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Slow Oscillations of In-plane Magnetoresistance in Strongly Anisotropic Quasi-Two-Dimensional Rare-Earth Tritellurides

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Abstract The slow oscillations of intralayer magnetoresistance in the quasi-2D metallic compounds $TbTe_3$ and $GdTe_3$ have been observed for the first time. These oscillations do not originate from small Fermi-surface pockets, as revealed usually by Shubnikov–de Haas oscillations, but from the entanglement of close frequencies due to a finite interlayer transfer integral t_z, either between the two Te planes forming a bilayer or between two adjacent bilayers, which allows to estimate its values. For $TbTe_3$ and $GdTe_3$, we obtain the estimate $t_z \approx 1$ meV.

Keywords Electronic conductivity · Magnetoresistance · Layered conductors

The Fermi surface (FS) of quasi-two-dimensional (Q2D) metals is a cylinder with weak warping $\sim 4t_z/v_F$, where v_F is the in-plane Fermi velocity [1]. If the interlayer transfer integral $t_z \gg \hbar\omega_c$, the standard 3D theory of galvanomagnetic properties of metals [1–3] based on the kinetic equation in the τ-approximation can be applied. This theory predicts several peculiarities of magnetoresistance (MR) in Q2D metals: the angular magnetoresistance oscillations (AMRO) [4–6] and the beats of the amplitude

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of magnetic quantum oscillations (MQO) [2] that give the possibility to extract the fine details of the FS.

When $t_z \sim \hbar\omega_c$, new qualitative features appear both in the monotonic [7–10] and oscillating parts of MR. The oscillating part of interlayer magnetoresistance at $\mu \gg t_z \geq \hbar\omega_c$, where $E_F = \mu$ is the in-plane Fermi energy, acquires the slow MR oscillations [11,12], and the phase shift of MQO beats between transport and thermodynamic quantities [12,13]. These two effects are missed by the standard 3D theory [1–3] because they appear in the higher orders in the parameter $\hbar\omega_c/t_z$.

The slow oscillations (SlO) qualitatively originate from the product of the oscillations with two close frequencies $F_0 \pm \Delta F$, which gives the oscillations with frequency $2\Delta F$. The SlO have many interesting and useful features as compared to the fast quantum oscillations [11]. 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. Third, the SlO allow to measure the interlayer transfer integral t_z. These features make the SlO to be a useful tool to study the electronic properties of Q2D metals. Until now, the SlO 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 [11]. At the same time, most of the 2D compounds as a rule have a shape of very thin flakes in which correct measurements of the interlayer conductivity are very difficult. The question we would like to answer in the present study is the following: is it possible to observe the SlO in the in-plain electrical transport, namely in transverse intralayer MR $R_{xx}(\mathbf{B})$?

In the present work, we study the possibility and usefulness of SlO in the *intralayer* electrical transport, choosing the non-organic layered Q2D rare-earth tritelluride compounds $R\mathrm{Te}_3$ (R = Y, La, Ce, Nd, Sm, Gd, Tb, Ho, Dy, Er, and Tm). These compounds well illustrate our goal: in addition to good metallic properties, their available single crystals have a very flat shape, allowing correct measurements of the intralayer conductivity [14].

Rare-earth tritellurides have an orthorhombic structure ($Cmcm$) in the normal state and exhibit a c-axis incommensurate charge-density wave (CDW) at high temperature, which was recently a subject of intense studies [14–20]. For the heaviest rare-earth elements, a second a-axis CDW occurs at low temperature. In addition to hosting incommensurate CDWs, magnetic rare-earth ions exhibit closed-spaced magnetic phase transitions below 10 K [21,22] leading to coexistence and competition of many ordered states at low temperatures. Therefore, any information about the FS on such small energy scale beyond the ARPES resolution is very important. An accurate measurement of t_z as function of temperature, provided by SlO, is also useful in these compounds. For the possible observation of SlO, the rare-earth tritellurides are very promising, because they have the appropriate anisotropy and good metallic conductivity up to low temperatures.

Single crystals of $GdTe_3$ and $TbTe_3$, with a high-T Peierls transition temperature 380 and 336 K correspondingly, were grown by a self-flux technique under purified argon atmosphere as described previously [17]. Thin-single crystal samples with a thickness typically 0.1–0.3 μm were prepared by micromechanical exfoliation of relatively thick crystals glued on a sapphire substrate. The quality of selected crystals, namely absence of twinning, was controlled by X-ray diffraction.

