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THE MULTIQUANTUM PHOTOEMISSIVE EFFECT

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1. INTRODUCTION

In recent years there has been a renewal of interest in the photoemissive effect, the emission of electrons by matter into the vacuum or into other matter under the influence of electromagnetic radiation. The revival of interest in the photoemissive effect has been caused by the development of methods of generating powerful coherent electromagnetic radiation, resulting in the appearance of new intensive light sources. Owing to the emergence of the powerful light sources, a number of new experiments have been set up in recent years to observe single-quantum photoemission on metal–dielectric and metal–semiconductor boundaries with the aim of investigating the size and structure of the potential barrier (see, for example, [1-5]). Experiments have been started on the boundary between a metal and an electrolyte solution.[5-8] Peculiarities in the structure of the boundary dividing the metal and electrolyte make it possible, applying a small potential difference, to change substantially the properties of the surface and its photoemissive characteristics. This opens up the possibility of using the photoemissive effect to obtain important information on the properties of the interfaces and absorption layers. On the other hand, the emergence of powerful light sources permits us to study the multiquantum photoemissive effect experimentally. This has an important theoretical significance.

In the present article we give a survey of work carried out in recent years which has been devoted to the theoretical and experimental study of the multiquantum photoemissive effect. The elemental event in the multiquantum photoemissive effect in metals in the visible and ultraviolet regions of the spectrum results from absorption of photons by conduction electrons; in semiconductors and dielectrics the determining factor is the excitation of electrons from bound states, from a valence band, and from defects or surface states. This leads to an essential difference between the photoemission properties of metals and those of semiconductors or dielectrics. In the following, therefore, we consider papers on the multiquantum photoemissive effect in metals and dielectrics separately (Secs. 2 and 3).

The authors of [9] on an experimental investigation of the multiquantum photoemissive effect discovered that the values of the probability of a two-photon transition in coherent and incoherent light beams of equal intensity and spectral composition were different. In view of the theoretical importance of this effect we consider this range of questions in detail in Sec. 4 of the survey.

Among the papers on the multiquantum effect, we should also include, strictly speaking, papers on the multiphoton ionization of separate atoms or molecules in gases and papers on the corresponding multiquantum photonuclear reactions. Such work, however, is not considered below. We thus limit ourselves to consideration of the multiquantum photoemissive effect in condensed media.

Papers published up to 1 January 1969 have been taken into consideration in the survey.

2. THE MULTIQUANTUM PHOTOEMISSIVE EFFECT IN METALS

2.1. Theory

The laws of conservation of energy and momentum exclude the possibility of absorption of a photon by a free electron. The absorption of photons by conduction electrons in a metal may occur, therefore, either in a surface layer, where the abrupt change in the potential and the exponential falling off of the electron wave function outside the metal render the conduction electrons bound, or inside the metal, where phonons or impurities act as third bodies in the interaction of the electrons and photons. Accordingly the photoemissive effect in metals may be a surface or volume effect. The surface photoeffect is dominant for light frequencies less than the ultraviolet transmission threshold ω^* of the metal; the photon energy corresponding to ω^* amounts to 8–10 eV.[10,11] In most of the theoretical papers devoted to the calculation of the multiquantum photocurrent in metals, it is the surface photoeffect that is considered. Clearly, in the first stage of the study of the question this is natural, since the photon energies of the optical quantum generators (lasers) used at present for the activation of the multiquantum photoeffect do not, as a rule, exceed 3.5 eV.

The problem of calculating the multiquantum photocurrent can be set in the following way. A plane monochromatic wave of frequency ω such that

$$1 < \frac{\chi}{\hbar\omega} < n, \qquad (2.1)$$

where χ is the work function of the metal, $\hbar$ is Planck's constant, and n is an integer greater than unity (the order of the photoeffect), impinges on the plane separation boundary, coinciding with the yz coordinate plane, of the metal and the medium. Assuming a definite model for the potential field in which the conduction electrons in the metal move, we want to find the photoelectron emission current into the vacuum or into another substance. This problem can be decomposed into two parts. In the first of these the x-component of the partial electron current density must be determined for $x \to \infty$ (the metal occupies the half-space $x < 0$)

$$\mathbf{j}_0 = \frac{ie\hbar}{2m}(\psi\nabla\psi^* - \psi^*\nabla\psi) - \frac{e^2}{mc}\mathbf{A}\psi^*\psi, \quad (2.2)$$

where **A** is the vector potential of the wave (below, the Coulomb gauge is chosen everywhere for the potentials: div **A** = φ = 0). For this it is necessary to solve the wave equation for the system "electron in the metal + electromagnetic radiation." The Hamiltonian of such a system may be represented in the form

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1, \quad (2.3)$$

where

$$\hat{\mathcal{H}}_0 = \hat{\mathcal{H}}_e + \hat{\mathcal{H}}_r = \frac{1}{2m}\hat{\mathbf{p}}^2 + V(\mathbf{r}) + \hat{\mathcal{H}}_r,$$

$\hat{\mathcal{H}}_r$ is the Hamiltonian of the free radiation field, and $\mathcal{H}_i$ is the electron-photon interaction Hamiltonian.

The function ψ in (2.2) satisfies the Schrödinger equation

$$i\hbar\frac{\partial\psi}{\partial t} = \hat{\mathcal{H}}\psi. \quad (2.4)$$

In the second part of the problem the value of the photocurrent is calculated by integrating the expression (2.2) over the momenta of the emitted electrons and over all the electron states in the metal.

The various theoretical papers on the surface photoemissive effect differ amongst themselves essentially by their different approaches to the solution of the first part of the problem of calculating the photocurrent.

We shall consider first the simplest case, that of the two-quantum photoeffect, n = 2 (cf. (2.1)). The possibility of observing a two-quantum surface photoeffect was first discussed in a paper by R. Makinson and M. Buckingham,[12] in which an estimate was made of the order of magnitude of the two-quantum photocurrent without specification of the form of the potential barrier on the metal-vacuum boundary.

The first paper in which the surface two-quantum photoeffect was analyzed in detail is, apparently, the paper by R. Smith.[13] We represent the Hamiltonian in the form (cf., e.g., [11])

$$\hat{\mathcal{H}} = -\frac{\hbar^2}{2m}\nabla^2 + V(x, y, z) + \frac{i\hbar e}{mc}\mathbf{A}\nabla + \frac{e^2}{2mc^2}\mathbf{A}^2. \quad (2.5)$$

Introducing the dimensionless quantities[13]

$$\left.\begin{aligned} x' &= \frac{x}{\lambda_0}\sqrt{2}, \quad \lambda_0 = \frac{\hbar}{mc}, \quad t' = \frac{t}{\lambda_0}c, \\ \nabla' &= \frac{\lambda_0}{\sqrt{2}}\nabla, \quad W = \frac{V}{mc^2}, \quad \mathbf{G} = \frac{e\mathbf{A}}{mc^2\sqrt{2}}, \end{aligned}\right\} \quad (2.6)$$

we write Eq. (2.4) in the form (the primes are omitted in the following)

$$\nabla^2\psi - W\psi + i\frac{\partial\psi}{\partial t} = 2i\mathbf{G}\nabla\psi + \mathbf{G}^2\psi. \quad (2.7)$$

We seek the solution of Eq. (2.7) in the form of a perturbation theory series

$$\psi = \sum_{n=-\infty}^{n=+\infty}\psi(n)\,e^{i(\alpha+n\omega)t}, \quad (2.8)$$

where α is a dimensionless constant corresponding to the electron energy when **A** = 0. Let

$$\mathbf{G} = \mathbf{K}e^{i\omega t} + \mathbf{M}e^{-i\omega t}, \quad (2.9)$$

where **K** = **M**. Putting (2.8) and (2.9) into (2.7) and equating to zero the coefficients in the terms of the form exp i(α + kω)t, we find

$$\begin{aligned}&[\nabla^2 - W - (\alpha + k\omega) - 2\mathbf{KM}]\,\psi(k) - 2i\mathbf{K}\nabla\psi(k-1)\\ &- 2i\mathbf{M}\nabla\psi(k+1) - \mathbf{M}^2\psi(k+2) - \mathbf{K}^2\psi(k-2) = 0. \quad (2.10)\end{aligned}$$

We assume that ψ(n) is of order $|\mathbf{G}|^{|n|}$; then, when terms of higher than first order are neglected, Eq. (2.10) gives

$$[\nabla^2 - W - (\alpha - \omega)]\,\psi(-1) - 2i\mathbf{M}\nabla\psi(0) = 0, \quad (2.11)$$

$$[\nabla^2 - W - \alpha]\,\psi(0) = 0, \quad (2.12)$$

$$[\nabla^2 - W - (\alpha + \omega)]\,\psi(1) - 2i\mathbf{K}\nabla\psi(0) = 0. \quad (2.13)$$

We note that the function ψ(1) corresponds to an energy level of energy $\hbar\omega$ less than that of the initial state; $\psi(-1)$ corresponds to a level of energy $\hbar\omega$ greater than that of the initial state, and the function ψ(0) corresponds to the state in the absence of the electromagnetic field.

We write the vector potential of the monochromatic wave impinging on the metal boundary in the form

$$\mathbf{A} = 2\mathbf{A}_0\cos(\omega t - \mathbf{kr}) = 2\mathbf{A}_0\cos(\omega t + kx\cos\theta + ky\sin\theta), \quad (2.14)$$

i.e.,

$$\mathbf{K} = \mathbf{G}(0)\,e^{i(x\cos\theta + y\sin\theta)\omega/\sqrt{2}}, \quad (2.15)$$

where

$$\mathbf{G}(0) = \frac{e\mathbf{A}_0}{mc^2\sqrt{2}}. \quad (2.16)$$

The wave vector **k** of the wave lies in the xy plane and makes an angle θ with the normal to the metal boundary. We assume that the wavelength of the radiation is significantly greater than that of the electron. This approximation corresponds essentially to neglecting the photon momentum in comparison with the Fermi momentum of the electron, which we can certainly do, since the former is two to three orders smaller than the latter.

Neglecting, by virtue of the above argument, the spatial dependence of the vector potential, we find an equation of second order for the function $\psi(-2)$:

$$[\nabla^2 - W - (\alpha - 2\omega)]\,\psi(-2) = 2i\mathbf{G}(0)\nabla\psi(-1) + \mathbf{G}^2(0)\,\psi(0). \quad (2.17)$$

Let the field V(x, y, z) be a "potential box:"

$$\begin{aligned} V &= 0, & x &> 0, \\ V &= -\hbar\omega_a, & x &< 0, \end{aligned} \quad (2.18)$$

i.e.,

$$\begin{aligned} W &= 0, & x &> 0, \\ W &= -W_a = -\frac{\omega_a\lambda_0}{c}, & x &< 0. \end{aligned}$$

Then the solution of (2.17) when $\mathbf{x} \to \infty$ is

$$\psi(-2)_{x\to\infty} = se^{i\gamma_{2x}x}YZ, \quad (2.19)$$

where

$$\begin{aligned} Y &\equiv e^{i\beta_y y} + e^{-i\beta_y y}, & Z &\equiv e^{i\beta_z z} + e^{-i\beta_z z}, \\ \beta &= \sqrt{W_a - \alpha}, & \beta^2 &= \beta_x^2 + \beta_y^2 + \beta_z^2, \\ \gamma_2 &= \sqrt{-(\alpha - 2\omega)}, & \gamma_2^2 &= \gamma_{2x}^2 + \beta_y^2 + \beta_z^2; \end{aligned}$$

and s is a constant determined from the boundary conditions. We note that γ_{2x} is a real quantity.

We calculate the current density of a single electron when $x \to \infty$. To this end, we notice that we may neglect the last term in expression (2.2), since it corresponds to to the current caused by an electron oscillating in the radiation field; this current therefore makes no contribution to the total current. From (2.2) we find an expression for the x-component of the partial current density.

$$j_{0x} = -\frac{2\zeta}{i\lambda_0^4}(\psi^*\nabla\psi - \psi\nabla\psi^*), \tag{2.20}$$

where $\zeta = |e|\hbar/m$.

