Slow oscillations of in-plane magnetoresistance in strongly anisotropic quasi-two-dimensional rare-earth tritellurides

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Slow oscillations of the in-plane magnetoresistance are observed in the rare-earth tritellurides and proposed as an effective tool to determine the parameters of electronic structure in various strongly anisotropic quasi-two-dimensional compounds. These oscillations do not originate from the small Fermi surface pockets, as revealed usually by the Shubnikov-de-Haas oscillations, but from the entanglement of close frequencies due to a finite interlayer transfer integral $t_z$, which allows to estimate its value. For TbTe$_3$ and GdTe$_3$ we obtain the estimate $t_z \approx 1$ meV.

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The angular and magnetic quantum oscillations (MQO) of magnetoresistance (MR) is a powerful tool to study electronic properties of various quasi-two-dimensional (Q2D) layered metallic compounds, such as organic metals (see, e.g., Refs. 1–4 for reviews), cuprate and iron-based high-temperature superconductors (see, e.g., 5–14), heterostructures 15 etc.

The Fermi surface (FS) of Q2D metals is a cylinder with weak warping $\sim 4t_z/E_F \ll 1$, where $t_z$ is the interlayer transfer integral and $E_F = \mu$ is the in-plane Fermi energy. The MQO with such FS have two close fundamental frequencies $F_0 \pm \Delta F$. In a magnetic field $B = B_z$ perpendicular to the conducting layers $F_0/B = \mu/h\omega_c$ and $\Delta F/B = 2t_z/h\omega_c$, where $h\omega_c = heB_z/m^*c$ is the separation between the Landau levels (LL).

The standard 3D theory of galvanomagnetic properties 16–18 is valid only at $t_z \gg h\omega_c$ and in the lowest order in the parameter $h\omega_c/t_z$. This theory predicts several peculiarities of MR in Q2D metals: the angular magnetoresistance oscillations (AMRO) 19–21 and the beats of the amplitude of MQO 17. One can even extract the fine details of the FS, such as its in-plane anisotropy 22 and its harmonic expansion 23, 24 from the angular dependence of MQO frequencies and from AMRO. For isotropic in-plane electron dispersion, AMRO can be described by the renormalization of the interlayer transfer integral: 22

$$t_z = t_z(\theta) = t_z(0) J_0(k_F d \tan \theta),$$

where $J_0(x)$ is the Bessel’s function, $p_F = \hbar k_F$ is the in-plane Fermi momentum, and $\theta$ is the angle between magnetic field $B$ and the normal to the conducting layers.

At $t_z \sim h\omega_c$ new qualitative features appear both in the monotonic and oscillating parts of MR. For example, the strong monotonic growth of interlayer MR $R_{zz}(B_z)$ was observed in various Q2D metals 26–32 and recently theoretically explained 33–36. The oscillating part of interlayer MR at $\mu \gg t_z \sim h\omega_c$ acquires the slow oscillations 34, 39 and the phase shift of beats.39, 40 These two effects are missed by the standard 3D theory 16–18 because they appear in the higher orders in $h\omega_c/t_z$.

The slow oscillations qualitatively originate from the product of the oscillations with two close frequencies $F_0 \pm \Delta F$, which gives the oscillations with frequency $2\Delta F$. Conductivity, being a non-linear function of the oscillating electronic density of states (DoS) and of the diffusion coefficient, has slow oscillations with frequency 2 $\Delta F$, while magnetization, being a linear functional of DoS, does not show slow oscillations 34, 39. The slow oscillations have many interesting and useful features as compared to the fast quantum oscillations. First, they survive at much higher temperature than MQO. Second, they are not sensitive to the long-range disorder, which has a strong action on fast MQO, similar to the influence of finite temperature. Third, the slow oscillations allow to measure the interlayer transfer integral $t_z$. These features make the slow oscillations to be a useful tool to study the electronic properties of Q2D metals. Until now, the slow oscillations where investigated only for the interlayer conductivity $\sigma_{zz}(B)$, when the current and the magnetic field are both applied perpendicularly to the 2D layers, and only in the organic compounds. In this paper, we study the slow oscillations of transverse intralayer MR $R_{yy}(B)$ both theoretically and experimentally in the non-organic rare-earth tritelluride Q2D layered compounds.