From high-quality $[R(300\,\mathrm{K})/R(10\,\mathrm{K}) > 100]$ untwinned single crystals, we cut bridges with a length 200–500 μm and a width 50–80 μm in well defined, namely [100] and [001] orientations along the conducting a and c axes [20]. 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 θ between the field direction and the normal b -axis to the highly conducting (a, c) plane.

In Fig. 1, we plot the derivative of MR $\mathrm{d}R/\mathrm{d}B$ as a function of magnetic field applied along b-axis for $TbTe_3$ (a) and $GdTe_3$ (d) with the current applied in the (a, c) plane up to $B = 8.2\,\mathrm{T}$ at $T = 4.2\,\mathrm{K}$. For both compounds, oscillations with a very weak amplitude are detectable. At $B > 2$ T, pronounced Shubnikov–de Haas (SdH) oscillations with a frequency $F \approx 55$–$58\,\mathrm{T}$ are observed in $\mathrm{d}R/\mathrm{d}B$ for $GdTe_3$. The temperature dependence of the amplitude of these oscillations gives the effective electron mass $m^* \approx 0.1m_\mathrm{e}$. At high field ($B > 7$ T), new oscillations with high frequency ($F \approx 0.7$–0.8 kT) appear in $TbTe_3$, indicating the existence of several types of pockets on the partially gapped FS. de Haas–van Alphen oscillations were previously observed [23] from a.c. susceptibility and torque measurements in $LaTe_3$ with three distinct frequencies $\alpha \sim 50\,\mathrm{T}$, $\beta \approx 520\,\mathrm{T}$, and $\gamma \sim 1600\,\mathrm{T}$. The β frequency was attributed to small FS pockets around the X point in the Brillouin zone, unaffected by the CDW, while the α frequency was assigned to a portion of the reconstructed FS. We can attribute the observed frequency $F \approx 56$ T of SdH oscillations above 2 T in $GdTe_3$ and $TbTe_3$ similarly to the α frequency in $LaTe_3$ [23].

In addition to rapid SdH oscillations, at low magnetic field ($B < 2$ T), the magnetoresistance exhibits prominent slow oscillations with very low frequency $F_\mathrm{slow} < 4$ T. To demonstrate the periodicity of these oscillations in B^{-1}, we plot the derivative $\mathrm{d}R/\mathrm{d}B$ as a function of the inverse magnetic field in Fig. 1 for $TbTe_3$ (b) and for $GdTe_3$ (e). In contrast to the usual SdH oscillations, the amplitude of which decreases rapidly as temperature increases, the slow oscillations are observable up to $T \approx 40$ K, as can be seen from Fig. 1c, f where we show the temperature evolution of slow oscillations for $TbTe_3$ and $GdTe_3$. The fitting of the temperature dependence of the SlO amplitude by standard Lifshits–Kosevich formula [24] gives the effective mass $m^* = 0.004m_\mathrm{e}$, which is unreasonably small. This suggests that the observed slow oscillations originate not from small FS pockets, but from the FS warping due to the interlayer transfer integral t_z, similarly to the slow oscillations of interlayer magnetoresistance in the organic superconductor β-$(BEDT\text{-}TTF)_2IBr_2$ [11]. If so, the observed slow oscillations give an excellent opportunity to find the value of interlayer transfer integral t_z and of the product $k_\mathrm{F}d$ at low temperature in tritellurides $TbTe_3$ and $GdTe_3$. Let us show that such oscillations can be observed in the in-plane magnetoresistance.

The intralayer electron conductivity at finite temperature is given by [25,26]

$$\sigma_{xx} = \int d\varepsilon \left[-n'_\mathrm{F}(\varepsilon)\right] \sigma_{xx}(\varepsilon), \tag{1}$$

where $n'_\mathrm{F}(\varepsilon)$ is the derivative of the Fermi distribution function, and the zero-temperature conductivity at energy ε is

$$\sigma_{xx}(\varepsilon) = e^2 g\,(\varepsilon)\, D_x\,(\varepsilon)\,. \tag{2}$$

