we consider the solution that goes through a point of the interval

$$\theta'=0, \ \theta, \ \varphi_{\theta}', \ \varphi_{\theta}; \quad 0 < \theta < \pi.$$
 (A.1)

From the expression (2.7) for the first integral, we find that

$$\varphi_0' = \pm (1 + \varepsilon \cos^2 \varphi_0)^{\frac{1}{2}}. \tag{A.2}$$

For definiteness, we choose the lower sign in (A.2). From points of the interval (A.1), which lies on the limiting surface  $\theta' = 0$ , we trace solutions of the Landau-Lifshitz equations to an intersection with the limiting surface. Thus we obtain a curve of first contacts  $\Gamma_1$  for the interval (A.1). A curve of last contacts  $\Gamma_{-1}$  for the interval (A.1) is constructed similarly. To a point of intersection of the interval (A.1) with the curve of last contacts corresponds a break in the curve  $\Gamma_1$ . Let the break occur at  $\theta = \theta_0(\varphi_0)$ . Then if the hypothesis of coincidence of curves of first and of last contacts is correct, the curve  $\Gamma_{-1}$  must also experience a break at the point  $\theta = \theta_0$ . A break point of the curve  $\Gamma_1$  was found and localized numerically; that is, values  $\theta_{\pm}$  of the polar angle were found that lay on opposite sides of the break:

$$\theta_{-} < \theta_{0} < \theta_{+}, \quad \theta_{+} - \theta_{-} < \delta,$$

and it was verified that the coordinates  $\varphi'$  of the points on the curve  $\Gamma_{-1}$  corresponding to the values  $\theta_{\pm}$  had different signs. Thus the curve  $\Gamma_{-1}$  also experiences a break. This procedure was carried out for various values of  $\varphi_0$ , with  $\delta \sim 10^{-5}$ , and everywhere the same result was obtained. Introduction of a parameter  $\beta > 0$  or of an external field leads to the result that the breaks in the curves  $\Gamma_1$  and  $\Gamma_2$  occur at different points of the interval (A.1); that is, there occurs a disintegration of the continuous set of self-localized solutions with a single nodal point with respect to the polar angle  $\theta$ .

<sup>1</sup>V. G. Bar'yakhtar, B. A. Ivanov, and A. L. Sukstanski<sup>ĭ</sup>, Pis'ma Zh. Eksp. Teor. Fiz. **27**, 226 (1978) [JETP Lett. **27**, 211 (1978)].

<sup>2</sup>V. M. Eleonskii, N. N. Kirova, and N. E. Kulagin, Zh. Eksp. Teor. Fiz. **74**, 1814 (1978) [Sov. Phys. JETP **47**, 946 (1978)].

<sup>4</sup>Yu. I. Neimark, Metod tochechnykh otobrazhenii v teorii nelineinykh kolebanii (Method of Point Mapping in the Theory of Nonlinear Oscillations), Nauka, 1972.

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### Calculation of particle mobility at high temperature

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The motion of a quantum particle in a stochastic time-dependent Gaussian potential  $\xi(x,t)$  is considered. Assuming the correlator to be  $\langle \xi(t), \xi(t') \rangle \sim \mu$  and the correlation time  $\tau_c$  to be small, the particle mobility  $\sigma(\omega)$  and the diffusion coefficient D are calculated and found to satisfy the Einstein relation. It is shown that  $\sigma \propto T^{-2}$  in the high-temperature limit.

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#### INTRODUCTION

The calculation of electron mobility in quasi-onedimensional systems in the presence of impurities is a timely problem. The main reason is that violation, even slight, of the translational invariance leads in the one-dimensional case to strong qualitative changes in many properties of the system: the energy spectrum, the localization of all the eigenstates,<sup>1,2</sup> vanishing of the static conductivity,<sup>3-6</sup> and others. It was shown in a number of papers<sup>3-6</sup> that the mobility of noninteracting electrons in a random static potential of impurities is equal to zero. On the other hand, it has been noted<sup>5</sup> that allowance for the electron-phonon interaction leads to a mobility that differs from zero. Diagrams that give a nonzero contribution to the mobility of an electron interacting with the lattice phonons were obtained and estimated.<sup>5</sup> The calculation methods used in a number of studies<sup>4-6</sup> consist of summing an infinite chain of principal diagrams and are technically quite complicated. Interest attaches therefore to methods that make it possible to calculate the mobility without

