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On the Nature of "Coherent Artifact"

M. V. Lebedev[a], O. V. Misochko[a], T. Dekorsy[b], and N. Georgiev[b]

[a]*Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, Moscow oblast, 142432 Russia*

[b]*Institute for Ion Beam Physics and Materials Research, Forschungszentrum Rossendorf, P.O. Box 510119, D-01314 Dresden, Germany*

e-mail: lebedev@issp.ac.ru

Abstract—The coherent interaction of femtosecond laser pulses in the pump–probe regime has been experimentally studied in the time domain by monitoring light reflection from a tellurium single crystal. The optical response of the probed medium exhibits periodic variations at a frequency equal to that of the exciting laser radiation. Experimental dependences of the observed "coherent artifact" on the pump/probe intensity ratio, the number of accumulated pulses, and the mutual orientation of the polarization vectors of electromagnetic fields and the crystallographic axes are well described by the proposed phenomenological model.

1. INTRODUCTION

Recent decades have seen considerably growing interest in the study of response of condensed media to the action of ultrashort laser pulses, in which the energy is concentrated within a short time, small space, and narrow spectral intervals [1]. This research activity has been stimulated to a considerable extent by the progress in laser technologies in reducing the laser pulse duration down to the subpicosecond range, thus opening the way to real-time investigation of many processes related to the properties of solids.

One of the main approaches to investigations of fast dynamics is based on the pump–probe method. According to this approach, a medium is first excited by a short powerful laser pulse and then the optical properties of this excited medium are studied using much weaker probing pulses, typically obtained by separating part of the main laser beam (with the aid of a beam splitter) and attenuating it to a considerable degree. The first (pumping) pulse prepares a certain nonequilibrium state of the medium, whereas the second (probing) pulse of very low intensity (practically incapable of modifying the state of the medium) is used to study the process of system relaxation to the equilibrium state. However, these notions are completely inconsistent with the experimental situation in the region of zero delay of the probing pulse relative to the pumping pulse, where the two pulses are superimposed in the sample. In this case, we can no longer consider one pulse as pumping and the other merely as probing, since the excitation of the medium substantially depends on the coherent superposition of pulses, and the smallness of the probing pulse by no means implies that its effect is negligibly small.

Experiments in the time domain in the region of zero delays frequently show unusual and poorly reproduced deviations from the curves measured at greater delays and then extrapolated to a zero delay (Fig. 1). In the literature devoted to coherent phonons [2, 3], all these phenomena have been given the name "coherent artifact." This term (in our opinion, rather inadequate) has to reflect, first, the connection of the observed effects to coherent superposition of laser pulses and, second, the irregular and poorly controlled character of these phenomena. Phonons excited by ultrashort laser pulses are usually called coherent, since the exciting pulse duration is shorter than the period of lattice oscillations. The excitation of oscillations in nontransparent media can

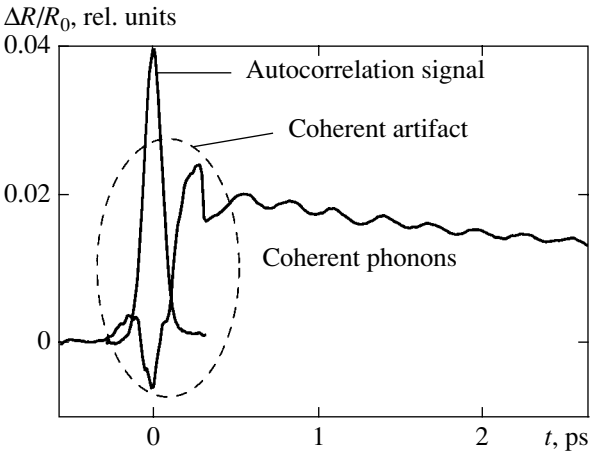

Fig. 1. Normalized time-resolved differential reflection signal $\Delta R/R_0$ from tellurium single crystal measured at room temperature for a time resolution of 6 fs. Laser pulse intensity, 0.1 mJ/cm^2.

be successfully described using phenomenological models based on the displacive excitation of coherent phonons and a mechanism close to the inelastic light scattering [2, 3]. It is a common practice to minimize the coherent artifact signal using orthogonal polarizations of the pumping and probing radiation, nondegenerate pump–probe scheme, etc. [2, 3]. There are several points of view on the nature of the coherent artifact. The formation of this signal is usually related to nonlinear optical effects (for collinear polarizations of the pumping and probing radiation) of the four-wave mixing [4]. According to this, the pumping pulse is scattered toward the probing pulse as a result of the phase grating formation.

This paper presents the results of thorough experimental and theoretical investigation of the coherent artifact in the time domain on a subfemtosecond resolution level. It is experimentally demonstrated that the "artifact" observed in the case of orthogonal polarizations of the pumping and probing radiation possesses a regular periodic structure, is perfectly reproduced, and can be described within the framework of a simple phenomenological model of the light–medium interaction. This analysis opens ways to investigating the response of a medium within ultimately short periods of time and, in addition, establishes the natural zero point for time count in the pump–probe method.

2. DESCRIPTION OF EXPERIMENT

Figure 2 shows a schematic diagram of the experimental arrangement. The experiments were performed with a tunable femtosecond Ti-sapphire laser of the Tsunami type (Spectra Physics) pumped at 532 nm from a Millenia Xs laser (Spectra Physics). The laser pulse shape was optimized using a double prism compressor and monitored with the aid of an autocorrelator. By shifting one of the two prisms, it was possible to adjust the phase modulation (chirp) so as to provide the optimum compensation of the group velocity dispersion in beam splitters, lenses, and other transparent elements in the pathways of both pumping and probing beams, thus ensuring the minimum pulse duration in the sample plane. This duration, as determined by monitoring the nonlinear transmission of a GaP crystal, was 90 fs. The spectral composition of the pulse was determined using a monochromator with a multichannel detection system. The pulse shape and spectrum were displayed in real time on the autocorrelator and computer displays. This setup allowed the pulse parameters to be continuously monitored and adjusted.

