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ELECTRONIC PROPERTIES OF SOLID

# THE INFLUENCE OF EXTERNAL PRESSURE ON THE BEHAVIOR OF THE METALLIC PHASE IN ORGANIC QUASI-TWO-DIMENSIONAL CONDUCTOR $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl. CONTRIBUTION OF CORRELATION EFFECTS


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**Abstract**. The quasi-two-dimensional organic metal $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl transfers to a Mott insulator state when cooled below T = 30 K. External hydrostatic pressure of $P > 0.7$ kbar restores the metallic state and enables the study of resistance, magnetoresistance, and Shubnikov–de Haas oscillations at helium temperatures in the external pressure range of $P$ = (1-8) kbar. The spectrum of observed Shubnikov–de Haas oscillations agrees well with theoretical calculations of the band structure. At the same time, the oscillation characteristics (cyclotron mass, frequency, amplitude) are significantly influenced by electronic correlations. Strongly correlated systems also exhibit specific temperature dependence of resistance. Pressure serves as the main tool controlling the strength of correlations. Various versions of pressure influence on the behavior of the nonoscillating part of magnetoresistance are discussed.

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## 1. INTRODUCTION

The organic low-dimensional metal of $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl (where BEDT-TTF — is bis(ethylenedithio)-tetrathiafulvalene) BEDT-TTF belongs to a well-known class of quasi-two-dimensional conductors, which are single-crystal samples of cation-radical salts, in which flat organic BEDT-TTF molecules form cationic layers alternating with inorganic anionic layers. During salt formation, an electron from every second BEDT-TTF molecule transfers to the anion. The remaining electrons can move between BEDT-TTF molecules, providing metallic conductivity within the layers, which is several orders of magnitude higher than the conductivity between layers [1,2]. This material has recently attracted increased research interest, primarily due to exotic quantum states of the electronic system at low temperatures. In particular, in $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl evidence has been found for the realization of dipole and spin liquid states [3-6]. The main reason for the emergence of such states lies in the peculiarities of the crystalline and electronic structures of the conducting layer. In organic metals with the general chemical formula (BEDT-TTF)$_2$X the conduction band is three-quarters filled. However, in metals with $\kappa$-type BEDT-TTF molecular packing, these molecules form dimers, and the conduction band splits into a fully filled and a half-filled band, i.e., metallic transfer occurs mainly through dimers [7]. Considering that in organic metals the conduction band is quite narrow, about 1000 K [8], in dimerized metals there is an instability towards transition to a Mott insulator state as temperature decreases. Such transition is determined by the ratio $U / W > 1$, where $U$ — is the Coulomb repulsion energy on one dimer, $W$ — is the electron kinetic energy in the conduction band, which includes the Fermi energy and temperature. In the classical case, below the transition temperature, electrons should localize on dimers, forming an ordered antiferromagnetic system. In the studied compound $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl a metal-insulator transition has been discovered at temperature $T \approx 30$ K [6]. However, in the conducting layer, dimers form a triangular frustrated lattice, which destroys long-range spin order and leads to the formation of a spin liquid and other quantum states at low temperatures, which were the main focus of research. At the same time, the metallic state that emerges under relatively small external hydrostatic pressure has not received sufficient attention so far.

According to the phase $T-P$-diagram [6], at pressure of $P \geqslant 0.7$ kbar, the metallic state of $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl is restored. Analysis of Shubnikov–de Haas (SdH) quantum oscillations observed at pressure $P = 0.7$ kbar (Fig.1) [9] showed fundamental frequencies $F_\alpha$ and $F_\beta$ and combination frequency $F_\beta - F_\alpha$, in the Fourier spectrum, which is consistent with theoretical calculations of the band structure and Fermi surface at room temperature [9,10]. The proposed work is devoted to studying the properties of the metallic phase $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl at various external hydrostatic pressures above 0.7 kbar, ensuring a stable state of such phase, and analysis of correlation effects in the electronic system.

