energy of vacancy formation and a rise in their

concentration in accordance with (3.2). Essentially all atoms instantly return to their original positions upon removal of the force that induces elastic strain; in so doing the vacancy formation energy and $1/f$-noise magnitude take the initial values.

In Cr films with large relative deformations ($\varepsilon \geqslant 0.4\%$), the noise level (Fig. 16) and resistance undergo irreversible changes [188], which give evidence of their plastic nature (it should be borne in mind that structural features of the films ensure a large elastic strain resource of up to $\varepsilon \simeq 0.5-1\%$ [190]). In this case, the inverse strain dependences of noise (see Fig. 16) and resistance do not coincide with the direct ones. At sufficiently high frequencies ($f > 1$ kHz), the noise PSD decreases during the relaxation time of 10 – 50 min [92]. Similar behaviour of flicker noise upon deformation of gold and platinum films was observed in Ref. [187].

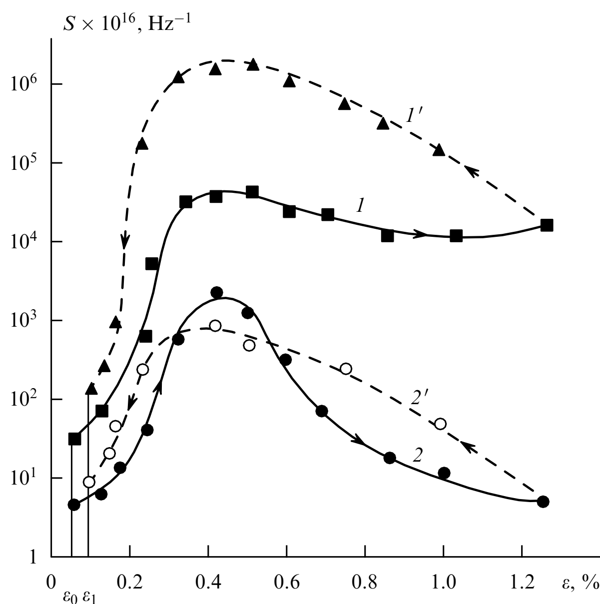


Figure 16. Strain effect on $1/f$-noise PSD in a Cr film ($h = 80$ nm) [188]: (*1*, *1'*) $f = 300$ Hz, (*2*, *2'*) $f = 2$ kHz (ε_0 is the initial strain due to internal macrostresses; ε_1 is the residual strain after unloading).

It is known [191 – 193] that plastic strain is associated with changes in the dislocation structure of a crystalline sample. The movements of dislocations by sliding and crawling are induced by mechanical stresses. On crawling dislocations absorb vacancies. This process reduces the number of nonequilibrium vacancies and their aggregates in the crystal [193, 194], which accounts for a lower noise PSD at strains $\varepsilon \geqslant 0.5\%$ (see Fig. 16).

Partial removal of the external force from the film results in a rise in vacancy concentration due to their emission by dislocations [194]. This increases the flicker-noise level at low frequencies (curves *1'* and *2'* in Fig. 16). In a completely unloaded film, the $1/f$-noise level is higher than initially, similar to what was observed in Ref. [187]. It is due to the appearance of additional microdefects. In this case, the film contains a residual strain ε_1 (see Fig. 16).

In experiments studying the effect of uniform tensile stress on $1/f$ noise in Al films on the elastic substrate [189], the stress was raised by applying a controlled tensile force to the free end of the film (aluminium was deposited at $T_{\text{cond}} = 400$ K on a thoroughly cleaned and previously heated substrate). Both the $1/f$-noise PSD and γ increased with increasing tensile stress (Fig. 17). Analysis of the dependence of the excess noise PSD $\Delta S = S - S_0$ (where S_0 and S are the noise PSD values before and after deformation, respectively) on the tensile stress [189] reveals its exponential character obeying the law (3.15) with $V_{\text{a}} \simeq 5\Omega_{\text{Al}}$, coinciding in magnitude with the activation volume derived in Ref. [106] from the dependence of the noise PSD on internal mechanical stresses.

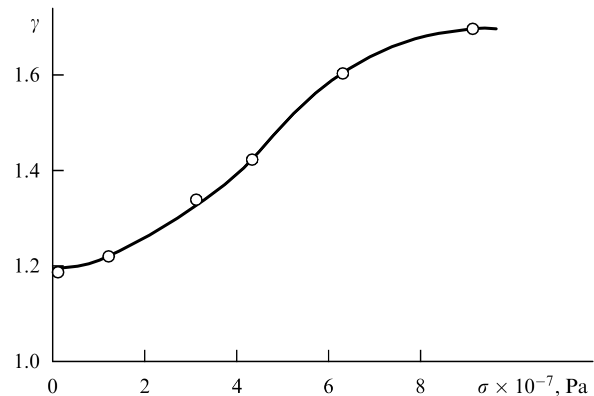


Figure 17. Frequency exponent vs. tensile stress caused by an external force in an Al film [189]; $h = 70$ nm.

3.3.6 Effect of structural factors on the $1/f$-noise level. Eberhard and Horn [34] reported a 2 – 6-fold decrease in the $1/f$-noise level after high-temperature annealing of Ag films, which resulted in an approximately 4-fold increase in the mean grain size. The effect of microstructural factors on $1/f$ noise was confirmed by a fall of the noise magnitude in aging platinum films. In selected samples, the noise abated by one or two orders of magnitude within a few days [37, 187]. In bismuth [181] and tin [101] films, a change in the noise magnitude took around 2 weeks. Fleetwood and Giordano [36, 187] observed variations of the $1/f$-noise level in minimally identical film samples, which exceeded one order of magnitude. Ref. [33] also reported a 3-fold difference of $1/f$ noise in identical films attributable to their specific microstructural features. At the same time, the noise level in gold films showed weak dependence on the microstructure suggesting that the effect of impurities is more pronounced than that of structural defects [195].

The effect of the microstructure on $1/f$ noise was examined in depth in Al, Cr, and Mo films [106, 156, 105, 172]. All these metals showed increasing noise with decreasing mean grain size.

Figure 18 presents results of a few studies designed to evaluate the effect of structure dispersion on the $1/f$-noise level in Al films [106, 156]. An increase in the mean grain size reduced the noise magnitude; the parameter α in samples with $d_{\text{av}} \simeq 200$ nm was close to $\alpha_{\text{H}} = 2 \times 10^{-3}$ (dashed line). A decrease in the flicker-noise level with increasing grain size was observed in Ag films [144], where an α value close to $\alpha_{\text{H}} = 2 \times 10^{-3}$ was achieved at the grain size $d_{\text{av}} \simeq 200$ nm.

An increase in the $1/f$-noise magnitude with decreasing grain size suggests the appearance of $1/f$ noise due to creation and annihilation of vacancies at grain boundaries.

It has been shown [105] that the $1/f$-noise level depends on the vacancy concentration inside grains (or the number of defective cells with respect to the total number of unit cells in the crystal). For example, in Cr films with an approximately

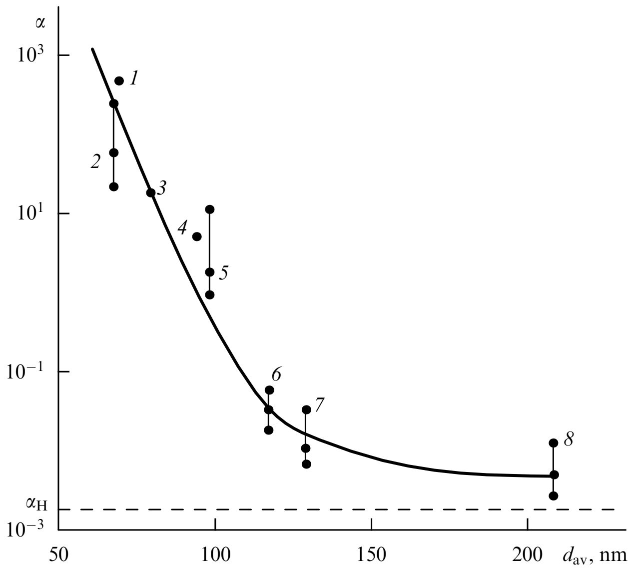


Figure 18. Relationship between the $1/f$-noise level and the mean grain size in Al films: series *1–4* were prepared at different condensation rates [106]; series *5–8*, at different argon pressures in the chamber [156]; line segments connect α values for different samples of the same set. The dashed line corresponds to the Hooge constant $\alpha_H = 2 \times 10^{-3}$.