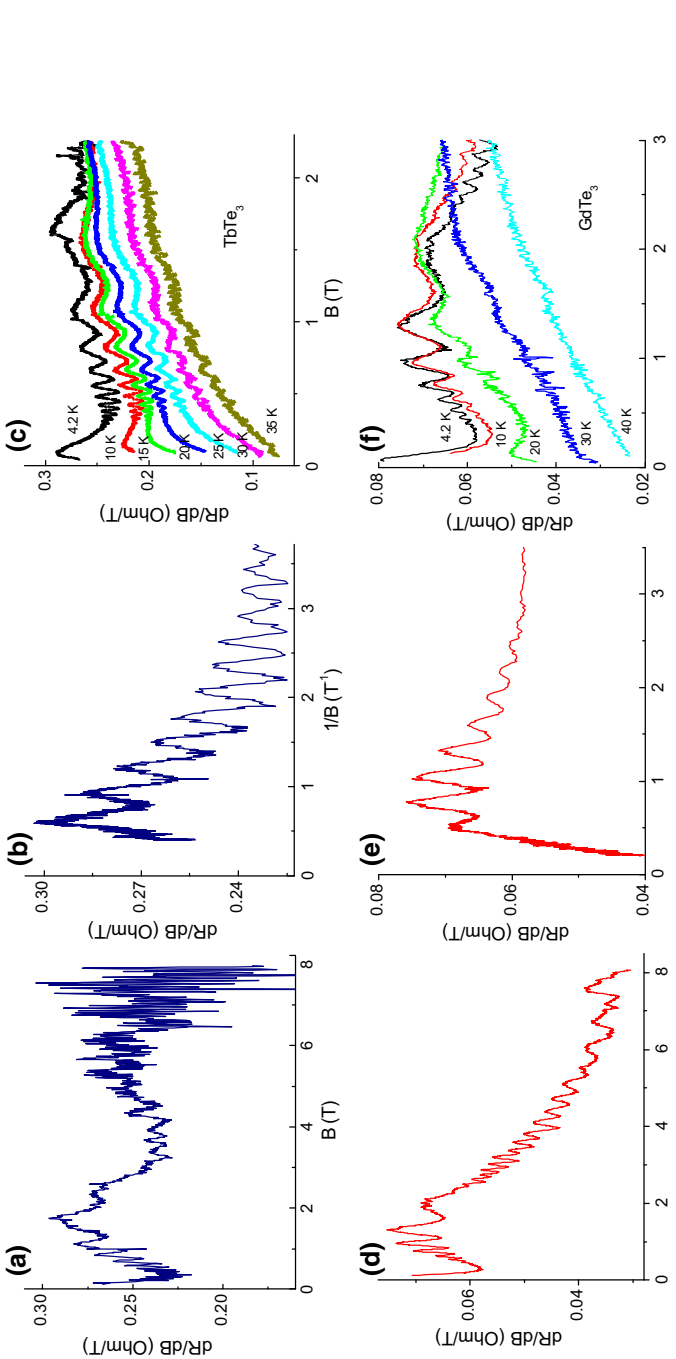

Fig. 1 $\mathrm{d}R(B)/dB$ dependencies at $T = 4.2$ K for $TbTe_3$ **a** and $GdTe_3$ **d** demonstrating clear rapid oscillations which appear in the field range $B > 2$ T. **b** and **e** $\mathrm{d}R/\mathrm{d}B$ as a function of inverse magnetic field, B^{-1}, at low field region demonstrating slow oscillations in $TbTe_3$ and $GdTe_3$ correspondingly. **c** and **f** temperature evolution of slow oscillations for $TbTe_3$ and $GdTe_3$ (Color figure online)

Here $g(\varepsilon)$ is the density of states (DoS) and $D_x(\varepsilon)$ is the diffusion coefficient of electrons along x-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 and momentum-independent interlayer hopping $t_z \sim \hbar\omega_c$ are given by [12,27–30]

$$g(\varepsilon) \approx g_0 \left[1 - 2\cos\left(\frac{2\pi\varepsilon}{\hbar\omega_c}\right) J_0\left(\frac{4\pi t_z}{\hbar\omega_c}\right) R_D\right], \tag{3}$$

where $g_0 = m^*/\pi\hbar^2 d$ is the DoS at the Fermi level per two spin components without magnetic field, $J_0(x)$ is the Bessel's function, the Dingle factor [2,31–33] $R_D \approx \exp[-\pi k/\omega_c\tau_0]$, and τ_0 is the electron mean free time without magnetic field.

