

Scientific session of the Physical Sciences Division of the Russian Academy of Sciences (12 April 2006)

A scientific session of the Physical Sciences Division of the Russian Academy of Sciences was held in the Conference Hall of the Lebedev Physics Institute, Russian Academy of Sciences, on 12 April 2006. The following reports were presented at the session:

(1) **Khokhlov D R** (Lomonosov Moscow State University) “High-sensitivity terahertz radiation detectors based on a new class of semiconductor materials”;

(2) **Mitin A V** (Kazan State Technological University) “Modulation gamma-resonance spectroscopy”;

(3) **Kurochkin V E** (Institute of Analytical Instrumentation, Russian Academy of Sciences, St. Petersburg) “Methods and tools for the express immunoassay. A new approach to solving the problem”;

(4) **Lukin V P** (Institute of Atmospheric Optics, Siberian Division of the Russian Academy of Sciences, Tomsk) “Adaptive optical imaging in the atmosphere.”

A brief presentation of the reports is given below.

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High-sensitivity terahertz radiation detectors based on a new class of semiconductor materials

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1. Introduction

Modern trends in the development of physics in the area of optoelectronics are moving towards an increase in the operating wavelength of the corresponding devices to several dozen or hundred micrometers. The terahertz range of electromagnetic waves occupies a special place among the existing problems. This is precisely the spectral range that corresponds to the characteristic phonon frequencies in solids and the rotational–vibrational modes of heavy molecules, which is of great interest to spectroscopic research. Furthermore, many important problems in entirely different fields of science are related to the investigation of terahertz radiation, e.g., in space research and biomedicine. Among the rapidly advancing areas of the practical application of terahertz spectroscopy is remote selective probing of dielectric objects. These include many organic materials and, in particular, explosives.

Progress in this area is moderated by the fact that both radiophysical techniques — on the longer wavelength side — and optical ones — on the shorter wavelength side — operate poorly in the terahertz range. In particular, the sensitivity of terahertz radiation detectors is substantially lower than the sensitivity of infrared and radio-frequency radiation detectors.

The majority of high-sensitivity photodetection systems operating in the terahertz electromagnetic wave range have been traditionally made on the basis of lightly doped germanium or silicon. The longest photoelectric threshold wavelength ($\sim 220 \mu\text{m}$) corresponds to uniaxially strained Ge(Ga) [1]. The most important advantage of germanium and silicon is the extremely well-proven technology of crystal growth, which enables producing materials with a very low uncontrollable impurity density. At the same time, external action, like hard radiation, may introduce a large number of defects in an initially extremely pure or perfect material, which degrades the performance of the corresponding photodetection devices.

This report is concerned with the opportunities furnished by the use of alternative materials — doped alloys based on lead tellurides — for recording low-intensity terahertz radiation.

2. Basic properties of doped alloys based on lead telluride

Lead chalcogenides are widely used in infrared optoelectronics, primarily in the fabrication of lasers and light-emitting diodes operating in the medium- and far-infrared (IR) ranges [2]. Furthermore, considerable progress has been reached in the production of photodiode arrays with a large number of elements, which are sputtered on silicon substrates via fluoride buffer layers [3]. The use of alloys on the basis of lead telluride doped with elements of group III opens up new interesting possibilities due to the emergence of qualitatively new physical properties in the doped material. The specific properties inherent in lead telluride-based alloys may provide improvement in the performance of the corresponding photodetector devices in comparison with the existing analogues.

2.1 Fermi-level stabilization

Lead telluride-based alloys are ternary or quadruple solid solutions of the IV–VI group semiconductors heavily doped with some impurities (which typically belong to group III): $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$, $\text{Pb}_{1-x}\text{Mn}_x\text{Te}(\text{In})$, $\text{Pb}_{1-x}\text{Mn}_x\text{Te}(\text{Ga})$, $\text{PbTe}(\text{Ga})$, $\text{Pb}_{1-x-y}\text{Sn}_x\text{Ge}_y\text{Te}(\text{In})$, $\text{Pb}_{1-x}\text{Ge}_x\text{Te}(\text{Ga}, \text{Yb})$, etc. (see the comprehensive review in Ref. [4]). This doping leads to the formation of deep local or quasilocal levels in the semiconductor spectrum. When an impurity is introduced in quantities exceeding the density of other electrically active

impurities and defects, the position of the Fermi level is stabilized by these deep levels, i.e., remains invariable under additional doping by other impurities. This results in a high degree of uniformity of electrophysical crystal properties and the temporal stability of their characteristics. In particular, the energy fluctuations ΔE_c of the bottom of the conduction band do not exceed ~ 0.1 meV for the Fermi energy ~ 100 meV above the bottom of the conduction band. These fluctuations are substantially smaller than even the fluctuations in undoped alloys or in other narrow-gap semiconductor alloys like $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$, in which the fluctuations in the width of the forbidden band are normally equal to about 20 meV.

