

Inhomogeneous structures in an excitonic ferromagnet

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The possibility of the formation of incommensurate magnetic structures in systems with electron-hole pairing are considered. The inhomogeneous magnetization is caused by the fact that the spin and charge density waves, whose coexistence results in ferrimagnetism, are incommensurate with the crystal lattice period. On the phase diagram there is a point of contact between the normal, commensurate, and incommensurate phases (the Lifshitz point), near which the Ginzburg-Landau functional is set up and investigated. Various types of magnetic structures are discussed and the extent to which they are energywise favored is estimated.

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1. INTRODUCTION

We continue here an earlier investigation of the magnetic ordering in an excitonic dielectric.¹⁻³ It was shown in a preceding paper⁴ that a region of "inhomogeneous" excitonic ferromagnetic state exists on the phase diagram at $T=0$. By inhomogeneous state we mean one in which the spatial periods of the order parameters Δ_s and Δ_t (singlet and triplet) are incommensurate with the period of the crystal lattice. For the case of single order parameter, the possibility of the transition of an excitonic dielectric into the incommensurate state was discussed in many papers.⁵⁻⁷ A particularly thorough study was made of the transition from the commensurate into the incommensurate state of chromium and its alloys. The region of the incommensurate phase is reached at sufficiently large disparity between the dimensions of the Fermi surfaces of the electrons and holes, when the inhomogeneous solution of the self-consistency equation for the order parameter turns out to be energywise favored over the homogeneous one. The phase diagram has a point of contact between the normal (i.e., disordered), commensurate, and incommensurate ordered phases (the so-called Lifshitz point). Systems with Lifshitz points are presently intensively investigated theoretically and experimentally (see, e.g., Ref. 8).

In the present study we investigate the phase diagram of an excitonic ferromagnet at finite temperatures, and take into account the fact that the spatial periods of the charge density wave (CDW) and the spin density wave (SDW) are not commensurate with the period of the crystal lattice. In Sec. 3 we develop a two-parameter Landau expansion near the Lifshitz point and investigate the role of the long-range part of the Coulomb interaction. In Sec. 4 the functional of the free energy, obtained in Sec. 3, is analyzed for the case of one order parameter. Certain possible types of inhomogeneous solution are indicated, and phase-transition lines between the normal, commensurate, and incommensurate phases are constructed. In Sec. 5 are considered some magnetic structures that are produced when noncommensurate CDW and SDW coexist. The period of the magnetic structure is macroscopic, i.e., much larger than the atomic distances. In Sec. 6 we analyze the influence of the magnetic field on the char-

acter of the magnetic ordering in the investigated system. Some directions of further development of the theory are indicated in the Conclusion.

2. THE MODEL HAMILTONIAN

We shall use here a model Hamiltonian of a semi-metal that has one electron band and one hole band, with the extrema of the bands separated by half the reciprocal-lattice vector. The spectra ε_1 and ε_2 of the electron (1) and hole (2) bands are similar:

$$\varepsilon_{1,2} = \pm(\varepsilon(\mathbf{k}) - \varepsilon_F). \quad (1)$$

The investigation will be carried out in the high-density approximation, when all the interactions can be regarded as between points.¹ It is known that in this case the phase transition in the system is accompanied by the appearance of either a spin density wave or a charge density wave, and the critical temperature of this transition is determined respectively by the triplet (g_t) or singlet (g_s) coupling constants. Definitions of these constants in terms of the bare Coulomb and electron-phonon interactions are given in Ref. 1:

$$g_s = g_1 + g_2 + 4(2g^2/\omega_0 - g_2), \quad g_t = g_1 + g_2. \quad (2)$$

Here g_1 is a constant that describes an interaction of the density-density type of the electrons from bands 1 and 2, g_2 is the interaction connected with the interband scattering of the electrons, and $2g^2/\omega_0$ is the magnitude of the interband electron-phonon interaction.

It is assumed that the concentrations of the electrons and holes are not equal because of the presence of a reservoir (i.e., other bands that overlap the investigated ones but do not take part in the pairing and ensure an overall electroneutrality). Therefore the Hamiltonian must be supplemented by a term in the form $-\mu(\hat{n}_1 + \hat{n}_2)$, where $\hat{n}_{1,2}$ are the electron and hole density operators, respectively. Because of this ($\mu \neq 0$) the Fermi surfaces of the electron and hole bands turn out not to be exactly congruent, so that inhomogeneous solutions (slowly varying in space) can appear for the SDW and CDW. To investigate these inhomogeneous solutions near the critical temperature of the phase transition, it is necessary to construct a functional of the free energy for the singlet Δ_s (for the CDW) and triplet Δ_t (for the SDW) order parameters.

3. THE TWO-PARAMETER LANDAU EXPANSION

1. It is known¹ that in the model system described in the preceding section a phase transition is accompanied by the appearance of charge- and spin-density waves whose amplitudes are determined by the singlet (for the CDW) and triplet (for the SDW) order parameters Δ_s and Δ_t respectively. The latter, in turn, are connected with the anomalous Green's functions by the self-consistency equations³:

$$2\Delta_s = g_s T \sum_{\omega_n} \text{Sp} \hat{G}_{21}, \quad 2\Delta_t = g_t T \sum_{\omega_n} \text{Sp} \sigma \hat{G}_{21}, \quad (3)$$

where \hat{G}_{ij} are the temperature Green's functions written in the form of matrices in the spin indices

$$\hat{G}^{\uparrow\downarrow} = \begin{pmatrix} G_{ij}^{\uparrow\uparrow} & G_{ij}^{\uparrow\downarrow} \\ G_{ij}^{\downarrow\uparrow} & G_{ij}^{\downarrow\downarrow} \end{pmatrix} \quad (4)$$

where, for example, $G_{ij}^{\uparrow\uparrow} = \langle T \psi_{i\uparrow}(\mathbf{r}) \psi_{j\uparrow}^*(\mathbf{r}') \rangle$, and $\psi_{i\uparrow}(\mathbf{r})$ is the operator of annihilation of an electron in the i -th band at the point \mathbf{r} with "up" spin. In the high-density approximation, the equations for the Green's functions (4) are of the standard form:

$$\begin{aligned} \{i\omega_n + \varepsilon(-i\nabla) + \varepsilon_F - \mu - \varphi(\mathbf{r})\} \hat{G}_{11}(\mathbf{r}, \mathbf{r}') + \hat{\Delta}(\mathbf{r}) \hat{G}_{21}(\mathbf{r}, \mathbf{r}') &= \delta(\mathbf{r} - \mathbf{r}'), \\ \{i\omega_n - \varepsilon(-i\nabla) - \varepsilon_F - \mu - \varphi(\mathbf{r})\} \hat{G}_{21}(\mathbf{r}, \mathbf{r}') + \hat{\Delta}^+(\mathbf{r}) \hat{G}_{11}(\mathbf{r}, \mathbf{r}') &= 0. \end{aligned} \quad (5)$$

