Theory of femtosecond photon-echo decay in semiconductors

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We investigate a mechanism responsible for the observed very short times of the photon-echo decay (of the order of a few femtoseconds) in semiconductors. It is associated with the loss of phase memory as a result of the interaction of the interband mixed state with an unscreened random Coulomb potential of the photocarriers and/or charged impurities. A time characteristic of a system of interacting electrons is found. This is the time of phase breaking τ_φ, which we calculate within the eikonal approximation using diagrammatic techniques. τ_φ is shown to be typically much shorter than both the time of electron-electron collisions and the period of plasma oscillations. We demonstrate that the screening of Coulomb potential cannot be built up during this time. τ_φ is proportional to $n^{-1/d}$ (where n is the carrier concentration and d the dimensionality of a system), which is consistent with the experimental results. The derived law of echo decay of the form $\exp[-(\tau/\tau_\varphi)^d]$ agrees with the results of numerical simulation, although it does not agree with the existing results of physical experiment. We believe that such a disagreement is of a fundamental nature and manifests a basic need for further experimental and theoretical work.

I. INTRODUCTION

The echo phenomenon in an ensemble of two-level electron systems excited by a sequence of electromagnetic pulses is well known.[1,2] Recent advances in ultrashort laser-pulse technique have made possible observation of the two-pulse femtosecond echo from interband (valence-conduction band) transitions in bulk semiconductors[3] as well as in quantum-well structures.[4] Knowledge of the ways of time evolution of the mixed quantum states responsible for the echo phenomena in semiconductors is of a great importance for the understanding of various mechanisms of phase relaxation of the electron states as well as its nature. The present paper is devoted to the investigation of a possible mechanism of the femtosecond echo decay in semiconductors and can be considered as a continuation and further development of an earlier paper.[5]

We will demonstrate that the decay of the interband femtosecond echo in semiconductors takes place when carriers in the mixed interband states lose their phase memory. This occurs due to the action of a random *unscreened* Coulomb field originated from either static impurities or photocarriers (provided they are randomly distributed in space) generated by the laser pulse. The field changing the carrier quasimomentum brings the particles out of resonance. This results in echo decay.

It is established that the phase-breaking time τ_φ is proportional to $n^{-1/d}$, where n is the carrier (impurity) concentration and d is the dimensionality of the system. One can say that τ_φ is a *time characteristic of an electron system.* This time usually appears to be shorter than other characteristic times such as time of electron-electron collisions τ_{ee} and period of plasma oscillations; it describes the rate of decay of the coherent properties of an electron-hole system in semiconductors.

The calculated phase-breaking times for a three-dimensional (3D) case are usually about 10 fs. The concentration dependence of τ_φ and its order of magnitude are in agreement with the experiment. We would like to note that it is difficult to make a direct comparison of the time of phase breaking τ_φ and the quantity measured in experiment, T_{echo}, because of different laws of decay observed on the experiment and predicted by the theory. However, the general conclusion $\tau_\varphi \sim n^{-1/d}$ is of major importance.

Echo is a nonlinear effect that in general can be described as follows. Let an observable quantity, say, a macroscopic electric dipole moment $\mathbf{D}$, be the sum of a large number of small contributions from N independent subsystems (in particular, particles). At the moment $t = 0$ an electromagnetic pulse of a very short duration Δt (a "shock") excites an individual particle j into a mixed quantum state between two stationary states with energies $\mathcal{E}_1^j$ and $\mathcal{E}_2^j$. Observables in such a mixed state oscillate with the frequency $\omega_j = \mathcal{E}_2^j - \mathcal{E}_1^j$ (to make expressions in the intermediate formulas less cumbersome we will often set $\hbar = 1$) and the total dipole moment evolves as

$$\mathbf{D}(t) = \sum_j \mathbf{d}_j \exp(-i\omega_j t) + \text{c.c.} \tag{1}$$

At $t = 0$ it has a macroscopic value $D \propto Nd$ ($N \gg 1$), but then decreases due to the differences in frequencies ω_j and practically vanishes for the times $t >$

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$\max\{\Delta t, 1/\Delta\omega\}$, where $\Delta\omega$ is a typical width of the frequency distribution. At the moment $t = \tau \gg \Delta t, 1/\Delta\omega$ a second pulse is applied. A time-reversed part (for which we use the same notation $\mathbf{D}$) appears in the mixed quantum state, i.e., the sign of the frequency is changed $\omega_j \to -\omega_j$ while the phase $(-\omega_j\tau)$ acquired at the moment $t = \tau$ is conserved. Therefore, for $t > \tau$,

$$\mathbf{D}(t) = \sum_j v_j^2 \mathbf{d}_j \exp\left[i\omega_j(t-\tau) - i\omega_j\tau\right] + \text{c.c.} \qquad (2)$$

Factors v_j^2 characterize the probability of time reversal in the dipole moment amplitudes d_j due to the second pulse. We see that at the moment $t = 2\tau$ the phases of all the contributions vanish, $\mathbf{D}(t)$ emerges again as a macroscopic quantity, and the corresponding echo pulse is generated.

Now let us consider echo decay. First of all, we note that there are two principal mechanisms leading to the decay of an echo signal. The first one is a simple damping resulting mainly from large (and fast) fluctuations ("collisions"). These are the processes that, in particular, bring about relaxation of the occupancies. In this case the echo decay is purely exponential

$$\mathbf{D}(2\tau) = \sum_j v_j^2 \mathbf{d}_j \exp\left(-2\tau\nu_j\right) + \text{c.c.}\ , \qquad (3)$$

where ν_j is the half sum of the damping rates, or inverse relaxation times, for energy levels 1 and 2.

The second decay mechanism is related to the phenomenon known as *spectral diffusion*,[6,7] where small frequency fluctuations play the main role. Equation (2) shows that the constancy of the oscillation frequencies ω_j with time is crucial for observation of the echo. However, these frequencies suffer random fluctuations $\delta\omega_j(t)$ because of the interaction of the particles with the surrounding medium.

$D(2\tau)$ is a random quantity that should be averaged over all possible frequency variations $\delta\omega_j(t)$. In Sec. II we will see that

$$\mathbf{D}(2\tau) = \sum_j v_j^2 \mathbf{d}_j \left\langle \exp\left(i\int_\tau^{2\tau} \delta\omega_j(t)dt - i\int_0^\tau \delta\omega_j(t)dt \right) \right\rangle + \text{c.c.} \qquad (4)$$

Here the averaging is denoted by angular brackets $\langle\,\rangle$. As a result of frequency fluctuations, the mean frequency values averaged over a finite time interval suffer a sort of random walk so that one can write

$$\mathbf{D}(2\tau) = \sum_j v_j^2 \mathbf{d}_j \left\langle \exp\left[i\tau(\delta\omega' - \delta\omega)\right] \right\rangle + \text{c.c.}\ , \qquad (5)$$

where $\delta\omega$ and $\delta\omega'$ are frequency fluctuation mean values during successive time intervals $0 < t < \tau$ and $\tau < t < 2\tau$, respectively. The result of the averaging procedure depends on the kind of random walk the frequency undergoes.

If we assume that the random variation of the frequency looks like the usual diffusion and take the probability distribution of the random variable $\xi = \delta\omega' - \delta\omega$ as given by

$$W_t(\xi) = \frac{1}{2\sqrt{\pi Dt}} \exp\left(-\frac{\xi^2}{4Dt}\right)\ , \qquad (6)$$

we get, irrespective to the real physical nature of the diffusion process, the following law for echo decay:

$$\mathbf{D}(2\tau) = \sum_j v_j^2 \mathbf{d}_j \int_{-\infty}^{+\infty} W_\tau(\xi) \exp\left(i\xi\tau\right) d\xi + \text{c.c.} = \sum_j v_j^2 \mathbf{d}_j \exp\left(-D_j\tau^3\right) + \text{c.c.} \qquad (7)$$

The diffusion constant D can be expressed through the mean square of the frequency fluctuation as $\langle\xi^2\rangle/2 = Dt$.

In such a way we can single out two extreme cases of echo decay: a simple exponential decay due to the usual damping (collisions) and decay (in general, nonexponential) due to the spectral diffusion. For various distributions of damping and diffusion constants the summation over j can change the result even for pure cases of damping or diffusion (see Ref. 7).