Putting (2.19) into (2.20) and averaging over the variables y and z we obtain*)

$$\overline{j}_{0x} = -8\zeta\gamma_{2x}\frac{ss^*}{\lambda_0^4},$$

or

$$\overline{j}_{0x} = -128\mu|a|^2 R(\beta_x), \tag{2.21}$$

where

$$\mu = \frac{\zeta G_x^4(0) W_a}{\lambda_0^4\omega^4},$$

and

$$R(\beta_x) = \frac{\beta_x^2(\beta_x^2+2\omega-W_a)^{1/2}}{4\omega - W_a + 2\beta_x^2 + 2[(2\omega+\beta_x^2)(2\omega-W_a+\beta_x^2)]^{1/2}}$$
$$\times\left\{\frac{5}{2}W_a + \omega + 2[(\omega+\beta_x^2)(2\omega+\beta_x^2)]^{1/2} + 2[(W_a-\beta_x^2)(W_a-\omega-\beta_x^2)]^{1/2}\right\}.$$

Associating the electron density in the metal, and thereby the Fermi–Dirac distribution, with the quantity $|a|^2$, we find

$$\langle\psi\psi^*\rangle = 8|a|^2 = \frac{2d\beta_x d\beta_y d\beta_z}{8\pi^3\left\{1+\exp\left[\frac{(\beta^2-W_F)c^2}{8\pi^2 T}\right]\right\}};$$

Here $W_F = \mathscr{E}_F/mc^2$ is the dimensionless Fermi energy.

If T = 0, then for the two-quantum photocurrent density we have the expression

$$j_2 = -\frac{4\mu}{\pi^3}\int R(\beta_x)\, d\beta_x d\beta_y d\beta_z, \tag{2.22}$$

when $\beta_x^2 + \beta_y^2 + \beta_z^2 < W_F$, and $j_2 = 0$ if $\beta_x^2 + \beta_y^2 + \beta_z^2 > W_F$. Integrating over β_y and β_z in (2.22) we find that

$$j_2 = -\frac{4\mu}{\pi^3}\int_{0 \text{ or } \sqrt{W_a-2\omega}}^{\sqrt{W_F}} (W_F - \beta_x^2) R(\beta_x)\, d\beta_x, \tag{2.23}$$

where the lower limit is equal to zero if $W_a < 2\omega$.

Finally the relation (2.23) may be written in the form

$$j_2 = -\frac{|e|^5 c}{4^5\pi^7 m^2\hbar^2}\left[\frac{4\pi m\nu_0}{\hbar} + (3\pi^2 n_e)^{2/3}\right]F_x^4$$
$$\times\frac{\nu_0^3}{\nu^8}\int_{0 \text{ or } (\mu-2\eta)^{1/2}}^{(\mu-1)^{1/2}} (\mu - 1 - \lambda^2) f(\lambda, \mu, \eta)\, d\lambda, \tag{2.24}$$

where

$$\lambda = \frac{\beta_x}{\sqrt{\omega_0}}, \quad \eta = \frac{\omega}{\omega_0}, \quad \mu = \frac{W_a}{\omega_a}, \quad \omega_0 = W_a - W_F,$$

$$\left.\begin{aligned} 2\pi\nu &= \frac{\omega c}{\lambda_0} \text{—dimensional angular frequency} \\ 2\pi\nu_0 &= \frac{(W_a - W_F)c}{\lambda_0} = \frac{V_a - \mathscr{E}_F}{\hbar} \text{—dimensional threshold frequency} \end{aligned}\right\} \tag{2.25}$$

$F_x = F\sin\theta$ is the component of the electric field intensity of the impinging electromagnetic wave,*) n_e is the electron density in the metal, and $\mathscr{E}_F = (\hbar^2/2m) \times (3\pi^2 n_e)^{2/3}$ (to derive the function $f(\lambda, \mu, \eta)$ we must substitute (2.25) in the expression for $R(\beta_x)$).

The integration of expression (2.24) for a specific metal may be carried out numerically. As an example we consider sodium:[17]

$$\chi = V_a - \mathscr{E}_F = 2.28 \text{ eV} \quad \mathscr{E}_F = 3.12 \text{ eV},$$

i.e., $V_a = 5.40$ eV and $\mu = V_a/\chi = 2.36$. If the wavelength of the radiation is 7000 Å, then $\eta = 0.82$. The numerical value of the integral in expression (2.24) under such conditions is 0.11. For the total density of the two-quantum photocurrent from the surface of sodium we have the expression

$$|j_2| = 7.8\cdot 10^{-31} F_x^4 \text{ A/cm}^2 \tag{2.26}$$

where F_x is expressed in V/m. If $F_x = 10^8$ V/m, then $|j_2| = 10^2$ A/cm^2.

Analogous calculations were performed later in the papers.[16, 18] In the paper by M. E. Marinchuk,[16] an expression was obtained in addition for the distribution function of the photoelectrons as a function of their total energies. On the basis of G. Fan's paper[19] on the single-quantum photoeffect, Bloch[18] calculated the value of the two-quantum photon current caused by direct transitions between bands, using a model with a spherical Fermi surface for the metals.

To determine the partial electron current we may also make use of the Green function method and the formal theory of scattering, as was done in the paper by I. Adawi.[15] In this the Hamiltonian of the free radiation field is written in the form

$$\hat{\mathscr{H}}_r = \sum_\beta \hat{a}_\beta^+ \hat{a}_\beta \hbar\omega_\beta, \tag{2.27}$$

where $\hat{a}_\beta^+$ and $\hat{a}_\beta$ are creation and annihilation operators for the radiation oscillator β with angular frequency ω_β ($\hat{a}_\beta\hat{a}_\beta^+ - \hat{a}_\beta^+\hat{a}_\beta = 1$). The electron-photon interaction Hamiltonian may be represented in the form

$$\hat{\mathscr{H}}_1 = \sum_\beta i\frac{e\hbar}{m}\left(\frac{2\pi\hbar}{\omega_\beta}\right)^{1/2}(\mathbf{e}_\beta\mathbf{n})\,\hat{a}_\beta\frac{\partial}{\partial x} \equiv \sum_\beta \gamma_\beta\hat{a}_\beta\hat{D}, \tag{2.28}$$

where $\mathbf{e}_\beta$ is the unit polarization vector parallel to $\mathbf{A}_\beta$, $\mathbf{n}$ is the unit vector in the direction of the x axis, $D \equiv \partial/\partial x$ and

$$\gamma_\beta = i\frac{e\hbar}{m}\left(\frac{2\pi\hbar}{\omega_\beta}\right)^{1/2}\mathbf{e}_\beta\mathbf{n}.$$

The radiation field is quantized in unit volume.

In the initial state let an electron be in the state Φ_0, so that $\hat{\mathscr{H}}_e\Phi_0 = E_0\Phi_0$ and all the occupation numbers of the radiation oscillators, apart from n, are equal to zero. It is convenient to write the wave function of the

*For the function $R(\beta_x)$ we adduce here an amended expression from the subsequent papers of other authors [14]-[16].

*We note that the photoeffect occurs only when the wave is polarized in the plane of incidence. The field component along the surface of the metal makes no contribution to the photoeffect probability, since in transverse motion the electrons behave as if they were free electrons.

initial state in the form

$$\psi_0 = |n_\beta, \Phi_0\rangle,$$

and the initial energy in the form

$$\mathscr{E}_0 = E_0 + n_\beta \hbar\omega_\beta.$$

The final state ψ^+ (for the outgoing wave) is the solution of the scattering equation

$$\psi^+ = |n_\beta, \Phi_0\rangle + \frac{1}{\mathscr{E}_0 - \hat{\mathscr{H}}_0 + i\varepsilon}\hat{\mathscr{H}}_1\psi^+. \quad (2.29)$$

We represent ψ^+ in the form

$$\psi^+ = \psi_0 + \psi_1 + \psi_2, \quad (2.30)$$

where

$$\psi_1 = \gamma_\beta n_\beta^{1/2}|n_\beta - 1, \Phi_1\rangle, \qquad \psi_2 = \gamma_\beta^2[n_\beta(n_\beta-1)]^{1/2}|n_\beta - 2, \Phi_2\rangle.$$

Substituting (2.30) in (2.29) we find

$$\Phi_1 = \hat{G}_1\hat{D}\Phi_0, \quad (2.31)$$

$$\Phi_2 = \hat{G}_2\hat{D}\hat{G}_1\hat{D}\Phi_0, \quad (2.32)$$

where $\hat{G}_1$ and $\hat{G}_2$ are the Green functions:

$$\hat{G}_1 = \frac{1}{E_0 + \hbar\omega_\beta - \hat{\mathscr{H}}_e + i\varepsilon}, \quad (2.33)$$

$$\hat{G}_2 = \frac{1}{E_0 + 2\hbar\omega_\beta - \hat{\mathscr{H}}_e + i\varepsilon}. \quad (2.34)$$

From (2.33) and (2.34) it follows that

$$\hat{G}_r\hat{D} = \hat{G}_r\hat{D}[E_r - \hat{\mathscr{H}}_e + i\varepsilon]\hat{G}_r = \hat{G}_r[-V' + (E_r - \hat{\mathscr{H}}_e + i\varepsilon)\hat{D}]\hat{G}_r$$
$$= \hat{D}\hat{G}_r - \hat{G}_r V'\hat{G}_r \qquad (r = 1, 2), \quad (2.35)$$

where $V' = \hat{\mathscr{H}}_e\hat{D} - \hat{D}\hat{\mathscr{H}}_e$ is the force acting on the electron.

Using the commutation relations (2.35) we may express the amplitude of the outgoing electron wave in the form of a series that includes both the effect of the potential and the effect of the force (the rate of change of the potential). In [15], the following examples of potential distribution V(x) are considered:

$$V(x) = \begin{cases} -V_a, & x < 0, \\ 0, & x > 0, \end{cases} \quad (2.36)$$

$$V(x) = \begin{cases} -V_a, & x \leqslant -a, \\ \frac{V_a}{2a}(x-a), & |x| \leqslant a, \\ 0, & x \geqslant a, \end{cases} \quad (2.37)$$

$$V(x) = \begin{cases} -V_a, & |x| < a, \\ 0, & |x| > a, \end{cases} \quad (2.38)$$

$$V(x) = \begin{cases} V_2(-2a - x), & x < -2a, \\ -V_a, & -2a < x < 0, \\ V_1(x), & x > 0. \end{cases} \quad (2.39)$$

The results of Adawi[15] for the potential distribution (2.36) coincide, of course, with those of Smith[13] (after correction of unfortunate errors in some formulas in the latter paper). The author made numerical estimates for ordinary metals; if $n_\beta \approx 10^{15}$ cm^{-3} and $\lambda_\beta \approx 2\pi \times 10^3$ Å, then $|j_2| \sim 10^{-3}$ A/cm^2.

In the papers [13, 15, 16, 18] considered above, the basic technique for calculating the value of the multiquantum photocurrent is perturbation theory (the methods of the stationary theory of scattering, used in the work of Adawi, are equivalent to perturbation theory). However, application of perturbation theory can be relied upon to be successful only when the two-quantum photoeffect is considered. For larger values of $n \sim \chi/\hbar\omega$ the difficulties in the calculation increase to such an extent that the use of perturbation theory becomes impracticable.

In connection with this, the paper by F. V. Bunkin and M. V. Fedorov,[20] in which the value of the n-quantum photocurrent is calculated without using perturbation theory, seems to be important.

For an electron moving in a potential box

$$V(x) = \begin{cases} -V_a, & x < 0, \\ 0, & x > 0, \end{cases}$$

the wave functions have the form[20]

$$\psi_p(x) = a_p \begin{cases} e^{\frac{ipx}{\hbar}}, & x > 0, \\ b_p^+ \exp\left[i\frac{(p^2+2mV_a)^{1/2}x}{\hbar}\right] + \\ \quad + b_p^- \exp\left[-i\frac{(p^2+2mV_a)^{1/2}x}{\hbar}\right], & x < 0, \end{cases}$$

$$\psi_k(x) = d_k \begin{cases} e^{\frac{ikx}{\hbar}}, & x > 0 \\ f_k^+ \exp\left[i\frac{(2mV_a - k^2)^{1/2}x}{\hbar}\right] + \\ \quad + f_k^- \exp\left[-i\frac{(2mV_a - k^2)^{1/2}x}{\hbar}\right], & x < 0, \end{cases} \quad (2.40)$$

where the numbers p define states of positive energy ($\mathscr{E}_p = p^2/2m$) and the numbers k define states of negative energy ($\mathscr{E}_k = -k^2/2m$); in this case

$$-\infty < p < \infty, \qquad 0 < k < \sqrt{2mV_a}.$$

The coefficients b_p^+, b_p^-, f_k^+ and f_k^- in (2.40) are determined from the boundary conditions when x = 0, while a_p and d_k are found from the normalization conditions

$$\left.\begin{aligned} &\int_{-\infty}^{+\infty} dx\psi_p^*\psi_{p'} = \delta(p-p') + \lambda_p\delta(p+p'), \\ &\int_{-\infty}^{+\infty} dx\psi_k^*\psi_{k'} = \delta(k-k'), \\ &\lambda_p = \frac{mV_a}{p^2 + |p|(p^2+2mV_a)^{1/2} + mV_a}. \end{aligned}\right\} \quad (2.41)$$

In formula (2.40), for brevity, we have not written out factors of the type

$$\exp\left[\frac{i}{\hbar}(p_y y + p_z z)\right],$$

corresponding to transverse motion of the electrons (it is possible to trace directly which results will be obtained by correctly taking such factors into account).

