FEMTOSECOND PHOTON ECHO IN SEMICONDUCTORS. DIAGRAMMATIC APPROACH

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Within the framework of the time ordered diagram technique a brief derivation of density matrix equations for the case of a two-band semiconductor is given. A diagram technique for the echo phenomenon is developed. The difference between single—atom dipole band (Yeh and Eberly model) and excitonic spectrum of semiconductors is discussed. A mechanism responsible for the observed very short times of the echo decay (of the order of a few femtoseconds) in semiconductors is investigated. Previous results of Gurevich *et. al.* concerning the femtosecond echo decay law are confirmed within the framework of the eikonal approximation.

The purpose of the present work is to develop a theory describing the early nonequilibrium stage of evolution of optically excited semiconductors. Particularly, this paper is motivated by the observations of two—pulse femtosecond photon echoes from the band–to–band transitions in semiconductors [1,2] and by the previous theoretical work [3] where the quasiclassical theory was developed describing the echo decay.

Briefly the echo phenomenon can be described as follows. The action of a short laser pulse can cause direct transitions binding electron states with a given quasimomentum $\mathbf{p}$ in the valence and conduction bands. As a result, a continuum of independent two—level systems in $\mathbf{p}$–space is excited.

After the first pulse, all the states have the same phases. But consequently a macroscopic observable, say, an induced polarization, will disappear because of the differences in the energies of the excited states. After the action of the second pulse, the states are changed in such a way that the evolution of the phases of the individual states is time reversed. As a result, at the moment $t = 2\tau$ a reconstruction of the coherence takes place and an echo pulse is observed.

Within the range of carrier concentrations investigated in the experiment [1], the time of the echo decay was smaller than 11 fs. According to the observations, the time of the decay was sensitive to the carrier concentration. It means that the interaction between the carriers should be crucial for the echo decay. In [1,2], to interpret their experimental findings, it has been assumed that the echo decay was determined by the time of quasimomentum relaxation due to the screened electron-electron interaction. However, as the following estimates show, this time is usually too long to support such an explanation for the echo decay.

We are giving now an estimate of the period of plasma oscillations $2\pi/\omega_p$, which is needed for the Debye screening to build up. The range of its variation on the experiment was $2\pi/\omega_p = 50 - 250$ fs. This means that the time of the experiment is usually not sufficient for the Debye screening to build up [4].

The second time is the time of intercarrier collisions, τ_{ee}. At $n = 2 \cdot 10^{17}$ cm^{-3} we have for this time the following lower estimate $\tau_{ee} > 100$ fs. This means that during the time of the echo decay no collisions can take place and the electron motion during this short time can be considered as free [4].

On the very short time scale, the interaction with a random Coulomb field the source of which is the electrons and holes appearing after the laser pulses (or already existing static impurities) cannot be treated as usual collisions and the corresponding expressions do not contain δ-functions describing the energy conservation. In this situation one can either rederive kinetic equations for so short times or totally avoid the kinetic equations approach and concentrate on the direct calculation of physical observables. Recent attempts mainly have been made to derive quantum kinetic equations [5–9], that turn out to be non—Markovian ones. Such equations have been derived using different formalisms. For instance, they have been derived within the framework of the Keldysh formalism using a set of Dyson equations as a starting point. Irrelevant which formalism has been used to get a closed set of kinetic equations some assumptions have to be made. In the case of the Keldysh formalism the generalized Kadanoff-Baym ansatz [5] has been used.

To be self–complete in the following section we give a brief derivation of the non-Markovian equations [5–9] for the density matrix using the slightly modified Konstantinov-Perel' technique [10] (see also [11]). It is convenient to use this diagram technique because it allows to visualize each physical process. Furthermore, we will use one—time density matrix approach as a starting point avoiding the lengthy algebra of the Keldysh nonequilibrium two—time Green function technique in the course of derivation of these equations and show how Markovian limit as well as the opposite limiting case can be obtained.

1. Equations for density matrix

First of all, let us fix the notations. We introduce the density matrix with the diagonal components $n_{c\mathbf{k}} = \langle a^+_{c\mathbf{k}} a_{c\mathbf{k}} \rangle$, $n_{v\mathbf{k}} = \langle a^+_{v\mathbf{k}} a_{v\mathbf{k}} \rangle$ and nondiagonal ones $p_{\mathbf{k}} = \langle a^+_{v\mathbf{k}} a_{c\mathbf{k}} \rangle$, $p^*_{\mathbf{k}} = \langle a^+_{c\mathbf{k}} a_{v\mathbf{k}} \rangle$, where $a^+_{i\mathbf{k}}(a_{i\mathbf{k}})$ is a creation (annihilation) operator for an electron in a state $i\mathbf{k}$, $n_{c\mathbf{k}}(n_{v\mathbf{k}})$ is a distribution function of carriers in the conduction (valence) band while $p_{\mathbf{k}}$ describes a mixed electron-hole state (coherent properties).

We will extensively use later on an operator $1/\partial_t$ defined as

$$\frac{1}{\partial_t} f(t) = \int_0^t dt\, f(t) \tag{1}$$

with the simple property

$$\frac{1}{\partial_t + i\omega} f(t) = e^{-i\omega t} \frac{1}{\partial_t} e^{i\omega t} f(t). \tag{2}$$

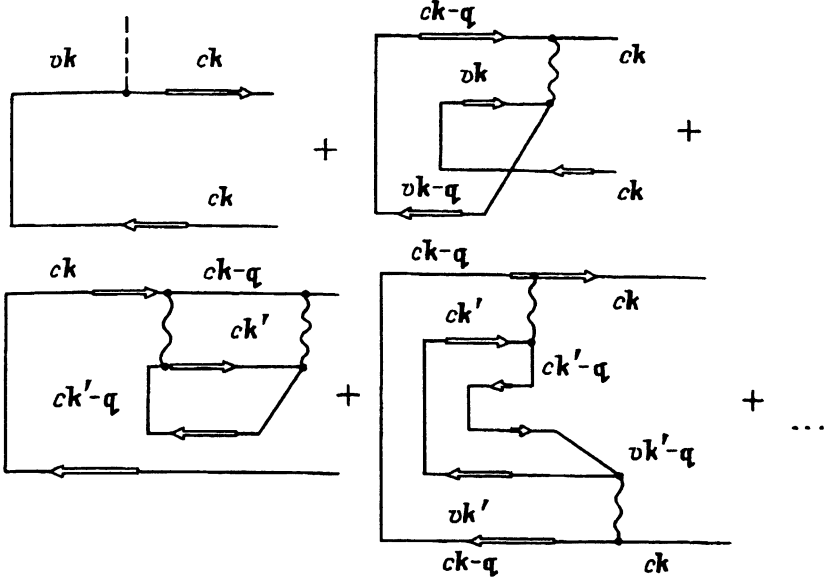

Fig. 1. Diagrammatic equation for $n_{\mathbf{ck}}$.