In the present paper we wish to indicate that the most important mechanism of echo decay due to band-to-band transitions in semiconductors is often associated with a sort of spectral diffusion rather than with the usual damping due to ordinary collisions. The physics of this mechanism can be described as follows. The echo is due to almost vertical transitions between the valence and conduction bands. The energy difference corresponding to such a transition is determined by the quasimomentum of a pairs of quantum states, or their "kinetic" energies $p^2/2m_{e,h}$. A long-range static field acting on the pair brings about variation of its quasimomentum and therefore the interlevel spacing of the corresponding two-level system. The variation equals the negative variation of their potential energies δU. It is this quantity that plays a role of $\delta\omega_j(t)$ in Eq. (4). A smoothness of the spatial variation of the field is important because in this case the processes of scattering of the carriers accompanied by a large variation of their quasimomenta would not happen.

We consider here a particular example of a long-range Coulomb interaction of the carriers in a semiconductor where one can look upon the phase variation as a sum of a large number of relatively small contributions. As a result, we get a sort of diffusional process that concerns the phase of the wave function in the mixed state. Such a process will be called here *phase wandering*. It is of major importance that the echo dies off during the time τ_φ, which may be much shorter than the time characterizing collisions of the particles interacting according to the Coulomb law. We are going to show that indeed for a number of cases of interest such random variations of the phase may be quite effective as a dephasing mechanism with the characteristic decay time much shorter than usual relaxation time due to collisions.

The echo decay, or optical dephasing, was studied in semiconductors in a number of papers (see Refs. 8–10). The results obtained usually correspond to the consid-

eration of damping, i.e., only the action of short-range and/or rapidly varying parts of the perturbations are taken into account. In this way the echo decay in disordered semiconductors is considered in the paper by Lonsky *et al.*[9] and an exponential law of decay is found. The difference between their and our case can be understood as follows. In Ref. 9, due to the short range of the potential, the time of echo decay is determined by the collision time and the law of decay turns out to be purely exponential. We point out another mechanism of decay that under certain conditions may be more effective and results in a different law of decay.

It is important to mention that recently El Sayed *et al.*[11] have performed a numerical simulation of the set of equations for the density matrix in semiconductor at the early nonequilibrium stage after optical excitation. They found the decay law to be nonexponential, in qualitative agreement with our results.

The present paper is organized as follows. In Sec. II we give a brief description of our diagram technique and time-operator method while applying them to the echo phenomena. In Sec. III, using these techniques, we demonstrate that the screening can be neglected at time intervals less than the inverse plasma frequency. In Sec. IV we consider the echo phenomenon in a two-band semiconductor. The use of the time-ordered diagrams enables one to visualize the physical mechanism of echo formation as well as possible mechanisms of its decay. We show that on a long-time scale the echo decay is purely exponential and due to the usual collisions. But this is probably not the case for the femtosecond echo experiments where the characteristic times are usually very short. On a short-time scale the wandering of phase can be the main mechanism responsible for the echo decay. This case is discussed in Sec. V, where we give a diagrammatic derivation of the echo decay law in the eikonal approximation. In this approximation we sum the terms of perturbation series in all orders in the perturbation potential and impurity concentration. The application of our formulas to the 3D and 2D situations is discussed. Then the result is generalized for the case where the perturbation field is due to mobile carriers excited by light. Section VI is devoted to the qualitative consideration of our results. The results are briefly discussed in Sec. VII.

II. DIAGRAMMATIC TECHNIQUES

To investigate the echo phenomenon in semiconductors and in particular the echo decay we will use in what follows a time-ordered diagrammatic techniques (cf. with Ref. 12; see also Ref. 13). Let us first define an inverse operator $1/\partial_t$ (which will be extensively used later) as

$$\frac{1}{\partial_t} f(t) \equiv \int_0^t dt'\ f(t')\ . \tag{8}$$

Making use of this definition one can show that

$$\frac{1}{\partial_t + i\omega} f(t) = e^{-i\omega t} \int_0^t dt'\ f(t') e^{i\omega t'} \tag{9}$$

and that the following operator identity holds:

$$\varphi(\partial_t) e^{i\omega t} = e^{i\omega t} \varphi(\partial_t + i\omega). \tag{10}$$

The use of the operator $1/\partial_t$ makes the formulas more compact and transparent. Moreover, if needed, one can easily get the Laplace (Fourier) transforms by simple substitutions of differentiation operators ∂_t with the corresponding variables.

The diagrams allow one to write down the analytical expressions by applying the following rules. Two lines going in the "positive" ("negative") direction with respect to the time variable (along time or against it) on a diagram describe the time evolution of the density matrix until the moment of observation $t = 2\tau$. Each line carries the corresponding quantum numbers. An initial occupancy is indicated by the bar that closes the loop in a remote past. A point on a positive (negative) line represents a matrix element of interaction together with an additional factor $-i$ $(+i)$. An evolution operator $(\partial_t + i\sum_j \mathcal{E}_j - i\sum_k \mathcal{E}_k)^{-1}$ should be brought into correspondence with each time interval between two adjacent interaction points and after the last one. Here j (k) stands for the positive (negative) lines. The extreme right point on the diagram corresponds to an operator of an observable physical quantity. One should ascribe to this point the corresponding matrix element. Note that to write down the analytical expression we should read a diagram from future to past, following the order in which all operators appear.

To illustrate the relation between pictures and formulas let us return to the echo effect in an ensemble of independent two-level systems (see Fig. 1). At $t < 0$ we have a stationary state. The first pulse at $t = 0$ creates an oscillating mixed quantum state. The second pulse reverses the state at $t = \tau$ and finally at $t = 2\tau$ a radiation pulse is observed. A positive (negative) line on the diagram carries energy $\mathcal{E}_2^j$ $(\mathcal{E}_1^j)$ from $t = 0$ to $t = \tau$ and $\mathcal{E}_1^j$ $(\mathcal{E}_2^j)$ from $t = \tau$ to $t = 2\tau$. To make our consideration as simple as possible, we assume that the time interval between the pulses τ is much longer than the durations of the pumping pulses Δt. As the duration of the pulses is the shortest time in the problem, we will consider here δ pulses. According to the aforementioned rules we have

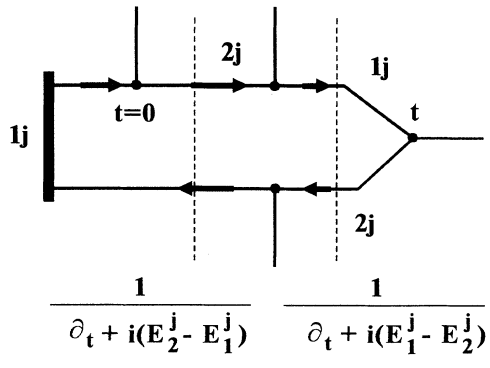

FIG. 1. Diagram for two-pulse echo. The bar before $t = 0$ represents the initial occupancy. For further explanation, see the text.

$$\mathbf{D}(t) = \sum_j \langle 2j|\mathbf{d}|1j\rangle \frac{1}{\partial_t + i(\mathcal{E}_1^j - \mathcal{E}_2^j)}\delta(t-\tau) \times(-i\langle 1j|V_2|2j\rangle)(i\langle 1j|V_2|2j\rangle) \times \frac{1}{\partial_t + i(\mathcal{E}_2^j - \mathcal{E}_1^j)}(-i\langle 2j|V_1|1j\rangle)n_1^j\delta(t) + \text{c.c.} ,$$

which is equivalent to Eq. (2) because of the identity $(\partial_t + a)^{-1}\delta(t) = \exp(-at)$ [see Eq. (9)]. Here $n_1^j(-i\langle 2j|V_1|1j\rangle)\langle 2j|\mathbf{d}|1j\rangle$ stands for the induced dipole moment $\mathbf{d}_j$ (n_1^j being the occupation number for the first level of jth particle) and $\langle 1j|V_2|2j\rangle$ stands for v_j. In principle, there are diagrams with an occupation number n_2^j as well, but for simplicity we consider here the case where $n_2^j = 0$. We can include the fluctuations in the general scheme, regarding them as amplitudes of instant shocks distributed randomly in time. Representing them by points on a diagram we get, for the evolution operator (see Fig. 2),

$$\frac{1}{\partial_t + i\omega_j} + \frac{1}{\partial_t + i\omega_j}[-i\delta\omega_j(t)]\frac{1}{\partial_t + i\omega_j} + \cdots = \frac{1}{\partial_t + i\omega_j + i\delta\omega_j(t)} . \quad (11)$$

Using Eq. (11) we obtain the total dipole moment as

$$\mathbf{D}(t) = \sum_j v_j^2 \mathbf{d}_j \frac{1}{\partial_t - i\omega_j - i\delta\omega_j(t)}\delta(t-\tau) \times \frac{1}{\partial_t + i\omega_j + i\delta\omega_j(t)}\delta(t) + \text{c.c.} \quad (12)$$

As we already mentioned, this is a random quantity that should be averaged over all possible frequency variations $\delta\omega_j(t)$. Making use of definitions Eqs. (8) and (9) and returning to exponents, we get a derivation of Eq. (4).