The measurements were performed using a fast scan system (AIXscan) comprising a shaker (angular retroreflector mounted on an electromagnetic vibrator driven by a high-precision oscillator), a delay line with a step motor, high-frequency twelve-digit analog-to-

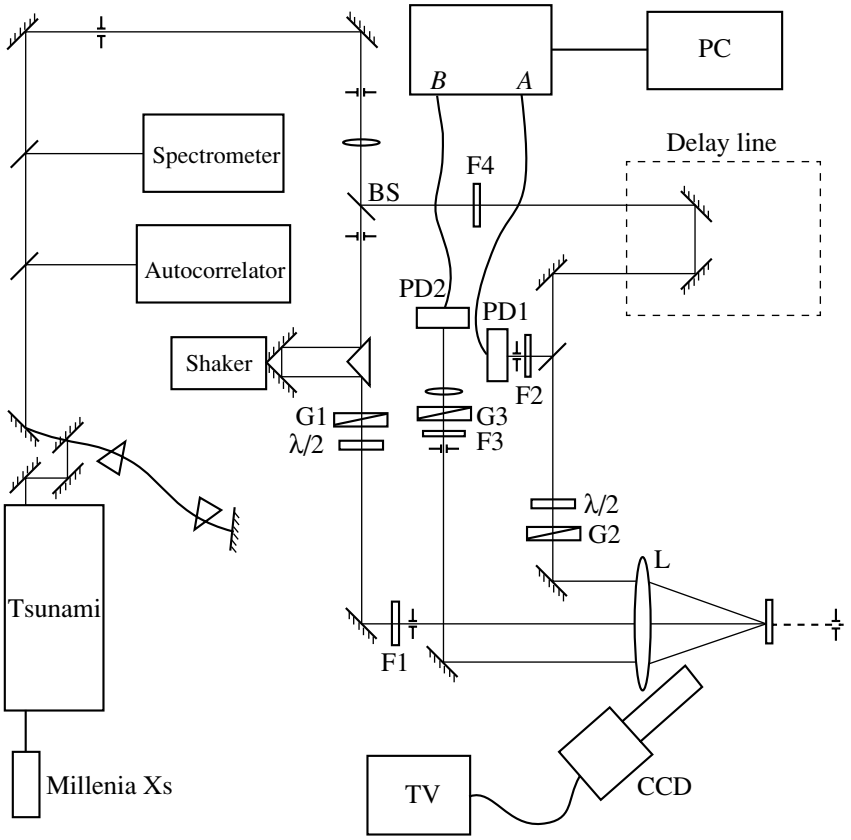

Fig. 2. Schematic diagram of the experimental arrangement (see the text for explanations).

digital amplitude converter (operating at 0.5 and 1.0 MHz), and a PC with special control software. The shaker made it possible to scan over and display a range of delay times with the initial value set by a delay line. The signal was digitized only during the forward run of the shaker. The measurements were performed at room temperature.

A sample crystal was placed at the focus of lens L (Fig. 2) with a focal distance of 90 mm, through which the pumping and probing pulses were transmitted parallel to the optical axis. The parallelism of laser beams entering the lens, which is a necessary prerequisite for the optimum spatial superposition of pumping and probing beams, was thoroughly checked at the system adjustment stage. The two beams were focused by the lens into one spot on the crystal surface. The imaging system, comprising a zoom objective, a CCD camera, and a TV monitor provided sharp beam focusing on the crystal surface and controlled matching of the pumping and probing beam spots. The pumping pulse was transmitted to the crystal via shaker, followed by a Glan prism G1 (eliminating parasitic polarization), a half-wave plate (rotating the polarization plate), and a neutral filter F1 of variable optical density (adjusting the required pump intensity).

The probing beam was obtained by separating a small part of the pumping beam with the aid of beam splitter BS. This beam passed through a delay line, a half-wave plate, and a Glan prism G2. The latter Glan prism was used to control polarization of the probing beam, while the intensity of this beam was adjusted by rotating the half-wave plate (changing the projection of the electric field vector onto the given polarization direction). The beams of pumping and probing pulses were separated by means of spatial filtration performed by diaphragm F3 mounted in front of a neutral light filter. Oscillations of the vibrator in the shaker ensured scanning of a certain interval of the time delay between pumping and probing pulses. The data were digitized in the high-frequency analog-to-digital amplitude converter and stored in a computer memory. The delay line was used for a rough leveling of the optical paths of the pumping and probing pulses and for calibration of the shaker sweep.

In order to increase the data accumulation rate and expand the working range of delay times provided by the shaker, it is necessary to increase the amplitude of the retroreflector and use a relatively high working frequency of the shaker. Evidently, just the opposite requirements have to be satisfied for reaching maximum time resolution of the system: this corresponds to the minimum velocity of the angular retroreflector that can be achieved by decreasing the amplitude and frequency of shaker vibrations. In our experiments, the shaker was operated with amplitude two to three times smaller than maximum and at a frequency of 40 Hz. The frequency was stable to within $\pm 10^{-3}$ Hz, which allowed the delay times in a range of 4 ps to be digitized at a step of 0.1 fs per point. The error caused by the frequency instability was comparable with the discreteness of digitization. A decrease in the range of scanned delay times and the frequency of shaker operation could increase the accuracy of digitization (under otherwise equal conditions). However, in our case this would detrimentally influence the conditions of operation of the program of automated time scale calibration. This program set a series of fixed time delays between the pumping and probing pulses, compared these delays to the shift of a pulse reflected from the shaker, and determined the coefficients of time scale calibration, thus taking into account nonlinearity of the sweep. The minimum parameters at which this calibration program could successfully operate corresponded to a shaker frequency of 34.76 Hz and a calibration range of about 4 ps. In a standard probe–pulse experiment with the response signal measured using a lock-in detector, the discreteness of digitization is determined by the delay line step and usually does not exceed 6.6 fs per point [2, 3].