We write the interaction Hamiltonian in the form

$$\hat{\mathscr{H}}_1 = \frac{e}{c}\dot{A}(t)x, \quad A(t) = -F\frac{\sin\theta}{\omega}\sin\omega t,$$

where F is the intensity of the electric field of the light wave and θ is the angle of incidence of the wave on the surface of the metal. We seek the wave function of an electron, which at the initial moment of time t = 0 is in state ψ_k in the form

$$\Psi(x, t) = \exp\left(\frac{i}{\hbar}\frac{k^2}{2m}t\right)\psi_k + \int dp C_p(t)\Psi_p(x, t), \quad C_p(0) = 0. \quad (2.42)$$

For our $\Psi_p(x, t)$ we choose the orthonormal system of functions[21]

$$\Psi_p(x, t) = \Psi_{p - \frac{e}{c}A(t)}\exp\left[\frac{i}{\hbar}\int_0^t \left(p - \frac{e}{c}A(\tau)\right)^2\frac{d\tau}{2m}\right]. \quad (2.43)$$

The electron transition probability from the state ψ_k to the state Ψ_p in unit time is

$$\lim_{t\to\infty}\frac{|C_p(t)|^2}{t}. \qquad (2.44)$$

Integrating the expression (2.44) over the momenta of the emitted electrons and over all electron states inside the metal we can find the value of the photocurrent density:

$$j=\frac{me\omega^{3/2}V_a^{1/2}}{32\pi^6\hbar^{3/2}}\gamma^{3/2}\lambda^{5/4}\int_{\gamma_F}^{\gamma}dq\left[\frac{q}{(1+q^2)^{1/2}}\right]^{5/4}\frac{\left(1-\frac{q}{\gamma}\right)^{1/2}}{q^2}(q^2-\gamma_F^2)$$
$$\times\left|J\left\{(\gamma-q)\frac{(q\lambda)^{1/2}}{(1+q^2)^{1/4}}\right\}\right|^2\sum_{s=n}^{\infty}\Theta\left[\left(\frac{s-\frac{\lambda}{2}}{\lambda}\right)^{1/2}-q\right]\left(s-\frac{\lambda}{2}-\lambda q^2\right)^{-1/2}$$
$$\times\exp\left\{-2\left[s\left(\operatorname{arsh}q-\frac{q}{(1+q^2)^{1/2}}\right)+\frac{\lambda q^3}{2(1+q^2)^{1/2}}\right]\right\}, \qquad (2.45)$$

where

$$\gamma=\frac{(2mV_a)^{1/2}\omega}{eF_x},\qquad \gamma_F=\frac{(2m\chi)^{1/2}\omega}{eF_x},\qquad \chi=V_a-\mathscr{E}_F,\qquad F_x=F\sin\theta,$$
$$\lambda=\frac{e^2F_x^2}{2m\hbar\omega^3}=\frac{\chi}{\hbar\omega\gamma_F^2},\qquad \Theta(x)=\begin{cases}1, & x>0,\\ 0, & x<0,\end{cases}$$
$$s=E\left[\lambda\left(\frac{1}{2}+\gamma_F^2\right)+1\right],$$

and E denotes the integral part.

The function J(x) is defined as the integral

$$J(x)=\lim_{\varepsilon\to 0}\int_{-\infty}^{+\infty}\frac{e^{-z^2}dz}{(z-i\varepsilon)^2(z-ix)^{1/4}}.$$

We consider the limiting case $\gamma_F \gg 1$. In this case the sum over s in expression (2.45) may be restricted to the first term. This gives

$$j=\frac{\delta e}{4\sqrt{6}}\frac{m\omega^2}{\hbar}\left(\frac{\chi}{\hbar\omega}\right)^{1/2}\exp(2n-\lambda\gamma_F^2)\left(\frac{e^2F_x^2}{8m\chi\omega^2}\right)^n, \qquad (2.46)$$

where

$$\delta=E\left[\lambda\left(\frac{1}{2}+\gamma_F^2\right)+1\right]-\lambda\left(\frac{1}{2}+\gamma_F^2\right).$$

The expression (2.46) permits us to compute the cold emission current caused by absorption of n quanta ($n \sim \lambda\gamma_F^2 = \chi/\hbar\omega$); for this, (2.46) is more accurate the greater the value of n. The inapplicability of relation (2.46) for small n is connected with the fact that the condition $\gamma_F \gg 1$ is not fulfilled simultaneously with the condition for the applicability, in the time integration, of the method of steepest descents, on which, essentially, the derivation of formula (2.45) rests. A comparison of the estimated values of the emission current found from formula (2.46) with the corresponding values obtained in Smith's paper[13] shows, however, that even for n = 2 the error is slight. Thus, for example, for sodium (χ = 2.28 eV) with $\omega = 3\cdot 10^{15}$ sec^{-1}, formula (2.46) gives $|j_2| \approx 10^{-29} F_x^4$ A/cm^2 (F_x is expressed in V/m). Smith's formula (2.24) for just this case leads to the value $|j_2| \approx 10^{-30} F_x^4$ A/cm^2. For platinum (χ = 6.2 eV), with $\omega = 3\cdot 10^{15}$ sec^{-1} (n = 4), on the basis of (2.46) we obtain $|j_4| \approx 10^{-72} F_x^8$ A/cm^2, which for $F_x \approx 10^9$ V/m corresponds to a photoemission current density of the order of 1 A/cm^2.

Thus it is clear from the above that in the theoretical study of the surface multiquantum photoeffect, as in the study of the single-quantum photoeffect, on the basis of calculations of the photocurrent density, an expression is set which is essentially of the form

$$j=\int\bar{j}_{0x}(E,\mathbf{p}_{\|},A)\left[1+\exp\left(\frac{E-\mathscr{E}_F}{kT}\right)\right]\rho(E,\mathbf{p}_{\|})\,\theta(p^2)\,dE\,d\mathbf{p}_{\|},$$

where $\bar{j}_{0x}(E, \mathbf{p}_{\|}, A)$ denotes the asymptotic value of the x-component of the partial electron current density for $x \to \infty$, averaged over the surface of the metal, E is the electron energy in the metal, $\mathbf{p}_{\|} = (p_y, p_z)$, $\mathscr{E}_F$ is the Fermi energy, $\rho(E, \mathbf{p}_{\|})$ is the density distribution function, $\theta(p^2)$ is the theta-function, and p is the value of of p_x far from the surface. As we have seen already, the basic problem is the calculation of the quantity $\bar{j}_{0x} \times (E, \mathbf{p}_{\|}, A)$. In the papers considered here, this quantity is found from the solution of a quantum-mechanical problem in which the "potential box" model is used to describe the motion of the electrons in the metal. In the paper by A. M. Brodskiĭ and Yu. Ya. Gurevich[22] attention is drawn to the fact that methods of calculating threshold production phenomena[23] can be used to determine the quantity $\bar{j}_{0x}(E, \mathbf{p}_{\|}, A)$. Such a treatment can be carried through without referring to a specific form of the potential barrier. The theory can also be extended to the surface photoeffect on a metal-electrolyte solution boundary. The single-quantum photoeffect was treated in [22] by this method. In another paper by the same authors[24] an expression for the single-quantum photocurrent density on the metal-electrolyte separation boundary is given without derivation:

$$j_n=A_0\frac{2\hbar^2}{5k^2\omega_0^{1/2}}\left(\frac{e^2}{\hbar c}\right)^{n-1}\varphi_n(n\omega-\omega_0)^{5/2}, \qquad (2.47)$$

if $\hbar(n\omega - \omega_0) \gg kT$, where

$$A_0=\frac{4\pi ek^2m}{(2\pi\hbar)^3}=120\ \text{A/cm}^2\,\text{deg}^2$$

and $\omega_0 = \chi/\hbar$ is the photoelectric threshold and $\varphi_n \sim I^n$ (I is the radiation intensity).

We shall make a few observations concerning the nature of the idealizations which lie at the basis of the theoretical papers considered here. All the papers start from the Sommerfeld model of a metal (a gas of electrons, obeying Fermi–Dirac statistics, in a potential box). The problem is assumed to be nonrelativistic. It is assumed that violation of thermodynamic equilibrium during the passage of the photocurrent may be neglected. No attempt is made in any of the papers to take account of reflection and refraction of the electromagnetic wave on the metal-vacuum boundary. Polarization selectivity (a vector effect) of the multiquantum photoeffect has, therefore, not been theoretically investigated either. The latter effect, as is well-known (see, e.g., [25]), contains important features for the case of single-quantum photoemission. A theory of the volume multiquantum photoeffect in metals is also lacking. Neither the paper by Bloch[18] nor its refinement[26] can be regarded as satisfactory in this respect.

2.2. Experiment

The experimental observation of the surface multiquantum photoeffect in metals is beset with a number of difficulties of a methodological nature. The chief of these is that the multiquantum photoeffect process,

which is characterized by small quantum yield, may be complicated by the occurrence of thermal emission (see, e.g., [27-34]). It is clear that, to observe the surface multiquantum photoeffect in metals, the experiment should be set up so that the number of electrons $(N_e)_{ph}$ appearing as a result of the multiquantum photoeffect exceeds appreciably the number of electrons $(N_e)_{th}$ appearing above the target as a result of the heating up caused by single-photon absorption of the radiation. Fulfillment of the condition $(N_e)_{ph} \gg (N_e)_{th}$ may be achieved by reducing the duration τ of the pulse of radiation and increasing its intensity I. In fact, if w_{th} denotes the probability of appearance of the thermoelectrons in unit time, and w_{ph} denotes the corresponding probability for the multiquantum photoeffect, then the following relations hold for the total number of particles emitted during time τ

$$(N_e)_{th} \sim w_{th}\{T(I,\tau)\}\tau, \quad (N_e)_{ph} \sim w_{ph}\tau \sim I^n\tau.$$

Since the phenomenon of thermal emission of electrons, along with absorption of radiation by the free electrons of a metal, has an essentially one-photon character, reducing τ and increasing I allows us to make the ratio $\beta = (N_e)_{th}/(N_e)_{ph} \sim w_{th}(I,\gamma)/w_{ph}$ as small as we choose, i.e., the point is reached where the multiquantum photoeffect will play the fundamental role in the appearance of free electrons over the surface of the metal, while the contribution of the competing thermoemission process will be negligibly small.