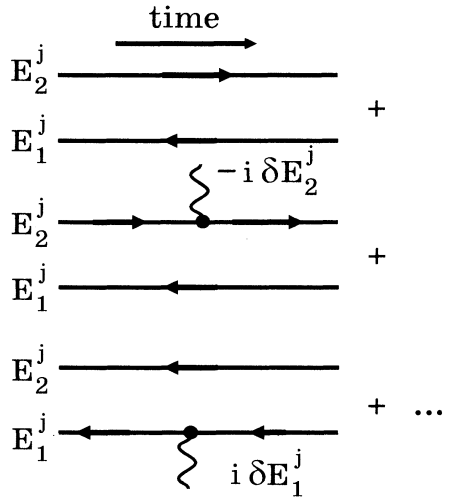

FIG. 2. Action of random forces on a two-level system.

III. SCREENING OF FAST PROCESSES

In this section we will show that the screening of the Coulomb potential cannot build up during the ultrafast processes we are discussing here. The Coulomb propagator renormalized both by the screening and by the lattice effects was calculated by Gurevich, Larkin, and Firsov in Ref. 14 and one could base consideration of the build up of the screening on the results of that paper. Here we will not be interested in the lattice effects because of the shortness of the time intervals we are going to consider. The dielectric susceptibility of the semiconductor may be considered as a time-independent constant ε_∞. We, however, prefer here to use the language based on the time-ordered diagram technique formulated in Sec. II. This will permit us to visualize the physical processes as developing just in the way they are represented in the diagrams. A diagram consists of horizontal lines with points on them. The lines represent the probability amplitudes (wave functions) whereas the points indicate time moments when a probability amplitude changes under the action of a perturbation.

In the nonrelativistic approximation the Coulomb interaction is instantaneous so that both points on the Coulomb line are characterized by the same interaction time, say, $t = t_0$, and bare interaction lines in our pictures become vertical. The screening may be interpreted as polarization of the medium and creation of a polarization field, which should be added to the bare interaction. The screened potential may be constructed step by step by the repetition of time-ordered events. Since the evolution of an electron system requires some time and the polarization field emerges at times $t > t_0$ the screened potential becomes time dependent. Its space Fourier transform $U(\mathbf{q}, t)$ is given by the series

$$U(\mathbf{q}, t-t_0) = U(\mathbf{q})[\delta(t-t_0) + \Pi(\mathbf{q}, t)U(\mathbf{q})\delta(t-t_0) + \cdots] = U(\mathbf{q})\frac{1}{1 - U(\mathbf{q})\Pi(\mathbf{q}, t)}\delta(t-t_0) , \quad (13)$$

where by definition

$$U(\mathbf{q}) = \frac{4\pi e^2}{\varepsilon_\infty q^2} .$$

Here the first term represents the "bare" momentary Coulomb interaction while other terms describe polarization obtained by the iterations. We see that the usual physical picture of polarization (the field displaces the charges from their equilibrium positions; as a result, a nonzero contribution to the local charge density emerges; this density gives rise to the polarization field, which in its turn displaces the charges) has a one to one correspondence to the diagrams.

First consider the contribution to the polarization operator of electrons in the conduction band. The operator is given by two simple response diagrams of the type shown on Fig. 1, but with only one interaction point at time t_0 and one point at the observation time t. It has the well-known form

$$\Pi(\mathbf{q},t)=\sum_{\mathbf{p}}\frac{1}{\partial_t+i(\epsilon_{\mathbf{p}+\mathbf{q}}-\epsilon_{\mathbf{p}})}i[F_{\mathbf{p}+\mathbf{q}}(t)-F_{\mathbf{p}}(t)]\ . \quad (14)$$

This expression may be read as follows. A perturbation with the amplitude proportional to the matrix element $\langle\mathbf{p}'|\exp(i\mathbf{qr})|\mathbf{p}\rangle=\delta_{\mathbf{p}',\mathbf{p}+\mathbf{q}}$ at some time creates a mixed $|\mathbf{p}'\rangle\langle\mathbf{p}|$ electron state out of the pure states $|\mathbf{p}\rangle\langle\mathbf{p}|$ or $|\mathbf{p}'\rangle\langle\mathbf{p}'|$ occupied by $F_{\mathbf{p}}$ or $F_{\mathbf{p}'}$ electrons, respectively. The evolution of the mixed state under the action of the unperturbed electron Hamiltonian is described by the denominator on the right-hand side of (14), where the use of the symbol ∂_t ensures the necessary time dependence. The sum over $\mathbf{p}$ gives the charge distribution at time t.

Let us consider small values of q as compared with the characteristic values of p. Then one can write

$$\epsilon_{\mathbf{p}+\mathbf{q}}=\epsilon_{\mathbf{p}}+\mathbf{qv}+\cdots$$

and

$$F_{\mathbf{p}+\mathbf{q}}=F_{\mathbf{p}}+\mathbf{q}\frac{\partial F_{\mathbf{p}}}{\partial\mathbf{p}}$$

so that the polarization operator becomes equal to

$$\Pi(\mathbf{q},t)=\sum_{\mathbf{p}}\frac{1}{\partial_t+i\mathbf{qv}}i\mathbf{q}\frac{\partial}{\partial\mathbf{p}}F_{\mathbf{p}}\ . \quad (15)$$

Using the identity

$$\frac{\partial}{\partial\mathbf{p}}\frac{1}{\partial_t+i\mathbf{qv}}F_{\mathbf{p}}=\frac{1}{\partial_t+i\mathbf{qv}}\frac{\partial}{\partial\mathbf{p}}F_{\mathbf{p}}+\left(\frac{\partial}{\partial\mathbf{p}}\frac{1}{\partial_t+i\mathbf{qv}}\right)F_{\mathbf{p}}$$
$$=\frac{1}{\partial_t+i\mathbf{qv}}\frac{\partial}{\partial\mathbf{p}}F_{\mathbf{p}}-\frac{i(\mathbf{q}/m)}{(\partial_t+i\mathbf{qv})^2}F_{\mathbf{p}} \quad (16)$$

we get

$$\Pi(\mathbf{q},t)=-\frac{q^2}{m}\sum_{\mathbf{p}}\frac{1}{(\partial_t+i\mathbf{qv})^2}F_{\mathbf{p}}(t)\ . \quad (17)$$

We assume that the characteristic time interval t_c (whose inverse gives in particular an order-of-magnitude estimate for ∂_t) is so short that t_c^{-1} is much larger than $\mathbf{qv}$. Omitting this term in the denominator, we obtain

$$\Pi(\mathbf{q},t)=-\frac{q^2}{m}\frac{1}{\partial_t^2}n(t)\ , \quad (18)$$

where

$$n(t)=\sum_{\mathbf{p}}F_{\mathbf{p}}(t)$$

is the number of carriers (per unit volume) in the conduction band. If we insert this expression into (13), we see that the acting potential after the switching at $t=t_0$ is developing as

$$[\partial_t^2+\omega_{\rm pl}^2(t)]U(\mathbf{q},t-t_0)=U(\mathbf{q})\partial_t^2\delta(t-t_0) \quad (19)$$

or

$$U(\mathbf{q},t-t_0)=U(\mathbf{q})\left[1-\frac{1}{\partial_t^2}\omega_{\rm pl}^2+\cdots\right]\delta(t-t_0)$$
$$=U(\mathbf{q})[\delta(t-t_0)$$
$$-(t-t_0)\omega_{\rm pl}^2(t_0)+\cdots]\ . \quad (20)$$

Here $\omega_{\rm pl}(t)=[4\pi e^2n(t)/m]^{1/2}$ is the (time-dependent) plasma frequency. The right-hand side of Eq. (19) takes into account the initial conditions to the differential equation (19).