The rare-earth tritelluride compounds $R$Te$_3$ ($R = \text{Y, La, Ce, Nd, Sm, Gd, Tb, Ho, Dy, Er, Tm}$) have the same orthorhombic structure ($C_{mmcm}$) in the normal state. These systems exhibit a c-axis incommensurate charge-density wave (CDW) at high temperature through the whole $R$ series that was recently a subject of intense studies 11–16. For the heaviest rare-earth elements, a second

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\(a\)-axis CDW occurs at low temperature. In addition to hosting incommensurate CDW, magnetic rare-earth ions exhibit closed-spaced magnetic phase transitions below 10 K \cite{44,45} leading to coexistence and competition of many ordered states at low temperature. Therefore, any information about the Fermi surface warping in the Q2D-compounds (determined by \(t_\perp\)) is very important. For the possible observation of the slow oscillations the rare-earth tritellurides are very promising, because they have appropriate anisotropy and good metallic conductivity up to low temperatures.

For experiments we choose GdTe\(_3\) and TbTe\(_3\). Single crystals of these compounds were grown by a self-flux technique under purified argon atmosphere as described previously \cite{44}. Thin single crystal samples with a thickness typically 0.1-0.3 \(\mu\)m were prepared by micromechanical exfoliation of relatively thick crystals glued on a sapphire substrate. The quality of selected crystals and the spatial arrangement of crystallographic axes were controlled by the X-ray diffraction. From high-quality \(R(300\,K)/R(10\,K) > 100\) untwinned single crystals we cut bridges with a width 50 – 80 \(\mu\)m in well defined, namely [100] and [001], orientations. Contacts for electrical transport measurements in the four-probe configuration have been prepared using gold evaporation and cold soldering by In. Magnetotransport measurements were performed at different orientations of the magnetic field in the field range up to 9 T using a superconducting solenoid. The field orientation was defined by the angle \(\theta\) between the field direction and the normal to the highly conducting \(ac\) plane.

In Figure 1 we plot the derivative of MR \(dR/dB\) as a function of magnetic field up to \(B = 8.2\) T applied along \(b\)-axis for TbTe\(_3\) (a) and GdTe\(_3\) (c) with the current applied in the \((a,c)\) plane at \(T = 4.2\) K. For both measured compounds, at \(B > 2\) T the pronounced Shubnikov - de Haas (SdH) oscillations with a frequency \(F \approx 55 – 58\) T are observed in the derivative of MR, \(dR/dB\). At high field \((B \gtrsim 7\) T) new oscillations with higher frequency \((F \approx 0.7 – 0.8\) kT) appear, indicating the existence of several types of pockets on the partially gapped FS, resulting from the CDW formation \cite{50}. Insets in Fig. 1a, c show the corresponding MR curves indicating very weak amplitude of oscillations. In addition to rapid SdH oscillations, at low magnetic field \((B < 2\) T) the MR exhibits prominent slow oscillations with very low frequency \(F_{\text{slow}} \lesssim 4\) T. To demonstrate the periodicity of these oscillations in \(B^{-1}\), we plot the derivative \(dR/dB\) as a function of inverse magnetic field in Fig. 1 for TbTe\(_3\) (b) and for GdTe\(_3\) (d). Insets show the Fourier transform of \(dR/dB(1/B)\).

