

Linear magnetoresistance in the charge density wave state of quasi-two-dimensional rare-earth tritellurides

A. A. Sinchenko,^{1,2,3} P. D. Grigoriev,^{4,5,6} P. Lejay,⁷ and P. Monceau⁷

¹*Kotel'nikov Institute of Radioengineering and Electronics of RAS, Mokhovaya 11-7, 125009 Moscow, Russia*

²*National Research Nuclear University (MEPhI), 115409 Moscow, Russia*

³*M.V. Lomonosov Moscow State University, 119991 Moscow, Russia*

⁴*L.D. Landau Institute for Theoretical Physics, 142432 Chernogolovka, Russia*

⁵*P.N. Lebedev Physical Institute, RAS, 119991 Moscow, Russia*

⁶*National University of Science and Technology "MISIS", 119049 Moscow, Russia*

⁷*Université Grenoble Alpes, CNRS, Grenoble INP, Institut NEEL, 38042 Grenoble, France*

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We report measurements of the magnetoresistance in the charge density wave (CDW) state of rare-earth tritellurides, namely TbTe₃ and HoTe₃. The magnetic field dependence of magnetoresistance exhibits a temperature dependent crossover between a conventional quadratic law at high T and low B and an unusual linear dependence at low T and high B . We present a quite general model to explain the linear magnetoresistance taking into account the strong scattering of quasiparticles on CDW fluctuations in the vicinity of “hot spots” of the Fermi surface (FS) where the FS reconstruction is the strongest.

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I. INTRODUCTION

Interaction between pairs of quasiparticles often leads to broken-symmetry ground states in solids. Typical examples are the formation of Cooper pairs in superconductors, or charge (CDW) and spin (SDW) density waves driven by electron-phonon and electron-electron interactions, respectively [1,2]. A CDW ground state is characterized by a spatial modulation $\sim \cos(Qx + \varphi)$ of the electron density and a periodic lattice distortion with the same $Q_{\text{CDW}} = 2k_F$ wave vector inducing opening of a gap, Δ , in the electron spectrum. From one-dimensional (1D) weak coupling mean-field theories, with $\Delta/E_F \ll 1$, the Peierls instability is driven by the electronic energy gain which originates mostly from the Fermi surface (FS) nesting with $Q = 2k_F$.

In the case of not complete nesting in quasi-one-dimensional (Q1D) compounds or in the case of quasi-two-dimensional (Q2D) or three-dimensional (3D) conductors the ground state in the CDW state is semimetallic because electron and hole pockets remain in the FS. Properties of these carriers can be modified by the CDW ordering. One of the methods to understand this possible modification is to study magnetoresistance.

In conventional metals, the Lorentz force caused by an applied magnetic field changes the electron trajectory and gives rise to a positive magnetoresistance (MR) which increases quadratically with the strength of the field [3–5]. Only in a few cases the MR may grow linearly with the field (LMR). For the first time such type of behavior was observed by Kapitza [6] in polycrystalline metals. It was shown that LMR is attributed to the presence of open Fermi surfaces. The quantum mechanism of LMR was proposed by Abrikosov [7,8]. In his model LMR is realized basically in gapless semiconductors or semimetals with a linear energy spectrum and with a very small carrier concentration, so that only one Landau band participates in the conductivity. Parish and Littlewood [9] considered a macroscopically disordered and strongly inhomogeneous semiconductor and showed that a classical mechanism will give LMR in this case. In Ref. [10] it was shown that LMR

may occur in weakly inhomogeneous systems, for fields where the cyclotron orbit period exceeds the scattering time.

From many published works one can also conclude that this unusual LMR may be a common feature of CDW systems. Indeed, LMR was observed in Q1D compounds exhibiting a CDW with incomplete FS nesting such as NbSe₃ [11,12] and in PdTeI [13]. The effect of LMR was reported for Q2D compounds with a CDW: transition metal dichalcogenides 2H-NbSe₂; 2H-TaSe₂ [14]; 1T-TaTe₂ [15], 1T-NbTe₂ [16], monophosphate tungsten bronzes (PO₂)₄(WO₃)_{2m} for $m = 4.6$ [17]; molybdenum purple bronze K_{0.9}Mo₄O₁₁ and molybdenum oxides η -Mo₄O₁₁ [18]. In the present work we have studied galvanomagnetic properties in another type of Q2D compound with a CDW, namely, rare-earth tritellurides. We have measured magnetoresistance in the temperature range across the Peierls transition temperature and show that effectively LMR appears below this temperature.