The Fermi level can be stabilized in both the allowed bands and the forbidden gap, depending on the alloy composition. Therefore, varying the alloy composition allows changing the properties of the system from ‘dielectric’ to ‘metallic’ ones. There is a possibility to vary the dark conductivity over very wide limits (from $\rho \sim 10^{-2}$ Ωcm to $\rho \sim 10^9$ Ωcm) with retention of high charge carrier mobility ($\mu \sim 10^5$ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$). The dark density of electrons or holes may vary between 10^{18} cm^{-3} and $n, p < 10^9$ cm^{-3} . The low values of n and p are realized in the semi-insulating state of alloys, when the Fermi level is stabilized inside the forbidden band. Because the impurity states that furnish the Fermi level stabilization are deep, the hydrogen-like states of impurity centers are not formed. That is why the dark currents in lead telluride-based alloys are substantially lower than in impurity photodetectors on the basis of Ge or Si for the same wavelength and temperature.

Therefore, the doping entails a highly unusual situation, whereby a heavily doped narrow-gap semiconductor with a large number of growth defects behaves as a virtually perfect semiconductor with a zero background density of charge carriers and a very high uniformity of electrophysical parameters. Such an ‘internal purification’ of the material underlies its relative cheapness in production and makes the idea of using these materials as IR detectors extremely attractive.

2.2 Persistent photoconductivity

A radically new feature of lead telluride-based alloys responsible for the occurrence of a high photoresponse is the existence of a barrier W between the impurity and band states in the energy spectrum. The existence of this barrier results in long lifetimes, $\tau \sim 10^5$ s, of nonequilibrium electrons. For the temperature $T < 20$ K (for $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$ -type alloys) or $T < 80$ K (for $\text{PbTe}(\text{Ga})$ -type alloys), the material is actually a photodetector with an internal signal integration, which eliminates the necessity of using complex external integrating circuits. Moreover, the Maxwellian relaxation time shortens in internal integration with time, which determines a faster response. Lead telluride-based alloys are also capable of operating as conventional photodetectors without internal signal integration in the temperature region $T \sim T_c$. The characteristic lifetime of nonequilibrium charge carriers may be varied over a wide range (from 10^{-4} to 10^5 s) for $T = 4.2$ K by changing the alloy composition, in addition to the possibility of controlling this parameter by changing the semiconductor temperature.

Internal signal integration emerges because the characteristic lifetime of a photoexcited free electron is much longer than the characteristic measurement time. In this situation, lengthening the exposure time leads to a linear increase in the

photoresponse because the excited electrons can hardly recombine.

2.3 Theoretical models

Several models have been proposed to account for the occurrence of slow relaxation processes in lead telluride-based alloys.

Kagan and Kikoin [5] proposed a model to account for the occurrence of slow relaxation processes at low temperatures. In the framework of the concepts described in Ref. [5], the long lifetimes of nonequilibrium charge carriers at low temperatures are related to the autolocalization barrier emerging in the change of impurity charge state. In the ionization of an impurity center, configuration restructuring of the local crystal environment of the impurity atom occurs. In this case, in order to recombine, an electron has to overcome the barrier in the configuration space associated with the need to polarize the crystal lattice in the vicinity of the impurity center in localization of the nonequilibrium electron. The authors of several subsequent papers [6–8] proposed different mechanisms of this polarization, which, however, basically differed little from each other.

It was shown in Refs [9–12] that the observed data cannot be explained in the framework of the model taking the existence of only one local state into account. In particular, this manifests itself in the photoresponse kinetics, upon turning out the illumination, exhibiting two portions — a fast (~ 1 – 10 ms) and slow (10 ms– 10^4 s, depending on the temperature) relaxation of conductivity. To describe this and several other results, the model in Ref. [5] was substantially augmented [9]. According to Zaslavitskii et al. [9], apart from the ground impurity state, which furnishes stabilization of the Fermi level and is a two-electron state, there is also a metastable one-electron impurity state. The latter state is separated by a barrier in the configuration space both from the ground two-electron local state and from the state with delocalized charge carriers in the allowed band. In this case, the localization of photoexcited charge carriers proceeds in two stages — fast and slow relaxation. The fast photoconductivity relaxation corresponds to recombination to the metastable local state, and the slow relaxation portion corresponds to recombination to the ground impurity state.

