Theory of Femtosecond Photon Echo Decay in Semiconductors

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Abstract

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 interaction of the mixed state (associated with inter-band transitions) with an unscreened random Coulomb potential of the photocarriers or with a random static potential of the impurities.

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Qualitative physical consideration enabling one to visualize the process of echo decay are presented. We have introduced a new time characteristic of a system of interacting electrons. This is the the time of phase breaking, τ_φ which we calculate within the eikonal approximation using a diagrammatic techniques. It is shown that τ_φ is typically much shorter than both the period of plasma oscillations and the time of electron-electron collisions. The screening of Coulomb potential cannot build up during this time. τ_φ is proportional to $n^{-1/d}$ (where n is the carrier concentration, d the dimensionality of a system) which is consistent with the experimental results. However, the derived law of echo decay of the form $\exp\left[-(\tau/\tau_\varphi)^d\right]$ does not agree with the existing experimental data.

as a

1 Introduction.

The echo phenomenon in 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 a quantum well structures [4]. The time evolution of the phase sensitive mixed quantum states responsible for echo phenomena in physical systems are of great importance for understanding of various mechanisms of phase relaxation of electron states as well as its nature. The present paper is devoted to investigation of the possible mechanism of the echo decay and can be considered as a continuation and further development of the earlier paper [5].

The echo is a nonlinear effect which can be in general described as follows. Let an observable quantity, say, a macroscopic electric dipole moment D be a sum of a great number of contributions from N independent subsystems (or particles). Then a perturbation of a very short duration (a "shock") excites each individual particle j into a mixed transition state between stationary states with energies $\mathcal{E}_1^j$ and $\mathcal{E}_2^j$. Observables in such a state oscillate with the frequency $\omega_j = \mathcal{E}_2^j - \mathcal{E}_1^j$ (to make expressions in the intermediate calculations less cumbersome we will often put $\hbar = 1$) and the total dipole moment is varying as

$$\mathrm{D}(t) = \sum_j \mathrm{d}_j \exp(-i\omega_j t) \qquad (1)$$

At $t = 0$ it has a macroscopic value $D \propto Nd$ ($N \gg 1$) but later due to differences in frequencies ω_j decreases and practically vanishes for times greater than the $1/\Delta\omega$, where $\Delta\omega$ is a typical frequency shift. For broad distribution of frequencies ω_j it is of the order of the width of the frequency distribution in the exciting pulse. After the time interval $t \gg 1/\Delta\omega$ there is a second pulse which reverses the quantum state in such a way that its frequency changes the sign $\omega_j \to \mathcal{E}_1^j - \mathcal{E}_2^j = -\omega_j$ while conserving the phase $(-\omega_j\tau)$ acquired to the moment $t = \tau$. Thus we have

$$\mathrm{D}(t) = \sum_j v_j^2 \mathrm{d}_j \exp[i\omega_j(t-\tau) - i\omega_j\tau]. \qquad (2)$$

Here we introduce factors v_j^2 which characterize the change in the dipole moment amplitude d_j due to the second pulse. We see that at the moment $t = 2\tau$ the phases of individual oscillators vanish and $\mathrm{D}(t)$ emerges again as a macroscopic quantity (and the corresponding echo pulse is generated).

This can be illustrated by a diagram (see Fig.1). We use a time-ordered diagrammatic techniques (see [6], cf. with [7]). A line on a diagram represents

the state of a jth particle and points indicate the actions of the pulses. At $t = 0$ we had a stationary state. The first shock creates an oscillating transition state. The second (double) shock reverses it at $t = \tau$ and finally at $t = 2\tau$ a radiation pulse is observed. Eq.(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 interaction of the particles with the surrounding medium. We can include the fluctuations into 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.)

$$\frac{1}{\partial_t + i\omega_j} + \frac{1}{\partial_t + i\omega_j}(-i\delta\omega_j)\frac{1}{\partial_t + i\omega_j} + \ldots = \frac{1}{\partial_t + i\omega_j + i\delta\omega_j}. \qquad (3)$$

Using Eq.(3) we obtain the total dipole moment as

$$\mathrm{D}(t) = \sum_j v_j^2 \mathrm{d}_j \frac{1}{\partial_t - i\omega_j - i\delta\omega_j(t)}\delta(t-\tau)\frac{1}{\partial_t + i\omega_j + i\delta\omega_j(t)}\delta(t). \qquad (4)$$

This is a random quantity that should be averaged over all possible frequency variations $\delta\omega_j(t)$:

$$< \mathrm{D}(2\tau) >= \sum_j v_j^2 \mathrm{d}_j \left\langle \exp\left(i\int_\tau^{2\tau} \delta\omega_j(t)dt - i\int_0^\tau \delta\omega_j(t)dt\right)\right\rangle. \qquad (5)$$

Here the averaging is denoted by the angular brackets. The averaging procedure will be considered in detail below. Now let us note that there are two principal mechanisms leading to the decay of the echo signal. The first one is a simple damping resulting mainly from large (and fast) fluctuations ("collisions"). These are the processes which, in particular, bring about relaxation of the occupancy numbers of energy levels. The law of the echo decay due to them is

$$< \mathrm{D}(2\tau) >= \sum_j v_j^2 \mathrm{d}_j \exp(-2\tau\nu_j) \qquad (6)$$

with $\nu_j = (\nu_j^{(1)} + \nu_j^{(2)})/2$, where $\nu_j^{(1)}$ and $\nu_j^{(2)}$ are the damping rates (or the inverse relaxation times) for the levels $\mathcal{E}_1^j$ and $\mathcal{E}_2^j$.

