

**Electronic-transport studies of the organic metals  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> and  $\alpha$ -(BETS)<sub>2</sub>KHg(SCN)<sub>4</sub> in high magnetic fields, where BETS is bis(ethylenedithio)tetraselenafulvalene**

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Electronic transport measurements in a magnetic field of the organic metals  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> and  $\alpha$ -(BETS)<sub>2</sub>KHg(SCN)<sub>4</sub> are presented. The parameters of the electron systems are estimated from experiments, and compared with both theoretical and measured values for analogous BEDT-TFF-based (ET) compounds. The results are interpreted as evidence of the influence of the electron-electron interactions on the properties of the ET and BETS families of organic conductors. [S0163-1829(97)09507-6]

### INTRODUCTION

The small crystallographic differences between the BETS- and the ET-based organic conductors and the very different low temperature properties of the members within the ET family motivate this study of the magnetic-field properties of  $\alpha$ -(BETS)<sub>2</sub>MHg(SCN)<sub>4</sub> ( $M$ =NH<sub>4</sub> and K) salts. We present experiments in high magnetic fields made on these organic metals, which were undertaken to gain further insights into how the increase in the bandwidth, resulting from replacing sulfur atoms by selenium, affects the properties of electron systems of BETS compounds.

Considerable attention has been devoted in recent years to the study of the mercury thiocyanate salts of bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET),  $\alpha$ -(ET)<sub>2</sub>MHg(SCN)<sub>4</sub> ( $M$ =NH<sub>4</sub>, Cs, Rb, K, Li, and Tl). The ammonium and potassium compounds have been examined most intensively.<sup>1,2</sup> Both salts are layered, quasi-two-dimensional (2D) electrical conductors. The ET radical-cation donor units are arranged in a herringbone packing motif ( $\alpha$ ), and form 2D conducting layers ( $ac$  plane) that alternate with sheets of insulating  $M\text{Hg}(\text{SCN})_4^-$  counterions.  $\alpha$ -(ET)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> ((ET)NH<sub>4</sub>) is a superconductor with a transition temperature  $T_c$  of 1.15 K (inductive onset).<sup>3</sup> The resistivity of  $\alpha$ -(ET)<sub>2</sub>KHg(SCN)<sub>4</sub> ((ET)K) decreases until at least 10 mK without reaching a superconducting state, although there has been a report of filamentary superconductivity in a few samples below 300 mK.<sup>4</sup> There is a kink in the temperature dependence of the resistivity of (ET)K near 8 K, signaling the onset of a magnetic phase transition, commonly associated with the formation of a spin-density-wave (SDW) state.<sup>1</sup> Anisotropic changes also occur in the magnetic susceptibility<sup>5</sup> and electron-spin resonance<sup>6</sup> at the transition temperature. Below 8 K the magnetoresistance increases dramatically, goes through a maximum, and has a negative slope at intermediate fields.<sup>1</sup> At higher fields an anomalous hysteretic ‘‘kink’’ occurs at about 22.5 T that is associated with the restoration of the metallic state.<sup>1</sup> Other noteworthy details of the magnetoresistance include the rich harmonic

content of the quantum oscillations,<sup>7</sup> especially in the low temperature phase, different behavior of the angle dependent magnetoresistance at constant fields below and above the kink transition,<sup>8</sup> and very deep and anharmonic dips at certain angles in both the field and angle dependence of the magnetoresistance.<sup>9</sup> The roles of various factors in forming the low-temperature magnetic state have been discussed, for example, nesting and reconstruction of the Fermi surface,<sup>10</sup> an increase of the resistivity of conducting sheets above the critical superconductor-insulator transition value,<sup>11</sup> and the formation of a magnetic superlattice in the layered  $b^*$  direction.<sup>12</sup>