<sup>&</sup>lt;sup>3</sup>V. M. Eleonskii, N. N. Kirova, and V. M. Petrov, Zh. Eksp. Teor. Fiz. 68, 1928 (1975) [Sov. Phys. JETP 41, 966 (1975)].

using a complicated diagram summation.

We consider in this paper the motion of an electron that interacts with lattice phonons in the case when the temperature T of the medium is much higher than the Debye frequency  $\Omega_D$ . The influence of the electron on the phonon field can then be nelgected, so that this field can be regarded as a classical field  $\xi(x, t)$  with a correlator in the form

$$\langle \xi(x, t) \rangle = 0, \quad \langle \xi(x, t) \xi(y, \tau) \rangle = \mu K(t-\tau) U(x-y), \tag{1}$$

where

$$K(t-\tau) = \Omega e^{-\Omega |t-\tau|}, \quad U(0) = 1, \quad U(x) = U(-x), \quad (2)$$

and the averaging  $\langle \ldots \rangle$  is over all the realizations of  $\xi(x,t)$ .

We note that  $K(t) \rightarrow 2\delta(t)$  as  $\Omega \rightarrow \infty$ , so that the quantity  $\tau_c = \Omega^{-1}$  plays the role of the correlation time of a random process. A correlator of this type occurs, for example, in a polar liquid,<sup>9</sup> when the relaxation processes are due to reorientation of the dipoles. A correlator of this type is obtained also when account is taken of the anharmonicity of the lattice vibrations in a solid.

When solving this problem, a distinction must be made between two cases: 1) the energy spectrum is bounded,  $|E(k)| < E_0$  (one-band approximation), and 2) the energy spectrum extends to infinity (all the bands are taken into account, or free motion in a random field). The first case was considered by us earlier,<sup>7</sup> and we have shown that at  $K(t) = 2\delta(t)$  the electron mobility is zero at all frequencies of the electric field. This is a consequence of the bounded spectrum, as a result of which the density matrix takes as  $t \to \infty$  the form  $\rho = \text{const} \cdot \delta(k - k')$ , and consequently the average electron velocity vanishes

$$\nu_{\rm sv} = \int_{-\pi}^{\pi} \frac{\partial \varepsilon}{\partial k} \rho(k,k) dk \xrightarrow[t \to \infty]{} \frac{\delta(0)}{2\pi} [\varepsilon(\pi) - \varepsilon(-\pi)] = 0.$$

It must be specially emphasized that it is precisely the bounded character of the spectrum which causes  $v_{av}$  to vanish, although the density matrix has solutions of the form  $\rho = \text{const} \cdot \delta(k - k')$  regardless of the character of the spectrum [see Eq. (9)]. The point is that the only solutions with physical meaning are those for which the normalization condition

$$\int \rho(k,k) dk = \delta(0)$$

where the integral is taken over all k, is satisfied. The solution  $\rho(k, k') = \text{const} \cdot \delta(k - k')$  satisfies this condition in the case of a bounded spectrum but not if the spectrum is unbounded. In otherwords, a state in which the quasimomentum is localized goes over in the course of time into a state with a quasimomentum that is uniformly spread over all of k-space. At the same time, if the process is  $\delta$ -correlated, any state localized in momentum space will spread out without limit over all of p-space. It is clear that in this case the diagonal elements of the density matrix  $\rho(p, p, t)$  tend to zero in the course of time, i.e.,  $\rho(p, p', t) \neq \delta(p - p')$ .

This difference is distinctly seen if one compares the character of the density-matrix asymptotic behavior

In view of the foregoing it is of interest to consider the problem with an unbounded energy spectrum, as will in fact be done here. We choose an energy spectrum with a square-law dispersion

$$E(k) = k^2/2m,$$
(3)

where k is the particle momentum. This model can describe the motion of a free electron in a liquid or crystal at high temperatures when, on the one hand, the reaction of the electron on the phonons is negligibility small, but on the other hand the Peierls transition is suppressed.