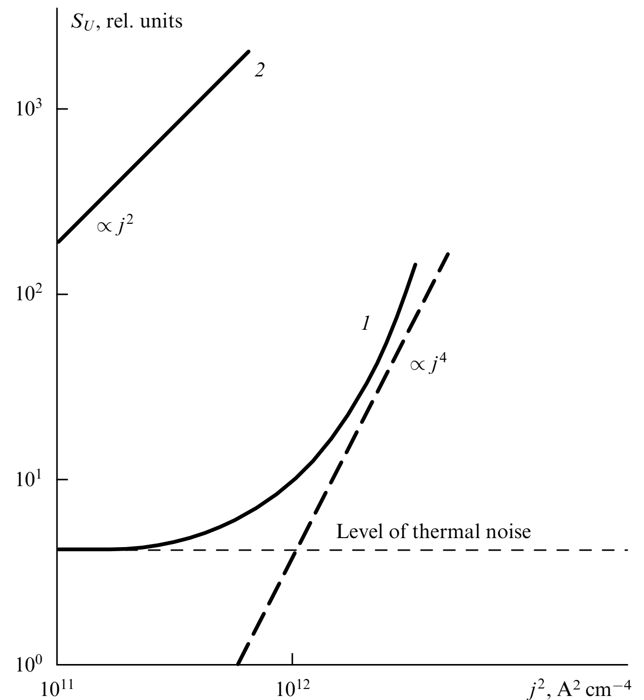


Figure 19. Noise PSD of a Cr film deposited in an atmosphere of nitrogen vs. current density squared (curve *1*) and the results of a computation using the Hooge formula (2.1) (curve *2*) [91].

similar grain size ($d_{av} \simeq 30-40$ nm) and differing degrees of crystallization, the difference in the noise levels amounted to three orders of magnitude. The degree of crystallization was qualitatively assessed from spreading and weakening of interference lines on roentgenograms and electronograms [105].

The Cr and Al films with inhomogeneous crystalline structures containing a fine-dispersed phase along with large grains exhibited strong flicker noise [106, 105].

It has already been mentioned that films deposited on one substrate by the same technique or even in the same technological cycle sometimes have different levels of $1/f$ noise. The microstructural examination of such samples revealed impurities at the film surface or various surface defects introduced by the substrate. These defects and admixtures were responsible for a 5–10-fold rise in the noise PSD [105] probably because they served as additional vacancy sources at contamination sites.

3.3.7 Flicker noise in metal films with elevated concentration of stable defects. Low $1/f$ noise corresponding to $\alpha_H = 2 \times 10^{-3}$ occurs in high-quality films with a low concentration of mobile defects [143, 183]. The resistivity of such films is similar to that of bulky metals. At the same time, metal films with a high concentration of stable defects and a low level of mobile ones also exhibit low $1/f$ noise. Specifically, the noise magnitude of $\alpha \simeq 10^{-4}-10^{-5}$ was recorded in Cr films obtained by thermal evaporation in an atmosphere of nitrogen [91]. These samples had a high concentration of stable defects, and their resistivity at TCR $\beta \simeq 5 \times 10^{-4}$ K^{-1} was $\rho \simeq 10\rho_0$ (ρ_0 is the specific resistance of bulky chromium).

Figure 19 shows the dependence of the $1/f$-noise PSD for such a film on the current density squared (curve *1*) and the dependence for $\alpha_H = 10^{-3}$ calculated by the formula (2.1) on the assumption that $n_c = 10^{22}$ cm^{-3} (curve *2*). The $1/f$ noise at a current density $j < 5 \times 10^5$ A cm^{-2} is slightly stronger than the thermal noise, whereas at $j > 10^6$ A cm^{-2} nonequilibrium resistance fluctuations predominate and the PSD of $1/f$ noise grows in accordance with the $S \propto j^4$ law. Such a dependence arises from fluctuations of the coefficient R_1 in the expansion of (2.6), probably due to local overheating of the film by Joule heat resulting from the nonuniform distribution of stable defects [91]. The domains of local overheating give rise to mobile defects and are therefore responsible for elevated $1/f$-noise level [91].

Stable defects cause a decrease in the relative SDF of film resistance. Formula (3.9) predicts a fall in the PSD of $1/f$ noise induced by fluctuations in the equilibrium vacancy concentration with the growth of resistivity ρ_f in the film due to additional scattering from stable defects.

Films of the Al/Si alloy AK-1 (1% Si) also have a lower noise magnitude than films of pure Al [91] obtained under identical technological regimes. Dimensionless parameters α for Al and the alloy are $\alpha \simeq 10^{-2}$ and $\alpha \simeq 10^{-3}$, respectively. The low noise level in the alloy films AK-1 compared with that in Al films can be accounted for by a lower concentration of vacancies at the grain boundaries, which are substituted by Si atoms. Taken together, this result and some others discussed earlier in this paper indicate that grain boundaries are the principal sources of vacancies in metal films.

4. Nonlinear effects in thin metal films and their relationship with flicker fluctuations

4.1 General remarks

A sinusoidal current $I_1 \sin(\omega_1 t)$ flowing through a film induces a signal-response of voltage for the third, $U_3 \sin(3\omega_1 t)$, or second, $U_2 \sin(2\omega_1 t)$, harmonics by virtue of cubic or quadratic nonlinearity of CVC. The degree of

CVC nonlinearity is characterized by nonlinearity coefficients defined as the ratio of the third U_3 (or second) harmonic amplitude to the amplitude of the signal of the fundamental frequency $U_1 = RI_1$:

$$K_{\mathrm{NL3}} = \frac{U_3}{U_1}\,, \tag{4.1}$$

or in decibels [196]:

$$K_{\mathrm{NL3}} = 20\log\frac{U_3}{U_1}\,. \tag{4.2}$$

Sometimes, the nonlinearity coefficient is presented as

$$K_{\mathrm{NL3}} = \frac{U_3}{I_1^3}\,. \tag{4.3}$$

In such a presentation, K_{NL3} does not depend on the signal amplitude I_1 [175], which allows samples for which the harmonics U_3 were measured at different currents to be compared in terms of the degree of nonlinearity.

Some authors agree that the dependences of the third harmonic amplitude U_3 and flicker noise magnitude in metal films on various technological factors (thickness, argon pressure in a vacuum chamber during condensation, bias voltage across the substrate during film production by the ion sputtering technique, etc.) are very similar. The minimum of U_3 in a Cr film [197], close to the minimum of $1/f$-noise PSD in Fig. 5, was recorded at a film thickness of 100 nm. Chromium films fabricated in an atmosphere of argon showed minima of $1/f$-noise PSD and nonlinearity coefficient at an argon pressure 10^{-2} Pa in the chamber [180, 198]. These findings suggest a similar physical nature of the mechanisms responsible for $1/f$ noise and cubic nonlinearity.