Belogorokhov et al. [13] proposed a somewhat different concept. The model in Ref. [13] relies on the fact that the actual bands in lead chalcogenides are built almost entirely of atomic p-orbitals. A group-III impurity atom replaces a lead atom in the doping of the above materials. The effect of Fermi level stabilization arises from the fact that the charge state of the impurity atom Im^{2+} , which is neutral relative to the crystal lattice, is unstable and decays according to the reaction $2\text{Im}^{2+} \rightarrow \text{Im}^+ + \text{Im}^{3+}$.

In terms of atomic orbitals, the Im^{2+} state corresponds to the s^1p^2 electron configuration, the Im^+ state corresponds to the s^2p^1 configuration, and the Im^{3+} state to the s^0p^3 configuration. The s^2p^2 configuration corresponds to the lead atom replaced by the impurity. The allowed bands in lead chalcogenides are almost entirely built of atomic p-orbitals, and therefore the electrons residing in the deep s-shell are localized for different charge states of the impurity atom, while p-electrons are delocalized.

The main idea of the model in Ref. [13] consists in the one-electron impurity state, in which only one electron is in the s-state, located (in the one-electron approximation) much higher in energy than the ground two-electron state and

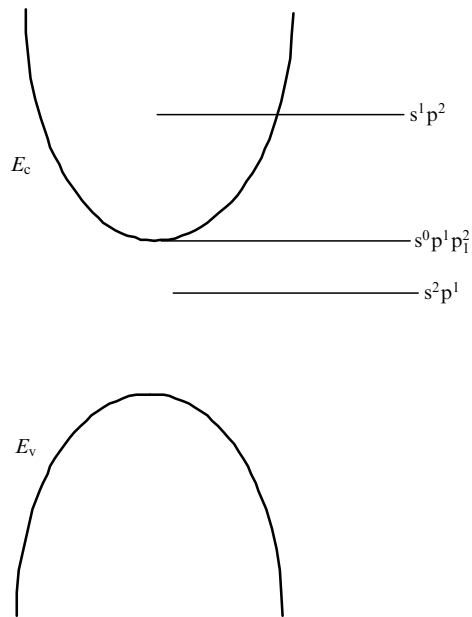


Figure 1. Model of the energy spectrum of an impurity center [13]; E_c corresponds to the edge of the conduction band; E_v is the edge of the valence band.

above the bottom of the conduction band (Fig. 1). Then, in the photoionization of the first electron from the impurity center, an energy sufficient, first, to transfer this electron to the conduction band and, second, to transfer the impurity center to the state corresponding to one s-electron localized on the impurity needs to be imparted. The single s-electron remaining at the impurity center rapidly transits to the conduction band, whose bottom lies lower in energy. Therefore, two nonequilibrium electrons emerge in the conduction band. Recombination is a one-electron process, but for the impurity center to capture an electron, the electron should be imparted an energy equal to the difference between the energies of the Fermi quasilevel and the one-electron impurity state. This energy gap is in fact the energy barrier that impedes fast electron recombination at the center.

However, the reasons why two portions of photoconductivity relaxation emerge remain unclear in the framework of the above model. A possible resolution of this contradiction is as follows. Stabilization of the Fermi level implies that a significant number of gallium atoms have an empty s-shell. Two p-electrons with oppositely directed spins may be localized in the attractive potential of this shell [14]. However, it may be that a single impurity center with an empty s-shell is unable to produce a bound state because of the large permittivity value and the low effective electron mass in PbTe. At the same time, the number of such centers is quite large, and one bound p-electron state may be formed with the simultaneous participation of a large number (up to $10^4 - 10^5$) of impurity centers having an empty s-shell [15]. Then, the fast photoconductivity relaxation may be caused by the localization of a part of the photoexcited electrons in these bound states, which are located in energy near the bottom of the conduction band and are shallow in this sense.

