Original Research Article

Fundamental Acoustic Wave Generation in Crystalline Organic Conductors with Two Conducting Channels

ABSTRACT

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> A linear thermoelectric generation of a fundamental acoustic wave in organic conductors with two conducting channels, guasi-one dimensional (g1D) and guasi-two dimensional (q2D), is analyzed theoretically. Specifically, the case when an acoustic wave with a fundamental frequency ω is generated along the most conducting axis of the multiband organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ is considered. The magnetic field and angular dependences of the wave amplitude for two boundary conditions, isothermal and adiabatic are obtained. Findings show that the wave amplitude for the isothermal boundary is much larger than the one for the adiabatic boundary although there is a heat flux trough the conductor's surface in the former. This is completely different compared to the case of a wave generated along the least conducting axis and the possible reasons behind this behavior are discussed. The angular oscillations of the fundamental wave amplitude are associated with the charge carriers motion on both the cylindrical part and quasi-planar sheets of the Fermi surface in a tilted magnetic field. The changes in the wave amplitude with the field orientation are correlated with the corresponding angular changes in the inplane thermoelectric coefficient and thermal conductivity. Following the magnetic field behavior of both the in-plane electromagnetic and thermal skin depth we find that the wave generation and propagation in the plane of the layers are determined mainly by the thermal wave as its skin depth is thousand times larger than the one of the electromagnetic wave. It is shown that both the q1D and q2D charge carriers contribute to the observation of the effect but the group of charge carriers with a q1D energy spectrum is significantly dominant in the generation of the fundamental acoustic wave in the plane of the layers.

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10 Keywords: Organic conductors, q1D and q2D group of charge carriers, thermoelectric effect, high-11 frequency fundamental acoustic wave, angular oscillations of the fundamental acoustic wave 12 (AOFAW), magnetotransport anisotropy.

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15 1. INTRODUCTION

17 Crystalline organic conductors have been among the most exciting objects in solid state 18 physics and chemistry over the last two decades, providing a laboratory not only for studying virtually all the ground states known in condensed matter physics but also for discovering new ones. The 19 20 organic conductors based on the bis(ethylenedithio)tetrath-iafulvalene molecule (BEDT-TTF, or, 21 shorter, ET) have initially attracted attention due to the discovery of the ambient pressure 22 superconductivity in a layered cation radical salt β -(ET)₂|₃ [1]. Further extensive efforts on the 23 synthesis and studies of new salts of ET and its derivatives gave rise to a new generation of guasi-24 two-dimensional compounds [2] with properties ranging from magnetic dielectric to superconducting. 25 depending on the chemical composition and external conditions such as temperature, pressure and 26 magnetic field.

In organic conductors large variations in the magnetoresistance are observed as the direction of the magnetic field is varied and are referred to as angular-dependent magnetoresistance oscillations (AMRO) [3]. These effects in quasi-one-dimensional systems are known as Danner [4], Lebed [5, 6], and third angular effects [7, 8], depending on whether the magnetic field is rotated in the *ac, bc,* or *ab* plane, respectively. (The *a* and *c* axes are the most- and least-conducting directions, respectively). Oscillations in quasi-two-dimensional systems include the Yamaji [9] oscillations and the

anomalous AMRO in the low-temperature phase (LTP) of two-band organic conductors α -(BEDT-33 34 TTF)₂MHg(SCN)₄ [M=K, Rb, TI] [3]. The family α -(BEDT-TTF)₂MHg(SCN)₄ [M=K, Rb, TI] are of 35 particular interest because they have a rich phase diagram and coexisting quasi-one dimensional and 36 quasi-two dimensional Fermi surface (FS) [10, 11]. Metallic, superconducting, and density wave 37 phases are possible, depending on temperature, pressure, magnetic field, and anion type. At ambient pressure, the family with M=K, Rb, TI undergo a transition from a metal to a charge density wave 38 (CDW) phase at a temperature T_{CDW} =8 [12], 10 [13], and 12 K [14], respectively. On passing through 39 40 the "kink-field" transition at $B_k=23$ T (for M = K), the CDW is removed and is replaced by a metallic 41 phase with a FS consisting of a quasi-two dimensional (q2D) hole cylinder, known as the α pocket, 42 and a pair of quasi-one dimensional (q1D) electronic sheets [15, 16]. The low-dimensional character of the organic conductors leads to important consequences in their response to a magnetic field. In 43 44 fact, numerous drastic deviations from the conventional three-dimensional behavior and even 45 qualitatively new effects, in particular related to the field orientation, have been found in these 46 materials (see Ref. [17]).

Generation of acoustic oscillations in solids by an electromagnetic wave can occur in a linear regime, when the frequency of the incident wave is equal to the frequency of the excited wave, and in a nonlinear regime, when the frequency of the excited elastic waves is a multiple of the frequency of the electromagnetic wave. In materials that are good conductors, both linear and nonlinear electromagnetic excitations of ultrasound occur as a result of interaction of the electromagnetic wave with the conduction electrons.

53 The mechanisms of linear transformation, which are responsible for the generation of high-54 frequency acoustic waves in conducting media at the frequency ω of an electromagnetic wave 55 incident on the surface of the metal, are a standard subject of investigation in electromagnetic-56 acoustic conversion problems. The induction [18-21] and deformation force [22-27] have been studied 57 in greatest detail as sources of linear generation of acoustic waves, i.e., fundamental wave generation 58 with frequency ω . Apart from the induction and deformation forces, longitudinal acoustic waves at 59 fundamental frequency can also be generated by thermoelectric forces [28-31]. In conducting media, 60 the mechanism occurs as follows: when an electromagnetic wave with frequency ω is incident on the 61 conductor, nonuniform temperature oscillations of the same frequency appear as a result of the 62 thermoelectric effect. These oscillations, in turn, generate acoustic oscillations in the conductor with a 63 frequency ω that coincides with the frequency of the incident electromagnetic wave (contactless 64 acoustic wave generation).