Here $\hat{\Delta} = \Delta_s + \Delta_t \sigma$, and the potential φ is connected with the long-range part of the Coulomb interaction due to the redistribution in space of the excess carriers that are present at $\mu \neq 0$ when inhomogeneous $\hat{\Delta}$ appear. Thus, the self-consistency equations (3) together with (5) and the equation for $\varphi(\mathbf{r})$, which will be derived below, form a closed system that makes it possible to reconstruct the form of the Landau two-parameter functional of the free energy.

2. In the derivation of the equation for $\varphi(\mathbf{r})$ we note that if an inhomogeneous order parameter $\hat{\Delta}(\mathbf{r})$ is produced in the system following the phase transition, then the local value of the produced dielectric gap $2\hat{\Delta} = |\hat{\Delta}_s(\mathbf{r}) \pm \Delta_t(\mathbf{r})|$ [see Eq. (5) of Ref. 4] varies from point to point. This causes a spatial redistribution of the electron density $n(\mathbf{r})$, as a result of which there appears in the system a slowly varying potential $\varphi(\mathbf{r})$:

$$\varphi(\mathbf{r}) = \int \frac{e^2}{|\mathbf{r} - \mathbf{r}'|} [n(\mathbf{r}') - n] d\mathbf{r}'. \quad (6)$$

It has already been taken into account here that since the system is on the average electrically neutral, the mean values are $\overline{\varphi(\mathbf{r})} = 0$ and $n = \overline{n(\mathbf{r})}$. The local change of the electron density can be easily determined knowing the Green's function and the state density N_p at the Fermi level in the bands that make up the reservoir. Since it is obvious that the change of the electron density in the bands of the reservoir is equal simply to $N_p[\mu - \mu_0 + \varphi(\mathbf{r})]$, where μ_0 is the chemical potential in the unchanged phase, it follows that

$$n(\mathbf{r}) = T \sum_{i\omega_n} \text{Sp} \hat{G}_{11} + N_p(\mu - \mu_0 + \varphi(\mathbf{r})). \quad (7)$$

Inasmuch as the total number of electrons is not changed as the result of the phase transition, Eq. (7)

must also be supplemented by the condition that the total number of particles be conserved:

$$n = \frac{1}{V} \int n(\mathbf{r}) d\mathbf{r}, \quad (8)$$

where n is the difference between the electron and hole concentrations in the unchanged phase and V is the volume of the system.

3. We proceed now to construct the free-energy functional at small values of the order parameters $\Delta_{s,t}$ accurate to $\hat{\Delta}^6$. It will be necessary to use Eqs. (3) and (5)–(7) and the condition (8). The calculation procedure is the following. Noting that the self-consistency equations can be regarded as equations for the minimization of the sought functional, we see that this functional will be obtained with the required degree of accuracy if these equations are expanded up to $\hat{\Delta}^5$. It will be shown later on that in the terms of first order in $\hat{\Delta}$ it suffices to retain the lowest derivatives of $\hat{\Delta}$ with respect to the coordinate up to the $\partial^4 \Delta / \partial r^4$, in the third-order terms up to $\partial^2 \hat{\Delta} / \partial r^2$, and in the fifth order they can be completely disregarded. The Landau functional is derived by the Gor'kov method,⁹ i.e., it is assumed that $\Delta_s(\mathbf{r})$ and $\Delta_t(\mathbf{r})$ are slowly varying functions [$v_F q < \pi T_F$, v_F is the velocity on the Fermi surface, q is a characteristic wave vector that determines the spatial periodicity of $\Delta(\mathbf{r})$]. It must also be noted here that when approximate equations are obtained for $\hat{\Delta}$ it is necessary to expand in powers of $\hat{\Delta}$ not only the Green's functions but also μ and $\varphi(\mathbf{r})$. The corresponding intermediate steps will not be presented here. We shall only demonstrate the importance of taking into account the long-range part of the potential if the form of the free-energy potential is to be correctly determined.

Accurate to terms quadratic in $\Delta_s(\Delta_t = 0)$, using (5), we obtain from (7)

$$n(\mathbf{r}) = 2N(0) \left\{ \frac{2\varphi_1}{\pi T} \Delta_s^2 + \frac{v_F^2 \varphi_3}{3(\pi T)^2} [(\text{grad } \Delta_s)^2 - 2\Delta_s \text{div grad } \Delta_s] - \mu - \varphi(\mathbf{r}) \right\} + N_p \{\mu - \mu_0 + \varphi(\mathbf{r})\}, \quad (9)$$

where $N(0)$ is the state density on the Fermi level in bands 1 and 2, while the coefficients φ_1 and φ_3 are determined by the sums of series for which expressions are given in the Appendix [(A.10) and (A.12)]. Taking into account the electroneutrality condition (8), we obtain from (9)

$$\mu - \mu_0 = \frac{1}{1+M} \left\{ \frac{2\varphi_1}{\pi T} \overline{\Delta_s^2} + \frac{v_F^2 \varphi_3}{3(\pi T)^2} [(\overline{\text{grad } \Delta_s})^2 - \overline{2\Delta_s \text{div grad } \Delta_s}] \right\}. \quad (10)$$

The superior bar denotes here averaging in the sense of (8), while $M = N_p / 2N(0)$ is the size of the reservoir. The quantity μ_0 is determined from (9) in which we put $\Delta_s = \varphi(\mathbf{r}) = 0$, $\mu = \mu_0$, and then integrate and use (8). It turns out that

$$n = 2N(0) \mu_0. \quad (11)$$

We obtain now the potential $\varphi(\mathbf{r})$, using Eq. (6), which is simply the Poisson equation integral form. Taking (9) and (20) into account, we obtain the following expression for $\varphi(\mathbf{r})$ in differential form:

$$\nabla^2 \varphi(\mathbf{r}) = 8\pi e^2 N(0) \left\{ \varphi(\mathbf{r}) - \left[\frac{2\varphi_s}{\pi T} \frac{\Delta_s^2 - \overline{\Delta_s^2}}{1+M} + \frac{v_F^2 \varphi_s}{(1+M)3(\pi T)^2} \right. \right. \\ \left. \left. \times [(\text{grad } \Delta_s)^2 - \overline{(\text{grad } \Delta_s)^2} - 2\Delta_s \text{ div grad } \Delta_s + 2\overline{\Delta_s} \text{ div grad } \Delta_s] \right] \right\}. \quad (12)$$