The expression for the screened potential can be simplified provided the electron concentration and therefore the plasma frequencies are time independent. Then we have

$$U(\mathbf{q},t-t_0)=U(\mathbf{q})\left[1-\omega_{\rm pl}^2\frac{1}{\partial_t^2}+\cdots\right]\delta(t-t_0)$$
$$=U(\mathbf{q})\frac{\partial_t^2}{\partial_t^2+\omega_{\rm pl}^2}\delta(t-t_0)\ . \quad (21)$$

Taking into account the identity

$$\frac{\partial_t^2}{\partial_t^2+\omega_{\rm pl}^2}=1+\frac{\omega_{\rm pl}}{2i}\left(\frac{1}{\partial_t+i\omega_{\rm pl}}-\frac{1}{\partial_t-i\omega_{\rm pl}}\right)\ , \quad (22)$$

one can get

$$U(\mathbf{q},t-t_0)=U(\mathbf{q})\{\delta(t-t_0)$$
$$-\Theta(t-t_0)\omega_{\rm pl}\sin[\omega_{\rm pl}(t-t_0)]\}\ . \quad (23)$$

We see that the screening of electric field by conduction electrons leads to the oscillations of their potentials with the frequency $\omega_{\rm pl}$. As we have pointed out, we limit ourselves to the lowest approximation in q^2, where there is no damping of plasma oscillations. One can check, however, that in higher approximations, the term describing screening would be even smaller.

We could include valence-band electrons into the scheme with the corresponding polarization operator that should be added to the polarization operator in (14). As a result, the total concentration of the carriers (electrons and holes) enters the expression for the plasma frequency. In contrast to the importance of the screening for lower frequencies, the above-written expressions clearly demonstrate that there is no reason to take the screening into account for very short time intervals in the femtosecond echo phenomenon where $\omega_{\rm pl}t_c\ll 1$ because it just has not enough time to build up.

An equation similar to Eq. (19) has been obtained for the nonsingular part of $U(\mathbf{q},t)$ by other methods and solved in Ref. 15. The authors of Ref. 15 assumed that the plasma frequency variation is sufficiently small during the period of oscillation (WKB approximation). We believe that, in fact, to neglect the screening there is no need for $\omega_{\rm pl}(t)$ to vary slowly. It is quite sufficient that the condition $\omega_{\rm pl}t_c\ll 1$ holds during the time interval of interest.

IV. ECHO PHENOMENON IN SEMICONDUCTORS

We consider a semiconductor with the energy gap E_g and the dispersion laws in the conduction and valence

bands given by $\mathcal{E}^c_{\mathbf{p}} = p^2/2m_e$ and $\mathcal{E}^v_{\mathbf{p}} = -E_g - p^2/2m_h$ where $\mathbf{p}$ is the electron quasimomentum. The interaction of light with electrons has the usual form

$$V_j = \frac{eE_j(t)}{2m\omega i}\left[e^{i(\omega t-\mathbf{k}_j\mathbf{r})}(\mathbf{e}_j\mathbf{P}) - e^{-i(\omega t-\mathbf{k}_j\mathbf{r})}(\mathbf{e}^*_j\mathbf{P})\right] , \tag{24}$$

where $\mathbf{P} = -i\nabla$ is an electron momentum operator and m is the mass of a bare electron. The indices $j = 1, 2$ correspond to the first and second pulses of an ac electric field and $E_j(t)$ are their envelopes.

The mixed quantum state that is responsible for the echo phenomenon is represented by the nondiagonal element of density matrix or, simply, by the product of the wave functions specified by the values of their quasimomenta

$$\psi^*_{\mathbf{p}c}\psi_{\mathbf{p}v} \sim \exp(i\Omega_{\mathbf{p}}t) , \tag{25}$$

$$\psi^*_{\mathbf{p}v}\psi_{\mathbf{p}c} \sim \exp(-i\Omega_{\mathbf{p}}t) , \tag{26}$$

where

$$\Omega_{\mathbf{p}} = E_g + \frac{p^2}{2m_e} + \frac{p^2}{2m_h}. \tag{27}$$

We assume that a short laser pulse creates such a state at $t = 0$ (see Fig. 3). Then another pulse at $t = \tau$ reverses it and at $t = 2\tau$ an echo pulse is observed just as it has been shown in Sec. III. The drawings in Fig. 3 reflect the fact that a state oscillating with frequency $\Omega_{\mathbf{p}}$ can be created by mixing of two pure quantum states (diagonal elements of the density matrix) $\psi^*_{\mathbf{p}c}\psi_{\mathbf{p}c}$ and $\psi^*_{\mathbf{p}v}\psi_{\mathbf{p}v}$. The bar at the bottom of the drawing corresponds to the occupancies of these states $F_{c\mathbf{p}}$ and $F_{v\mathbf{p}}$.

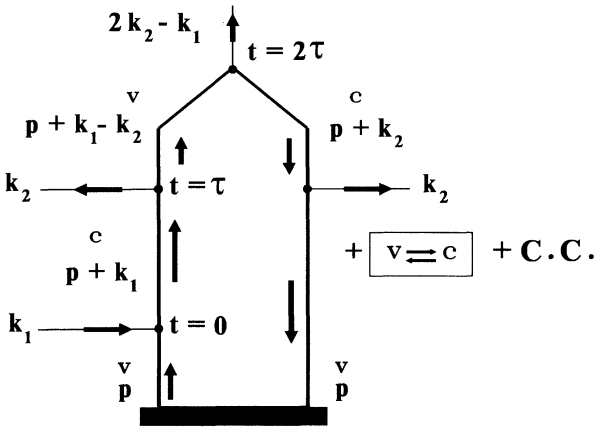

FIG. 3. Echo in semiconductors.

The diagram in Fig. 3 enables one to write an expression for the spatial Fourier transform of the polarization current. The current is represented by the point at the moment $t = 2\tau$ (connected with an upper arrow) in the drawing. The corresponding interband matrix element is $e\mathbf{v}_{cv} = e\mathbf{P}_{cv}/m$. Other points correspond to the matrix elements of electron interband transitions and describe the action of the two laser pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. Due to spatial homogeneity the total quasimomentum is conserved at any interaction point. Thus we have

$$\mathbf{j}(t, \mathbf{k}_1 - 2\mathbf{k}_2) = 2e\sum_{\mathbf{p}} \mathbf{v}_{cv}\frac{1}{\partial_t + i(\mathcal{E}^v_{\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2} - \mathcal{E}^c_{\mathbf{p}+\mathbf{k}_2})}\delta(t-\tau)(-i\langle v, \mathbf{p}+\mathbf{k}_1-\mathbf{k}_2|V_2|c, \mathbf{p}+\mathbf{k}_1\rangle)$$
$$\times(i\langle v, \mathbf{p}|V_2|c, \mathbf{p}+\mathbf{k}_2\rangle)\frac{1}{\partial_t + i(\mathcal{E}^c_{\mathbf{p}+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}})}(i\langle c, \mathbf{p}+\mathbf{k}_1|V_1|v, \mathbf{p}\rangle)\delta(t)(F_{c\mathbf{p}+\mathbf{k}_1} - F_{v\mathbf{p}})$$

or

$$\mathbf{j}(t, \mathbf{k}_1 - 2\mathbf{k}_2)$$
$$= 2e\sum_{\mathbf{p}} \mathbf{v}_{cv}\frac{V^2_{2\mathbf{p}}e^{2i\omega\tau}}{\partial_t - i[\Omega_{\mathbf{p}} - (\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}_h + \mathbf{k}_2\mathbf{v}_e]}\delta(t-\tau)$$
$$\times\frac{iV^*_{1\mathbf{p}}}{\partial_t + i(\Omega_{\mathbf{p}} + \mathbf{k}_1\mathbf{v}_e)}\delta(t)(F_{c\mathbf{p}+\mathbf{k}_1} - F_{v\mathbf{p}}). \tag{28}$$