## 2. EXPERIMENTAL RESULTS

Resistance and magnetoresistance were measured using the standard four-probe method with a lock-in amplifier on single crystal samples with characteristic dimensions of $0.5 \times 0.3 \times 0.03$ mm$^3$. The measuring current with a frequency of 20 Hz, not exceeding 10 μA, was directed perpendicular to the conducting layers of the crystal. The sample was

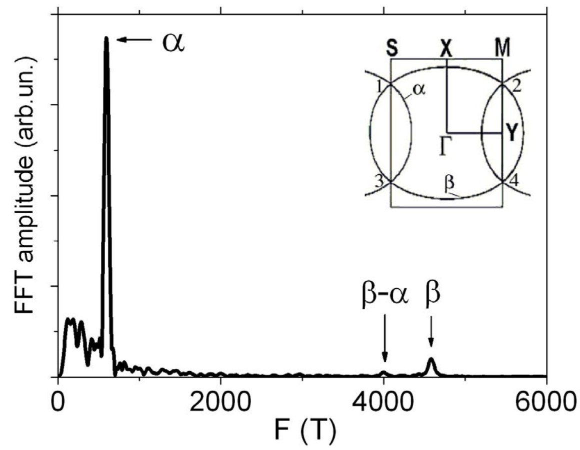


**Fig.1.** Fourier spectrum of SdH oscillations in $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl at $P = 0.7$ kbar, $T = 0.5$ K. Inset: schematic representation of the Fermi surface in the conducting plane [9]

mounted in a highpressure cell filled with silicone fluid, which provided quasi-hydrostatic conditions up to pressures of $P = 8$ kbar at low temperatures. Measurements were carried out in a pumped insert, allowing operation in the temperature range from room temperature to 0.5 K. The magnetic field $B$ up to 16.5 T was created by a superconducting solenoid and was directed normal to the conducting layers along the current direction.

Figure 2 shows the temperature dependencies of interlayer resistance in $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl as a function of external pressure. In the absence of pressure at room temperature, the resistance is $R \approx 130$ Ohm. The application of pressure reduces the resistance to $R \approx 30$ Ohm at $P = 8$ kbar. Temperature decrease leads to a monotonic reduction in resistance by approximately one and a half orders of magnitude at helium temperatures In the temperature range 2–15 at all pressures, the resistance is well described by the relation (Fig. 3)

$$R(T) = R_0 + AT^2. \quad (1)$$

The behavior of magnetoresistance as a function of pressure is shown in Fig. 4.

At minimum pressure $P = 1$ kbar, the field dependence curve of resistance has positive curvature and is well described by the relation

$$R(B) = R(0) + bB^n,$$

where $n \approx 2$, across the entire field range. Pressure increase leads to a decrease in $n$ at high fields, and

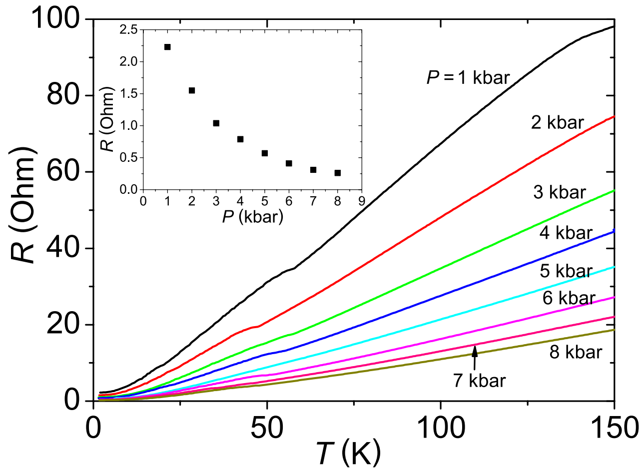


**Fig. 2.** Temperature dependencies of interlayer resistance at various pressures. Inset: resistance dependence at $T = 2$ K on pressure

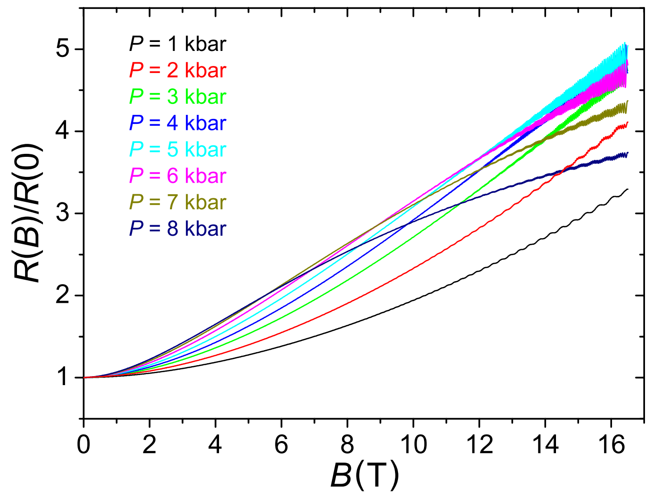


**Fig. 4.** Field dependencies of interlayer resistance at different pressures at $T = 0.5$ K. The field is directed perpendicular to the conducting layers