Recently, mercury thiocyanate salts of bis(ethylenedithio)tetraselenafulvalene (BEDT-TSF or BETS),  $\alpha$ -(BETS)<sub>2</sub>MHg(SCN)<sub>4</sub> ( $M$ =NH<sub>4</sub> and K) have been synthesized that are isostructural with the corresponding  $\alpha$ -(ET)<sub>2</sub>MHg(SCN)<sub>4</sub> systems.<sup>13</sup> The crystal packing in the two series is remarkably similar. For a given  $M$ , the interplane donor distances and angles differ by no more than a few percent. In BETS the four inner tetrathiafulvalene sulfur atoms of ET are replaced with selenium atoms. The atomic radii of selenium atoms are 0.1 Å greater than those of sulfur atoms,<sup>14</sup> and the intermolecular transfer integrals of the selenium centers with other sites are somewhat larger than those of analogous sulfur atoms.<sup>15</sup> In spite of these modest differences, selenium-atom substitution has a profound effect on the electrical transport properties of the  $\alpha$ -(BETS)<sub>2</sub>MHg(SCN)<sub>4</sub> salts. The resistivities of both compounds decrease smoothly and monotonically to at least 0.4 K.<sup>13</sup>  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> is not a superconductor above this temperature, and there is no anomaly in the resistivity profile of the potassium salt. The band-structure calculations<sup>16</sup> for  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> and  $\alpha$ -(BETS)<sub>2</sub>KHg(SCN)<sub>4</sub> [hereafter called (BETS)NH<sub>4</sub> and (BETS)K] based on the extended Hückel tight-binding model predict a Fermi surface and a band structure similar to those of (ET)NH<sub>4</sub> and (ET)K. The bandwidths for the 2D part of the Fermi surface for BETS compounds are about 50% larger than the bandwidths for ET, and the BETS also has a larger warping of the 1D sheets.

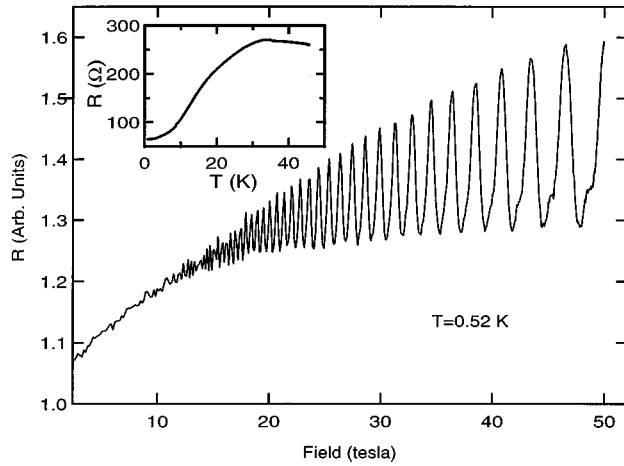


FIG. 1. The magnetoresistance of the (BETS)NH<sub>4</sub>Hg sample at  $T=0.52$  K and the magnetic field perpendicular to the most conducting layers. The inset shows part of the cooldown curve.

### EXPERIMENTAL DETAILS

The magnetoresistance of single-crystal samples of (BETS)NH<sub>4</sub> and (BETS)K was measured at temperatures between 0.4 and 4.2 K using the Clark University pulsed-magnetic-field apparatus with fields up to 50 T. The samples were platelets with average dimensions of about  $1 \times 0.5 \times 0.02$  mm<sup>3</sup>. Gold wires of 18- $\mu$ m diameter were attached to the samples with gold paste. The measurements were performed using a high frequency four-contact ac method at 832 kHz. A current of about 80  $\mu$ A was applied perpendicularly to the most conducting planes. A rotating probe was used to position the sample, with the conducting planes perpendicular to the magnetic field. To reduce eddy current heating effects, a plastic <sup>3</sup>He cryostat capable of reaching temperatures down to 400 mK is used in conjunction with the pulsed magnet. The magnet is driven by a 350-kJ capacitor bank, producing a magnetic field up to 50 T with a rise time of about 11 ms. 50-T pulses were performed once for each sample, at  $T=0.4$  K. To conserve the pulsed magnet resources, angular and temperature dependent data were taken in magnetic fields up to 35.5 T.