65 The purpose of the present work is to study the magnetic field and angular dependence of the amplitude of a fundamental high-frequency acoustic wave ($\omega = 10^9$ Hz) generated through linear 66 67 thermoelectric effect in the metallic phase of organic conductors with two conducting channels, quasi-68 one dimensional (q1D) and quasi-two dimensional (q2D). By far, this phenomenon has been 69 considered in organic conductors with only q2D group of charge carriers [32] and in two-band organic 70 conductors for an acoustic wave that is generated along the least conducting axis, z - axis [33]. The 71 present work analyzes the generation of a high-frequency acoustic wave in two-band organic 72 conductors along the most conducting x – axis. The results obtained show that the amplitude of the 73 fundamental acoustic wave is by far larger than the one of a wave that is generated along the least 74 conducting axis. In addition, for generation along the most conducting axis the amplitude of the 75 fundamental wave for the isothermal boundary is always larger than the one for the adiabatic boundary although there is a heat flux trough the conductor's surface in the former. We suggest that 76 77 the distinct behavior of the fundamental wave for both geometries is correlated with the high 78 magnetotransport anisotropy in these materials. The changes in the amplitude of the induced acoustic 79 wave with the magnetic field orientation are associated with the angular dependent changes in both 80 the in-plane thermoelectric coefficient and thermal conductivity. The period of angular oscillations of 81 the acoustic wave generated along the layers is half the period of angular oscillations of the wave 82 generated across the layers indicating that generation of a fundamental acoustic wave in the plane of 83 the layers is significantly affected by the q1D charge carriers. In highly anisotropic organic conductors 84 due to the small electron mean free-path the thermoelectric generation of high-frequency fundamental 85 acoustic waves can be observed in a wide range of fields and angles providing possibilities for 86 experimental studies using non-contact ultrasonic techniques. Such studies will give new insights into 87 the unusual electronic properties of these systems.

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2. LINEAR FUNDAMENTAL ACOUSTIC WAVE GENERATION: FORMULATION OF THE PROBLEM

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93 A high-frequency fundamental acoustic wave with frequency ω is generated as a result of 94 temperature oscillations which are induced by an electromagnetic wave with the same frequency. 95 Here we consider a case when an electromagnetic wave $\mathbf{E} = (0, E_{v}, 0)$ with frequency $\boldsymbol{\omega}$ is incident 96 normally on the conductor's surface along the most conducting axis (x - axis), $\mathbf{k} = (k, 0, 0)$ of a multi-97 band organic conductor. In that case, the only nonzero component of the current density is the 98 y-component, $\mathbf{j} = (0, j_y, 0)$. The fundamental wave is generated and propagating along the most 99 conducting axis (x – axis), and therefore all of the quantities depend only on the x – component. The 100 conductor is placed in magnetic field oriented at an angle θ from the normal to the plane of the 101 layers, in the xz plane, $B = (B\sin\theta, 0, B\cos\theta)$. A temperature oscillating with frequency ω occurs 102 only if the condition $\omega \tau \ll 1$ is satisfied, where τ is the relaxation time of the conduction electrons. 103 We study the case of a normal skin effect when the condition $k_T l \ll 1$, where l is the electron mean-104 free path length and k_T is the thermal wave number, is fulfilled.

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2.1 System of equations. Calculation of the temperature distribution $\Theta(x)$

109 The complete system of partial differential equations, describing the generation of longitudinal 110 fundamental wave at frequency ω includes Maxwell's equations for the magnetic **B** and electric 111 **E** field, the kinetic equation for the nonequilibrium correction Ψ to the electron distribution function 112 $f_0(\varepsilon)$, and the equations of heat conduction and the theory of elasticity for ionic displacement 113 $\mathbf{U} = (U_{\omega}, 0, 0)$:

114
$$\operatorname{curl} \mathbf{B} = \mu_0 \mathbf{j}; \quad \operatorname{curl} \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t},$$
 (2.1)

115
$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + e\left[\mathbf{v}\mathbf{B}\right]\frac{\partial}{\partial \mathbf{p}} + \frac{1}{\tau}\right)\Psi = -e\mathbf{v}\mathbf{E} + \frac{\varepsilon - \mu}{k_B T}\mathbf{v}\nabla T, (2.2)$$

116
$$C \frac{\partial \Theta}{\partial t} + \operatorname{div} \mathbf{Q} = 0, \ Q_i = k_B T \alpha_{ik} \frac{\partial j_k}{\partial x_k} - \kappa_{ik} \frac{\partial \Theta}{\partial x_k},$$
 (2.3)

117
$$\rho \frac{\partial^2 U_i}{\partial t^2} - \lambda_{iklm} \frac{\partial U_{lm}}{\partial x_k} = -\rho s^2 \beta \delta_{ik} \frac{\partial \Theta}{\partial x_k}.$$
 (2.4)

Here μ_0 is the magnetic permeability of the vacuum, v and p are the electron velocity and 118 119 momentum, e is the electron charge, Q is the heat flux, Θ is the high-frequency addition to the 120 mean temperature T of the crystal, μ is the chemical potential, C is the volumetric heat capacity, k_{R} is the Boltzmann constant, α_{ik} is the thermoelectric coefficient, κ_{ik} is the thermal conductivity, ρ is 121 the density of the crystal, δ_{ik} is the Kronecker delta, s is the fundamental acoustic wave velocity and 122 123 β is the volumetric expansion coefficient. $U_{lm} = (\partial U_l / \partial x_m + \partial U_m / \partial x_l)/2$ is the deformation tensor and λ_{iklm} are components of the elastic tensor of the crystal. The subscripts in U and x describe the 124 125 wave polarization and direction of wave propagation, respectively. The wave is taken to be 126 monochromatic, so the differentiation with respect to the time variable is equivalent to multiplication by 127 $(-i\omega)$.

