Quantum magnetoresistance of layered semimetals

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A model is proposed for the linear magnetoresistance recently observed in layered rare-earth diantimonides. It is based on a graphitelike energy spectrum with a small hopping between the layers and the assumption that the distance between the lowest-Landau bands exceeds both the temperature and the bandwidth along the main axis. [S0163-1829(99)13829-3]

I. INTRODUCTION

Recently a huge linear positive magnetoresistance was discovered in nonstoichiometric silver chalcognides.[1] Its characteristic feature was a surprising stability with magnetic field and temperature. The interpretation[2] was based on a rather exotic model of an inhomogeneous material consisting of clusters of metallic atoms imbedded in a medium with a very small electron concentration and a gapless spectrum with a linear dependence of energy on momentum. The linearity of the magnetoresistance with magnetic field was a consequence of the extreme quantum situation in such a model, when only one Landau band was filled with electrons. The author proposed to call it ''quantum magnetoresistance (QMR).'' A natural question appeared, whether this phenomenon could take place in a more general situation, not requiring such an exotic model.

The answer came again from experiment. In a recent publication[3] Bud'ko *et al.* discovered a linear magnetoresistance in layered rare-earth diantimonides. First of all, the zero-field resistance increases with temperature, and this shows that these substances are metals. The linear magnetoresistance effect is most strongly pronounced at low temperatures with the magnetic field perpendicular to the layers and the current parallel to the layers.

It is very simple to understand why this could be a favorable situation. Indeed the conditions for QMR are that the distance between the bottoms of the Landau bands should be larger than the temperature and the Fermi energy in the lowest band. The effect was observed at fields higher than 1 T at temperatures of a few K. The condition $\hbar\Omega \gg T$, where $\Omega = eH/mc$ is the Larmor frequency, does not require a very small effective mass. On the other hand the small hopping between the layers mans that the effective mass for the motion perpendicular to the layers is large, and hence the Fermi energy in the Landau band is reduced. For a quadratic spectrum it is easy to obtain an estimate (here we write $\hbar$ explicity; in the future we set $\hbar = 1$):

$$n \lessapprox \left(\frac{M}{m}\right)^{1/2}\left(\frac{eH}{\hbar c}\right)^{3/2}, \tag{1}$$

where n is the electron density, M is the large hopping mass, and m is the mass in the layers. For $H \sim 1$ T and $M \sim 1000\,m$ this gives $n \lessapprox 10^{18}\,\mathrm{cm}^{-3}$, and this is not very small for doped semiconductors. However, the substances under consideration are definitely metals, and for them the electron density is too small. In principle, there exists such a possibility in complex compounds, as, e.g., layered superconducting cuprates, where some atoms, or layers, play the role of ''charge reservoirs'' for conducting layers, but this is unlikely to happen in relatively simple compounds. Therefore, one can think about semimetals, where the carriers are created by a small intersection of neighboring bands.

II. MODEL

A very attractive candidate is graphite, where the spectrum, according to Slonczewski and Weiss,[4] is obtained as the result of a slight overlap of the wave functions of adjacent layers, which originally have a two-dimensional gapless spectrum with a linear dependence of the energy on momentum in both matching branches. Due to an additional dependence on p_z, graphite becomes a semimetal with alternating electron and hole ''pockets'' along p_z. The small density of carriers is defined by the weak overlap of the wave functions (or small hopping between the layers).

We will consider a model of this type, first, because it involves in a natural way a small carrier density, second, because there definitely exists a substance with a similar spectrum, and, third, since in real substances the overlap, defining the carrier density, can be further reduced by intercalation. In order to avoid complications we assume a simplified version with the Hamiltonian

$$\mathcal{H} = v\left[\sigma_x\left(p_x - \frac{e}{c}A_x\right) + \sigma_y\left(p_y - \frac{e}{c}A_y\right)\right] - t\cos(p_z d), \tag{2}$$

where we supposed that the magnetic field is directed along the z axis, and $A_y = Hx$, $A_x = A_z = 0$. The problem in the plane is similar to the one considered in Ref. 2. The bands in the absence of the field are