It is easy to verify that we can neglect the left-hand side of (12) at the required degree of accuracy, and set it equal to zero. This is easiest to do formally by solving Eq. (12) with the aid of a Fourier transformation and recognizing that the characteristic wave vector of the change of Δ_s is $q < T/v_F$, while the quantity $8\pi e^2 N(0)$ is the inverse square of the Debye radius in the high-density limit $8\pi e^2 N(0) \leq (\epsilon_F/v_F)^2$, whereas $T \ll \epsilon_F$ in the case of weak interaction. As a result we get

$$\mu - \mu_0 + \varphi(\mathbf{r}) = \frac{1}{1+M} \left\{ \frac{2\varphi_s}{\pi T} \Delta_s^2(\mathbf{r}) + \frac{v_F^2 \varphi_s}{3(\pi T)^2} [(\text{grad } \Delta_s)^2 - 2\Delta_s \text{ div grad } \Delta_s] \right\}. \quad (13)$$

It is now easy to understand why allowance for the long-range part of the potential is important for the determination of the form of the state-energy functional. In fact, assume that $e^2 = 0$ in (6). Then the series expansion of the self-consistency equations (3) will then also contain [because of the dependence of the Green's function on μ , see Eqs. (5) and (10), at $\varphi(\mathbf{r}) = 0$] terms of the type $\hat{\Delta}(\mathbf{r}) \overline{\Delta^2(\mathbf{r}')}^T$. Because of this there should appear in the functional the terms $\overline{\Delta^2(\mathbf{r}) \Delta^2(\mathbf{r}')}^T$, which are patently unphysical and correspond to an effective potential of infinite radius. On the other hand, if $e^2 \neq 0$, then no unphysical nonlocal terms will appear in the functional, since the equations (5) for the Green's functions contain μ and $\varphi(\mathbf{r})$ only in the combination $\mu + \varphi(\mathbf{r})$, and the quantity $\mu + \varphi(\mathbf{r})$ is determined according to (13) [in contrast to (10) and (12)] only by the local values of $\hat{\Delta}(\mathbf{r})$. It is also of interest that this conclusion is valid for all values of the charge e , since (13) does not contain the charge at all.

4. Finally, we write down the general form of the Landau two-parameter free-energy functional, which follows from (3) and (5). We represent this functional in the form

$$\mathcal{F} = \int F(\mathbf{r}) d\mathbf{r}, \quad (14)$$

where $F(\mathbf{r})$ is the free-energy density

$$F(\mathbf{r}) = 2N(0) \{f_s(\mathbf{r}) + f_t(\mathbf{r}) + f_{st}(\mathbf{r})\}. \quad (15)$$

Here $f_s(\mathbf{r})$ and $f_t(\mathbf{r})$ are the free-energy density components, which depend only on Δ_s (for the CDW) or Δ_t (for the SDW), and $f_{st}(\mathbf{r})$ is the interference term. Accurate to Δ_s^6 , the density $f_s(\mathbf{r})$ can be expressed in the form

$$f_s(\mathbf{r}) = \alpha_s \Delta_s^2 + \gamma_s \beta_s \Delta_s^4 + \gamma_{s,2} \Delta_s^6 + a_1 (\text{grad } \Delta_s)^2 \\ + a_2 (\text{div grad } \Delta_s)^2 + b_s \Delta_s^2 (\text{grad } \Delta_s)^2. \quad (16)$$

The density $f_t(\mathbf{r})$ has a somewhat more unique form because of the pseudovector character of the order parameter Δ_t . A distinction can be made between two cases. In the first only Δ_t^z (the z -component of Δ_t) differs from zero, corresponding to a linearly polarized SDW. This was the case considered in our preceding paper⁴ at a temperature $T = 0$. The corresponding density is completely analogous to $f_s(\mathbf{r})$ (16)

with the substitutions $\Delta_s \rightarrow \Delta_t^z$, $\alpha_s \rightarrow \alpha_t$. The second case is realized at $\Delta_t^z = 0$ and when Δ_t^x, Δ_t^y are not equal to zero. It is convenient in this case to change to the linear combinations $\Delta_{\perp} = \Delta_t^x + i\Delta_t^y$. Then

$$f_t(\mathbf{r}) = \alpha_t |\Delta_{\perp}|^2 + \gamma_t \beta_t |\Delta_{\perp}|^4 + \gamma_{t,2} |\Delta_{\perp}|^6 + a_1 |\text{grad } \Delta_{\perp}|^2 \\ + a_2 |\text{div grad } \Delta_{\perp}|^2 + b_t |\Delta_{\perp}|^2 |\text{grad } \Delta_{\perp}|^2 + b_2 (\Delta_{\perp} \text{ grad } \Delta_{\perp} - \Delta_{\perp}^* \text{ grad } \Delta_{\perp})^2. \quad (17)$$

The interference term takes in both cases the same form apart from the substitution $|\Delta_{\perp}| \rightarrow \Delta_t^z$:

$$f_{st} = \beta_{st} \Delta_s^2 |\Delta_{\perp}|^2 + \gamma_{st} (\Delta_s^2 |\Delta_{\perp}|^4 + |\Delta_{\perp}|^2 \Delta_s^4) \\ + b_{st} \{ \Delta_s^2 |\text{grad } \Delta_{\perp}|^2 + |\Delta_{\perp}|^2 (\text{grad } \Delta_s)^2 \}. \quad (18)$$

The coefficients $\alpha_s, \alpha_t; \beta_{1,2}; \gamma_{1,2}; a_{1,2}; b_{1,2}$ are defined by Eqs. (A.1)–(A.9) of the Appendix. We note here that the coefficients α_s and α_t reverse sign at the critical temperatures T_s and T_t , which can be obtained from the equation

$$2[g_{s,t} N(0)]^{-1} = \int_0^{\infty} \frac{d\xi}{\xi} \left[\text{th} \frac{\xi + \mu_0}{T_{st}} + \text{th} \frac{\xi - \mu_0}{T_{st}} \right]. \quad (19)$$

It is now appropriate to point out the following circumstance. As seen from (16) and (17), the cases of a linearly polarized SDW ($\Delta_{\perp} = 0$) and a circularly polarized one ($\Delta_{\perp} \neq 0$) differ substantially because of the term with the coefficient b_2 in (17). It will be shown below that in a definite part of the phase diagram (in the variables μ_0 and T) there exists a region where a helicoidal SDW structure is realized. It was stated earlier in some papers^{6,7} that no helical SDW structure can be realized. No account was taken in these papers, however, of the long-range part of the Coulomb potential (6), i.e., they used essentially a nonlocalizable functional with an infinite effective radius.