128 For the given geometry the above system of equations (2.1-2.4) takes the following form

129
$$\frac{\partial B_z}{\partial x} = \mu_0 j_y; \quad \frac{\partial E_y}{\partial x} = i\omega B_z,$$
 (2.5)

130
$$\left(\frac{\partial}{\partial t_B} + \frac{1}{\tau}\right) \Psi = v_y (eE_y + \frac{\varepsilon - \mu}{k_B T} \frac{\partial T}{\partial x}),$$
 (2.6)

131
$$-i\omega C\Theta - \kappa_{xx} \frac{\partial^2 \Theta}{\partial x^2} = -k_B T \alpha_{xy} \frac{\partial j_y}{\partial x}, \qquad (2.7)$$

132
$$\frac{\partial^2 U_{\omega}}{\partial x^2} + q^2 U_{\omega} = \beta \frac{\partial \Theta}{\partial x}.$$
 (2.8)

Here t_B is the time of motion of the conduction electrons in a magnetic field under the influence of the Lorentz force with a period $T_p = 2\pi/\omega_c$ and cyclotron frequency $\omega_c = eB\cos\theta/m^*$. κ_{xx} and α_{xy} are the in-plane thermal conductivity and thermoelectric coefficient and $q = \omega/s$ is the acoustic wave vector.

137 The above system of equations must be supplemented with the corresponding boundary 138 conditions for the temperature distribution and fundamental acoustic wave amplitude at the 139 conductor's surface. Two types of boundary are considered, isothermal and adiabatic for which the 140 boundary conditions are defined as follows:

141
$$\Theta|_{x=0} = 0, \qquad \kappa_{xx} \frac{\partial \Theta}{\partial x}|_{x=0} = 0,$$
 (2.9)

142
$$U_{\omega}|_{x=0} = 0, \quad \frac{\partial U_{\omega}}{\partial x}|_{x=0} = -\beta \frac{\partial \Theta}{\partial x}|_{x=0}.$$
 (2.10)

143 By using Maxwell's equations one obtains the following expression for the current density

144
$$j_y = \frac{ik_E^2}{\omega\mu_0} Exp[i(k_E x - \omega t)],$$
 (2.11)

145 where $k_E = (1+i)/\delta_E$ and $\delta_E = (2\rho_{yy}/\omega\mu_0)^{1/2}$ are the wave vector and skin depth of the 146 electromagnetic field, respectively.

147 Substituting eq. (2.11) into the heat conduction equation (2.7) we obtain the following partial 148 differential equation for the temperature distribution within the conductor

149
$$\frac{\partial^2 \Theta}{\partial x^2} + k_T^2 \Theta = \frac{k_E^3 k_B T \alpha_{xy}}{\kappa_{xx} \omega \mu_0} Exp \ [ik_E x], \qquad (2.12)$$

150 where $k_T = (1+i)/\delta_T$ and $\delta_T = (2\kappa_{xx}/\omega C)^{1/2}$ are the wave vector and skin depth of the thermal field 151 under the conditions of a normal skin effect.

Crucial part in further theoretical analysis is to obtain the solutions of the eq. (2.12) and 152 153 substitute them in the eq. (2.8) for the amplitude of the generated fundamental acoustic wave to be 154 determined. The linear acoustic wave generation due to thermoelectric effect, i.e., generation of an acoustic wave with fundamental frequency ω , can be observed only when the coupling between the 155 electromagnetic and temperature oscillations is weak, i.e., when the parameter $a = k_B T(\alpha_{xy}^2 / \rho_{yy} \kappa_{xx})$ 156 that determines the coupling between the two oscillations is much smaller than 1, $a \ll 1$. In that 157 158 case, the temperature distribution that results from the oscillations of the current density i can be 159 determined. Using the boundary conditions for the temperature (eq. (2.9)) the following solution for the 160

161 temperature distribution within the conductor is obtained

162
$$\Theta(x) = \frac{k_E k_B T \alpha_{xy}}{\kappa_{xx} \omega \mu_0} \frac{1}{k_T^2 - k_E^2} (Exp \ [ik_E x] - b \ Exp \ [ik_T x]), (2.13)$$

163 where b is 1 for an isothermal boundary condition, and k_T/k_E for an adiabatic boundary condition.

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2.2 Calculation of the fundamental acoustic wave amplitude $U_{\omega}(x)$

By substituting the obtained expression for the temperature distribution $\Theta(x)$ (eq. (2.13)) and using the boundary conditions for the wave amplitude (eq. (2.10)) into the equation of the theory of elasticity (2.8), calculations yield the following expressions for the complex amplitude of the fundamental acoustic wave excited by the temperature oscillations for both the isothermal and adiabatic boundary:

172
$$U_{\omega}^{i}(x) = i \frac{k_{B} T \beta \alpha_{xy} \kappa_{xx}}{C \omega (\rho_{yy} \omega \mu_{0})^{1/2}} \frac{1}{(\rho_{yy} / \omega \mu_{0})^{1/2} + (\kappa_{xx} / \omega C)^{1/2}}$$
(2.14)

173
$$U_{\omega}^{a}(x) = (1-i)\frac{\sqrt{2\mu_{0}k_{B}T\beta\alpha_{xy}\kappa_{xx}}}{q(\rho_{yy}\omega)^{1/2}}\frac{1}{\rho_{yy}C + (\rho_{yy}\kappa_{xx}C\mu_{0})^{1/2}}$$
(2.15)

174 The components of the conductivity tensor which relate the current density to the electric field 175 can be calculated by using the Boltzmann transport equation for the charge carrier distribution 176 function, based on the tight binding approximation band structure within the single relaxation time 177 approximation τ (eq. (2.6)) [34]. The components of the electrical conductivity and thermoelectric 178 tensor are determined as follows

179
$$\sigma_{ij} = \frac{2e^3B}{(2\pi\hbar)^3} \int \frac{\partial f_0}{\partial \varepsilon} d\varepsilon \int dp_B \int_0^{T_p} dt v_i(t) \Psi_j.$$
(2.16)

180
$$\alpha_{ij} = \frac{2e^3B}{(2\pi\hbar)^3} \int \frac{\varepsilon - \mu}{k_B T} \frac{\partial f_0}{\partial \varepsilon} d\varepsilon \int dp_B \int_0^{T_p} dt v_i(t) \Psi_j. \quad (2.17)$$

181 Here \hbar is the Planck's constant divided by 2π , $p_B = p_x \sin \theta + p_z \cos \theta = \text{const}$ is the momentum 182 projection in the magnetic field direction.

