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Cite as: AIP Conference Proceedings **1815**, 050006 (2017); https://doi.org/10.1063/1.4976384
Published Online: 17 February 2017

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Abstract. Magnetic field dependence of the thermopower and Nernst effect of the multiband organic conductor $\alpha - (BEDT - TTF)_2KHg(SCN)_4$ is theoretically studied at low temperatures in the charge density wave (CDW) state, to fields of 30 T and several field directions. A theoretical model of quantum interlayer tunneling for the q1D charge carriers is used to probe the thermoelectric effects in the CDW state. The contribution from the q2D carriers is calculated by using the Boltzmann transport theory. The background components of the thermopower and Nernst effect as well as the quantum oscillations that originate from the closed Fermi surface orbits are analyzed. The model implies that in the CDW state, the properties of $\alpha - (BEDT - TTF)_2KHg(SCN)_4$ are determined mostly by the orbits on the new open Fermi sheets. This is in accord with the previously reported CDW scenario of the low temperature state of $\alpha - (BEDT - TTF)_2KHg(SCN)_4$ with imperfect nesting of the open Fermi surface sections.

INTRODUCTION

The quasi-two-dimensional (q2D) organic conductor, $\alpha - (BEDT - TTF)_2KHg(SCN)_4$ (where BEDT-TTF denotes bis(ethylenedithio)tetrathiafulvalene), has been extensively investigated in recent years for unusual electronic properties [1, 2]. The Fermi surface (FS) of this salt contains both a q2D cylinder and a pair of weakly warped open q1D sheets. The q1D and q2D bands are separated by a substantial gap near the Fermi level. At $T_p = 8$ K, the system undergoes a phase transition from metallic to an insulating charge density wave (CDW) state due to the nesting of sheet-like FS. Although in the CDW phase where no quasi-one-dimensional (q1D) FS sheet should survive, there appear clear angle-dependent magnetoresistance oscillations (AMRO) similar to the Lebed resonance, which is characteristic to q1D FS. Also, below T_p and at intermediate magnetic field ($B \sim 23$ T), there are profound changes in the magnetoresistance which are indicative of a first-order phase transition in the electronic structure at the so-called 'kink transition field', B_k [3, 4]. The kink field B_k defines the high field regime where the zero-field state CDW_0 is transformed into the CDW_x state with a field-dependent wavevector. This critical field clearly indicates that a magnetic field has a profound effect on the ground state of this compound.

Over past few years there has been a rising interest in experimental and theoretical research of the magnetothermopower , i.e. Seebeck (longitudinal magnetothermopower) and Nernst (transverse magnetothermopower) thermoelectric effect in organic conductors with q1D and q2D electron energy spectrum in a magnetic field [5, 6, 7, 8]. Magnetothermopower studies yield information about both the thermodynamic and transport properties of charge carriers. The investigation of magnetothermopower opens new possibilities of studying the electronic structure of the organic conductors since the thermoelectric effects are significantly more sensitive to the electron energy spectrum. It may also be used to explain electron-electron and phonon interactions in organic metals as well as the acoustic energy absorption at high frequencies.

The purpose of the present study is to probe the low-temperature state, i.e. the CDW state of the multiband organic conductor $\alpha - (BEDT - TTF)_2KHg(SCN)_4$ through how the magnetothermopower changes with the magnetic field magnitude and direction when the temperature gradient is directed along the least conducting axis in this material, the *b*-axis.

Turkish Physical Society 32nd International Physics Congress (TPS32)
AIP Conf. Proc. 1815, 050006-1–050006-4; doi: 10.1063/1.4976384
Published by AIP Publishing. 978-0-7354-1483-9/$30.00

THEORETICAL MODEL FOR THE MAGNETOTHERMOPOWER

In general, magnetothermopower contains two components: the background which is sensitive to the FS topology, and an oscillatory component which is a manifestation of the Landau quantization of the closed orbit FS. A theoretical model which involves a magnetic breakdown effect (MB) between the q1D and q2D bands is proposed in order to simulate the total and background magnetothermopower of $\alpha-(BEDT-TTF)_2KHg(SCN)_4$ in the CDW state.