Rare-earth tritellurides $R\text{Te}_3$ ($R = \text{Y, La, Ce, Nd, Sm, Gd, Tb, Ho, Dy, Er, Tm}$) exhibit an incommensurate CDW through the whole R series with a wave vector $\mathbf{Q}_{\text{CDW1}} = (0, 0, \sim 2/7c^*)$ with a Peierls transition temperature above 300 K for the light atoms (La, Ce, Nd). For the heavier R (Dy, Ho, Er, Tm) a second CDW occurs with the wave vector $\mathbf{Q}_{\text{CDW2}} = (\sim 2/7a^*, 0, 0)$. The superlattice peaks measured from x-ray diffraction are very sharp and indicate a long range 3D CDW order [19–21].

Below the Peierls transition, in all $R\text{Te}_3$ compounds, the Fermi surface is partially gapped resulting in a metallic behavior at low temperature. The layered $R\text{Te}_3$ compounds exhibit a large anisotropy between the resistivity along the b axis and that in the (a, c) plane, typically $\sim 10^2$ below T_{CDW1} and much higher at low temperature [22]. Because of the unidirectional character of the upper CDW [23–25], a conductivity anisotropy in the (a, c) plane arises in the CDW state as was observed experimentally and explained theoretically in Ref. [26]. The effect of the upper CDW on the in-plane resistivity observed in experiments is very weak, no more than a few percent of the total resistance [19,22,26].

For our study we chose two compounds: TbTe_3 as a system with unidirectional CDW and HoTe_3 exhibiting a bidirectional CDW. In TbTe_3 the CDW ordering is observed well above room temperature ($T_{\text{CDW1}} = 336$ K). In HoTe_3 the first and the second CDW transitions take place at $T_{\text{CDW1}} = 283$ K and $T_{\text{CDW2}} = 110$ K correspondingly [19].

II. EXPERIMENT

Single crystals of TbTe_3 and HoTe_3 were grown by a self-flux technique under purified argon atmosphere as described previously [27]. Thin single-crystal samples with a square shape and with a thickness less than $1 \mu\text{m}$ were prepared by micromechanical exfoliation of relatively thick crystals glued on a sapphire substrate. The untwinned character of selected crystals and the spatial arrangement of crystallographic axes were controlled by x-ray diffraction. Room temperature resistivity of crystals was $26\text{--}28 \mu\Omega \text{cm}$ for TbTe_3 and $12\text{--}13 \mu\Omega \text{cm}$ for HoTe_3 that is in accordance with previously reported results [19,26]. The quality of crystals was confirmed by high value of resistance residual ratio (RRR), $R(300 \text{ K})/R(4 \text{ K})$: $70\text{--}90$ for HoTe_3 and more than 100 for TbTe_3 .

The magnetic field was applied parallel to the b axis, and in-plane magnetoresistance was recorded using the van der Pauw method, sweeping the field between $+6.5$ and -6.5 T. Measurements were performed at fixed temperature in the temperature range $350\text{--}20$ K with the step $\Delta T = 10$ K.

III. EXPERIMENTAL RESULTS

The temperature variation of the field dependence of magnetoresistance, defined as $\text{MR} = [R_{xx}(B) - R_{xx}(0)]/R_{xx}(0)$, in the temperature range from 10 K up to the temperature well above T_{CDW} is shown in Fig. 1 for HoTe_3 (a) and for TbTe_3 (b) in a log-log plot. Both compounds demonstrate nearly the same behavior: magnetoresistance changes by more than four orders of magnitude as temperature T decreases from 300 K to 20 K. Simultaneously, the power-law field dependence of MR changes monotonically from quadratic (red straight line segment) at high T and at low B to linear (blue straight line segment) at low T and high B . Note that in the studied magnetic field range (up to 6.5 T) we never observed any deviation of MR from quadratic law at temperatures above the Peierls transition temperatures. The examples of $\text{MR}(B)$ dependencies measured at T above T_{CDW} (330 K and 290 K for TbTe_3 and HoTe_3 , respectively) and below T_{CDW} (40 K for both compounds) are shown in Fig. 2. Note that, at the same temperature $T = 40$ K, the linear $R_{xx}(B)$ is more pronounced for HoTe_3 in which two CDWs exist at this temperature.