We set $\mathcal{E}^{c,v}_{\mathbf{p}+\mathbf{k}} \simeq \mathcal{E}^{c,v}_{\mathbf{p}} + \mathbf{k}\mathbf{v}_{e,h}$ because of the inequality $k \ll p$. Here $V_{j\mathbf{p}}$ stands for the matrix elements of the light-electron interaction [$j = 1$ (2) for the first (second) pulse]. In what follows we consider rectangular pulses. Then

$$V_{j\mathbf{p}} = \frac{e}{2m\omega}(\mathbf{E}_j\mathbf{P}_{vc})\frac{1 - e^{i(\omega-\Omega_{\mathbf{p}})\Delta t_j}}{\omega - \Omega_{\mathbf{p}}} , \tag{29}$$

where Δt_j is the pulse duration. Making use of the operator identity

$$\exp(-at)(1/\partial_t)\exp(at) = 1/(\partial_t + a) \tag{30}$$

[see Eq. (9)], we have for $t = 2\tau$

$$\mathbf{j}(2\tau, \mathbf{k}_1 - 2\mathbf{k}_2) = 2e\sum_{\mathbf{p}} \mathbf{v}_{cv}\exp[i(\mathbf{k}_2 - \mathbf{k}_1)(\mathbf{v}_h + \mathbf{v}_e)\tau]$$
$$\times iV^2_{2\mathbf{p}}e^{2i\omega\tau}V^*_{1\mathbf{p}}(F_{c\mathbf{p}} - F_{v\mathbf{p}}). \tag{31}$$

Here we neglect $\mathbf{k}_1$ in the argument of the distribution function $F_{c\mathbf{p}}$.

Let us discuss the mechanisms leading to the echo decay in a more formal way. We assume that the electrons are influenced by a random field $\mathcal{U}(\mathbf{r}, t)$ produced either by impurities (and lattice vibrations) or by other electrons created by the laser pulse and randomly distributed in space. Field $\mathcal{U}(\mathbf{r}, t)$ can be represented by its space and time harmonics

$$\mathcal{U}(\mathbf{r}, t) = \sum_{\mathbf{q},\omega}\mathcal{U}_{\mathbf{q}\omega}e^{-i\omega t+i\mathbf{q}\mathbf{r}}. \tag{32}$$

The mean value of $\mathcal{U}(\mathbf{r},t)$ vanishes while

$$\langle \mathcal{U}^*(\mathbf{r}_1,t_1)\mathcal{U}(\mathbf{r},t)\rangle = \sum_{\mathbf{q},\omega} \langle |\mathcal{U}_{\mathbf{q}\omega}|^2\rangle\, e^{-i\omega(t-t_1)+i\mathbf{q}(\mathbf{r}-\mathbf{r}_1)} \ . \tag{33}$$

For example, for the potential of randomly distributed static impurities we have

$$\mathcal{U}(\mathbf{r}) = \sum_j U(\mathbf{r}-\mathbf{r}_j) = \sum_{j,\mathbf{q}} U_{\mathbf{q}} e^{i\mathbf{q}(\mathbf{r}-\mathbf{r}_j)} \ , \tag{34}$$

where $\mathbf{r}_j$ is the position of the jth impurity.

At first we will study the role of short-range fast field fluctuations with wave vectors $q \simeq p$ and frequencies $\omega \simeq \mathcal{E}_p$. Due to the action of such fluctuations the electron states become damped. We illustrate this by the following diagram (see Fig. 4). There are two fluctuations with the space and time dependence of the type $\exp(\pm i\omega t \pm i\mathbf{qr})$ that act on the wave functions of the considered state at arbitrary time moments as two successive momentary shocks. The first diagram corresponds to an expression of the type

$$-\sum_{\mathbf{q}} \langle |\mathcal{U}_{\mathbf{q}\omega}|^2\rangle \frac{1}{\partial_t + i\Omega_{\mathbf{p}} + i(\mathcal{E}^c_{\mathbf{p}+\mathbf{q}} - \mathcal{E}^c_{\mathbf{p}} - \omega)} \equiv -i\delta\mathcal{E}^c_{\mathbf{p}} \tag{35}$$

and there is an analogous expression for the second diagram. In the final expressions for the polarization current [of the same type as in Eq. (31)] a large frequency $\Omega_{\mathbf{p}}$ can be removed from all the denominators making use of operator identity Eq. (10). An order-of-magnitude estimate gives $\partial_t \simeq 1/t$. For times $t \gg 1/\mathcal{E}_p$ this expression is none other than a contribution to the electron self-energy and to the damping due to field fluctuations:

$$-i\delta\mathcal{E}^c_{\mathbf{p}} = -\pi \sum_{\mathbf{q}} \langle |\mathcal{U}_{\mathbf{q}\omega}|^2\rangle\, \delta(\mathcal{E}^c_{\mathbf{p}+\mathbf{q}} - \mathcal{E}^c_{\mathbf{p}} - \omega) + i\sum_{\mathbf{q}} \langle |\mathcal{U}_{\mathbf{q}\omega}|^2\rangle \frac{1}{\mathcal{E}^c_{\mathbf{p}+\mathbf{q}} - \mathcal{E}^c_{\mathbf{p}} - \omega} \ . \tag{36}$$

Here, for $\partial_t \to 0$ we have used the symbolic relation

$$\frac{1}{\partial_t + i\Delta\mathcal{E}} \to -i\,P\,\frac{1}{\Delta\mathcal{E}} + \pi\delta(\Delta\mathcal{E}) \ . \tag{37}$$

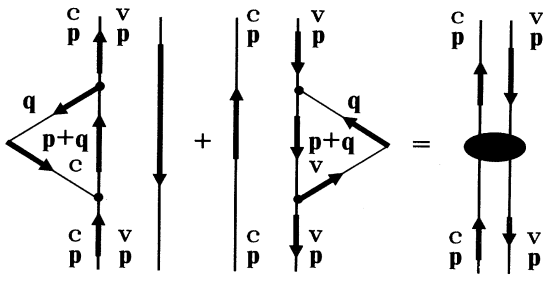

FIG. 4. Damping due to "collisions."

The second drawing in Fig. 4 represents the term $i\delta\mathcal{E}^v_{\mathbf{p}}$. The diagram in Fig. 4 describes interaction of an electron in the mixed quantum state with the time-dependent local field fluctuations caused by the scatterers. We can include the interactions in all lines of the echo diagrams in Fig. 3. This results in the substitution

$$\Omega_{\mathbf{p}} \Rightarrow \Omega_{\mathbf{p}} + \delta\Omega_{\mathbf{p}} \pm i\frac{\nu_{\mathbf{p}}}{2}. \tag{38}$$

The frequency variation $\delta\Omega_{\mathbf{p}}$ vanishes in the final expression so that only the damping is important. As a result, there is an exponential echo decay in this case.

For the fast echo phenomena such as the femtosecond echo an estimate shows that decay takes place on the time scale $\tau \simeq 1/\mathcal{E}_p$. Therefore one usually cannot use the δ-function approximation [Eq. (37)]. Moreover, during the delay time for the echo signal τ, the electron collisions are improbable because of the inequality $\tau \ll \tau_{\mathbf{p}}$, where $\tau_{\mathbf{p}} = 1/\nu_{\mathbf{p}}$ is the momentum relaxation time. If the source of the random potential are the mobile carriers, then $\tau_{\mathbf{p}}$ has the physical meaning of the electron-electron collision time τ_{ee}. For the carrier concentration of the order of 10^{17} cm^{-3}, τ_{ee} is greater than 100 fs. Since the time of echo decay is of the order of 10 fs (see Ref. 3), we should exclude these collisions as a cause of the echo decay. On the other hand, for such carrier concentrations the period of plasma oscillations is also about 100 fs, so that the Debye screening of field fluctuations cannot be built up during the echo evolution (see Sec. III). This was pointed out in Ref. 16 and confirmed recently in Ref. 15. The experiment shows[3,4] that the time of echo decay depends on the carrier concentration n, as $n^{-1/d}$. This fact indicates that field fluctuations are mainly due to the unscreened Coulomb field of the carriers created by the laser pulses. Thus the long-range and slowly varying part of the field fluctuations should play the main role in the decay. We will consider in detail this recently proposed mechanism of phase wandering in the next section.