(BETS)NH<sub>4</sub>, unlike (ET)NH<sub>4</sub>, is metallic down to 0.4 K with a residual resistance ratio,  $\rho(300 \text{ K})/\rho(0 \text{ K})$ , of the order

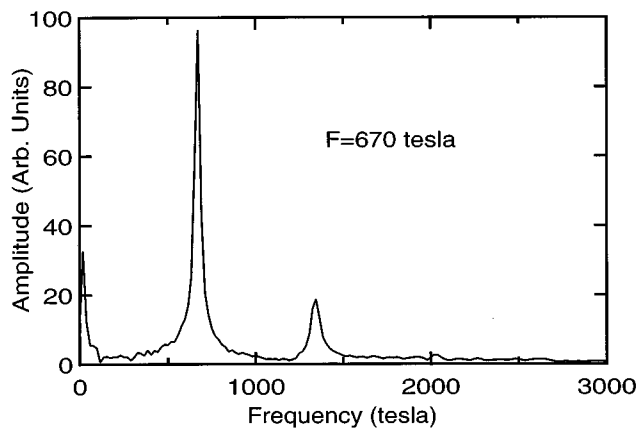


FIG. 2. The power spectrum of the data in Fig. 1.

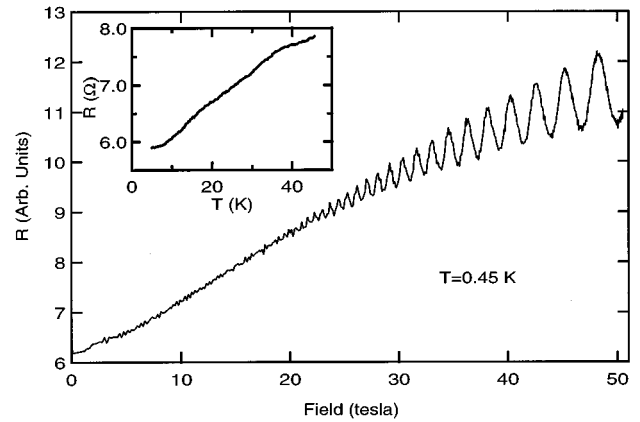


FIG. 3. The magnetoresistance of the (BETS)K sample at  $T=0.45$  K and the magnetic field perpendicular to the most conducting layers. The inset shows part of the cooldown curve.

of 10 and no indication of superconductivity. rf penetration depth measurements were performed, but failed to detect any sign of a superconducting transition down to 550 mK. Figure 1 shows resistance vs magnetic field for the (BETS)NH<sub>4</sub> sample; the inset in Fig. 1 demonstrates a part of the cooldown curve revealing metallic behavior. The power spectrum obtained from the resistance plotted as a function of the inverse field is shown in Fig. 2. The frequency of the fundamental harmonic is  $670 \pm 10$  T [15.6% of first Brillouin zone (FBZ)]; second and third harmonics are also resolved.

When cooled down to 0.4 K, (BETS)K does not undergo a phase transition to the spin-density-wave state that takes place in (ET)K. The inset in Fig. 3 shows a part of the cooldown curve having metallic character. Figure 3 represents the magnetoresistance up to 50 T at  $T=0.45$  K, showing the smooth background and the absence of the features characteristic of (ET)K (i.e., a large magnetoresistance with a maximum at about 10 T, a complex harmonic composition of Shubnikov-de Haas (SdH) oscillations, and a negative slope of the magnetoresistance sometimes associated with SDW state). Figure 4 shows the power spectrum where the frequency of the fundamental harmonic is  $684 \pm 10$  T; the second harmonic is also seen.