First we consider equation for $n_{\mathbf{ck}}$. We represent this quantity by the set of the diagrams depicted in Fig.1. The events are time ordered in our diagrams, the time variable t running from the left to the right (in the «positive» direction) in each diagram. The interaction events can occur at the moments $t_1, t_2, \ldots$ $(0 < t_1 < t_2 < \ldots t)$ and the consecutive integration is needed over $t_1, t_2, \ldots$. The integration gives rise to a number of operators $(\partial_t + i\sum_j \mathcal{E}_j - i\sum_k \mathcal{E}_k)^{-1}$, where j stands for the «positive» propagator lines and k for the «negative» ones (each propagator carrying the quasimomentum $\mathbf{p}$ and the energy $\mathcal{E}_{\mathbf{p}}$). The operators can be obtained by the following rule. One should impose vertical lines upon the diagram between each two adjacent vertices and after the last one. The operator which contains the energies of the propagator lines dissected should then be brought into correspondence with each of the obtained vertical section. Note that diagrams that can be obtained from the presented ones by changing the arrangement of interaction points are not depicted. An interaction point (in our case of the Coulomb potential represented by a wavy line in the diagrams) on the upper (lower) line (propagator or antipropagator) contributes an additional factor $-i(i)$. According to the band indices of the two propagators (running in opposite directions) which are connected with the vertical bar in the diagram, each vertical bar represents a definite density matrix component. Notice that the nondiagonal density matrix component running in the «negative» direction contributes an additional factor (-1). Finally, note that the diagrams should also be supplemented by the similar ones with $c\mathbf{k}' \to v\mathbf{k}'$, $c\mathbf{k}' - \mathbf{q} \to v\mathbf{k}' - \mathbf{q}$.

In order to illustrate these rules, we consider the fourth diagram in Fig.1.

$$\frac{1}{\partial_t} iU_{\mathbf{q}} \frac{1}{\partial_t - i\Delta\mathcal{E}_{cv}} (-iU_{\mathbf{q}}) n_{\mathbf{ck}-\mathbf{q}} (-p_{\mathbf{k}'}) p^*_{\mathbf{k}'-\mathbf{q}} (1 - n_{\mathbf{ck}}). \tag{3}$$

Here $U_{\mathbf{q}} = 4\pi e^2/\epsilon q^2$ is the Fourier transform of the Coulomb potential and $\Delta\mathcal{E}_{\alpha,\beta}$ means the sum of two single—particle energy differences

$\Delta\mathcal{E}_{\alpha,\beta} = \mathcal{E}_{\alpha\mathbf{k}-\mathbf{q}} - \mathcal{E}_{\alpha\mathbf{k}} + \mathcal{E}_{\beta\mathbf{k}'} - \mathcal{E}_{\beta\mathbf{k}'-\mathbf{q}}$. Further on we omit higher order terms, so in the written expression we neglect $n_{c\mathbf{k}}$ as compared with unity. The diagram under consideration describes the following process: an electron in the pure state $c\mathbf{k}-\mathbf{q}$ and electrons in mixed states $p_{\mathbf{k}'}$ and $p_{\mathbf{k}'-\mathbf{q}}$ interact via the Coulomb field. This results in the creation of states in the conduction band with $\mathbf{k}, \mathbf{k}-\mathbf{q}$ and in the valence band with $\mathbf{k}'$, $\mathbf{k}'-\mathbf{q}$; the subsequent «scattering» of these states finally creates the electron in the state $c\mathbf{k}$. Briefly, this diagram visualizes the appearance of the electron in the state $c\mathbf{k}$ after «scattering» of the electron in the state $c\mathbf{k}-\mathbf{q}$ on the polarization $p_{\mathbf{k}'}p^*_{\mathbf{k}'-\mathbf{q}}$. In the same way one can interpret each diagram. Diagrams in Fig.1. allow to write the equation for the diagonal component of the density matrix

$$\partial_t n_{c\mathbf{k}} = (\partial_t n_{c\mathbf{k}})_{coh} + (\partial_t n_{c\mathbf{k}})_{occ} + (\partial_t n_{c\mathbf{k}})_{pol} \tag{4}$$

where $(\partial_t n_{c\mathbf{k}})_{coh}$ is a coherent part

$$(\partial_t n_{c\mathbf{k}})_{coh} = \Big(-iV^{cv}_{\mathbf{k}} - i\sum_{\mathbf{q}} U_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}\Big)p^*_{\mathbf{k}} + c.c. \tag{5}$$

Here $V^{cv}_{\mathbf{k}}$ is a matrix element of an external field and the second term in the parentheses $\sum_{\mathbf{q}} U_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}$ is responsible for the renormalization of the Rabi frequency [12,13]. On the other hand, the same term is responsible for the excitonic effect (see the following section and the second diagram in Fig.1). $(\partial_t n_{c\mathbf{k}})_{occ}$ is connected with the particle-particle «scattering» while $(\partial_t n_{c\mathbf{k}})_{pol}$ describes the «scattering» on the polarization.

$$(\partial_t n_{c\mathbf{k}})_{occ} = \sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cc}} + c.c.\right)(n_{c\mathbf{k}'} n_{c\mathbf{k}-\mathbf{q}} - n_{c\mathbf{k}} n_{c\mathbf{k}'-\mathbf{q}})$$

$$+ \sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cv}} + c.c.\right)\Big((1-n_{v\mathbf{k}'-\mathbf{q}})n_{c\mathbf{k}-\mathbf{q}} - n_{c\mathbf{k}}(1-n_{v\mathbf{k}'})\Big), \tag{6}$$

$$\begin{aligned}(\partial_t n_{c\mathbf{k}})_{pol} = &-\sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cv}} p^*_{\mathbf{k}'} p_{\mathbf{k}'-\mathbf{q}} + c.c.\right)(n_{c\mathbf{k}-\mathbf{q}} - n_{c\mathbf{k}}) \\ &-\sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cc}} p_{\mathbf{k}'} p^*_{\mathbf{k}'-\mathbf{q}} + c.c.\right)(n_{c\mathbf{k}-\mathbf{q}} - n_{c\mathbf{k}}) \\ &-\sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cc}} p^*_{\mathbf{k}} p_{\mathbf{k}-\mathbf{q}} + c.c.\right)(n_{c\mathbf{k}'} - n_{c\mathbf{k}'-\mathbf{q}}) \\ &-\sum_{\mathbf{q}\mathbf{k}'} U^2_{\mathbf{q}} \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cv}} p^*_{\mathbf{k}} p_{\mathbf{k}-\mathbf{q}} + c.c.\right)(n_{v\mathbf{k}'} - n_{v\mathbf{k}'-\mathbf{q}}).\end{aligned} \tag{7}$$