Without scattering, the electron diffusion in the direction perpendicular to **B** is absent. Scattering by impurities leads to the electron diffusion, because it changes the electronic states. For simplicity, we consider only short-range impurities, described by the δ-function potential: $V_i(r) = U\delta^3(r - r_i)$. Scattering by impurities is elastic, i.e., it conserves the electron energy ε, but changes the electron quantum numbers. At $\mu \gg t_z$, the change of the Landau level (LL) number Δn does not exceed $4t_z/\hbar\omega_c$, because otherwise the electron energy cannot be conserved. 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 m-th state. During each scattering, the typical change $\Delta x = -\Delta P_y c/eB_z$ of the mean electron coordinate x_0 perpendicular to **B** is of the order of Larmor radius $R_L = p_F c/eB_z$ [34]. The diffusion coefficient is then

$$D_x(\varepsilon) \approx \left\langle (\Delta x)^2 \right\rangle / 2\tau(\varepsilon), \tag{4}$$

where $\tau(\varepsilon)$ is the energy-dependent electron mean scattering time by impurities, and the angular brackets in Eq. (4) are 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. (3). The MQO of $\left\langle (\Delta x)^2 \right\rangle \approx R_L^2$ are, usually, weaker and in 3D metals they are neglected [25]. Then $D_x(\varepsilon) \approx R_L^2/2\tau(\varepsilon) \propto g(\varepsilon)$. However, in Q2D metals, when $t_z \sim \hbar\omega_c$, the MQO of $\left\langle (\Delta x)^2 \right\rangle$ can be of the same order as MQO of the DoS, and at $R_D \ll 1$ one has

$$D_x(\varepsilon) \approx D_0 \left[1 - 2\alpha\cos\left(\frac{2\pi\varepsilon}{\hbar\omega_c}\right) J_0\left(\frac{4\pi t_z}{\hbar\omega_c}\right) R_D\right], \tag{5}$$

where $D_0 \approx R_L^2/2\tau_0$, and the number $\alpha \sim 1$.

Substituting Eqs. (2), (3), and (5) to Eq. (1) and performing the integration over ε one obtains

$$\frac{\sigma_{xx}(B)}{e^2 g_0 D_0} \approx 1 + 2\alpha J_0^2(4\pi t_z/\hbar\omega_c) R_D^2 - 2(\alpha + 1)\cos\left(\frac{2\pi\mu}{\hbar\omega_c}\right) J_0\left(\frac{4\pi t_z}{\hbar\omega_c}\right) R_D R_T, \tag{6}$$

where $R_T = \left(2\pi^2 k_B T/\hbar\omega_c\right) / \sinh\left(2\pi^2 k_B T/\hbar\omega_c\right)$ is the temperature damping factor of MQO. The slow oscillations, described by the first line of Eq. (6), are not

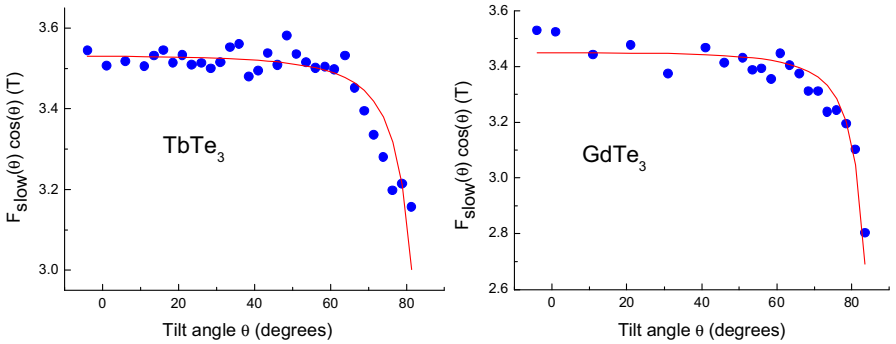

Fig. 2 Angular dependence of the frequency $F_{\rm slow}(\theta)$ of slow oscillations of intralayer magnetoresistance in $TbTe_3$ and $GdTe_3$, multiplied by $\cos(\theta)$. The experimental data are shown by *black filled squares*, and the theoretical prediction according to Eq. (8) at $k_{\rm F}d = 0.12$ is shown by *solid red lines* (Color figure online)

damped by temperature within our model. Approximately, one can use the asymptotic expansion of the Bessel function in Eq. (6) 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 $F_{\rm slow} = 4t_{\rm z}B/\hbar\omega_{\rm c}$, the first line in Eq. (6) simplifies to

$$\frac{\sigma_{\rm xx}^{\rm slow}(B)}{e^2 g_0 D_0} \approx 1 + \frac{\alpha\hbar\omega_{\rm c}}{2\pi^2 t_{\rm z}} \sin\left(\frac{2\pi F_{\rm slow}}{B}\right) R_{\rm D}^2. \tag{7}$$

In a tilted magnetic field **B** at angle θ with respect to the normal to conducting layers and at constant $|\mathbf{B}|$, $\omega_{\rm c} \propto \cos\theta$. For the cylindrical FS with straight (momentum-independent) warping along k_z and interlayer distance d, the frequency of slow oscillations depends on tilt angle θ as [5,11]:

$$F_{\rm slow}(\theta)/F_{\rm slow}(0) = J_0(k_{\rm F}d\tan\theta)/\cos(\theta). \tag{8}$$

To clarify this effect, we experimentally study the angular dependence of the observed slow oscillations frequency. In Fig. 2, we plot the angular dependence of the product $F_{\rm slow}(\theta)\cos(\theta)$ in $TbTe_3$ (a) and $GdTe_3$ (b). 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. (8) at $k_{\rm F}d = 0.12$ and are shown by solid red lines in Fig. 2b, d.