#### 1. DERIVATION OF BASIC EQUATIONS

To calculate the average particle velocity and its spatial moments  $\langle x \rangle$  and  $\langle x^2 \rangle$ , etc. it is convenient to work with a density matrix  $\hat{\rho}$ . To determine these quantities we use the averaging methods described in the review of Klyatskin and Tatarskii.<sup>8</sup> We assume hereafter that the parameters  $\mu$  and  $\tau_c = \Omega^{-1}$  in (2) are small quantities, and we can expand in their terms, retaining in the kernel of the equation for the density matrix  $\hat{\rho}$  only the smallest nonvanishing powers of  $\mu$  and  $\tau_c$ . The equations for  $\hat{\rho}$  and its functional derivative  $\delta \rho / \delta \xi(z, \tau)$  are (the employed methods call for calculation of  $\delta \rho / \delta \xi$ )

$$i\frac{\partial\rho}{\partial t} = \frac{1}{2m} \left[ -\frac{\partial^2}{\partial x^2} + \frac{\dot{\sigma}^2}{\partial y^2} \right] \rho(x, y, t) + \left[ \xi(x) - \xi(y) \right] \rho(x, y, t),$$

$$i\frac{\partial}{\partial t} \left[ \frac{\delta\rho}{\delta\xi(z, \tau)} \right] = \frac{1}{2m} \left[ -\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right] \frac{\partial\rho}{\delta\xi(z, \tau)}$$

$$\left[ \xi(x, t) - \xi(y, t) \right] \frac{\delta\rho}{\delta\xi(z, \tau)} + \left[ \delta(x-z) - \delta(y-z) \right] \delta(t-\tau) \rho(x, y, t).$$
(4)

We use in (4) a system of units with  $\hbar = 1$ .

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Averaging the system (4) over all the realizations of  $\xi(x,t)$  we obtain, taking (2) into account, the following equations for the averaged  $\langle \hat{\rho} \rangle$  and its functional derivative  $\delta \rho / \delta \xi$ :

$$\frac{\partial \langle \rho \rangle}{\partial t} = \frac{i}{2m} \left[ \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} \right] \langle \rho \rangle$$

$$-i\mu \int_{0}^{t} K(t-\tau) d\tau \int_{-\infty}^{+\infty} \left[ U(x-z) - U(y-z) \right] \left\langle \frac{\delta \rho}{\delta \xi(z,\tau)} \right\rangle dz,$$

$$\frac{\partial}{\partial t} \left\langle \frac{\delta \rho}{\delta \xi(z,\tau)} \right\rangle = \frac{i}{2m} \left[ \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} \right] \left\langle \frac{\delta \rho}{\delta \xi(z,\tau)} \right\rangle$$

$$-i\delta(t-\tau) \left[ \delta(x-z) - \delta(y-z) \right] \langle \rho(x,y,t) \rangle.$$
(5)

To deduce (5) from (4) we have used the known averaging rule<sup>9</sup>

$$\langle \xi(x,t)\Phi\{\xi\}\rangle = \int_{-\infty}^{+\infty} dy \int_{-\infty}^{+\infty} d\tau \langle \xi(x,t)\xi(y,\tau)\rangle \left\langle \frac{\delta\Phi}{\delta\xi(y,\tau)} \right\rangle$$

which is valid if  $\xi(x,t)$  is a Gaussian process. We assume hereafter that the field  $\xi(x,t)$  is Gaussian. In addition, we have left out of (5) the terms linear in  $\mu$ , since they lead to quadratic terms  $\mu^2$  in the kernel of the equation for the averaged matrix  $\langle \hat{\rho} \rangle$  (see below).