To make this quadratic-to-linear MR crossover clearer, in Figs. 3(a) and 3(b) we plot MR as a function of square magnetic field, B^2 for HoTe_3 and for TbTe_3 correspondingly. Solid black lines are quadratic dependencies which coincide with the experimental curves at low magnetic fields. At a certain magnetic field, B^* , experimental dependencies deviate from these lines. Temperature dependence of this characteristic field B^* is shown in Fig. 4. As can be seen, B^* increases rapidly or even diverges when T approaches the CDW transition temperature.

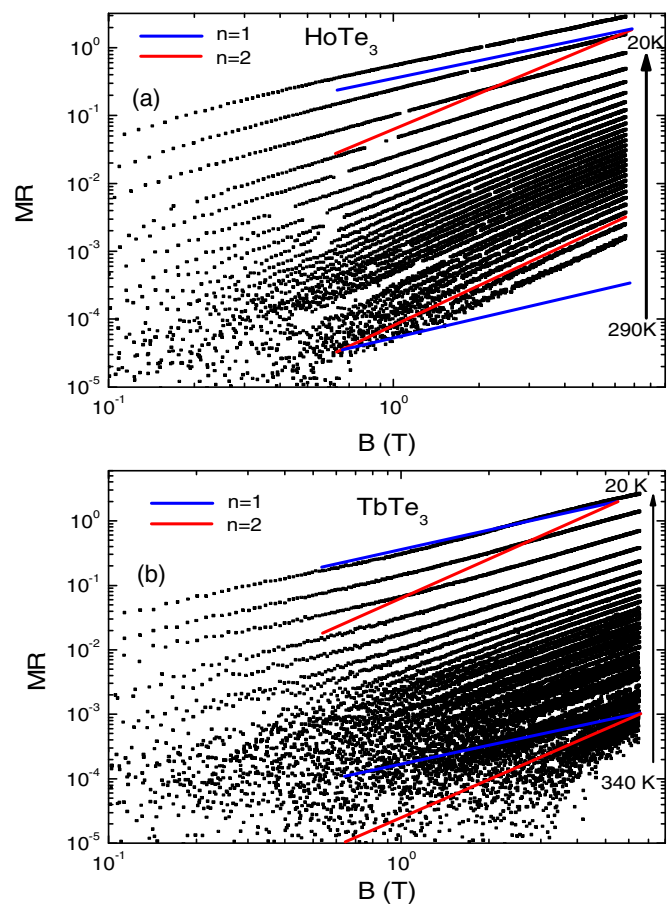


FIG. 1. Magnetoresistance of HoTe_3 (a) and TbTe_3 (b) as a function of magnetic field, B , in log-log scale at different temperatures. Blue and red straight line segments indicate linear and square dependencies correspondingly.

IV. THEORETICAL MODEL AND DISCUSSION

Thus most charge density wave systems with imperfect nesting exhibit a linear magnetoresistance that is, probably, related to the CDW electronic structure. To propose a possible explanation of this linear MR we invoke a usually neglected scattering mechanism of quasiparticles on the fluctuations of the order parameter of a charge density wave, which violate the space uniformity and lead to the momentum relaxation of quasiparticles. The scattering on CDW fluctuations is the strongest near the so-called “hot spots” on the Fermi surface (FS). Somewhat similar mechanism of linear MR but above the CDW transition temperature was proposed in Refs. [28,29]. In Ref. [28] the scattering in the hot spots, with large momentum and low energy transfer, involves umklapp processes. In Ref. [29] it involves the scattering by soft phonons, appearing due to the proximity to Peierls instability. In our case these hot spots are the FS areas where the FS reconstruction due to CDW is the strongest. Usually the hot spots are the ends of the ungapped FS parts. The electron dispersion in such hot spots depends strongly on the CDW structure, and the electrons in such hot spots may be easily scattered by CDW fluctuations. Thus, in cuprate high- T_c superconductors such hot spots are the ends of Fermi