Diagrams for $p_{\mathbf{k}}$ can be depicted in a similar way and we have

$$\Big(\partial_t + i(\mathcal{E}_{c\mathbf{k}} - \mathcal{E}_{v\mathbf{k}})\Big)p_{\mathbf{k}} = (\partial_t p_{\mathbf{k}})_{coh} + (\partial_t p_{\mathbf{k}})_{occ} + (\partial_t p_{\mathbf{k}})_{pol}, \tag{8}$$

$$(\partial_t p_{\mathbf{k}})_{coh} = \left(iV_{\mathbf{k}}^{cv} - i\sum_{\mathbf{q}} U_{\mathbf{q}} p_{\mathbf{k}-\mathbf{q}}\right)(n_{c\mathbf{k}} - n_{v\mathbf{k}}) - ip_{\mathbf{k}} \sum_{\mathbf{q}} U_{\mathbf{q}}(n_{v\mathbf{k}-\mathbf{q}} - n_{c\mathbf{k}-\mathbf{q}}). \quad (9)$$

The term in Eq.(9) $\sum_{\mathbf{q}} U_{\mathbf{q}}(n_{c\mathbf{k}-\mathbf{q}} - n_{v\mathbf{k}-\mathbf{q}})$ can be regarded as an exchange contribution to the energy renormalization [12,13].

$$\begin{aligned}
(\partial_t p_{\mathbf{k}})_{occ} = &- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t + i\Delta\mathcal{E}_{cc} + i\mathcal{E}_{cv\mathbf{k}}} p_{\mathbf{k}} n_{c\mathbf{k}'-\mathbf{q}} \\
&- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t + i\Delta\mathcal{E}_{cv} + i\mathcal{E}_{cv\mathbf{k}}} p_{\mathbf{k}}(1 - n_{v\mathbf{k}'}) \\
&+ \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t - i\Delta\mathcal{E}_{cc} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} p_{\mathbf{k}-\mathbf{q}} n_{c\mathbf{k}'} \\
&+ \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t - i\Delta\mathcal{E}_{cv} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} p_{\mathbf{k}-\mathbf{q}}(1 - n_{v\mathbf{k}'-\mathbf{q}}) \\
&- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t - i\Delta\mathcal{E}_{vc} + i\mathcal{E}_{cv\mathbf{k}}} p_{\mathbf{k}} n_{c\mathbf{k}'} \\
&- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t - i\Delta\mathcal{E}_{vv} + i\mathcal{E}_{cv\mathbf{k}}} p_{\mathbf{k}}(1 - n_{v\mathbf{k}'-\mathbf{q}}) \\
&+ \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t + i\Delta\mathcal{E}_{vc} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} p_{\mathbf{k}-\mathbf{q}} n_{c\mathbf{k}'-\mathbf{q}} \\
&+ \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \frac{1}{\partial_t + i\Delta\mathcal{E}_{vv} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} p_{\mathbf{k}-\mathbf{q}}(1 - n_{v\mathbf{k}'}), \qquad (10)
\end{aligned}$$

$$\begin{aligned}
(\partial_t p_{\mathbf{k}})_{pol} = &\sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cc} + i\mathcal{E}_{cv\mathbf{k}}} + \frac{1}{\partial_t - i\Delta\mathcal{E}_{vv} + i\mathcal{E}_{cv\mathbf{k}}}\right) p_{\mathbf{k}} p_{\mathbf{k}'} p^*_{\mathbf{k}'-\mathbf{q}} \\
&+ \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \left(\frac{1}{\partial_t + i\Delta\mathcal{E}_{cv} + i\mathcal{E}_{cv\mathbf{k}}} + \frac{1}{\partial_t - i\Delta\mathcal{E}_{vc} + i\mathcal{E}_{cv\mathbf{k}}}\right) p_{\mathbf{k}} p^*_{\mathbf{k}'} p_{\mathbf{k}'-\mathbf{q}} \\
&- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \left(\frac{1}{\partial_t - i\Delta\mathcal{E}_{cc} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} + \frac{1}{\partial_t + i\Delta\mathcal{E}_{vv} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}}\right) p_{\mathbf{k}-\mathbf{q}} p^*_{\mathbf{k}'} p_{\mathbf{k}'-\mathbf{q}} \\
&- \sum_{\mathbf{q}\mathbf{k}'} U_{\mathbf{q}}^2 \left(\frac{1}{\partial_t - i\Delta\mathcal{E}_{cv} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}} + \frac{1}{\partial_t + i\Delta\mathcal{E}_{vc} + i\mathcal{E}_{cv\mathbf{k}-\mathbf{q}}}\right) p_{\mathbf{k}-\mathbf{q}} p_{\mathbf{k}'} p^*_{\mathbf{k}'-\mathbf{q}}. \qquad (11)
\end{aligned}$$

Here we introduced the interband oscillation frequency $\mathcal{E}_{cv\mathbf{k}} = \mathcal{E}_{c\mathbf{k}} - \mathcal{E}_{v\mathbf{k}}$. Equations (4) and (8) are still integral-differential equations. These equations coincide with the quantum kinetic equations which were derived by other methods in Ref. [5–9] if we restore conventional notations using the definition of the operator $1/\partial_t$. They can be simplified and reduced to the effective differential Bloch equations in the case when the characteristic energy transfer in

two-particle collisions is much larger than the inverse time variation scale of $n_{c\mathbf{k}}$ and $P_{\mathbf{k}} = p_{\mathbf{k}} \exp(+i(\mathcal{E}_{c\mathbf{k}} - \mathcal{E}_{v\mathbf{k}})t)$. In this case, because of a weak time dependence of the abovementioned quantities we can consider them as ones commutating with operators $(\partial_t + i\Delta\mathcal{E}_{\alpha\beta})^{-1}$. Remembering the definition of $1/\partial_t$, the remaining operators can be converted into

$$\frac{1}{\partial_t \pm i\Delta\mathcal{E}_{\alpha\beta}} = \mp iP\frac{1}{\Delta\mathcal{E}_{\alpha\beta}} + \pi\delta(\Delta\mathcal{E}_{\alpha\beta}) = \pi\delta_{\mp}(\Delta\mathcal{E}_{\alpha\beta}) \tag{12}$$

and we get the generalization of the kinetic equations for the two band case [12,13].

Therefore, the scattering integral approach is adequate only for describing fast processes while slow fields acting on particles should be treated in a different way. It is obvious that such a simplification is not justified at sufficiently short times, particularly to describe such memory sensitive coherent effect as the echo.

The second limiting case of these equations can be obtained by discarding the differences of the single particle energies $\Delta\mathcal{E}_{\alpha\beta}$ in operators $(\partial_t + i\Delta\mathcal{E}_{\alpha\beta})^{-1}$. In this case Eq.(4) and (8) coincide with the equations derived by another method in [9]. Even after such a simplification the derived quantum kinetic equations appear to be too complicated to get the analytical solution of the echo decay problem but prove to be useful in numerical simulations. Haug *et. al* [9] analyzed the obtained set of equations numerically and found the decay law which turned out to be in a qualitative agreement with the previous results [3,4].