To conclude this section, we discuss the general form of the functional (15)–(18). The reason why it is necessary to retain in the functional, besides the ordinary gradient term $(\text{grad } \Delta)^2$ also the next derivative $(\text{div grad } \Delta)^2$ is that the coefficient a_1 can vanish and reverse sign. The need for retaining the terms with the second derivative in the Landau functional in similar cases was noted by Abraham and Dzyaloshinskii.¹⁰ The point Θ on the phase diagram (μ_0, T) , which is characterized by the system of equations $a_1 = 0, \alpha_{s,t} = 0$, is called the Lifshitz point. It is the point of contact of three phases: symmetrical (high temperature), commensurate (SDW or CDW), and incommensurate. To determine correctly the position of the phase-transition lines and their order in the vicinity of the Lifshitz point and in the inhomogeneous phase, it was necessary to retain in the functional terms of the form $\Delta^2 (\text{grad } \Delta)^2$ and Δ^6 . The role played by the latter was observed in Refs. 3 and 5. It turns out that allowance for sixth-order terms near the Lifshitz point is important for the determination of the line of ferromagnetic transitions between commensurate phases. We proceed now to investigate the functional (15).

4. INVESTIGATION OF THE FREE-ENERGY FUNCTIONAL IN THE CASE OF ONE ORDER PARAMETER

We shall investigate the solutions of the functional (14) for three cases: a) $\Delta_s \neq 0, \Delta_t = 0$; b) $\Delta_s = 0, \Delta_t = 0, \Delta_t^* \neq 0$; c) $\Delta_s = 0, \Delta_t^* = 0, |\Delta_t| = \text{const}$. The first two cases are mathematically equivalent and correspond to phase transitions into a state with a CDW ($\Delta_s \neq 0$) or into a state with a linearly polarized SDW ($\Delta_t^* \neq 0$). The last solution ($|\Delta_t| = \text{const}$) corresponds to a helicoidal structure of an SDW.

1. Let, for example, $T_s \gg T_t$ (19). Then the phase transition produces in the system CDW and an order parameter Δ_s . The corresponding density of the free-energy functional takes the form (16). The dependence of the temperature T_s on μ_0 at which the coefficient α_s reverses sign, is known (see, e.g., Ref. 3, p. 8 with $n \equiv \mu_0$). At this temperature, according to (16), a commensurate ordering parameter is produced ($\Delta_s = \text{const}$, doubling of the period), with an equilibrium value

$$\Delta_0 = -\alpha_s/\beta_1 \quad (20)$$

at $\alpha_s < 0, \beta_1 > 0$. This solution is universally known. Near the Lifshitz point Θ , the coefficient β_1 at infinite size of the reservoir ($M \rightarrow \infty$) goes through zero (A.2), (A.11) and with decreasing temperature it reverses sign and becomes negative. Therefore a solution of the type (20) is unsatisfactory and it is necessary to retain in the functional (16) terms of sixth order in Δ_s . It turns out therefore that in contrast to the case of a reservoir of low capacity ($M \rightarrow 0$), at temperatures lower than the tricritical value T_Θ , the phase transition in a system with a large reservoir proceeds via a first-order transition, and the temperature at which β_1 vanishes is the supercooling temperature of the symmetrical (semimetallic) phase. We shall investigate hereafter mainly the case of a small reservoir ($M \rightarrow 0$), and the changes in the form of the phase diagram (μ_0, T), due to the increased reservoir size, will be discussed only qualitatively.

We now must investigate the (μ_0, T) phase-diagram curves corresponding to the phase transition from the symmetrical phase into an inhomogeneous (incommensurate) state and the lines of phase transitions from the homogeneous (commensurate, $\Delta_s = \text{const}$) phase into an inhomogeneous one. The entire investigation will be carried out in the vicinity of the Lifshitz point, whose coordinates are μ_Θ, T_Θ and $\mu_\Theta/\pi T_\Theta \approx 0.61$, where in fact the free-energy expansion (16) is valid. We shall use a variational principle. The gist of the application of the variational principle to the determination of the line of the absolute instability of a certain phase relative to its transition to another state consists in the following. Let the equilibrium value of the order parameter Δ into the initial phase be known and equal to Δ_0 . It is then necessary to substitute in the functional (14) with the density (16) the trial value of Δ in the form

$$\Delta = \Delta_0 + \varepsilon(\mathbf{r}), \quad (21)$$

where $\varepsilon(\mathbf{r})$ is a small quantity having the symmetry of

the new phase, and calculate the functional (14) accurately to ε^2 . The boundary of the region of the absolute instability of the old phase is in this case determined by the inequality

$$\delta^2 \mathcal{F} / \delta \varepsilon^2 \leq 0. \quad (22)$$

A similar procedure can be used also to determine the boundaries of the region of the absolute instability of the phase with symmetry $\varepsilon(\mathbf{r})$ relative to its transition to the "old" phase. Therefore if it turns out that the regions of the instabilities of both phases overlap, then the phase transition between them is of first order, and the line of the thermodynamic equilibrium of these phases lies between the lines of their absolute instability. These lines have the meaning of the superheating and supercooling lines.

