Collisionless dynamics of a nonequilibrium semiconductor-metal phase transition

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The dynamics of a nonequilibrium semiconductor-metal (SM) phase transition is considered over time intervals shorter than the inelastic collision time. Two cases are discussed separately: 1) when the SM transition is produced under equilibrium conditions by interband Coulomb interaction; 2) when the SM transition and the associated structural transition are due to the electron-phonon interaction. In the case of small perturbations of the semiconductor phase the deviation of the order parameter Σ (Σ represents the gap in the spectrum of elementary excitations) oscillates at the frequency 2Σ and its amplitude relaxes in time in accordance with the law $t^{-1/2}$ tending to a state which depends on the initial conditions. In the second case the amplitude of the shift of the sublattices includes a term which oscillates at the frequency 2Σ and decreases in amplitude like $t^{-3/2}$, as well as a term which is oscillatory but undamped and has a frequency of the order of the phonon frequency. In the case of finite perturbations above the critical value the system exhibits coherent quantum oscillations between the semiconductor and metal phases.

INTRODUCTION

Reports of experiments on the action of powerful femtosecond laser pulses on covalent semiconductors have appeared recently.[1–3] The available experimental technique make it possible to time-resolve the processes of duration amounting to several picoseconds.[1–3] The power of such pulses is sufficient to cause a nonequilibrium semiconductor-metal (SM) transition as a result of excitation of the electron subsystem.[4] This raises the question of the dynamics and characteristic times of a nonequilibrium SM phase transition.

If the duration of a laser pulse is less than the time required for modification of the system, the action of a laser source is limited to the establishment of a nonequilibrium state of the system at the initial moments. However, if the pulse duration is considerably longer than the time constants of the transient processes but the trailing edge of the pulse is much shorter than the time needed for the modification of the system, the process of switching off of the source can be regarded as instantaneous. After the end of the pulse the system is again in a nonequilibrium state. From the theoretical point of view both situations reduce to a study of the evolution of the system (in the absence of a source) from a strongly nonequilibrium state set up by the preceding pulse.

This evolution occurs in several stages. During the first collisionless stage when the deviation from equilibrium is greater than critical we can expect coherent macroscopic transitions of a crystal between the semiconductor and metal phases. During the next stage the inelastic relaxation processes reduce the system to a state of local equilibrium. This very state depends on the concentration of nonequilibrium excitations and changes during the recombination time in this state. If the system is in a local minimum corresponding to the metal phase, then this minimum disappears in the recombination time of carriers in the metal and the transition to the semiconductor phase occurs without an activation energy. The disappearance of a barrier has been observed in a system of nonequilibrium carriers on transition to an electron-hole liquid.[5] This effect is known to be a common feature of first-order transitions.

A detailed description of the evolution of covalent semiconductors, such as Si and Ge, is quite difficult and this is why we shall consider some models. There are examples of systems which undergo a structural transition as well as an SM transition and for which there are well-developed theoretical models capable of revealing the qualitative features of the dynamics of a nonequilibrium transition. Such substances are, for example, IV-VI semiconductors[6] and vanadium oxides.[7] In the case of vanadium oxides, there are also experimental data for picosecond pulses indicating that initiation of the SM transition is of nonthermal nature and occurs because of the excitation of the electron subsystem.[8]

A common feature of these substances is a special form of a one-particle electron spectrum in the symmetric phase or in the precursor phase (in the case of IV-VI compounds). These substances have parts of the Fermi surface which nest in one another when translated by a certain vector. A special form of the spectrum obtained on allowance for the electron-electron and electron-phonon interactions may result in Bose condensation of electron-hole pairs and of phonons, i.e., it may produce a structural transition.[9,10]

We shall consider the collisionless stage of the evolution process (for time intervals shorter than the time constants of inelastic processes) of the initial nonequilibrium state in systems exhibiting the SM transition. We shall consider separately two cases: 1) when the SM transition is due to the electron-electron interaction; 2) when it is due to the electron-phonon interaction. In the former case the order parameter relaxes to a certain state which depends on the initial conditions. The gap oscillates at a frequency 2Σ (where Σ is the gap in the spectrum of one-particle excitations) and its amplitude decreases in time in accordance with the $t^{-1/2}$ law. In the second case the amplitude of the shift of the sublattices has terms which relax in accordance with the power

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law $t^{-3/2}$ and oscillate at the frequency 2Σ. Moreover, there are also undamped terms oscillating at a frequency ω_D (ω_D is the characteristic phonon frequency). The electron distribution functions have undamped oscillatory terms associated with the thermodynamically reversible process of collisionless evolution.

Suslov[11] studied qualitatively the dynamics of changes in a Peierls insulator subjected abruptly to an external perturbation.