2. Echo in diagram representation

The echo phenomenon can be represented by a set of time-ordered diagrams (Fig.2). One sees from this figure that the echo formation can be understood as the sequence of light—electron interactions (straight lines with the points on ends in Fig.2) changing the electron states (lines in the figure). The corresponding operator is given by

$$V_j = \frac{e}{2m\omega i} e^{i(\omega t - \mathbf{k}_j\mathbf{r})}\mathbf{E}_j(t)\mathbf{p} + c.c. \tag{13}$$

Here $j = 1(2)$ for the first (second) pulse. The first pulse at $t = 0$ creates a mixed state from the initial one which is supposed to be pure and described by an occupation number (a bar in the diagram). At the points of interaction the band index is changed (v→ c or vice versa). An individual state acquires the phase $(\mathcal{E}_{c\mathbf{p}+\mathbf{k}_1} - \mathcal{E}_{v\mathbf{p}})\tau$ during the time between the pulses. The double action of the second pulse at $t = \tau$ (see Fig.2) changes the state in such a way that it looks as reversed one in time and the phase shift acquired from $t = \tau$ to t is $(\mathcal{E}_{v\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2} - \mathcal{E}_{c\mathbf{p}+\mathbf{k}_2})(t - \tau)$. As a consequence, at $t = 2\tau$ the total phase $(\mathcal{E}_{v\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2} - \mathcal{E}_{c\mathbf{p}+\mathbf{k}_2} + \mathcal{E}_{c\mathbf{p}+\mathbf{k}_1} - \mathcal{E}_{v\mathbf{p}})\tau$ vanishes (we neglect the term $(\mathbf{k}_1 - \mathbf{k}_2)(\mathbf{v}_v + \mathbf{v}_c)\tau$) and the coherence is restored. This results in an echo signal generated by the total dipole momentum of all the states (the final point in Fig.2 represents an observable).

For the polarization current we have from the diagrams

$$\mathbf{j}(t,\mathbf{r}) = -2e^{i\omega t}\sum_{\mathbf{p}}\langle c\mathbf{p}+\mathbf{k}_2|\mathbf{j}|v\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2\rangle e^{-it(\mathcal{E}_{v\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2} - \mathcal{E}_{c\mathbf{p}+\mathbf{k}_2}+\omega)} \cdot V_2^2 \cdot V_1^*$$

$$\times\, e^{-i\tau(\mathcal{E}_{c\mathbf{p}+\mathbf{k}_1} - \mathcal{E}_{v\mathbf{p}+\mathbf{k}_1-\mathbf{k}_2}-\omega)} e^{-i\tau(\mathcal{E}_{c\mathbf{p}+\mathbf{k}_2} - \mathcal{E}_{v\mathbf{p}}-\omega)}(n_{c\mathbf{p}+\mathbf{k}_1} - n_{v\mathbf{p}}) + c.c.$$

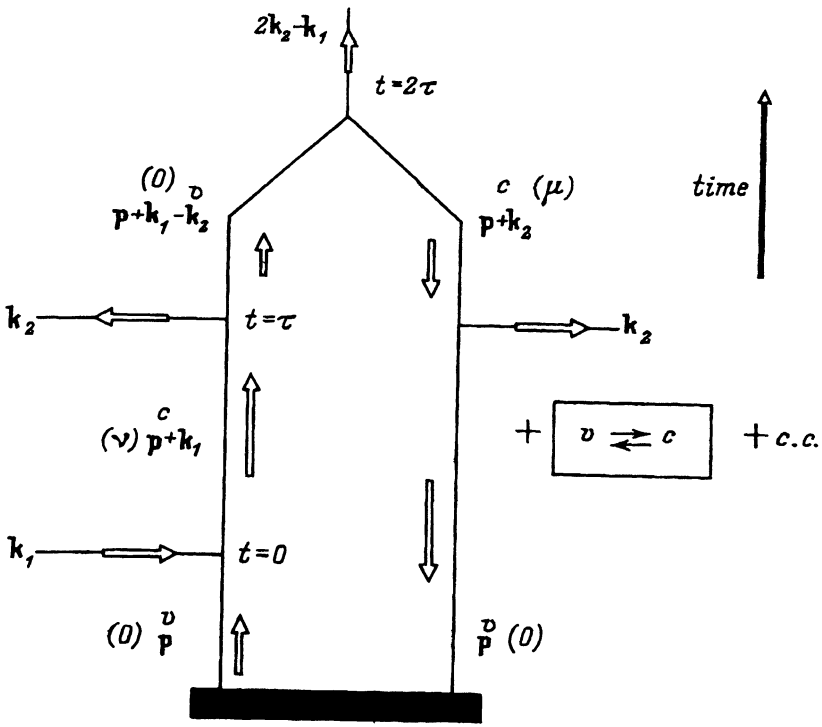

Fig. 2. Set of diagrams representing the formation of the two-pulse echo.

Here $V_j = e\tau_j(\mathbf{E}_j\mathbf{p}_{vc})/(2m\omega)$ are the matrix elements of the light—electron interaction, $n_{c\mathbf{p}}$ and $n_{v\mathbf{p}}$ are the initial electron occupation numbers in the conduction and valence bands, $\mathbf{j}$ is the current density operator. It is clear from the diagram that the direction of the echo pulse propagation is $\mathbf{k} = 2\mathbf{k}_2 - \mathbf{k}_1$, if one takes into account the quasimomentum conservation at each point of interaction.

In Appendix, we present a calculation of the polarization current beyond the perturbation theory in the case of rectangular pulses of external driving fields [14] (see also [15]). This case also can be visualized making use of the diagram technique.

We now show how our diagram formalism may be extended to take into account the excitonic spectrum of excitations in semiconductors. In this case the following question arises. As it was shown by Yeh and Eberly [16], the externally stimulated reconstruction of the dipole coherence is not possible under the conditions of the single-atom dipole band (an «atom» with a single ground state and an excited band containing a set of levels). The result can be explained in this way (see Fig.2, note indices in brackets). Let the first pulse of the external field create a superposition of ground and excited ν-states. The second pulse leads to the transition from the ν state to the ground one and from the ground state to the μ state (the states ν and μ are, in general, not the same). To calculate an observable one should sum over ν and μ the product of exponents $\exp(i\omega_\nu\tau)\exp(-i\omega_\mu(t-\tau))$ (quantum beats) and the observable will vanish if one assumes a sufficiently broad distribution of energies ω_ν.