V. FEMTOSECOND ECHO DECAY

For such a short time interval the potential created by randomly distributed charges (electrons or impurities) may be treated as static or quasistatic. The change of the electron (hole) quasimomentum q under the action of a smooth and long-range Coulomb potential is small compared to the average quasimomentum p (this makes the so-called high-energy or *eikonal* approximation[17]), so that

$$|\mathcal{E}_{\mathbf{p}-\mathbf{q}} - \mathcal{E}_{\mathbf{p}}| \ll \mathcal{E}_{\mathbf{p}}.$$

Neglecting the terms proportional to q^2 we have

$$\mathcal{E}_{\mathbf{p}-\mathbf{q}} - \mathcal{E}_{\mathbf{p}} = -\mathbf{q}\mathbf{v}. \tag{39}$$

It was shown[17] that within this approximation it is possible to sum all the diagrams in all orders of the perturbation theory with respect to the concentration of scatterers and their potential energy.

Let us begin with the consideration of the echo decay due to the random potential of static impurities. To take into account the influence of charged impurities, one should insert points of interaction with impurities into the diagrams and take the average. In the second order in the perturbation potential U and in the first order in the impurity concentration n one gets 12 terms of the perturbation theory. As an example, three of them are represented by diagrams in Fig. 5. In the expressions corresponding to the diagrams given below we will omit the common factor

$$2e\mathbf{v}_{cv}iV_{2\mathbf{p}}^2 e^{2i\omega\tau}V_{1\mathbf{p}}^*(F_{c\mathbf{p}}-F_{v\mathbf{p}}). \tag{40}$$

After taking into account Eqs. (30) and (39) the diagrams are equal to the expressions

$$n\frac{1}{\partial_t}\delta(t-\tau)\frac{1}{\partial_t}(-iU_{\mathbf{q}})e^{i\mathbf{q}\mathbf{v}_e t}\frac{1}{\partial_t}(-iU_{-\mathbf{q}})e^{-i\mathbf{q}\mathbf{v}_e t}\frac{1}{\partial_t}\delta(t), \tag{41}$$

$$n\frac{1}{\partial_t}\delta(t-\tau)\frac{1}{\partial_t}(-iU_{-\mathbf{q}})e^{-i\mathbf{q}\mathbf{v}_e t}\frac{1}{\partial_t}(iU_{\mathbf{q}})e^{i\mathbf{q}\mathbf{v}_e t}\frac{1}{\partial_t}\delta(t), \tag{42}$$

$$n\frac{1}{\partial_t}(iU_{-\mathbf{q}})e^{-i\mathbf{q}\mathbf{v}_e t}\frac{1}{\partial_t}e^{i\mathbf{q}\mathbf{v}_e t}$$

$$\times e^{-i\mathbf{q}\mathbf{v}_h t}\delta(t-\tau)\frac{1}{\partial_t}(iU_{\mathbf{q}})e^{i\mathbf{q}\mathbf{v}_h t}\frac{1}{\partial_t}\delta(t). \tag{43}$$

It is worthwhile to mention that in addition to the diagrams well known in kinetics describing the usual "in" and "out" terms, there are some special types describing correlation of the carriers via impurities during various time intervals. For example, the third diagram takes into account such a correlation in the electron motion in the conduction and valence bands during the time intervals from 0 to τ and from τ to 2τ. After summation over $\mathbf{q}$ we get for the sum of all 12 diagrams

$$\frac{n}{2}\int d\mathbf{r}\left\{-i\int_0^\tau U(\mathbf{r}-\mathbf{v}_e t)dt\right.$$
$$+i\int_\tau^{2\tau} U\left[\mathbf{r}-\mathbf{v}_e(t-\tau)-\mathbf{v}_h\tau\right]dt$$
$$+i\int_0^\tau U(\mathbf{r}-\mathbf{v}_h t)dt$$
$$\left.-i\int_\tau^{2\tau} U\left[\mathbf{r}-\mathbf{v}_h(t-\tau)-\mathbf{v}_e\tau\right]dt\right\}^2.$$

Taking into account all orders in U and n we obtain the evolution law in the form

$$\exp\left[-n\int d\mathbf{r}(1-e^{i\phi})\right], \tag{44}$$

where

$$\phi=-\int_0^\tau U\left[\mathbf{r}-(\mathbf{p}/m_e)t\right]dt$$
$$+\int_\tau^{2\tau} U\left[\mathbf{r}-(\mathbf{p}/m_e)t+(\mathbf{p}/m_{eh})\tau\right]dt$$
$$+\int_0^\tau U\left[\mathbf{r}+(\mathbf{p}/m_h)t\right]dt$$
$$-\int_\tau^{2\tau} U\left[\mathbf{r}+(\mathbf{p}/m_h)t-(\mathbf{p}/m_{eh})\tau\right]dt\,.$$

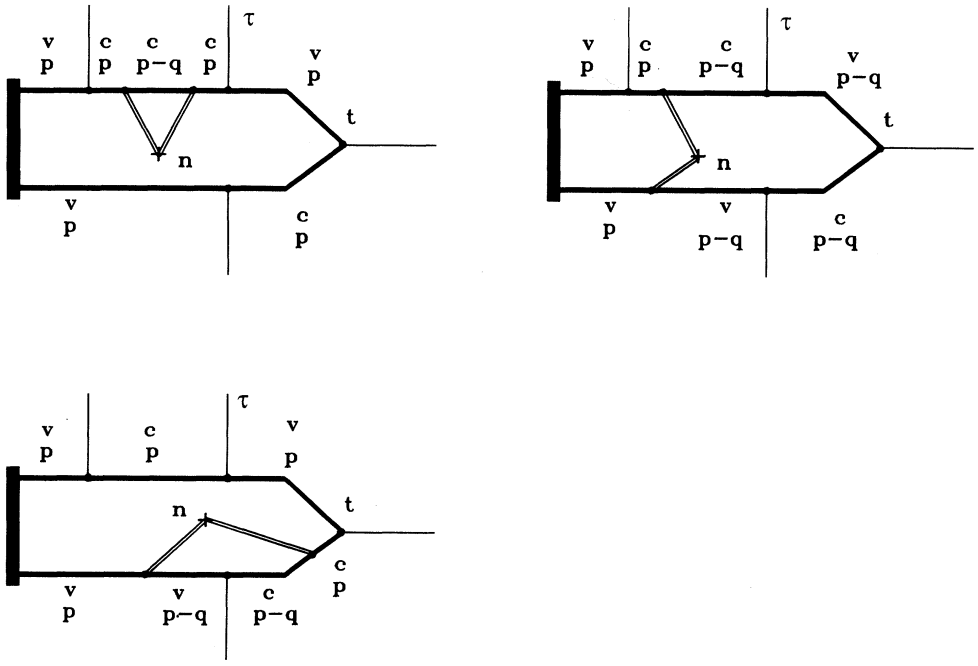

FIG. 5. Influence of impurities on the nonlinear polarization.

Here we introduced the reduced electron-hole mass

$$m_{eh} = m_e m_h/(m_e + m_h).$$

We wish to emphasize that even under the condition $m_e = m_h$ the total phase of a mixed state does not vanish.