183 We shall assume that the velocities $\pm \mathbf{v}_1$ of the electrons belonging to the plane sheets of the 184 FS, i.e., the q1D group of charge carriers are predominantly oriented in a direction determined by an 185 angle ϕ so that their velocities are $v_{1x} = \pm v_1 \cos \phi$, $v_{1y} = \pm v_1 \sin \phi$ and the dispersion law for the 186 charge carriers belonging to a weakly warped FS cylinder has the form

187
$$\varepsilon(p) = \frac{p_x^2 + p_y^2}{2m^*} + \eta \frac{v_F \hbar}{c} \cos(\frac{cp_z}{\hbar})$$
(2.18)

188 where m^* is the electron cyclotron effective mass, η is the quasi-two dimensionality parameter, v_F 189 is the characteristic Fermi velocity of the electrons along the layers and c is the lattice constant.

190 We shall assume that the angle of deviation of the magnetic field from the direction normal to 191 the layers is not too close to $\pi/2$ so that all orbits of electrons with a quadratic dispersion law are 192 closed and do not contain self-intersections. The components of the conductor's kinetic coefficients 193 σ_{ij} are a sum of the contributions of quasi-one dimensional q1D and quasi-two dimensional q2D 194 charge carriers, which are calculated using the eq. (2.16). Specifically, the following expressions are 195 obtained for the components of the conductivity tensor

196
$$\sigma_{xx} = \sigma_{xx}^{q2D} + \sigma_{xx}^{q1D} = \frac{\gamma^2 \sigma_2}{B^2 \cos^2 \theta} (1 - \sigma_{zz}^{q2D} \tan^2 \theta) + \sigma_1 \cos^2 \phi,$$
 (2.19)

197
$$\sigma_{yy} = \sigma_{yy}^{q2D} + \sigma_{yy}^{q1D} = \left(\frac{\gamma^2 \sigma_2}{B^2 \cos^2 \theta} + \sigma_{zz}^{q2D} \tan^2 \theta\right) + \sigma_1 \sin^2 \phi, \qquad (2.20)$$

198
$$\sigma_{xy} = \sigma_{xy}^{q2D} + \sigma_{xy}^{q1D} = \frac{\gamma \sigma_2}{B \cos \theta} (1 - \sigma_{zz}^{q2D} \tan^2 \theta) + \sigma_1 \cos \phi \sin \phi, \qquad (2.21)$$

199
$$\sigma_{zz}^{q2D} = \sigma_2 \cos\theta J_0^2 (\frac{cD_p \tan\theta}{\hbar}).$$
(2.22)

Here σ_1 and σ_2 are the contributions to the electrical conductivity along the layers with **B** = 0 of q1D and q2D group of charge carriers, $D_p = 2p_F$ is the averaged diameter of the FS along the p_x – axis, J_0 is the zeroth order Bessel function and $\gamma = m^* / e\tau$.

203 In τ – approximation, it is sufficient to calculate the components of the electrical conductivity 204 tensor and the rest of the kinetic and thermoelectric coefficients, describing the heat flux and 205 thermoelectric effects, are obtained as follows

206
$$\rho_{yy} = \sigma_{yy}^{-1} = 1 + \frac{\sigma_1}{\sigma_2} \frac{B^2 \cos^2 \theta \cos^2 \phi}{\gamma^2},$$
 (2.23)

207

$$= \frac{\pi^2 k_B T}{3e^2 \mu} \left(\frac{\gamma \sigma_2}{B \cos \theta} + \sigma_1 \cos \phi \sin \phi \right) - \frac{\pi^2 k_B T}{3e^2} \frac{\gamma \sigma_2}{B \cos \theta} \alpha_{zz} \tan^2 \theta,$$
(2.24)

$$208 \qquad \alpha_{zz} = \frac{\pi^2 k_B T}{3e^2} \frac{\partial \sigma_{zz}^{q2D}}{\partial \varepsilon} |_{\varepsilon=\mu} = \frac{\pi^2 k_B T}{3e^2} \frac{\sigma_2}{\mu} \left(4\sin\theta J_0 \left(\frac{cD_p \tan\theta}{\hbar}\right) J_1 \left(\frac{cD_p \tan\theta}{\hbar}\right) \right), \quad (2.25)$$

209
$$\kappa_{xx} = \frac{\pi^2 k_B^2 T}{3e^2} \sigma_{xx}.$$
 (2.26)

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211 3. RESULTS AND DISCUSSION

 $\alpha_{xy} = \frac{\pi^2 k_B T}{3e^2} \frac{\partial \sigma_{xy}}{\partial \varepsilon}|_{\varepsilon = \mu} =$

213 **3.1 Angular oscillations of the fundamental acoustic wave (AOFAW)**

215 The amplitude of the generated acoustic wave with fundamental frequency due to 216 thermoelectric effect is a function of the frequency of applied electric current ω , the magnetic field B, 217 the angle between the normal to the layers and the magnetic field θ as well as of the kinetic and thermoelectric characteristics of the conductor: electrical and thermal conductivity as well as the 218 thermoelectric coefficient. For the multi-band organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ the 219 cyclotron mass extracted from data on magnetic quantum oscillations is $m^* = 3.5m_e$ (m_e is the free 220 electron mass) and the relaxation time is of the order of $\tau = 2 \times 10^{-12}$ s [17]. We assume that the q1D 221 222 plane sheets and g2D FS cylinder are not strongly corrugated and the guasi-two dimensionality 223 parameter is of order $\eta = 0.01$.