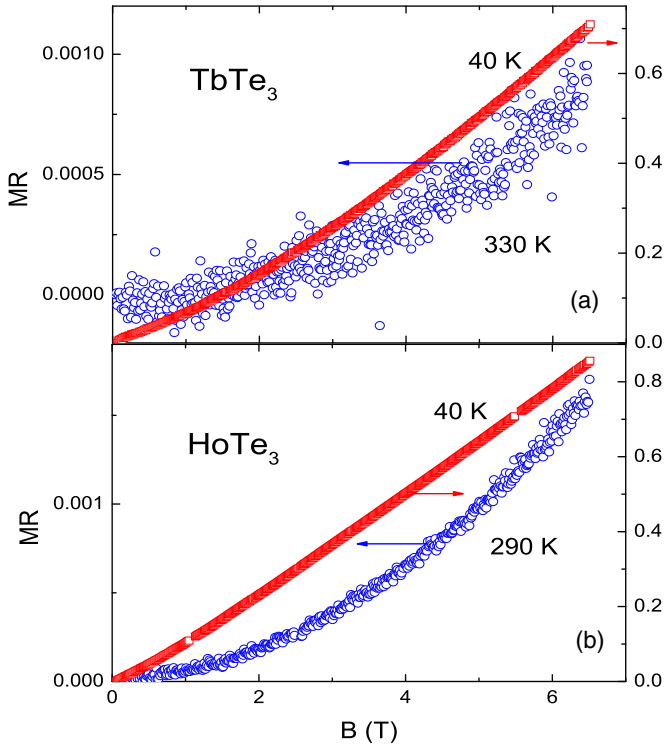


FIG. 2. MR(B) for TbTe₃ (a) and HoTe₃ (b), at temperatures above T_{CDW} (open blue circles) and below T_{CDW} (open red squares).

arcs, but the FS reconstruction is driven by the pseudogap or antiferromagnetic ordering, rather than by CDW. In organic metals, e.g., α -(BEDT-TTF)₂KHg(SCN)₄, where the CDW leads to the FS reconstruction and changes its topology [30], such hot spots are the points of intersection of the original FS and the FS shifted by the CDW wave vector. In these hot spots the electron dispersion changes strongly, somewhat similar to the change of electron dispersion at the boundaries of the Brillouin zone in the weak-coupling approximation [3], where the energy gap is formed due to periodic potential, which is the CDW in our case. Since the periodic potential and the formed energy gap in the electron spectrum is of the order of CDW order parameter and much less than the Fermi energy, in high magnetic field the electron trajectories in these hot spots are subject to magnetic breakdown in addition to the direct scattering by CDW fluctuations. This leads to an additional indirect scattering mechanism of conducting electrons, which may be rather strong [30].

The electron scattering in the hot spots leads to the linear field dependence of the scattering rate and, hence, to the linear magnetoresistance. To show this linear field dependence of the electron mean-free time τ we assume that in each hot spot an electron is scattered with some probability $w_{hs} < 1$; the possible origin of this scattering is discussed later. If this hot-spot scattering is the main scattering mechanism of conducting electrons, the corresponding electron mean-free time $\tau_{hs} = t_{hs}/w_{hs}$, where t_{hs} is the mean time of electron motion between these hot spots. This time t_{hs} is determined by the FS details [3]. In magnetic field \mathbf{H} electrons move in momentum space along the Fermi surface due to the Lorentz force, $dp/dt = (e/c)[\mathbf{v}_{\perp} \times \mathbf{H}]$, and periodically pass through such hot spots.