In order to explore the changes in the background thermopower and Nernst effect in the CDW state, a quantum model of the interlayer magnetotunneling for the q1D charge carriers [6] for an arbitrary magnetic field direction $\mathbf{B} = (B\cos\phi\sin\theta, B\sin\phi\sin\theta, B\cos\theta)$ is used

$$\sigma_{zz}^{1D}(B,\theta,\phi) = \frac{\sigma_1}{2\pi}\sum_N J_N^2(ck_F\tan\theta)\int_0^{2\pi} dx \frac{1}{1+(N\Omega+\frac{\Delta_g}{\hbar}\alpha(\mathbf{B},\mathbf{Q})\sin x)^2\tau^2}, \tag{1}$$

where

$$\alpha(\mathbf{B},\mathbf{Q}) = 4\exp\Big(-\frac{l^2Q_x^2}{4}\Big)J_0^2(k_FQ_xl^2)\sin\left[\frac{cQ_x}{2}\left(\tan\theta\sin\phi+\frac{Q_z}{Q_x}\right)\right]. \tag{2}$$

Here, σ_1 is the electrical conductivity of the q1D carriers in the absence of a magnetic field, c and l are the interlayer and in-plane spacing, τ is the electron relaxation time, k_F is the Fermi wave number, Ω is the cyclotron frequency, Δ_g is the CDW gap energy, Q_x and Q_z are the in- and out-of-plane components of the wave vector Q of the CDW state, J_0 and J_N are the 0th and Nth order Bessel function.

The interlayer electrical conductivity σ_{zz}^{2D} from the q2D charge carriers is calculated by using the Boltzmann transport theory

$$\sigma_{zz}^{2D} = \sigma_2\left\{J_0^2(\zeta x)J_0^2(\eta x) + 2h^2\left(J_0^2(\zeta x)\sum_{k=1}^{\infty}\frac{J_k^2(\eta x)}{h^2+k^2} + J_0^2(\eta x)\sum_{n=1}^{\infty}\frac{J_k^2(\zeta x)}{h^2+k^2}\right)\right\}. \tag{3}$$

where $h = \frac{m^*}{eB\tau}\sqrt{1+x^2}$, $x=\tan\theta$, σ_2 is the electrical conductivity of the q2D carriers along the layers in the absence of a magnetic field and $\zeta = ck_F\cos\phi$, $\eta = ck_F\sin\phi$.

At low enough temperatures where the condition $\Omega\tau \gg 1$ is satisfied, the quantization of the energy of electrons whose motion in the plane orthogonal to the magnetic field is finite should be taken into account. Their states belong to the q2D closed orbits of the FS since the q1D part of the FS is an open orbit, and therefore does not undergo Landau quantization. The oscillatory part of the coefficients is deteremined mainly by the oscillatory dependence on the inverse magnetic field $1/B$ of the relaxation time $\tau(\varepsilon)$ that result from the summation over the electron states in the incoming term of the collision integral [9]. In the Born approximation, for $\hbar\Omega \ll t_c$, the relaxation time is given by

$$\frac{1}{\tau(\varepsilon)} = \frac{1+\Delta_{\rm osc}}{\tau_0}, \tag{4}$$

where

$$\Delta_{\rm osc} = \left(\frac{\hbar\Omega}{t_c}\right)^{1/2}\sum_{k=1}^{\infty}(-1)^k k^{-1/2}\Phi(k\Lambda)\cos\left(\frac{\pi k}{\cos\theta}\right)\left\{\cos\left(\frac{kS_{\rm max}}{e\hbar B}-\frac{\pi}{4}\right)+\cos\left(\frac{kS_{\rm min}}{e\hbar B}+\frac{\pi}{4}\right)\right\}. \tag{5}$$

Here, $\hbar$ is the Planck constant divided by 2π, τ_0 is the nonoscillatory part of the relaxation time, t_c is the interlayer transfer integral, $\Phi(k\Lambda) = k\Lambda/\sinh(k\Lambda)$ is the temperature damping factor where $\Lambda = 2\pi^2k_BT/\hbar\Omega \ll 1$, and k_B is the Boltzmann constant. $S_{\rm max}$ and $S_{\rm min}$ are two extremal cross-sections of the Fermi surface by the plane p_B=const. As a result, the thermoelectric coefficient α_{zz}, which is proportional to the relaxation time τ, acquires an oscillatory component $\alpha_{zz}^{\rm osc} \sim \sigma_0\partial\Delta_{\rm osc}/\partial\mu$. μ is the chemical potential.