224 The angular oscillations are characteristic of the kinetic and thermoelectric coefficients of 225 layered organic conductors and do not occur in isotropic metals. Since the amplitude of the acoustic 226 wave is determined by both the kinetic and thermoelectric coefficients angular oscillations of the 227 amplitude of the generated fundamental acoustic wave are expected to emerging when a constant 228 magnetic field is turned from the direction normal to conducting layers toward the plane of the layers. 229 The angular oscillations are associated with the charge carriers motion on both the cylindrical part 230 and quasi-planar sheets of the FS in a tilted magnetic field. The existence of points at which the 231 interaction with the wave is most effective on different trajectories leads to a resonant dependence of 232 the acoustic wave amplitude on magnetic field B and the angle between the normal to the layers and 233 magnetic field θ . Resonant oscillations of the amplitude U_{ω} on the tangent of the angle between the normal to the layers and magnetic field for both the isothermal and adiabatic boundary at T = 30 K, 234

B = 0.6 T and $\phi = 85^{\circ}$ are shown in Fig. 1. The amplitude $U_{\omega}(\tan \theta)$ exhibits giant oscillations as the value at the peaks is much larger (about two times larger) than the one at the minimum. The angular oscillations of the fundamental wave are observed when the field is tilted close to the plane of the layers, i.e., for $\tan \theta > 7$ ($\theta > 82^{\circ}$) as expected since we consider a wave generation in the plane of the layers. The amplitude of the wave increases very fast with tilting the angle towards the plane of the layers, i.e., towards the x-axis, in proportion to $(\tan \theta)^4$ since $U_{\omega}^{i,a} \sim \alpha_{xy} \kappa_{xx} \sim (\tan \theta)^4$. In

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addition, with tilting the field from the *z*-axis, the drift of charge carriers along the *z*-axis, \overline{v}_z , decreases but the drift along the *x*-axis, $\overline{v}_x = \overline{v}_z \tan \theta$, is rather large at angles close to $\theta = 90^{\circ}$.

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Fig. 1. Angular oscillations of the fundamental acoustic wave amplitude U_{ω} in the plane of the layers for isothermal (blue curve) and adiabatic (red curve) boundary at T = 30 K, $\eta = 0.01, B = 0.6$ T and $\phi = 85^{\circ}$. The dashed lines indicate the positions of the maxima $U_{\omega, \text{max}}^{i,a}$ and minima $U_{\omega, \text{min}}^{i,a}$ of the wave amplitude for both boundaries.

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251 An important feature to emphasize is that the amplitude of the fundamental wave for the 252 isothermal boundary is larger than the one for the adiabatic boundary although there is a heat flux 253 trough the conductor's surface in the former. Our suggestion is that the reason for this behavior is the 254 larger adiabatic resistivity in the case of a fundamental acoustic wave generated along the layers. In fact, when the thermoelectric coefficient is not too small (as in the case of the in-plane thermoelectric 255 256 coefficient α_{xy}), a distinction must be made between isothermal ρ_i and adiabatic ρ_a resistivities, i.e., $\rho^a = \rho^i (1 + k_B T \alpha_{xy}^2 / \rho^i \kappa_{xx})$. Due to the larger resistivity in the adiabatic case the fundamental 257 258 wave generated under the conditions of adiabatic boundary is more strongly attenuated than in the

259 isothermal case. 260 The minima in the $U_{\alpha}(\tan\theta)$ dependence correspond to the zero amplitude of the 261 fundamental acoustic wave and their positions coincide with the positions of the zeros in the angular 262 dependence of the in-plane thermoelectric coefficient $\alpha_{xy}(\tan\theta)$ as shown in Fig. 2. The in-plane 263 thermal conductivity is negative in the whole range of angles which indicates that the in-plane 264 conductivity is also negative as $\kappa_{xx} \sim \sigma_{xx}$ implying that the dominant carriers in the fundamental wave 265 generation are the electron-like carriers, i.e., the q1D group of charge carriers. This is expected as in 266 the multi-band organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ the g1D charge carriers drift mainly 267 along the x – axis and this is the direction along which the wave is generated.

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Fig. 2. Angular positions of the fundamental acoustic wave amplitude U_{ω} for isothermal boundary (blue curve), in-plane thermoelectric coefficient a_{xy} (green curve), in-plane thermal conductivity κ_{xx} (red curve) and the term $a_{xy}\kappa_{xx}$ (black curve) at T = 30 K, $\eta = 0.01$, B = 0.6 T and $\phi = 85^{\circ}$. The dashed lines indicate the positions of the peaks of the amplitude U_{ω} and in-plane thermal conductivity κ_{xx} .

276 The in-plane conductivity σ_{xx} is determined by the interlayer conductivity σ_{zz} (eq. 2.19) and the direction of the g1D charge carriers ϕ in the plane of the layers. When $\tan \theta >> 1$ electrons may 277 278 execute many orbits before dephasing, resulting in the resonance. In that case, the first term in eq. 279 (2.19) for the in-plane conductivity σ_{xx} is dominant. It follows from eq. 2.(19) that when $cD_p \tan\theta/\hbar$ 280 equals a zero of the zeroth-order Bessel function, then at that angle the electrical conductivity along 281 the less conducting axis, $\sigma_{zz}(\theta)$, will be negligible and consequently the in-plane conductivity $\sigma_{xx}(\theta)$ 282 vanishes at the same angle. These are known as Yamaji angles [9]. If $cD_p \tan\theta/\hbar >> 1$, then the zeros in $\sigma_{xx}(\theta)$ and $\kappa_{xx}(\theta)$ occur at angles $\theta = \theta_n^{\min}$, given by $cD_p \tan \theta_n^{\min} / \hbar = \pi (n-1/4)$, 283 n = 0, 1, 2, 3... For $\sigma_{xx}(\theta)$ and $\kappa_{xx}(\theta)$ to be a maximum it should be $\theta = \theta_n^{\max}$, where 284 $cD_n \tan \theta_n^{\max} / \hbar = \pi (n+1/4)$. The positions of the peaks in the $U_{\omega}(\tan \theta)$ dependence do not 285 286 coincide with the position of the maxima of the thermal conductivity as seen in Fig. 2. Instead, their 287 positions coincide with the positions of the extremes of the term $\alpha_{xy}\kappa_{xx}(\tan\theta)$ as expected since this 288 term is the one that determines the field and angular behavior of the wave amplitude. The term 289 $\alpha_{xy}\kappa_{xx}$ shows resonant like behavior which is existence of two close to each other extremes, with 290 sign change at angles that correspond to the angles where the in-plane thermal conductivity has maximum or minimum, $\theta = \theta_n^{\max}, \theta_n^{\min}$. This, in turn, reflects as appearance of two close maxima in 291 292 the $U_{\omega}(\tan \theta)$ dependence. In the vicinity of angles $\theta = \theta_n^{\max}$, the drift velocity \overline{v}_x of charge carriers 293 along the acoustic wavevector coincides with the velocity s of the acoustic wave, and their interaction 294 with the wave is most effective. As a result, at these angles the amplitude is the largest (especially 295 this trend is apparent for $\tan\theta >>1$) and $U_{\alpha}(\tan\theta)$ exhibits giant oscillations. The narrow peaks that appear in the $U_{\omega}(\tan\theta)$ dependence repeat with a period $\Delta(\tan\theta) = 2\pi\hbar/cD_{p}$ which is the same as 296 297 the period of oscillations of the in-plane thermal conductivity $\kappa_{xx}(\tan\theta)$ as evident from Fig. 2. This is 298 different from the case of an organic conductor with only g2D charge carriers where the period of the 299 acoustic wave amplitude generated along the least conducting axis is half the period of angular oscillations of the inverse interlayer conductivity $\kappa_{zz}^{-1}(\tan\theta)$ [32]. This indicates that the presence of 300