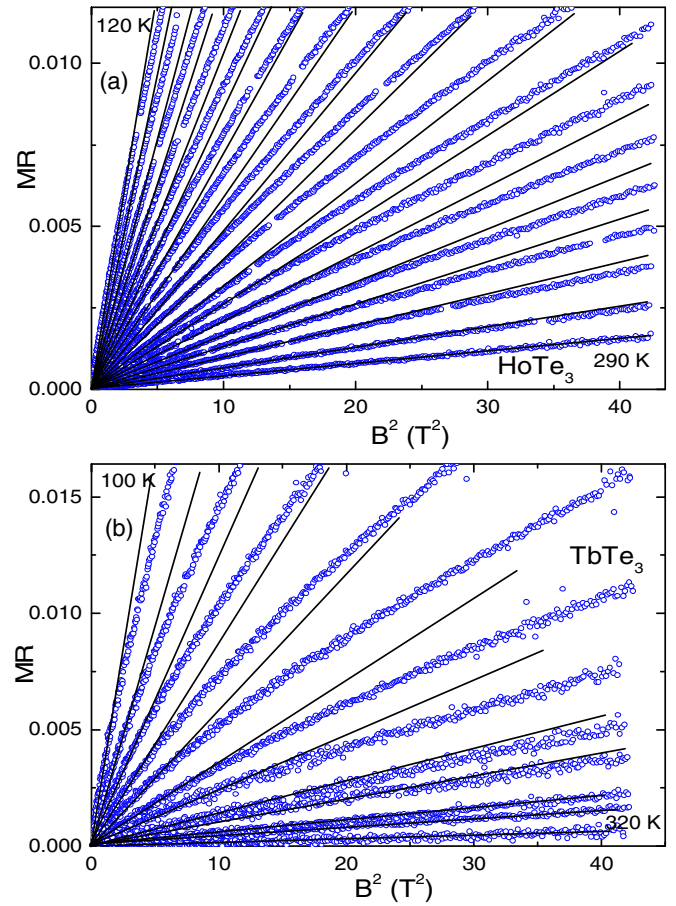


FIG. 3. Magnetoresistance of HoTe₃ (a) and TbTe₃ (b) as a function of square magnetic field, B^2 , at different temperatures. Solid black lines demonstrate the deviation of $MR(B)$ dependencies from a square law at some value of magnetic field, B^* .

Hence the mean-free time τ_{hs} is proportional to the length of the Fermi surface between hot spots divided by magnetic-field H strength and by electron velocity in real space v_{\perp} [3]:

$$\tau_{hs} = (c/eHw_{hs}) \int dl/v_{\perp}. \quad (1)$$

If the electron trajectory in magnetic field is closed, its motion is periodic given by the cyclotron (or Larmor) period $T_L = 2\pi/\omega_c$, where the cyclotron frequency $\omega_c = e\hbar H/m^*c$ and m^* is the electron effective mass. Then $\tau_{hs} \approx T_L/w_{hs}n_{hs}$, where n_{hs} is a number of hot spots along the cyclotron period. If the electron trajectory in the magnetic field is open, it also periodically passes through hot spots and the length of the Fermi surface between hot spots is approximately given by the length $2\pi/a^*$ of the first Brillouin zone divided by the number of hot spots on this open trajectory, where a^* is the lattice constant. Then, according to Eq. (1), $\tau_{hs} \sim (c/eHw_{hs})2\pi/a^*|v_{\perp}|n_{hs}$. We see that both for closed and open electron trajectories the hot-spot mean-free time is inversely proportional to magnetic field: $\tau_{hs} \propto 1/H$.