We shall estimate the critical duration τ_{cr} of the radiation pulse and the corresponding critical radiation intensity I_{cr} at which the roles of the thermal and photoelectron mechanisms in the appearance of free electrons over the surface of the target are exchanged.[35] The values of the quantities τ_{cr} and I_{cr} may be determined from the solution of the system of equations

$$\tau = \tau_{th}(I), \quad j_n(I)\tau = q_{thr}, \tag{2.48}$$

where q_{thr} is the threshold value of the charge emitted from unit area of the target in the photoeffect process (in the final analysis this value is determined by the threshold of sensitivity of the charge detector); j_n is the current density of the n-quantum photoemission; τ is the duration of the radiation pulse of intensity I, in the course of which thermal emission of charge βq_{thr} is observed. Here β is a constant characterizing the degree of suppression of the thermal emission. The total charge q emitted during the time τ for which the radiation pulse acts may be found after integrating the expression for the thermal emission current density over time from zero to τ with the condition that a temperature

$$T = \frac{2(1-r)I}{\varkappa}\left(\frac{at}{\pi}\right)^{1/2}$$

is realized on the surface of the sample. Here κ and a are respectively the coefficients of thermal conductivity and thermal diffusivity, r is the coefficient of reflection and t is the time elapsed after the initial moment of action of the light pulse. Calculations show that for the quantity q the correct expression is

$$q = A_0\left[\frac{2(1-r)I}{\varkappa}\right]^3 \frac{2\pi k}{a\chi}\left(\frac{a\tau}{\pi}\right)^{5/2}\exp\left[-\frac{\chi\varkappa\left(\frac{\pi}{a\tau}\right)^{1/2}}{2k(1-r)I}\right], \tag{2.49}$$

where

$$A_0 = \frac{4\pi k^2 em}{\hbar^3} = 120 \text{ A/cm}^2 \text{ deg}^2$$

The expression (2.49) enables us to determine the function $\tau_{th}(I)$ if we substitute into it the quantity βq_{thr} in place of q and the quantity τ_{th} in place of τ and solve the resulting equation for τ_{th}. To estimate the photocurrent density we must make use of formula (2.46) which may be written in the more convenient form ($n \geq 3$)

$$j_n(I) = 2^{-3n}B\frac{em\omega^2}{\hbar}n^{1/2}\left(\frac{\omega_t}{\omega}\right)^{2n}, \tag{2.50}$$

where B is a dimensionless quantity of order unity and $\omega_t^2 = e^2F^2/m\chi = 8\pi e^2 I/mc\chi$. Substituting the functions $\tau_{th}(I)$ and $j_n(I)$ in the initial system of equations (2.48) and eliminating the quantity I from them, we can obtain the following equation for an estimate of the pulse duration τ_{cr}:

$$\omega\tau_{cr} = n^{\frac{1}{n-2}}\left(\frac{em\omega}{\hbar q_{thr}}\right)^{\frac{2}{n-2}} \times \left\{\frac{\pi e^2\varkappa}{2mc\omega^2 k(1-r)}\left(\frac{\pi\omega}{a}\right)^{1/2}\left[\ln Z + \frac{5n-6}{2n}\ln(\omega\tau_{cr})\right]^{-1}\right\}^{\frac{2n}{n-2}}, \tag{2.51}$$

where Z is defined by the expression

$$Z = \frac{16A_0k\chi^2}{\pi^2 n^{3\cdot 2n}a\beta q_{thr}}\left(\frac{a}{\pi\omega}\right)^{5/2}\left[\frac{mc\omega^2(1-r)}{e^2\varkappa}\right]^3\left(\frac{q_{thr}\hbar}{em\omega}\right)^{3/n}.$$

The critical intensity I_{cr} corresponding to the pulse duration τ_{cr} is then equal to

$$I_{cr} = \frac{mc\omega^2\chi}{\pi e^2 n^{1/2n}}\left(\frac{q_{thr}\hbar}{em\omega}\right)^{1/n}\frac{1}{(\omega\tau_{cr})^{1/n}}. \tag{2.52}$$

If the radiation pulse parameters satisfy the conditions $\tau < \tau_{cr}$ and $I \gtrsim I_{cr}$, then it becomes possible to observe the multiquantum photoeffect against a background of thermal emission, since the number of electrons appearing over the surface of the target as a result of its being heated up does not exceed a fixed fraction $(\beta/(1+\beta))$ of the total number of emitted particles. Numerical estimates of the threshold quantities τ_{cr} and I_{cr} for a target of gold (χ = 4.8 eV, κ = 0.7 cal/cm^1 sec^1 deg^1, a = 1.2 cm^2/sec^1, r = 0.9) irradiated by a light pulse from a ruby laser (n = 3, $\hbar\omega$ = 1.78 eV) leads to the values $\tau_{cr} = 10^{-8}$ sec and $I_{cr} = 10^6$ W/cm^{-2} when $\beta = 10^{-2}$ and $q_{thr} = 10^{-16}$ Coul/cm^2. The laser radiation pulse parameters used in [36, 37], where the three-quantum photoeffect from the surface of gold was studied, are in satisfactory agreement with the estimates given above.

Although the experimental setups used by authors in observation and study of the multiquantum photoemissive effect differ slightly in details, they are on the whole similar to each other, regardless of whether the photoeffect being studied is in metals, dielectrics, or semiconductors. Figure 1 depicts a schematic diagram of an experimental arrangement for observing the multiquantum photoemissive effect from the surface of irradiated solids. As a rule a laser serves as the radiation source since the current density of the light should, in this case, exceed the value $I \approx 10^{22}$ photons/cm^2 · sec^1. Only then is it possible to observe processes of second (and higher) order, such as the multiquantum

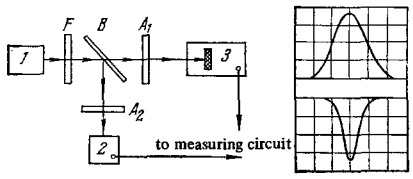


FIG. 1. Schematic diagram of the experimental setup for observation of the surface multiquantum photoeffect: 1 – intense radiation source –laser; 2 – calibrated PM recording the pulse parameters of the laser radiation; 3 – vacuum vessel with a target and an electron detector (often the target is prepared in the form of a photocathode PM); F – light filter eliminating the radiation from the laser pump lamp; A_1 and A_2 – calibrated light flux attenuators; B – light beam divider. The lenses and slits are not shown in the diagram. On the right are typical oscillograms for the laser radiation pulses (above) and the photocurrent (below).

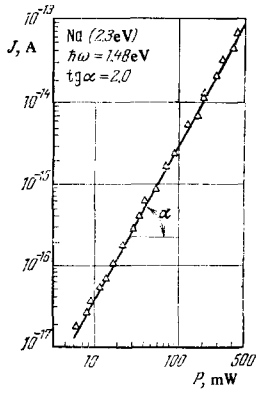


FIG. 2. The dependence of the photocurrent J from the surface of a sodium target on the radiation power P of a gallium arsenide semiconductor laser operating in the pulse regime. A part where $J \sim P$ is sometimes observed in the region of small light flux values. Its existence is explained by the fact that for $P \lesssim 10$ mW the contribution of the two-quantum photoeffect to the total current will be so small that the single-quantum photoeffect of emission of photoelectrons from the tail of the Fermi distribution becomes noticeable[14].

photoemissive effect. To detect the electrons emitted from the surface of the target, either an electron multiplier or a collector placed in immediate proximity with the surface of the sample is used. Sometimes the target is prepared in the form of the photocathode of a photomultiplier (PM). The parameter of the laser radiation pulse and of the emission current pulse are measured by means of a carefully calibrated detection system; the duration of the emission current pulse is, as a rule, less than the duration of the laser radiation pulse (see Fig. 1) and this furnishes evidence of the photoelectron nature of the emission current. A set of calibrated reducers for the light beam enables us to plot the dependence of the emission current J on the light flux intensity I and thus to evaluate the contribution of the multiquantum photoeffect to the measured value of the emission current.

A preliminary report of the first observation of the multiquantum (two-photon) photoemissive effect from the surface of a metal (sodium) is given in [14]. The results of a more thorough investigation were published by the authors later.[26] As a radiation source they used a gallium arsenide semiconductor injection laser, operating at a temperature of 77°K ($P \approx 400$ mW, $\lambda \approx 8450$ Å, f = 2.2 kHz, $\hbar\omega = 1.48$ eV). The laser radiation was sharply focused ($S \approx 5 \times 10^{-5}$ cm^2) on the surface of the photocathode of a photomultiplier (multiplication factor $\sim 5 \times 10^4$) made in the form of a thin metallic film of sodium obtained by vacuum deposition ($\chi \approx 2.3$ eV). A calibrated filter permitted controlled change of the peak radiation power of the laser within the limits $1 \lesssim P \lesssim 400$ mW, corresponding to change in the light flux intensity on the surface of the sample within the limits $2 \times 10^2 \lesssim I \lesssim 10^4$ W/cm^2. The results of investigating the dependence of the emission current J on the peak radiation power P of the laser are given in Fig. 2, which is taken from [26]. As is easily seen, the dependence of J on P found experimentally is, in logarithm coordinates ln J and ln P, a straight line of slope equal to 2. This fact is a result of the two-quantum nature of the photoeffect since the contribution of the competing single-quantum processes of thermal emission and of the photoeffect under the action of second harmonic radiation is negligibly small. As the results of the measurements showed, when laser radiation is focused on the surface of a metallic film of sodium, a temperature increase of up to 2° is observed. The thermoelectronic emission current corresponding to this temperature increase of the sample (the initial temperature was ~300°K) is of order of 10^{-23} A, which is considerably smaller than the value observed in the experiment. The energy of the second harmonic laser radiation ($\lambda = 4225$ Å, $2\hbar\omega = 2.96$ eV) is very small ($P \lesssim 5 \times 10^{-12}$ W) and, it would seem, cannot explain the experimental results by single-photon absorption. If we use the results of the calculations of Smith,[13] then for the experiments carried out by the authors, the dependence of the photocurrent caused by two-quantum absorption on the power of the laser radiation and on the dimensions of the light spot on the surface of the target must have the form $J = 9.7 \times 10^{-24}\ P^2/S$.* The dependence of J on P with S = constant is shown in Fig. 2. In order to verify the inverse proportionality of the photocurrent to the area of the light spot the authors carried out additional investigations, in the course of which, by means of a small adjustment of the lens focusing the laser radiation, they succeeded in changing the dimensions of the light spot on the target. As a result of the experiments it was found that $J \sim S^{-1}$, which agrees with the predictions of the theory. However, estimates of the value of the photoemission current made using formula (2.26) at the peak value of the laser radiation power $P \approx 400$ mW give a photocurrent value $J \approx 3 \times 10^{-16}$ A, whereas the experimentally found value is equal to 9×10^{-14} A. Thus the agreement between the calculated and experimental values of the photocurrent cannot be considered satisfactory. This disparity is caused, apparently, by the volume character of the two-quantum photoeffect in alkali metals, in the same way as this is found in the case of the single-quantum photoeffect.[25, 38]

An interesting experiment, devised to study the dependence of the photocurrent caused by the multiquantum photoeffect on the direction of polarization of the incident radiation, is described in [39]. The sample, a carefully degassed and well-polished silver disc, was placed in a vacuum flask ($p_0 \approx 10^{-8}$ mm Hg) and was irradiated with unfocused light from a ruby laser ($S \approx 1$ cm^2) operating in the "giant pulse" regime ($\tau \approx 25$–30 nsec). To avoid the side effect of thermal emission the sample was irradiated with a glancing light ray with angle of incidence $\theta = 87°$. The control measurements and the calculations have shown that, under similar experimental conditions, the heating of the sample surface

*In the MKS system of units.

caused by absorption of a certain fraction of the energy of the laser pulse is slight and does not exceed 10° (the measurements were carried out at room temperature). Consequently the contribution of the thermal emission process is negligibly small. To detect the photoelectrons they used a cylindrical collector, to which a voltage U = +1000 V was applied, close to the target surface. As was shown by the results of the experiments, free electrons ($J_{max} \approx 10$ mA) appear over the surface of the target under the action of laser radiation pulses ($I = 0.5$ MW/cm^2), and the duration of the emission current is always less than the duration of the laser pulse. Since the work function for silver is 4.7–4.8 eV and the energy of a quantum from a ruby laser is $\hbar\omega = 1.78$ eV, the three-quantum photoemissive effect is the only possible mechanism causing the appearance of the cold emission current. The results of measuring the dependence of the peak value of the photoemission current J_{max} on the direction of polarization of the incident radiation are shown in Fig. 3. It is clear that the photoemission current is a maximum when the component $F_{\parallel}$ of the electric field intensity vector of the laser radiation parallel to the plane of incidence reaches a maximum (a Glan–Thompson prism was used to rotate the plane of polarization). In the case where the angle φ between the plane of incidence and the plane of polarization of the laser radiation is equal to $\pi/2$, the photoemission current is practically absent, although the density of the exciting light flux remains unchanged. This is explained by the fact that, for $\varphi = \pi/2$, the component $F_{\parallel}$ of the electric field intensity is practically equal to zero and, consequently, we may consider the vector of the total intensity of the laser radiation field to be parallel to the surface of the target. For $\varphi \approx 20°$, on the other hand, the value of the component $F_{\parallel}$ reaches a maximum and, consequently, the photoeffect probability increases.