4.2 Mechanisms of cubic nonlinearity in metal films

Refs [143, 183] report the computation of the third harmonic amplitude taking into account the scattering of carriers from phonons and quasi-equilibrium vacancies in the zero-memory approximation. The effects of inertia were discussed in Ref. [175]. Taking these into consideration leads only to a change in the voltage phase and amplitude of the third harmonic which is unessential for the elucidation of the nonlinearity mechanism. Therefore, only the results of Refs [143, 183] are reviewed below.

When a sinusoidal signal $I_1\sin(\omega_1 t)$ is carried to a film, the excess of film temperature ΔT over the equilibrium temperature T_0 in the zero-memory approximation is defined through the thermal resistance R_T and dissipated power of the signal P:

$$\Delta T = T - T_0 = R_T P = \frac{R_T K_1 \rho_{\mathrm{f}}}{h}\, I_1^2 \sin^2(\omega_1 t)\,, \tag{4.4}$$

where T is the film temperature taking into account current heating by the signal of the fundamental frequency ($\Delta T \ll T_0$); $K_1 = L/b$ is the form factor; L and b are the length and the width of the film, respectively; h is the thickness, and ρ_{f} is the resistivity defined by expression (3.12) (it is assumed that heat relaxation time of the film–substrate system $\tau_{RT} \ll \omega_1^{-1}$, which is fulfilled for the system examined in the above papers at a signal frequency $f_1 \leqslant 30$ kHz).

A change in film temperature induced by a sinusoidal signal in accordance with (4.4) leads to periodic changes of the vacancy concentration in the film according to (3.2). Assuming that only vacancies with the activation energy E_{v} contribute to the part of the specific resistance related to mobile defects, the temperature dependence of ρ_{f} in view of (3.1) and (3.12) can be written as

$$\rho_{\mathrm{f}}(T) = \rho_{01}(1 + \beta\Delta T) + AA_{\mathrm{v}}\exp\left[-\frac{E_{\mathrm{v}}}{k(T_0 + \Delta T)}\right]. \tag{4.5}$$

Here, ρ_{01} is the temperature-independent part of the film resistivity due to scattering from phonons at T_0, stable defects, and the film surface boundaries; β is the film TCR. The first and second items in (4.5) define the contribution of scattering from phonons and vacancies to the film resistance, respectively; E_{v} is the lowest activation energy of mobile defects.

The following expression holds true for a drop of voltage across the film:

$$U = \rho_{\mathrm{f}} K I_1 \sin(\omega_1 t)\,, \tag{4.6}$$

where $K = L/bh$.

Substitution of (4.5) and (4.4) into (4.6) and expansion first in powers of ΔT with allowance made for $E_{\mathrm{v}}\Delta T/kT_0^2 \ll 1$ ($E_{\mathrm{v}}\Delta T/kT_0^2 = 0.06$ at $E_{\mathrm{v}} = 0.5$ eV, $\Delta T = 1$ K) and then in Fourier series yields the expression for the third harmonic

$$U_3 = 0.25 R_T K^2 \rho_{01}\rho_{\mathrm{f}}\left[\beta + \frac{AA_{\mathrm{v}}E_{\mathrm{v}}}{kT_0^2\rho_{01}}\exp\left(-\frac{E_{\mathrm{v}}}{kT_0}\right)\right] I_1^3\,. \tag{4.7}$$

The first term in (4.7) is proportional to the film TCR and is related to the mechanism of carrier scattering by phonons, and the second term to the process of vacancy activation by the signal. The first term in (4.7) may be neglected when the vacancy concentration in the film is sufficiently high. In such a case, the harmonic U_3 is proportional to the quasi-equilibrium vacancy concentration which accounts for the exponential dependence of U_3 on the temperature and mechanical stresses, since $E_{\mathrm{v}} = u_{\mathrm{v}} - \sigma V_{\mathrm{v}}$:

$$U_3 = 0.25 R_T K^2 \rho_{\mathrm{f}}\,\frac{AA_{\mathrm{v}}E_{\mathrm{v}}}{kT_0^2}\, I_1^3 \exp\left(-\frac{u_{\mathrm{v}} - \sigma V_{\mathrm{v}}}{kT_0}\right). \tag{4.8}$$

4.3 Experimental studies of CVC cubic nonlinearity in metal films

Temperature and mechanical stress dependences of CVC nonlinearity coefficient have been investigated in Mo and Ta films affected by a weak sinusoidal signal of not more than 10 μW at 10 kHz [172].

Films with enhanced $1/f$-noise level showed the activation-law dependence of the third harmonic amplitude U_3 on the temperature [172, 175]. The activation energies derived from the temperature dependences of nonlinearity coefficients (4.2) for Mo and Ta films were $E_{\mathrm{a}} = 0.3 - 0.35$ eV and $E_{\mathrm{a}} = 0.4 - 0.45$ eV, respectively (Fig. 20) [172] and coincided with the activation energies for $1/f$-noise PSD at frequencies $f > 10^3 - 10^4$ Hz (see Figs 11 and 12).

Figure 21 displays the experimentally found dependence of U_3/I_1^3 on the logarithm of mechanical stress measured in the same film samples, which were used to study the noise PSD dependence shown in Fig. 13 [183, 143]. The activation volumes obtained from these plots were $(1.3 \pm 0.4) \times 10^{-29}$ and $(1.1 \pm 0.3) \times 10^{-29}$ m^3 for Cr and Mo, respectively. These

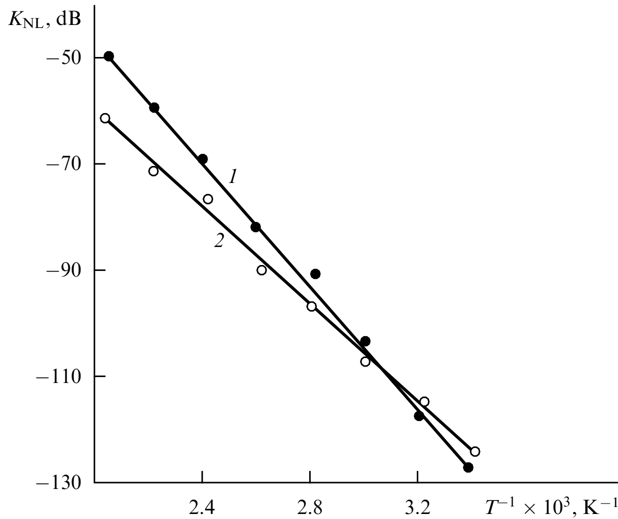


Figure 20. CVC nonlinearity coefficient in Ta (*1*) and Mo (*2*) films vs. inverse temperature [172]; $h = 0.5$ μm, $f_1 = 10$ kHz, $U_1 = 250$ mV.

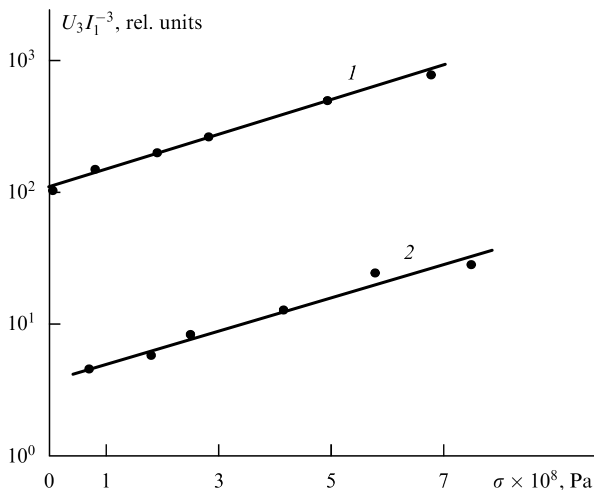


Figure 21. CVC nonlinearity coefficient vs. internal mechanical stresses in Cr (*1*) and Mo (*2*) films [143, 183].

values coincide with activation volumes found from the dependences $S(\sigma)$ (see Fig. 13), which confirms the vacancy mechanism of cubic CVC nonlinearity and $1/f$ noise in films with an elevated concentration of mobile defects.