In contrast to the usual SdH oscillations, the amplitude of which decreases rapidly as temperature increases, the slow oscillations of MR are observable up to \(T \approx 40\) K, as can be seen from Fig. 2 where we show the temperature evolution of slow oscillations for GdTe\(_3\) and TbTe\(_3\). This suggests that the observed slow oscillations originate not from small FS pockets, but from the FS warping due to \(t_\perp\), similarly to the slow oscillations of interlayer MR in the organic superconductor \(\beta-(\text{BEDT-TTF})_2\text{InBr}_3\) \cite{34}. If so, the observed slow oscillations give an excellent opportunity to find the value of \(t_\perp\) and of the product \(k_F d\) at low temperature in tritellurides TbTe\(_3\) and GdTe\(_3\) from the experimental data on the intralayer MR.

According to Eq. (90.5) of Ref. \cite{49} the intralayer conductivity at finite temperature is given by \cite{49}

\[
\sigma_{yy} = \int d\varepsilon \left[ -n_F'(\varepsilon) \right] \sigma_{yy}(\varepsilon), \tag{2}
\]

where the derivative of the Fermi distribution function \(n_F'(\varepsilon) = -1/\{4T \cosh^2[\varepsilon - \mu/2T]\}\), and the zero-temperature electron conductivity at energy \(\varepsilon\) is

\[
\sigma_{yy}(\varepsilon) = e^2 g(\varepsilon) D_y(\varepsilon). \tag{3}
\]

Here \(g(\varepsilon)\) is the DoS and \(D_y(\varepsilon)\) is the diffusion coefficient of electrons along \(y\)-axis. It is convenient to use the harmonic expansion for the oscillating DoS \(g(\varepsilon)\). Below we will need only the first terms in this harmonic series, which at finite \(t_\perp \sim \hbar \omega_c\) are given by \cite{39,51,52}

\[
g(\varepsilon) \approx g_0 \left[ 1 - 2 \cos \left( \frac{2\pi \varepsilon}{\hbar \omega_c} \right) J_0 \left( \frac{4\pi t_\perp}{\hbar \omega_c} R_D \right) \right], \tag{4}
\]

where \(g_0 = m^* / \pi \hbar^2 d\) is the DoS at the Fermi level in the absence of magnetic field per two spin components, \(J_0(x)\) is the Bessel’s function, the Dingle factor \cite{53,54} \(R_D = \exp[-\pi k/\omega_c \tau_0]\), \(\tau_0 = \hbar/2\Gamma_0\) is the electron mean free time without magnetic field, and \(\Gamma_0\) is the LL broadening.
The calculation of the diffusion coefficient $D_y(\varepsilon)$ is less trivial and requires to specify the model. At $\mu \gg \hbar \omega_c$ the quasi-classical approximation is applicable. In an ideal crystal in a magnetic field $B$ the electrons move along the cyclotron orbits with a fixed center and the Larmor radius $R_L = p_c/eB_z$. Without scattering the electron diffusion in the direction perpendicular to $B$ is absent. Scattering by impurities changes the electronic states and leads to the electron diffusion. For simplicity, we consider only short-range impurities, described by the $\delta$-function potential: $V_i(r) = U\delta^3(r-r_i)$. Scattering by impurities is elastic, i.e. it conserves the electron energy $\varepsilon$, but the quantum numbers of electron states may change. The matrix element of impurity scattering is given by

$$T_{mm'} = \Psi^*_m(r_i) U \Psi_m(r_i) ,$$

where $\Psi_m(r)$ is the electron wave function in the state $m$. During each scattering, the typical change $\Delta y = \Delta P_z c/eB_z$ of the mean electron coordinate $y_0$ perpendicular to $B$ is of the order of $R_L$, because for larger $\Delta y \gg R_L$ the matrix element in Eq. (5) is exponentially small because of small overlap of the electron wave functions $\Psi^*_m(r_i) \Psi_m(r_i) \sim \Psi^*_m(r_i+\Delta y) \Psi_m(r_i)$. The diffusion coefficient is approximately given by