Investigations into the dependence of the three-quantum photoemission current on the intensity I of the radiation from a ruby laser for targets of gold (χ = 4.8 eV),[36, 37] silver (χ = 4.7–4.8 eV),[37, 40] and nickel (χ = 5.1 eV)[37] showed that it has a complicated nonlinear character. The results of these experiments are shown in Figs. 4 and 5, taken from [36, 37]. It is not difficult to see that in the initial part for not too high values of the intensity of the light flux, $J \sim I^3$ and then, as the light flux intensity increases, the function J(I) changes and may be approximated by a function of the form $J = \text{const} \cdot I^{\alpha}$, where $\alpha \approx 6$.[37] The fact that the recorded emission current in the region where $J \sim I^3$ has a photoelectron nature is corroborated both by calculations and by test measurements, which indicate the small heating up of the target under the action of the laser radiation ($\Delta T \approx 10°$). In addition, if we assume that $J \sim I^3$, then, when the form of the laser radiation pulse is close to Gaussian and has a width τ in time, the photocurrent pulse must have a width $\tau' = \tau/\sqrt{3}$. Just such a relation between the pulse widths τ and τ' was also recorded experimentally in [36, 40, 41]. The apparent considerable discrepancy in the values I* up to which the dependence $J \sim I^3$ is observed experimentally (I* ≈ 1.1 MW/cm^2,[36] I* ≈ 45 MW/cm^2 [37]) is the result of two causes, each of which is due to a difference in the conditions of the experiment. First, in the work,[36] the angle of incidence of the laser radiation on the surface of the sample was equal to 60°, whereas in the work[37] it was 85°. The values of the light flux intensities shown in Figs. 4 and 5 characterize the total intensity of the laser radiation. If we recalculate the stated values I* taking the angle of incidence θ into account, then the maximum light flux intensities on the target surface will be respectively equal to 0.55 MW/cm^2 and 3.7 MW/cm^2. Second, and this is perhaps the most important reason, the experiment in the work[37] was set up in such a way that the component of the electric field intensity vector of the incident radiation perpendicular to the surface of the sample had its maximum value for the given angle of incidence θ. In this case, as was shown earlier,[39] the conditions for observing the multiquantum photoemissive effect are optimal. It was just this optimization of the experimental conditions that enabled the authors to observe the dependence $J \sim I^3$ at higher values of the density of the light flux on the surface of the

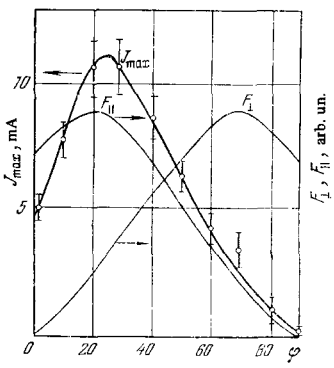


FIG. 3. The dependence of the peak value of the emission current J_{max} and of the components $F_{\parallel}$ and $F_{\perp}$ of the electric field intensity of light wave on the angle φ (φ is the angle between the plane of polarization of the incident radiation from the ruby laser and the plane of incidence). Experimental conditions: angle of incidence $\theta = 87°$, light flux intensity I = 0.5 MW MW/cm^2, target material silver (χ = 4.8 eV).

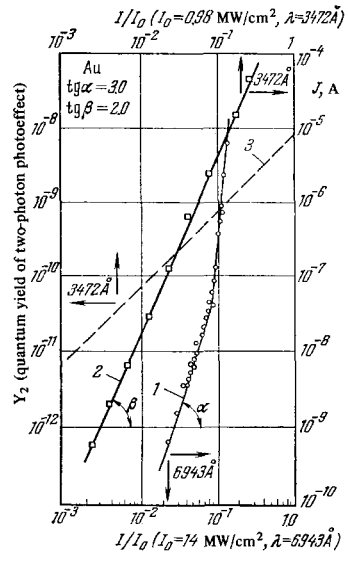


FIG. 4. The dependence of the emission current (1 and 2) and of the quantum yield Y_2 (3) of the two-quantum photoeffect on the relative intensity I/I_0 of the light flux. Upon irradiation of the target with the fundamental harmonic (λ = 6943 Å) of the ruby laser radiation, the three-quantum photoeffect (1) was observed, whereas in the case of irradiation with the second harmonic (λ = 3472 Å) the two-quantum photoeffect was observed. The light beam diameter was equal to 4.6 and 6 mm for cases 1 and 2 respectively. The laser pulse parameters were: E = 1 J, τ = 40 nsec.

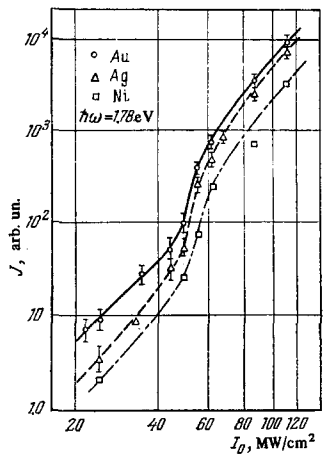


FIG. 5. The dependence of the emission current from the surfaces of gold, silver and nickel targets on the intensity of the ruby laser radiation ($\tau \approx 25$ nsec).

target. The experimentally observed dependence (which is stronger than cubic) of the emission current on I for light flux densities $I > I^*$ remained obscure. In the opinion of the authors of [36], in this region of values of I heating up of the target material begins to play a role and the observed current is thus of a thermal nature. However, measurements of the energy of the electrons carried out by the retarding potential method showed[40] that the average energy of the electrons and their energy distribution cannot be explained by the thermal emission mechanism.

The hypothesis advanced in turn by Farkas et al.,[37] that the stated deviation of the function J(I) at light flux densities $I > I^*$ may arise as a result of increase of the quantum yield of the multiphoton photoeffect is also not free from deficiencies, although, according to calculations,[20] the cold emission current density in a strong radiation field may be described by formula (2.46), from which, in particular, it follows that $j_n \sim f(I)I^n$ and therefore, for the quantum yield of the photoemission process at high light flux densities, the expression $Y_n \sim f(I)I^{n-1}$ is correct, and not $Y_n \sim I^{n-1}$.

Putting aside the question of the nature of the emission current at light flux densities $I > I^*$, we shall discuss estimations of the quantum yield Y_3 in the case of the three-quantum photoemissive effect on the surface of a gold target. If we assume that the photocurrent density j obeys the relation $j = CI^3$ (here C is a constant that takes into account the stochastic character of the three-photon absorption process and the stochastic character of the emission of an electron into free space), then it follows from the experimental data (see Fig. 4.1), that $C = 1.02 \times 10^{-7}$ A/MW)(MW/cm^2)$^{-2}$. In this case the relation $Y_3 = 1.8 \times 10^{-6}\, CI^2$ MW/cm^2 (Y_3 is the ratio of the numbers of emitted electrons and incident photons) is valid for the quantum yield Y_3 of the three-photon photoeffect. For a light flux intensity of, for example, $I = 0.77$ MW/cm^2, we have $Y_3 = 1.1 \times 10^{-13}$ electrons/photon. The investigation of the two-quantum photoemissive effect carried out incidentally by the authors of [36] enabled them to evaluate also the value of the quantum yield for the two-photon photoeffect in gold at wavelength $\lambda = 3472$ Å. For this purpose the samples were irradiated with an unfocused light beam of second harmonic radiation, obtained by means of a KDP crystal, from a ruby laser. The results of measuring the dependence of J on I are shown in Fig. 4.2. The energy of one photon in this case is equal to 3.57 eV ($\chi = 4.8$ eV) and the photoeffect may have only a two-quantum character. If, by analogy with the three-photon effect, we write the relation $j = C'I^2$, then it follows from the experimental data that $C' = 2.35 \times 10^{-3}$ (A/MW)(MW/cm^2)$^{-1}$. The quantum yield Y_2 for a light flux density $I = 0.77$ MW/cm^{-2} is found to be equal to 6.5×10^{-9} electrons/photon (see Fig. 4.3).

The photoemissive effect on a metal-solution interface was studied in paper.[42] The setup used to make the measurements was as follows: a ray of light from a ruby ($\hbar\omega = 1.785$ eV, $E \approx 1$ J, $\tau \approx 100$ μsec) or a neodymium ($\hbar\omega = 1.18$ eV, $E \approx 2$ J, $\tau \approx 100$ μsec) laser was focused using a long-focus lens (f = 94 mm) onto the surface of a mercury drop placed in a decinormal aqueous solution of perchloric acid ($HClO_4$). There was practically no back current of the electrons ejected by the light (i.e., current from the solution to the electrode) since protons, dissociation products of the $HClO_4$, were acting as electron acceptors in the solution. In view of the fact that the one-photon photoeffect work function from mercury into the solution is equal to 3.3 ± 0.15 eV at the potential at which the charge on the metal is zero, and depends additively on the target potential, two-quantum photoemission of electrons into the solution should begin at potential $\varphi = 0.2$ V relative to the zero-charge point in the case when the drop is irradiated by a light pulse from a ruby laser ($2\hbar\omega = 3.57$ eV). For a neodymium laser $2\hbar\omega = 2.36$ eV and $3\hbar\omega = 3.54$ eV and, consequently, two-quantum photoemission should begin at $\varphi = -1.0$ V and three-quantum photoemission at $\varphi = 0.2$ V. All the enumerated effects have been observed experimentally. Investigations of the dependence of the photocurrent on the laser radiation intensity and on the electrode potential φ showed that for the three-quantum photoeffect, $J \sim I^\alpha$ ($\alpha = 2.7$ to 3.0) while for the two-quantum case, $J \sim I^2$. This result coincides qualitatively with expression (2.47), which describes the multiquantum photoeffect process from a metal into an electrolyte solution. It is not possible to trace more exact agreement, since the exact value of the radiation intensity of a laser working in the peak generation regime is not known. The experimentally found dependence on the electrode potential φ of the charge q emitted from the surface of the mercury drop under the influence of radiation from a ruby laser has the form $q^{0.4} \sim \varphi$ (Fig. 6). The value of the two-quantum photoeffect work function extrapolated from these data turns out to be close to 3.3 eV, the value obtained for the one-

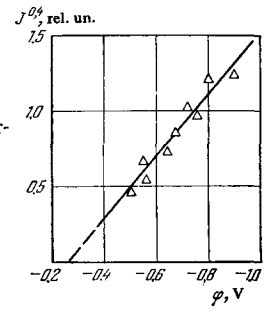


FIG. 6. Voltage-current plot of the two-quantum photoemissive effect from the surface of a mercury drop in a decinormal solution of perchloric acid ($HClO_4$) under the action of radiation from a ruby laser (E = 1 J, $\tau \approx 10^{-4}$ sec). φ is the potential of a saturated calomel electrode.

quantum photoeffect. On exposure to light from a ruby laser, the charge emitted from the surface of the drop reached a value 3×10^{-8} Coul/cm^2 at $\varphi = 0$ and increased to 2.4×10^{-7} Coul/cm^2 at $\varphi = -0.3$ V.

Summarizing the results of the experimental work in which the multiquantum photoemissive effect in metals has been observed, we should say that this work is basically of a preliminary character. It is clear that from now on the number of investigations on the non-linear photoeffect in metals will increase and will enable us to broaden significantly our ideas about both the structure of metals and the elemental act of the photoeffect process.

3. THE MULTIQUANTUM PHOTOEFFECT IN SEMICONDUCTORS AND DIELECTRICS

3.1 Theory

The photoemissive effect in semiconductors and dielectrics is caused by direct and indirect optical transitions from bound states (from a valence band or discrete levels) to free states (conduction bands). In pure and weakly-alloyed semiconductors transitions between bands play the basic role (Fig. 7).