301 group of charge carriers with a q1D energy spectrum significantly affects the generation of the 302 fundamental acoustic wave in the plane of the layers.

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3.2 Magnetic field dependence of the fundamental acoustic wave amplitude

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Fig. 3 shows the magnetic field dependence of the amplitude of the generated fundamental acoustic wave in α -(BEDT-TTF)₂KHg(SCN)₄ for both the isothermal and adiabatic boundary at $\theta = 85^{\circ}$ and $\phi = 89^{\circ}$.

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Fig. 3. Magnetic field dependence of the fundamental acoustic wave amplitude $U_{\omega}(B)$ for isothermal (blue curve) and adiabatic (red curve) boundary at T = 30 K, $\eta = 0.01$, $\theta = 85^{\circ}$ and $\phi = 89^{\circ}$. The dashed lines indicate the B^{-4} field dependence of the wave amplitude for both boundaries. The inset shows the magnetic field dependence of $\alpha_{xy}\kappa_{xx}$ (red dashed curve) compared to $U_{\omega}(B)$ (blue solid curve). $\alpha_{xy}\kappa_{xx}$ is following the same B^{-4} field dependence as the amplitude $U_{\omega}(B)$.

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319 The fundamental wave generation starts at zero field and the amplitude decreases with 320 increasing field for both boundaries. It is obvious that the in-plane generated fundamental wave is strongly attenuated with increasing field and its amplitude decreases in proportion to B^{-4} . The 321 322 observed field dependence originates from the magnetic field behavior of the term $\alpha_{\rm m}\kappa_{\rm m}$ (that determines the amplitude $U^{i,a}_{\omega}$) which is also following the B^{-4} dependence as seen from the inset in 323 Fig. 3. This is completely different form the case of an acoustic wave generated along the least 324 conducting axis, i.e., the interlayer fundamental wave previously considered in [33] where the wave 325 amplitude is decreasing with increasing field approximately as B^{-1} . It is instructive to discuss here 326 presented results in the context of previous studies on the interlayer acoustic wave generation. We 327 first note that the amplitude of the in-plane wave is much larger than the one of the interlayer 328 329 fundamental wave of order of 10^{-3} . This is conditioned by the high magnetotransport anisotropy, i.e., 330 by the high conductivity anisotropy ratio of the interlayer and in-plane conductivity in α -(BEDT-TTF)₂KHg(SCN)₄. It follows that the larger amplitude of the in-plane acoustic wave is due to the large 331 332 in-plane electrical conductivity and consequently due to the large number of charge carriers (g1D and 333 a2D) included in the process of the thermoelectric acoustic wave generation. Another difference 334 between the two geometries is that the isothermal wave amplitude exceeds the adiabatic in a whole 335 range of fields. This implies that when performing experiments on in-plane acoustic wave generation 336 the isothermal boundary is preferable in the whole range of fields. For the interlayer wave generation 337 the preference of one type of a boundary over another depends on the magnetic field strength. In 338 addition, the in-plane acoustic wave is more strongly attenuated with increasing field (despite the

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much larger amplitude) than the interlayer acoustic wave. This is evident from the stronger field dependence (B^{-4}) in the former compared to the B^{-1} dependence in the latter [33] and is correlated with the magnetic field behavior of the thermal skin depth as well as the corresponding contributions from the q1D and q2D charge carriers as discussed below.

344 **3.3 In-plane electromagnetic and thermal skin depth**

346 Fig. 4 shows the magnetic field dependence of the total in-plane electromagnetic and thermal 347 skin depth as well as the corresponding contributions from the g1D and g2D charge carriers. The 348 total electromagnetic skin depth is linear in field as it is the one for the g2D carriers but is field 349 independent for the q1D carriers. On the other hand, the total thermal skin depth is decreasing with 350 increasing field as it is one for the g2D carriers and is field independent for the g1D carriers as in the 351 case of the electromagnetic skin depth. The field independence of both skin depths for the q1D 352 carriers follows from the field independence of the in-plane electrical conductivities for the q1D carriers $\sigma_{xy}^{q1D} = \sigma_1 \cos\phi \sin\phi$ and $\sigma_{xx}^{q1D} = \sigma_1 \cos^2\phi$ that determine the electromagnetic and thermal 353 skin depth, respectively. 354

Following the changes in skin depths with field allows to distinguish if the wave generation is 355 356 deter mined by the thermal or electrodynamic characteristics of the conductor. As evident from Fig. 4 the thermal skin depth exceeds by far the electromagnetic one (it is about 20 times larger) indicating 357 that the fundamental wave generated in the plane of the layers is mainly transmitted by the thermal 358 359 waves. Our suggestion is that in the case of linear wave generation due to the weak coupling between 360 electromagnetic and temperature oscillations the thermal skin depth is always larger than the electromagnetic one and the fundamental wave is transmitted mainly by the thermal wave that 361 362 dissipates at distance $\sim \delta_T$. We find that the thermal skin depth for the q1D charge carriers is larger 363 than the one for the q2D charge carriers (except for very low fields $B \le 0.05$ T) corroborating above 364 mentioned that both the g1D and g2D charge carriers contribute to the observation of the effect but 365 the q1D charge carriers are the dominant carriers in generation and propagation of a fundamental 366 acoustic wave along the most conducting axis in α -(BEDT-TTF)₂KHg(SCN)₄. 367





Fig. 4. Magnetic field dependence of the total in-plane a) electromagnetic and b) thermal skin depth (black curves) and the corresponding contributions from the q1D (blue curves) and q2D charge carriers (red curves) at T = 30 K, $\eta = 0.01$, $\theta = 85^{\circ}$ and $\phi = 89^{\circ}$.