To find the absolute instability lines of the homogeneous and high-temperature phases relative to a transition into an incommensurate state, it suffices to choose $\varepsilon(\mathbf{r})$ in the form

$$\varepsilon(\mathbf{r}) = \varepsilon \cos \mathbf{q}\mathbf{r}. \quad (23)$$

In the calculation of the limit of absolute instability (supercooling line) of the high-temperature ($\Delta_0 = 0$) phase it suffices to retain in the free-energy density f_s the terms of (16) quadratic in Δ . For \mathcal{F} we obtain in this case

$$\mathcal{F} = (\alpha_s + a_1 q^2 + a_2 q^4) \varepsilon^2 N(0). \quad (24)$$

Minimizing this expression with respect to q^2 ; we find that the phase transition into an inhomogeneous state from the high-temperature phase occurs for a characteristic inhomogeneity wave vector

$$q_0^2 = -a_1/2a_2 \geq 0 \quad (25)$$

and at a temperature T_{13} determined from the condition (22)

$$4\alpha_s - a_1^2/a_2 \leq 0. \quad (26)$$

The subscripts of the critical temperature T_{13} mean that when the temperature is lowered the phase transition proceeds from the symmetrical phase 1 ($\Delta_0 = 0$) to the inhomogeneous phase 3 ($\Delta_0 \neq \text{const}$). For the commensurate phase ($\Delta_0 = \text{const}$) we shall use the subscript 2.

Near the Lifshitz Θ point the coefficients α and a_1 [(A.1), (A.1)] pass through zero and become negative with decreasing temperature. The coefficient a_2 (A.7) is larger than zero at this point. Therefore the incommensurate phase is produced only at a temperature lower than tricritical (T_Θ). It is easy to verify that the critical temperatures T_s (the temperature of the transition from the symmetrical to the commensurate phase) and T_{13} (the temperature of the transition from the symmetrical to the incommensurate phase) as functions of μ_0 have at the Lifshitz point identical derivatives $\partial T_s / \partial \mu_0 = \partial T_{13} / \partial \mu_0$ (see the figure). To prove this it suffices to differentiate Eq. (26) at the Θ point.

The temperature T_{23} , which determines the lines of the absolute instability of the restructured ($\Delta_s = \text{const}$) commensurate phase relative to its transition into the

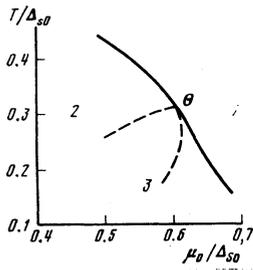


FIG. 1.

incommensurate state is determined in similar manner. However, since the trial solution takes the form (21) with $\Delta_0 \neq 0$ (20), the free-energy functional \mathcal{F} must be calculated accurate to Δ^4 . We find that the region of the absolute instability of the commensurate phase is determined by the conditions

$$q_0^2 = -\frac{a_1 + b_1 \Delta_0^2}{2a_2} \geq 0, \quad \alpha_s + 3\beta_1 \Delta_0^2 + a_1 q_0^2 + a_2 q_0^4 \leq 0. \quad (27)$$

The derivatives $\partial T_{23}/\partial \mu_0$ and $\partial T_s/\partial \mu_0$ likewise coincide here (see the figure).

We consider now the limits of the absolute-instability region of the incommensurate phase. To this end it is necessary to know the equilibrium value of the order parameter $\Delta_0(\mathbf{r})$ in this phase. It is impossible to determine it exactly. We shall therefore use a variational procedure, choosing $\Delta_0(\mathbf{r})$ in the form

$$\Delta_0(\mathbf{r}) = \Delta \cos \mathbf{q}\mathbf{r}. \quad (28)$$

The functional \mathcal{F} (14) with the density (16) has a minimum when

$$q_0^2 = -\frac{1}{2a_2} \left(a_1 + \frac{1}{4} b_1 \Delta^2 \right), \quad \Delta^2 = -\frac{\alpha_s + a_1 q_0^2 + a_2 q_0^4}{\frac{3}{4} \beta_1 + \frac{1}{2} b_1 q_0^2}. \quad (29)$$

It follows from these equations that the phase-transition line T_{13} (26), corresponding to absolute instability of the high-temperature phase, coincides with the condition for thermodynamic equilibrium between this phase and the incommensurate one (29). Thus, in the absence of a reservoir the corresponding phase transition is of second order. In the presence of a sufficiently large reservoir the coefficient ($\frac{3}{4} \beta_1 + \frac{1}{2} b_1 q^2$) in the second equation of (29) becomes negative. The transition then becomes of first order.

Another situation arises when we consider the transition between the commensurate and incommensurate phases. The absolute-instability line T_{32} of the incommensurate phase with respect to its transition to a commensurate one does not coincide here with the absolute instability line T_{23} of the commensurate phase, and lies higher. This can be easily seen if it is recognized that the line T_{32} is determined by the condition that the system (26) have a nontrivial solution in the limit $q_0^2 \rightarrow 0$. This condition is of the form

$$a_1 - b_1 \alpha_s / 3\beta_1 = 0, \quad (30)$$

and the line T_{32} is determined by the inequality (27) in the same limit:

$$a_1 - b_1 \alpha_s / \beta_1 = 0. \quad (31)$$

It is from this that the existence of superheating and supercooling temperatures T_{32} and T_{23} respectively exist in transitions between the commensurate and in-

commensurate phases. It must be noted that near the Lifshitz point a solution of the type (28) is quite satisfactory (owing to the presence of harmonics of the type $\Delta_3 \cos 3\mathbf{q}\mathbf{r}$). This, however, is insignificant for the qualitative conclusions drawn here concerning the character of the phase transitions, because they follow from the variational principle.

The total phase diagram in the vicinity of the Θ point in terms of the variables μ_0 and T is shown for the case of CDW in the figure, where the superheating and supercooling lines are shown dashed, while the solid line shows the temperature of the thermodynamic equilibrium of the phases. The phase diagram corresponding to the formation of a linearly polarized SDW is exactly of the same form.

2. We proceed now to investigate the helicoidal solution for the SDW. The corresponding density of the free-energy functional takes in this case the form (17). Choosing the solution for Δ_{\perp} in the form

$$\Delta_{\perp}(\mathbf{r}) = \Delta_{\perp} \exp(i\mathbf{q}\mathbf{r}), \quad (32)$$

which is exact for the functional (14), (17), we obtain

$$q_0^2 = -\frac{a_1 + (b_1 - 4b_2)\Delta_{\perp}^2}{2a_2} \geq 0, \quad \Delta_{\perp}^2 = -\frac{\alpha_s + a_1 q_0^2 + a_2 q_0^4}{\beta_1 + 2q_0^2(b_1 - 4b_2)}. \quad (33)$$

A comparison must be made between the energy of the helicoidal phase \mathcal{F}_{\perp} and the linearly polarized phase \mathcal{F}_{\parallel} . To this end it is necessary to know the correct solution of Δ_{\perp} in the linearly polarized SDW phase.