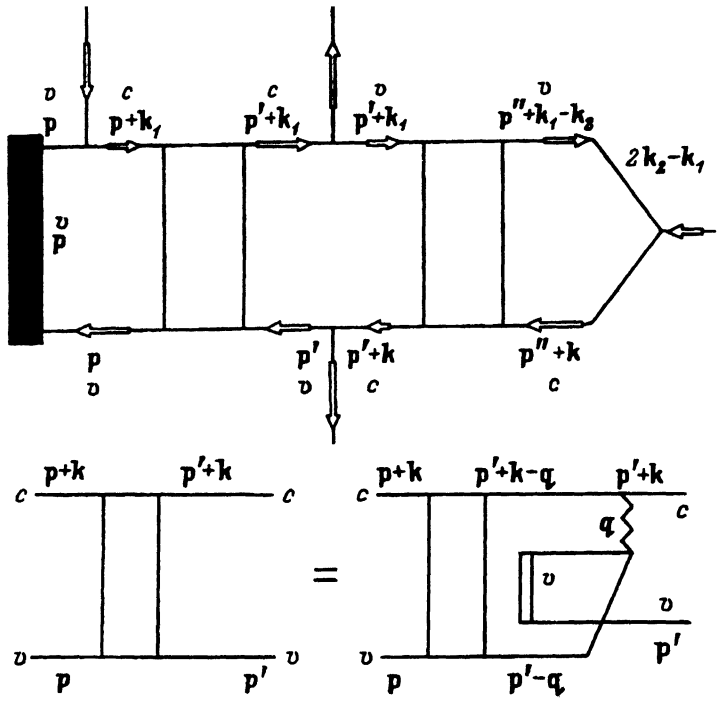

Fig. 3. Echo diagram including the electron—hole Coulomb attraction.

So one could come to the conclusion that if one takes into account the Coulomb attraction of holes and electrons there would be no echo because of the analogy between the Wannier-exciton and the «atom» spectrum.

However, there is a difference between the Yeh and Eberly model and the excitonic spectrum of semiconductors. Namely, in the case of semiconductors we have a selection rule for the quasimomentum. It is the quasimomentum conservation and the completeness of the exciton wave functions that brings about the condition that the states ν and μ must be the same. As a consequence one can describe the interaction of semiconductors with light by a set of equations identical to the Bloch equations for independent two-level systems [13] even if one takes into account the Coulomb interaction between electrons and holes.

Now we present how one can incorporate the excitonic structure of excitations in our general diagram formalism. To take into account excitonic effects we insert additional blocks and the echo in this situation is described by the diagram in Fig.3. The blocks in the diagram contain the result of the Coulomb attraction between the electrons and holes. For the blocks one can obtain the equation (in a diagram representation the equation is presented in Fig.3).

$$\frac{1}{\partial_t + i(\mathcal{E}^c_{\mathbf{p}'+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}'})} W^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1} =$$

$$= \frac{1}{\partial_t + i(\mathcal{E}^c_{\mathbf{p}'+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}'})} \sum_{\mathbf{q}} \frac{iU_q}{\partial_t + i(\mathcal{E}^c_{\mathbf{p}'-\mathbf{q}+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}'-\mathbf{q}})} W^{\mathbf{p}'-\mathbf{q},\mathbf{p}'-\mathbf{q}+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1}. \quad (14)$$

We rewrite Eq.(14)

$$\left[\partial_t + i(\mathcal{E}^c_{\mathbf{p}'+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}'})\right]\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1} = \sum_{\mathbf{q}} iU_{\mathbf{q}}\Gamma^{\mathbf{p}'-\mathbf{q},\mathbf{p}'-\mathbf{q}+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1} \tag{15}$$

where we introduce

$$\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1} = \frac{1}{\partial_t + i(\mathcal{E}^c_{\mathbf{p}'+\mathbf{k}_1} - \mathcal{E}^v_{\mathbf{p}'})} W^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1}. \tag{16}$$

Adopting the notation for the center of mass $\mathbf{R}$ and for the relative coordinates $\mathbf{r}$ we obtain the differential equation describing the excitonic spectrum

$$\left[\partial_t - i\left(\frac{\nabla^2_r}{2m_{eh}} + \frac{\nabla^2_R}{2M}\right)\right]\Gamma(\mathbf{r},\mathbf{R}) = iU(\mathbf{r})\Gamma(\mathbf{r},\mathbf{R}). \tag{17}$$

The solution is

$$\Gamma(\mathbf{r},\mathbf{R}) = \sum_{\mathbf{K},\nu} e^{i\mathbf{K}\mathbf{R}}\psi_\nu(\mathbf{r})e^{-iE_{\nu,\mathbf{K}}t}C_{\nu,\mathbf{K}} \tag{18}$$

where ψ_ν are the eigenfunctions of the Coulomb problem and $E_{\nu,\mathbf{K}}$ are the eigenvalues of Eq.(17). For $\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1}$ we obtain

$$\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1} = \sum_\nu \psi_\nu\left(\mathbf{p}' + \mathbf{k}_1\frac{m_h}{M}\right)e^{-iE_{\nu,\mathbf{k}_1}t}C_{\nu,-\mathbf{k}_1}. \tag{19}$$

The initial condition

$$\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1}|_{t=0} = \delta_{\mathbf{p}\mathbf{p}'} \tag{20}$$

determines the coefficients $C_{\nu,\mathbf{K}}$

$$\sum_\nu \psi_\nu\left(\mathbf{p}' + \mathbf{k}_1\frac{m_h}{M}\right)C_{\nu,-\mathbf{k}_1} = \delta_{\mathbf{p}\mathbf{p}'}, \tag{21}$$

$$C_{\nu,-\mathbf{k}_1} = \psi^*_\nu\left(\mathbf{p} + \mathbf{k}_1\frac{m_h}{M}\right). \tag{22}$$

The result taking into account the Coulomb attraction can be presented as

$$\Gamma^{\mathbf{p}',\mathbf{p}'+\mathbf{k}_1}_{\mathbf{p},\mathbf{p}+\mathbf{k}_1}(t) = \sum_\nu \frac{\psi^*_\nu(\mathbf{p}+\mathbf{k}_1 m_h/M)\psi_\nu(\mathbf{p}'+\mathbf{k}_1 m_h/M)}{\partial_t + iE_{\nu,\mathbf{k}_1}}\delta(t). \tag{23}$$

As it should be expected we get the expression for the excitonic Green function. One can easily obtain the expression for the second block noting that the latter is the complex conjugate counterpart of the first (with the corresponding change of the arguments)

$$\sum_\nu \frac{\psi^*_\nu(\mathbf{p}''+\mathbf{k}_1-\mathbf{k}_2+(2\mathbf{k}_2-\mathbf{k}_1)m_h/M)\psi_\nu(\mathbf{p}'+\mathbf{k}_1-\mathbf{k}_2+(2\mathbf{k}_2-\mathbf{k}_1)m_h/M)}{\partial_t - iE_{\nu,2\mathbf{k}_2-\mathbf{k}_1}}\delta(t) \tag{24}$$