It is convenient to change over to the momentum representation. We introduce for this purpose new functions G, f, and  $\rho(k, k')$  in accord with the formulas

$$G(q) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} U(x) e^{-iqx} dx,$$

$$\rho(k, k') = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dx \, dy \exp(-ikx + ik'y) \langle \rho(x, y, t) \rangle,$$

$$\rho(k) = \rho(k, k),$$

$$f(k, k', t | q, \tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dx \, dy \, dz \exp[iqz - i(k+q)x \qquad (6)$$

$$+ik'y] \left\langle \frac{\delta\rho(x, y, t)}{\delta\xi(z, \tau)} \right\rangle,$$

$$f(k, t | q, \tau) = f(k, k, t | q, \tau).$$

Using (5), we obtain the following system of equations for the functions  $\rho(k, k', t)$  and  $(k, k', t|q, \tau)$ :

$$\frac{\partial \rho(k, k', t)}{\partial t} = -\frac{i}{2m} (k^2 - k'^2) \rho - i\mu \int_{0}^{t} K(t-\tau) d\tau \int_{-\infty}^{t+\sigma} G(q) \\ \times [\tilde{f}(k, k', t|q, \tau) - \tilde{f}^*(k, k', t|q, \tau)] dq, \\ \frac{\partial \tilde{f}}{\partial t} = -\frac{i}{2m} [(k+q)^2 - k'^2] f(k, k', t|q, \tau) \\ + i\delta(t-\tau) [\rho(k+q, k'+q) - \rho(k, k')].$$
(7)

at  $t < \tau$  we have

 $\tilde{f}(k, k', t | q, \tau) = 0.$ 

Equations (7) will be used to calculate the spatial moments  $\langle x \rangle$ ,  $\langle x^2 \rangle$ , etc. To calculate the average velocity and the momentum of the particle it suffices to know the function  $\rho(k)$ , which satisfies the equation

$$\frac{\partial \rho(k)}{\partial t} = -i\mu \int_{-\infty}^{t} K(t-\tau) d\tau \int_{-\infty}^{+\infty} dq G(q) [f(k,t|q,\tau) - f^{*}(k,t|q,\tau)], \quad (8)$$
$$\frac{\partial f}{\partial t} = -\frac{i}{2m} (2kq+q^{2}) f^{*} i\delta(t-\tau) [\rho(k+q) - \rho(k)];$$

at  $t < \tau$  we have

 $f(k, t \mid q, \tau) = 0.$ 

In the next section we use Eqs. (7) and (8) to calculate the mobility and the diffusion coefficient of the particle.

### 2. CALCULATION OF THE MOBILITY OF A PARTICLE IN A FIELD

We calculate now the average particle momentum  $\langle p(t) \rangle$ . To this end we obtain a closed equation for the diagonal part of the matrix  $\rho(k)$ , using the system (8) for this purpose. Solving the second equation of the system (8) and substituting the solution in the first equation of this system, we obtain the following equation for the function  $\rho(k, t)$ :

$$\frac{\partial \rho}{\partial t} = 2\mu \int_{-\infty}^{t} K(t-\tau) d\tau \int_{-\infty}^{t-\tau} dq \, G(q) \cos\left[\frac{(q^2+2kq)(t-\tau)}{2m}\right] \times \{\rho(k+q,\tau)-\rho(k,\tau)\}.$$
(9)

Solving (9) by the Laplace method, we obtain the following equation for the Laplace transform of the density matrix  $\rho(k, \eta)$ :

$$\eta \rho(k,\eta) = \rho_0(k) + \frac{2\mu\Omega(\Omega+\eta)}{(\Omega+\eta)^2} \int_{-\infty}^{+\infty} \frac{G(q-k) \{\rho(q) - \rho(k)\}}{1 + (q^2 - k^2)^2/4m(\Omega+\eta)^2} dq, \quad (10)$$

where  $\rho_0(k)$  is the initial value of the function  $\rho(k, t)$ . In the derivation of (10) we used expression (2) for the function  $K(t - \tau)$ .