Using the solution (29), which is valid at $q_0 v_F > \Delta$, we find with the aid of (14), (16), (17), (29), and (33) that the energy difference between the helical and linearly polarized phases is

$$\mathcal{F}_{\parallel} - \mathcal{F}_{\perp} \approx 2N(0) \left[\alpha_s - \frac{a_1^2}{4a_2} \right]^2 \frac{1}{6\beta_1}. \quad (34)$$

Thus, on the line of transition to the inhomogeneous state, which is precisely the line on which the solution (29) is valid, a helicoidal SDW is produced, and furthermore via a second-order phase transition.

It should be noted that this conclusion is valid only in the limit of a small reservoir ($M \ll 1$). This follows from the equation for q_0^2 (33). In fact, with increasing M , in accord with (A.8) and (A.9), the difference $b_1 - 4b_2$ in (33) becomes positive, and at $a_1 + (b_1 - 4b_2)\Delta_{\perp}^2 \geq 0$ no helicoidal solution is realized at all, since q_0^2 turns out to be less than zero. In addition, an increase in the size of the reservoir (as already noted above) makes the phase transition of first order. Therefore in systems with a large reservoir a first-order phase transition takes place into a state with linearly polarized SDW. A good example of this is chromium.

To conclude this section, we note that the physical reason why the helicoidal solution (32) is favored over the inhomogeneous linearly polarized solution in the case of a small reservoir lies in the long-range Coulomb interaction (6). It is seen from (13) that in the case of an inhomogeneous linearly polarized SDW there is produced in the system a potential $\varphi(\mathbf{r})$ which hinders the formation of the sinusoidal structure, and

this potential is maximal when the reservoir is small, On the other hand, formation of a helicoidal structure, in which $|\Delta_1|$ is constant, does not cause at all a spatial redistribution of the carriers, so that in this structure $\varphi(\mathbf{r}) \equiv 0$ and there is no Coulomb repulsion.

5. INHOMOGENEOUS MAGNETIC STRUCTURES

In this section we consider the question of the possible magnetic structures that are produced when singlet and triplet order parameters coexist. The case of coexistence of homogeneous SDW and CDW was considered previously in Refs. 1-3. It turned out that this case leads to formation of a ferrimagnetic phase (excitonic ferromagnet). We shall therefore investigate here the situation when at least one of the order parameters is inhomogeneous. Out of the entire manifold of possible structures, we investigate the existence of inhomogeneous CDW and SDW, when the SDW is plane-polarized, and discuss briefly the possible coexistence of a CDW with a helicoidal inhomogeneous SDW.

1. Let the critical temperature for the SDW T_t be much higher than the temperature T_s , and let a state with plane-polarized SDW of the type (29) be realized in the system. We examine the stability of such a solution with respect to formation of CDW at a certain temperature T_k . To this end it is necessary to calculate the functional (14) accurate to terms quadratic to Δ_s , which can be chosen in the form

$$\Delta_s = \varepsilon \cos(q_0 r + \Phi). \quad (35)$$

Proceeding in the usual manner we find that an incommensurate sinusoidal structure with SDW is unstable to a transition into a magnetic state at a temperature lower than T_k :

$$\alpha_s(T_k) + a_1 q_0^2 + a_2 q_0^4 = -\ln \delta^2 \frac{3\beta_1 + 2b_1 q_0^2}{3(\beta_1 - \beta_2) + 2b_1 q_0^2}, \quad \delta^2 = \frac{\Delta_{s0}}{\Delta_{s0}}, \quad (36)$$

at $\Phi = 0$ and $3(\beta_1 - \beta_2) + 2b_1 q_0^2 > 0$; alternately

$$\alpha_s(T_k) + a_1 q_0^2 + a_2 q_0^4 = -\ln \delta^2 \frac{3\beta_1 + 2b_1 q_0^2}{3\beta_1 - \beta_2 - 4b_1 q_0^2} \quad (37)$$

at $\Phi = \pi/2$ and $3\beta_1 - \beta_2 - 4b_1 q_0^2 > 0$. Taking into account the expressions for β_1 and β_2 [(A.2) and (A.3)], we see that the magnetic instability (the appearance of Δ_s against the background of Δ_t) sets in usually at the temperature (36), at which a CDW that is shifted in phase by $\pi/2$ relative to the SDW appears. We note that Eq. (36) goes over at $q_0 \rightarrow 0$ into the corresponding formula of Refs. 2 and 3, where the Curie temperature is determined for the coexistence of homogeneous order parameters Δ_s and Δ_t . The coexistence of CDW and SDW with a relative phase shift $\pi/2$ does not lead to a homogeneous magnetization. In fact, it is shown in Ref. 4 that the magnetic moment of the system is $\mathbf{M} \sim \Delta_s(\mathbf{r})\Delta_t(\mathbf{r}) \sim \cos q_0 \mathbf{r} \sin q_0 \mathbf{r}$, i.e., it oscillates at double the period of the incommensurate structure. Such a picture might be realized in chromium, but it is very difficult to observe because of the small magnitude of the effect (of the wavelength of the magnetic superstructure $\sim \pi/q_0$).

2. We now discuss briefly the conditions for the coexistence of a helicoidal SDW with a homogeneous CDW. The helicoidal SDW can be visualized as a superposition of SDW that are plane-polarized in perpendicular planes and whose phases are shifted by $\pi/2$.

Assume that against the background of such a helicoidal SDW there is produced a CDW having the same period. According to conditions (36) and (37) for the coexistence of SDW and CDW with different phase shift Φ between Δ_t and Δ_s , we see that for one of the phase relations, $\Phi = \pi/2$, (37), the coexistence of SDW and CDW is favored, for the other (36), at the specified numerical values of the coefficients β_1 and β_2 [(A.2) and (A.3)] the coexistence conditions are not satisfied. And since the helicoidal structure consists of two sets of SDW, it is quite natural for the coexistence of an inhomogeneous order parameter Δ_s with a helicoidal SDW to be always energywise unfavored. This does not hinder in any way the coexistence of a commensurate CDW with a helicoidal SDW. The result obtained in this case is practically obvious from the form of the densities of the free-energy functional (16), (17), (18), and Eqs. (33). The Curie temperature for a magnetic transition from a helicoidal structure, following the appearance of a commensurate CDW near the Lifshitz Θ point, agrees with the results (36) if we let in the latter $q_0^2 \rightarrow 0$. In this case, in accordance with Eq. (38) for the magnetic moment $\mathbf{M} \sim \Delta_s \Delta_t$, large-scale magnetization is produced in the system and rotates in a plane perpendicular to the helicon axis with a period $2\pi/q_0$.