It is worthwhile to note that the creation and annihilation of vacancies associated with the generation of quasi-equilibrium $1/f$ noise occur at a sacrifice in the internal energy of the crystal. The process of vacancy creation and annihilation obeys therewith statistical laws and involves defects with a variety of relaxation times (activation energies), the defects with the lowest activation energy contributing to the formation of the $1/f$-noise spectrum at higher frequencies.

Generation of the third harmonic of voltage is associated with the activation of microdefects due to Joule heating of the film by a weak signal of the fundamental frequency. The signal with frequency f_1 changes the film temperature in accordance with the law $\sin^2(\omega_1 t)$, which results in an altered vacancy concentration in conformity with (3.2) and the occurrence of nonlinear effects. In the first place, defects with the lowest activation energy are generated, which accounts for the coincidence of activation energies for the third harmonic of the signal-response and $1/f$-noise PSD at high frequencies ($f > 10^3 - 10^4$ Hz).

For films with a low vacancy concentration, the second term in (4.7) may be neglected and the amplitude of the third harmonic is defined as

$$U_3 = 0.25 R_T K^2 \rho_0 \rho_f \beta I_1^3 \,. \tag{4.9}$$

Here, the temperature dependence of U_3 is determined by the weak dependence of $\rho_f(T)$. Such films are characterized by a low amplitude of the third harmonic and its linear temperature dependence [175]. $1/f$ noise in these films is also weak [143, 156, 183].

Molybdenum films with an elevated content of reactive gas contaminants have a higher U_3 level than can be expected from (4.7) [175] due to the manifestation of nonmetallic conduction mechanisms, which make an additional contribution to the third harmonic amplitude and induce second harmonic along with nonequilibrium flicker noise [89, 90].

The third harmonic in Al films was observed by Jones et al. [199, 200] by reacting the film with the combined sinusoidal signal and a direct current of density 10^6 A cm^{-2} at 260 °C.

4.4 Some data on CVC quadratic nonlinearity in metal films

It follows from the calculations in Section 4.2 that carrier scattering from phonons and equilibrium vacancies gives rise to cubic CVC nonlinearity but fails to produce quadratic nonlinearity. At the same time, Jones et al. [199, 200] reported the appearance of the second harmonic, besides the third one, in the signal-response from Al films subjected to an enhanced current and temperature exposures. The mechanism of generation of the second harmonic was not clarified by these authors, but their experimental data suggest a close relationship between the electromigration film damage and the harmonic assembly.

The quadratic CVC nonlinearity was also observed in Mo films prepared by magneton sputtering at a relatively low effective density of sinusoidal current through the sample ($j < 0.5 \times 10^4$ A cm^{-2}) [89, 90]. It was demonstrated in Ref. [90] that the conduction mechanism related to overbarrier emission could be one of the causes of quadratic nonlinearity in metal films even if it accounts for a minor part of the total (metallic) conduction of the film. In metals showing a high affinity to oxygen, such an electric conductivity mechanism is feasible because thin (~ 1 nm) oxide interlayers are likely to develop at boundaries of isolated grains due to the capture of oxygen molecules by the film in the course of condensation.

5. $1/f^2$ noise

Recently, a large number of studies have been devoted to the relationship between the electromigration immunity of thin metal films and the level of $1/f$ noise [17–19, 93]. In such experiments, $1/f$ noise is measured under high current and thermal exposures, and the concomitant $1/f^\gamma$ noise has $\gamma \simeq 2$.

The method of forecasting the stability of thin metal films experiencing electromigration damage, based on the evaluation of $1/f$ noise, was first suggested in Ref. [201]. Its validity was confirmed in experiments demonstrating that Al films with enhanced excess noise are characterized by smaller median time to failure [202]. Thereafter, Neri et al. [20]

estimated the temperature dependence of $1/f^2$-noise PSD in Al films. Starting from these data they determined the activation energy which turned out to correspond to that of vacancy diffusion along the grain boundaries (around 0.6 eV) and correlate with the values obtained by other methods.

At a current density of $j \sim 10^6 - 10^7$ A cm^{-2}, the current dependence of noise PSD differed from quadratic, and $S \propto j^n$, where $n = 3, 4$ and even 7. Also, the frequency exponent γ increased with increasing current [20, 21, 93, 203–210]. For example, n in Nichrome films grew from $n \simeq 2$ at weak currents to 3.5–4.5 at larger ones, while the frequency exponent γ rose from 1 to 2.5 [21]. The $1/f^2$-noise component is sometimes referred to as excess electromigration noise. Vrachev [211] was the first to suggest the causative role of electromigration in the development of low-frequency noise in metal films.

The deviation of the current dependence of the electromigration noise PSD from quadratic indicates that this component of flicker noise originates from nonequilibrium fluctuations of conduction and is related to fluctuations of the coefficient of the nonlinear terms of CVC in expression (2.6) [89, 90]. The nonequilibrium $1/f^2$ component of noise appears at high current and thermal exposures, each component of equilibrium $1/f$ noise and nonequilibrium $1/f^2$ noise playing different roles in the film samples [208]. If $1/f$ noise induced by fluctuations in the number of quasi-equilibrium vacancies in a sample (annealed and undeformed) is stationary, then the electromigration noise (associated with electromigration leading to structural changes in the film) must be nonstationary.

Some studies have demonstrated that the electromigration component of $1/f^2$ noise arises from the diffusive motion of atoms along the grain boundaries [20, 93]. Indeed, the activation energy determined using the temperature dependence of the noise PSD corresponds to the activation energy of diffusion along the grain boundaries. It was confirmed by the experiments of Koch et al. [212] in which the activation energy of $1/f^2$ noise in films of Al and its alloys increased in parallel to a rise in the activation energy of diffusion along the grain boundaries with the growing content of impurities. Since impurities are segregated at the grain boundaries, they do not appreciably influence the bulk properties of crystallites [213, 214].

In submicron metal conductors with bamboo structure, the activation energy determined from the temperature dependence of the $1/f^2$-noise PSD proved to be equal to the activation energy of diffusion across a crystal lattice (1.45 eV) [210]. This finding indicates that in this case $1/f^2$ noise is associated with the diffusion of atoms in the lattice [213, 214].

Measurements with direct current normally reveal a higher PSD of $1/f^2$ noise compared with the PSD of equilibrium $1/f$ noise (for which $\gamma \simeq 1$) only at very low frequencies (a few mHz to 1 Hz). This hampers its study. Therefore, investigations of nonequilibrium electromigration noise may take advantage of the method of measuring amplitude fluctuation spectra of signal-response harmonics. This signal is generated in response to the action of a sinusoidal current on the sample and allows nonequilibrium $1/f^2$ noise to be separated from the total noise [89, 90].

6. Conclusions

It has been shown in many experiments that equilibrium $1/f$ noise in metal films is generated by fluctuations in current carrier mobility. Two mechanisms of equilibrium $1/f$ noise are hypothesized: one related to scattering from phonons, and the other to scattering by quasi-equilibrium vacancies. The latter mechanism provides a broad range of relaxation times necessary to explain the $1/f$ spectrum, whereas in the former case the mechanism responsible for a similarly wide range of relaxation times remains to be elucidated despite a few arguments advanced by Hooge [32] in an attempt to obtain a pertinent explanation.

