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Since the relaxation parameter associated with the unequal reorientational motion under discussion is T_1^{II}, it follows from Eqs. (1) and (3) that the relevant temperature dependence gives information on the potential barrier E, which applies to the shallower potential well, i.e., to the position of the orientational defect. Therefore, in Eq. (5) the spin−lattice relaxation rate governed by the unequal-well reorientations is

$$(T_1^{-1})_{reor} = (T_1^{II})^{-1} = b\exp(-E/RT). \tag{6}$$

The physical meaning of the relationship between the rate of relaxation and the smaller of the two orientational potential barriers reflects the dominant role of the higher of the probabilities of molecular jumps in the spin−lattice relaxation process.

Our determination of the temperature dependence $T_1(T)$ thus gave information on the potential barrier which the molecules have to overcome in the course of the transition from the orientational defect state to the main equilibrium position in the crystal lattice. The results of our investigation carried out in the temperature range from 77 K to the melting points of the samples are presented in Fig. 1 and in Table I: they demonstrate the thermally activated molecular mobility in the investigated compounds. Reorientations of the molecules between the "defect" and main positions result in an exponential reduction in the spin−lattice relaxation time and "fading" of the resonance signals of the ^{35}Cl nuclei. The heights of the barriers E representing the orientational motion of molecular defects are 8.9 and 7.6 kcal/mole for $Cl(CN)C=CCl_2$ and $Cl(CN)C=C(C_6H_5)[N(CH_2\cdot CH_2)_2O]$, respectively. We can compare the values of E obtained in this way with the barrier hindering the reorientation of the molecules as a whole in crystalline $Cl_2C=CCl_2$ (according to Ref. 6, this barrier is 12.4 kcal/mole), in which the motion occurs most likely in an equal-well potential (the molecule has the twofold symmetry axis), i.e., without a change in the main equilibrium orientation in the molecular crystal lattice.

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Field dependence of the anomalous magnetoresistance

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It is shown that quantum corrections to the conductivity in fields for which the magnetic length is shorter than the mean free path but the inequality $\omega_c\tau < 1$ holds (ω_c is the cyclotron frequency and τ is the mean free time) decrease with increasing field as $H^{-1/2}$ in the two-dimensional case and logarithmically in the three-dimensional case.

The physical origin of the anomalous negative magnetoresistance is related to the fact that magnetic field suppresses quantum corrections to the conductivity.[1] The quantum corrections are due to interference of electron wave packets scattered from impurities and moving on the same classical trajectory but in opposite directions. The electrons moving along such trajectories acquire the same phase difference, which increases the probability that an electron returns to the initial point and, therefore, the mobility of electrons decreases. When closed trajectories are traversed in a magnetic field in one or the other direction, a finite phase difference depending on the shape of the trajectory is acquired. Since the coherence is broken, the probability that an electron returns to the initial point is reduced and, therefore, the conductivity increases. The negative magnetoresistance in weak fields satisfying the condition $l_H \gg l$ ($l_H = \sqrt{\hbar c/2eH}$ is the magnetic length of a particle with a charge 2e and l is the mean free path of electrons) was studied in Refs. 1-3. The weak fields in question satisfy the conditions $\omega_c\tau \ll \hbar/p_F l \ll 1$, where ω_c is the cyclotron frequency, p_F is the Fermi momentum, and τ is the mean free time.

We shall study the negative magnetoresistance in two-dimensional and three-dimensional systems subjected to magnetic fields satisfying the conditions that the magnetic length is shorter than the mean free path and the inequality $\omega_c\tau < 1$ holds.

This range of fields was studied theoretically in

Refs. 4 and 5, but we believe that the results obtained in Refs. 4 and 5 are incorrect.

We shall first quote the principal results and discuss their physical interpretation and then present their derivation in the second half of the paper.