The second decay mechanism is related to the phenomenon known as *spectral diffusion* [8],[9], in which small frequency fluctuations play a principal role. As a result of such fluctuations the oscillation frequency suffers a sort of random walk so that one can write

$$< \mathrm{D}(2\tau) >= \sum_j v_j^2 \mathrm{d}_j < \exp\left[i\tau(\delta\omega' - \delta\omega)\right] > \qquad (7)$$

where $\delta\omega'$ and $\delta\omega$ are the frequency value after independent wandering during the time τ. If we assume that frequency wandering looks like a usual diffusion and take the probability distribution of random variable $\xi = \delta\omega' - \delta\omega$ as given by

$$W_t(\xi) = \frac{1}{2\sqrt{\pi D t}} \exp\left(-\frac{\xi^2}{4Dt}\right) \qquad (8)$$

we obtain for the echo decay due to the spectral diffusion:

$$< \mathrm{D}(2\tau) >= \sum_j v_j^2 \mathrm{d}_j \int_{-\infty}^{+\infty} W_\tau(\xi) \exp(i\xi\tau) d\xi = \sum_j v_j^2 \mathrm{d}_j \exp\left(-D\tau^3\right) \qquad (9)$$

The diffusion constant D can be expressed through the mean square of the frequency wandering as $< \delta\omega_t^2 >=< \xi_t^2 > /2 = Dt$. Thus there are two limiting cases of echo decay: the law $\exp(-\nu t)$ and the law $\exp(-Dt^3)$ where ν and D are the damping and diffusion constants, respectively. For various distributions of damping and diffusion constants the summation over j can change the resulting formula even for pure cases of damping or diffusion (it is interesting to compare our results also with [9] where the law $\exp(-At^2)$ where A is another constant is obtained for two- level systems in glasses).

The echo decay, or optical dephasing, was studied in semiconductors in a number of papers (see [10], [11], [12]). The obtained results usually corre-

spond to consideration of damping, i.e. only the action of short range and/or rapidly varying parts of the perturbations are finally taken into account. In this way, in paper by Lonski et al.[11] the echo decay in disordered semiconductors is considered and an exponential law of decay is found. The difference between their and our case can be understood as follows. In Ref.[11], due to a 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 consider here a long range Coulomb interaction where one can look upon the phase variation as a sum of a great number of relatively small contributions. As a result, we get a sort of diffusional motion of the phase and the echo dies off during the time τ_φ, which may be much smaller than a time characterizing collisions of the particles interacting according to the Coulomb law. We are going to show that for a number of cases of interest the spectral diffusion may be quite effective as a dephasing mechanism with the characteristic decay time much smaller than the usual relaxation time due to "collisions".

2 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

$$\mathcal{E}^c_{\rm p} = p^2/2m_e, \quad \mathcal{E}^v_{\rm p} = -E_g - p^2/2m_h$$

where p is the electron quasimomentum. The mixed quantum state which is responsible for the echo phenomenon in this case is represented by the nondiagonal element of density matrix or, simply, by the product of the

wave function specified by the values of their quasimomenta:

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

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

where Ω_{p} is equal to

$$\Omega_{\mathrm{p}} = E_g + \frac{p^2}{2m_e} + \frac{p^2}{2m_h} \tag{10}$$

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 signal is observed just as it has been shown in the previous section. Two drawings in Fig.3. reflect the fact that a state with the frequency of oscillation $+\Omega_{\mathrm{p}}$ can be created by mixing two pure quantum states (diagonal elements of the density matrix) $\psi^*_{\mathrm{p}c}\psi_{\mathrm{p}c}$ and $\psi^*_{\mathrm{p}v}\psi_{\mathrm{p}v}$. The bar at the bottom of the drawing corresponds to the occupancy numbers of these states, $F_{c\mathrm{p}}$ and $F_{v\mathrm{p}}$.

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 uppermost arrow in the drawing. Other points correspond to the matrix elements of electron interband transitions and describe the actions of the two laser pulses with the wave vectors k_1 and k_2. Thus we have:

$$\begin{aligned} \mathrm{j}(t, \mathrm{k}_1 - 2\mathrm{k}_2) &= \mathrm{e}\sum_{\mathrm{p}} < v|\mathrm{v}|c > \frac{V^2_{2p}}{\partial_t - i(\Omega_{\mathrm{p}} + (\mathrm{k}_1 - \mathrm{k}_2)\mathrm{v}_v + \mathrm{k}_2\mathrm{v}_c)}\delta(t-\tau) \\ &\times \frac{iV_{1p}}{\partial_t + i(\Omega_{\mathrm{p}} + \mathrm{k}_1\mathrm{v}_c)}\delta(t)(F_{c\mathrm{p}+\mathrm{k}_1} - F_{v\mathrm{p}}). \end{aligned} \tag{11}$$

Here we put $\mathcal{E}_{\mathrm{p}+\mathrm{k}} \simeq \mathcal{E}_{\mathrm{p}} + \mathrm{kv}$ because of the inequality $k \ll p$. Note that a point in a line going along the time (the wave function) contributes $(-i)$ while a point in a line going in the opposite direction (the complex conjugated wave function) contributes $(+i)$. The energy in the resolvent enters with the plus

sign for a wave function line and with the minus sign for a complex conjugated wave function line. Due to spatial homogeneity the total quasimomentum is conserved at any interaction point. The current Eq.(11) has the form typical for the echo phenomena and at $t = 2\tau$ is equal to

$$\mathrm{j}(t, \mathrm{k}_1 - 2\mathrm{k}_2) = \mathrm{e} \sum_{\mathrm{p}} < v|\mathrm{v}|\mathrm{c} > \exp\left[i(\mathrm{k}_1 - \mathrm{k}_2)(\mathrm{v}_v - \mathrm{v}_c)\tau\right] i V_{2p}^2 V_{1p} (F_{\mathrm{cp}} - F_{v\mathrm{p}}) \tag{12}$$

In Eq. (11) we put $F_{\mathrm{cp+k}} \simeq F_{\mathrm{cp}}$.

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

$$U(\mathrm{r}, t) = \sum_{\mathrm{q}\omega} U_{\mathrm{q}\omega} e^{-i\omega t + i\mathrm{qr}}. \tag{13}$$

The mean value of $U(\mathrm{r}, t)$ vanishes so that $< U_{\mathrm{q}\omega} >= 0$ and

$$< U^*(\mathrm{r}_1, t_1) U(\mathrm{r}, t) >= \sum_{\mathrm{q}\omega} |U_{\mathrm{q}\omega}|^2 e^{-i\omega(t - t_1) + i\mathrm{q}(\mathrm{r} - \mathrm{r}_1)} \tag{14}$$

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

$$U(\mathrm{r}) = \sum_j u(\mathrm{r} - \mathrm{r}_j) = \sum_{j\mathrm{q}} u_{\mathrm{q}} e^{i\mathrm{q}(\mathrm{r} - \mathrm{r}_j)} \tag{15}$$

where r_j is the position of the jth impurity. At first we study the role of short range fast field fluctuations with wave vectors $q \sim p$ and frequencies $\omega \sim \mathcal{E}_p$. Due to the action of such fluctuations damping constants appear for electron states. We illustrate it by the following diagram (see Fig. 4.). There are two fluctuations with the space and time dependence of the type $\exp(\pm i\omega t \pm i\mathrm{qr})$