$$\varepsilon^{(\pm)} = \pm v p_\perp - t\cos(p_z d). \tag{3}$$

In the presence of the magnetic field we obtain the eigenvalues

$$\varepsilon_0 = -t\cos(p_z d),$$

$$\varepsilon_n^{(\pm)} = \pm v\sqrt{2eHn/c} - t\cos(p_z d), \quad n \geqslant 1, \tag{4}$$

i.e., a sequence of Landau bands with the width $2t$. The eigenfunctions are $\psi_{n\alpha}^{(\pm)} e^{ip_z z + ip_y y}$ ($\alpha=1,2$) with

$$\psi_{01}=\psi_0(x-cp_y/eH),\psi_{02}=0,$$

$$\psi_{n1}^{(+)}=\frac{1}{\sqrt{2}}\psi_n(x-cp_y/eH),$$

$$\psi_{n2}^{(+)}=-\frac{i}{\sqrt{2}}\psi_{n-1}(x-cp_y/eH), \tag{5}$$

$$\psi_{n1}^{(-)}=\frac{1}{\sqrt{2}}\psi_n(x-cp_y/eH),$$

$$\psi_{n2}^{(-)}=\frac{i}{\sqrt{2}}\psi_{n-1}(x-cp_y/eH);n\geqslant 1.$$

Here the ψ_n are the usual normalized Landau eigenfunctions of a free electron in a magnetic field

$$\psi_n=(2^n n!)^{-1/2}(\beta/\pi)^{1/4}e^{-(\beta/2)[x-(p_y/\beta)]^2} \times H_n[\sqrt{\beta}(x-p_y/\beta)] \tag{6}$$

with $\beta=eH/c$, and H_n being the Hermite polynomials.

We will consider the case when the following conditions are observed: the temperature and the width of each Landau band are much less than the magnetic splitting. In the model under consideration with $v\sim 10^8$ cm/s in a field of 1 kOe, the temperature has to be less than 100 K and the bandwidth less than 10 meV. These requirements are not very restrictive.

In the case of a pure metal the densities of electrons and holes must be equal, i.e., the band $n=0$ is half-filled. Let us consider a more general case of a slightly doped metal with an excess of some carriers, say electrons. The chemical potential is defined by the relation (we assume the spin splitting to be small).

$$2\frac{eH}{2\pi c}\int_{-\pi/d}^{\pi/d}\left\{\frac{1}{e^{[-t\cos(p_z d)-\mu]/T}+1} - \theta[-\cos(p_z d)]\right\}\frac{dp_z}{2\pi}=n_0, \tag{7}$$

n_0 being the excess electron density and d being the interlayer distance. Transforming the integral and introducing dimensionless variables we obtain

$$\int_0^{1/2}\left\{\frac{1}{e^{[-\cos(\pi x)-m]/\theta}+1}-\frac{1}{e^{[-\cos(\pi x)+m]/\theta}+1}\right\}dx=\frac{1}{h}, \tag{8}$$

where $\theta=T/t$, $m=\mu/t$, $h=H/H_0$, and $H_0=\pi n_0 cd/e$.

For $T=0$ the solution of Eq. (8) is

$$m=\sin\left(\frac{\pi}{h}\right). \tag{9}$$

The band edges are reached at $h=\pm 2$. The function $m(h)$ for different values of θ is presented in Fig. 1. In the case of a pure metal $n_0=0$, and hence, the right-hand side of Eq. (8) vanishes. This means that the solution is $m=0$ at any temperature.

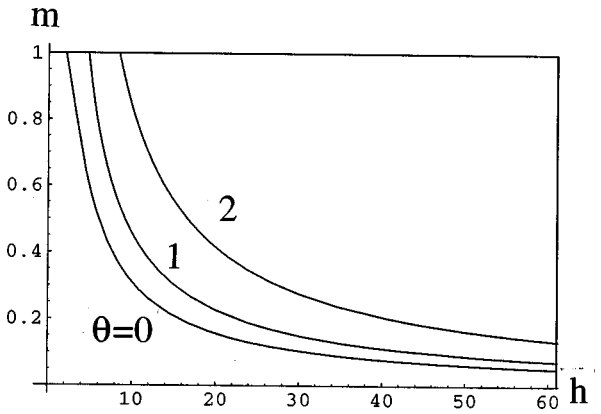

FIG. 1. Dependence of the chemical potential on magnetic field at different temperatures. Here $m=\mu/t$, $\theta=T/t$, $2t$ is the bandwidth; $h=H/H_0$, $H_0=\pi n_0 cd/e$, n_0 is the excess carrier density, d is the interlayer distance. The curves correspond to $\theta=0,1,2$.