In the two-dimensional case, the magnitude of the quantum correction to the resistivity decreases logarithmically with increasing field in weak magnetic fields satisfying $l_H \gg l$ (see Refs. 1 and 2). We shall show that such corrections decrease as $H^{-1/2}$ in strong fields satisfying $l_H < l$. The corresponding quantum correction to the conductivity is given by

$$\frac{\Delta\sigma_2(H)}{\sigma_0} = -1.58\frac{\hbar}{p_F l}\frac{l_H}{l}. \tag{1}$$

In the three-dimensional case, the quantum correction to the resistivity decreases as $H^{1/2}$ with increasing field in the limit $l_H \gg l$ (see Ref. 3). We shall show that, in fields satisfying $l_H < l$, the square-root dependence changes to a logarithmic decay. The quantum correction to the conductivity is given by

$$\frac{\Delta\sigma_3(H)}{\sigma_0} = -\frac{\pi^2}{8}\left(1-\frac{4}{\pi^2}\right)\frac{\ln(p_F l_H/\hbar)}{(p_F l/\hbar)^2}. \tag{2}$$

The quantity σ_0 in Eqs. (2) and (3) is, respectively, the conductivity of three-dimensional and two-dimensional systems neglecting quantum corrections.

The dependence of the quantum corrections on the magnetic field is shown in Fig. 1 in the three-dimensional case.

Such field dependences can be interpreted physically as follows. For $l_H < l$, interfering wave packets make significant contribution only for the simplest closed trajectories with a nonzero magnetic flux across the trajectory (Fig. 2). The probability of finding a particle at a distance R from the initial point is proportional to R^{-2} in the three-dimensional case and to R^{-1} in the two-dimensional case or, in the momentum representation, we obtain $\sim q^{-1}$. The probability that a particle traverses the closed trajectory shown in Fig. 2 (and, therefore, the quantum correction to the conductivity) is proportional to the following integral:

$$\int\limits_{q l_H > \hbar}\frac{d^d q}{q^3} \sim \begin{cases} H^{-1/2}, & d=2; \\ \ln H, & d=3. \end{cases}$$

The lower limit in this integral is $\hbar l_H^{-1}$ since the maximum size of the trajectory which preserves coherence when traversed in opposite directions is of the order of l_H.

We note that, in addition to the contributions defined by Eqs. (1) and (2), there is always the standard magnetoresistance related to the curvature of electron trajectories in a magnetic field $\Delta\sigma/\sigma_0 \sim -(\omega_c\tau)^2$. While this contribution in weak magnetic fields $\omega_c\tau < \hbar/p_F l$ is smaller than the negative magnetoresistance, the positive magnetoresistance for $\omega_c\tau \gtrsim \hbar/p_F l$ becomes of the order of or greater than the negative magnetoresistance defined by Eqs. (1) and (2) but the latter contribution can be still detected experimentally since, in contrast to the classical magnetoresistance, the negative magnetoresistant is independent of the orientation of the magnetic field relative to the electric field.[6]

Interference corrections to the conductivity which are sensitive to the magnetic field are acquired along these trajectories which have the property that the magnetic field flux across the trajectory is nonzero. It follows that only fan diagrams with a number of impurity lines greater than two need be considered in the calculation of the negative magnetoresistance. We note that diagrams with two impurity lines were incorrectly included in Ref. 4; since such diagrams are independent of H, they do not contribute to the negative magnetoresistance.

Localization corrections to the conductivity which determine the negative magnetoresistance are described by diagrams shown in Fig. 3. For fields satisfying $l_H > l$, the main contribution to the conductivity is due to the diagrams shown in Fig. 3a. When the magnetic length is shorter than or of the order of the mean free path, the diagrams in Figs. 3a and 3b become comparable. We note that it is then necessary to consider both the magnetic field dependences of the cooperon and of the electron Green functions.

Since we are mainly concerned with the range of fields satisfying $l_H < l$, we shall further neglect the process of relaxation of the phase[7] due to inelastic scattering which is important in weak fields satisfying $l_H \gtrsim L_\varphi$ (L_φ is the length in which dephasing of the wave function occurs[7]).

The corresponding correction to the conductivity can be written in the form[1)]

$$\Delta\sigma(H) = -\frac{2De^2\tau}{\pi}\,\mathrm{Tr}\{(K^3 + \Gamma\Gamma K)(1-K)^{-1}\}. \tag{3}$$

The first and second terms in Eq. (3) are the contributions of the diagrams shown in Figs. 3a and 3b. The quantity $D = v_F l/d$ is the diffusion coefficient and d is the dimensionality of space.