COLLISIONLESS EVOLUTION OF THE DENSITY MATRIX OF AN EXCITON INSULATOR

It is convenient to use the method of Kukharenko and Tikhodeev[12] in describing the evolution of the density matrix from a given initial state. This method is a generalization of the Keldysh technique[13] developed for the problems of relaxation allowing for the initial correlations. Collisionless evolution in a superconductor was studied by Volkov and Kogan[14] using the Keldysh technique. In this section we shall consider the case when the SM transition is due to the electron-electron interaction. The Hamiltonian of the system is

$$H=\sum_{i,\mathbf{p},\sigma}\varepsilon_i(\mathbf{p})\,a^{\pm}_{i\mathbf{p}\sigma}a_{i\mathbf{p}\sigma}+\frac{1}{2}\sum_{\substack{\mathbf{p}_1,\mathbf{p}_2,\mathbf{k}\\ \sigma_1,\sigma_2}}V(\mathbf{k})\,a^{+}_{1\mathbf{p}_1\sigma_1}a^{+}_{2\mathbf{p}_2\sigma_2}a_{2\mathbf{p}_2-\mathbf{k}\sigma_2}a_{1\mathbf{p}_1+\mathbf{k}\sigma_1}$$
$$+\frac{1}{2}\sum_{\substack{\mathbf{p}_1,\mathbf{p}_2,\mathbf{k}\\ \sigma_1,\sigma_2}}V_1(\mathbf{k})\,[a^{+}_{1\mathbf{p}_1\sigma_1}a^{+}_{1\mathbf{p}_2\sigma_2}a_{2\mathbf{p}_2-\mathbf{k}\sigma_2}a_{2\mathbf{p}_1+\mathbf{k}\sigma_1}+a^{+}_{1\mathbf{p}_1\sigma_1}a^{+}_{2\mathbf{p}_2\sigma_2}$$
$$\times a_{1\mathbf{p}_2-\mathbf{k}\sigma_2}a_{2\mathbf{p}_1+\mathbf{k}\sigma_2}+\text{H.a.}]; \qquad (1)$$

here, $i=1$ and 2 are the band indices; $a^{+}_{1\mathbf{p}\sigma}$ and $a^{+}_{2\mathbf{p}\sigma}$ are the operators describing creation of electrons with momenta $\mathbf{p}$ and spins σ in the bands 1 and 2. The unrenormalized (bare) spectrum of one-particle excitations satisfies the congruence conditions

$$\varepsilon_1(\mathbf{p})=-\varepsilon_2(\mathbf{p}+\mathbf{Q}),$$

where $\mathbf{Q}$ is a reciprocal lattice vector. The second term in the Hamiltonian describes the Coulomb density-density interaction between different bands and the third term corresponds to the interaction associated with a transition involving the scattering of electrons from one band to another. In the high density limit $na_B^3\gg 1$ (n is the particle density and a_B is the Bohr radius of an exciton) the potentials $V(\mathbf{k})$ and $V_1(\mathbf{k})$ can be regarded as constants and are denoted by g and g_1, respectively.

Information on the system at the initial moment is given by the density matrix $\hat{\rho}(t_0)$. The evolution is described by the behavior of one-particle and many-particle Green functions. We shall confine ourselves to one-particle Green functions, because in the self-consistent Hartree-Fock approximation, when the interaction is allowed for, the higher correlation functions are unimportant in the description of the evolution with the exception of some special cases. A generalization of the method of Ref. 12 to the case of two electron bands does not present any difficulty and reduces essentially to introduction of additional band indices of the Green function. The zeroth Green function is a matrix

$$\hat{G}_{ij}(\mathbf{1},\mathbf{1}')=\begin{vmatrix}0 & G_{ij}^{a}\\ G_{ij}^{r} & \mathcal{F}_{ij}\end{vmatrix}=-i\exp[i\varepsilon_j(\mathbf{p}_1)(t_1'-t_0)$$
$$-i\varepsilon_i(\mathbf{p}_1)(t_1-t_0)]\begin{vmatrix}0 & -\delta_{ij}\theta(t_1'-t_1)\\ \delta_{ij}\theta(t_1-t_1') & \delta_{ij}-2f_{ij}(p_1t_0)\end{vmatrix}, \qquad (2)$$

where $\mathbf{1}=(p_1,t,\alpha)$ and $p_1=(\mathbf{p}_1,\sigma)$; here, α is an index describing the position on the time profile; $f_{ij}(\mathbf{p}_1t_0)$ is a one-particle distribution function at the initial moment in time. The complete Green function considered in the self-consistent Hartree-Fock approximation satisfies the equation

$$\check{G}(t,t')=\check{G}_0(t,t')+\int_{t_0}^{t_{max}}\int dt_1\,dt_1'\check{G}_0(t,t_1)\check{\Sigma}(t_1,t_1')\check{G}(t_1',t'). \qquad (3)$$

The self-energy part is of the form

$$\hat{\Sigma}_{12}(t_1,t_1')=\hat{\nu}\Big\{\int\frac{d\mathbf{p}}{(2\pi)^3}\Big[g\hat{G}_{12}(t_1,t_1')+g_1\hat{G}_{21}(t_1,t_1')$$
$$-g_1\sum_{\sigma}(\hat{G}_{12}(t_1,t_1')+\hat{G}_{21}(t_1,t_1'))\Big]\Big\}, \qquad (4)$$

$$\check{G}(\mathbf{1},\mathbf{1}')=\begin{vmatrix}\hat{G}_{11} & \hat{G}_{12}\\ \hat{G}_{21} & \hat{G}_{22}\end{vmatrix},\quad \check{\Sigma}(\mathbf{1},\mathbf{1}')=\begin{vmatrix}0 & \hat{\Sigma}_{12}\\ \hat{\Sigma}_{21} & 0\end{vmatrix}. \qquad (5)$$

The matrix $\hat{\nu}$ has nonzero components $\nu_{1112}=\nu_{2221}$ with all possible transpositions of the indices 1 and 2 on the time profile; $t_{\max}=\max(t,t')$.