Let us note that the same result can be derived in a somewhat different way. To begin with, we calculate the phase acquired by electron-hole state in a field of a single jth impurity center. Within the quasiclassical approximation and assuming that the electron (or hole) kinetic energy is much larger than its potential energy U, we see that the electron and hole during the time interval τ between two successive pulses acquire phases[5]

$$-\int_0^\tau U(\mathbf{R}_j - \mathbf{v}_e t)dt, \quad \int_0^\tau U(\mathbf{R}_j - \mathbf{v}_h t)dt\ , \tag{45}$$

respectively. Here $\mathbf{R}_j = \mathbf{r}_j - \mathbf{r}(0)$ is the relative carrier position at $t = 0$. The first light pulse creates an electron with quasimomentum $\mathbf{p}$ and a hole with quasimomentum $-\mathbf{p}$. The corresponding velocities that should be inserted in Eq. (45) are $\mathbf{v}_e = \mathbf{p}/m_e$ and $\mathbf{v}_h = -\mathbf{p}/m_h$. At the moment $t = \tau$ the second light pulse changes the band indices. Therefore, after the second pulse during the time interval between $t = \tau$ and $t = 2\tau$ the electron state acquires the phase

$$\int_\tau^{2\tau} U[\mathbf{R}_j - \mathbf{v}_h\tau - \mathbf{v}_e(t-\tau)]dt\ , \tag{46}$$

whereas the hole state gets the phase

$$-\int_\tau^{2\tau} U[\mathbf{R}_j - \mathbf{v}_e\tau - \mathbf{v}_h(t-\tau)]dt. \tag{47}$$

The total phase at $t = 2\tau$ is the sum over all the impurities randomly distributed in space. To calculate an observable one should take the configurational average of the expression

$$A = \exp\left(i\sum_j \phi_j\right)\ , \tag{48}$$

where

$$\phi_j = -\int_0^\tau U(\mathbf{R}_j - \mathbf{v}_e t)dt + \int_0^\tau U(\mathbf{R}_j - \mathbf{v}_h t)dt + \int_\tau^{2\tau} U[\mathbf{R}_j - \mathbf{v}_h\tau - \mathbf{v}_e(t-\tau)]dt - \int_\tau^{2\tau} U[\mathbf{R}_j - \mathbf{v}_e\tau - \mathbf{v}_h(t-\tau)]dt.$$

The number of impurities N in a volume $\mathcal{V}$ obeys the Poisson distribution

$$P(N) = \frac{\overline{N}^N}{N!}e^{-\overline{N}}, \tag{49}$$

where $\overline{N}$ is the average number of impurities. The coordinates of impurities are uniformly distributed with the probability density $1/\mathcal{V}$.

The exponent in Eq. (48) is then a product of exponents and we have for the configurational average denoted by $\langle\,\rangle_c$

$$\langle A\rangle_c = \left\langle \left(\frac{1}{\mathcal{V}}\int_{\mathcal{V}} d\mathbf{r}e^{i\phi}\right)^N \right\rangle_N\ , \tag{50}$$

where $\langle\,\rangle_N$ means an average over the Poisson distribution: Since

$$\langle a^N\rangle_N = \sum_{N=0}^{\infty} P(N)a^N = e^{\overline{N}(a-1)}\ , \tag{51}$$

we get for $\langle A\rangle_c$

$$\langle A\rangle_c = \exp\left[\frac{\overline{N}}{\mathcal{V}}\int_{\mathcal{V}} d\mathbf{r}\left(e^{i\phi}-1\right)\right]. \tag{52}$$

Introducing the concentration of impurities $\overline{N}/\mathcal{V} = n$ we see that Eq. (52) coincides with Eq. (44).

For the 3D case where the impurities of two types (donors and acceptors) in equal concentrations are present, we get that the echo signal decay is determined by Eq. (44), where $\exp(i\phi)$ is replaced by $\cos\phi$. Taking for the impurity potential $U(\mathbf{r}) = e/\varepsilon_\infty r$, we transform the decay factor to dimensionless variables

$$\exp\left[-2\pi n(p/m_{eh})^3\tau^3\int_0^\infty R^2dR\int_{-1}^1 dx(1-\cos\phi)\right], \tag{53}$$

where

$$\phi = e^2 f(m_e/m_h, R, x)/\varepsilon_\infty\hbar(v_e + v_h).$$

Here $f(m_e/m_h, R, x)$ is a function of the effective mass ratio m_e/m_h and dimensionless distance variables R and x (another variable that is equal to the cosine of the angle between $\mathbf{p}$ and $\mathbf{r}$),

$$f(m_e/m_h, R, x) = \int_0^1 \frac{dt}{\sqrt{R^2 + (m_e/M)^2t^2 + 2R(m_e/M)xt}} - \int_1^2 \frac{dt}{\sqrt{R^2 + [(m_e/M)t-1]^2 + 2Rx[(m_e/M)t-1]}}$$

$$-\ [\text{terms obtained by replacement } m_e \to m_h, x \to (-x)]\ ,$$

where $M = m_e + m_h$. Equation (53) can be presented in the form

$$\exp\left[-(\tau/\tau_\varphi)^3\right], \tag{54}$$

where τ_φ has the physical meaning of the time of phase breaking.

Let us consider particular cases where the general formula can be simplified. In the quasiclassical case

$$\alpha = e^2/\varepsilon_\infty \hbar(v_e + v_h) \gg 1,$$

we have

$$\tau_\varphi = [15(2\pi)^{1/2}/16\pi^2]^{1/3}\tau_f/\alpha^{1/2}. \tag{55}$$

Then the phase-breaking time is much smaller than the time of flight τ_f, which is given by

$$\tau_f = n^{-1/3}/(v_e + v_h). \tag{56}$$

Roughly, this time can be defined as the time for a particle to traverse the average distance between the Coulomb centers.

Let us turn to the second case where $\alpha \ll 1$. In this case the Coulomb potential can be considered as a perturbation. We can expand $\cos\phi$ and obtain the same law for the echo decay with

$$\tau_\varphi = \tau_f/\alpha^{2/3}[\pi g(m_e/m_h)]^{1/3}, \tag{57}$$

where

$$g(m_e/m_h) = \int_0^\infty R^2 dR \int_{-1}^1 dx\,[f\,(m_e/m_h, R, x)]^2 \ . \tag{58}$$

For $m_e/m_h \to 0$ we have $\tau_\varphi = \tau_f/\alpha^{2/3}(2\pi)^{1/3}$. Now the phase-breaking time is larger than τ_f. In this case, as well as in the previous one, the deviation of the carrier trajectory from the straight line is small during the time τ_φ. In other words, the quasimomentum relaxation time τ_{ee} is much larger than the time of phase breaking τ_φ.

We wish to emphasize that the same form for the law of echo decay given by Eq. (7) for the spectral diffusion and by Eq. (54) for the phase wandering is purely coincidental. One can see this, for instance, by considering within the framework of the same method a *two-dimensional situation.* In this case we get instead of Eq. (53)

$$\exp\left[-n\left(\frac{p}{m_{eh}}\right)^2 \tau^2 \int_0^\infty RdR \int_0^{2\pi} d\theta(1-\cos\phi)\right], \tag{59}$$

with

$$\phi = \frac{e^2}{\varepsilon_\infty \hbar(v_e + v_h)} f(m_e/m_h, R, \cos\theta) \ . \tag{60}$$

We arrive at a general conclusion that in the 2D case the decay law becomes

$$\exp\left[-(\tau/\tau_\varphi)^2\right], \tag{61}$$

with the time of phase breaking τ_φ being proportional to $n^{-1/2}$.

Finally, let us see how our results are changed if the random Coulomb potential is produced by the moving carriers. Instead of Eq. (44) we have

$$\exp\left\{-\sum_{\mathbf{p}'}(F_{c\mathbf{p}'} + 1 - F_{v\mathbf{p}'}) \times \int d\mathbf{r}[1 - \tfrac{1}{2}(e^{i\phi_1} + e^{-i\phi_2})]\right\}, \tag{62}$$

where

$$\begin{aligned}\phi_1 = &-\int_0^\tau U\left[\mathbf{r} - \left(\frac{\mathbf{p}}{m_e} - \frac{\mathbf{p}'}{m_e}\right)t\right]dt \\ &+\int_\tau^{2\tau} U\left[\mathbf{r} - \left(\frac{\mathbf{p}}{m_e} - \frac{\mathbf{p}'}{m_e}\right)t + \frac{\mathbf{p}}{m_{eh}}\tau\right]dt \\ &+\int_0^\tau U\left[\mathbf{r} + \left(\frac{\mathbf{p}}{m_h} - \frac{\mathbf{p}'}{m_e}\right)t\right]dt \\ &-\int_\tau^{2\tau} U\left[\mathbf{r} + \left(\frac{\mathbf{p}}{m_h} - \frac{\mathbf{p}'}{m_e}\right)t - \frac{\mathbf{p}}{m_{eh}}\tau\right]dt,\end{aligned} \tag{63}$$

and one gets ϕ_2 by the replacement $\mathbf{p}'/m_e \to \mathbf{p}'/m_h$ in Eq. (63). Instead of the electron velocity we now have the difference of the carrier velocities and instead of the concentration of the carriers $\sum_{\mathbf{p}'}(F_{c\mathbf{p}'} + 1 - F_{v\mathbf{p}'})$ enters our formulas where one should sum over the quasimomenta $\mathbf{p}'$ of the carriers that produce the Coulomb field. Note that in this case both the law of decay and the conclusion that $\tau_\varphi \propto n^{-1/d}$ remain the same as for the scattering by static impurities.