An estimate of the localization radius for this state yields a huge value, of the order of 100 nm. The filling of these states may occur either at low temperatures under photoexcitation or at a higher temperature in darkness. Because the electron

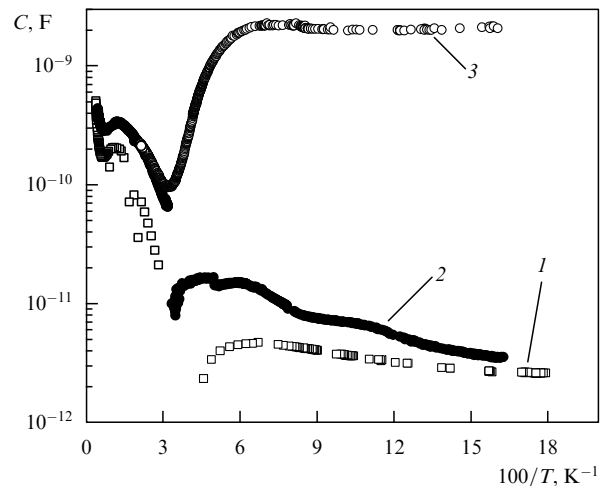


Figure 2. Temperature dependences of the capacitance C for a $\text{Pb}_{0.75}\text{Sn}_{0.25}\text{Te}(\text{In})$ sample obtained at the frequencies 1 MHz (1) and 50 kHz (2, 3). Curves 1 and 2 were measured under dark screening conditions, curve 3 was obtained under illumination by a thermal radiation source [17].

localization radius is long, the filling of metastable states is expected to entail a giant dielectric response. This effect is indeed observed [16, 17]: the effective material permittivity increases by more than two orders of magnitude on raising the temperature and under infrared illumination (Fig. 2).

2.4 Spectral characteristics

The characteristic energy values in the spectrum, which can be varied by changing the material composition, determine a high photosensitivity of the materials in a wide wavelength range (from 1 μm to several hundred micrometers). The characteristic energies of the semiconductor spectrum and, accordingly, the threshold photoresponse values are determined by the alloy composition.

Kristovskii et al. [18] reported the discovery of a photoresponse in a $\text{Pb}_{0.75}\text{Sn}_{0.25}\text{Te}(\text{In})$ film at the wavelengths 176 and 241 μm . The $\text{Pb}_{0.75}\text{Sn}_{0.25}\text{Te}(\text{In})$ film was grown by molecular-beam epitaxy on a BaF_2 substrate. The thermal activation energy of the ground impurity state was calculated from the relation $\rho \sim \exp(E_a/2kT)$ and was equal to 20 meV. The experiment was carried out at a facility with complete screening of background radiation. A blackbody with the temperature 77 K or 300 K was the source of infrared radiation. Several cooled filters formed the narrow spectral band of radiation directed onto the sample. The temporal kinetics of the current flowing through the sample was recorded under different voltages across the sample and different blackbody temperatures. The experimental data obtained for the voltage 10 mV and the blackbody temperature 300 K are depicted in Fig. 3.

Appreciable photoresponse was recorded for both wavelengths of radiation incident on the sample. We note several features of the photoconduction. First of all, the temporal current buildup kinetics is strongly nonlinear. Removing the illumination results in a fast photocurrent decay with a subsequent slow relaxation to the dark value. However, turning on the illumination shortly after its removal has the effect that the photocurrent rapidly, in a time comparable to the 'fast' relaxation time, builds up to the value that existed prior to the removal of illumination, with the subsequent

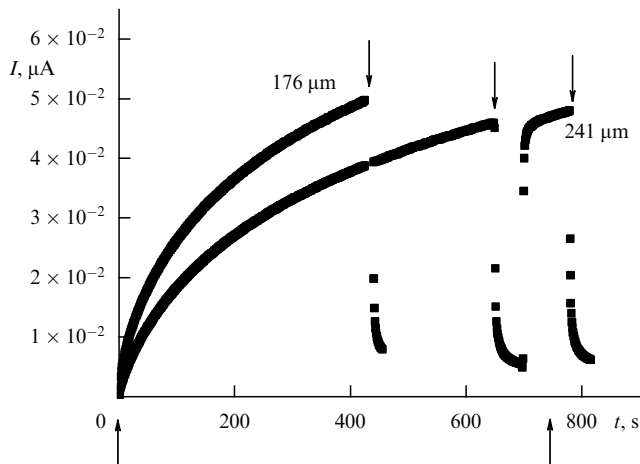


Figure 3. Photocurrent buildup and decline kinetics for the voltage 10 mV across the sample and different wavelengths of exciting radiation, which are indicated in the figure. Arrows indicate the moments at which infrared illumination was engaged and turned off [18].

resumption of the previous, relatively slow, dynamics of photocurrent buildup. Clearly, the fast and slow relaxation processes are essentially different in nature.