The vacancy model of $1/f$ noise has been confirmed by numerous experiments on the annealing and aging of films, the effects of mechanical stresses and strains, temperature, and structural features on noise level. Metal films with homogeneous structures and a low concentration of mobile defects exhibit low $1/f$ noise described by the Hooge formula at $\alpha_H \simeq 2 \times 10^{-3}$. Films with an elevated concentration of stable defects and a low level of mobile ones also have low $1/f$-noise level (parameter $\alpha \simeq 10^{-5} - 10^{-3}$). An important argument in support of the vacancy mechanism of $1/f$ noise in metal films is the relationship between the $1/f$-noise and cubic CVC nonlinearity, which was both observed in experiments and calculated. The mechanism of electron scattering from quasi-equilibrium vacancies is responsible for cubic CVC nonlinearity and equilibrium $1/f$ noise. The vacancy mechanism of $1/f$ noise accounts for many experimental findings concerning $1/f$ noise in metals. The known dependences of the $1/f$-noise PSD on various factors constitute the scientific basis for manufacturing metal films with low $1/f$-noise levels.

For all that, a more comprehensive mathematical analysis of the vacancy model is needed to account for the dependence of the frequency exponent γ on the internal mechanical stresses, temperature, and other factors.

Also, further studies are necessary to better understand nonequilibrium flicker noise, different varieties of which, along with equilibrium noise, can be observed in conducting films: (a) in metal films containing contaminants of reactive gases [89, 90]; (b) in films with a high concentration of stable defects at elevated currents [91], and (c) in films subjected to large current and temperature exposures, which give rise to electro-mass transfer [203–210]. Studies on the statistical properties of electromigration $1/f^2$ noise are of special interest for the development of new methods for predicting the electromigration immunity of conducting films. Equally important is the in-depth investigation of nonequilibrium temperature fluctuations arising in overheated films [91, 128].

References

1. Johnson J B *Phys. Rev.* **26** 71 (1925)
2. Schottky W *Phys. Rev.* **28** 74 (1926)
3. Van der Ziel A *Fluctuation Phenomena in Semi-Conductors* (London: Butterworths, 1959) [Translated into Russian (Moscow: Inostrannaya Literatura, 1961)]
4. Naryshkin A K, Vrachev A S *Teoriya Nizkochastotnykh Shumov* (The Theory of Low-Frequency Noises) (Moscow: Energiya, 1972)
5. Van der Ziel A *Sources, Characterization, Measurement* (Englewood Cliffs: Prentice-Hall, 1970) [Translated into Russian (Moscow: Sovetskoe Radio, 1973)]
6. Van der Ziel A *Adv. Electron. Phys.* **49** 225 (1979)
7. Van der Ziel A *Proc. IEEE* **76** (4) 28 (1988)
8. Buckingham M *Noise in Electronic Devices and Systems* (Chichester: Ellis Horwood, 1983) [Translated into Russian (Moscow: Mir, 1986)]
9. Luk'yanchikova N B *Fluktuatsionnye Yavleniya v Poluprovodnikakh i Poluprovodnikovykh Priborakh* (Fluctuation Phenomena in Semiconductors and Devices Containing Semiconductors) (Moscow: Radio i Svyaz', 1990)