To solve (10) we use the smallness of the correlation time  $\tau_c = \Omega^{-1}$ , i.e., the smallness of the parameter

$$q^4/m^2\Omega^2 \ll 1,\tag{11}$$

where

$$\overline{q^n} = \int_{-\infty}^{+\infty} G(q) q^n dq.$$
 (12)

 $\overline{q}^4 \propto l_c^{-4}$ , where  $l_c$  is the correlation length of the process  $\xi(x, t)$ .

Taking (11) into account, we expand (10) in powers of  $\Omega^{-1}$  and retain the first two terms of this expansion. As a result we have

$$\eta \rho(k,\eta) = \rho_0(k) + \frac{2\mu\Omega}{\Omega+\eta} \int_{-\infty}^{+\infty} dq \ G(q-k) \left\{ 1 - \frac{(q^2 - k^2)^2}{4m^2(\Omega+\eta)^2} \right\} \{\rho(q,\eta) - \rho(k,\eta) \}.$$
(13)

We multiply both halves of (13) by k and integrate with respect to k. As a result we obtain after simple transformations the following equation for the average momentum  $\langle P(\eta) \rangle$ :

$$\eta \langle P(\eta) \rangle = p_{0} - \frac{2\mu q^{2}}{m^{2} \Omega^{2}} \langle P(\eta) \rangle^{*}.$$
(14)

In the case of an external field  $F_0 e^{i\omega t}$  Eq. (14) takes the form

$$\eta \langle P(\eta) \rangle = p_0 - \frac{2\mu \overline{q^*}}{m^2 \Omega^2} \langle P(\eta) \rangle + \frac{F_0}{\eta - i\omega}.$$
 (15)

Hence

$$\langle P(\eta) \rangle = \frac{p_0 + F_0 / (\eta - i\omega)}{\eta + 2\mu \overline{q^*} / m^2 \Omega^2},$$
(16)

and consequently  $\langle p(t) \rangle$  takes as  $t \to \infty$  the form

$$\langle p(t) \rangle = \frac{F_0 e^{i \cdot t}}{i \omega + 2\mu \overline{q}^4 / m^2 \Omega^2}.$$
 (17)

From (17) it follows that the mobility  $\sigma(\omega)$  is given by

$$\sigma(\omega) = \frac{1}{im\omega + 2\mu \overline{q^*}/m\Omega^2}, \quad \sigma_0 = \sigma(0) \sim \frac{ml_c^4}{\mu \tau_c^2}.$$
 (18)

Similar calculations lead to the following expression for the mean squared momentum  $\langle p^2(t) \rangle$ :

$$\langle p^2(t)\rangle = \frac{m^2 q^2 \Omega^2}{3q^4} \sim \frac{m^2}{3} \frac{l_c^2}{\tau_c^2}.$$
 (19)

The deviation of the static mobility  $\sigma_0$  from zero agrees with the results of Ref. 5. It is curious to note that if we use formally formula (18) we obtain in the case of a static field ( $\tau_c = \infty$ ) the value  $\sigma_0 = 0$  for any spatial correlation  $l_c$ . In the particular case  $l_c = 0$ ,

the equality  $\sigma_0 = 0$  agrees with the results of Refs. 3-7.

Formulas (18) and (19) make it possible to determine the character of the temperature dependence of  $\sigma(T)$ . In fact, from (19) we have  $(l_c/\tau_c)^2 \propto T$ , where T is the temperature of the system. Furthermore,  $\mu \propto T$  and  $l_c \propto T^{-1}$ . The former follows from the fact that the parameter  $\mu$  is proportional to the square of the phonon operators (see also Ref. 7), and the latter from the fact that in the one-dimensional case  $l_c \propto 1/n_{\rm ph}$  $\propto \Omega_D/T$ , where  $n_{\rm ph}$  is the phonon-number density. Taking the foregoing into account, we find that

 $\sigma_0(T) \sim T^{-2}.$  (20)