III. HALL CONSTANT AND RESISTIVITY

To define the Hall constant we act very similarly to Ref. 2. After some calculations we obtain a natural result

$$\sigma_{xy}=\frac{ecn_0}{H},\quad \rho_{xy}=\frac{H}{ecn_0},\quad R=\frac{1}{ecn_0}. \tag{10}$$

The simplicity of this result at large fields must not be a source of confusion. It is possible to show that at smaller fields, when the band with $n=1$ starts to be filled, the Hall constant will depend on magnetic field at $n_0=\text{const}$. The same is true for a tilted magnetic field. If $n_0=0$, $\sigma_{xy}=0$, and there is no Hall effect. We will see below that such an assumption does not fit the experimental data on magnetoresistance.

The conductivity depends on electron scattering. If the scatterers are ions, the screening is important. In the usual way we obtain

$$\kappa^2=-\frac{8\pi e^2}{\varepsilon_\infty}\frac{eH}{2\pi c}T\sum_n\int_{-\pi/d}^{\pi/d}\frac{dp_z}{2\pi}\frac{1}{[i\omega_n+\mu+t\cos(p_z d)]^2}$$
$$=\frac{eH}{c}\frac{e^2}{\varepsilon_\infty d}\frac{1}{t}\int_0^{1/2}\{\cosh^{-2}[(m+\cos(\pi x))/2\theta] + \cosh^{-2}[(m-\cos(\pi x))/2\theta]\}\frac{dx}{\theta}, \tag{11}$$

where we used the same dimensionless variables, as in Eq. (8); ε_∞ is the part of the dielectric constant associated with the ion cores. At $\theta=0$ the integral is equal to $(2/\pi)(1-m^2)^{-1/2}$, and at $\theta\gg 1$ it equals 1. The spread of the electron wave functions in the plane is the magnetic length $(c/eH)^{1/2}$, and the value of the ratio $\kappa^2/(eH/c)$ is, according to Eq. (11), $(e^2/\varepsilon_\infty d)/t$, or larger, when μ is close to the boundary of the band. If ε_∞ and d are not too large, this ratio is always large, since t is the small hopping energy. This means that

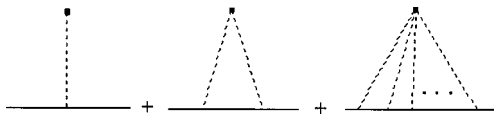

FIG. 2. Diagrams corresponding to impurity scattering in the non-Born approximation.

even if the scatterers are ions, the Coulomb interaction is screened at distances smaller than the spread of the wave function, and hence, it can be regarded, as a point interaction. This is also true for neutral scatterers. Therefore, we take it as $U\delta(\mathbf{r})$.

In our previous work[5] it was mentioned that in strong magnetic fields the Born approximation can fail. Therefore, we sum up the diagrams in Fig. 2. The result is a geometric progression containing powers of

$$U\frac{eH}{2\pi c}\int_{-\pi/d}^{\pi/d}[\omega+i\delta+\mu+t\cos(p_z d)]^{-1}(dp_z/2\pi)$$
$$=-iU\frac{eH}{2\pi cd}\frac{\theta(t-|m+w|)}{[t^2-(m+w)^2]^{1/2}}.$$

Summing up the whole series we obtain

$$\frac{1}{2\tau}=-\operatorname{Im}N_i U\left[1+iU\frac{eH}{2\pi cd}\frac{\theta(t-|\mu+\omega|)}{[t^2-(\mu+\omega)^2]^{1/2}}\right]^{-1},$$

where N_i is the concentration of scattering centers.