10. Kogan Sh M *Usp. Fiz. Nauk* **145** 285 (1985) [*Sov. Phys. Usp.* **28** 170 (1985)]
11. Hooge F N, Kleinpenning T G M, Vandamme L K J *Rep. Prog. Phys.* **44** 479 (1981)
12. Dutta P, Horn P M *Rev. Mod. Phys.* **53** (3) 497 (1981)
13. Weissman M B *Rev. Mod. Phys.* **60** 537 (1988)
14. Bochkov G N, Kuzovlev Yu E *Usp. Fiz. Nauk* **141** 151 (1983) [*Sov. Phys. Usp.* **26** 829 (1983)]
15. Pryanikov V S *Prognozirovanie Otkazov Poluprovodnikovykh Priborov* (Forecasting of Failures in Semiconductor Devices) (Moscow: Energiya, 1978)
16. Luk'yanchikova N B *Elektronnaya Prom.* (6) 28 (1983)
17. Celik-Butler Z, in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 200
18. Sikula J, Touboul A, in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 206
19. Jones B K *Adv. Electron. Electr. Phys.* **87** 201 (1994)
20. Neri B, Diligenti A, Bagnoli P E *IEEE Trans. Electron. Dev.* **ED-34** 2317 (1987)
21. Touboul A, Verdier F, Herve Y, in *Proc. Int. Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Kyoto, Japan, 1991) p. 73
22. Hooge F N, Hoppenbrouwers A M H *Physica* **45** 386 (1969)
23. Handel P H *Phys. Rev. Lett.* **34** 1492 (1975)
24. Handel P H *Phys. Rev. Lett.* **34** 1495 (1975)
25. Handel P H *Phys. Rev. A* **22** 745 (1980)
26. Handel P H, in *Noise in Physical Systems and 1/f Noise* (Eds A d'Amico, P Mazzetti) (New York: Elsevier, 1986) p. 465
27. Van Viet K M *Solid-State Electron.* **34** 1 (1991)
28. Bychikhin S, Gertsenstein M, Potemkin V, in *Proc. 13 Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Eds V Bareikis, R Katilius) (Singapore: World Scientific, 1995) p. 269
29. Black R D, Snow W M, Weissman M B *Phys. Rev. B* **25** 2955 (1982)
30. McWhorter A L, in *Semiconductor Surface Physics* (Ed. R H Kingston) (University of Pennsylvania Press, 1957) p. 207 [Translated into Russian (Moscow: Inostrannaya Literatura, 1959) p. 263]
31. Hooge F N *Phys. Lett. A* **29** 139 (1969)
32. Hooge F N *Physica B* **162** 343 (1990)
33. Voss R F, Clarke J *Phys. Rev. B* **13** 556 (1976)
34. Eberhard J W, Horn P M *Phys. Rev. B* **18** 6681 (1978)
35. Fleetwood D M, Giordano N *Phys. Rev. B* **25** 1427 (1982)
36. Fleetwood D M, Giordano N *Phys. Rev. B* **27** 667 (1983)
37. Fleetwood D M, Mosden J T, Giordano N *Phys. Rev. Lett.* **50** 450 (1983)
38. Hooge F N *Physica* (Netherlands) **60** 130 (1972)
39. Hooge F N *Physica B* **83** 14 (1976)
40. Kleinpenning T G M *Physica B* **113** 189 (1982)
41. Bosman G, Zijlstra R J J, Van Rheenen A *Phys. Lett. A* **80** 57 (1980)
42. Weissman M B *J. Appl. Phys.* **51** 5872 (1980)
43. Black R D, Restle P J, Weissman M B *Phys. Rev. Lett.* **51** 1476 (1983)
44. Black R D, Weissman M B, Restle P J *J. Appl. Phys.* **53** 6280 (1982)
45. Weissman M B *Physica B* **100** 157 (1980)
46. Hooge F N, Vandamme L K J *Phys. Lett. A* **66** 315 (1978)
47. Palenskis V, Shoblitskas Z *Solid State Commun.* **43** 761 (1982)
48. Hooge F N, Kedzia J, Vandamme L K J *J. Appl. Phys.* **50** (12) 8087 (1979)
49. Jevtic M M *Solid-State Electron.* **31** 1049 (1988)
50. Musha T *Phys. Rev. B* **26** 1042 (1982)
51. Dutta P, Eberhard J W, Horn P M *Solid State Commun.* **21** 679 (1977)
52. Leemann C, Skove M J, Stillwell E P *Solid State Commun.* **35** 97 (1980)
53. Voss R F, Clarke J *Phys. Rev. Lett.* **36** 42 (1976)
54. Clarke J, Voss R F *Phys. Rev. Lett.* **33** 24 (1974)
55. Scofield J H, Mantese J V, Webb W W *Phys. Rev. B* **32** 736 (1985)
56. Scofield J H, Webb W W *Phys. Rev. Lett.* **54** 353 (1985)
57. Kittel Ch *Introduction to Solid State Physics* (New York: Wiley, 1967) [Translated into Russian (Moscow: Nauka, 1978)]
58. Palenskis V P, Shoblitskas Z P, Mikolaĭtis G S, in *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 4 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 4th All-Union Conf.) (Pushchino, Moscow Region, 1985) p. 102
59. Vandamme L K J, Kedzia J *Thin Solid Films* **65** 283 (1980)
60. Hooge F N, in *Proc. Int. Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Kyoto, Japan, 1991) p. 7
61. Kleinpenning T G M *Physica B* **98** 289 (1980)
62. Hooge F N, Hoppenbrouwers A M H *Phys. Lett. A* **29** 642 (1969)
63. Hoppenbrouwers A M H, Hooge F N *Philips Res. Rep.* **25** 69 (1970)
64. Tandon J L, Bilger H R *J. Appl. Phys.* **47** 1697 (1976)
65. Rollin B V, Templeton I M *Proc. Phys. Soc. B* **66** Part 3 259 (1953)
66. Firle T E, Winston H *J. Appl. Phys.* **26** 716 (1955)
67. Caloyannides M A *J. Appl. Phys.* **45** 307 (1974)
68. Van Viet K M et al. *Physica* **20** 481 (1954)
69. Malakhov A N *Radiotekh. Elektron.* **4** (1) 54 (1959)
70. Brophy J J *Phys. Rev.* **166** 827 (1968)
71. Malakhov A N, Yakimov A V *Radiotekh. Elektron.* **19** 2436 (1974) [*Radio Eng. Electron. Phys.* **19** 137 (1974)]
72. Yakimov A V *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **23** 238 (1980) [*Radiophys. Quantum Electron.* **23** 170 (1980)]
73. Brophy J J *J. Appl. Phys.* **40** 3551 (1969)
74. Brophy J J *J. Appl. Phys.* **41** 2913 (1970)
75. Purcell W E *J. Appl. Phys.* **43** 2890 (1972)
76. Dell R A, Epstein M, Kannewurf C R *J. Appl. Phys.* **44** 472 (1973)
77. Moore W J *J. Appl. Phys.* **45** 1896 (1974)
78. Stoisiek M, Wolf D *J. Appl. Phys.* **47** 362 (1976)
79. Potemkin V V, Stepanov A V *Radiotekh. Elektron.* **25** 1269 (1980)
80. Potemkin V V, Stepanov A V, in *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 2 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 2nd All-Union Conf.) (Vilnius, USSR, 1979) p. 66
81. Dutta P, Dimon P, Horn P M *Phys. Rev. Lett.* **43** 646 (1979)
82. Klimontovich Yu L *Statisticheskaya Fizika* (Moscow: Nauka, 1982) [Translated into English *Statistical Physics* (New York: Harwood Acad. Publ., 1986)]
83. Stratonovich R L *Izbrannye Voprosy Teorii Fluktuatsyĭ v Radiotekhnike* (Selected Topics of Fluctuation Theory in Radioengineering) (Moscow: Sovetskoe Radio, 1961)
84. Bell D A, Dissanayake S P B *Electron. Lett.* **11** 274 (1975)
85. Nelkin M, Tremblay A M S *J. Stat. Phys.* **25** 253 (1981)
86. Aleksandrov A A et al. *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **29** 980 (1986) [*Radiophys. Quantum Electron.* **29** (1986)]
87. Aleksandrov A A et al. *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **31** 507 (1988) [*Radiophys. Quantum Electron.* **31** (1988)]
88. Garfunkel G A, Weissman M B *Bull. Am. Phys. Soc.* **32** 480 (1987)
89. Zhigal'skii G P, in *Fluctuation Phenomena in Physical Systems* (Ed. V Palenskis) (Vilnius: University Press, 1994) p. 285
90. Zhigal'skiĭ G P *Zh. Fiz. Khim.* **69** 1355 (1995)
91. Potemkin V V, Stepanov A V, Zhigal'skii G P, in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 61
92. Zhigal'skii G P , in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 81
93. Dagge K et al., in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 603
94. Briggmann J et al., in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 607
95. Bull C S, Bozic S M *Brit. J. Appl. Phys.* **18** 883 (1967)
96. Lorteije J H J, Hoppenbrouwers A M H *Philips Res. Rep.* **26** 29 (1971)
97. Jones B K, Francis J D *J. Phys. D.: Appl. Phys.* **8** 1172 (1975)
98. Palenskis V P, Leont'ev G E, Mikolaĭtis G S *Radiotekh. Elektron.* **21** 2433 (1976)
99. Jones B K *Electron. Lett.* **12** 110 (1976)
100. Beck H G E, Spruit W P *J. Appl. Phys.* **49** 3384 (1978)
101. Dutta P, Eberhard J W, Horn P M *Solid State Commun.* **27** 1389 (1978)
102. Clarke J, Hsiang T Y *Phys. Rev. Lett.* **34** 1217 (1975)
103. Clarke J, Hsiang T Y *Phys. Rev. B* **13** 4790 (1976)
104. Weissman M B, Dollinger G D *J. Appl. Phys.* **52** 3095 (1981)
105. Zhigal'skiĭ G P et al. *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **33** 1181 (1990) [*Radiophys. Quantum Electron.* **33** (1990)]
106. Andrushko A F, Bakshi I S, Zhigal'skiĭ G P *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **24** 498 (1981) [*Radiophys. Quantum Electron.* **24** 343 (1981)]