We shall assume that $g>3g_1$, i.e., that the instability in the exciton channel[15] and not in the interband plasmon channel predominates.

Equations (3) and (4) contain the distribution functions $f_{ij}(\mathbf{p}t_0)$ describing the situation at the initial moment in time. The regularization technique from Ref. 12 can be used to provide a closed description of the evolution of the system by a slowly varying distribution function $f_{ij}(\mathbf{p}t)$ at a given moment. The procedure of elimination of $f(\mathbf{p}t_0)$ and adoption of the slowly varying distribution function was first used by Bogolyubov.[16] The equations for $f_{ij}(\mathbf{p}t)$ are obtained by separation of those components from the matrices $\hat{G}_{11}$ and $\hat{G}_{12}$ which have the indices 22; i.e., $\mathcal{F}_{11}$ and $\mathcal{F}_{12}$.

The consistency conditions for the renormalized system of integral equations (3) and (4) are

$$\Big[i\frac{\partial}{\partial t}-(\varepsilon_i(\mathbf{p})-\varepsilon_j(\mathbf{p}))\Big]f_{ij}(\mathbf{p}t)$$
$$=-\frac{i}{2}\{[G_{0i}^{-1}(1)-G_{0j}^{-1*}(1')]\mathcal{F}_{ij}(t_1,t_1')\}_{t_1=t_1'=t}, \qquad (6)$$
$$G_{0i}^{-1}(1)=\Big(i\frac{\partial}{\partial t}-\varepsilon_i(\mathbf{p})\Big).$$

This can be represented in the form of the Liouville equation for the density matrix:

$$i\partial\hat{\rho}/\partial t=[\hat{H},\hat{\rho}],$$
$$\hat{\rho}(\mathbf{p}t)=\begin{vmatrix}f_{11}(\mathbf{p}t) & f_{12}(\mathbf{p}t)\\ f_{21}(\mathbf{p}t) & f_{22}(\mathbf{p}t)\end{vmatrix},\quad \hat{H}(\mathbf{p}t)=\begin{vmatrix}\varepsilon_1(\mathbf{p}) & \Sigma_{12}(t)\\ \Sigma_{21}(t) & \varepsilon_2(\mathbf{p})\end{vmatrix}. \qquad (7)$$

It will be now convenient to adopt the distribution functions averaged over the angular variables:

$$f_{ij}(\xi t)=\frac{1}{4\pi}\int d\Omega_{\mathbf{p}}f_{ij}(\mathbf{p}t),\quad \xi=v_F(|\mathbf{p}|-p_F).$$

In the stationary case the self-energy function $\Sigma_{12}(t)$ plays the role of a gap in the spectrum of elementary excitations: by definition, we have

$$\Sigma_{12}(t)=N(0)\int_0^{\varepsilon_F} d\xi\,[gf_{12}(\xi t)+g_1 f_{21}(\xi t)-2g_1(f_{12}(\xi t)+f_{21}(\xi t))], \tag{8}$$

where $N(0)$ is the density of states at the Fermi level. We can readily show that the energy integral of the system can be represented in the form

$$E=2N(0)\left\{-\mathrm{Sp}\int d\xi(\hat{\rho}(\xi t)\hat{H}(\xi t))+\Sigma_{\mathrm{Re}}^2/\lambda_{\mathrm{Re}}+\Sigma_{\mathrm{Im}}^2/\lambda_{\mathrm{Im}}\right\}; \tag{9}$$

here, Σ_{Re} and Σ_{Im} are the real and imaginary parts of the order parameter, and

$$\lambda_{\mathrm{Re}}=N(0)(g-3g_1),\quad \lambda_{\mathrm{Im}}=N(0)(g-g_1)$$

are the corresponding coupling constants. The first term represents the energy of quasiparticles and the second the energy of a condensate of electron-hole pairs. The stationary values of the phase of the gap are: $\varphi_{\mathrm{Re}}=0,\pi;\varphi_{\mathrm{Im}}=\pm\pi/2$. The phase φ_{Re} corresponds to the real order parameter (charge density wave) and φ_{Im} to the imaginary parameters (current density wave).[17] An energy minimum under thermodynamic equilibrium conditions corresponds to a real value of Σ if $g_1<0$ and to an imaginary Σ if $g_1>0$. We shall consider the specific case when the relationship between the coupling constants is such that a state with a charge density wave is realized under equilibrium conditions.

Arbitrary initial conditions can be applied to a system of equations (7)–(8). We shall consider initial perturbations which are symmetric in respect of electrons and holes. It then follows from the equations of motion that the phase of the gap is constant in time. If there is an initial unbalance between the electron and hole branches, then (in addition to the time dependence of the gap width) the phase of the gap is "rotated." In contrast to a superconductor, when spatially homogeneous phase rotation does not result in the observed physical consequences, in the present case the different values of the phase correspond to fundamentally different physical states of the system. In particular, the establishment of an unbalance between the electron and hole branches (for example, by tunnel injection) may facilitate realization of a nonequilibrium state with a current density wave.