The second significant point is as follows. The photon energy corresponding to radiation wavelengths 176 and 241 μm is substantially lower than the thermal activation energy of the ground impurity state. Therefore, the results in Ref. [18] are direct evidence that the persistent photoconductivity in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$ may be caused by the photoexcitation of metastable impurity states. The threshold energy for the optical excitation of these states is very low. The wavelength of the corresponding photon is longer than 241 μm , which is the greatest value for the photoelectric threshold in nonthermal radiation detectors. The photosensitivity cutoff for the materials under consideration is supposedly in a substantially longer wavelength region. It is not unlikely that the working range of $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$ -based photodetectors spans the entire terahertz range. On the other hand, the nature of the emergence of two portions of free-electron generation and recombination remains unclear in this case, because the ground impurity state should be ‘excluded from play’ because the exciting photon energy is low.

2.5 Radiometric characteristics

Internal signal integration is an advantage only when a possibility to nullify the stored photosignal exists. We have developed an efficient technique of quenching the persistent photoconductivity in lead telluride-based alloys. Microwave pulses (200–400 MHz) applied to the sample were found to completely quench the persistent photoconductivity (PC) in 10^{-5} – 10^{-6} s [19]. This signifies that the possibility exists of implementing the regime of periodic storage of a photosignal followed by its fast nullification.

Furthermore, under a certain regime of quenching of the residual PC by microwave pulses, the quantum efficiency of the material increases to ~ 100 upon exposure to the quenching pulse [19], i.e., every incident photon produces at least 100 free electrons in the conduction band. This effect is related to the specific character of impurity states. It furnishes a high current responsivity ($10^9 \text{ A}^2 \text{ W}^{-1}$), which was recorded for a single photodetector based on $\text{Pb}_{0.75}\text{Sn}_{0.25}\text{Te}(\text{In})$

operating in the above regime with the readout frequency 10 Hz. In this test, it has been possible to record the radiation power 10^{16} W at the wavelength 18 μm (determined by a mesh filter), even though the measuring technique in use did not enable measuring a current below 10^{-7} A . The above parameters correspond to recording about 10^3 photons per 1 cm^2 in 1 s [20].

2.6 ‘Continuous’ focal-plane array

It was established that local illumination of a part of a photodetector sample results in a local excitation of non-equilibrium charge carriers, which do not diffuse into the unilluminated domains at low temperatures. Therefore, there is a one-to-one correspondence between the radiation intensity distribution over the sample surface and the distribution of the nonequilibrium charge carrier density [21]. In other words, the alloys under consideration allow making photodetector arrays in which each effective element effects internal signal accumulation. The dimension of the shaded interval between the elements of the array required to eliminate their interference is theoretically estimated at about 10 μm . This is the characteristic dimension corresponding to the spatial distribution of the free charge carriers at the edge of a ‘light spot.’ Our experiments have shown that this characteristic dimension is below 200 μm . The geometric noise in arrays of this type is minimized due to the high degree of spatial uniformity of the electrophysical material parameters. The idea of a simple technique for information readout from such an array has been elaborated, which so far has not been realized because of technical problems.

2.7 Radiation hardness

The position of the Fermi level in crystals depends neither on the impurity density fluctuations nor on the fluctuations of intrinsic lattice defects, including radiation-induced ones, over a wide range. The possibility of dividing the impurity states into states with different charges underlies the high radiation hardness of the parameters of lead telluride-based alloys, which is at least four orders of magnitude higher than in the known analogues (with the exception of blocked-impurity-band structures). Exposure of the alloys to fast 6 MeV electrons in an accelerator showed that the photoconduction parameters in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$ and $\text{PbTe}(\text{Ga})$ (with the In, Ga content ~ 0.2 – 0.4 at. %) remained invariable (to within $\sim 5\%$) under electron fluxes Φ up to 10^{18} cm^{-2} [22].