Finally we compute the polarization current (see Fig.3)

$$\mathbf{j}(\mathbf{r},t) = -2e^{i\omega t} \sum_{\nu,\nu',p,p',p''} \langle cp'' + \mathbf{k}_2|\mathbf{j}|vp'' + \mathbf{k}_1 - \mathbf{k}_2\rangle$$
$$\times \frac{\psi^*_\nu(\mathbf{p}'' + \mathbf{k}_1 - \mathbf{k}_2 + (2\mathbf{k}_2 - \mathbf{k}_1)m_h/M)\psi_\nu(\mathbf{p}' + \mathbf{k}_1 - \mathbf{k}_2 + (2\mathbf{k}_2 - \mathbf{k}_1)m_h/M)}{\partial_t - i(E_{\nu,2\mathbf{k}_2-\mathbf{k}_1} - \omega)}$$
$$\times \delta(t-\tau)\langle vp'|V^{(2)}|cp' + \mathbf{k}_2\rangle\langle vp' + \mathbf{k}_1 - \mathbf{k}_2|V^{(2)}|cp' + \mathbf{k}_1\rangle$$
$$\times \frac{\psi^*_{\nu'}(\mathbf{p} + \mathbf{k}_1 m_h/M)\psi_{\nu'}(\mathbf{p}' + \mathbf{k}_1 m_h/M)}{\partial_t + i(E_{\nu',\mathbf{k}_1} - \omega)} \delta(t)\langle cp + \mathbf{k}_1|V^{(1)}|vp\rangle + c.c.$$

The latter expression can be simplified under the following assumptions: i) the momentum dependence of the external perturbation matrix elements is weak, ii) the light wave length is much larger than the Bohr radius of the exciton. The first assumption allows us to factor out matrix elements in the sum over p' and this expression due to the second assumption and the wave function completeness gives

$$\sum_{\mathbf{p}'} \psi_\nu\Big(\mathbf{p}' + \mathbf{k}_1 - \mathbf{k}_2 + (2\mathbf{k}_2 - \mathbf{k}_1)m_h/M\Big)\psi_{\nu'}(\mathbf{p}' + \mathbf{k}_1 m_h/M) \simeq \delta_{\nu\nu'}. \quad (25)$$

The rest sums over p and p'' give the well-known Sommerfeld or Elliot excitonic enhancement factor.

$$\sum_{\mathbf{p}''} \psi_\nu(\mathbf{p}'') \sum_{\mathbf{p}} \psi_\nu(\mathbf{p}) = \psi^2_\nu(\mathbf{r})|_{r=0}. \quad (26)$$

Due to Eq.(25) and (26) one can be convinced that we have again two level systems but discriminated in the last case by the excitonic quantum number ν.

3. Echo decay

Let us discuss the influence of charged impurities randomly distributed in the space on the echo signal. Then we shall generalize the result of our consideration to the case of moving charged particles (i.e. carriers). The impurity potential is assumed to be static and long ranged. We shall work within the framework of the high energy or eikonal approximation, i.e. we suppose that the potential is sufficiently smooth so that the electron quasimomentum variation under its action, q, is much smaller than the quasimomentum p [17] (see also [4]). In the expressions like $\mathcal{E}_{\mathbf{p}-\mathbf{q}} - \mathcal{E}_{\mathbf{p}}$ we omit terms quadratic in q, i.e. $\mathcal{E}_{\mathbf{p}-\mathbf{q}} - \mathcal{E}_{\mathbf{p}} = -\mathbf{qp}/m$. To take into account the influence of impurities, one should insert the points of interaction with impurities in 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 we have the terms of the perturbation theory each represented by a diagram of the type shown in Fig.4. To make a consideration as simple as possible, we assume that the time interval between the pulses is much longer than the durations of any pumping pulse. In our further calculations we will consider δ–type pulses.

The presented diagrams with omission of a common factor

$$2e\mathbf{v}_{cv}e^{2i\omega\tau}V_1V_2^2 \quad (27)$$

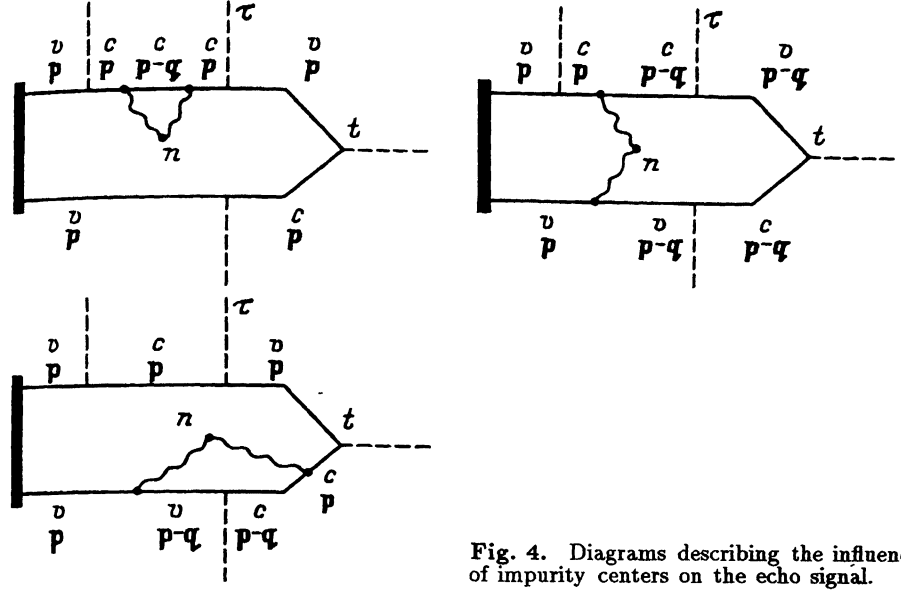

Fig. 4. Diagrams describing the influence of impurity centers on the echo signal.

are equal to the following expressions

$$n\frac{1}{2}\int_0^\tau dt_1 \int_0^\tau dt_2(-i)^2 U_{\mathbf{q}} U_{-\mathbf{q}} e^{-i\mathbf{q}\mathbf{v}_c t_1} e^{i\mathbf{q}\mathbf{v}_c t_2}, \tag{28}$$

$$n\int_0^\tau dt_1 \int_{-\infty}^\tau dt_2(i)(-i) U_{\mathbf{q}} U_{-\mathbf{q}} e^{-i\mathbf{q}\mathbf{v}_c t_1} e^{i\mathbf{q}\mathbf{v}_v t_2}, \tag{29}$$

$$n\int_0^\tau dt_1 \int_\tau^{2\tau} dt_2(-i)^2 U_{\mathbf{q}} U_{-\mathbf{q}} e^{-i\mathbf{q}\mathbf{v}_c t_1} e^{i\mathbf{q}\mathbf{v}_v t_2} e^{i\mathbf{q}(\mathbf{v}_c - \mathbf{v}_v)\tau} \tag{30}$$

where $U_{\mathbf{q}}$ is the Fourier transform of the potential. Let us note that in addition to the well known in kinetics diagrams describing the usual «in—» and «out—terms» there are some special types describing the correlation of propagators through impurities during various time intervals. For example, the third diagram takes into account such a correlation in the electron motion in the conduction band and valence band at the time intervals from zero to τ and from τ to 2τ. Summing over $\mathbf{q}$ we have for the sum of all diagrams

$$\frac{1}{2}\Big[-i\int_0^\tau U(\mathbf{r}-\mathbf{v}_c t)dt + i\int_\tau^{2\tau} U\Big(\mathbf{r}-\mathbf{v}_c(t-\tau)-\mathbf{v}_v\tau\Big)dt$$
$$+ i\int_0^\tau U(\mathbf{r}-\mathbf{v}_v t)dt - i\int_\tau^{2\tau} U\Big(\mathbf{r}-\mathbf{v}_v(t-\tau)-\mathbf{v}_c\tau\Big)dt\Big]^2.$$

Taking into account all orders in U and n we obtain as the sum of all diagrams within the eikonal approximation

$$\exp\Big(-n\int d\mathbf{r}(1-e^{i\phi})\Big), \tag{31}$$

where

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

Here we introduced the masses of electron and hole, m_e and m_h respectively ($m_h > 0$) and the reduced electron—hole mass $m_{eh} = m_e m_h/(m_e+m_h)$.