Let us assume that a perturbation symmetric in respect of electrons and holes

$$\delta f_{11}(\xi t=0)=-\delta f_{22}(\xi t=0)=f^{(0)}(\xi)$$

is applied at the initial moment in time and let us assume that the perturbations of the gap and the correlation function $f_{12}(\xi)$ are absent at $t=0$. Linearization of the system (7)–(8) and the Laplace transformation gives the following expression at $T=0$:

$$\delta\Sigma(p)=-\frac{4\Sigma}{p(p^2+4\Sigma^2)}\left[\int_0^{\varepsilon_F}\frac{d\xi\,\xi f^{(0)}}{p^2+4\varepsilon^2}\right]\left[\int_0^{\varepsilon_F}\frac{d\xi}{\varepsilon(p^2+4\varepsilon^2)}\right]^{-1}, \tag{10}$$

where p is the Laplace variable and Σ is the equilibrium value of the gap. The function $\delta\Sigma(p)$ has a hole at $p=0$ in the plane of the complex variable p and exhibits cuts from $\pm 2i\Sigma$ to $\pm i\infty$. Bearing in mind that the imaginary part of the denominator in Eq. (10) becomes infinite in the limit $p\to\pm 2i\Sigma$, we find that the main contribution in inversion of the Laplace transform $\delta\Sigma(p)$ is made in the region of $p=\pm 2i\Sigma$.

The final answer corresponding to $t\to\infty$ can be expressed in terms of a zeroth-order Neumann function and it is of the form

$$\delta\Sigma(t)\approx\frac{\varphi_1(0)}{2\pi\Sigma\varphi_2(0)}+2(2\pi)^{1/2}\Sigma\varphi_1(2\Sigma)\frac{\sin(2\Sigma t-\pi/4)}{(2\Sigma t)^{1/2}}, \tag{11}$$

where

$$\varphi_1(x)=\int_0^{\varepsilon_F}\frac{d\xi\,\xi f^{(0)}(\xi)}{4\varepsilon^2-x^2},\quad \varphi_2(x)=\int_0^{\varepsilon_F}\frac{d\xi}{\varepsilon(4\varepsilon^2-x^2)}.$$

Therefore, the amplitude of the gap relaxes in accordance with the power law and it exhibits oscillations of frequency 2Σ to a certain state which depends on the initial conditions.

The distribution functions corresponding to a given quasiparticle energy have a damped part associated with $\delta\Sigma(t)$ as well as undamped, in the limit $t\to\infty$, oscillatory terms which are associated with the thermodynamically reversible relaxation process. We then have

$$\delta f_{11}(\xi t)=-\frac{f^{(0)}(\xi)\Sigma\xi}{\varepsilon^2}(1-\cos 2\varepsilon t),$$

$$\mathrm{Re}\,\delta f_{12}(\xi t)=f^{(0)}(\xi)\left[1-\frac{\Sigma^2}{\varepsilon^2}(1-\cos 2\varepsilon t)\right],$$

$$\mathrm{Im}\,\delta f_{12}(\xi t)=f^{(0)}(\xi)\sin 2\varepsilon t,\quad \varepsilon=(\xi^2+|\Sigma|^2)^{1/2}.$$

Collective oscillations of the order parameter are associated with the following circumstance. When we specify the density matrix at the initial moment of time, this fixes uniquely the energy of the system which is conserved in the electron subsystem at times shorter then the time for inelastic collisions with phonons. Since the initial state is arbitrary, it follows that in general it is not stationary. Energy is transferred between the Bose condensate of electron-hole pairs and quasiparticles. The binding energy of excitons is $\sim 2\Sigma$ and the pairing time is $\sim 1/2\Sigma$, which governs the oscillation period. When the exciton condensate dissociates, it breaks up into quasiparticles with momenta in the interval $\sim\Sigma/v_F$, resulting in the loss of coherence and collisionless damping. Clearly, the pairing and dissociation processes do not disturb the electrical neutrality of the system.

In constrast to the exponential Landau damping,[18] the damping is now of the power-law and this is due to the following physical factors. The exponential law of the Landau damping is due to the fact that in the range of small wave numbers k the phase volume of the state which can facilitate exchange of energy between electrons and the plasma mode is exponentially small in respect of the parameter ka_D, where a_D is the Debye-Hückel screening radius. There is no damping for $k=0$. In our case, even when oscillations are homogeneous, there is always a possibility of energy transfer from the condensate to quasiparticles in an energy interval

$\sim\Sigma$ and vice versa. The phase volume is governed by the gap itself and this gives rise to a power-law relaxation process.