3. Conclusions

Thus, a unique combination of physical properties makes lead telluride-based alloys highly attractive for constructing extremely sensitive focal-plane arrays for the photodetection of terahertz radiation. The $\text{Pb}_{1-x}\text{Sn}_x\text{Te}(\text{In})$ photodetectors exhibit the following distinguishing features that permit them to successfully compete with the existing analogues:

- internal integration of the incident light flux;
- the possibility of efficient and rapid quenching of the accumulated signal;
- microwave stimulation of quantum efficiency up to 10^2 ;
- the feasibility of realizing a ‘continuous’ focal-plane array;
- the possibility of realizing a simple readout method;
- a high radiation hardness.

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Modulation gamma-resonance spectroscopy

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1. Introduction

Thanks to the Mössbauer effect, the gamma range of electromagnetic waves is being mastered more and more in both fundamental and technological areas and is beginning to compete, in terms of its complexity of problems and exactness of measurements, with the optical range with its laser light sources [1, 2]. The standard Mössbauer spectroscopy is being replaced with gamma optics, an important part of which is the modulation gamma-resonance spectroscopy. The idea behind this spectroscopy is the possibility of controlling dynamical processes in matter by modulating a gamma resonance via external varying magnetic fields with frequencies exceeding the reciprocal lifetimes of excited states of Mössbauer nuclei

(i.e., in the megahertz frequency range). This frequency range of dynamical processes covers the range of high-frequency fields of nuclear magnetic resonance (NMR), quadrupole, ferromagnetic, electron paramagnetic, and optical resonances. These perturbations are detected by measuring changes in gamma-resonance responses, which manifest themselves in distortions of the Mössbauer spectrum, such as shifts and the broadening of lines, and the emergence of satellite lines, the collapse of the hyperfine structure of lines under rapid magnetization reversal of ferromagnets, as well as in the onset of quantum beats caused by the interference of nuclear states.

But the intensities of the existing sources of gamma radiation (natural or synchrotron) are much lower than the values needed for producing inverse population in isomeric nuclear states. One encouraging fact here is the recently discovered nuclear excitation of ^{197}Au caused by an electron transition stimulated by the photoionization of the K-shell by X-ray radiation from a synchrotron source [3].

The modern method of theoretical analysis of gamma-resonance processes has had a great impact on the development of modulation gamma-resonance spectroscopy. The method combines the solution of the Maxwell equations in a medium and of the equations for the nuclear density matrix and showed its usefulness in examining processes accompanying the transmission of gamma radiation [4, 5]. However, to describe gamma-resonance scattering, I used a method for solving equations for the density matrix with second quantization of the gamma-radiation field [6, 7]. This approach has made it possible to solve problems related to the effect of alternating fields and relaxation on gamma resonances.

2. Ultrasonic modulation of gamma radiation

Already in their first Mössbauer experiments, Ruby and Bolef [8] recorded frequency modulation of gamma radiation with satellites whose distances from the spectral lines were found to be integer multiples of the ultrasonic frequency of the vibrating source or absorber of gamma radiation. However, the situation with a thick vibrating Mössbauer absorber was still unclear. The theory developed in Refs [1, 9, 10] showed that when the absorber or source vibrates with a frequency Ω , the intensity of gamma radiation is given by

$$P = \sum_{n=-\infty}^{\infty} J_n^2(k_1 a) \text{Tr} \left\{ \exp(ik_1 z \hat{b}_n) \hat{\chi} \exp(-ik_1 z \hat{b}_n^\dagger) \right\}, \quad (1)$$

where k_1 is the wave number of the gamma radiation, a is the amplitude of the ultrasonic vibrations, z is the thickness of the absorber, J_n is the n th Bessel function, \hat{b}_n is the tensor refractive index for a gamma wave with a shift in the gamma-radiation frequency by $n\Omega$, with Ω being the ultrasonic frequency, $\hat{\chi}$ is the polarization density matrix of the incident gamma radiation, and \dagger stands for Hermitian conjugation.

If the source and absorber vibrate simultaneously with the same frequency and amplitude, the argument $k_1 a$ of the Bessel function can be replaced with the expression

$$w = 2k_1 a \sin \left\{ \frac{1}{2} \left[\varphi_1 - \varphi_2 + \frac{\Omega}{2} (z_2 - z_1) \right] \right\}, \quad (2)$$

where $z_2 - z_1$ is the distance between the source and absorber and $\varphi_2 - \varphi_1$ is the difference between their ultrasonic phases.