With inclusion of $\tau_{\rm OSC}$, calculations yield the following asymptotic expressions for the oscillatory with $1/B$ part of the interlayer thermoelectric coefficients $\alpha_{zz}^{\rm osc}$ and $\alpha_{yz}^{\rm osc}$

$$\alpha_{zz}^{\rm osc} = \frac{\pi^2k_B^2T}{3e\mu}\sigma_0\frac{\mu}{\left(t_c\hbar\Omega\cos\theta\right)^{1/2}}J_0^2\left(\frac{cp_F\tan\theta}{\hbar}\right)\sum_{k=1}^{\infty}(-1)^k k^{3/2}\Phi(k\Lambda)R_S(k)R_D(k)R_W(k)\sin\left(2\pi k\left(\frac{F}{B}-\frac{1}{2}\right)\right), \tag{6}$$

$$\alpha_{yz}^{\rm osc} = \alpha_{zz}^{\rm osc}\tan\theta\sin\phi. \tag{7}$$

Here $F = \mu\, m/\hbar e \cos\theta$ is the fundamental frequency, $R_S(k) = \cos(k\pi g/\cos\theta)$ is the spin splitting damping factor, g is the conduction electron g-factor. $R_D(k) = \exp(-\pi k/\Omega\tau)$ is the Dingle factor and $R_W(k) = J_0(4\pi k t_c/\hbar\Omega)$ describes the interlayer coupling. When two types of charge carriers are involved in the transport one should also sum up the corresponding contributions to obtain the total magnetothermopower.

The corresponding components of the total interlayer thermopower (Seebeck effect) and Nernst effect are determined as following

$$S_{zz} \simeq \frac{\pi^2 k_B^2 T}{3e}\frac{1}{\sigma_{zz}}\frac{\partial\sigma_{zz}}{\partial\mu} + \frac{\alpha_{zz}^{\mathrm{osc}}}{\sigma_{zz}} \qquad \text{and} \qquad N_{yz} \simeq \frac{\pi^2 k_B^2 T}{3e}\frac{1}{\sigma_{zz}\tan\theta\sin\phi}\frac{\partial\sigma_{zz}}{\partial\mu} + \frac{\alpha_{yz}^{\mathrm{osc}}}{\sigma_{zz}}. \tag{8}$$

RESULTS AND DISCUSSION

Fig. 1 shows the theoretically simulated curves of the magnetic field dependence of total thermopower (Seebeck effect) and Nernst effect, by using the eqs. (8), for temperature gradient along the crystallographic direction b (or coordinate axis z), S_{zz} and N_{yz}, at $T = 0.6$ K, in the CDW state of $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$. The magnetic field is rotated from the b-axis to the a-axis (or coordinate axis y) for $\theta = 26°$ and $\theta = 45°$ at fixed in-plane angle $\phi = 30°$, counted from the c-axis. The selected θ angles correspond to the first dip and peak in the angular magnetoresistance oscillations (AMRO) as obtained from the AMRO studies in this material in the CDW state [7, 8].

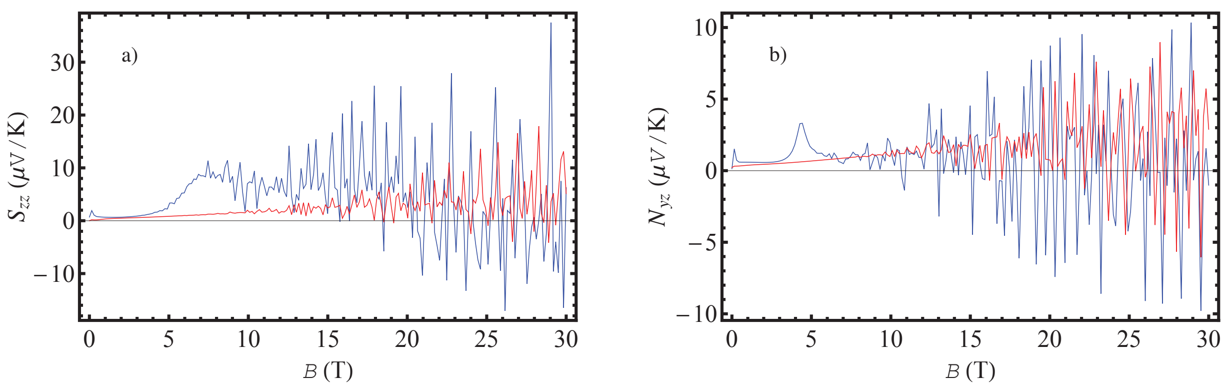

FIGURE 1. Magnetic field dependence of the total a) Seebeck and b) Nernst effect, $S_{zz}(B)$ and $N_{yz}(B)$, at $T = 0.6$ K in the CDW state of $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$ at field tilted for $\theta = 26°$ (blue curves) and $\theta = 45°$ (red curves) from the b-axis and fixed in-plane angle $\phi = 30°$.