6. INFLUENCE OF EXTERNAL MAGNETIC FIELD

The magnetic field \mathbf{H} is introduced into the functional (14) by adding to the free-energy density functional the term³

$$f(\mathbf{H}) = -\gamma \Delta_s(\mathbf{r}) [\mathbf{H} \times \Delta_t(\mathbf{r})], \quad (38)$$

where $\gamma = (4\pi \mu_B / \pi T) \varphi_1$, μ_B is the Bohr magneton.

The influence of the homogeneous field on the linearly polarized SDW coexisting with the CDW in the inhomogeneous phase is obvious. The external field \mathbf{H} simply shifts them relative to the phase, so that a magnetic moment is induced in the system. A more peculiar action is exerted by an external homogeneous field on the critical temperature T_c , at which the coexistence of the commensurate CDW and the helicoidal SDW sets in.

To determine this influence, we consider a system in a field \mathbf{H} parallel to the z axis, in which in the absence of the field there is only a commensurate CDW. If a magnetic field \mathbf{H} is superimposed on such a system, then with increasing \mathbf{H} there will be induced in the system a commensurate triplet order parameter Δ_t (SDW). We now change the parameters of the system (for example, the temperature) near the Lifshitz Θ point without changing the field, so that the system tends to become unstable relative to the appearance in it of a helicoidal SDW. The influence of the external homogeneous field H on T_c of the helicoidal instability can

be obtained by calculating the functional \mathcal{F} accurate to terms quadratic in \mathbf{H} , Δ_s^0 , and Δ_s^1 . This can be easily done near the Lifshitz point, where the gradient terms in the functional make a small contribution. It should be noted that inasmuch with such a formulation of the problem the triplet order parameter turns out to be three-dimensional, an additional term appears in the total density of the functional

$$f(\mathbf{r}) = \beta_s (\Delta_s^1)^2 |\Delta_s^0|^2. \quad (39)$$

Minimizing the total functional (14) at the required degree of accuracy, we find that in an external magnetic field H there is induced in the system the triplet parameter of the homogeneous SDW:

$$\Delta_s^1 = \frac{1}{2} \gamma \Delta_s^0 H (\alpha_s - \alpha_s')^{-1}, \quad (40)$$

where

$$(\Delta_s^0)^2 = -\alpha_s / \beta_s. \quad (41)$$

In the derivation of (40) we used the approximate equality $\beta_1 \approx \beta_2$, which is valid near the Lifshitz point [(A.2), (A.3)] at which $\varphi_2 = 0$ (A.11). This gives rise to a correction to the singlet order parameter, $\Delta_s = \Delta_s^0 + \Delta_s^1$, where

$$\Delta_s^1 = -\frac{1}{2} (\gamma H)^2 \alpha_s^{-1} (\alpha_s - \alpha_s')^{-1}. \quad (42)$$

Ultimately, taking into account the results of (33), we find that whereas in the absence of a magnetic field the temperature T_c of the transition from the homogeneous state with CDW into a state with coexistence of a homogeneous CDW and a helicoidal SDW is determined by the condition

$$\alpha_s + a_1 q_0^2 + a_2 q_0^4 - \alpha_s \leq 0, \quad (43)$$

in a magnetic field this condition becomes much more stringent

$$\alpha_s + a_1 q_0^2 + a_2 q_0^4 - \alpha_s + \frac{1}{4} (\gamma H)^2 (\alpha_s - \alpha_s')^{-1} \left[1 - \frac{\alpha_s}{\alpha_s - \alpha_s'} \right] \leq 0. \quad (44)$$

Therefore an external magnetic field should lead to a sudden change of the helicoidal magnetism into ferromagnetism, in analogy with what was observed in M and Si.¹¹ We stipulate immediately that the present paper does not claim to explain the properties of M and Si, all the more since the band structure of this compound has not been sufficiently well studied.

7. CONCLUSION

The Landau two-parameter expansion method used in the present paper makes it possible to determine qualitatively the character of the magnetic solutions in the incommensurate phase. A substantial shortcoming of this analysis is, however, that with increasing distance from the Lifshitz point, on going into the region of noncommensurate solutions, it is necessary to take into account an increasingly larger number of higher gradient terms with respect to Δ , owing to the strong increase of the role of the harmonic components of the SDW and CDW, and owing to the rapid increase of the quantity q which characterizes their spatial periodicity. In a recent paper, Kotani¹² discusses the role of higher

harmonics of CDW and SDW at $T=0$, and shows that in the case of the two-band model the helicoidal SDW structure is not favored at any value of q . This result was obtained without allowance for the long-range Coulomb interaction. As shown above, allowance for this interaction makes the helicoidal SDW structure favorable near the Lifshitz point at a small reservoir size.

We have neglected in our analysis the spin-orbit interaction, which can exert a noticeable influence on the form of the magnetic solutions (in particular, a preferred direction of the magnetization appears). The absence of sufficient experimental data on the band structure and the Fermi surfaces of weak ferromagnets (ZrZn₂, Sc₃In, MnSi, and others) does not make it possible at present to carry out calculations within the framework of a more realistic model. Nonetheless, it is clear even now that many data on NMR, on the relaxation of the nuclear spin, and results of neutron diffraction investigations of these substances fit qualitatively within the scheme of "excitonic ferromagnetism."³