## 3. CALCULATION OF THE SPATIAL MOMENTS $\langle x \rangle$ AND $\langle x^2 \rangle$ AND OF THE DIFFUSION COEFFICIENT

The spatial moments  $\langle x \rangle$  and  $\langle x^2 \rangle$  are expressed in the following manner in terms of the density matrix  $\rho(k, k')$ :

$$\langle x(t) \rangle = \int_{-\infty}^{+\infty} \hat{L}\rho \, dk, \quad \langle x^2(t) \rangle = \int_{-\infty}^{+\infty} \hat{L}^2\rho \, dk, \tag{21}$$

where the operator  $\hat{L}$  is defined by the formula

$$(\hat{L}\hat{\rho})(k) = \frac{i}{2} \left( \frac{\partial}{\partial k} - \frac{\partial}{\partial k'} \right) \rho(k, k')|_{k=k'}.$$
 (22)

Applying the operators  $\hat{L}$  and  $\hat{L}^2$  to both halves of the first equation of the system (7), we find that the functions

$$\rho_1(k) = \hat{L}\rho(k, k'), \quad \rho_2(k) = \hat{L}^2\rho(k, k')$$
 (23)

satisfy the system of equations

$$\frac{\partial \rho_1}{\partial t} = \frac{k}{m} \rho(k) - i\mu \int_0^t K(t-\tau) d\tau \int_{-\infty}^{+\infty} dq G(q) \{f_1(k,t|q,\tau) - f_1^*(k,t|q,\tau)\},$$

$$\frac{\partial \rho_2}{\partial t} = \frac{2k}{m} \rho_1(k),$$
(24)

where

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$$f_{1}(k, t|q, \tau) = Lf(k, k', t|q, \tau).$$
(25)

The terms linear in  $\mu$  were omitted from the second equation of the system (24), since their contribution to the spatial moments is of higher order in  $\mu$ .

The equation for the function  $\overline{f}_1$  is obtained by applying the operator  $\hat{L}$  to the second equation of the system (7). As a result we get

$$\frac{\partial f_{1}}{\partial t} = -\frac{t}{2m} (q^{2} + 2kq) f_{1} + i\delta(t-\tau) [\rho_{1}(k+q) - \rho_{1}(k)] + \frac{(q+2k)}{2m} f(k, t|q, \tau),$$

$$f_{1}(k, t|q, \tau) = 0 \text{ at } t < \tau,$$
(26)

where the function  $f(k, t|q, \tau)$  is the solution of the second equation of the system (8).

Solving (26) and substituting the obtained solution in (24), we get the following system of equations for the functions  $\rho_1(k)$  and  $\rho_2(k)$ :

$$\frac{\partial \rho_{1}}{\partial t} = \frac{k\rho}{m} + 2\mu \int_{\sigma}^{t} K(t-\tau) d\tau \int_{-\infty}^{+\infty} dq \ G(q) \cos\left[\frac{(q^{2}+2kq)(t-\tau)}{2m}\right] \\ \times \left\{ \left[\rho_{1}(k+q,\tau) - \rho_{1}(k,\tau)\right] + \frac{(t-\tau)(q+2k)}{2m} \left[\rho(k+q,\tau) - \rho(k,\tau)\right] \right\},$$
(27)

$$\frac{\partial \rho_2}{\partial t} = \frac{2k}{m} \rho_1.$$

It is seen directly from (27) that the average particle displacement, accurate to terms  $\sim \mu$ , is

$$\langle x(t)\rangle = \int_{-\infty}^{+\infty} \rho_1(k) dk = \int_{0}^{t} \int_{-\infty}^{+\infty} \frac{k\rho(k,\tau)}{m} dk d\tau = \frac{1}{m} \int_{0}^{t} \langle p(\tau)\rangle d\tau, \qquad (28)$$

as it should be in accord with the general theory.

We obtain now the quantity

$$\langle p_1(t) \rangle = \int_{0}^{+\infty} k \rho_1(k) dk.$$
(29)

To this end, we multiply both halves of the first equation of (27) by k and integrate with respect to k from  $-\infty$  to  $+\infty$ . We then take the Laplace transform of the resultant equation and retain the terms of principal order in  $\Omega^{-1}$ . This yields the following equation for the Laplace transform  $\langle P_1(\eta) \rangle$ :

$$\eta\langle P_{i}(\eta)\rangle = p_{i0} + \frac{\langle P^{2}(\eta)\rangle}{m} - \frac{2\mu \overline{q^{i}}}{m^{2}\Omega^{2}}\langle P_{i}(\eta)\rangle, \qquad (30)$$

where  $p_{10}$  is the initial value of  $\langle p_1(t) \rangle$ , and  $\langle P^2(\eta) \rangle$  is the Laplace transform of the mean squared momentum  $\langle p^2(t) \rangle$ .