The operators K and Γ which appear in Eq. (3) originate from integration of the electron Green functions. They are given by[1)]

$$K = \langle (1 + (\mathbf{q}l)^2)^{-1} \rangle, \tag{4}$$

$$\Gamma = \left\langle \frac{1}{l}(1 - i\mathbf{q}l)^{-1} \right\rangle, \tag{5}$$

$$\mathbf{q} = i\nabla - \frac{2e}{c}\mathbf{A}(\mathbf{r}). \tag{6}$$

Here, $\mathbf{A}(\mathbf{r})$ is the vector potential of the magnetic field which is assumed to be parallel to the z axis in a three-dimensional system or normal to the plane of a two-dimensional system. The angular brackets in Eqs. (4) and (5) indicate averaging over the directions of the vector $\boldsymbol{l}$.

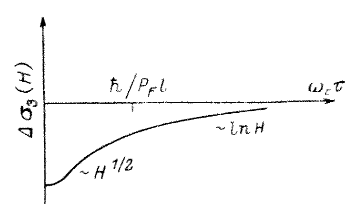

FIG. 1. Field dependence of the negative magnetoresistance in the three-dimensional case.

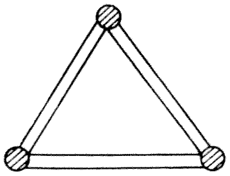

FIG. 2. Simplest trajectories which have the property that the field flux across the trajectory is nonzero.

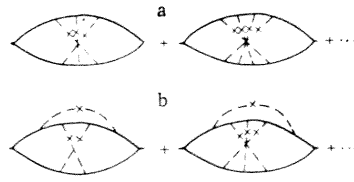

FIG. 3. Diagrams contributing to the quantum correction to the conductivity.

We shall not quote explicit expressions for the functions K and Γ in the coordinate representation but merely note that their dependence on the magnetic field is concentrated in the phase factor $\exp\left\{i\frac{2e}{c}\int_{r}^{r'}\mathbf{A}d\mathbf{r}\right\}$, where the integral is over a straight line connecting $\mathbf{r}$ and $\mathbf{r}'$.

It is convenient to use the representation of eigenfunctions of a particle with a charge 2e in a magnetic field.[3] The quantities $M \equiv \{K, \Gamma^z\}$ can be then written in the form

$$M(r, r') = \sum_{n\geq 0}\int\frac{dp_x}{2\pi}M_n\chi_{np_x}(\rho)\chi^*_{np_x}(\rho'). \tag{7}$$

The components of Γ perpendicular to the magnetic field can be written in the form

$$\Gamma^{x,y}(r, r') = \sum_{n,m\geq 0}\int\frac{dp_x}{2\pi}\Gamma^{x,y}_{n,m}\chi_{np_x}(\rho)\chi^*_{mp_x}(\rho'), \tag{8}$$

where only the following off-diagonal components are nonzero:

$$\Gamma^x_{n+1,n} = \Gamma^x_{n,n+1};\quad \Gamma^y_{n+1,n} = -\Gamma^y_{n,n+1}. \tag{9}$$

The quantity ρ in Eqs. (7) and (8) is the coordinate in the plane perpendicular to the magnetic field.

The eigenfunctions of a particle with a charge 2e in a magnetic field $\chi_{np_x}(\rho)$ are given by[8]

$$\chi_{np_x}(\rho) = \frac{\exp\left\{ip_x x - \frac{(y - p_x l_H^2)^2}{2l_H^2}\right\}H_n\left(\frac{y - p_x l_H^2}{l_H}\right)}{\pi^{1/4}\sqrt{2^n n!\, l_H}}, \tag{10}$$

where $H_n(x)$ are the Hermite polynomials.

Integrating the product of the functions $\chi_{np_x}(\rho)$ in Eqs. (7) and (8) with respect to p_x, we can isolate a factor $\exp\left\{i\frac{2e}{c}\int_{r}^{r'}\mathbf{A}d\mathbf{r}\right\}$, which cancels the same phase factor included in K and Γ. Equations (7) and (8) then reduce to a series in terms of the Laguerre polynomials.