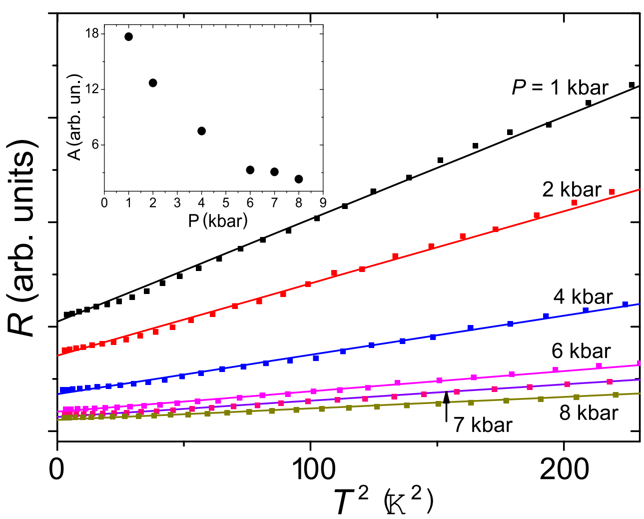


**Fig. 3.** Resistance dependencies on temperature squared at various pressures. Inset: dependence of coefficient $A$ in formula (1) on pressure temperatures

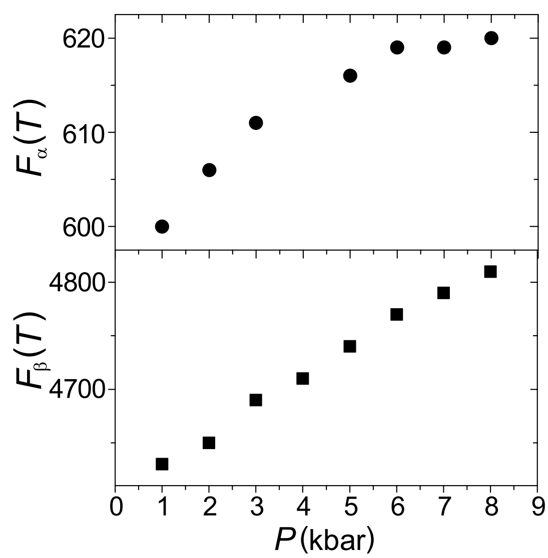


**Fig. 5.** Pressure dependencies of α- and $\beta$-SdH oscillation frequencies

at pressure $P \approx 5$ kbar, the power-law dependence degenerates into an almost linear one. At even higher pressures, the dependence acquires negative curvature in fields of $B > 5$ T. At all pressures in fields of $B > 9$ T, SdH quantum oscillations are observed, with an example of their Fourier spectrum shown in Fig. 1. The dependencies of frequencies $F_\alpha$ and $F_\beta$ and corresponding amplitudes of α- and $\beta$-SdH oscillations on applied pressure are shown in Figures 5 and 6. Both frequencies increase monotonically with pressure up to 8 kbar by approximately 3–4 %. In contrast, the pressure dependencies of amplitudes are non-monotonic and demonstrate nearly zero amplitude: for $\beta$-frequency at $P = 1$ kbar and for α-frequency at $P = 4$ kbar. Fig. 7 shows the pressure dependencies of cyclotron masses for α-and $\beta$-oscillations. The mass values were determined from temperature dependencies of the corresponding oscillation amplitudes. Both masses approximately linearly depend on inverse pressure (inset in Fig. 7) and decrease by 7–8 % when pressure increases to 8 kbar.