We have measured the relaxation of changes in the reflection coefficient (or in the transmission coefficient, in the case of a transparent GaP crystal used for the system calibration) caused by the pumping pulse. Since these changes are relatively small, we have used a special differential technique in order to increase the accuracy of measurements. According to this, a part of the probing beam was separated (before striking the sample crystal) by the beam splitter and directed to photodiode PD1 (Fig. 2). The probing beam reflected from the crystal was detected by photodiode PD2. The signals from both detectors were fed to the inputs (A and B) of a differential amplifier, where the difference $A–B$ was amplified and measured using a high-frequency analog-to-digital converter of the AIXscan system. The A and B signal channels were thoroughly balanced (whereby the signals from photodiodes were equilibrated using a neutral filter F2 with variable optical density mounted in front of photodiode PD1) in the absence of pumping pulses. As a result, we monitored the signal proportional to a change in the reflection coefficient, which was on a level from 10^{-3} to 10^{-5} of the absolute value of this coefficient. The differential amplifier was equipped with a built-in frequency filter, which allowed the working signal to be measured in a preset frequency band and thus increased the signal to noise ratio. For the study of coherent phonons, it was sufficient to use a frequency band from 1 Hz to 3 kHz, where virtually no distortions were introduced into the corresponding oscillations (with a period of about 300 fs). In investigations of the coherent artifact, the period of signal variation was about 3 fs and this required the working frequency band of the amplifier to be increased (in our measurements, the frequency band was typically from 1 Hz to 300 kHz).

All measurements were performed for a basal plane of a tellurium single crystal crystallized at room

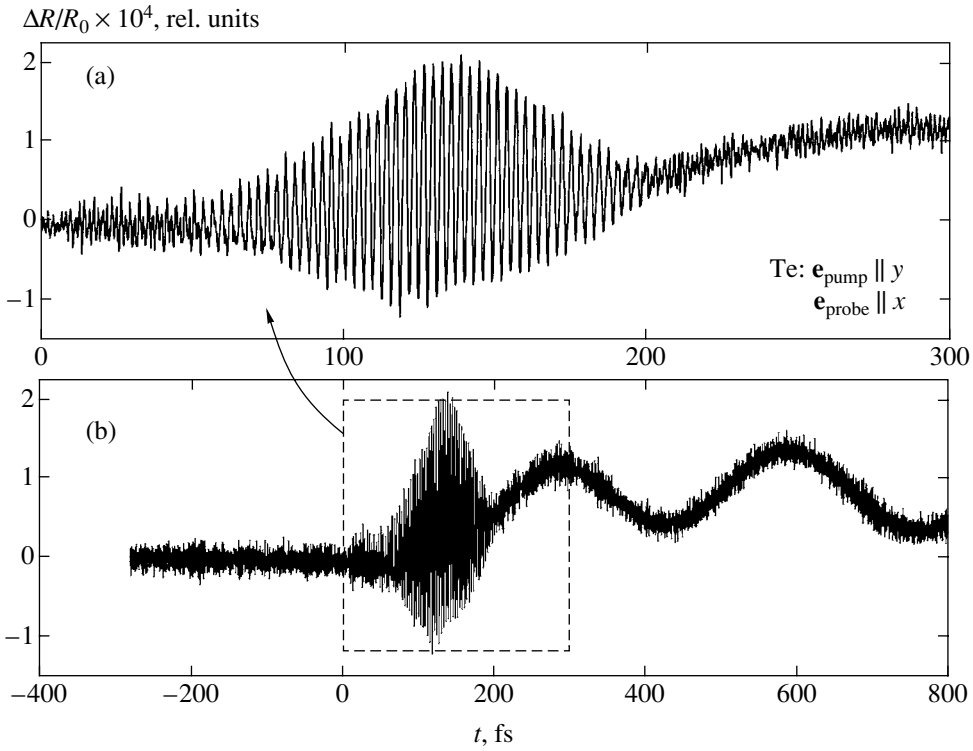

Fig. 3. (a) The shape of a coherent artifact signal with well-pronounced regular structure measured at a high resolution; (b) the coherent artifact on the background of first two oscillations of the reflection coefficient, related to the excitation of fully symmetric (A_1) coherent phonons in a tellurium single crystal. Pumping laser beam power, 37 mW; probing laser beam power, 3.2 mW; number of accumulated runs, 100; polarization: $\mathbf{e}_{pump} \parallel y$, $\mathbf{e}_{probe} \parallel x$.

temperature in a hexagonal lattice belonging to space group D_3^6.

3. EXPERIMENTAL RESULTS

In the measurements of coherent phonons, the coherent artifact is usually manifested by a special region in the initial part of relaxation curves oscillating at a frequency of the phonon mode [2, 3]. This region is clearly seen in Fig. 3b together with two initial oscillations of the reflection coefficient caused by excitation of the coherent fully symmetric phonons in the tellurium crystal [5]. A comparison of Figs. 1 and 3 shows that an increase in the time resolution reveals a regular periodic structure of the artifact (see Fig. 3a). It is seen at first glance that the rapid oscillations are performed not relative to the zero line, but relative to a certain slowly varying (on the given time scale) function. This function can be readily separated from the oscillating curve by averaging over the oscillation period. The results of such separation for various pump/probe intensity ratios are presented in Fig. 4. At equal intensities of the pumping and probing beams, rapid oscillations are virtually symmetric relative to the zero level. If the pumping pulse intensity is significantly higher than that of the probing pulse, the artifact exhibits a slowly varying positive additive to the reflection coefficient. The magnitude of this additive also significantly depends on the orientation of light polarization relative to the crystallographic axes. The ratio of the slowly varying and rapidly oscillating components of the response also changes when the beams change their roles, whereby the probing beam is used for pumping and vice versa (in experiment, this is achieved by exchanging photodiodes between the probing and pumping beams, while retaining the same beam polarizations).

Figure 5 shows a change in the artifact in the course of a gradual increase in the probing beam intensity in the case when the polarizations of both beams were rotated by 90° as compared to their orientations in the experiment presented in Fig. 4. As can be seen, a change in the ratio of beam intensities leads to a change in the ratio of slowly varying and rapidly oscillating components of the response (analogous to that shown in Fig. 4). However, in this geometry, the contribution of the slowly varying component is greater than in the case presented in Fig. 4. This was specially checked for the same number of accumulated data, which is an important circumstance since it was found that the shape of the artifact depends on the number of accumulated runs. This dependence is illustrated in Fig. 6, which shows that an increase in this number is accompanied by a decrease in the relative contribution of the oscillating part of the response. The effect is related to