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

$$\sum_{\mathrm{q}}(-i)^2 < \mathrm{p}+\mathrm{q}|U(\mathrm{r},t)|\mathrm{p} > \frac{1}{\partial_t + i\Omega_{\mathrm{p}} + i(\mathcal{E}^c_{\mathrm{p+q}} - \mathcal{E}^c_{\mathrm{p}})} < \mathrm{p}|U(\mathrm{r},t)|\mathrm{p}+\mathrm{q} > \tag{16}$$

Assuming for the first point the time dependence $e^{-i\omega t}$ and for the second point $e^{+i\omega t}$ we have

$$-\sum_{\mathrm{q}}|U_{\mathrm{p+q,p}}|^2 \frac{1}{\partial_t + i\Omega_{\mathrm{p}} + i(\mathcal{E}^c_{\mathrm{p+q}} - \mathcal{E}^c_{\mathrm{p}} - \omega)} = -i\delta\mathcal{E}^c_{\mathrm{p}}, \tag{17}$$

and an analogous expression for the second drawing. Considering the time evolution of the envelope function one can exclude the large frequency Ω_{p} from the denominator, We see that this expression is none other than a contribution to the electron energy and to the damping due to field fluctuations:

$$-i\delta\mathcal{E}^c_{\mathrm{p}} = -\pi\sum_{\mathrm{q}}|U_{\mathrm{p+q,p}}|^2\delta(\mathcal{E}^c_{\mathrm{p+q}} - \mathcal{E}^c_{\mathrm{p}} - \omega) + i\sum_{\mathrm{q}}|U_{\mathrm{p+q,p}}|^2\frac{1}{\mathcal{E}^c_{\mathrm{p+q}} - \mathcal{E}^c_{\mathrm{p}} - \omega} \tag{18}$$

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

$$\Omega_{\mathrm{p}} \Rightarrow \Omega_{\mathrm{p}} + \delta\Omega_{\mathrm{p}} - \frac{\nu}{2}. \tag{19}$$

The frequency variation $\delta\Omega_{\mathrm{p}}$ vanishes in the final expression so that only damping is important. For the fast echo phenomena such as the femtosecond echo an estimate shows that during the delay time for the echo signal, τ, the electron collisions are very improbable because of the inequality $\tau \ll \tau_{\mathrm{p}}$.

Thus the main role in the decay should play the long range and slowly varying part of the field fluctuations. For carrier concentration of the order of $10^{17}\mathrm{cm}^{-3}$ the electron-electron collision time τ_{ee} is greater than 100 fs. Since the time of echo decay is of the order of 10 fs [3] we should exclude these collisions as a cause of echo decay. On the other hand, for such carrier concentrations the period of plasma oscillations is also about 100 fs so that Debye screening of field fluctuations cannot build up during the echo evolution. The experiment shows [3] that the time of echo decay depends on the carrier concentration, n, as $n^{-1/d}$. This fact points out that field fluctuations are mainly due to Coulomb field of the carriers created by the laser pulses.

3 Femtosecond echo decay.

For so 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 quasimomentum, q, under the action of a smooth and long-range Coulomb potential is small as compared with quasimomentum p (this makes the so called high energy or *eikonal* approximation [13]), so that

$$|\mathcal{E}_{\mathrm{p-q}} - \mathcal{E}_{\mathrm{p}}| \ll \mathcal{E}_{\mathrm{p}} \tag{20}$$

where

$$\mathcal{E}_{\mathrm{p-q}} - \mathcal{E}_{\mathrm{p}} = -\mathrm{qp}/m.$$

To take into account the influence of charged impurities , one should insert 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 one gets 16 terms of the perturbation theory. As

an example, three of them are represented by diagrams in Fig. 5. To make our consideration as simple as possible, we assume that the time interval between the pulses is much longer than the durations of the pumping pulse. In our further calculations we will consider δ-pulses.

The presented diagrams are equal to the following expressions

$$n\frac{1}{\partial_t}\delta(t-\tau)\frac{1}{\partial_t}(-iU_{\mathrm{q}})e^{i\mathrm{q}\mathrm{v}_c t}\frac{1}{\partial_t}(-iU_{-\mathrm{q}})e^{-i\mathrm{q}\mathrm{v}_c t}\frac{1}{\partial_t}\delta(t), \tag{21}$$

$$n\frac{1}{\partial_t}\delta(t-\tau)\frac{1}{\partial_t}(-iU_{-\mathrm{q}})e^{-i\mathrm{q}\mathrm{v}_c t}\frac{1}{\partial_t}(iU_{\mathrm{q}})e^{i\mathrm{q}\mathrm{v}_c t}\frac{1}{\partial_t}\delta(t), \tag{22}$$

$$n\frac{1}{\partial_t}(iU_{-\mathrm{q}})e^{-i\mathrm{q}\mathrm{v}_c t}\frac{1}{\partial_t}e^{i\mathrm{q}\mathrm{v}_c t}e^{-\mathrm{q}\mathrm{v}_v t}\delta(t-\tau)\frac{1}{\partial_t}(iU_{\mathrm{q}})e^{i\mathrm{q}\mathrm{v}_v t}\frac{1}{\partial_t}\delta(t). \tag{23}$$

Let us note that in addition to the diagrams commonly 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 band and valence band during the time intervals from 0 to τ and from τ to 2τ. Summing over q we get for the sum of all 16 diagrams

$$\frac{n}{2}\{-i\int_0^{\tau}U(\mathrm{r}-\mathrm{v}_c t)dt+i\int_{\tau}^{2\tau}U\left[\mathrm{r}-\mathrm{v}_c(t-\tau)-\mathrm{v}_v\tau\right]dt$$
$$+i\int_0^{\tau}U(\mathrm{r}-\mathrm{v}_v t)dt-i\int_{\tau}^{2\tau}U\left[\mathrm{r}-\mathrm{v}_v(t-\tau)-\mathrm{v}_c\tau\right]dt\}^2.$$

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

$$\exp\left[-n\int d\mathrm{r}(1-e^{i\phi})\right], \tag{24}$$

where

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

Here we introduced the reduced electron-hole mass $m_{eh} = m_e m_h/(m_e + m_h)$.