If a perturbation in the distribution of the particles is initially strongly localized in energy at the edge of the gap (for example, if it is created by illumination with a source near the interband absorption edge), in an interval $\gamma \ll \Sigma$ [$f^{(0)}(\xi) \propto \Sigma^2/(\xi^2+\gamma^2)$], the relaxation time of the gap increases. It follows from Eq. (10) that

$$\delta\Sigma(t)\sim\Sigma\cos 2\Sigma t, \quad t\ll\tau_\gamma,$$
$$\delta\Sigma(t)\sim\Sigma\sin(2\Sigma t-\pi/4)(\pi t/\tau_\gamma)^{-1/2}, \quad t\gg\tau_\gamma,$$
$$\tau_\gamma=\Sigma/\gamma^2;$$

the time τ_γ may become considerably greater than $1/\Sigma$. This property of localized perturbations are pointed out by Kulik[19] in the case of superconductors. Slowing down of the relaxation may occur also when inelastic processes are activated (if the intraband energy relaxation time is shorter than the interband time, which is possible if $\omega_D \ll \Sigma$). In the case of a smooth initial distribution the relaxation of excitations to the gap edge localizes the distribution. A change in γ and in the distribution function due to inelastic processes occur during the "slow" intraband relaxation time. The gap is modified in accordance with the distribution function because of collisionless relaxation in a time τ_γ, where γ is the width of the energy distribution at a given moment in time.

These solutions are valid at times shorter than the time constants of inelastic processes. Oscillations should occur also when inelastic collisions become important, but their damping is exponential until a local equilibrium state is attained. This state itself changes during the inelastic interband relaxation (recombination) time and the gap follows these changes in a quasistatic manner. If during the process of evolution the system reaches a minimum corresponding to the metal phase ($\Sigma = 0$), then this minimum disappears during the recombination time of the metal phase (since this minimum originates from excess quasiparticles). The system returns in an activation-free manner to the semiconductor phase. The process of evolution is illustrated n Fig. 1 as a function of the concentration of excitations n_e and curves 1–3 show how a local equilibrium changes in time ($n_{e1} > n_{e2} > n_{e3}$). The time for the recovery back to the original semiconductor phase with the energy Σ from the intermediate state is equal to the gap relaxation time during the kinetic stage and it can be estimated from $\tau_\Sigma \sim (\Sigma_0/\Sigma)\tau_{\text{inel}}$ (Ref. 20), where τ_{inel} is the characteristic inelastic collision time. The disappearance of a barrier in the case of a finite carrier lifetime is clearly a common property of first-order transitions. A similar disappearance of a barrier in the case of a transition from an exciton gas to an electron-hole liquid has been confirmed experimentally.[5]

We shall now consider the nature of relaxation of the initial perturbation in the semiconductor phase under strongly nonequilibrium conditions. If the interband relaxation time is long compared with the intraband inelastic processes (which is possible if $\omega_D \ll \Sigma$), then a state of local equilibrium is established. The order parameter "fits" the distribution function $\eta_0(\varepsilon)$ in this state which is stationary for $t \ll \tau_R$ (τ_R is the recombination time in this phase). The

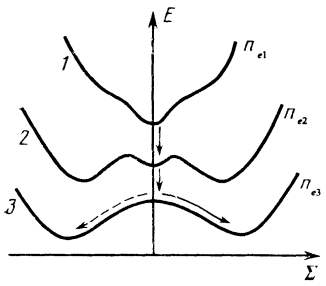

FIG. 1.

gap is found from the self-consistency equation

$$\frac{1}{\lambda_{Re}}=\int_0^{\varepsilon_F}\frac{1-2n_0(\varepsilon)}{\varepsilon}d\xi. \tag{12}$$

The distribution function n_0 causes vanishing of the intraband collision integrals. In this case there may be undamped (again at times shorter than those of inelastic intraband processes) collective modes absent under equilibrium conditions. The condition for the appearance of undamped oscillations Σ is the vanishing of the imaginary and real parts of the function $\varphi_2(n_0,\omega)$, which is analogous to Eq. (11) for imaginary $p = i\omega + 0$:

$$\varphi_2(n_0,\omega)=\int_0^{\varepsilon_F}\frac{1-2n_0(\varepsilon)}{\varepsilon[4\varepsilon^2-(i\omega+0)^2]}=0, \quad \omega^2<4\Sigma^2. \tag{13}$$

A solution of Eq. (13) exists if $1-2n_0(\varepsilon)$ exhibits an alternating sign, i.e., if an inverted distribution is realized in a certain range of energies. All those requirements are satisfied by a quasi-Fermi distribution with zero temperature and we shall consider this distribution by way of example.

The solutions of Eqs. (12) and (13) are

$$\omega_1^2=4\Sigma^2(1-n_e^2\Sigma_0/\Sigma^3),$$
$$\omega_2^2=4\Sigma^2\left[1-n_e^2\frac{\Sigma_0}{\Sigma^3}\left(1-2n_e\left(\frac{\Sigma_0}{\Sigma}\right)^{1/2}\frac{1}{\Sigma_0}\right)^2\right], \tag{14}$$
$$\Sigma(\Sigma-\Sigma_0)^2=4n_e^2\Sigma_0.$$

The total number of excitation is

$$n_e=\frac{1}{2N(0)}\int d\xi\, n_0(\varepsilon), \quad n_0(\varepsilon)=\theta(\mu-\varepsilon), \quad n_e=(\mu^2-\Sigma^2)^{1/2}, \tag{15}$$

where μ is the quasi-Fermi chemical potential. The dependence $\Sigma(n_e)$ of Eq. (15) has two branches which merge at $n_{es} = \Sigma_0/3\sqrt{3}$, and we find that $\Sigma(n_{es}) = \Sigma_0/3$. In the case of the upper branch ($\Sigma > \Sigma_0/3$) both modes are stable: $\omega_{1,2}^2 > 0$, whereas for the lower branch [$\Sigma(n_e) < \Sigma_0/3$] the first mode is unstable: $\omega_1^2 < 0$. For a critical value of n_{es} the frequency ω_1^2 is a linear function of $n_e - n_{es}$ and passes through zero, and $\omega_2^2(n_{es}) > 0$. These modes have been found by Aronov and Gurevich for a superconductor by a different and in our opinion more complicated method.[21] However, an inverted distribution of quasiparticles does not usually appear in superconductors.