According to Eq. (7), with the effective electron mass $m^* \approx 0.1m_{\rm e}$ and SlO frequency $F_{\rm slow} \approx 3.5$T, we obtain $t_{\rm z} \approx 1$ meV. The effective electron mass was determined from the temperature dependence of the amplitude of SdH oscillations in $GdTe_3$ at $B = 4.5$ T and is in a agreement with previously reported measurements for $LaTe_3$ [23]. The small values of t_z compared to the transfer integrals $t_{\parallel} \approx 2$ eV along the chains and $t_{\perp} \approx 0.37$ eV perpendicular to the chains in the conducting (a, c) plane, as obtained by the band structure calculations [16], illustrate the quasi-2D character of these rare-earth tritellurides and justify that the dispersion along b-axis is neglected in ARPES measurements.

The angular dependence of the frequency $F_{\rm slow}(\theta)$ of SlO allows also to determine the value of the Fermi momentum of the open FS pockets. Fitting the experimental data of $F_{\rm slow}(\theta)$ shown in Fig. 2 to Eq. (8) gives $k_{\rm F}d \approx 0.11$ for $GdTe_3$ and $k_{\rm F}d \approx 0.12$ for $TbTe_3$. There are two possible origins of the observed SlO in RTe_3: the bilayer splitting t_b and the inter-bilayer coupling t_z. The first double splits the Fermi energy, while the latter leads to the k_z energy dispersion and to the FS warping. Correspondingly, there are two interlayer distances: $d^\star \approx 3.64$ Å and $c^\star/2 = h + d^\star \approx 12.9$ Å. With $d = d^\star = 3.64$ Å we obtain $k_{\rm F} \approx 3.3 \cdot 10^6 {\rm cm}^{-1}$, and with $d = c^\star/2 = h + d^\star \approx 12.9$ Å we obtain $k_{\rm F} \approx 9.3 \cdot 10^5 {\rm cm}^{-1}$. The FS reconstruction by two CDWs leads to the formation of various FS pockets of different size. If one assumes that these small FS pockets are not elongated but almost circular, the corresponding FS cross section areas are $S_{\rm ext} \approx \pi k_{\rm F}^2$. For the obtained value $k_{\rm F} \approx 3.3 \cdot 10^6 cm^{-1}$ for bilayer splitting ($d = d^\star$), this gives the MQO frequency $F_0 = S_{\rm ext}\hbar c/2\pi e \approx 36T$, a value close to the frequency 55–58 T of oscillations we have measured. The difference between the calculated 36 T and the experimental value 55–58 T can be accounted by considering the elongation of the FS pockets or another non-circular shape. On the other hand, taking $d = h + d^\star$ and $k_{\rm F} \approx 9.3 \times 10^5 {\rm cm}^{-1}$ gives only $F_0 \approx 3T$.

The observed frequency of the SlO does not depend on temperature, which is another argument in favor of their proposed origin as due to the interlayer hopping rather than due to small ungapped FS pockets. Indeed, if the observed SlO originated from small ungapped FS pockets, their frequency would strongly depend on temperature on the scale of the CDW transition temperature, because the size of the ungapped FS pockets depends on the temperature-dependent CDW energy gap. For $TbTe_3$, the second CDW transition temperature is $T_{c2} = 41$ K [35], but we do not observe any change in the frequency of SlO up to 35 K which is inconsistent with the small FS-pocket origin of SlO. On the contrary, the interlayer transfer integral t_z is not sensitive to the in-plane electronic phase transitions. In particular, it does not depend on the in-plane Fermi-surface reconstruction. The interlayer transfer integrals t_z and t_b are determined mainly by the strong (1 eV) crystalline potential in the interlayer direction, which is not affected by the CDW or other in-plane electronic orderings.

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 $t_z \approx 1$ meV in the rare-earth tritelluride compounds $TbTe_3$ and $GdTe_3$. The value of t_z is very important for quantum corrections to conductivity [36,37] and for other physical properties of strongly anisotropic compounds.

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