Let us note that the same result can be derived in a somewhat different way. To begin with, let us calculate the phase acquired by electron-hole state in a field of single ith impurity center. In the field U electron and hole during the time interval τ between pulses acquire phases

$$-\int_0^\tau U(\mathbf{R}_i - \mathbf{v}_e t)dt \quad \text{and} \quad \int_0^\tau U(\mathbf{R}_i - \mathbf{v}_h t)dt \tag{25}$$

respectively. The light creates electron with momentum $\mathbf{p}$ and hole with momentum $-\mathbf{p}$. The corresponding velocities which should be inserted in (25) 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 the band indices. After the second pulse during the time interval between $t = \tau$ and $t = 2\tau$ the electron state acquires the phase

$$\int_\tau^{2\tau} U(\mathbf{R}_i - \mathbf{v}_h\tau - \mathbf{v}_e(t-\tau))dt \tag{26}$$

whereas the hole state gets the phase

$$-\int_0^\tau U(\mathbf{R}_i - \mathbf{v}_e\tau - \mathbf{v}_h(t-\tau))dt \tag{27}$$

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

$$A = \exp\,(i\sum_i \phi_i) \tag{28}$$

where

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

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

$$P(N) = \frac{\overline{N}^N}{N!} e^{-\overline{N}}, \tag{29}$$

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

The exponent in Eq.(28) is then a product of exponents and we have for the configurational average denoted by $< ... >_c$

$$< A >_c = \left\langle \left(\frac{1}{V} \int_V d\mathrm{r} \exp \phi \right)^N \right\rangle_N \tag{30}$$

where $< ... >_N$ means the average over the Poisson distribution. Since

$$< a^N >_N = \sum_{N=0}^{\infty} P(N) a^N = e^{\overline{N}(a-1)} \tag{31}$$

we get for $< A >_c$

$$< A >_c = \exp \left[\frac{\overline{N}}{\mathcal{V}} \int_{\mathcal{V}} d\mathrm{r} \left(e^{i\phi} - 1 \right) \right] \tag{32}$$

Introducing the concentration of impurities $\overline{N}/\mathcal{V} = n$ we see that Eq.(32) coincides with Eq.(24). We wish to emphasize that even under the condition $m_e = m_h$ the total phase of mixed state does not vanish.

For 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.(24) where $\exp(i\phi)$ is replaced by $\cos\phi$. Inserting for the impurity potential $U(\mathrm{r}) = e/\varepsilon r$ (ε being the dielectric susceptibility) we rewrite the decay factor through dimensionless variables

$$\exp \left[-2\pi n (p/m_{eh})^3 \tau^3 \int_0^{\infty} R^2 dR \int_{-1}^{1} dx (1 - \cos\phi) \right], \tag{33}$$

where $\phi = e^2 f(m_e/m_h, R, x)/\varepsilon\hbar(v_e + v_h)$. Here $f(m_e/m_h, R, x)$ is a function of the effective mass ratio, m_e/m_h, dimensionless distance variable R and x (another variable which is equal to the cosine of the angle between p and r).

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

$$- \int_1^2 \frac{dt}{\sqrt{R^2 + ((m_e/M)t - 1)^2 + 2Rx((m_e/M)t - 1)}}$$

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

where $M = m_e + m_h$. Eq.(33) can be presented in the form $\exp(-(\tau/\tau_\varphi)^3)$ where τ_φ is the time of phase breaking.

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

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

we have

$$\tau_\varphi = [15(2\pi)^{1/2}/16\pi^2]^{1/3} \tau_f/\alpha^{1/2}. \tag{34}$$

Then the phase breaking time is much smaller than the time of flight τ_f i.e. the time it takes a particle to traverse mean distance between the Coulomb centers,

$$\tau_f = n^{-1/3}/(v_e + v_h). \tag{35}$$

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

$$\tau_\varphi = \tau_f/\alpha^{2/3}[\pi g(m_e/m_h)]^{1/3}. \tag{36}$$

where

$$g(m_e/m_h) = \int_0^\infty R^2 dR \int_{-1}^{1} dx \left[f\left(m_e/m_h, R, x\right)\right]^2 \qquad (37)$$

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 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 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. Instead of Eq.(24) we have

$$\exp\left[-\sum_{\mathrm{p_1}}(F_{c\mathrm{p_1}} + F_{v\mathrm{p_1}}) \int d\mathrm{r}(1 - \frac{1}{2}(e^{i\phi_1} + e^{-i\phi_2}))\right], \qquad (38)$$

where

$$\begin{aligned}
\phi_1 = &-\int_0^\tau U\left[\mathrm{r} - \left(\frac{\mathrm{p}}{m_e} - \frac{\mathrm{p_1}}{m_e}\right)t\right] dt \\
&+ \int_\tau^{2\tau} U\left[\mathrm{r} - \left(\frac{\mathrm{p}}{m_e} - \frac{\mathrm{p_1}}{m_e}\right)t + \frac{\mathrm{p}}{m_{eh}}\tau\right] dt \\
&+ \int_0^\tau U\left[\mathrm{r} + (\frac{\mathrm{p}}{m_h} - \frac{\mathrm{p_1}}{m_e})t\right] dt \\
&- \int_\tau^{2\tau} U\left[\mathrm{r} + (\frac{\mathrm{p}}{m_h} - \frac{\mathrm{p_1}}{m_e})t - \frac{\mathrm{p}}{m_{eh}}\tau)\right] dt, \qquad (39)
\end{aligned}$$

and one gets ϕ_2 by the replacement $\mathrm{p_1}/m_e \to \mathrm{p_1}/m_h$ in Eq.(39). Instead of the electron velocity we now have the difference of the carrier velocities and instead of the concentration of the carriers $\sum_{\mathrm{p_1}}(F_{c\mathrm{p_1}} + F_{v\mathrm{p_1}})$ enters our formulae where one should sum over the quasimomenta $\mathrm{p_1}$ of the carriers which produce the Coulomb field. Our formulae are applicable also to a two-dimensional situation. In this case we get instead of Eq. (39)

$$\exp\left[-n\left(\frac{p}{m_{eh}}\right)^2 \tau^2 \int_0^\infty RdR \int_0^{2\pi} d\theta(1 - \cos\phi)\right]. \qquad (40)$$

with

$$\phi = \frac{e^2}{\varepsilon\hbar(v_e + v_h)} f(m_e/m_h, R, \cos\theta) \tag{41}$$

We come to the general conclusion that in this case the time of phase breaking is proportional to $n^{-1/2}$.