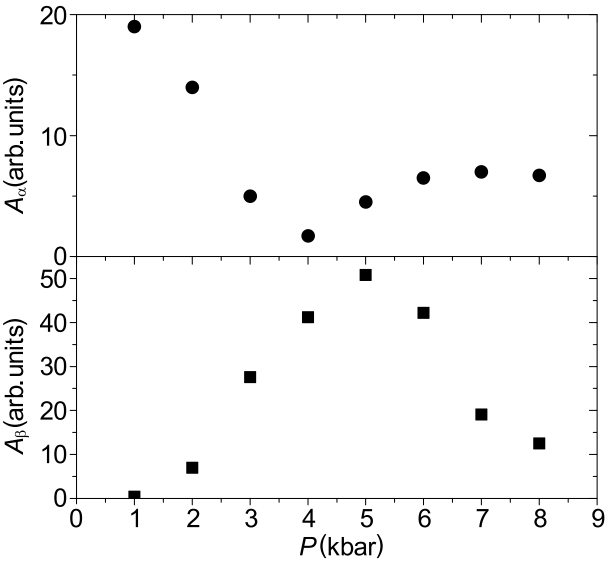


**Fig. 6.** Pressure dependence of α- and $\beta$-SdH oscillation amplitudes at $T = 0.5$ K $P = 4$ kbar

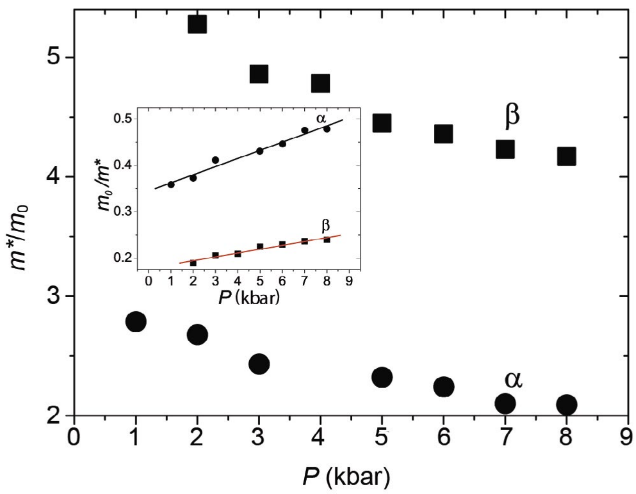


**Fig. 7.** Dependences of cyclotron masses of α- and $\beta$-ShdH oscillations on pressure. Inset: dependences of inverse cyclotron masses of α- and $\beta$-ShdH oscillations on pressure

## 3. DISCUSSION OF RESULTS

Temperature dependence of interlayer resistance in organic metal $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl shows metallic behavior at atmospheric pressure. At temperature $T \approx 30$ K a metal-insulator transition is observed, interpreted as a Mott-type transition [6]. Pressure of $P > 0.7$ kbar suppresses the transition, restores the metallic state, which allows observing the features of temperature dependences of resistance at various pressures down to helium temperatures (see Fig. 2). Such features include quadratic temperature dependence of resistance (1), characteristic of electron-electron scattering (see Fig. 3). The contribution of such scattering is usually small in normal metals, however in low-dimensional organic metals, where $U \sim W$, this phenomenon is quite common [11] and is one of the signs of strong electronic correlations. Application of external pressure weakens correlations, reducing coefficient $A$, mainly due to the increase in kinetic energy $W$, caused by hydrostatic compression of the crystal lattice. Pressure increase up to 8 kbar reduces A almost by an order of magnitude (see inset in Fig. 3).

### 3.1. Magnetoresistance (non-oscillating part)

The metallic state of the electronic system of compound $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl provides observation of magnetoresistance characteristic of strongly anisotropic quasi-twodimensional metals. Initially, one could expect a square root field dependence of longitudinal magnetoresistance in high fields, $\Delta R / R(0) \propto B^{1/2}$ [12], typical for most organic layered metals. However, at low pressures, a power-law dependence with exponent $n \approx 2$ is observed even in maximum available fields. Two scenarios seem reasonable to explain such behavior.

**First scenario.** Taking into account the combination of large longitudinal magnetoresistance, $\Delta R(16\ \mathrm{T}) / R(0) \sim 3$–5 see Fig. 4, and the "metallic" nature of the temperature dependence of resistance $R(T)$, it can be assumed that the interlayer charge transfer in $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl occurs in an incoherent mode through resonant impurities [13,14]. (It should be noted that important additional information about the interlayer transport mode can be obtained from angular dependencies of magnetoresistance [8], however, such studies represent a separate very complex task.) In this case, electron movement across layers also includes their movement along the conducting layers [14]. Conductivity along the layers has a quadratic dependence on the magnetic field perpendicular to the layers, which gives quadratic longitudinal magnetoresistance at least in fields for which $\omega\tau_c < 1$, where $\omega$ — is the cyclotron frequency, $\tau_c$ — is the electron lifetime between collisions. The application of external pressure reduces the effective mass of electrons (Fig. 7) and resistance (see inset in Fig. 2), probably also increasing $\tau_c$. This leads to an increase in $\omega\tau_c$ and, consequently, shifts the interval of quadratic magnetoresistance towards lower fields, which is observed in Fig. 4.