375 The generation of fundamental waves is constraint by the condition $\omega \tau \ll 1$. In the organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ this condition is always fulfilled, even at high frequencies 376 $(\omega = 10^9 \text{ Hz})$ as the relaxation time of charge carriers τ is very small $(\tau = 2 \times 10^{-12} \text{ s})$. In addition, 377 for the generation of waves to be the most effective the condition $l\delta_T \ll 1$ must be satisfied. A Fermi 378 velocity of $v_F = 6.5 \times 10^4$ m/s [35] gives an electron mean free-path of order $l = 13 \times 10^{-8}$ m. We have 379 obtained from Fig. 4 the following values for the electromagnetic and thermal skin depth at 380 381 B = 0.6 T, $\delta_E = 0.03 \text{ mm}$ and $\delta_T = 0.6 \text{ mm}$, respectively. It is evident that the electron mean free-382 path l is much smaller than both skin depths providing the fundamental wave generation to be 383 studied in detail experimentally that will give new insights into the complex electronic properties of 384 these systems. 385

- 386 4. CONCLUSION
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The linear generation of high-frequency acoustic waves ($\omega = 10^9$ Hz) in layered organic 388 conductors with two groups of charge carriers, q1D and q2D, due to thermoelectric effect is 389 390 considered. Only a fundamental wave is generated due to the thermoelectric stresses caused by the 391 non-uniform temperature oscillations and its amplitude is analyzed as a function of the magnetic field 392 B, the angle θ between the normal to the layers and the magnetic field as well as of the conductor's 393 thermoelectric characteristics. Specifically, the parameter values for the organic conductor *a*-(BEDT-TTF)₂KHg(SCN)4 are used to obtain the fundamental wave amplitude U_{ω} at T = 30 K and for not 394 strongly warped open plane sheets and closed cylindrical FS. We find that the oscillatory dependence 395

396 of the fundamental amplitude is determined mainly by the angular oscillations of both the in-plane 397 thermoelectric coefficient thermal conductivity, i.e., by the angular behavior of the term $\alpha_{vv}\kappa_{vr}$ and are associated with the periodic charge carriers motion on both the cylindrical part and quasi-planar 398 sheets of the FS in a tilted magnetic field. At angles where $U_{\omega}(\tan\theta)$ is maximum, the average drift 399 400 velocity of charge carriers along the wave vector coincides with the wave velocity s. Therefore narrow 401 peaks appear in the angular dependence $U_{w}(\tan\theta)$ that correspond to the most effective interaction 402 of the charge carriers with the wave. It has been shown that the positions of the peaks of the 403 fundamental wave angular oscillations are shifted from those of the in-plane thermal conductivity and 404 the positions of the peaks in the $U_{w}(\tan\theta)$ dependence coincide with the positions of the extremes of 405 the term $\alpha_{xy}\kappa_{xx}(\tan\theta)$. It is important to note that for a fundamental acoustic wave generated in the 406 plane of the layers in organic conductors with two conducting channels the period of angular

406 plane of the layers in organic conductors with two conducting channels the period of angular 407 oscillations is larger compared to the one of a wave generated along the normal to the layers in 408 organic conductors with only q2Dgroup of charge carriers.

409 As regards the magnetic field behavior of the wave amplitude we find that the in-plane 410 generated fundamental wave is strongly attenuated with increasing field and its amplitude decreases 411 in proportion to B^{-4} . The observed field dependence originates from the magnetic field behavior of 412 the term $\alpha_{xy}\kappa_{xx}$. Comparison of the magnetic field dependence of the in-plane and interlayer

413 generated fundamental wave shows that both waves exhibit different field dependence, B^{-4} in the 414 former and B^{-1} in the latter. We find that the thermal skin depth is larger than the electromagnetic 415 one and the fundamental wave is transmitted mainly by the thermal wave that dissipates at distance 416 $\sim \delta_T$. Both the q1D and q2D charge carriers contribute to the observation of the effect but the q1D 417 charge carriers are the dominant carriers in generation and propagation of a fundamental acoustic 418 wave along the most conducting axis (*x* – axis) in the multi-band organic conductor α –(BEDT-419 TTF)₂KHg(SCN)₄.