These collective modes can appear at times shorter than

the recombination time τ_R if a quasi-Fermi distribution is established during the evolution. Since the formation of this distribution is possible only if an allowance is made for inelastic intraband collisions, in reality these modes may be damped.

We shall consider the case of amplitude oscillations of a charge density wave. We can readily find the frequency of phase oscillations in a charge density wave from Eqs. (7) and (8). We shall give the answer directly:

$$\omega^2 \approx 4\Sigma^2 (1/\lambda_{\mathrm{Im}} - 1/\lambda_{\mathrm{Re}}), \quad 1/\lambda_{\mathrm{Im}} - 1/\lambda_{\mathrm{Re}} \ll 1. \tag{16}$$

The presence of phase-locking terms in the Hamiltonian (1) leads to gaps in Goldstone modes. In particular, it follows from Eq. (16) that if $\lambda_{\mathrm{Im}} > \lambda_{\mathrm{Re}}$, then the state with a charge density wave is unstable against fluctuations of the gap phase which transfer the system to a state with a current density wave.

In the case of triplet pairing a spin density wave appears, i.e., the band antiferromagnetism is observed.[15] A deviation from equilibrium gives rise to a similar collisionless relaxation law of the sublattice magnetizations.

The thermodynamically reversible nature of the evolution in such systems should give rise to collective effects of the echo type, known from collisionless plasma studies.[22]

COLLISIONLESS EVOLUTION OF PERTURBATIONS IN A SYSTEM WITH A STRUCTURAL TRANSITION

The characteristic features of the spectrum of a semimetal $\varepsilon_1(\mathbf{p}) = -\varepsilon_2(\mathbf{p}+\mathbf{Q})$ [and of a one-band metal with $\varepsilon(\mathbf{p}) = -\varepsilon(\mathbf{p}+\mathbf{Q})$] not only give rise to an instability of the electron subsystem against Bose condensation of electron-hole pairs considered in the preceding section, but are also responsible for an instability of the phonon system due to the interband electron-phonon interaction.[15] One of the examples of such systems are II-VI semiconductor compounds.[6]

In this section we shall consider the dynamics of a transition in a system in which the SM transition is due to the electron-phonon interaction. For simplicity, we shall consider the case when the polarization vector of unstable phonons has one component. For example, in the case of IV-VI semiconductors such an instability corresponds to the tetragonal deformation of the cubic lattice.

The Hamiltonian of the system is of the form

$$H = \sum_{i,\mathbf{p},\sigma} \varepsilon_i(\mathbf{p}) a^+_{i\mathbf{p}\sigma} a_{i\mathbf{p}\sigma} + g_{ph} \sum_{\mathbf{p},\sigma} [(b_{\mathbf{Q}} + b_{-\mathbf{Q}}^+) \times (a^+_{1\mathbf{p}\sigma} a_{2\mathbf{p}+\mathbf{Q}\sigma} + a^+_{2\mathbf{p}\sigma} a_{1\mathbf{p}+\mathbf{Q}\sigma}) + \text{H.a.}], \tag{17}$$

where $b_{\mathbf{Q}}$ and $B_{-\mathbf{Q}^+}$ are the phonon operators; g_{ph} is the electron-phonon interaction constant.

In this case, in addition to the electron correlation functions, we shall specify the condensate averages at the initial moment:

$$\langle b_{\mathbf{Q}}(t)\rangle = \langle b_{\mathbf{Q}}(t_0)\rangle \exp\{i\omega_{\mathbf{Q}}(t-t_0)\},$$
$$\langle b_{-\mathbf{Q}}^+(t)\rangle = \langle b_{-\mathbf{Q}}^+(t_0)\rangle \exp\{-i\omega_{\mathbf{Q}}(t-t_0)\}. \tag{18}$$

The structure of the equations for the Green functions is similar to that in the preceding section except that, instead of the self-energy part $\hat{\Sigma}_{12}$, we now have the phonon condensate averages. The collisionless kinetic equations for the renormalized distribution functions averaged over the angular variables are

$$\frac{i\partial\hat{\rho}}{\partial t} = [\hat{H}_{ph}, \hat{\rho}],$$
$$\hat{\rho}(\xi t) = \begin{vmatrix} f_{11}(\xi t) & f_{12}(\xi t) \\ f_{21}(\xi t) & f_{22}(\xi t) \end{vmatrix}, \quad \hat{H}_{ph}(\xi t) = \begin{vmatrix} \xi & g_{ph}u(t) \\ g_{ph}u(t) & -\xi \end{vmatrix},$$
$$u(t) = \langle b_{\mathbf{Q}}(t)\rangle + \langle b_{-\mathbf{Q}}^+(t)\rangle. \tag{19}$$