**Second scenario.** In this scenario, during electron tunneling between adjacent layers and $j+1$ with a low charge spreading rate along the layer, an exciton analog appears: there will be a hole on layer $j$ and an excess electron on layer $j+1$ — there will be Coulomb attraction between them with energy $E_c$. The magnetic field localizes electrons in the layer and slows down their spreading, which enhances their Coulomb interaction $E_c \sim e^2 / r$. The localization radius is of the order of the Larmor radius $r_L$ and is inversely proportional to the magnetic field $B$. This leads to strong magnetoresistance [15]. However, in metals with high conductivity, the charge spreading rate is so high that during the time $\tau \ll h / E_c$ ($h$ — is Planck's constant) such an exciton will decay, and this "Coulomb polaron" effect will be insignificant. The closer to the Mott transition, the lower the conductivity in the layer, and therefore, the longer the lifetime $\tau$ of such an exciton and its influence on interlayer conductivity by analogy with the effect studied in work [16]. Thus, in the metallic phase (at high pressure), this source of longitudinal magnetoresistance $\Delta R(B) / R(0)$ does not play a role, but at low pressure, when the exciton dissociation rate is not high enough, it can make a significant contribution.

### 3.2. Shubnikov–de Haas Oscillations

In fields of $B > 9$ T SdH oscillations were observed at all pressures. The oscillation spectrum at $P = 1$ kbar contains two fundamental frequencies, $F_\alpha \approx 600$ T and $F_\beta \approx 4630$ T (an example of the spectrum at $P = 0.7$ kbar is shown in Fig.1). It agrees well with theoretical calculations of the band structure and Fermi surface for layered organic metals with $\kappa$-type cation layer packing [11]. The initial hole $\beta$-orbit, encompassing 100 % of the first Brillouin zone (FBZ) (two electrons per unit cell), undergoes reflection when crossing the zone boundary, forming a second hole $\alpha$-orbit with an area of 13 % FBZ (see inset in Fig. 1). At the zone boundary intersection points 1, 2, 3, 4 between these orbits, a gap appears, which, generally speaking, should tend to zero due to the presence of an inversion center in the studied crystals [17]. However, the fact of coexistence of both SdH oscillation frequencies at all pressures indicates a non-zero magnitude of such gap. The most likely cause of this is spin-orbital interaction [17]. Thus, in a magnetic field, a magnetic breakthrough $\beta$-orbit with four magnetic breakthrough transitions and a closed $\alpha$-orbit with two reflections in the transition region are formed.

Fig. 7 shows the dependence of cyclotron masses $m_\alpha^*$ and $m_\beta^*$, corresponding to $\alpha$- and $\beta$-oscillations, on pressure. Pressure reduces the masses while maintaining their ratio $m_\beta^* / m_\alpha^* \approx 2$ in accordance with theoretical calculations [18]. The general expression for both cyclotron masses is well described by the relation

$$m^* \propto (P - P_0)^{-1}, \tag{2}$$

where $P_0 \approx -20$ kbar for both types of oscillations (inset in Fig. 7). This result can be compared with a similar theoretical relation for the effective mass in strongly correlated metals near the metal-insulator transition, taking into account the band mass renormalization due to electronic correlations [19,20]:

$$m^* \propto (1 - u / u_0)^{-1}. \tag{3}$$

Here $u = U / t$ — is the strength of electronic correlations, where $U$ — is the Coulomb repulsion of electrons on one dimer, $t$ — is the integral of electronic transfer between nearest dimers, $u_0$ — is the critical correlation strength at which the metal-insulator transition occurs. Relations (2) and (3) appear equivalent under the condition of linear dependence of correlation strength $u$ on pressure and weak pressure dependence of the electron-phonon interaction contribution to the cyclotron mass. Evidence of linear dependence $u(P)$ was presented in [20]. They were obtained in the study of organic metal $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. However, it should be noted that the pressure range in this work did not exceed 2 kbar. The pressure dependence of cyclotron mass renormalization due to electron-phonon interaction was evaluated in [21] during the study of organic metal $\kappa$-(BEDT-TTF)$_2$Cu(NCN)$_2$. Arguments are presented in favor of small pressure influence on the contribution of this interaction compared to the electron-electron contribution. Thus, there are reasons to believe that the decrease in cyclotron mass in $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl with increasing external pressure occurs mainly due to the weakening of electronic correlations.