4 Qualitative consideration.

Let us begin with analysis of the case

$$\alpha \gg 1 \tag{42}$$

Consider the electrostatic potential $U(t)$ in the reference frame moving with the electron under consideration (see Fig.6). The characteristic scale of time variation is $\tau_f = n^{-1/d}/v$, while the characteristic amplitude is $n^{1/d}e^2/\varepsilon$. Therefore for $t \ll \tau_f$ one can expand $U(t)$ retaining the linear term

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

The corresponding phase variation is

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

Hence,

$$\tau_\varphi = (\hbar\tau_f\varepsilon/e^2 n^{1/d})^{1/2}$$

which coincides with Eq.(34). We should, however, make here the following important point. The theory developed in the present paper is based on the assumption that the spatial distribution of the scatterers is random. Such an assumption is usually valid for charged impurities. However, one can expect that the carriers excited by light in the course of band-to-band transitions

are strongly correlated immediately after their excitation and it takes roughly time τ_f for the correlation to die off. This means that for the case of Eq.(42) our theory is applicable only for τ bigger than τ_f. For shorter times further development of the theory is needed.

The opposite case $\alpha \ll 1$ can be understood in the following way. One can assume that, together with short-scale fluctuations, the 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

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

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

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

It brings about the phase variation

$$e^2 n^{1/2}(v)^{d/2-1}t^{d/2}/\hbar\varepsilon$$

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 3D case. It depends on the average carrier energy. For $\mathcal{E} \simeq 0.2 \cdot 10^{-12}$ erg. and $n = 7 \cdot 10^{18}$ cm^{-3}, we get $\tau_\varphi = 15$ fs.

5 Conclusion

One can ask as to what is the physical nature of the contribution to the echo decay due to the interaction between the electrons and impurities. Indeed, when calculating the echo amplitude we could have used the exact electron

wave function in the impurity field. One could come to the conclusion that in such a case there would be no decay. The point, however, is that the plane waves of light interact with a pair of electron states having *definite values of quasimomenta* (because of the quasimomentum conservation). The exact states are superposition of the states with the definite values of quasimomenta. Since our treatment of the echo phenomenon rests on the concept of independent states, the coupling among them brings about the decay (cf. with Ref.[14] where irreversibility of *energy conserving* dipole dephasing for a simple atomic system was found). The impurity concentration being 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.

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 random unscreened Coulomb field originated either in the static impurities or photocarriers (provided that they are randomly distributed in space) generated by the laser pulse. We do not take into account the effects caused by the finite duration of the laser pulses. We believe, however, that these effects do not change qualitatively our results. 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 for a 3D case are up to about a dozen of femtoseconds. The concentration dependence of such a time and its order of magnitude are in agreement with 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, $T_{\rm echo}$, 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.

One of the principal results of our paper is that we have established a *new time characteristic of an electron system*, τ_φ. This time 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.

We wish to emphasize once again that the physical considerations put forward in this paper are based on some generic concepts. As for the calculations, they are presented, with the help of a 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 opinion that such a disagreement is of fundamental nature and that manifests basic need for further experimental and theoretical work.

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References

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

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

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

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

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

[6] O. V. Konstantinov, V. I. Perel', Sov. Phys. — JETP. 12, 142 (1961).

[7] S. V. Gantsevich, V. D. Kagan, R. Katilius, E. Rzaev, Sov.Phys. — JETP, 60, 997 (1984)

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

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

[10] L. Root and J. L.Skinner, J. Chem. Phys. 81, *12*, 5310 (1984).

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

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

[13] S. L.Ginzburg. Sov. Phys. — JETP, 36, 1198 (1973).

[14] J. Yeh, J. H. Eberly, Phys. Rev.A. 22, 1124 (1980)

Figure captions

Fig. 1. General scheme for echo phenomena. Interaction constants are omitted. The "tail" before $t = 0$ represents the particle distribution function.

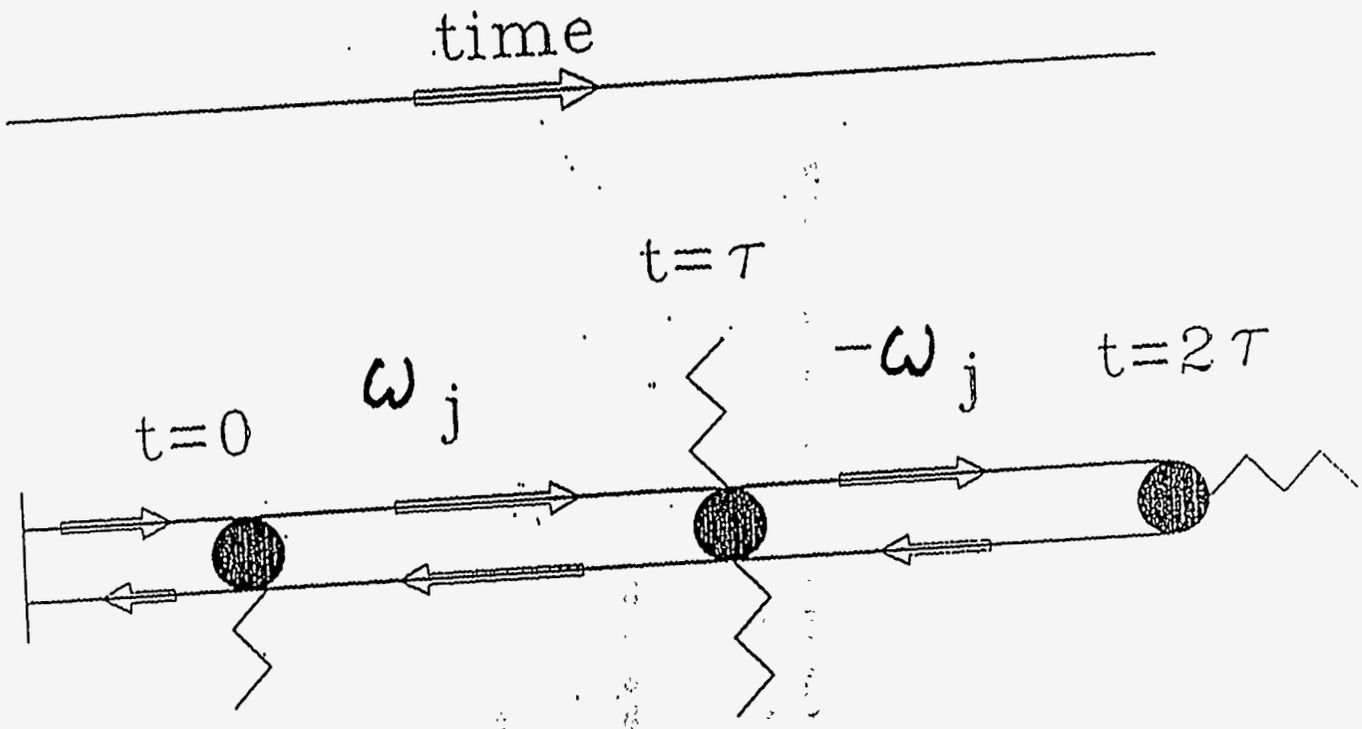
Fig. 2. Action of random forces on an oscillator.