The equation for the phonon condensate average is as follows:

$$\left(\frac{\partial^2}{\partial t^2} + \omega_{\mathbf{Q}}^2\right) u(t) = 2g_{ph}N(0) \sum_{v} \int d\xi \,\mathrm{Re}\, f_{12}(\xi t). \tag{20}$$

In this case the energy integral is

$$E = 2\left\{-N(0)\mathrm{Tr}\int d\xi (\hat{\rho}(\xi t)\hat{H}_{ph}(\xi t)) + \frac{1}{4}\left[\left(\frac{\partial u}{\partial t}\right)^2 + \omega_{\mathbf{Q}}^2 u^2(t)\right]\right\}. \tag{21}$$

The first term in Eq. (21) represents the energy of quasiparticles, the second and third terms are the kinetic and potential energies of the classical phonon condensate field. Under equilibrium conditions ($\dot{u} \equiv 0$, $u \neq 0$) Eq. (21) gives the energy of the insulator (semiconductor) phase. The equilibrium values at $T = 0$ are

$$f_{11}(\xi) = \xi/\varepsilon, \quad f_{12}(\xi) = \Sigma/\varepsilon, \quad \Sigma = g_{ph}u, \quad \varepsilon = (\xi^2+\Sigma^2)^{1/2}, \tag{22}$$

and the energy is

$$E = -2N(0)\left\{2\int_0^{\varepsilon_F} \varepsilon d\xi - \frac{\omega_{\mathbf{Q}}^2 u^2}{4N(0)}\right\},$$
$$\Sigma = g_{ph}u = \frac{4g_{ph}^2 N(0)}{\omega_{\mathbf{Q}}^2} \int_0^{\varepsilon_F} d\xi \,\mathrm{Re}\, f_{12}(\xi). \tag{23}$$

Variation of the energy over the gap gives the usual equation for the gap with the coupling constant $\lambda_{ph} = 4g_{ph}^2 N(0)/\omega_{\mathbf{Q}}^2$.

We shall now consider the evolution of small perturbations. We shall assume that initially a symmetric perturbation of the distribution functions of particles (electrons and holes) is applied. The gap in the electron spectrum and the anomalous phonon averages are assumed to be in equilibrium at $t = 0$. Linearization of Eqs. (19) and (20) gives, after Laplace transformation,

$$\delta u(p) = \frac{4\omega_{\mathbf{Q}}^2 \Sigma}{\lambda_{ph} p}\left[\int_0^{\varepsilon_F} \frac{d\xi\, \xi f^{(0)}}{p^2+4\varepsilon^2}\right] \times \left[\int_0^{\varepsilon_F} \frac{p^4 + p^2(4\varepsilon^2+\omega_{\mathbf{Q}}^2) + 4\Sigma^2\omega_{\mathbf{Q}}^2}{\varepsilon(p^2+4\varepsilon^2)} d\xi\right]^{-1},$$
$$\delta f_{11}(\xi t=0) = -\delta f_{22}(\xi t=0) = (\xi/\varepsilon)\delta n, \tag{24}$$

where δn is a perturbation of the quasiparticle distribution function. The time dependence $\delta u(t)$ is determined by ana-

lytic properties in the complex plane of p corresponding to the right-hand side of Eq. (24).

Near the imaginary axis we find that the denominator of Eq. (24) is described by ($p = i\omega + 0$):

$$\varphi_3(\omega) = \int_0^{\varepsilon_F} \frac{\omega^4 - \omega^2(4\varepsilon^2+\omega_Q^2) + 4\Sigma^2\omega_Q^2}{\varepsilon(4\varepsilon^2-\omega^2)}\, d\xi + \frac{i\pi}{\omega}\omega_Q^2 \times (\omega^2-4\Sigma^2)^{1/2}\theta(\omega^2-4\Sigma^2)\operatorname{sign}\omega. \quad (25)$$

The function $\varphi_3(\omega)$ has cuts on the imaginary axis from $\pm 2i\Sigma$ to $\pm i\infty$. Moreover, $\varphi_3(\omega)$ has two poles for real values of ω ($\omega^2 < 4\Sigma^2$). The poles satisfy the equation

$$\frac{\omega^2}{\omega_Q^2}\ln\left(\frac{\varepsilon_F}{\Sigma}\right) = \frac{[\Sigma^2-(\omega/2)^2]^{1/2}}{\omega/2}\operatorname{arctg}\frac{\omega/2}{[\Sigma^2-(\omega/2)^2]^{1/2}}. \quad (26)$$

The left-hand side of Eq. (26) is a parabola and the right-hand side is a function which decreases from 1 at $\omega = 0$ to 0 at $\omega = \pm 2\Sigma$; therefore, there are always solutions with a frequency $0 \leqslant \Omega_{ph}^2 \leqslant 4\Sigma^2$. In the case when $\omega_Q/\Sigma \to \infty$, the poles $\pm\Omega_{ph}$ are driven to the edges of the cuts $\pm 2\Sigma$.