Figure 5 shows the frequency dependencies of α- and β-oscillations on pressure. The frequencies increase monotonically at 3–4 % due to the reduction of the elementary crystal cell and, consequently, the increase in the FBZ area. However, there are minor qualitative and quantitative differences in the behavior of $F_\beta(P)$ and $F_\alpha(P)$. They may also be related to correlation effects. In paper [21], it is noted that electron-phonon interaction does not affect the orbit shape and the area size encompassed by it. At the same time, electron-electron interaction changes the shape while preserving the size. For β-orbit, the shape change does not affect the area size – it equals 100% of the FBZ area at all pressures and grows according to the changes in elementary cell dimensions. Conversely, the, α-orbit, being part of the β-orbit (see inset in Fig. 1), will slightly change its area size when the β-orbit shape changes and, accordingly, the frequency $F_\alpha(P)$ as correlations weaken with increasing external pressure. The pressure dependence of the SdH oscillation amplitude for α- and β-orbits is shown in Fig. 6. In the pressure range of $P = 1$–$8$ kbar, both amplitudes undergo radical changes, nearly vanishing. The expression for the magnetoresistance oscillation amplitude in a twodimensional electronic system has the form [22]

$$A \propto R_T R_D R_{MB} R_S. \tag{4}$$

In particular, for the β-orbit, the following relations can be used:

$$R_T \approx \frac{2C\mu^* T}{B}\exp\left(-\frac{C\mu^* T}{B}\right)$$

under the condition

$$C\mu^* T[\mathrm{K}] / B[\mathrm{T}] > 2,$$

where $C = 14.69$ [T/K], $\mu^* = m^* / m_0$ — reduced cyclotron mass, $m_0$ — free electron mass;

$$R_D = \exp\left(-\frac{C\mu^* T_D}{B}\right),$$

where $T_D$ — Dingle temperature;

$$R_{MB} = \exp\left(-2\frac{B_0}{B}\right)$$

considering the fourfold passage through the magnetic breakdown gap, where $B_0$ — magnetic breakdown field;

$$R_S = \cos\left(\pi\frac{g^* \mu_b^*}{2}\right),$$

where $g^*$ — $g$-factor, renormalized by electron-electron interaction, $\mu_b^*$ — cyclotron mass without electron-phonon contribution. Thus, expression (4) can be reduced for the β-orbit to expression

$$A \propto \frac{R_S}{B}\exp\left(-\frac{B^*}{B}\right), \tag{5}$$

where the effective field

$$B^* = C\mu^*(T + T_D) + 2B_0.$$

From this expression, it is evident that pressure can significantly affect the amplitude of β-oscillations by substantially changing the Dingle temperature and/or the magnetic breakdown field magnitude. Figure 8 shows the dependence of the effective field $B^*$ on pressure at $T = 0.5$ K. It is clearly visible that throughout the pressure range, the effective field changes insignificantly. Therefore, the effect of notable pressure impact on oscillation amplitude through changes in the magnetic breakdown gap magnitude, proportional to $B_0^{1/2}$, or Dingle temperature can likely be excluded. Thus, it can be considered that in relation (4), the first three factors do not participate in the observed significant changes in oscillation amplitude. The fourth factor

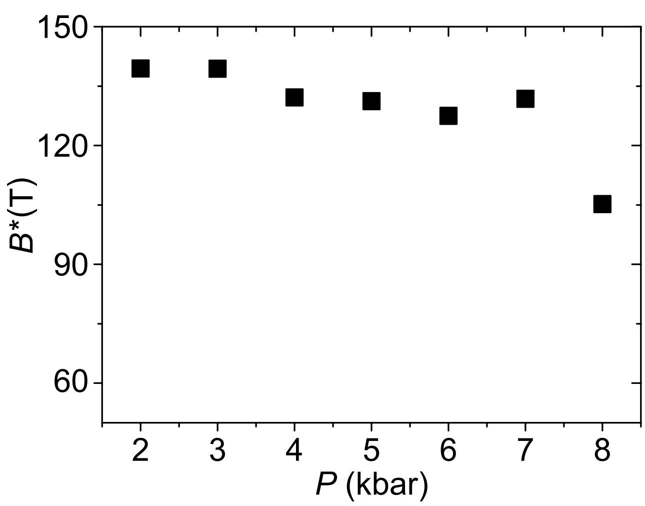


**Fig. 8.** The dependence of effective field on pressure at $T = 0.5$ K

remains $R_S$, the so-called spin reduction factor, related to the Zeeman splitting of Landau levels. Its magnitude directly depends on the cyclotron mass, and it periodically vanishes, forming so-called "spin zeros" under the condition