Fig. 3. Echo in semiconductors.

Fig. 4. Damping due to "collisions".

Fig. 5. Diagrams for nonlinear polarization in the eikonal approximation.

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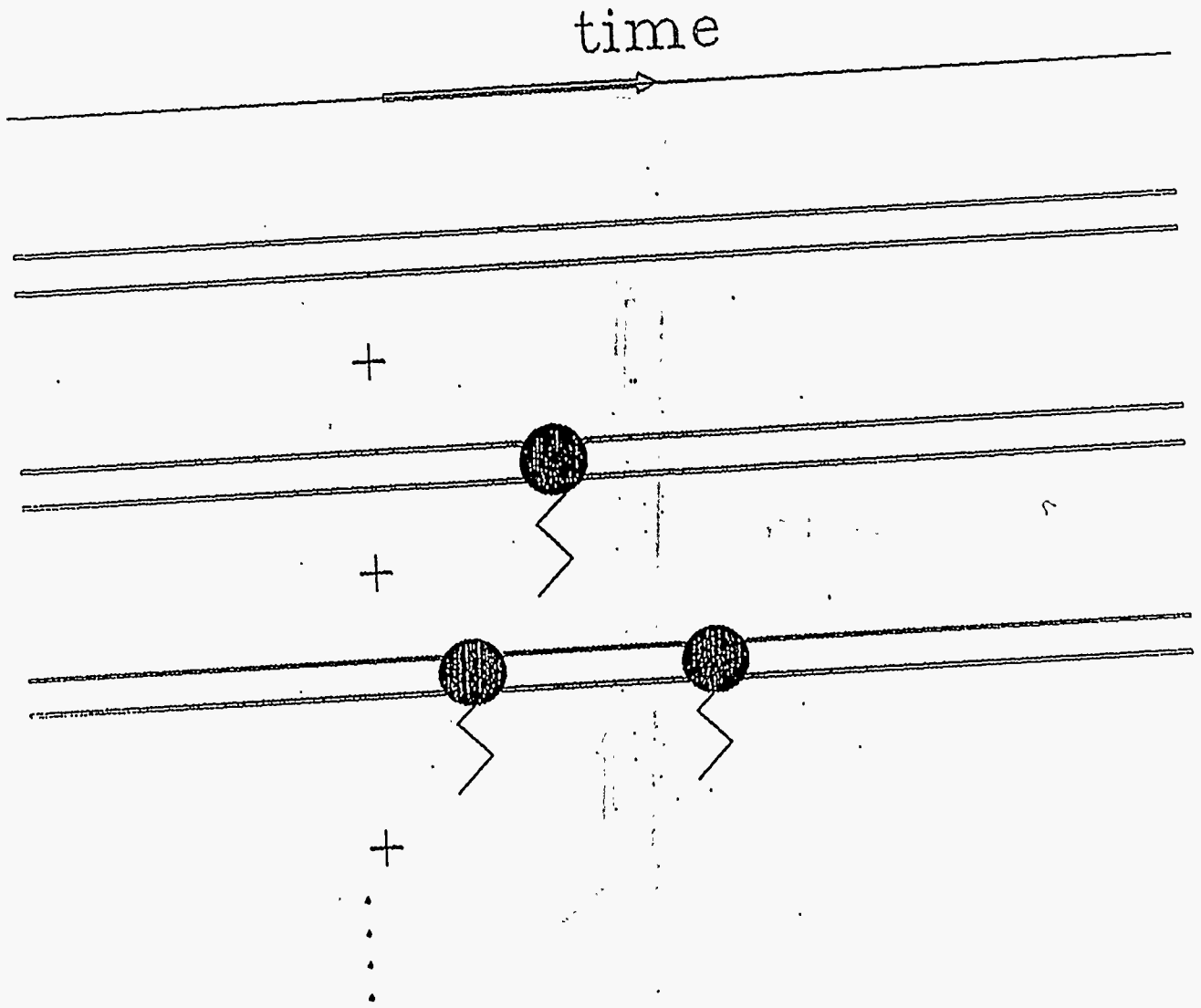
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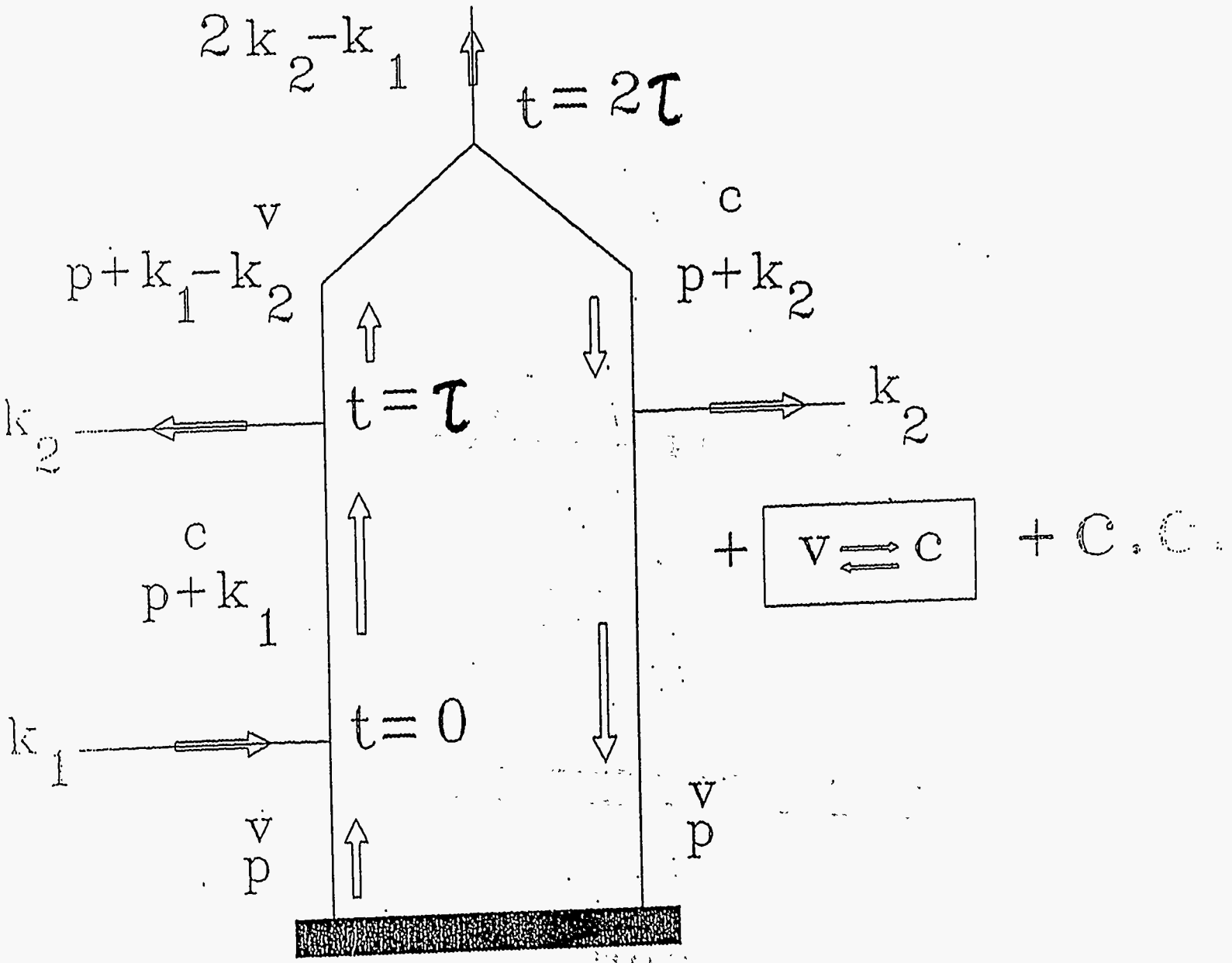