The evaluation of the second term in the bracket depends on the assumption about U. If we use a strongly screened Coulomb interaction, then

$$U\approx 4\pi e^2/(\varepsilon_\infty\kappa^2)\approx 4\pi dt/(eH/c),$$

and hence the second term will have an absolute value of the order of unity (actually, it is 2). Therefore, we can leave only the second term in the bracket (in the imaginary part the mistake is 1/4) and get

$$\frac{1}{2\tau}=N_i\frac{2\pi cd}{eH}[t^2-(\mu+\omega)^2]^{1/2}\theta(t-|\mu+\omega|). \quad (12)$$

The conductivity can be calculated similarly to Ref. 2, and so we arrive at the formula

$$\sigma_{xx}=\frac{e^2v^2}{T}\frac{eH}{2\pi c}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\cosh^{-2}\frac{\omega}{2T}\int_{-\pi/d}^{\pi/d}\frac{dp_z}{2\pi}$$
$$\times\operatorname{Im}G_{01}^R(\omega,p_z)\operatorname{Im}G_{12}^R(\omega,p_z), \quad (13)$$

where we have already integrated out the eigenfunctions entering the Green functions [the normalization in Eq. (5) contributes a factor 1/2], and we are left only with the energy denominators. The $-\operatorname{Im}G_{12}^R(\omega,p_z)$ in Eq. (13), is under our assumptions

$$\frac{1/2\tau}{[\omega+\mu-v(2eH/c)^{1/2}+t\cos(p_z d)]^2+(1/2\tau)^2}$$
$$\approx\frac{1}{2\tau}\frac{c}{2eHv^2}.$$

Integrating over p_z, we get

$$\sigma_{xx}=\frac{e^2v^2}{T}\frac{eH}{2\pi c}\frac{1}{4\tau^2}\frac{c}{2eHv^2}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\cosh^{-2}\frac{\omega}{2T}\frac{1}{\pi d}\int_0^{\pi}$$
$$\times\frac{dy}{(\omega+\mu+t\cos y)^2+(1/2\tau)^2}$$
$$=\frac{e^2}{8\pi}\frac{1}{T}\int_{-\infty}^{\infty}\frac{d\omega}{\cosh^2(\omega/2T)}\frac{1}{2\tau(\omega)}\frac{\theta(t-|\omega+\mu|)}{[t^2-(\omega+\mu)^2]^{1/2}}.$$

Substituting $1/2\tau$ according to Eq. (12) and integrating over ω we obtain the final result

$$\sigma_{xx}=\frac{ecN_i}{\pi H}\frac{\sinh(1/\theta)}{\cosh(m/\theta)+\cosh(m/\theta)}, \quad (14)$$

where we used dimensionless variables introduced before.

Two situations are possible: either $N_i\ll n_0$, or $N_i\gg n_0$. In the first case $\sigma_{xx}\ll\sigma_{xy}$ and $\rho_{xx}=\sigma_{xx}/\sigma_{xy}^2$. In the second case $\sigma_{xx}\gg\sigma_{xy}$ and $\rho_{xx}=1/\sigma_{xx}$. Both assumptions lead to a resistivity linear in H. However, the dependence on temperature is different. In the model considered in Ref. 2 the doping was external, and the first case was definitely more adequate. In the present situation this is not so clear. Since σ_{xx} decreases with temperature, and so does the magnetoresistivity in Ref. 3, our model can fit the experiment only in the case that $\sigma_{xx}\ll\sigma_{xy}$, i.e., if the excess density of carriers exceeds the concentration of impurities. Whether this is true can be decided only from the Hall measurements which were not performed on rare-earth antimonides. Particularly, in the case of an exactly half-filled band $n_0=0$, and there is no linear in field Hall effect. This would mean that our model is wrong.

If the model is applicable, we get

$$\rho_{xx}=\frac{HN_i}{\pi ecn_0^2}\frac{\sinh(1/\theta)}{\cosh(m/\theta)+\cosh(m/\theta)}. \quad (15)$$

The dependence on H is essentially linear, except for the variation of m. Introducing the dimensionless variable h instead of H we obtain

$$\rho_{xx}=\rho_0\frac{h\sinh(1/\theta)}{\cosh(m/\theta)+\cosh(m/\theta)};\quad \rho_0=\frac{N_i}{n}\frac{d}{e^2}. \quad (16)$$

The plot of ρ_{xx}/ρ_0 for $\theta=0.05$ and $\theta=2$ is presented in Fig. 3 [m is defined from Eq. (8)]. At low temperatures the curve bends down around $h=2$, i.e., close to the band edge.