107. Kurov G A, Zhigal'skiĭ G P *Fizika Plenok* Chast' 2 (Uchebnoe Posobie) [Physics of Films Part 2 (Text-Book)] (Moscow: MIET Publ., 1980)
108. Boltaks B I *Diffuziya v Poluprovodnikakh* (Diffusion in Semiconductors) (Moscow: Fizmatgiz, 1961)
109. Keshner M S *Proc. IEEE* **70** 212 (1982)
110. Orlov V B, Yakimov A V *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **33** 196 (1990) [*Radiophys. Quantum Electron.* **33** (1990)]
111. Van der Ziel A *Physica* **16** 359 (1950)
112. Du Pre F K *Phys. Rev.* **78** 615 (1950)
113. Kogan Sh M, Nagaev K E *Pis'ma Zh. Tekh. Fiz.* **10** 313 (1984) [*Sov. Tech. Phys. Lett.* **10** 132 (1984)]
114. Kogan Sh M, Nagaev K E *Solid State Commun.* **49** 387 (1984)
115. Yakimov A V *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **28** 1071 (1985)
116. Kozub V I, in *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 4 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 4th All-Union Conf.) (Pushchino, Moscow Region, 1985) p. 73
117. Vaĭnshteĭn L A *Dokl. Akad. Nauk SSSR* **285** 609 (1985) [*Sov. Phys. Dokl.* **30** 967 (1985)]
118. Halford D *Proc. IEEE* **56** 251 (1968)
119. Palenskis V et al., in *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 3 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 3rd All-Union Conf.) (Vilnius, USSR, 1983) p. 131
120. Radeka V *IEEE Trans. Nucl. Sci.* **NS-16** 17 (1969)
121. Landau L D, Lifshitz E M *Statisticheskaya Fizika* Chast' 2 (Moscow: Nauka, 1976) [Translated into English *Statistical Physics* Part 2 (Oxford: Pergamon Press, 1984)]
122. Zhigal'skiĭ G P, Karev A V *Radiotekh. Elektron.* **22** 2610 (1977) [*Radio Eng. Electron. Phys.* **22** 112 (1977)]
123. Kleinpenning T G M *Physica B* **84** 353 (1976)
124. Scofield J H, Darling D H, Webb W W *Phys. Rev. B* **24** 7450 (1981)
125. Black R D, Weissman M B, Fliegel F M *Phys. Rev. B* **24** 7454 (1981)
126. Yakimov A V *Zh. Tekh. Fiz.* **56** 1657 (1986) [*Sov. Phys. Tech. Phys.* **31** 987 (1986)]
127. Ketchen M B, Clarke J *Phys. Rev. B* **17** 114 (1978)
128. Zhigal'skiĭ G P, Fedorov A S *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **28** 1192 (1985) [*Radiophys. Quantum Electron.* **28** 831 (1985)]
129. Zhigal'skiĭ G P, Bakshi I S *Radiotekh. Elektron.* **25** 771 (1980) [*Radio Eng. Electron. Phys.* **25** 61 (1980)]
130. Sato H *J. Phys. Soc. Jpn.* **44** 49 (1978)
131. Sato H *Physica B* **97** 194 (1979)
132. Van Viet K M, Van der Ziel A, Schmidt R R *J. Appl. Phys.* **51** 2947 (1980)
133. Vaĭnshteĭn L A *Zh. Eksp. Teor. Fiz.* **83** 1841 (1982) [*Sov. Phys. JETP* **56** 1064 (1982)]
134. Van Viet K M, Mehta H *Phys. Status Solidi B* **106** 11 (1981)
135. Klimontovich Yu L *Zh. Eksp. Teor. Fiz.* **80** 2243 (1981) [*Sov. Phys. JETP* **53** 1171 (1981)]
136. Klimontovich Yu L *Pis'ma Zh. Tekh. Fiz.* **9** 406 (1983) [*Sov. Tech. Phys. Lett.* **9** 174 (1983)]
137. Lundström I, McQueen D, Klason C *Solid State Commun.* **13** 1941 (1973)
138. Klason C *Solid State Commun.* **13** 1945 (1973)
139. Zhigal'skiĭ G P, Fedorov A S *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **37** 161 (1994)
140. Yakimov A V *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **25** 308 (1982)
141. Masoero A, Mazzetti P, Stepanescu A *Vacuum* **27** 349 (1977)
142. Potemkin V V et al., in *Proc. Int. Conf. "Noise in Physical Systems and 1/f Fluctuations"* (Kyoto, Japan, 1991) p. 607
143. Zhigalskii G P, in *Proc. Int. Conf. "Noise in Physical Systems and 1/f Fluctuations"* (Kyoto, Japan, 1991) p. 39
144. Eberhard J W, Horn P M *Phys. Rev. Lett.* **39** 643 (1977)
145. Robinson F N H *Phys. Lett. A* **97** 162 (1983)
146. Pelz J, Clarke J *Phys. Rev. Lett.* **55** 738 (1985)
147. Pelz J, Clarke J *Phys. Rev. B* **36** 4479 (1987)
148. Fleetwood D M, Giordano N *Phys. Rev. B* **31** 1157 (1985)
149. Miller S C *Phys. Rev. B* **24** 3008 (1981)
150. Kogan Sh M, Nagaev K E *Fiz. Tverd. Tela* (Leningrad) **24** 3381 (1982)
151. Damask A, Dienes J *Point Defects in Metals* (New York: Pergamon Press, 1963) [Translated into Russian (Moscow: Mir, 1966)]
152. Thompson M *Defects and Radiation Damage in Metals* (Cambridge: Cambridge University Press, 1969) [Translated into Russian (Moscow: Nauka, 1983)]
153. Girifalko L *Statistical Physics of Materials* (New York: Wiley, 1973) [Translated into Russian (Moscow: Mir, 1975)]
154. Palatnik L S, Fuks M Ya, Kosevich V M *Mekhanizm Obrazovaniya i Struktura Kondensirovannykh Plenok* (Mechanism of Fabrication and Structure of Condensed Films) (Moscow: Nauka, 1972)
155. Chopra K *Thin Film Phenomena* (New York: Wiley, 1969) [Translated into Russian (Moscow: Mir, 1972)]
156. Potemkin V V, Bakshi I S, Zhigal'skiĭ G P *Radiotekh. Elektron.* **28** 221 1 (1983) [*Radio Eng. Electron. Phys.* **28** 89 (1983)]
157. Celasco M, Fiorillo F, Mazzetti P *Phys. Rev. Lett.* **36** 38 (1976)
158. Bokshteĭn B S *Diffuziya v Metallakh* (Diffusion in Metals) (Moscow: Metallurgiya, 1978)
159. Zhigal'skiĭ G P et al., in *Nauchnye Trudy po Problemam Mikroelektroniki. Poluprovodnikovye Pribory i Integral'nye Skhemy* Vyp. 27 (Problems of Microelectronics. Semiconductor Devices and Integrated Circuits. Issue 27) (Moscow: MIET Publ., 1976) p.134
160. Zhil'kov É A et al., in *Nauchnye Trudy po Problemam Mikroelektroniki. Poluprovodnikovye Pribory i Integral'nye Skhemy* Vyp. 27 (Problems of Microelectronics. Semiconductor Instruments and Integrated Circuits. Issue 27) (Moscow: MIET Publ., 1976) p. 127
161. Dobrynina G M, Zhigal'skiĭ G P, Novikov V V, in *Nauchnye Trudy po Problemam Mikroelektroniki. Poluprovodnikovye Pribory i Integral'nye Skhemy* Vyp. 27 (Problems of Microelectronics. Semiconductor Devices and Integrated Circuits. Issue 27) (Moscow: MIET Publ., 1976) p. 141
162. Bakshee I S et al., in *Proc. 10th Int. Conf. on Noise in Physical Systems* (Ed. A Ambrozy) (Budapest, Hungary, 1990) p. 389
163. Bakshi I S et al. *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 5 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 5th All-Union Conf.) (Vilnius, USSR, 1988) p. 117
164. Zhigal'skiĭ G P, Karev A V, Siranashvili I Sh, in *Fluktuatsionnye Yavleniya v Fizicheskikh Sistemakh. Tez. Dokl. 5 Vses. Konf.* (Fluctuation Phenomena in Physical Systems. Abstracts of the 5th All-Union Conf.) (Vilnius, USSR, 1988) p. 133
165. Zimmerman N M et al. *Phys. Rev. B* **34** 773 (1986)
166. Celasco M, Fiorillo F, Masoero A *Phys. Rev. B* **19** 1304 (1979)
167. Verbruggen A H, Koch R H, Umbach C P *Phys. Rev. B* **35** 5864 (1987)
168. Kurov G A, Zhil'kov E A *Mikroelektronika* **1** 144 (1972)
169. Kurov G A et al. *Dokl. Akad. Nauk SSSR* **219** 582 (1974) [*Sov. Phys. Dokl.* **19** 772 (1975)]
170. Zhigal'skiĭ G P, Dobrynina G M, Fedorov A S, in *Nauchnye Trudy MIET po Problemam Mikroelektroniki. Ser. Fiz.-Mat.* Vyp. 35 (Problems of Microelectronics. No. 35) (Moscow: MIET Publ., 1977) p. 150
171. Agrikov Yu M et al. *Elektronnaya Tekhnika Ser. 6 Mikroelektronika* **4** (25) 69 (1970)
172. Zhigal'skiĭ G P, Markaryants É A, Fedorov A S *Poverkhnost': Fizika, Khimiya, Mekhanika* (4) 78 (1993)
173. Geguzin Ya E *Diffuzionnaya Zona* (Diffusion Zone) (Moscow: Nauka, 1979)
174. Zhigal'skiĭ G P, Brylov I N, Markaryants É A, in *Nauchnye Trudy MIET "Fizika i Modelirovanie Tekhnologicheskikh Protsessov, Marshrutov i Elementov SBIS"* [Physics and Models of Technological Processes, Routes, and VLSI Elements (Ed. Yu A Parmenov)] (Moscow: MIET Publ., 1991) p. 46
175. Akhromushkin E A, Zhigal'skiĭ G P, in *Nauchnye Trudy MIET "Fizika i Modelirovanie Tekhnologicheskikh Protsessov, Marshrutov i Elementov SBIS"* [Physics and Models of Technological Processes, Routes, and VLSI Elements (Ed. Yu A Parmenov)] (Moscow: MIET Publ., 1991) p. 37
176. Potemkin V V et al., in *Proc. 6th Sci. Conf. on Fluctuation Phenomena in Physical Systems* (Palanga, Lithuania, 1991) p. 79
177. Holland L (Ed.) *Thin Film Microelectronics* (London: Chapman and Hall, 1965) [Translated into Russian (Moscow: Mir, 1968)]
178. *Tablitsy Fizicheskikh Velichin. Spravochnik* (Tables of Physical Quantities. Reference Book) (Ed. I K Kikoin) (Moscow: Atomizdat,1976)