VI. QUALITATIVE CONSIDERATION

Let us begin with analysis of the case

$$\alpha \gg 1 \ . \tag{64}$$

Consider the electrostatic potential $U(t)$ in the reference frame moving with an electron under consideration (see Fig. 6). The characteristic scale of time variation is

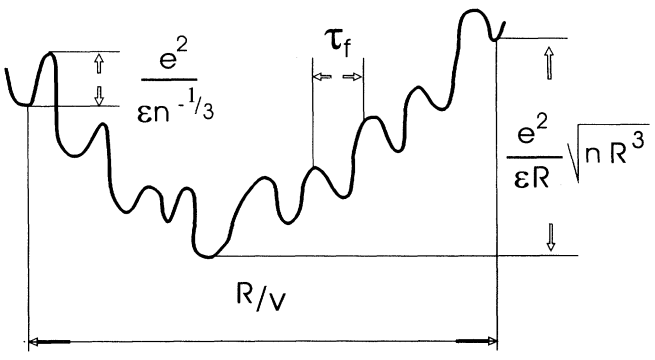

FIG. 6. $U(t)$ in the electron reference frame.

$\tau_f = n^{-1/d}/v$, where v is the larger of the two velocities v_e and v_h [see Eq. (56)], while the characteristic amplitude is $n^{1/d}e^2/\varepsilon_\infty$. We will call this short-scale fluctuations of the long-range Coulomb potential (in contrast to the long-scale fluctuations that are considered below). Therefore for $t \ll \tau_f$ one can expand $U(t)$, retaining the linear term only

$$\delta U \simeq t n^{1/d} e^2/\tau_f \varepsilon_\infty .$$

The corresponding phase change is

$$\delta\phi \simeq t^2 n^{1/d} e^2/\tau_f \hbar \varepsilon_\infty .$$

Hence

$$\tau_\varphi = (\hbar \tau_f \varepsilon_\infty / e^2 n^{1/d})^{1/2} , \tag{65}$$

which coincides with Eq. (55).

The opposite case $\alpha \ll 1$ can be understood in the following way. One can assume that, together with comparatively short-scale fluctuations, the Coulomb potential U has also long-scale fluctuations due to the excess number of carriers with a charge of a particular sign. Let the characteristic spatial scale of such a fluctuation be R. Then the characteristic energy is

$$e^2 (nR^d)^{1/2}/\varepsilon_\infty R = e^2 (nR^{d-2})^{1/2}/\varepsilon_\infty .$$

Due to such a fluctuation the time variation of electron energy is

$$e^2 n^{1/2} (vt)^{d/2-1}/\varepsilon_\infty .$$

It brings about the phase variation

$$e^2 n^{1/2} (v)^{d/2-1} t^{d/2}/\hbar\varepsilon_\infty , \tag{66}$$

which gives $\tau_\varphi \simeq \tau_f/\alpha^{2/d}$ in this case. Let us give an example of order of magnitude estimate of τ_φ in the 3D case. It depends on the average carrier energy. For $\mathcal{E} \simeq 0.2 \times 10^{-12}$ ergs and $n = 7 \times 10^{18}$ cm^{-3}, we get $\tau_\varphi = 15$ fs.

VII. CONCLUSION

One can question the physical nature of the echo decay due to the interaction between the electrons and static impurities. Indeed, when calculating the echo amplitude we could have used the exact electron wave function in the impurity field. This approach, however, might bring us to a misleading conclusion that there is no decay. This seeming contradiction has the following origin. The plane waves of light interact with a pair of electron states having *certain values of quasimomenta* (because of the quasimomentum conservation). The exact states are superpositions of the states with certain values of quasimomenta. Since our treatment of the echo phenomenon relies on the concept of independent states, the coupling among them brings about the decay (cf. with Ref. 18 where irreversibility of *energy-conserving* dipole dephasing for a simple atomic system was found). Since impurity concentration is small, the exact wave functions are close to the plane waves and can be built up by the perturbation method. Mathematically such an approach is totally identical to that we used while starting with plane waves and considering the impurity potential as the cause of phase breaking.

We wish to emphasize once again that the physical considerations put forth in this paper are based on some generic concepts. As for the calculations, they are presented, with the help of diagrammatic techniques, in a straightforward way. The law of the echo decay we derived does not agree with the existing experimental data. We are of the opinion that such a disagreement is of a fundamental nature and manifests the basic need for further experimental and theoretical work.

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[1] E. L. Hahn, Phys. Rev. **80**, 580 (1950).

[2] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. **6**, 567 (1964).

[3] P. C. Becker, H. L. Fragnito, C. H. Brito Cruz, R. L. Fork, J. E. Cunningham, J. E. Henry, and C. V. Shank, Phys. Rev. Lett. **61**, 1647 (1988).

[4] J.-Y. Bigot, M. T. Portella, R. W. Schoenlein, J. E. Cunningham, and C. V. Shank, Phys. Rev. Lett. **67**, 636 (1991).

[5] V. L. Gurevich, M. I. Muradov, and D. A. Parshin, Europhys. Lett. **12**, 375 (1990).

[6] J. R. Klauder and P. W. Anderson, Phys. Rev. **125**, 912 (1962).

[7] J. L. Black and B. I. Halperin, Phys. Rev. B **16**, 2879 (1977).

[8] L. Root and J. L. Skinner, J. Chem. Phys. **81**, 5310 (1984).

[9] Ch. Lonsky, P. Thomas, and A. Weller, Phys. Rev. Lett. **63**, 652 (1989); J. Lumin. **45**, 77 (1990).

[10] D. Bennhard, P. Thomas, A. Weller, M. Lindberg, and S. W. Koch, Phys. Rev. B **43**, 8934 (1991).

[11] K. El Sayed, L. Banyai, and H. Haug, Phys. Rev. B **50**, 1541 (1994).

[12] O. V. Konstantinov and V. I. Perel', Zh. Eksp. Teor. Fiz. **39**, 197 (1960) [Sov. Phys. JETP **12**, 142 (1961)].
[13] S. V. Gantsevich, V. D. Kagan, R. Katilius, and E. Rzaev, Zh. Eksp. Teor. Fiz. **87**, 1734 (1984) [Sov. Phys. JETP **60**, 997 (1984)].
[14] V. L. Gurevich, A. I. Larkin, and Yu. A. Firsov, Fiz. Tverd. Tela (Leningrad) **4**, 131 (1962) [Sov. Phys. Solid State **4**, 185 (1962)].
[15] K. El Sayed, S. Shuster, H. Haug, F. Herzel, and K. Henneberger, Phys. Rev. B **49**, 7337 (1994).
[16] V. L. Gurevich, M. I. Muradov, and D. A. Parshin, in *Proceedings of 20th International Conference on the Physics of Semiconductors, Thessaloniki, 1990*, edited by E. M. Anastassakis and J. D. Joannopulos (World Scientific, Singapore, 1990), p. 1843.
[17] S. L. Ginzburg, Zh. Eksp. Teor. Fiz. **63**, 2084 (1973) [Sov. Phys. JETP **36**, 1198 (1973)].
[18] J. Yeh and J. H. Eberly, Phys. Rev. A **22**, 1124 (1980).