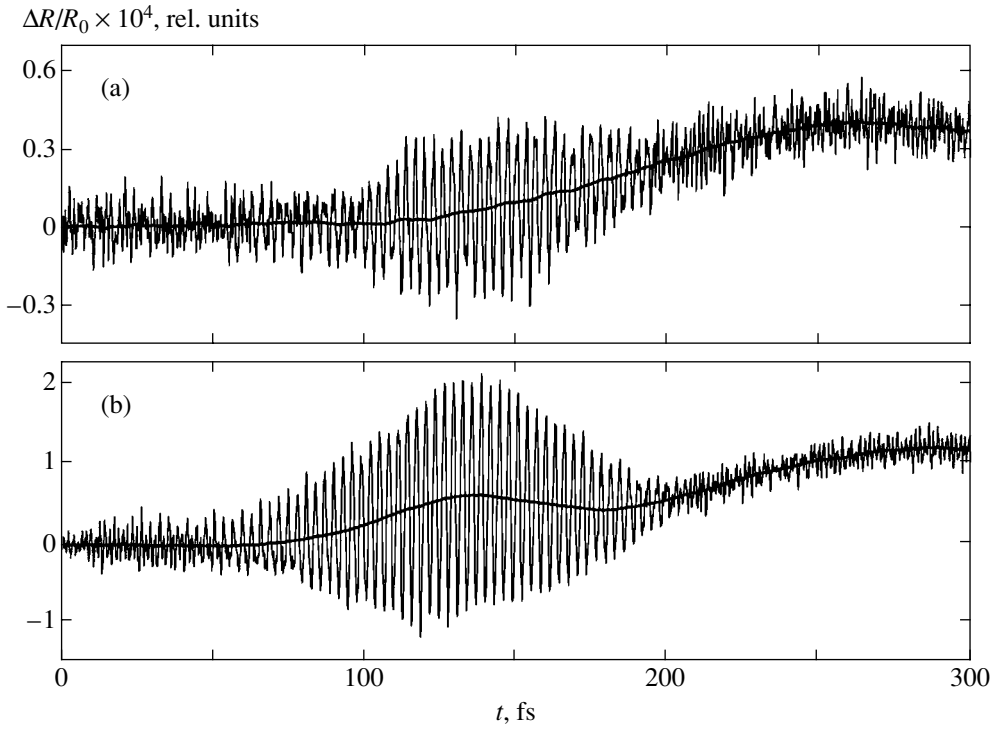

Fig. 4. The influence of the ratio of the pumping and probing beam intensities on the shape of the coherent artifact. An increase in the pumping beam intensity leads to the appearance of a slowly varying positive component. Slowly varying background curves were obtained by smoothening over the period of the artifact. Pump/probe intensity ratio (mW): (a) 7.2/7; (b) 37/3.2; number of accumulated runs, 1000; polarization: $\mathbf{e}_{\text{pump}} \parallel y$, $\mathbf{e}_{\text{probe}} \parallel x$.

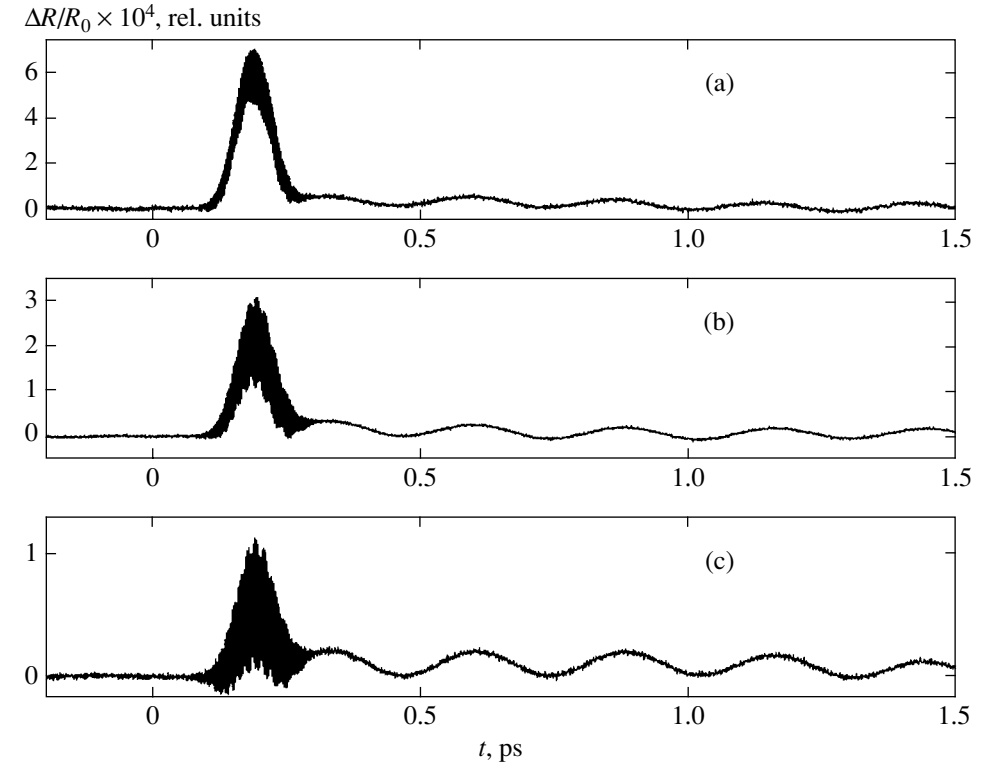

Fig. 5. The influence of the ratio of the pumping and probing beam intensities on the shape of the coherent artifact measured for a laser beam polarization other than that in Fig. 4: $\mathbf{e}_{\text{pump}} \parallel x$, $\mathbf{e}_{\text{probe}} \parallel y$; number of accumulated runs, 10000; pumping laser beam power, 28.4 mW; probing laser beam power (mW): (a) 7.2; (b) 18.2; (c) 28.7.