In the two-dimensional case, K_n and $\Gamma^{x,y}_{n,m}$ are given by

$$K_n = \frac{\Delta}{2}\int_0^\infty\frac{dt\,L_n(t)}{\sqrt{t}}\exp\left\{-\frac{t}{2} - \Delta\sqrt{t}\right\}, \tag{11}$$

$$\Gamma^y_{n+1,n} = i\Gamma^x_{n+1,n} = \frac{\Delta}{4\sqrt{n+1}}\int_0^\infty dt\,L^1_n(t)\exp\left\{-\frac{t}{2} - \Delta\sqrt{t}\right\}, \tag{12}$$

where $\Delta = \sqrt{2}\,l_H/l$ and $L^p_n(t)$ are the Laguerre polynomials.

In the three-dimensional case, we obtain

$$K_n(z - z') = \frac{1}{4l}\int_0^\infty\frac{dt\,L_n(t)}{t + \left(\frac{z - z'}{\Delta l}\right)^2}\exp\left\{-\frac{t}{2} - \Delta\sqrt{t + \left(\frac{z - z'}{\Delta l}\right)^2}\right\}, \tag{13}$$

$$\Gamma^z_n(z - z') = \frac{z - z'}{4l^2\Delta}\int_0^\infty\frac{dt\,L_n(t)}{\left[t + \left(\frac{z - z'}{\Delta l}\right)^2\right]^{3/2}}\exp\left\{-\frac{t}{2} - \Delta\sqrt{t + \left(\frac{z - z'}{\Delta l}\right)^2}\right\}, \tag{14}$$

$$\Gamma^y_{n+1,n} = i\Gamma^x_{n+1,n} = \frac{1}{8l\sqrt{n+1}}\int_0^\infty\frac{dt\,L^1_n(t)}{\left[t + \left(\frac{z - z'}{\Delta l}\right)^2\right]^{3/2}}\exp\left\{-\frac{t}{2} - \Delta\sqrt{t + \left(\frac{z - z'}{\Delta l}\right)^2}\right\}. \tag{15}$$

We shall now discuss the two-dimensional case. For $\Delta < 1$, it follows from Eq. (11) that the terms even in n are given by

$$K_{2n} = \Delta\sqrt{\frac{\pi}{2}}\frac{(2n-1)!!}{(2n)!!} + o(\Delta^2). \tag{16}$$

The leading term decreases with increasing n as $\Delta n^{-1/2}$ and, therefore, the terms $\sim\Delta^2$ can be neglected for $n < \Delta^{-2}$. We can also neglect in the same limit the terms K_{2n+1} which are of the order of Δ^2.

For $\Delta < 1$ and $n < \Delta^{-2}$, Eq. (12) yields

$$\Gamma^y_{2n+1,2n} = i\Gamma^x_{2n+1,2n} = \frac{\Delta}{2\sqrt{2n+1}}. \tag{17}$$

Substituting Eqs. (7) and (8) in which K_n and $\Gamma_{n,m}$ are given by Eqs. (16) and (17) in Eq. (3) and retaining only the leading terms in Δ, we obtain

$$\frac{\Delta\sigma_2(H)}{\sigma_0} = -\frac{l_H}{\sqrt{\pi}\,p_F l^2}\sum_{n\geq 0}\frac{(2n-1)!!}{(2n)!!}\times\left\{\pi\left(\frac{(2n-1)!!}{(2n)!!}\right)^2 - \frac{1}{2n+1}\right\} = \frac{2.80\,l_H}{\sqrt{\pi}\,p_F l_H^2}. \tag{18}$$

The series in Eq. (18) converges rapidly and, therefore, the upper limit which is of the order of Δ^{-2} for $\Delta < 1$ can be set equal to infinity.