Inverse Laplace transformation of $\delta u(p)$, subject to Eqs. (25) and (26), gives (for $t \to \infty$)

$$\delta u(t) = \delta u_1(t) + \frac{4\Sigma\omega_Q^2\varphi_1(0)}{\lambda_{ph}\varphi_3(0)} - \frac{8\pi^{1/2}\omega_Q^2\Sigma\varphi(2\Sigma)}{\lambda_{ph}}\frac{\cos(2\Sigma t - \pi/4)}{(2\Sigma t)^{1/2}}, \quad (27)$$

$$\varphi(\omega) = \left(\omega_Q^2 \operatorname{Re}\varphi_1 + {}^1/_4\delta n\left(\frac{\omega}{2}\right)\operatorname{Re}\varphi_3(\omega)\right)|\varphi_3(\omega)|^{-2},$$

where $\delta u_1(t)$ is an undamped oscillatory function:

$$\delta u_1(t) = 8\Sigma\varphi_1(\Omega_{ph})\omega_Q^2\left(\lambda_{ph}\Omega_{ph}\frac{d\varphi_3(\Omega)}{d\Omega}\Big|_{\Omega_{ph}}\right)^{-1}\sin(\Omega_{ph}t).$$

Therefore, when the electron subsystem is excited, the amplitude of the sublattice displacement $\delta u(t)$ exhibits, after a long time, undamped oscillations (if no allowance is made for inelastic processes) at the renormalized phonon frequency Ω_{ph}. The terms in $\delta u(t)$ which show a power-law decay with time (damping) are related to the loss of coherence in the electron subsystem. This analysis is valid for any value of the adiabaticity parameter ω_0/Σ.

In reality, the formation of a gap as a result of the SM transition is influenced by the Coulomb and electron-phonon interactions at the same time. A study of collisionless evolution of perturbations allows us, in principle, to separate the contributions of the Coulomb and electron-phonon interactions because of their different time asymptotes.

Perturbations which are not small were modeled numerically. The initial state of the system was described by the density matrix

$$f_{11}(\xi t=0) = -f_{22}(\xi t=0) = \frac{\xi}{\varepsilon}(1-2n(\varepsilon)),$$

$$f_{12}(\xi t=0) = f_{21}(\xi t=0) = \frac{\Sigma}{\varepsilon}(1-2n(\varepsilon)),$$

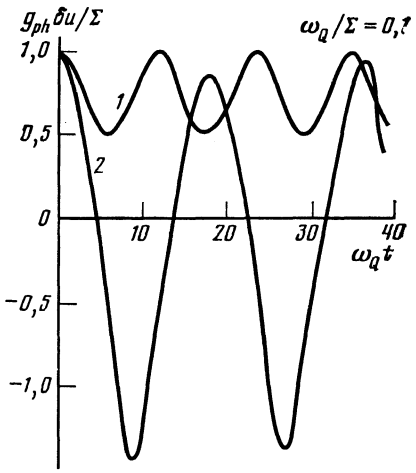

FIG. 2.

where Σ is the gap in the absence of excitations and $n(\varepsilon)$ is the distribution function of quasiparticles at $t = 0$, selected in the form

$$n(\varepsilon) = \exp(-\varepsilon^2/\varepsilon_0^2).$$

The total number of excitations is

$$n_e = \frac{1}{\Sigma N(0)}\int n(\varepsilon)\, d\xi.$$

In the case when the number of nonequilibrium excitations is less than the critical value, the system exhibits coherent oscillations and remains in the semiconductor phase (the gap does not pass through zero, as demonstrated by curve 1 in Fig. 2, corresponding to $n_e = 0.1$). On increase in n_e the system passes through states with $\Sigma = 0$, i.e., through the metal phase (curve 2 in Fig. 2, $n_e = 0.2$). The critical value of n_e depends on the nature of the density matrix at the initial moment in time.

CONCLUSIONS

We shall now consider the qualitative features of the dynamics of a transition in group IV semiconductors (Si, Ge). In the case of these substances there are several phases with similar energies. Under normal conditions only the diamond phase is stable and the rest are unstable or metastable. Transitions to other phases are possible under equilibrium conditions when pressure and temperature are varied.[23] It is shown in Ref. 4 that transitions to other phases may occur when the density of nonequilibrium carriers is altered. The analysis in Ref. 4 is valid at times longer than the time constants of transient processes in a system and the system is already in a local energy minimum.

It follows from our investigation that, in principle, transitions between different phases are possible even before the kinetic stage. In the case of Si and Ge such oscillations should be strongly damped for the following reasons. The characteristic times of transitions in the electron subsystem are of the order of $1/E_g \sim 10^{-15}$ sec, where E_g is the gap in the electron spectrum. During such a time interval there are oscillations of the covalent charge at the bonds (for fixed positions of the nuclei). The damping time of these oscillations due to "inelastic" collisions within the electron subsys-

tem is $\sim 10^{-14}$–10^{-15} sec (when the concentrations are $\sim 10^{21}$ cm^{-3}—see Refs. 3 and 24). The changes in the lattice occur in a time of $\sim 10^{-13}$ sec, and, therefore, because of rapid relaxation in the electron subsystem the lattice vibrations should be strongly damped.

Clearly, the most suitable semiconductors for the observation of coherent transitions between phases are those of the IV-VI type.[6] Structural transitions occur in them in accordance with the Peierls mechansim and, moreover, the displacements of the sublattices amount to a few percent of the lattice constant. Therefore, the concentrations of excitations at which transitions between phases become possible amount to $\sim 10^{19}$–10^{20} cm^{-3}, which are one or two orders of magnitude less than for Si and Ge.

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