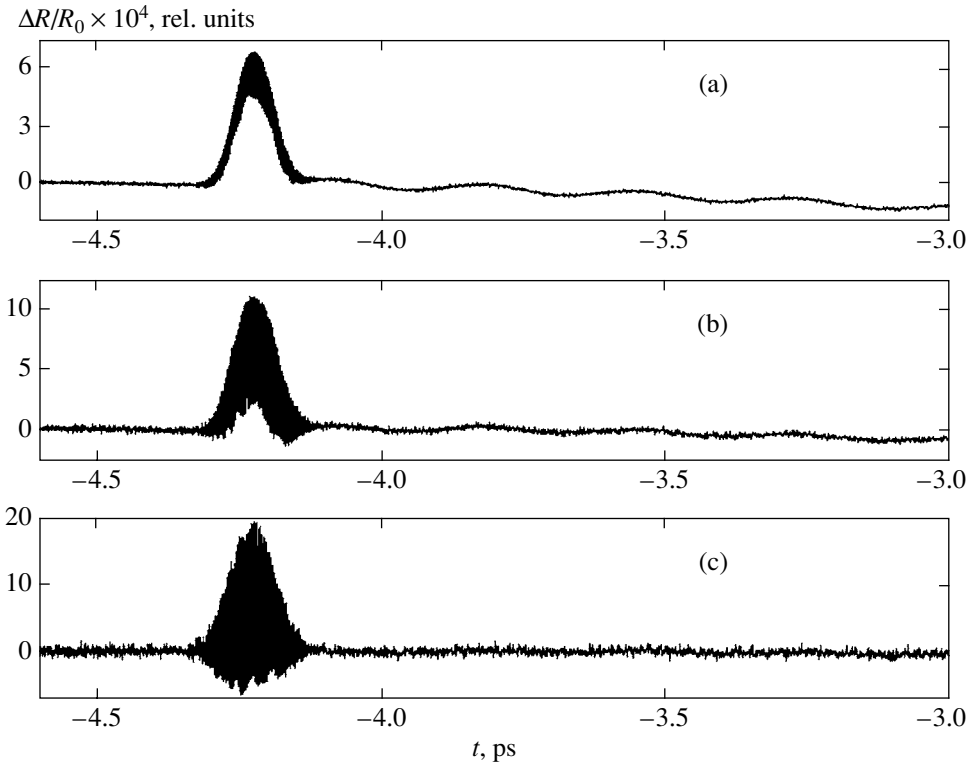

Fig. 6. Variation of the coherent artifact depending on the number of accumulated runs: (a) 10000; (b) 1000; (c) 100. An increase in this number leads to a decrease in the relative contribution of the rapidly oscillating component. Polarization: $\mathbf{e}_{\text{pump}} \parallel x$, $\mathbf{e}_{\text{probe}} \parallel y$; pumping laser beam power, 28.4 mW; probing laser beam power, 7.2 mW.

averaging of the oscillating curve with time as a result of uncontrolled phase fluctuations appearing, probably, because of imperfection (the lack of interferometric precision) of the experimental setup.

Figure 7 shows the results of two sequential experiments performed using the same number of accumulated runs and equal values of all controlled experimental parameters. As can be seen, one of the curves exhibits clearly pronounced oscillations with a single period of 2.67 fs. The amplitude of these oscillations in the other curve is significantly decreased and additional small peaks are observed between the main maxima. The change is most probably caused by a spontaneous fluctuation in the phase of shaker oscillations. The period of the oscillating component in the response (2.67 fs) coincides to within 4.8% with the period of the carrier wave of the light pulse (2.80 fs). The latter is determined to within 1.3% (for an excitation laser wavelength of 841 ± 11 nm).

The above experimental data elucidate the reasons for which the artifact was previously observed in the form of an irregular and poorly reproducible behavior of the relaxation curves in the region of zero delay times. First, the standard time resolution (not better than 6.6 fs) did not allow detecting oscillations with a period shorter than 3.0 fs. Second, the goal of increasing the signal to noise ratio in the investigation of features of the relaxation kinetics required using large signal accumulation times, selecting sufficiently high shaker frequency, and employing optimum signal filtration regimes. All these factors lead to effective averaging of the oscillating component of the artifact and strongly distort its shape, the more so that it is highly sensitive to small uncontrollable phase fluctuations. The sensitivity of the artifact to intensities of the beams and their polarization relative to the crystal axis still complicates the pattern.

All the experimental results considered above referred to differential measurements of the relaxation of a change in the reflection coefficient of tellurium single crystal exposed to a high-power femtosecond laser pulse. It should be emphasized that very similar results were obtained when the metal crystal was replaced by a crystal of high-temperature superconductor or manganite. However, for the sake of brevity, the consideration is restricted to the analysis of data obtained for tellurium.

In the experiments with all nontransparent (strongly absorbing) crystals, the time sweep of the shaker was calibrated using a standard procedure, whereby a sample crystal in the lens focus was replaced by a transparent (in the laser wavelength range employed) crystal of GaP (an indirect-band semiconductor with a bandgap width of 2.8 eV at the Γ point; $n = 3.24$ at 850 nm [6]). The photodiode was moved from the reflected beam to the transmitted beam and the same differential tech-

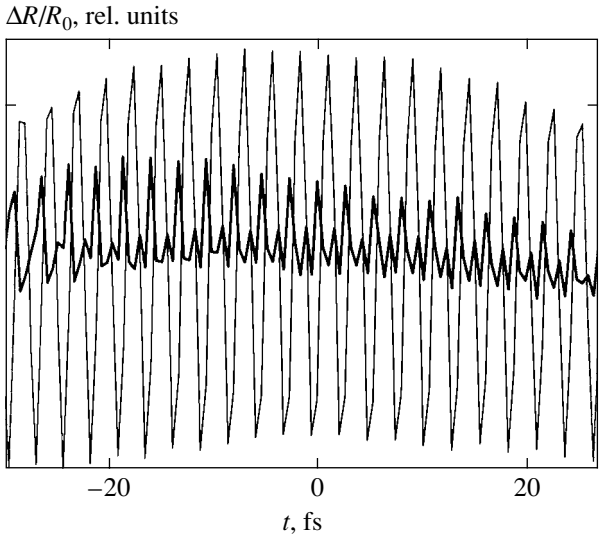

Fig. 7. Two time series of the coherent artifact recorded for the same fixed experimental parameters. A sharp decrease in the magnitude of oscillations and the appearance of additional peaks in one of the curves is probably related to a spontaneous fluctuation in the phase of shaker vibrations. Number of accumulated runs, 300; pumping laser beam power, 28 mW; probing laser beam power, 18 mW; polarization: $\mathbf{e}_{\text{pump}} \parallel x$, $\mathbf{e}_{\text{probe}} \parallel y$.