The depth from which photoelectrons are ejected from semiconductors is determined by the energy losses of the electrons to impact ionization. To effect the elemental act of impact ionization an electron in the conduction band must have energy greater than the width $\Delta\mathcal{E}$ of the forbidden band. To be emitted into the vacuum its energy must be not less than the electron affinity δ for the given semiconductor. Therefore, in semiconductors for which $\delta < \Delta\mathcal{E}$, the photoelectrons rapidly lose energy to impact ionization and the depth from which they emerge is small (10^{-6} cm). On the other hand, in semiconductors for which $\delta < \Delta\mathcal{E}$ and also in dielectric ionic crystals, photoelectrons with sufficient energy for emission into the vacuum cannot effect impact ionization and are therefore able to emerge from a greater depth. This is one of the reasons for the large quantum yield of sensitive photocathodes of Cs_3Sb, CsBi, Cs_2Te, etc.[43]

The velocity distribution of the photoelectrons reflects the structure of the energy bands from which they are excited by the radiation. In metals the photoelectrons are scooped out of the conduction band, in which the density of quantum states increases with increase of energy. In semiconductors the density of quantum states in the valence band from which the photoelectrons are excited decreases towards the band edge. In the energy spectrum of the photoelectrons of metals there are, therefore, relatively more fast electrons than in the corresponding spectrum of semiconductors.

In semiconductors containing appreciable amounts of impurities, distinctive features appear in the long-wavelength part of the photoemissive effect spectral characteristic, representing excitation from impurity levels.

The multiplicity of possible mechanisms of excitation and scattering. leading to substantial change in the photoelectron quantum yield in semiconductors, is illustrated to some extent by Table I, taken from the paper by Kane.[44] This table, in particular, is also evidence of the fact that much greater difficulties arise in the theoretical consideration of the multiquantum photoemissive effect in semiconductors than in the case of metals.

This apparently explains the absence of theoretical papers* on the multiquantum photoemissive effect in semiconductors and dielectrics. In the interpretation of experimental results on the multiquantum photoemissive effect in semiconductors and dielectrics at the present time a formula is used which was given without proof in [56] and according to which the following relation for the two-quantum photocurrent density is valid

$$j_2 = \left[\frac{e^5}{\pi}\frac{l}{m^{1/2}\hbar^2\omega^2}\frac{(2\hbar\omega - W_v)^{1/2}}{(W_c - \hbar\omega)^2}\right]F^4, \qquad (3.1)$$

where l is the average emission depth of the photoelectrons, W_c and W_v are the energy differences between the vacuum level and the bottom edge of the conduction band or the top edge of the valence band. From (3.1) it is clear that the two-quantum photoeffect occurs when $\hbar\omega > W_v/2$.

The calculations show that for Cs_3Sb (the energy level scheme is shown in Fig. 8)

$$|j_2| = 1.5 \cdot 10^{-25} F^4 \text{ A/cm}^{-2} \qquad (3.2)$$

where F is expressed in V/m (in the calculation the values $l = 200$ Å, $\hbar\omega = 1.17$ eV, $W_c = 0.45$ eV, $W_v = 2.05$ eV were used). For a light flux density of the order of 35 W/cm^2 (i.e., $F^2 = 1.32 \times 10^8$ V^2/m^2) we have $|j_2| \approx 10^{-9}$ A/cm^{-2}.

3.2 Experiment

Several papers are devoted to the experimental study of the multiquantum photoemissive effect from the surface of semiconductors; all the experiments were car-

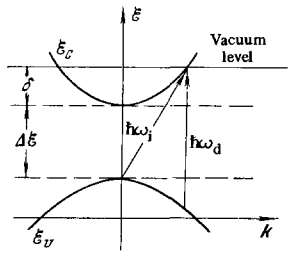


FIG. 7. Direct and indirect interband transitions in a semiconductor. ϵ_c and ϵ_v are the dependences of the electron energy on its quasimomentum k in the conductance and valence bands; $\Delta\epsilon$ is the width of the forbidden band; δ is the electron affinity; $\hbar\omega_d$ is the minimum photon energy for which a direct transition (no change of k) can induce the photoemissive effect; $\hbar\omega_i$ is the minimum photon energy equal to $\Delta\epsilon + \delta$, for which an indirect transition can induce the photoemissive effect.

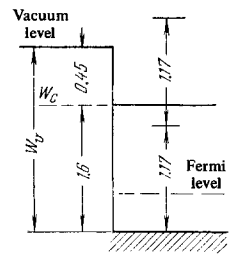
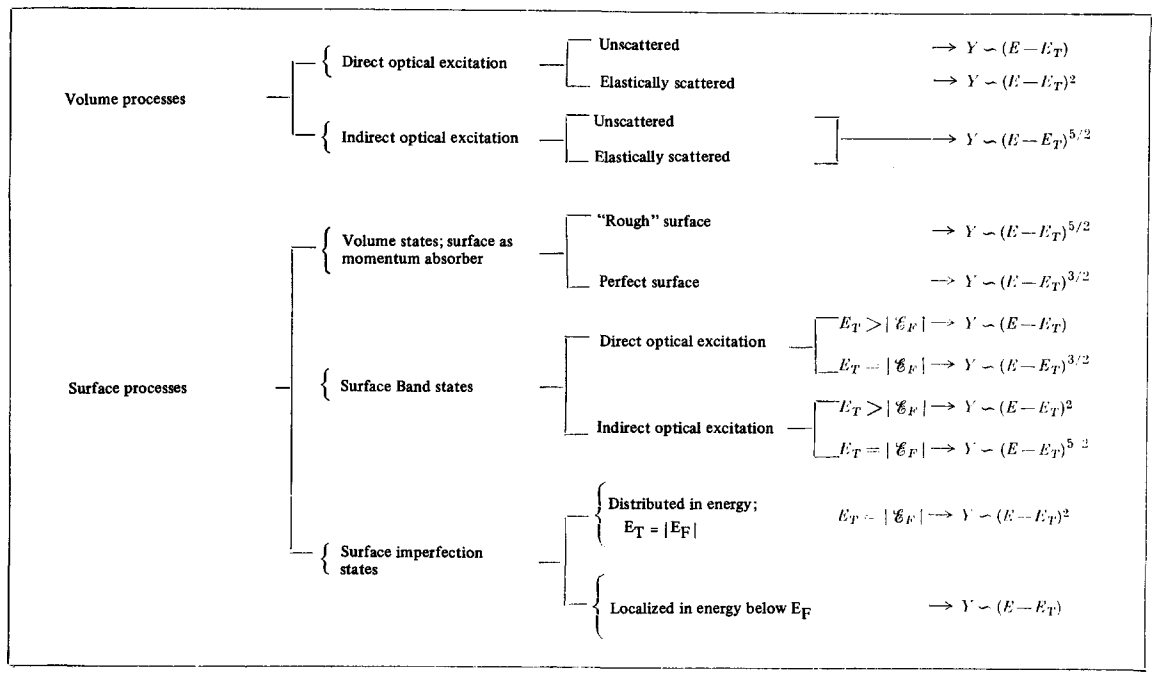


FIG. 8. The energy level scheme in Cs_3Sb (energy in eV).

*There exist, however, a number of papers on other optical multiquantum processes in semiconductors and dielectrics (see, e.g., [45-55]).

Table I. Dependence of photoelectric yield, Y, on photon energy, E, near threshold, E_T, for a variety of production and scattering mechanisms. $\mathcal{E}_F$ is the Fermi level measured from the vacuum



ried out on targets of compounds of antimony with alkali metals, normally used as the coating of photomultiplier photocathodes (Cs_3Sb,[9,56-58] K_3Sb[58]). To investigate the multiquantum photoemissive effect from a Cs_3Sb surface ($1.8 \lesssim \chi \lesssim 2.0$ eV), both a pulsed neodymium glass laser ($\lambda = 1.059\ \mu$, $\hbar\omega = 1.17$ eV, $5 \times 10^{-1} \lesssim I_0 \lesssim 10^3$ W/cm^2)[9,56,58] and a CW helium-neon gas laser ($\lambda = 1.153\ \mu$, $\hbar\omega = 1.07$ eV, $P \approx 1.35$ mW, $5 \lesssim I_0 \lesssim 10^2$ W/cm^2)[57] were used. To study the multiquantum photoemissive effect from a K_3Sb surface ($2.2 \lesssim \chi \lesssim 2.9$ eV) a ruby laser working in the pulse regime ($\hbar\omega = 1.78$ eV, $5 \lesssim I_0 \lesssim 10^3$ W/cm^2) was used.

The results of investigating the dependence of the photocurrent density j_{ph} on the intensity I_0 of the laser radiation incident on the target are shown in Fig. 9. It is clear from the behavior illustrated in the graph that it is possible to divide the curve $j_{ph}(I_0)$ into two parts, in the first of which $j_{ph} \sim I_0$ and in the second $j_{ph} \sim I_0^2$. The presence of parts with a quadratic dependence of the photocurrent emission density j_{ph} on the intensity I_0 of the existing light, observed at light flux densities $I_0 \gtrsim 100$ W/cm^2 for Cs_3Sb samples and $I_0 \gtrsim 100$ W/cm^2 for K_3Sb targets, confirms the two-quantum character of the photoemissive effect, since the energy and power of the light pulses in all cases were such that they did not induce noticeable heating of the samples. Some scattering in the values of j_{ph} for Cs_3Sb targets (see Fig. 9, curves 1–3) is explained, firstly, by the fact that, for the irradiation, lasers with various energy values of the radiation quanta (1,[58] 2,[56] $\hbar\omega = 1.17$ eV; 3,[57] $\hbar\omega = 1.07$ eV) were used, and secondly, by the fact that the work function for Cs_3Sb targets, which depends strongly on the method of preparation and on the purity of the photocathode, varied

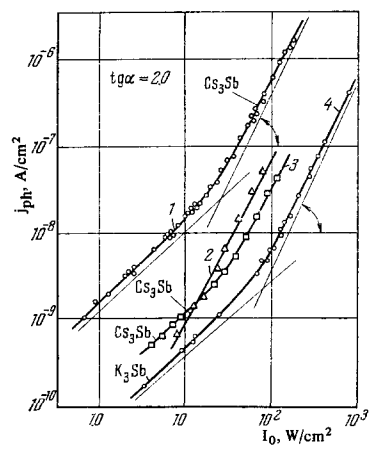


FIG. 9. The dependence of the photocurrent density j_{ph} on the laser radiation intensity I_0 for Cs_3Sb (1 [56], 2 [56], 3 [57]) and K_3Sb (4 [58]) samples.

from sample to sample within wide limits of up to 0.2 eV.

As the investigations carried out in [58] have shown, the presence of a linear portion in the curve of $j_{ph}(I_0)$ for Cs_3Sb and K_3Sb samples is caused by the existence of a long-wavelength "tail" in the single-photon absorption spectrum, which even at small values of the absorption coefficient k makes a significant contribution to the photoemission current because of the large density of the exciting light flux ($j_{ph} \sim kI_0$).

Changes in the energy of the emitted electrons, effected by the retarding potential method,[58] made it possible to find the velocity distribution function of the photoelectrons N(E)dE = f(E) in the case of the surface photoeffect from Cs_3Sb (Fig. 10). From the figure, it is

clear that: 1) the distribution function of the photoelectrons appearing in the two-quantum photoeffect process is virtually identical with the photoelectron distribution function for the single-quantum photoeffect, if the added energy of the two quanta taking part in the two-quantum photoeffect process is equal to the energy of the quantum inducing the single-quantum photoeffect, i.e., $[N(E)dE]_{\lambda=1.06} \approx [N(E)dE]_{\lambda=0.53}$ 2) the maximum of the photoelectron velocity distribution function $[N(E)dE]_{\lambda=1.06}$ lies in the energy region $E \approx 2\hbar\omega - \chi$.

Although both the experimental papers[59,60] in which the multiquantum photoemissive effect from the surface of dielectrics was investigated are of a preliminary character, they nevertheless differ markedly from one another, both in experimental technique and in the nature and depth of the results obtained. Muray's work work,[59] which was devoted to a study of multiquantum photoemission from surfaces of dielectrics transparent to laser radiation—from quartz (χ = 4.2–5.1 eV) and from borosilicate glass (χ = 4.9 eV)—was carried out on a low experimental level. The author investigated the dependence of the number N_e of electrons emitted from the sample surface on the pulse energy of radiation from a ruby laser ($0.2 \lesssim E \lesssim 4.0$ J, $\tau \approx 1.4$ μsec) and succeeded only in establishing the presence of the multiquantum photoeffect (n = 3) at light flux densities $I \gtrsim 10^5$ W/cm^2. Neither the photo-excitation mechanism nor the nature of the secondary phenomena which complicate the observation of the photoeffect were elucidated by the author.