Let us note that the result can be derived in a slightly different way [3]. To begin with, let us calculate the phase acquired by an electron—hole state in a field of the single i-th impurity center. In the field U, during the time interval τ between pulses an electron and a hole acquire phases

$$-\int_0^{\tau} U(\mathbf{R}_i-\mathbf{v}_e t)dt, \qquad \int_0^{\tau} U(\mathbf{R}_i-\mathbf{v}_h t)dt. \tag{32}$$

The light creates an electron with a momentum $\mathbf{p}$ and a hole with a momentum $-\mathbf{p}$. The corresponding velocities which should be inserted in (32) are $\mathbf{v}_e = \mathbf{p}/m_e$, $\mathbf{v}_h = -\mathbf{p}/m_h$. At the time $t=\tau$ the second light pulse changes band indices. After the second pulse from $t=\tau$ to $t=2\tau$ the electron state acquires the phase

$$\int_{\tau}^{2\tau} U\Big(\mathbf{R}_i-\mathbf{v}_h\tau-\mathbf{v}_e(t-\tau)\Big)dt \tag{33}$$

while the hole state acquires the phase

$$-\int_{\tau}^{2\tau} U\Big(\mathbf{R}_i-\mathbf{v}_e\tau-\mathbf{v}_h(t-\tau)\Big)dt. \tag{34}$$

Note, that just before the second pulse at $t=\tau$ the relative electron—impurity radius-vector was $\mathbf{R}_i-\mathbf{v}_h\tau$ and after the pulse due to the band indices exchange it becomes $\mathbf{R}_i-\mathbf{v}_h\tau-\mathbf{v}_e(t-\tau)$. The total phase at $t=2\tau$ is a sum over spatial coordinates of impurities randomly distributed in space . To compute an observable one should take a configurational average of the expression

$$A = \exp\Big(i\sum_i \phi_i\Big) \tag{35}$$

where

$$\phi_i = -\int_0^{\tau} U(\mathbf{R}_i - \mathbf{v}_e t)dt + \int_0^{\tau} U(\mathbf{R}_i - \mathbf{v}_h t)dt$$
$$+ \int_{\tau}^{2\tau} U(\mathbf{R}_i - \mathbf{v}_h\tau - \mathbf{v}_e(t-\tau))dt$$
$$- \int_{\tau}^{2\tau} U(\mathbf{R}_i - \mathbf{v}_e\tau - \mathbf{v}_h(t-\tau))dt.$$

Making use of the Holtsmark method [18] in the course of computing a configurational average we obtain for $\langle A\rangle_c$

$$\langle A\rangle_c = \exp\left(\frac{\bar{N}}{V}\int_V d\mathbf{r}\Big(\exp(i\phi) - 1\Big)\right). \tag{36}$$

Introducing the concentration of impurities $\bar{N}/V = n$ we note that Eq.(36) coincides with Eq.(31).

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

For the impurities of two types (donors and acceptors) with equal concentrations we get that the echo signal decay is determined by the expression (31) where $\exp(i\phi)$ is replaced by $\cos\phi$. Inserting for the impurity potential $U(\mathbf{r}) = e/\varepsilon r$ (ε is the dielectric susceptibility) we rewrite the decay factor through the dimensionless variables

$$\exp\left(-2\pi n(p/m_{eh})^3\tau^3\int_0^{\infty} r^2 dr\int_{-1}^{1} dx(1-\cos\phi)\right), \tag{37}$$

where $\phi = e^2 f(m_e/m_h, r, x)/\varepsilon\hbar(v_e + v_h)$ and $f(m_e/m_h, r, x)$ is a function of m_e/m_h, dimensionless r and x, which is equal to the cosine of the angle between $\mathbf{p}$ and $\mathbf{r}$. This equation can be presented in the form $\exp\left(-(\tau/\tau\varphi)^3\right)$ where τ_φ is the time of phase breaking.

The result obtained deserves some comments. In the course of the decay law derivation a system dimensionality was essential. So, if we have a two—dimensional situation we will get a decay law $\exp(-(\tau/\tau_\varphi)^2)$, τ_φ being proportional to $n^{-1/2}$. Note also, that the obtained expression Eq.(37) should be multiplied by Eq.(27) and summation over $\mathbf{p}$ should be performed.

Now let us consider particular cases where the general formula can be simplified. In the quasiclassical case, $\alpha = e^2/\varepsilon\hbar(v_e + v_h) \gg 1$, we have

$$\tau_\varphi = \left(15(2\pi)^{1/2}/16\pi^2\right)^{1/3}\tau f/\alpha^{1/2}. \tag{38}$$

The phase breaking time is much smaller than the time of particle flight over the mean distance between the impurities, $\tau_f = n^{-1/3}/(v_e + v_h)$.

Now 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 $\tau_\varphi = \tau_f/\alpha^{2/3}(2\pi)^{1/3}$ when $m_e/m_h \to 0$. Now the phase breaking time is larger than the time of flight. 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 momentum relaxation time, τ_{ee}, is much larger than the phase breaking time τ_φ.

Finally let us see how our results are changed if the random Coulomb potential is produced by the moving carriers. The only change is that, instead of the electron velocity the difference in the carrier velocities and instead of n the concentration of carriers $n(\mathbf{p}')$ enter our formulae and one should take a sum over the momenta $\mathbf{p}'$ of the carriers which produce the Coulomb field.

Let us give an estimate of τ_φ in the three—dimensional case. It depends on the average carrier energy. In order to estimate the energy, we use uncertainty principle $\mathcal{E} \sim \hbar/\tau$. For $\mathcal{E} \sim 10^{-13}$ erg and $n = 7\cdot 10^{18}$ cm^{-3}, we get $\tau_\varphi \simeq 20$ fs.

The derived quantum kinetic equations appear to be too complicated to get the analytical solution of the echo decay problem. However, they seem to be useful in the numerical simulation. Recently the work of H. Haug *et. al.* [9] have attracted our attention where authors have neglected in quantum kinetic equations the differences of single particle energies and reduced them to a set of simplified equations. Obtained set of equations was analyzed numerically. They found the decay law which turned out to be in a qualitative agreement with previous results of papers [3,4].