$$g^* \mu_b^* = 2n + 1,$$

where $n$ — integer number [22]. In all probability, this mechanism is responsible for significant changes in the amplitudes of α- and $\beta$-oscillations, arising from the weakening of electronic correlations.

Thus, it can be considered established that strong electronic correlations significantly influence the characteristics of quantum oscillations inherent in Fermi liquid, such as oscillation frequency, amplitude, and cyclotron mass. Support for this statement can be found in the analysis of the theoretical relationship obtained for strongly correlated systems [23],

$$\frac{A}{\gamma^2} \propto a, \qquad (6)$$

where $A$ — coefficient at $T^2$ in formula (1), γ — coefficient at the linear part of temperature dependence of heat capacity, $a$ — elementary crystal cell parameter. In a very rough approximation

$$\gamma \propto \frac{1}{E_F} \propto \frac{m^*}{F},$$

where $E_F$ — Fermi energy, and $F$ — oscillation frequency. Parameter $a$ is inversely proportional to the square root of oscillation frequency:

$$a \propto \frac{1}{F^{1/2}}.$$

Now expression (6) takes the form

$$A \propto \frac{m^{*2}}{F^{5/2}}.$$

Figure 9 shows the dependence of coefficient $A(P)$ on $Y(P) = m^{*2}(P) / F^{5/2}(P)$ for $\beta$-oscillations at different pressures, which satisfactorily agrees with the theoretical dependence (6) and serves as confirmation of the significant role of correlation effects in the metallic properties of organic metal $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl at suppressed Mott transition.

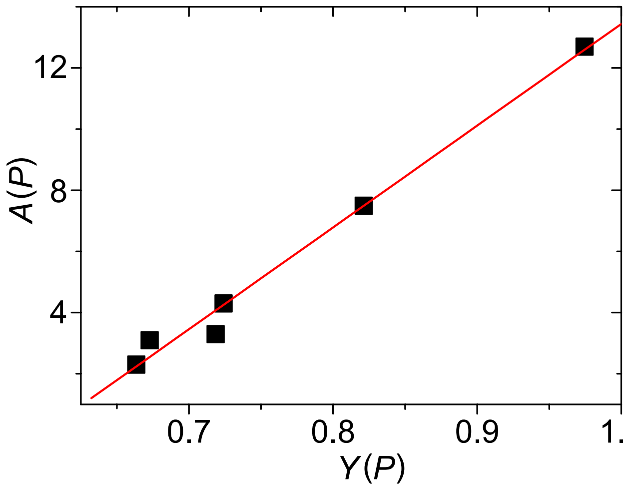


**Fig. 9.** Dependence of coefficient $A(P)$ on $Y(P) = m^{*2}(P) / F^{5/2}(P)$ for $\beta$-oscillations at different pressures. $A$ and $Y$ in arbitrary units

## 4. CONCLUSION

The organic quasi-two-dimensional metal $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl upon cooling and at normal pressure undergoes a Mott-type metal-insulator transition at temperature $T \approx 30$ K. Application of external pressure $P > 0.7$ kbar suppresses the metal-insulator transition, restoring the metallic state, which corresponds to theoretical calculations of the electronic structure at room temperature. This is confirmed by the observation of quantum oscillations in magnetoresistance. Analysis of the non-oscillating part of magnetoresistance showed that interlayer charge transfer occurs most likely in an incoherent regime and at low pressures can be additionally limited by the influence of polarons emerging in the magnetic field. The effective cyclotron mass decreases with increasing pressure mainly due to the weakening of correlation strength of interacting electrons. This decrease is reflected in the pressure dependence of oscillation amplitude, which demonstrates sharp minima – so-called spin zeros. The oscillation frequency dependence also suggests the involvement of correlation effects that weaken with increasing pressure. Thus, electron-electron interactions directly or indirectly have a significant impact on the main characteristics of SdH oscillations in the strongly correlated electronic system $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl .

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