From (30) we get

$$\langle P_i(\eta) \rangle = \frac{p_{i0} + m^{-i} \langle P^2(\eta) \rangle}{\eta + 2\mu q^{\overline{i}} / m^2 \Omega^2}.$$
(31)

Recognizing that  $\langle P^2(\eta) \rangle$  has a pole at  $\eta = 0$  [see (19)], we find that  $\langle p_1(t) \rangle$  takes as  $t \to \infty$  the form

$$(p_1(t)) = \langle p^2(\infty) \rangle m \Omega^2 / 2 \mu \overline{q^4}.$$
 (32)

It follows from (21), (27), and (32), that the mean squared displacement  $\langle x^2(t) \rangle$  is given as  $t \to \infty$  by the formula

$$\langle x^{i}(t)\rangle = \frac{2}{m} \int_{0}^{t} \langle p_{i}(\tau)\rangle d\tau = \langle p^{i}(\infty)\rangle \Omega^{i}t/\mu q^{i}.$$
(33)

From (18) and (33) it follows that the diffusion coefficient is

$$D = \langle p^2(\infty) \rangle \Omega^2 / 2\mu \overline{q^4} = \sigma_0 \langle p^2(\infty) \rangle / m.$$
(34)

Recognizing further that at equilibrium the average kinetic energy is  $E_{\rm kin} = \langle p^2(\infty) \rangle / 2m = T/2$  (one-dimensional case), we arrive at the Einstein ratio

$$D/\sigma_0 = T. \tag{35}$$

We note in conclusion that if the momenta in (9) and (27) are taken to be three-dimensional, then the Einstein equation is also satisfied. We note also that at  $\tau_c = 0$ , i.e., when the correlator is  $\langle \xi(x,t) \cdot \xi(x',t') \rangle$  $= 2\mu \delta(t-t')$ , then Eq. (9) for the density matrix  $\rho(k,k)$ is exact [see (4), (5), and (7)]. In this case, as follows from (8), particle diffusion takes place in pspace, and the average particle momentum  $\langle p(t) \rangle$  obeys the law  $\langle p(t) \rangle = F(t)$ , where F(t) is the external force, and the mean squared momentum  $\langle p^2(t) \rangle$  increases without limit like  $\langle p^2(t) \rangle = \langle p_Q^2 \rangle + 2\mu q^2 \overline{t}$ . This character of the motion leads to a mobility corresponding to the free mobility  $\sigma = 1/im\omega$  of the particle. This is precisely the relation that follows from (18) when  $\tau_c = 0$ . <sup>1)</sup> All terms of order higher than  $(\eta/\Omega)^2$  have been omitted from Eq. (14).

<sup>2</sup>R. E. Borland, Proc. R. Soc. London Ser. A 27Y, 529 (1963).

<sup>3</sup>V. L. Bychkov, Zh. Eksp. Teor. Fiz. **65**, 427 (1973) [Sov. Phys. JETP **38**, 209 (1974)].

- <sup>4</sup>V. L. Berezinskii, Zh. Eksp. Teor. Fiz. **65**, 1251 (1973) [Sov. Phys. JETP **38**, 620 (1974)].
- <sup>5</sup>A. A. Gogolin, V. I. Mel'nikov, and E. I. Rashba, Zh. Eksp. Teor. Fiz. **69**, 327 (1975) [Sov. Phys. JETP **42**, 168

(1975)].