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423 **REFERENCES**

424

- 425 1. Yagubskii EB, Shchegolev IF, Laukhin VN, Kononovich PA, Kartsovnik MV, Zvarykina AV et al.
 426 Normal-pressure superconductivity in an organic metal (BEDT-TTF)₂I₃ [bis (ethylene dithiolo) tetrathiof
 427 ulvalene triiodide]. JETP Lett. 1984;39(1):12.
- 428 2. Williams JM, Ferraro JR, Thorn RJ, Carlson K, Geiser U, Wang HH et al. Organic Superconductors:
 429 Synthesis, Structure, Properties and Theory. Prentice Hall: Englewood, Cliffs, NJ: 1992.
- 430 3. Wosnitza J. Fermi Surfaces of Low Dimensional Organic Metals and Superconductors. 1st ed.
 431 Springer: Berlin; 1996.
- 432 4. Danner KG, Chaikin PM. Non-Fermi-Liquid Behavior in Transport in (TMTSF)₂PF₆. Phys Rev Lett.
 433 1995;75: 4690.
- 434 5. Chashechkina EI, Chaikin PM. Magic angles and the ground states in (TMTSF)₂PF₆. Phys Rev Lett.
 435 1998;80: 2181.
- 6. Osada T, Kagoshima S, Miura N. Resonance effect in magnetotransport anisotropy of quasi-one dimensional conductors. Phys Rev B. 1992;46: 1812.
- 438 7. Osada T, Kagoshima S, Miura N. Resonance effect in magnetotransport anisotropy of quasi-one 439 dimensional conductors. Phys Rev Lett. 1996;77: 5261.
- 440 8. Lebed AG, Bagmet NN. Nonanalytical magnetoresistance, the third angular effect, and a method to 441 investigate Fermi surfaces in quasi-two-dimensional conductors. Phys Rev B. 1997;55: 8654.
- 442 9. Yamaji K. On the angle dependence of the magnetoresistance in quasi-two-dimensional organic
 443 superconductors. J Phys Soc Jpn. 1989;58:1520.
- 444 10. Sasaki T, Toyota N, Tokumoto M, Kinoshita N, Anzai N. Transport properties of organic conductor
 445 (BEDT-TTF)₂KHg(SCN)₄: I. Resistance and magnetoresistance anomaly. Solid State Commun.
 446 1990;75:93.
- 447 11. Kinoshita N, Tokumoto M, Anzai N. Electron Spin Resonance and Electric Resistance Anomalyof
 448 (BEDT-TTF)₂RbHg(SCN)₄. J Phys Soc Jpn. 1991;60:2131.
- 449 12. Kusch ND, Buravov LV, Kartsovnik MV, Laukhin VN, Pesotskii SI, Shibaeva RP et al. Resistance
- and magnetoresistance anomaly in a new stable organic metal $(ET)_2TIHg(SCN)_4$. Synth Met. 1992;46:271.

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- 452 13. Mori H, Tanaka S, Oshima M, Saito G, Mori T, Murayama Y, Inokuchi H. Crystal and Electronic
 453 Structures of (BEDT–TTF)₂[MHg(SCN)₄](M=K and NH₄). Bull Chem Soc Jpn. 1990;63:2183.
- 454 14. Ducasse L, Fritsch A. Effect of basis set on the band structure and Fermi surface of BEDT-TTF-455 based organic conductors. Solid State Comm. 1994;91:201.
- 456 15. Qualls JS, Balicas L, Brooks JS, Harrison N, Montgomery LK, Tokumoto M. Competition between
 457 Pauli and orbital effects in a charge-density-wave system. Phys Rev B. 2000;62:10008.
- 458 16. Proust C, Audouard A, Kovalev A, Vignolles D, Kartsovnik M, Brossard L et al. Quantum oscillations and phase diagram of α -(BEDT-TTF)₂TIHg(SCN)₄. Phys Rev B. 2000;62:2388.
- 460 17. Kartsovnik MV. High Magnetic Fields: A Tool for Studying Electronic Properties of Layered 461 Organic Metals. Chem Rev. 2004;104:5737.
- 462 18. Kontorovich VM, Glushyuk AM. Transformation of sound and electromagnetic waves at the 463 boundary of a conductor in a magnetic field. JETP. 1962;14(4):852.
- 464 19. Kravchenko VYa. Electromagnetic excitation of sound in a metallic plate. JETP. 1968;27(5):801.
- 465 20. V. L. Fal'ko VL. Electromagnetic generation of sound in a metal plate in a perpendicular magnetic
 466 field. JETP. 1983;58(1):175.
- 467 21. Vasil'ev AN, Gaidukov YuP. Electromagnetic excitation of sound in metals. Soviet Physics
 468 Uspekhi. 1983;26(11):952.
- 469 22. Turner G, Thomas RL, Hsu D. Electromagnetic generation of ultrasonic shear waves in 470 potassium. Phys Rev B. 1971;3:3097.
- 471 23. Wallace WD, Gaerttner MR, Maxfield BW. Low-temperature electromagnetic generation of
 472 ultrasound in potassium. Phys Rev Lett. 1971;27:995.
- 473 24. Chimenti DE, Kukkonen CA, Maxfield BW. Nonlocal electromagnetic generation and detection of
 474 ultrasound in potassium. Phys Rev B. 1974;10:3228.
- 475 25. Chimenti DE. Nonlocal electromagnetic generation of acoustic waves in aluminum films. Phys Rev
 476 B. 1976;13:4245.
- 477 26. Banik NC, Overhauser AW. Electromagnetic generation of ultrasound in metals. Phys Rev B.478 1977;16:3379.
- 479 27. Kartheuser E, Rodriguez S. Deformation potentials and the electron-phonon interaction in metals.
 480 Phys Rev B. 1986;33:772.
- 481 28. Kaganov MI, Tsukernik VM. Effect of thermoelectric forces on the skin effect in a metal. JETP.
 482 1959;8:327.
- 483 29. Kaganov MI. Thermoelectric mechanism of electromagnetic-acoustic transformation. JETP.484 1990;71:1028.
- 485 30. Kaganov MI, Maallavi FM. Importance of Nernst effect in electromagneto-acoustic transformation.
 486 Low Temp Phys. 1992;18:737.
- 487 31. Vasil'ev AN, Maallavi FM, Kaganov MI. Thermoelastic stresses as a mechanism of 488 electromagnetic-acoustic transformation. Soviet Physics Uspekhi. 1993;36:968.
- 489 32. Krstovska D, Galbova O, Sandev T. Thermoelectric mechanism of electromagnetic-acoustic
 490 transformation in organic conductors. Europhys Lett. 2008;81:37006.
- 491 33. Krstovska, D. Ultrasonic wave generation in two-band organic conductors due to thermoelectric
 492 effect. Int J Mod Phys B. 2017;31: (*In press*).
- 493 34. Abrikosov AA. Fundametals of the theory of metals. 2nd ed. North-Holland: Amsterdam; 1988.
- 494 35. Kovalev AE, Hill S, Qualls JS. Determination of the fermi velocity by angle-dependent periodic
- 495 orbit resonance measurements in the organic conductor α -(BEDT-TTF)₂KHg(SCN)₄. Phys Rev B. 496 2002;66:134513.