We shall now consider the three-dimensional case. For $q_z l_H > 1$ (q_z is the momentum in the direction of the field), we find that the Fourier components with respect to $(z - z')$ calculated from Eqs. (13), (14), and (15) in the leading order in Δ are given by

$$K_n(q_z) = \frac{\pi\Delta}{2\sqrt{2}\sqrt{(q_z l_H)^2 + 2n + 1}}, \tag{19}$$

$$\Gamma^z_n(q_z) = i\frac{q_z l_H}{(q_z l_H)^2 + 2n + 1}\frac{\Delta}{\sqrt{2}}, \tag{20}$$

$$\Gamma^y_{n+1,n}(q_z) = i\Gamma^x_{n+1,n}(q_z) = \frac{\sqrt{n+1}}{(q_z l_H)^2 + 2n + 2}\frac{\Delta}{2}. \tag{21}$$

Substituting Eqs. (7) and (8) in which the expansion coefficients are given by Eqs. (19)–(21) in Eq. (3), we obtain the following results which hold in the leading order in Δ:

$$\frac{\Delta\sigma_3(H)}{\sigma_0} = -\left(\frac{\pi}{4p_F l}\right)^2\left(1-\frac{4}{\pi^2}\right)\sum_{n\geqslant 0}^{N_0}\int\frac{dq_z l_H}{[(q_z l_H)^2+2n+1]^{3/2}} \qquad (22)$$
$$= -\frac{\pi^2}{8}\left(1-\frac{4}{\pi^2}\right)\frac{\ln p_F l_H}{(p_F l)^2}.$$

Equations (3), (4), and (5) hold provided $q < p_F$ and, therefore, the upper limit in Eq. (22) is $N_0 \sim (p_F l_H)^2$.

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1)Here, we set $\hbar = 1$.

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Electrical conductivity and thermoelectric power in a two-dimensional percolation region at a metal–semiconductor phase transition

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Experimental results are reported of a study of the electrical conductivity and thermoelectric power near the metal–semiconductor phase transition in vanadium dioxide films. They are well accounted for by the exact expressions for the effective properties of the medium in terms of the properties of its components. It is shown that at this phase transition in the films used there is two-dimensional percolation in the metal and semiconductor regions.

There is considerable scientific and practical value in studying the transport properties (electrical conductivity, thermoelectric properties, galvanomagnetic properties, and so on) of inhomogeneous media. However, their interpretation, especially for randomly inhomogeneous media, meets with great mathematical difficulties as regards the theoretical investigation of such systems. Nevertheless, for two-component media, exact relations have been derived[5,6] which express the effective electrical conductivity, thermoelectric power, and thermal conductivity of the medium in terms of the properties and concentrations of the components. These relations can be used, in particular, to examine transport phenomena in systems consisting of two components with very different conductivities, near a metal–insulator phase transition. This problem is one that is considered in percolation theory.

In the present investigation, the relations[5,6] mentioned were tested experimentally for the first time with a system having a metal–semiconductor phase transition as the temperature varies. The experiments used vanadium dioxide films $\sim$100 nm thick on pyroceramic glass substrates $8 \times 6 \times 0.6$ mm.

The temperature dependences were measured for the dc layer resistance and the thermoelectric power of the same samples.

The film resistance R was found by the standard four-probe method in the range 170–370 K. The error of measurement for R did not exceed 5%. The sample temperature was varied in heating at $\sim$3.5 K/min, and in cooling at $\sim$2 K/min.

To determine the thermoelectric power α in the range 280–370 K, the temperature difference $T_1 - T_2$ was 8–10 K in the plane of the sample between electrodes used to measure the thermo-emf U. The dependence $\alpha(T)$ was calculated from $\alpha(T) = \alpha[(T_1 + T_2)/2] = U/(T_1 - T_2)$. The error of measurement for α did not exceed 5%. The sample temperature was varied in heating at $\sim$3 K/min, and in cooling at $\sim$10 K/min.

Figure 1 shows the observed dependences R(T) and α(T) for a VO_2 film in the phase transition region. In the low-temperature semiconductor phase, R and α have an activation dependence, as is usual for n-type semiconductors. In the range 310–350 K, R and α decrease considerably as a result of the phase transition. In the high-temperature metal phase, α stabilized at -30 μV/K, while R continues to decrease as T rises; that is, it shows "semiconductor" behavior, which has been explained[7] as being due to the localization of conduction electrons in the metal phase because of structural disorder.

At first sight, the observed dependences R(t) and α(T) are in conflict with each other, since the corresponding branches of the hysteresis loop for R(T) are at temperatures averaging 4–7 K higher than those for α(T). This conclusion would be incorrect, however. To see

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