The work of Logothetis,[60] on the other hand, was carried out on a high experimental level and contains a number of important results and ideas. To investigate the spectral characteristics of the multiquantum photoemissive effect in dielectrics, the author used the two-photon spectroscopy method, first suggested by Hopfield et al.[61,62] The essence of the method is as follows. The sample being investigated is irradiated with two independent light beams, one of which is produced by a laser ($\hbar\omega_1$ = const) and the other is continuous radiation, passed through a monochromator ($\hbar\omega_2$ is a variable quantity), from a flash lamp. Thus the total energy of the two quanta $E = \hbar\omega_1 + \hbar\omega_2$ may vary within broad limits, making it easy to plot the spectral characteristics of multiquantum processes (the initial energies $\hbar\omega_1$ and $\hbar\omega_2$ of the quanta are chosen to lie outside the region of possible single-photon absorption in the sample). The radiation sources used in the work[60] were a ruby laser, working in the "giant pulse" regime (E = 1 J, $\hbar\omega_1$ = 1.786 eV, τ = 40 nsec) and a xenon flash lamp ($\tau \approx 50$ μsec) and the synchronization of the laser radiation pulse relative to the flash lamp pulse was effected so that the laser pulse occurred on the "plateau" of the flash-lamp radiation pulse. The use of the monochromator in the path of the light ray from the xenon lamp made it possible to vary the photon energy within the limits $4.0 \lesssim \hbar\omega_2 \lesssim 6.2$ eV, while the spectral width of the radiation which had passed through the monochrometer did not exceed 33 Å. A CsI film of thickness of the order of a few thousand Å, obtained by vacuum deposition onto a backing of stainless steel, was used as a target. The sample was placed in a vacuum cell ($p_0 \approx 10^{-6}$ mm Hg) in such a way that the angle of incidence of the unfocused laser radiation was 60°. To detect the electrons emitted from the surface of the target an electron multiplier was used and was placed close to the sample surface in such a way that scattered radiation from the two exciting light beams did not fall on it. As the experiments demonstrated, the emission current is characterized by the following properties:

1) Emission current pulses are observed only when both light beams are impinging on the target; the photon energy of the UV radiation of the flash lamp must exceed the value $\hbar\omega_2 \gtrsim 4.4$ eV (λ = 2770 Å).

2) The shape and duration of the emission current pulses are similar to the corresponding characteristics of the laser radiation pulse.

3) The emission current pulse is directly proportional both to the laser radiation intensity and to the flash-lamp radiation intensity.

The above-mentioned facts are evidence that the detected photocurrent has a photoelectron nature and that the free electrons over the surface of the CsI sample appear as a result of the two-quantum photoemissive effect. The experimentally determined dependence of the relative value of the quantum yield Y for the two-photon photoeffect on the total energy of the two photons of the exciting radiation is shown in Fig. 11. It is clear that the curve consists of two parts. For $E \gtrsim 6.9$ eV, a sharp increase in the photocurrent is observed, approaching saturation at $E \approx 7.3$ eV. In the energy region $E \lesssim 6.9$ eV the function Y(E) decreases monotonically in proportion to the decrease of E; the lower threshold value, which it was not possible to find because of the smallness of the signal, lies, probably, in the region $E \approx 6.2$–6.3 eV.

To elucidate the nature of the two-quantum photoeffect a comparative analysis of the one- and two-photon

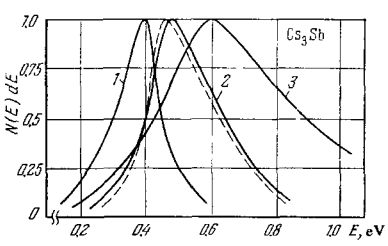


FIG. 10. The normalized photoelectron energy distribution function N(E)dE found by the retarding potential method during irradiation of a Cs_3Sb sample with light of different wavelengths. The solid curve is for the case of irradiation of the sample with light of wavelengths 0.7, 0.53, and 0.4 μ from a thermal source for curves 1, 2, and 3, while the dotted curve is for the case of irradiation of the sample with light from a neodymium laser (λ = 1.06 μ).

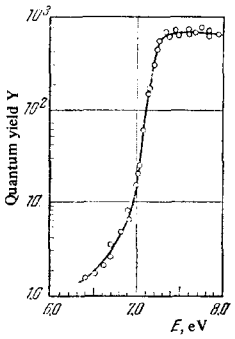


FIG. 11. The dependence of the quantum yield Y of the photoeffect from a film of CsI on the total energy of two photons of the radiation incident on the target ($\hbar\omega_1$ = 1.78 eV, $\hbar\omega_2$ is variable). Y is the number of emitted electrons divided by the number of UV photons from the flash lamp. The intensity of the ruby laser radiation $I_0 = 8 \times 10^{-2}$ MW/cm^2.

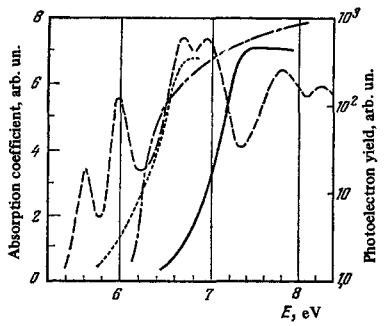


FIG. 12. Spectral functions for one- and two-photon absorptions and photoemission for CsI samples at room temperature. —— one-photon absorption [67]; · · · two-photon absorption [62]; –·– one-photon photoeffect [63]; — two-photon photoeffect [60].

absorption spectra was carried out using data on the one- and two-quantum photoeffect (Fig. 12). It is not difficult to see that the long-wavelength boundary of the two-quantum photoeffect ($E \approx 6.1$–6.3 eV) practically coincides with the corresponding value for the one-quantum photoeffect[63] and is correlated with the two-photon absorption spectrum.[61,62] It is known that the photoemissive effect in alkali halide crystals has an exciton mechanism.[63,64] From a comparison of the one- and two-photon absorption spectra it is clear that in the energy region $5.5 \lesssim E \lesssim 6.4$ eV the spectra differ markedly from each other, whereas in the energy region $6.4 \lesssim E \lesssim 6.9$ eV they differ very slightly. This means that in the region $E \lesssim 6.9$ eV the two-quantum photoeffect may be explained by two-photon "forbidden-forbidden" transitions from the ground state to the first conduction band close to the Γ-point of the Brillouin zone ($\mathbf{k} = 0$)[50,61,65,66] while in the region $E > 6.9$ eV, it is explained by the so-called "allowed-allowed" transitions at some point of the Brillouin zone without inversion symmetry, i.e., at the Σ-line.[50,61] In other words, in the region $E \gtrsim 6.9$ eV there are excited in the crystal states the symmetry of which is identical to the symmetry of the ground state; consequently, direct two-photon excitation of exciton states in the crystal is possible. Although the author did not succeed in identifying concretely the types of the observed transitions responsible for the appearance of the emission current, the investigations which he completed made it possible to draw a number of important conclusions, the essence of which is as follows. The study of the multiquantum photoemissive effect in dielectrics enables us:

1) to study the structure and symmetry type of the energy bands in irradiated media, in the same way as is done in the analysis of single-quantum photoeffect characteristics;[68,69]

2) to understand and evaluate the role of the two-photon-excited states (excitons) which have symmetry identical with the ground state symmetry and which are not observed in one-photon excitation;

3) to study the role of F-centers and of the various exciton-dissociation mechanisms which lead to the appearance of photoemission, and, finally,

4) to trace the influence of the characteristics of the internal multiquantum photoeffect (the angular and polarization dependences of the photoconductivity, etc.) on the process of electron emission from the surfaces of irradiated bodies.

All these questions are still far from being solved and the proposed method of investigating the interaction of radiation with matter undoubtedly helps towards an elucidation of a detailed picture of the phenomenon.

4. STATISTICAL PROPERTIES OF RADIATION AND THE MULTIQUANTUM PHOTOEFFECT PROBABILITY

As a result of the invention of the laser and of the possibility of experimental investigation of electromagnetic fields with a high order of coherence, in recent years a considerable number of theoretical papers devoted to the development and refinement of the concept of coherence has appeared.[70-89] In some of these papers[79-86] the connection between the elemental act of multiquantum absorption and the coherence of the electromagnetic radiation is considered. From the point of view of the theory of optical coherence and photon statistics, a photon counter, and, consequently, a coherence detector, are manifestations of the elemental act of the multiquantum photoeffect. The multiquantum photoemissive effect was also considered from exactly this point of view in [9,78].

Following Glauber[70,71] we define a set of correlation functions, describing the statistical properties of the radiation, as follows:

$$G^{(n)}(x_1, \ldots, x_n, x_{n+1}, \ldots, x_{2n}) = \mathrm{Sp}\,[\hat{\rho}\hat{E}^{(-)}(x_1) \ldots \hat{E}^{(-)}(x_n)\hat{E}^{(+)}(x_{n+1}) \ldots \hat{E}^{(+)}(x_{2n})], \tag{4.1}$$

where $\hat{\rho}$ is the density operator, $x_n = (\mathbf{r}_n, t_n)$, $\hat{E}^{(-)}$ and $\hat{E}^{(+)}$ are non-Hermitian parts of the electric field intensity operator, with negative and positive frequencies respectively.

The function $G^{(n)}(x_1, \ldots, x_n, x_{n+1}, \ldots, x_{2n})$ is called a correlation function of n-th order. As the condition for complete coherence, we require that the normalized n-th order correlation function

$$g^{(n)}(x_1, \ldots, x_{2n}) = \frac{G^{(n)}(x_1, \ldots, x_{2n})}{\prod_{j=1}^{2n} [G^{(1)}(x_j, x_j)]^{1/2}}$$

be of modulus unity for all n and for all combinations of the arguments x. If this condition is fulfilled for $n \leq M$, we shall speak of M-th order coherence.

It is possible to show that the average count rate of n-fold coincidences for ideal (small dimensions, broad band, etc.) photodetectors at the points $x_1, x_2, \ldots, x_n$ is proportional to the function $G^{(n)}(\mathbf{r}_1 t_1, \ldots, \mathbf{r}_n t_n, \mathbf{r}_n t_n, \ldots, \mathbf{r}_1 t_1)$. Since the function $G^{(n)}$ is real and positive, the condition $|g^{(n)}| = 1$ for $n \leq M$ means that

$$g^{(n)}(x_1, \ldots, x_n, x_n, \ldots, x_1) = 1$$

for $n \leq M$. Therefore, by definition of the function $g^{(n)}$ for a field with M-th order coherence, the relation

$$G^{(n)}(x_1, \ldots, x_n, x_n, \ldots, x_1) = \prod_{j=1}^{n} G^{(1)}(x_j, x_j) \tag{4.2}$$

is valid for $n \leq M$.

From the experimental point of view this means that the count rate of n-fold coincidences is equal to the product of the count rates which each counter would give in the absence of the others. Thus, for radiation fields of coherence of order $M \geq n$, n photon counters give statistically independent counts.

For simplicity we shall consider the two-quantum photoeffect as a detector of the coherence of the radia-

tion.[78] The average count rate at the space-time point $x_0 = (r_0, t_0)$ for an ideal two-photon detector is proportional to the quantity

$$G^{(2)}(x_0, x_0, x_0, x_0) = \mathrm{Sp}\,[\hat{\rho}\hat{E}^{(-)}(x_0)\,\hat{E}^{(-)}(x_0)\,\hat{E}^{(+)}(x_0)\,\hat{E}^{(+)}(x_0)].$$

We assume that the radiation has first-order coherence, i.e., that

$$G^{(2)}(x_i, x_j, x_j, x_i) = g_2\,[G^{(1)}(x_i, x_i)]\,[G^{(1)}(x_j, x_j)], \qquad (4.3)$$

where g_2 is the coherence parameter ($g_2 \neq 1$).

Information about the radiation field may be obtained by means of experiments of two types: a) by absolute measurement of the two-quantum photocurrent; b) by means of relative measurements when the detector is irradiated with two superimposed light fluxes.

The output signal of the two-quantum detector, when the latter is irradiated with a single light flux, is proportional to

$$G^{(2)}(x_0, x_0, x_0, x_0),$$

where $x_0 = (r_0, t_0)$. When the detector is irradiated with the same flux delayed in time, the output signal at the moment of time t_1 is proportional to

$$G^{(2)}(x_1, x_1, x_1, x_1),$$

where $x_1 = (r_1, t_1)$ and $t_1 > t_0$.