Quantum oscillations in magnetothermopower, associated with Landau quantization are observed above B=8 T, the threshold field at which the α-frequency oscillations emerge. In the present case, at $T = 0.6$ K, the energy spectrum consist of Landau levels of only the α orbit since the higher harmonics of the fundamental α orbit have not been revealed in the oscillation frequency spectrum. Calculations are performed with the corresponding fundamental frequency for each field direction from the b-axis: $F_{\alpha 1} = 845$ T for $\theta = 26°$ and $F_{\alpha 2} = 1135$ T for $\theta = 45°$. The suppression of the quantum oscillations at higher angles from the b-axis is associated with the decrease of the amplitude of the fundamental α orbit with tilting the field towards the layers plane. The attenuation of the oscillations below $B_k \sim 23$ T is due to the so-called 'kink field'- induced phase transition.

The background magnetothermopower is determined by the first terms of S_{zz} and N_{yz} in eq. (8). The total background Seebeck and Nernst effect (solid curves) are hole-like, i.e., positive for selected field directions as shown in Fig. 2. The Nernst effect dominates by far which can be attributed to the phonon drag effect resulting from the electron-phonon coupling. Calculated contributions from the two types of charge carriers show that at low temperatures the behavior of the CDW state in $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$ is determined completely by the q1D carriers (doted curves in Fig. 2) whose states belong to the new open orbits of the FS due to the reconstruction below the transition temperature T_p. The existence of the open sheets on the FS below T_p is in a good agreement with the model given in Ref. [10]. Previous reports on low-temperature magnetoresistance indicate that it is determined by the open orbits only in fields below 10-15 T [11, 12, 13].

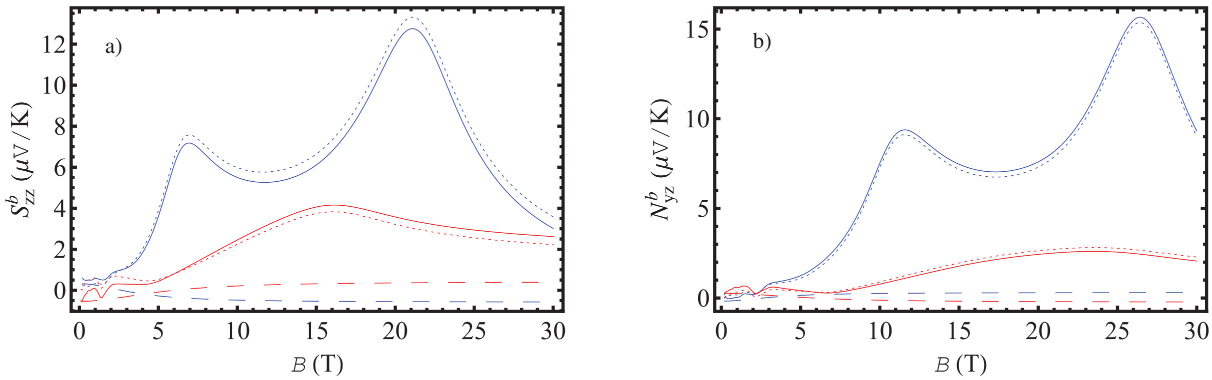

FIGURE 2. Magnetic field dependence of the background a) Seebeck and b) Nernst effect, $S^b_{zz}(B)$ and $N^b_{yz}(B)$, at $T = 0.6$ K in the CDW state of $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$ for magnetic field tilt $\theta = 26°$ (blue curves) and $\theta = 45°$ (red curves) from the b-axis and fixed in-plane angle $\phi = 30°$. The solid curves are the total background Seebeck and Nernst effect. Doted lines represent the contribution from the q1D carriers and dashed lines the corresponding contribution from the q2D carriers.

Present results on low-temperature magnetothermopower in $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$ indicate that it is determined completely by the q1D carriers on the new open FS orbits to fields of 30 T, including the $\mathrm{CDW_x}$ state above the 'kink field', $B_k \sim 23$ T. This indicates that the open FS sheets can still survive in the $\mathrm{CDW_x}$ state but their effect on the magnetothermopower is weakened (S^b_{zz} and N^b_{yz} decrease with increasing filed above B_k) compared to that in the $\mathrm{CDW_0}$ state. Since the magnetothermopower is non-vanishing at given field directions it follows that the nesting of the original q1D open sheets is imperfect.

CONCLUSION

A theoretical model for the q1D and q2D band is proposed in order to simulate the magnetothermopower in $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$. Analysis of the background components imply that at low temperatures, in the $\mathrm{CDW_0}$ and $\mathrm{CDW_x}$ state, the properties of $\alpha-(\mathrm{BEDT-TTF})_2\mathrm{KHg(SCN)}_4$ are determined by the electrons on the new open Fermi orbits. Quantum oscillations observed in the both thermoelectric effects, at fields above 8 T, originate only from the α orbit. These results corroborate and advance previous findings on magnetothermopower in this material.

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