APPENDIX

The equations presented in the Appendix were obtained with the aid of Gor'kov's standard procedure (Ref. 9, Sec. 38). In our case it reduces to a solution of Eqs. (3) and (5)–(8) by a series expansion in powers of $\hat{\Delta}$ with accuracy to $\hat{\Delta}^5$, with $\hat{\Delta}$ assumed to be a slowly varying function of the coordinate r . We have

$$\alpha_{s,t} = \tau_{s,t} \left(1 - \frac{4\mu_0}{\pi T} \varphi_1 \right), \quad \tau_{s,t} = \frac{T - T_{s,t}}{T_{s,t}}, \quad (A.1)$$

$$\beta_1 = (\pi T)^{-2} \varphi_2 + 8(1+M)^{-1} (\pi T)^{-2} \varphi_1^2, \quad (A.2)$$

$$\beta_2 = 3(\pi T)^{-2} \varphi_2 + 8(1+M)^{-1} (\pi T)^{-2} \varphi_1^2, \quad (A.3)$$

$$\gamma_1 = \frac{1}{4} (\pi T)^{-1} \varphi_1 + 12(1+M)^{-1} (\pi T)^{-1} \varphi_1 \varphi_2 + 8(1+M)^{-2} (\pi T)^{-1} \varphi_1 \varphi_3, \quad (A.4)$$

$$\gamma_2 = \frac{1}{4} (\pi T)^{-1} \varphi_1 + 84(1+M)^{-1} (\pi T)^{-1} \varphi_1 \varphi_2 + 24(1+M)^{-2} (\pi T)^{-1} \varphi_1 \varphi_3, \quad (A.5)$$

$$a_1 = \frac{1}{6} \nu_F^2 (\pi T)^{-2} \varphi_2, \quad (A.6)$$

$$a_2 = \frac{1}{10} \nu_F^4 (\pi T)^{-4} \varphi_1, \quad (A.7)$$

$$b_1 = \frac{1}{12} \nu_F^2 (\pi T)^{-1} \varphi_1 + \frac{2}{3} \nu_F^2 (1+M)^{-1} (\pi T)^{-1} \varphi_1 \varphi_3, \quad (A.8)$$

$$b_2 = \frac{1}{24} \nu_F^2 (\pi T)^{-1} \varphi_1 + \frac{1}{2} \nu_F^2 (1+M)^{-1} (\pi T)^{-1} \varphi_1 \varphi_3. \quad (A.9)$$

Here

$$\varphi_1 = \sum_{k \geq 0} \frac{\mu_0}{\omega_k} (2k+1)^{-2} \left(1 + \frac{\mu_0^2}{\omega_k^2} \right)^{-2}, \quad (A.10)$$

$$\varphi_2 = \sum_{k \geq 0} \left(1 - 3 \frac{\mu_0^2}{\omega_k^2} \right) (2k+1)^{-3} \left(1 + \frac{\mu_0^2}{\omega_k^2} \right)^{-3}, \quad (A.11)$$

$$\varphi_3 = \sum_{k \geq 0} \frac{\mu_0}{\omega_k} \left(\frac{\mu_0^2}{\omega_k^2} - 1 \right) (2k+1)^{-4} \left(1 + \frac{\mu_0^2}{\omega_k^2} \right)^{-4}, \quad (A.12)$$

$$\varphi_4 = \sum_{k \geq 0} \left(10 \frac{\mu_0^2}{\omega_k^2} - 5 \frac{\mu_0^4}{\omega_k^4} - 1 \right) (2k+1)^{-5} \left(1 + \frac{\mu_0^2}{\omega_k^2} \right)^{-5}, \quad (A.13)$$

where $\omega_k = \pi T(2k+1)$. In (A.1)–(A.13) the temperature T is determined by the investigated region of the phase diagram (μ_0, T) .

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Propagation of high-frequency acoustic phonons in germanium and their interaction with electron-hole drops

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The propagation of nonequilibrium transverse acoustic phonons ($T \approx 5-10$ K, $v_{\max} \approx 4 \times 10^{11}$ Hz) in Ge is investigated at 1.8 K by a technique in which heat pulses are applied and detected with a superconducting bolometer. If the sample pumping is pulsed, the signals registered by the bolometer reflect the picture of heat propagation in Ge (at $q \parallel [100]$) at velocities smaller than those of transverse sound, and characterize the change from ballistic phonon propagation to diffuse propagation with decay followed by ballistic propagation. The bolometer-signal fall-off time ($\tau \sim 10-18$ μ sec) is evidence that the phonons are produced by a relaxation mechanism connected with the cooling of the photoexcited carriers, since the phonons emitted following Auger recombination of the carriers in the EHD would produce signals with times of the order of the recombination time of the carriers in the EHD, i.e., $\tau \sim 40$ μ sec.

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The problem of heat relaxation and heat dissipation is of essential interest both for the solution of the basic problem of the behavior of a system of nonequilibrium phonons in a crystal, and for practical applications, say heat dissipation in microelectronics. A natural need arises for tracing the behavior of nonequilibrium phonons, studying their propagation in a crystal and their passage through a crystal-liquid helium interface, their interaction with impurities or nonequilibrium carriers, etc.¹

Few direct experimental observations were made of the interaction of nonequilibrium phonons with nonequilibrium carriers.²⁻⁴ The heat-pulse technique used in some studies makes it possible to observe separately in time the arrival of longitudinal and transverse mode in a ballistic regime (when the phonon mean free path Λ is less than the sample length l), and thus study directly the action of the medium on Λ and hence on the phonon propagation.

Absorption of longitudinal sound of high frequency $\nu \approx 10^{11}$ Hz by electron-hole drops (EHD) in germanium, and hence the dragging of EHD by a flux of such phonons, was observed by Hensel and Dynes.² They have also shown that transverse phonons interact weakly with EHD, the *TA*-phonon absorption being not more

than 7%. On the other hand, judging from the distribution obtained by Greenstein and Wolfe⁵ for the EHD cloud in a crystal, it is seen that a narrow flare of this cloud stretches out in the $[100]$ direction. This anisotropy of the distribution can be due either to anisotropy of the longitudinal phonons along the $[100]$ axis, which is denied in Ref. 3, or anyhow to the dragging of the drops by the *TA* phonons, although theoretically the interaction of the *TA* phonons with the carriers can be due only to nonsphericity of the conduction band or to a complex structure of the valence band of Ge.

One of the tasks undertaken in the present study was to produce a larger volume of the liquid phase than obtained by Hensel and Dynes² who used for the excitation a cw laser of low power, 6 mW (volume excitation), since we wanted to register reliably the damping of the *TA* phonons. The phonon propagation direction was specially chosen to be the $[100]$ axis of the crystal, since it is precisely in this direction that the heat propagates in practice only in the form of a transverse mode.⁶

Hensel and Dynes² cited surprisingly long EHD motion times under the influence of the *LA* phonons (up to 6 μ sec). It might seem that this time can in fact not