The average density of the two-quantum photocurrent j_2 when the detector is irradiated with two superposed light fluxes from the same source is proportional to the following sum.

$$j_2 \sim G^{(2)}(x_0, x_0, x_0, x_0) + G^{(2)}(x_1, x_1, x_1, x_1) + G^{(2)}(x_0, x_1, x_1, x_0) + G^{(2)}(x_1, x_0, x_0, x_1). \qquad (4.4)$$

The first two terms in the sum (4.4) correspond to absorption of two photons at the same space-time point (both photons from the same light flux); the first term corresponds to one light flux and the second to the other; the last two terms in (4.4) correspond to absorption of two photons at different space-time points (each photon from a different light flux).

Putting (4.3) into (4.4) and remembering that the correlation function $G^{(1)}(x_j, x_j)$ is proportional to the average single-quantum count rate at the point x_j, we find

$$j_2 = g_2 C\,(I_0^2 + 2I_0I_1 + I_1^2), \qquad \tau_\delta < \tau_c, \qquad (4.5)$$

where I_j is the intensity of the j-th flux and C is a constant. Here it is assumed that the delay time τ_δ between the fluxes is less than the coherence time τ_c of the radiation. The formula (4.5) means, in particular, that the two-quantum photocurrent induced by radiation from thermal sources ($g_2 = 2$, $g_m = m!$) is twice as large as the two-quantum photocurrent caused by radiation from an ideal single-mode laser ($g_1 = 1$, $g_m = 1$). Physically this is explained by the fact that the probability of simultaneous arrival of two photons is greater when there is no correlation. This is valid both for a single light flux and for two superposed light fluxes. The same results were arrived at by the authors of the theoretical paper,[80] in which the dependence of the two-photon absorption probability on the coherence of the radiation was studied.

If the delay time between the fluxes is greater than the coherence time of the radiation ($\tau_\delta > \tau_c$) then there is no correlation of the times of arrival of photons from one flux and the other. In the given case we may write

$$G^{(2)}(x_0, x_1, x_1, x_0) = [G^{(1)}(x_0, x_0)]\,[G^{(1)}(x_1, x_1)],$$

or

$$j_2 \sim g_2\,[G^{(1)}(x_0, x_0)]^2 + g_2\,[G^{(1)}(x_1, x_1)]^2 + 2\,[G^{(1)}(x_0, x_0)]\,[G^{(1)}(x_1, x_1)], \qquad \tau_\delta > \tau_c. \qquad (4.6)$$

We note that the two-quantum photocurrent caused by absorption of one photon from each light flux is here the same as for the radiation from a single-mode laser; it is equal to $2CI_0I_1$.

For random fields, including also the thermal radiation field ($g_m = m!$), we have

$$j_2 = 2C\,(I_0^2 + I_0I_1 + I_1^2), \qquad \tau_\delta > \tau_c, \qquad (4.7)$$

while, for a single mode laser ($g_2 = 1$)

$$j_2 = 2C\left(\frac{1}{2}I_0^2 + I_0I_1 + \frac{1}{2}I_1^2\right). \qquad (4.8)$$

Again the photocurrent for random fields is greater than the photocurrent for the laser radiation field. Comparing (4.7) and (4.8) we can see that the ratio of the coefficient of the cross term I_0I_1 to the coefficient of I_0^2 or I_1^2 is equal to unity in (4.7) and equal to 2 in (4.8). This means that in the case of random fields the count increases for each flux

$$G^{(2)}(x_0, x_0, x_0, x_0) \text{ and } G^{(2)}(x_1, x_1, x_1, x_1).$$

The reason for this is the tendency of the photons to arrive as correlated pairs. So long as $\tau_\delta > \tau_c$, there is no correlation between the arrival time of a photon from one flux and the arrival time of a photon from the other flux. Thus it is clear from the above that the multiquantum photoeffect can give us information about the correlation functions of electromagnetic radiation and, conversely, for full information on the multiquantum photoeffect it is necessary to know the statistical character of the radiation which causes it.

Using the apparatus of quantum electrodynamics and the above-mentioned ideas, the authors of [81, 84] solved the problem of calculating the probability of multiphoton absorption under the action of a laser radiation pulse and thermal radiation. If the thermal radiation, appropriately filtered, has the same frequency spectrum and total energy as the laser radiation pulse, the ratio of the probability of an n-photon transition for thermal radiation to the corresponding probability for coherent light is equal to

$$\eta_n = \frac{n!\,n^{1/2}}{(4\pi)^{(n-1)/2}}\left(\frac{\tau_c}{\tau}\right)^{n-1}, \qquad (4.9)$$

where τ is the time for which the atomic system is irradiated with the thermal radiation and τ_c is the coherence time of the laser radiation ($\tau_c \Delta\nu \sim 1$). Here it is assumed that the laser radiation pulse is a pure coherent polychromatic state, for which the pulse duration τ_1 is equal to the coherence time τ_c, whereas the thermal radiation is stationary and may be described by a density operator which is diagonal in the occupation numbers. The latter is justified by the fact that the frequency spectrum and total energy of the laser and thermal radiation pulses are equal and, therefore, to compensate the difference in the spectral intensities,

the inequality $\tau \geq \tau_c$ must be fulfilled. From expression (4.9) we find

$$\frac{\eta_n}{\eta_{n-1}} \approx \frac{n}{\sqrt{2}} \frac{\tau_c}{\tau}, \quad (4.10)$$

$$\eta_2 = \sqrt{\frac{2}{\pi}} \frac{\tau_c}{\tau_c}, \quad (4.11)$$

$$\eta_1 = 1. \quad (4.12)$$

The relation (4.11) shows that $\eta_2 < 1$. On the other hand, it is clear from (4.10) that the ratio η_n/η_{n-1} increases with increase of n. However, because, usually, $n < \tau/\tau_c$, the ratio η_n/η_{n-1} remains less than unity. This means that when the probabilities of multiphoton absorption in coherent and incoherent light fluxes are compared, when $\tau \gg \tau_1$ the probability of n-photon absorption for a laser radiation pulse is usually greater than that for thermal radiation.

Only one experimental paper in which the dependence of the multiquantum photocurrent on the statistical properties of the radiation was investigated is known at the present time. In this paper[9] the two-quantum photoemissive effect from the surface of cesium antimonide (Cs_3Sb) was studied. A schematic diagram of the experimental setup on which the measurements were carried out is given in Fig. 13. A ray of light from a neodymium laser ($\hbar\omega = 1.17$ eV) working in the giant pulse regime impinged on the surface of a photocathode PM prepared from Cs_3Sb ($\chi \approx 2$ eV). To compare the photoemission currents in coherent (1) and incoherent (2) light fluxes, the authors used two independent methods of irradiating the sample. In the first of these (1) the photocathode was irradiated by a light pulse from the neodymium laser and in the second (2), by the same laser radiation which had previously been passed through plates (~1200 lines/mm) which diffused the radiation; the object of the gratings was to make the light flux completely incoherent, while only slightly weakening it, i.e., to make it such that the phase of the electromagnetic wave would vary randomly in the interval $[0, 2\pi]$. In other words, the diffusion plates were used to bring about an essential change in the phase characteristics of the field and to convert the coherent laser radiation field, characterized by insignificant fluctuation of the phase about a certain fixed value, into a radiation field with a random spread of phases in the interval $0 \leq \varphi \leq 2\pi$. The use of calibrated light flux attenuators enabled the authors to irradiate the photocathode with

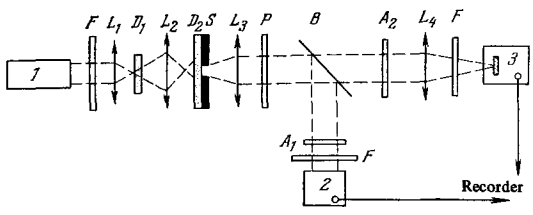
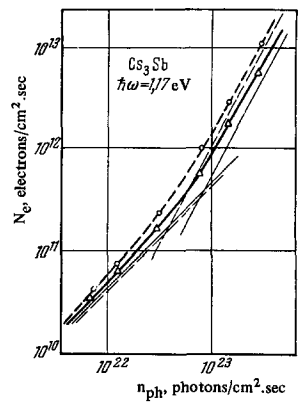


FIG. 13. Schematic diagram of experimental setup: 1 – neodymium glass laser operating in the giant pulse regime; 2 – photomultiplier recording the light flux density of the laser radiation; 3 – electron multiplier with a Cs_3Sb target as photocathode; L – lenses; F – filter eliminating the radiation from the laser pumping-lamp; A_1 and A_2 – calibrated light flux density attenuators; P – polarizer making it possible to keep constant the ratio of the light fluxes which have passed through the light beam divider B; D_1 and D_2 – diffusing plates intended to convert a coherent light flux from the laser into an incoherent one; S – slit.

FIG. 14. The dependence of the number of electrons N_e emitted from unit surface of a Cs_3Sb target in unit time on the light flux density n_{ph}. The points O and Δ correspond to averaged experimental values for irradiation with incoherent and coherent light fluxes. The experimentally determined functions $N_e = \alpha n_{ph} + \beta n_{ph}^2$ are shown as dotted and solid lines for the cases of excitation by incoherent and coherent radiation respectively. The faint dotted straight lines are asymptotes of the type $N_e = \alpha_{incoh} n_{ph}$ and $N_e = \beta_{incoh} n_{ph}^2$, and the faint solid lines are asymptotes of the type $N_e = \alpha_{coh} n_{ph}$ and $N_e = \beta_{coh} n_{ph}^2$.



light fluxes whose intensities coincided but whose statistical properties were different. As was shown by the experiments (Fig. 14) which were carried out for light flux densities in the interval $10^{21} \lesssim n_{ph} \lesssim 10^{24}$ photons/$cm^{-2} \cdot sec^{-1}$, the number of photoelectrons N_e emitted from unit surface of the target in unit time depends on the flux density n_{ph} and $N_e^{coh} < n_e^{incoh}$ always. When the dependences obtained in the form $N_e = \alpha n_{ph} + \beta n_{ph}^2$ were analyzed, it was found that $\alpha_{coh} \approx \alpha_{incoh}$ ($\alpha_{coh} \approx 4.66 \times 10^{-12}$, $\alpha_{incoh} \approx 4.55 \times 10^{-12}$), whereas $\beta_{incoh} > \beta_{coh}$ always ($\beta_{incoh} \approx 1.24 \times 10^{-34}$, $\beta_{coh} \approx 6.57 \times 10^{-35}$) in the region of the quadratic dependence of the photocurrent ($N \sim n^2$). In other words, the number of photoelectrons emitted in the two-quantum photoeffect process under the action of incoherent electromagnetic radiation is greater than the corresponding number of photoelectrons produced under the action of coherent laser radiation of the same intensity and spectral composition; the photoemission currents satisfy the relation $N_e^{incoh} = (1.88 \pm 0.05) N_e^{coh}$, which agrees well with the value $N_e^{incoh} = 2N_e^{coh}$ predicted theoretically.[80, 83] We note that the conditions of this experiment do not correspond to those of the problem solved in [84].

The results of this work illustrate clearly the importance of taking account of the statistical properties of the laser radiation field in the correct calculation of the multiquantum photoeffect probability and are evidence of the fact that in this way it is possible in principle to measure and determine the coherence of optical radiation.

5. CONCLUSION

The multiquantum photoemissive effect is one of the non-linear interaction processes of electromagnetic radiation with matter. It is essentially a quantum phenomenon.

Investigations of the single-quantum photoemissive effect have played a fundamental role in the experimental basis of the quantum theory of light. The multiquantum photoeffect evidently has a no less important role. This is concerned in particular with establishing and studying the connection between multiquantum absorp-

tion of photons and radiation coherence. On the other hand, the study of multiquantum photoemission of electrons from condensed media enables us to investigate the structural features of energy bands, the symmetry type of electron wave functions in a crystal, the structure of the potential barrier at the interface separating two media, etc.

The investigation of the multiquantum photoeffect also enables us to determine the precise limits of applicability of the basic laws of the linear photoeffect, the laws of Stoletov and Einstein.

At the present time only a few steps have been taken in the theoretical and experimental study of the multiquantum photoeffect. Evidently the number of papers and the depth of the investigations on the multiquantum photoeffect will grow steadily from now on. The possibility of practical use of the multiquantum photoeffect as a detector of the coherence order of radiation is also not excluded.

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