In the τ approximation for isotropic in-plane dispersion the conductivity tensor σ in magnetic field \mathbf{H} is given by the

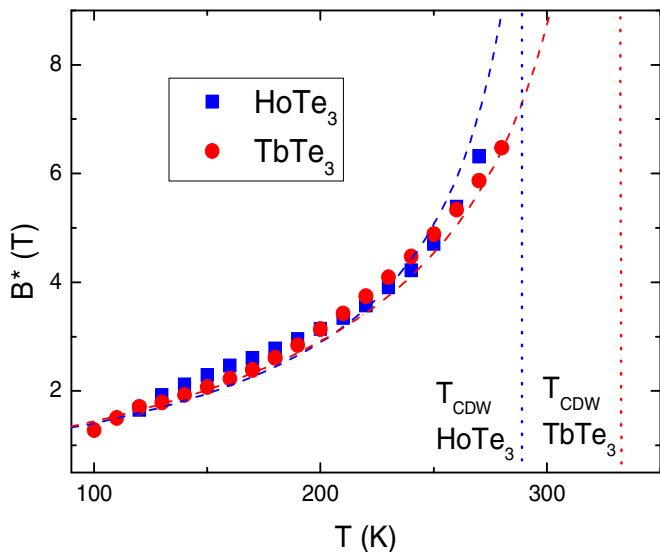


FIG. 4. Temperature dependence of the characteristic field B^* for HoTe₃ (blue) and TbTe₃ (red). Dashed lines are guides to the eye.

well-known formula [3]

$$\sigma = \frac{n_e e^2}{m^* (\omega_c^2 + 1/\tau^2)} \begin{pmatrix} \tau^{-1} & \omega_c \\ -\omega_c & \tau^{-1} \end{pmatrix}, \quad (2)$$

which gives for the resistivity tensor

$$R = \sigma^{-1} = \frac{m^*}{n_e e^2} \begin{pmatrix} \tau^{-1} & -\omega_c \\ \omega_c & \tau^{-1} \end{pmatrix}. \quad (3)$$

When τ is independent of magnetic field, as in the simplest models of electron scattering by impurities or by phonons, Eq. (3) predicts no magnetoresistance: $\Delta R_{xx}(H) \equiv R_{xx}(H) - R_{xx}(0) = 0$. The absence of magnetoresistance in this model is the result of the Hall electric field, which balances the Lorentz force. This balance can be maintained and leads to zero magnetoresistance only if the drift velocity v , included in the equations of motion, is the same for all charge carriers. Therefore, in metals with several types of charge carrier, e.g., electrons or holes from different Fermi-surface parts, the quadratic magnetoresistance appears, which saturates at high magnetic field. Thus, for the simplest isotropic model of only two types of carriers the calculation based on the kinetic equation gives (see Eq. 7.163 of Ref. [4])

$$\frac{\Delta R_{xx}(H)}{R_{xx}(0)} = \frac{\sigma_1 \sigma_2 (\omega_{c1} \tau_1 - \omega_{c2} \tau_2)^2}{(\sigma_1 + \sigma_2)^2 + (\omega_{c1} \tau_1 \sigma_1 + \omega_{c2} \tau_2 \sigma_2)^2}, \quad (4)$$

where the subscripts 1 and 2 denote the charge carriers of the first and second type, respectively. The quadratic dependence here comes from ω_c in the numerator and the saturation at $\omega_c \tau \gtrsim 1$ comes from ω_c in the denominator. Usually, the relaxation time depends on the speed v_i of an individual charge carrier, so that one cannot describe the motion of the carriers in terms of a single drift velocity even for metals with a single electron band. Therefore, even in single-band metals in weak field one observes the quadratic magnetoresistance [5]

$$\frac{\Delta R_{xx}(H)}{R_{xx}(0)} \equiv \frac{R_{xx}(H) - R_{xx}(0)}{R_{xx}(0)} = \frac{\alpha (\omega_c \tau)^2}{1 + \beta (\omega_c \tau)^2}, \quad (5)$$

where the coefficients $\alpha \sim \beta \sim 1$, which saturates in strong field when $\omega_c \tau \gtrsim 1$. This quadratic magnetoresistance in weak fields can be understood also in terms of the curvature of electron trajectories, which geometrically reduces the electron mean-free path $l_i = \tau v_i$ by the quantity $\sim l_i (l_i / r_L)^2$, where $r_L \gg l_i$ is the Larmor radius [3]. Since $l_i \propto \tau$, this effect can be taken into account in Eq. (3) by the renormalization of the electron mean-free time $\tau(H)$ in weak magnetic field H according to [3]

$$\frac{\tau(0)}{\tau(H)} = 1 + \frac{\alpha (\omega_c \tau)^2}{1 + \beta (\omega_c \tau)^2}, \quad (6)$$

which leads to Eq. (5).