179. Dagge K et al., in *Proc. 13 Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Eds V Bareikis, R Katilius) (Singapore: World Scientific, 1995) p. 603
180. Zhigal'skiĭ G P, Kurov G A, Siranashvili I Sh *Izv. Vyssh. Uchebn. Zaved. Radiofiz.* **26** 207 (1983) [*Radiophys. Quantum Electron.* **26** 162 (1983)]
181. Dimon P, Dutta P, Horn P M *Bull. Am. Phys. Soc.* **24** 358 (1979)
182. Potemkin V V, Gertsenshteĭn M E, Bakshi I S *Izv. Vyssh. Uchebn. Zaved. Fiz.* **26** (4) 114 (1983)
183. Zhigal'skiĭ G P *Pis'ma Zh. Eksp. Teor. Fiz.* **54** 510 (1991) [*JETP Lett.* **54** 513 (1991)]
184. Zhurkov S N *Fiz. Tverd. Tela* (Leningrad) **25** 3119 (1983) [*Sov. Phys. Solid State* **25** 1797 (1983)]
185. Timashev S F *Dokl. Akad. Nauk SSSR Ser. Fiz. Khim.* **279** 1407 (1984)
186. Zhigal'skiĭ G P, Sokov Yu E, Tomson N G *Radiotekh. Elektron.* **24** 410 (1979) [*Radio Eng. Electron. Phys.* **24** 137 (1979)]
187. Fleetwood D M, Giordano N *Phys. Rev. B* **28** 3625 (1983)
188. Kurov G A, Zhigal'skiĭ G P, Siranashvili I Sh, in *Puti Povysheniya Stabil'nosti i Nadezhnosti Mikroelementov i Mikroskhem. Materialy 2 Vses. Nauchno-Tekh. Seminara* (Approaches to Improving Stability and Reliability of Microelements and Microcircuits. Proc. 2nd All-Union Scientific and Technical Workshop) (Ryazan': RRTI Publ., 1982) p. 41
189. Zhigal'skiĭ G P, Sokov Yu E, Tomson N G, in *Fizika Poluprovodnikov i Mikroelektronika* (Physics of Semiconductors and Microelectronics) (Ryazan': RRTI Publ., 1977) p. 63
190. Hoffman R U, in *Physics of Thin Films. Advances in Research and Development* Vol. 3 (Eds G Hass, R Thun) (New York: Academic Press, 1966) [Translated into Russian (Moscow: Mir, 1968) p. 225]
191. Friedel J *Dislocations* (Oxford, 1964) [Translated into Russian (Moscow: Mir, 1967)]
192. Van Bueren H G *Imperfections in Crystals* (Amsterdam: North-Holland, 1960) [Translated into Russian (Moscow: Inostrannaya Literatura, 1961)]
193. Hirth J, Lothe J *Theory of Dislocations* (New York: McGraw-Hill, 1968) [Translated into Russian (Moscow: Atomizdat, 1972)]
194. Prokhorov N N *Fizicheskie Protsessy v Metallakh pri Svarke* Vol. 2 (Physical Processes in Metals Under Welding) (Moscow: Metallurgiya, 1976)
195. Scofield J H, Mantese J V, Webb W W *Phys. Rev. B* **34** 723 (1986)
196. Kirby P L *Electronic Engineering* **37** 722 (1965)
197. Dobrynina G M, Zhigal'skiĭ G P, Mozgin A A, in *Fizika Poluprovodnikov i Mikroelektronika* (Physics of Semiconductors and Microelectronics) (Ryazan': RRTI Publ., 1979) p. 79
198. Zhigal'skiĭ G P, Siranashvili I Sh, Khaletskiĭ V G, in *Metody i Sredstva Integral'noĭ Diagnostiki* (Methods and Tools for Integral Diagnosis) (Kiev: KPI, 1984) p. 28
199. Jones B K, Xu Y Z, Emonts P G A, in *Conf. Proc. ESREF 92* (Schwabisch Gmund, Germany, 1992) p. 363
200. Jones B K, Xu Y Z *Microelectron. Reliab.* **33** 1829 (1993)
201. Potemkin V V, Bakshi I S, Zhigal'skiĭ G P, in *Elektrofluktuatsionnaya Diagnostika Materialov i Izdeliĭ Elektronnoĭ Tekhniki* (Electrofluctuation Diagnosis of Materials and Appliances for Electronic Devices) (Moscow: TsNIIITEI Publ., 1981) p. 52
202. Chen T M, Djeu T P, Moore R D, in *Proc. Reliability Phys. 1985, Orlando* (New York: IEEE Publ., 1985) p. 87
203. Bagnoli P E et al. *J. Appl. Phys.* **63** 1448 (1988)
204. Neri B et al., in *Proc. 10th Int. Conf. "Noise in Physical Systems"* (Ed. A Ambrozy) (Budapest, Hungary, 1990) p. 237
205. Chen T M, Fang P, Cottle J G, in *Proc. 10th Int. Conf. "Noise in Physical Systems"* (Ed. A Ambrozy) (Budapest, Hungary, 1990) p. 515
206. Schwarz J A et al. *J. Appl. Phys.* **70** 1561 (1991)
207. Smith R G, Biery G A *Appl. Phys. Lett.* **65** 315 (1994)
208. Chen T M, in *Proc. AIP Conf. 285 "Noise in Physical Systems and* $1/f$ *Fluctuations"* (St. Louis, New York, 1993) p. 17
209. Yassine A M, Chen T M, in *Proc. 13th Int. Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Eds V Bareikis, R Katilius) (Singapore: World Scientific, 1995) p. 614
210. Ciofi C, Diligenti A, Neri B, in *Proc. 13th Int. Conf. "Noise in Physical Systems and* $1/f$ *Fluctuations"* (Eds V Bareikis, R Katilius) (Singapore: World Scientific, 1995) p. 618
211. Vrachev A S *Trudy Moskovskogo Energeticheskogo Instituta* Vyp. 522 (Proc. Moscow Power Engineering Institute No. 522, 1981) p. 19
212. Koch R H, Lloyd J R, Cronin J *Phys. Rev. Lett.* **55** 2487 (1985)
213. D'Erl F, Rosenberg R, in *Physics of Thin Films: Advances in Research and Development* Vol. 7 (Eds G Hass, M Francombe, R Hoffman) (New York: Academic Press, 1973) [Translated into Russian (Moscow: Mir, 1977)]
214. Poate J, Tu K, Mayer J (Eds) *Thin Films. Interdiffusion and Reactions* (New York: Wiley, 1978) [Translated into Russian (Moscow: Mir, 1982)]