IV. DISCUSSION

The results obtained here fit qualitatively the experimental curves in Ref. 3, although, due to the presence of rare-earth atoms various magnetic phase transitions can take place and induce deviations from pure linearity (see, e.g., Ref. 3, Fig. 23). It should be mentioned that in our model the *total* resistance is linear in field at low temperatures, and this means that it is large compared to the value at $H=0$. This agrees with experimental data.

According to our theory, at high fields only one Landau band participates in the conductivity, and this contradicts the observation of the Shubnikov–de Haas oscillations. In the range between 150 and 160 kG the period of these oscilla-

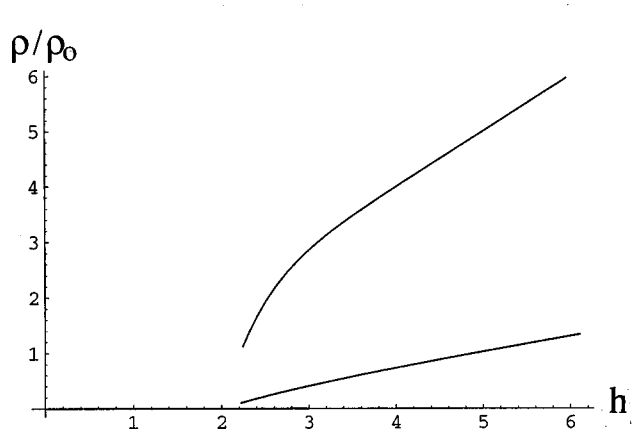

FIG. 3. Dependence of ρ/ρ_0 with $\rho_0=(N_i/n_0)(d/e^2)$, N_i is the density of scatterers on a magnetic field at low ($\theta=0.05$) and high ($\theta=2$) temperatures.

tions is 2 kG. The connection with the spectrum is (see, e.g., in Ref. 6)

$$\Delta H=2\pi\frac{e\bar{h}H^2}{cS_m}, \tag{17}$$

where S_m means the extremal area of the Fermi surface cross section by a plane perpendicular to the magnetic field. Substituting the data we get for the Fermi momentum $p_0\approx 2\times 10^{-20}\,\mathrm{g\,cm\,s^{-1}}$, and this corresponds to a large Fermi surface and an electron concentration of the order of 1 electron per atom. Hence, either our assumption about the spectrum is completely incorrect, or the oscillations have a different origin.

The possibilities are (a) the oscillations are due not to the material under investigation but to metallic leads, and (b) they are associated with the magnetic breakdown in a strong field. The objection to the first possibility could be that the leads are not single crystals, and so the oscillations are smeared out, except for the case where the leads are of a noble metal, whose Fermi surface is almost spherical. The objection to magnetic breakdown is that such a phenomenon can hardly be imagined for a graphitelike spectrum. Some light on the nature of the spectrum could come from Hall measurements. Although it is generally believed that for a metal with an even number of electrons per unit cell the amounts of carriers with opposite charges are equal, and the Hall effect is absent, this is true either at very high fields, or for an idealized model. Measurements at moderate fields would give an estimate of the electron concentration and establish the relevance of the present model.

The alternative to this model is the freeze-out model, according to which in strong magnetic fields the electrons are bound to the impurity ions, and the latter transform into neutral centers (see Ref. 1 and references therein). However, as we have shown, the screening length in strong fields is short, and therefore no large difference between charged and neutral scatterers can be expected. There are also other objections to this model discussed in Ref. 3.

The picture would be much clearer if the experiments were performed on graphite, preferably with intercalation. There are no magnetic atoms, and hence no magnetic phase transitions. The energy spectrum is definitely of the type considered here. Unfortunately the only experiments were performed over 20 years ago.[7] The samples were either polycrystalline, or badly characterized, and hence, the results obtained there (linear magnetoresistance with saturation in strong fields) cannot be considered reliable. In future experiments it would be possible, probably, to use clean and doped samples, and see both possible cases with different temperature dependences, mentioned in the previous section.

Note added in proof. Recently in the work of K. Matsabara *et al.* [J. Phys. Condens. Matter **11**, 3149 (1999)] the magnetoresistance was measured in graphite intercalated by $MoCl_5$. In the most relevant case (Fig. 4, stage 2) the magnetoresistance varied linearly with field, although it represented only a small part of the total resistance, which could be due to the presence of other, large parts of the Fermi surface.

ACKNOWLEDGMENTS

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