Fig. 3

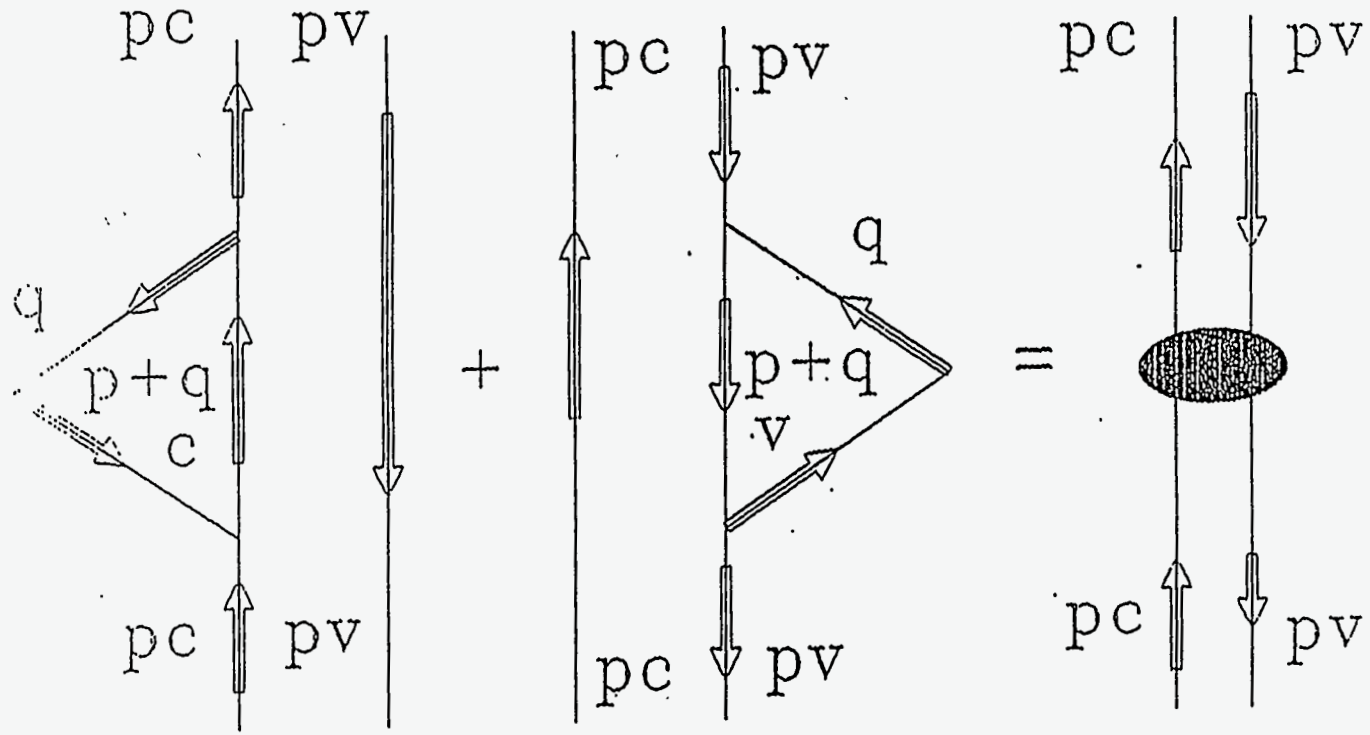
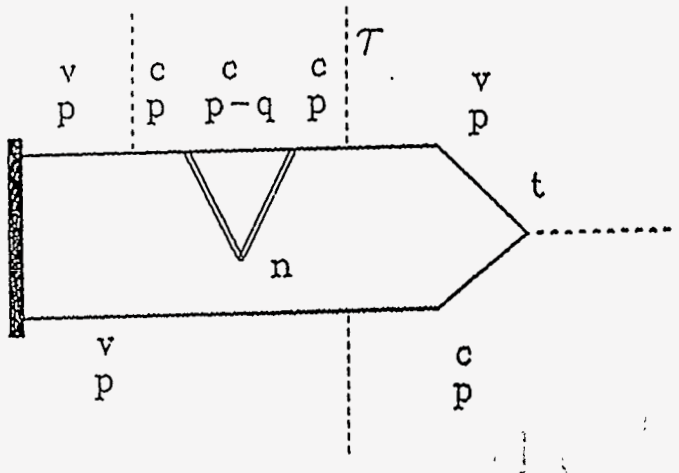