We treat the echo decay by a rather different approach. The main assumption consists of regarding the field produced by photocarriers in an optically excited semiconductor and interband polarization as a field originated from charges randomly distributed in space. Furthermore, within the framework of eikonal approximation we take into account the influence of randomly distributed charges on the coherent state. It is demonstrated that the phase breaking time is proportional to $n^{-1/d}$ where n is the carrier (impurity) concentration and d is the dimensionality of the system. The calculated phase breaking times are of the typical order of tens of femtoseconds. The time τ_φ appears to be usually shorter than other characteristic times such as τ_{ee}; it describes the rate of decay of the coherent properties of an electron—hole system.

The concentration dependence of these times and their order of magnitude are in agreement with experiment while the nonexponential decay law is not consistent with experiment where an exponential decay law has been reported. The situation can be regard as far from being comprehensive. There is a need for further work in this direction.

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Appendix

The two-pulse echo in the case of rectangular pumping pulses (neglecting the decay processes) can be presented by the diagram in Fig.5. For the first block we have

$$\frac{1}{\partial_t + i\mathcal{E}_1}\delta(t-t_b)+\frac{1}{\partial_t + i\mathcal{E}_1}(-iV_{12})e^{+i\omega t}\frac{1}{\partial_t + i\mathcal{E}_2}(-iV_{21})e^{-i\omega t}\frac{1}{\partial_t + i\mathcal{E}_1}\delta(t-t_b)+\ldots$$

$$= e^{-i\mathcal{E}_1(t_e-t_b)}\sum_{n=0}^{\infty}\frac{1}{\partial_t}\left(\frac{-|V_{12}|^2}{\partial_t + i\mathcal{E}}\frac{1}{\partial_t}\right)^n\delta(t-t_b).$$

Here we introduce the detuning $\mathcal{E} = \mathcal{E}_2 - \mathcal{E}_1 - \omega$. Summation over n gives

$$\exp\left(-i\mathcal{E}_1(t_e-t_b)\right)\frac{1}{\partial_t + \frac{|V_{12}|^2}{\partial_t + i\mathcal{E}}}\delta(t-t_b). \tag{39}$$

The operator can be simplified

$$\frac{1}{\partial_t + \frac{|V_{12}|^2}{\partial_t + i\mathcal{E}}} = \frac{\mathcal{E}}{4\Omega}\left(\frac{1}{\partial_t + i(\frac{\mathcal{E}}{2}-\Omega)} - \frac{1}{\partial_t + i(\frac{\mathcal{E}}{2}+\Omega)}\right)$$

$$+\frac{1}{2}\left(\frac{1}{\partial_t + i(\frac{\mathcal{E}}{2}-\Omega)} + \frac{1}{\partial_t + i(\frac{\mathcal{E}}{2}+\Omega)}\right).$$

Here Ω stands for the generalized detuning $\sqrt{|V_{12}|^2 + \mathcal{E}^2/4}$. We can rewrite this expression as

$$\exp\left(-i\mathcal{E}_1(t_e-t_b)\right)\exp\left(-i\mathcal{E}/2(t_e-t_b)\right)$$

$$\times\left(\cos\Omega(t_e-t_b) + \frac{i\mathcal{E}}{2\Omega}\sin\Omega(t_e-t_b)\right)\theta(t_e-t_b). \tag{40}$$

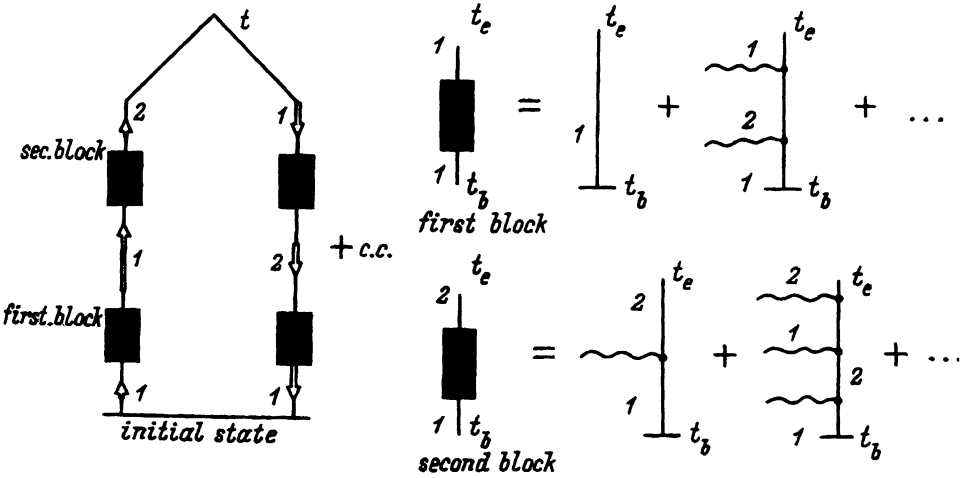

Fig. 5. Echo in the case of rectangular external pulses.

For the second block we have

$$(-iV_{21})e^{-i\omega t_e}e^{-i\mathcal{E}_1(t_e-t_b)}\frac{1}{\partial_t+i\mathcal{E}}\frac{1}{\partial_t+\frac{|V_{12}|^2}{\partial_t+i\mathcal{E}}}\delta(t-t_b) \tag{41}$$

or in the conventional way

$$\frac{(-iV_{21})}{\Omega}e^{-i\omega t_e}e^{-i\mathcal{E}_1(t_e-t_b)}e^{-i\mathcal{E}/2(t_e-t_b)}\sin\Omega(t_e-t_b)\theta(t_e-t_b). \tag{42}$$

The expressions for the rest blocks are obvious now and we have all we need to write an analytical expression for the diagram describing polarization

$$\begin{aligned}\mathbf{P} = i\sum\langle 1|\mathbf{d}|2\rangle e^{-i\mathcal{E}(t-2\tau-\tau_1-\tau_2)}e^{-i\omega t}\frac{V_{12}}{\Omega}\left(\frac{V_{21}^s}{\Omega}\right)^2 \\ \times\frac{1}{2}\left(\sin 2\Omega\tau_1 + i\frac{\mathcal{E}}{\Omega}\sin^2\Omega\tau_1\right)\sin^2\Omega^s\tau_2(F_2-F_1)+c.c.\end{aligned} \tag{43}$$

Here, τ_1, τ_2 are the first and the second external pulse durations, the superscript s stands for the second pulse, F_1 and F_2 are occupation numbers for levels 1 and 2. In the case of semiconductors, $\mathcal{E}$ should be replaced by $\mathcal{E}_{c\mathbf{k}}-\mathcal{E}_{v\mathbf{k}}-\omega$, V_{12} by $\mathbf{E}\mathbf{d}_{cv}$ etc.

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