nique was used to measure a change in the transmission of the probing beam after excitation of the GaP crystal by the pumping pulse. It is commonly accepted that (i) a decrease in the probing beam transmission observed at the moment of superposition of the pumping and probing pulses is related to the two-photon absorption and (ii) the shape of the detected signal is proportional to the autocorrelation function of laser pulse intensities. This provides a simple and convenient method of optimization of the excitation pulse duration immediately at the site where the sample will be placed, while rather large signal amplitude makes it possible to use this method for calibration of the shaker sweep. The two-photon absorption is insensitive to the phase of the light wave. The interference between the pumping and probing beams was excluded by using orthogonal polarizations of the two beams. Despite this, our study of the nonlinear transmission of a GaP crystal at a high time resolution revealed the presence of oscillating component in the response signal, which had a shape quite similar to that presented in Fig. 4, with a period close to that of the carrier wave. Further investigation showed that, under certain conditions, it is possible to observe a change in the sign of the differential transmission, whereby the pumping pulse induces an increase (rather than a decrease) in transmission of the crystal. Moreover, the response of a GaP crystal has proved to be sensitive not only to the phase difference between the pumping and probing beams, but also to the phase modulation of the laser pulse that could be varied with the aid of a prism compressor. All these features are difficult to explain within the framework of a simple two-photon absorption model, although this process can still play a certain role in the phenomena under consideration.

Consistent theoretical analysis of the propagation of a high-power femtosecond laser pulse in a nonlinear medium is a highly complicated problem and there are a huge number of publications on this subject. As for the task of description of the nonlinear reflection of a short high-power laser pulse, this problem admits a simple solution within the framework of a simple phenomenological model considered below.

4. DISCUSSION OF RESULTS

It should be noted that the observed phenomena cannot be explained by simple interference between the pumping and probing beams. Indeed, polarizations of the two beams in all experiments were orthogonal and the reflected signal was detected at a site distant from the region of beam overlap, so that no superposition of light waves took place in the detector.

The coherent phonons excited in a tellurium crystal by the pumping laser pulse are manifested in the experimental curves by a periodic time variation of the reflection coefficient of the sample (Fig. 3). We naturally assumed that the observed periodic temporal structure of the coherent artifact is also related to a periodic time variation of the reflection coefficient under the action of electromagnetic field of the pumping pulse. Based on this assumption, it is possible to provide a phenomenological description of the properties of a coherent artifact observed in experiment.

4.1. Phenomenological Model

Let us describe a laser pulse at the crystal surface by a wave packet,

$$\Phi(t) = A(t)\exp[-i(\omega_0 t + \psi(t))], \qquad (1)$$

where ω_0 is the carrier frequency and $A(t)$ and $\psi(t)$ are slowly varying amplitude and phase. Since only a surface layer of the crystal with a thickness much smaller than the wavelength contributes to the reflection coefficient, we may decline from analysis of the character of light propagation in the crystal and solve an essentially local problem. This circumstance makes possible a simple description of the experiment. In order to simplify the formulas, we will not take into account the vector character of electromagnetic fields. If necessary, the form of $A(t)$ and $\psi(t)$ functions can be specified. These functions are quite rigidly set by the condition of femtosecond laser pulse generation using synchronized modes of a Ti-sapphire laser. In particular, the carrier frequency is a higher harmonic of the fundamental mode of the laser cavity and the amplitude $A(t)$ is directly related to the number of synchronized modes. The phase $\psi(t)$ includes the linear and quadratic terms, the latter determining the phase modulation (chirp) of

the pulse. The carrier frequency and the spectral width of a pulse can be directly measured in experiment. The chirp determining (together with the slowly varying amplitude) the spectral width can be varied with the aid of the prism compressor.

We assume that a laser pulse produces a change in the coefficient of reflection of the crystal surface, which varies with time as

$$R(t) = R_0 + r(t), \tag{2}$$

where R_0 is the background reflection coefficient. Accordingly, the amplitude of the reflected pulse can be written as

$$\begin{aligned}\tilde{\Phi}(t) &= \Phi(t)R(t)\\ &= R(t)A(t)\exp[-i(\omega_0 t + \psi(t))].\end{aligned} \tag{3}$$

Let us express the variable increment of the reflection coefficient as

$$r(t) = a(\mathbf{e})\Phi(t), \tag{4}$$

where $a(\mathbf{e})$ is a coefficient taking into account the dependence of the increment on the orientation of the polarization of light relative to the crystallographic axes. In the general case, this coefficient is a complex quantity whose modulus determines the magnitude of the increment and the argument reflects the possible phase shift of the laser-induced change in the reflection coefficient relative to the phase of the electric field vector of the pulse:

$$a(\mathbf{e}) = |a|e^{i\varphi}. \tag{5}$$

Under the conditions of superposition of the pumping and probing laser pulses, the increment of the reflection coefficient can be written as

$$r(t) = a_{\text{pump}}\Phi_{\text{pump}}(t+\tau) + a_{\text{probe}}\Phi_{\text{probe}}(t), \tag{6}$$

where τ is the time delay between the pumping (Φ_{pump}) and probing (Φ_{probe}) pulses. Then, the amplitude of the reflected probing pulse is

$$\begin{aligned}\tilde{\Phi}_{\text{probe}}(t) &= \Phi_{\text{probe}}(t)\\ &\times (R_0 + a_{\text{pump}}\Phi_{\text{pump}}(t+\tau) + a_{\text{probe}}\Phi_{\text{probe}}(t)).\end{aligned} \tag{7}$$

Since the photodiodes measure the light intensity averaged over the response time, the measured differential signal S can be determined by multiplying the amplitude (7) by its complex conjugate, averaging the product over the diode time constant, and rejecting the terms not containing Φ_{pump} (which are compensated in a balanced differential scheme). As a result, we obtain

$$\begin{aligned}S \propto{}& \langle a_{\text{pump}}^2 A_{\text{pump}}^2(t+\tau)A_{\text{probe}}^2(t)\rangle\\ &+ 2\langle a_{\text{pump}}A_{\text{pump}}(t+\tau)a_{\text{probe}}A_{\text{probe}}^3(t)\\ &\times \cos(\omega_0\tau + \Delta\psi + \Delta\varphi)\rangle,\end{aligned} \tag{8}$$

where the angle brackets denote averaging with respect to time, the a_i and A_i are assumed to be positive (all signs are included into the phase factors), and the terms $\Delta\psi$ and $\Delta\varphi$ are given by the formulas

$$\begin{aligned}\Delta\psi &= \psi_{\text{pump}}(t+\tau) - \psi_{\text{probe}}(t),\\ \Delta\varphi &= \varphi_{\text{pump}}(t+\tau) - \varphi_{\text{probe}}(t).\end{aligned} \tag{9}$$