If the electron scattering is dominated by the scattering in the hot spots, instead of Eq. (6) we have $\tau(H) \approx \tau_{hs} \propto 1/H$, and Eq. (3) gives $R_{xx} \propto H$. However, in real compounds the total scattering rate τ^{-1} is a sum of the contribution from various mechanisms, including those from the scattering by impurities τ_i^{-1} and by phonons τ_{ph}^{-1} . The scattering by phonons at high temperature is somewhat similar to scattering by short-range impurities, as in both cases the momentum transfer during each scattering is large and comparable to the Fermi momentum. In rather weak magnetic field, when the Landau levels are not separated and the magnetic quantum oscillation can be neglected [31], the scattering rates τ_i^{-1} and τ_{ph}^{-1} depend on magnetic field according to Eq. (6). Then $\tau^{-1} \approx \tau_i^{-1} + \tau_{ph}^{-1} + \tau_{hs}^{-1}$, and the linear MR appears only in rather strong magnetic field, when $\tau_{hs}^{-1} > \tau_i^{-1} + \tau_{ph}^{-1} \equiv \tau_{i+ph}^{-1}$, i.e., when $\omega_c \tau_{i+ph} \gtrsim 2\pi / \omega_{hs} n_{hs}$, or when the quadratic magnetoresistance saturates. At high temperature, when the scattering rate by phonons τ_{ph}^{-1} becomes larger than $\tau_{hs}^{-1} \approx \omega_c \omega_{hs} n_{hs} / 2\pi$, one should observe a usual quadratic MR. On the contrary, at lower temperature and in higher field in the CDW state, when $\tau_{hs}^{-1} > \tau_{i+ph}^{-1}$, the linear MR should be observed as a general phenomenon. This crossover is clearly seen on experimental data in Figs. 1–3 and, according to the theoretical model, the crossover field increases with temperature, as shown in Fig. 4.

Let us discuss the possible microscopic origin of the electron scattering in the hot spots in more detail. The mechanism of linear MR above the CDW transition temperature T_{CDW} , proposed in Ref. [29], assumes a strong scattering by soft phonons with a wave vector close to the nesting vector Q_N due to the Peierls instability. Then the hot spots are those connected by the CDW wave vector, as shown in Fig. 5(a). However, in our experiment the linear MR is observed much below T_{CDW} . Then the expected hot spots are the ends of the ungapped FS parts, as shown in Fig. 5(b). In these hot spots the FS reconstruction due to CDW is the strongest, and the electron dispersion depends strongly on the CDW order parameter. Therefore, electrons in such hot spots may be easily scattered by CDW fluctuations. Unfortunately, the available experimental data do not give detailed information about the Fermi surface in RTe₃ compounds in the CDW state: the ARPES data [21] do not have sufficient resolution to determine even the FS topology in the CDW state, while the magnetic quantum oscillations in the CDW state are complicated by the second CDW transition in some RTe₃ materials and by magnetic breakdown. Therefore, there are