Fig. 4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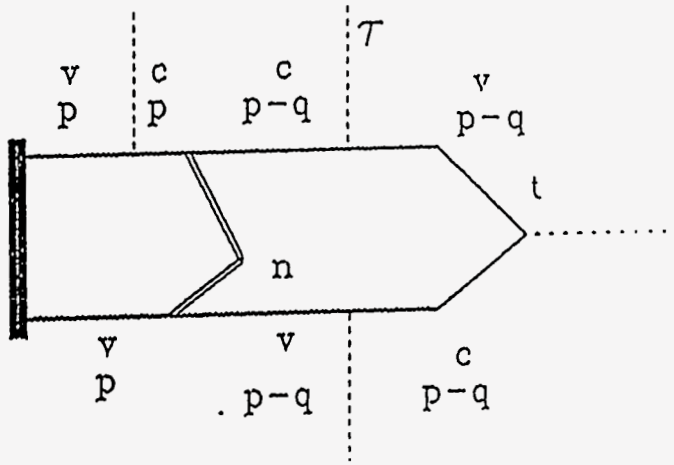
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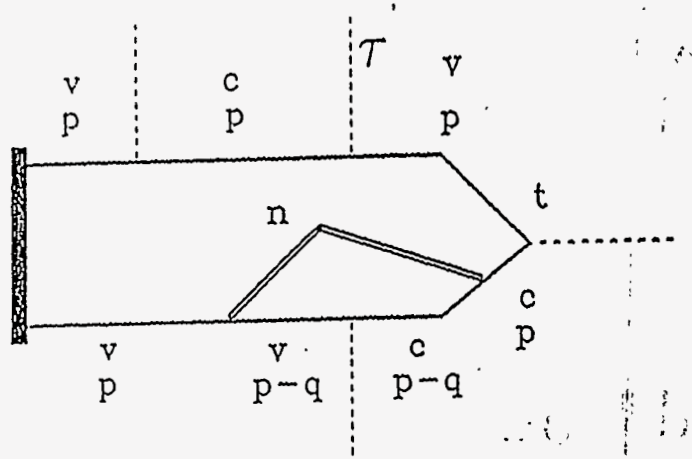
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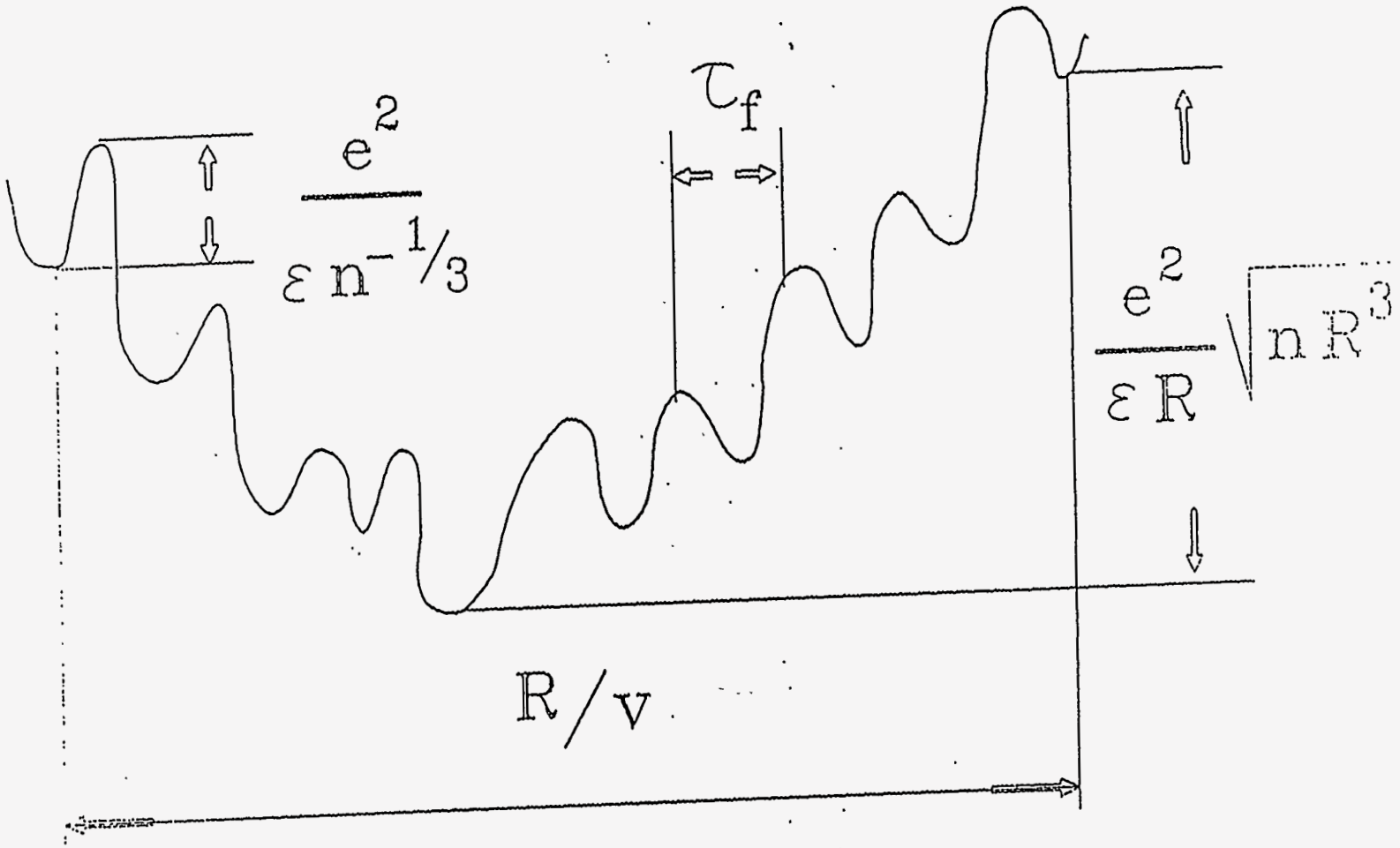
Fig. 5


$\frac{e^2}{\varepsilon n^{-1/3}}$
τ_f
$\frac{e^2}{\varepsilon R}\sqrt{n R^3}$
R/v

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