The linear term in the pulse phase increment, which is related to a difference in optical paths between the pumping and probing beams, is explicitly taken into account by the delay time τ. Then, $\Delta\psi$ is determined entirely by the quadratic term, that is, by the chirp. For the sake of simplicity, we will assume the phase modulation (chirp) in the sample plane to be zero. The second formula (9) implies that the delay of the induced increment in the reflection coefficient depends on the slowly varying amplitude and on the pulse phase (chirp). It is natural to assume that this dependence is weak and can be ignored to the first approximation. Under these assumptions, expression (8) can be rewritten as

$$\begin{aligned}S \propto{}& \langle a_{\text{pump}}^2 A_{\text{pump}}^2(t+\tau)A_{\text{probe}}^2(t)\rangle\\ &+ 2\langle a_{\text{pump}}A_{\text{pump}}(t+\tau)a_{\text{probe}}A_{\text{probe}}^3(t)\rangle\cos\omega_0\tau.\end{aligned} \tag{10}$$

This expression qualitatively describes all the main features of the coherent artifact observed in experiment. First, it contains the rapidly oscillating component of the response varying with the period of the carrier wave. The slowly varying component is always positive (as in experiment) and coincides to within a factor (a_{pump}^2) with the correlation function of the laser pulse intensities. The presence of the factor a_{pump}^2 explains why the slowly varying contribution depends on the orientation of the polarization of the pumping pulse relative to the crystal axes. The ratio of the second term to the first term in expression (10) is proportional to

$$2\frac{a_{\text{probe}}A_{\text{probe}}}{a_{\text{pump}}A_{\text{pump}}},$$

which implies that the contribution of the rapidly oscillating component increases with the probing pulse amplitude. Not that this ratio also depends on the orien-

tation of the polarization of the pumping pulse relative to the crystallographic axes.

Although the proposed model quite satisfactorily describes the trends observed in experiment, it is necessary to develop a consistent microscopic description of these phenomena, since the phenomenological model is self-contradictory. Indeed, the reflection coefficient is a single-valued function of the susceptibility $\chi(\omega)$ of the medium. The susceptibility, characterizing the optical properties of the medium, is defined as the coefficient of proportionality between the Fourier components of the electric field and the polarization of the medium at the same frequency ω. Therefore, the susceptibility $\chi(\omega)$ and, hence, the reflection coefficient cannot explicitly depend on the time, as it was assumed in proposed phenomenological model. It should be noted that, in the case of coherent phonon excitation, the susceptibility implicitly depends on the time because of nonstationary excitation of lattice vibrations in the medium [2, 3].

4.2. Analysis of the Adequacy of the Phenomenological Model

We believe that the physical reason ensuring the adequacy of the proposed phenomenological model is the conservation of the total probability of finding the electron subsystem of a crystal either in the ground or in the excited state in the time scale on the order of laser pulse duration. This, in turn, is a consequence of the smallness of the laser pulse duration as compared to the characteristic times of the energy and phase relaxation of the electron subsystem. This can be illustrated by a simple example of a two-level model of the medium, despite the fact that this model is oversimplified and does not aspire to provide a detailed description of real experiments.

Consider a medium consisting of identical two-level systems with a transition frequency ω_{21}, which are distributed in the volume with a density of N. As is known, the susceptibility of such a medium initially occurring in a ground (nonexcited) state can be expressed in a quasi-stationary case as [7]

$$\chi'(\omega) = \frac{d^2N}{\hbar}\frac{\Delta}{\Delta^2 + \Gamma_2^2 + \frac{\Gamma_2}{\Gamma_1}\Omega^2},$$
$$\chi''(\omega) = \frac{d^2N}{\hbar}\frac{\Gamma_2}{\Delta^2 + \Gamma_2^2 + \frac{\Gamma_2}{\Gamma_1}\Omega^2}, \tag{11}$$

where $\chi'(\omega)$ and $\chi''(\omega)$ are the real and imaginary parts of the susceptibility, respectively; d is the matrix element of the dipole moment of the transition; $\Delta = \omega_{21} - \omega$ is the frequency detuning; Γ_2 and Γ_1 are the transverse and longitudinal relaxation constants, respectively; and Ω is the Rabi frequency. These formulas for the real and imaginary parts of the susceptibility were obtained assuming that a dipole transition is allowed between levels of the two-level system. For $\Gamma_2 = \Gamma_1 \longrightarrow 0$, formulas (11) give correct expressions for the susceptibility in the absence of relaxation:

$$\chi'(\omega) = \frac{d^2N}{\hbar}\frac{\Delta}{\Delta^2 + \Omega^2}, \tag{12}$$
$$\chi''(\omega) = 0.$$

These expressions can be also obtained directly, by solving a nonstationary Schrödinger equation with harmonic time-dependent perturbation:

$$i\hbar\frac{\partial\Psi}{\partial t} = [H_0 + V(t)]\Psi, \tag{13}$$
$$V(t) = V\cos\omega t.$$

Let us consider a somewhat more general case of this problem with a perturbation of the following type:

$$V(t) = V_1\cos\omega t + V_2\cos(\omega t + \phi). \tag{14}$$

As can be readily seen, perturbation (14) with real V_1 and V_2 values can be reduced to the form (13) by substitution

$$V = \sqrt{V_1^2 + 2V_1V_2\cos\phi + V_2^2},$$
$$\omega t \longrightarrow \omega t + \theta, \tag{15}$$
$$\sin\theta = -\frac{V_2\sin\phi}{\sqrt{V_1^2 + 2V_1V_2\cos\phi + V_2^2}}.$$