$$D_y(\varepsilon) \approx \langle (\Delta y)^2 \rangle / 2\tau(\varepsilon) ,$$

where $\tau(\varepsilon)$ is the energy-dependent electron mean scattering time by impurities, and the angular brackets in Eq. (6) mean averaging over impurity scattering events. In the Born approximation, the mean scattering rate

$$1/\tau(\varepsilon) = 2\pi n_i U^2 g(\varepsilon) ,$$

where $n_i$ is the impurity concentration. This scattering rate has MQO, proportional to those of the DoS in Eq. (4). The MQO of $\langle (\Delta y)^2 \rangle \approx R^2_D$ are, usually, weaker and in 3D metals they are neglected [49]. Then

$$D_y(\varepsilon) \approx R^2_D/2\tau(\varepsilon) \propto g(\varepsilon) .$$

However, in Q2D metals, when $t_z \sim \hbar \omega_c$, the MQO of $\langle (\Delta y)^2 \rangle$ can be of the same order as MQO of the DoS. Then instead of Eq. (8) at $R_D \ll 1$ one has

$$D_y(\varepsilon) \approx D_0 \left[ 1 - 2\alpha \cos \left( \frac{2\pi \varepsilon}{\hbar \omega_c} \right) J_0 \left( \frac{4\pi t_z}{\hbar \omega_c} \right) R_D \right] ,$$

where $D_0 \approx R^2_D/2\tau_0$, and the number $\alpha \sim 1$. Substituting Eqs. (3), (1) and (9) to Eq. (2) after integration over $\varepsilon$ one obtains

$$\sigma_{yy}(B) \approx 1 + 2\alpha^2 J_0^2 (4\pi t_z/\hbar \omega_c) R^2_D - 2(\alpha + 1) \cos \left( \frac{2\pi \mu}{\hbar \omega_c} \right) J_0 \left( \frac{4\pi t_z}{\hbar \omega_c} \right) R_D R_T ,$$

where the temperature damping factor of MQO is

$$R_T = (2\pi^2 k_B T/\hbar \omega_c) / \sinh (2\pi^2 k_B T/\hbar \omega_c) .$$

The slow oscillations, described by the first line of Eq. (10), are not damped by temperature within our model. Approximately, one can use the asymptotic expansion of the Bessel function in Eq. (11) for large values of the argument: $J_0(x) \approx \sqrt{2/\pi x} \cos(x-\pi/4)$, $x \gg 1$. Then, after introducing the frequency of slow oscillations $F_{slow} = 4t_z B/\hbar \omega_c$, the first line in Eq. (10) simplifies to

$$\sigma_{yy}^{slow}(B) \approx 1 + \frac{\alpha \hbar \omega_c}{2\pi^2 t_z} \sin \left( \frac{2\pi F_{slow}}{B} \right) R^2_D .$$

In tilted magnetic field at constant $|B|$, $\omega_c \propto \cos \theta$ and $t_z$ changes according to Eq. (11). Then the frequency of slow oscillations must depend on tilt angle $\theta$ as:

$$F_{slow}(\theta) / F_{slow}(0) = J_0 \left( k_F d \tan \theta \right) / \cos(\theta) .$$

To clarify this effect, we experimentally study the angular dependence of observed slow oscillations frequency. The evolution of the slow oscillations in GdTe$_3$ with the change of the tilt angle $\theta$ of magnetic field at $T = 4.2$ K is shown in Fig. 3 where the derivative $dR/dB$ is plotted as function of the perpendicular-to-layers component of magnetic field $B_\perp = B \cos(\theta)$. Note, that magnetic field
rotation in the \((b-c)\) and \((b-a)\) planes demonstrated the same results for \(\text{TbTe}_3\).