- <sup>6</sup>A. A. Abrikosov and I. A. Ruzhkin, Zh. Eksp. Teor. Fiz. **71**, 1204 (1976) [Sov. Phys. JETP **44**, 630 (1976)].
- <sup>7</sup>A. A. Ovchinnikov and N. S. Erikhman, Zh. Eksp. Teor. Fiz. 67, 1474 (1974) [Sov. Phys. JETP 40, 733 (1975)].
- <sup>8</sup>V. I. Klyatskin and V. I. Tatarskii, Usp. Fiz. Nauk **110**, 499 (1973) [Sov. Phys. Usp. **494** (1974)].

<sup>9</sup>H. Fröhlich, Theory of Dielectrics, Oxford Univ. Press, 1958.

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# Antiferromagnet–ferromagnet and semiconductor–metal phase transitions in gadolinium sesquisulfide

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An investigation was made of the magnetic and electrical properties of  $Gd_2S_3$  crystals in the temperature range 4.2-300°K and magnetic fields up to 70 kOe. It was found that stoichiometric  $Gd_2S_3$  crystals are antiferromagnetic and semiconducting. Excess gadolinium increases the electrical conductivity and gives rise to indirect exchange via the conduction electrons. Increase in the excess gadolinium concentration in  $Gd_2S_3$  crystals produces first an inhomogeneous magnetic state (a mixture of antiferromagnetic and ferromagnetic phases) and then a homogeneous ferromagnetic state. Localized ferron states of the conduction electrons appear in the doped crystals: this is equivalent to introduction of a compensating impurity. Such compensation delays the Mott transition from the semiconducting to the metallic state. An analysis is made of possible low-temperature mechanisms of conduction in magnetic semiconductors.

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### **1. INTRODUCTION**

The indirect exchange interaction between magnetic ions via the conduction electrons in antiferromagnetic semiconductors tends to establish and maintain ferromagnetic ordering in a crystal.<sup>1</sup> Consequently, when the carrier density n exceeds a certain value  $n_F$ , such a semiconductor becomes ferromagnetic. A distinguishing feature of antiferromagnetic semiconductors is that the carrier-density-dependent magnetic transition does not occur abruptly at the point  $n = n_F$ . As shown by Nagaev,<sup>2</sup> there is a certain range of carrier densities  $[n_A, n_F]$  in which a canted antiferromagnetic order is preferred (for energy reasons) to ferromagnetic and collinear antiferromagnetic orders. However, in the range  $n > 4n_A$  this canted order is unstable if shortwavelength magnons are generated.<sup>3</sup> Therefore, at least in the range  $4n_A < n < n_F$  and possibly even at lower carrier densities the magnetic structure of a crystal should be different and have a lower energy. This structure may correspond to inhomogeneous magnetization of a crystal within the framework of a single crystal lattice which becomes split into antiferromagnetic regions with  $n < n_A$  and ferromagnetic regions with  $n > n_F$ (Ref. 4). The conduction electrons collect in the ferromagnetic part of the crystal and this gives rise to

special features of the semiconductor-metal phase transition in antiferromagnetic semiconductors.

An inhomogeneous magnetic state has been observed earlier in europium monoselenide,<sup>5</sup> europium monotelluride,<sup>6</sup> and gadolinium sulfides.<sup>7</sup> The present paper reports an investigation of the characteristics of the magnetic and semiconductor-metal phase transitions in gadolinium sesquisulfide crystals.

### 2. EXPERIMENTAL METHOD

Gadolinium sesquisulfide  $Gd_2S_3$  (or, equivalently,  $GdS_{1.50}$ ) is a wide-gap semiconductor with a high electrical resistivity. One of the modifications of this compound is known as the high-temperature phase and has the  $Th_3P_4$ -type structure in which  $\frac{1}{9}$  of the sites in the metal sublattice are unoccupied, i.e., are stoichiometric vacancies. The presence of such vacancies makes it possible to dissolve considerable amounts of gadolinium in excess of the stoichiometric formula. The outer-shell electrons of the excess atoms do not form valence bonds but their ionization energy decreases under the influence of the dielectric properties of the medium and they can easily be transferred to the conduction band. Therefore, introduction of excess gado-

<sup>&</sup>lt;sup>1</sup>N. E. Mott and W. D. Twoose, Adv. Phys. 10, 137 (1961).