This implies that a solution of the problem with perturbation (14) reduces to solution of the problem (13) with then effective Rabi frequency

$$\tilde{\Omega} = \sqrt{\Omega_1^2 + 2\Omega_1\Omega_2\cos\phi + \Omega_2^2}, \tag{16}$$

where $\Omega_i = V_i/\hbar$. Not that, since the form of the operator of interaction between the electromagnetic field and the two-level system was not specified, formulas (14)–(16) are valid not only for the electric dipole interaction, but for the other (e.g., quadrupole) interactions as well. In the case of the electric dipole interaction, the susceptibility in the absence of relaxation is

$$\chi'(\omega) = \frac{d^2N}{h}\frac{\Delta}{\Delta^2 + \Omega_1^2 + 2\Omega_1\Omega_2\cos\phi + \Omega_2^2}, \tag{17}$$
$$\chi''(\omega) = 0.$$

The fact that the susceptibility is real allows us to use a relation between the susceptibility and refractive index, which is valid for transparent media:

$$n = \sqrt{1 + 4\pi\chi'(\omega)} = \sqrt{1 + 4\pi\frac{d^2N}{\hbar}\frac{\Delta}{\Delta^2 + \tilde{\Omega}^2}}. \quad (18)$$

Accordingly, for $\tilde{\Omega} \ll \Delta$, the reflection coefficient is

$$R(\omega) = \frac{1-n}{1+n} = \frac{\pi d^2 N}{\hbar} \times\left[-\frac{1}{\Delta} + \frac{\Omega_1^2}{\Delta^3} + \frac{\Omega_2^2}{\Delta^3} + \frac{2\Omega_1\Omega_2\cos\phi}{\Delta^3}\right]. \quad (19)$$

A comparison of expression (19) to the phenomenological formulas (2) and (6) shows their close similarity. In the case of a quasi-stationary excitation, Ω_1 and Ω_2 depend on the slowly varying amplitude and phase, that is, are also slowly varying functions of time. If the phased modulation is absent, we have

$$\Omega_i = \frac{V_i}{\hbar} = \frac{d}{\hbar}A_i(t),$$

where $A_i(t)$ are slowly varying pulse amplitudes. The first term in square brackets of expression (19) corresponds to the background reflection coefficient R_0, while the second and third terms correspond to changes in the reflection coefficients induced separately by the pumping and probing pulses. The last term (which is absent in the phenomenological formula) describes the mutual influence of the two pulses. Formula (19) does not contain explicit dependence of the reflection coefficient on the time with a period of the carrier wave, but it naturally reflects the influence of the relative phase of the two waves, $\phi = \omega_0\tau$, which removes the inconsistency of the phenomenological approach. Expression (19) shows that, during the coherent interaction of two laser pulses with the medium. The reflection coefficient cannot be independently determined for each of the waves and has a physical meaning only for their combination. Even a small change in amplitude of one of the waves will lead to a change in the reflection coefficient for the other wave as well. Using relation (19), we readily obtain an expression for the reflected signal measured using our differential technique:

$$S = -\frac{2\pi^2 d^6 N^2}{\hbar^4\Delta^4}[\langle A_{\text{pump}}^2(t+\tau)A_{\text{probe}}^2(t)\rangle + 2\langle A_{\text{pump}}(t+\tau)A_{\text{probe}}^3(t)\rangle\cos\omega_0\tau]. \quad (20)$$

This expression coincides in form with relation (10) obtained within the framework of the phenomenological model. It should be noted that the dependence of the signal on the orientation of the polarization of laser beams relative to the crystal axes can be also determined taking into account the tensor character of the dipole moment of the transition. We did not perform this analysis for the sake of simplicity. The only significant difference between formula (20) and relation (10) is related to the sign. According to the two-level model, a nonlinear additive to the reflection coefficient must always lead to a decrease in reflection of the probing beam, whereas experiment shows the opposite trend.[1]

Thus, a simple two-level model unexpectedly provides a good qualitative description of the observed features of the coherent artifact. The physical reason of the mutual influence of laser pulses in this model is absolutely clear, being related to the conservation of the total probability. As is known, the interaction of a two-level system with an external harmonic field leads to periodic oscillations in the probability to find the system in an excited state. Therefore, a delayed probing pulse finds the system in the state of a superposition with a certain phase whose value determines the interaction of the probing pulse with the excited system.

5. CONCLUSIONS

The results presented in this paper show that the so-called coherent artifact possesses a regular structure, has a reproducible character, and appears as a result of conservation of the total probability of finding the electron system in one of the two possible (ground or excited) states in the course of the interaction of the medium with spatially superimposed pumping and probing pulses. A good qualitative description of the experimentally observed properties of the coherent artifact, provided by the simple phenomenological model, makes it possible to use this artifact as a natural marker determining the zero point on the relaxation curve. This approach to determining the zero point offers considerable advantages to the other known methods (e.g., reversal of the time axis by means of changing the role of the beams with the transfer of photodiodes from probing to pumping beam channel or use of an auxiliary crystal replacing the sample [2]). Indeed, the method employing the artifact allows the position of the zero delay time to be determined immediately in the course of measurements, without introducing additional changes into the experimental setup. The slow component of the coherent artifact is proportional to the correlation function of the laser pulse intensities and can be used for optimization of these intensities in the course of measurements.

[1] The authors are grateful to the referee for drawing their attention to the fact that the discrepancy between the two-level model and experiment in the sign of the effect is quite reasonable, since a field-induced decrease of the average population difference in the two-level system always leads to a decrease in the resonance additive to the permittivity and, hence, in the refection coefficient.

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REFERENCES

1. J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer, New York, 1996), Springer Series in Solid State Sciences, Vol. 115.
2. T. Dekorsy, G. C. Cho, and H. Kurz, in *Light Scattering in Solids VIII*, Ed. by M. Cardona and G. Güntherodt (Springer, Berlin, 2000), p. 169.
3. O. V. Misochko, Zh. Éksp. Teor. Fiz. **119**, 285 (2001) [JETP **92**, 246 (2001)].
4. P. Borri, F. Romstad, W. Langbein, *et al.*, Opt. Express **7**, 107 (2000).
5. H. Z. Zeiger, J. Vidal, T. K. Cheng, *et al.*, Phys. Rev. B **45**, 768 (1992).
6. *Optical Properties of Semiconductors: a Handbook*, Ed. by V. I. Gavrilenko, A. M. Grekhov, D. V. Korbutyak, and V. G. Litovchenko (Naukova Dumka, Kiev, 1987) [in Russian].
7. D. N. Klyshko, *Physical Principles of Quantum Electronics* (Nauka, Moscow, 1986) [in Russian].

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