In Fig. 4 we show the \(\theta\)-dependence of the frequency of slow oscillations at \(T = 4.2\) K for \(\text{TbTe}_3\) (a) and for \(\text{GdTe}_3\) (c). The solid curves give the cosine dependence \(F(\theta) = F(0)/\cos(\theta)\) typical for MQO. According to Eq. 13, the angular dependence of the frequency \(F_{\text{slow}}(\theta)\) of slow oscillations differs from this standard cosine dependence, especially at high tilt angle. In Fig. 4 (b) we plot the angular dependence of the product \(F_{\text{slow}}(\theta)\cos(\theta)\) in \(\text{TbTe}_3\). If the origin of slow oscillations is due to small FS pockets, the product \(F_{\text{slow}}(\theta)\cos(\theta)\) would be independent of the tilt angle \(\theta\). The experimental data, shown by black filled squares, clearly indicate the deviation from the horizontal line. These experimental data can be reasonably fitted by Eq. 13 at \(k_Fd = 0.1\), shown by solid red lines in Figs. 4 (b,d).

From the angular dependence of the frequency \(F_{\text{slow}}(\theta)\) of slow oscillations, shown in Fig. 4 we conclude that these oscillations originate not from small FS pockets as usual SdH oscillations, but from the entanglement of close frequencies due to a finite \(t_z\), similar to that observed in the interlayer MR in the organic metal \(\beta-(\text{BEDT-TTF})_2\text{IBr}_2\) [34]. The strong additional argument in favor of this origin of the observed slow oscillations is the very weak temperature dependence of their amplitude. To our knowledge, the data obtained are the first observation of such slow oscillations in the intralayer magnetotransport.

The frequency of slow oscillations at \(\theta = 0\) can be used to estimate the value of \(t_z\). According to Eq. 12, with the effective electron mass \(m^* \approx 0.1m_e\) determined from the temperature dependence of the amplitude of SdH oscillations [54], and \(F_{\text{slow}} \approx 3.5T\) (see Fig. 4 (b,d)), we obtain \(t_z \approx 1\text{meV}\). The small values of \(t_z\) compared to the transfer integrals \(t_\parallel \approx 2\text{eV}\) along the chains and \(t_\perp \approx 0.37\text{eV}\) perpendicular to the chains, as obtained by the band structure calculations [42], illustrate the quasi-2D character of these rare-earth tritellurides and justify that the dispersion along \(b\)-axis is neglected in ARPES measurements. The value of \(t_z\) is very important for quantum corrections to conductivity [57, 58] and for other physical properties of strongly anisotropic compounds.

The angular dependence of the frequency \(F_{\text{slow}}(\theta)\) of slow oscillations allows also to determine the value of the Fermi momentum of the open FS pockets. Fitting experimental data on \(F_{\text{slow}}(\theta)\) shown in Fig. 4 to Eq. 13 gives \(k_Fd \approx 0.11\) for \(\text{GdTe}_3\) and \(k_Fd \approx 0.12\) for \(\text{TbTe}_3\). With \(d \approx 25\text{Å}\) this gives \(k_F \approx 4.5 \cdot 10^7\text{cm}^{-1}\). Such a small value of \(k_F\) means that the FS reconstruction due to CDW is very strong and leaves only very small ungapped FS pockets. These FS pockets are most probably elongated and directed differently, which gives different \(F_{\text{slow}}(\theta)\) dependence. Their total contribution to the slow oscillations, being a sum of the contributions from the individual FS pockets, has a smeared angular dependence of the frequency of slow oscillations \(F_{\text{slow}}(\theta)\) as compared to the case of only one elliptical FS pocket observed in \(\beta-(\text{BEDT-TTF})_2\text{IBr}_2\) [34].

To summarize, we report the first observation and qualitative theoretical description of the slow oscillations of intralayer magnetoresistance in quasi-2D metallic compounds. These slow oscillations allow to measure the interlayer transfer integral \(t_z\), which is hard to measure by any other ways. We obtain the value \(t_z \approx 1\text{meV}\) in the rare-earth tritelluride compounds \(\text{TbTe}_3\) and \(\text{GdTe}_3\).
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