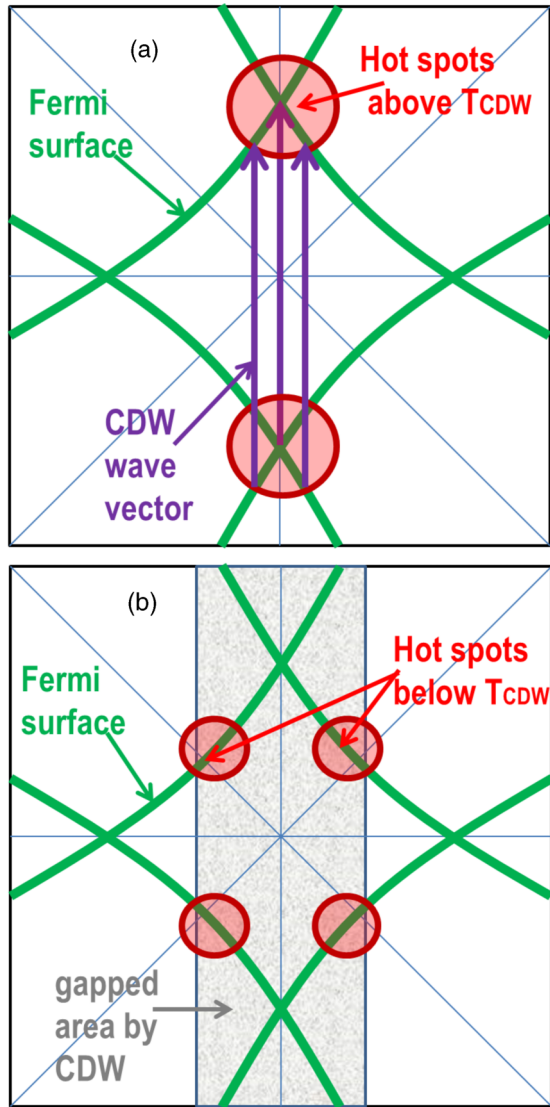


FIG. 5. Position of hot spots on the Fermi surface of $R\text{Te}_3$ above (a), as proposed in Ref. [29], and below (b) the CDW transition temperature (our work).

possibly other hot spots in the ungapped FS parts, and without detailed information about the FS in the CDW state we cannot predict the coefficient in the dependence $\tau_{\text{hs}} \propto 1/H$.

There are two types of CDW fluctuations: amplitude and phase fluctuations. Both may arise, e.g., due to the CDW pinning by crystal defects or local inhomogeneities. The amplitude CDW defects may strongly scatter the conducting electrons, e.g., due to the inhomogeneous magnetic breakdown (MB) [30], because the MB probability depends exponentially [29,32] on the gap opened in the electron spectrum due to the CDW. At the ends of the gapped region the gap values decrease,

as explicitly shown by ARPES data [21], and the magnetic breakdown become possible even in low field. Moreover, the amplitude fluctuations of the CDW gap may lead to spatial variations of the boundary between gapped and ungapped FS parts. Therefore, in Fig. 5(b) we place the hot spots at the ends of the gapped region. The MB amplitude depends strongly not only on the gap value, but also on the electron velocity and dispersion in the MB region [32], which is also affected by the amplitude fluctuations of CDW. The inhomogeneous MB probability leads to the strong electron scattering in the MB regions, playing the role of hot spots. Note that this strong scattering mechanism may be important not only for transverse but also for the longitudinal MR and may even lead to the phase inversion of magnetic quantum oscillations [30]. The CDW phase fluctuations mean local variations of the CDW wave vector, which also change the electron dispersion in our hot spots and affect the MB probability in addition to the direct scattering in these hot spots by the CDW periodic potential with inhomogeneous wave vector. Hence the CDW fluctuations may indeed lead to the strong electron scattering in the hot spots of the Fermi surface and, consequently, to linear MR.

Our theoretical model is in many aspects similar to that in Ref. [29], but there are some important differences. First, the model in Ref. [29] is developed and applied only slightly above the CDW transition temperature T_{CDW} , while our model can be applied much below T_{CDW} . Second, the model in Ref. [29] is applied only to the unreconstructed FS, while we apply our model to the strongly reconstructed FS. Third, in Ref. [29] the CDW fluctuations have the wave vector equal to the nesting vector, while in our model any Fermi-surface reconstruction due to CDW leads to scattering by CDW fluctuations, even if there are no ungapped Fermi-surface points connected by the CDW wave vector. Fourth, we propose a temperature-driven crossover between the quadratic and linear magnetoresistance, which cannot be found in the model of Ref. [29] where the CDW fluctuations are considered only in the vicinity of transition temperature.

In conclusion, we have shown that in the CDW state of $R\text{Te}_3$ compounds there is a crossover from linear magnetoresistance at low temperature to the usual quadratic magnetoresistance at higher temperature. We propose a general explanation of this phenomenon as being related to the electron scattering in the hot spots of the Fermi surface due to the spatial fluctuations or inhomogeneity of the charge density wave order parameter.

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