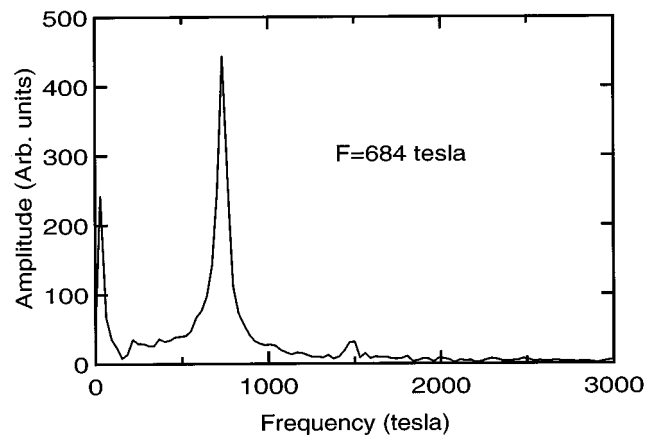


FIG. 4. The power spectrum of the data in Fig. 3.

## DISCUSSION

Because the Fermi surfaces for both compounds of this study are expected to have cylindrical parts, we adopt the approach of Harrison *et al.*<sup>17</sup> for the de Haas–van Alphen (dHvA) effect due to a 2D Fermi surface. Although they treated magnetization and we measure magnetoresistance, we simply modify their formula for the dHvA effect, noting that the oscillatory part of magnetoresistance is<sup>18</sup>  $\tilde{\rho} \propto \tilde{D} \propto B^2 (dM/dB)$ , where  $\tilde{D}$  is the density of states,  $B$  is the magnetic field, and  $M$  is the magnetization. Since the voltage in the detection coils measured in dHvA experiments is  $V \propto (dM/dB)(dB/dt)$ , the oscillatory part of the magnetoresistance is described by

$$\tilde{\rho} \propto \sum_p \alpha(p, B, T) \cos \left[ 2\pi p \left( \frac{F}{B} - \frac{1}{2} \right) \right], \quad (1)$$

with

$$\alpha(p, B, T) \propto p K_B \frac{F^2}{B} \frac{T}{\sinh X} R_D R_S, \quad (2)$$

where  $p$  is a harmonic number,  $K_B$  is the component of the reciprocal lattice parallel to  $B$ ,  $X = (2\pi^2 p k_B T) / (\hbar \omega_c) \cong 14.69 (m^*/m_e) (T/B)$ ,  $\omega_c = eB/m^*$ ,  $m^*$  is the effective mass, and  $R_D$  and  $R_S$  are the Dingle and spin-splitting factors in the Lifshitz-Kosevich (LK) formula.<sup>18</sup>

An important difference between Eqs. (1) and (2) and the LK formula<sup>18</sup> is that the power of  $B$  in Eq. (2) is  $-1$ ,

whereas in the LK formula it is  $\frac{1}{2}$  for the oscillatory behavior of the density of states and  $-\frac{1}{2}$  for the magnetization. Using the 2D formulas (1) and (2), we plot the measured amplitudes  $\alpha_m$  as  $\ln(\alpha_m B \sinh(X)/T)$  as a function of  $1/B$ . The Dingle temperatures for the (BETS)NH<sub>4</sub> and (BETS)K salts are found to be 2.8 and 3.8 K respectively.

In contrast, the procedure usually performed to determine the effective mass is the same for both the conventional LK formula describing 3D systems and the 2D model.<sup>17</sup> Considering the fact that the harmonic content of the oscillations observed in our experiments is simple, we fitted amplitudes of individual oscillations as a function of temperature to the usual LK term<sup>18</sup>  $R_T = X/\sinh X$ . Figure 5 shows the resulting effective mass as a function of field for both (BETS)NH<sub>4</sub> and (BETS)K. The effective masses appear to be independent of the magnetic field. The effective masses measured from SdH oscillations in dc magnetic fields may be somewhat larger than the pulsed field data presented here. For example, Singleton *et al.*<sup>19</sup> showed that SdH measurements carried out in pulsed fields can underestimate the effective mass due to carrier heating, with roughly a 10% underestimate observed for an (ET)K sample.

We summarize some parameters of the electron systems for BETS and analogous ET compounds in Tables I and II. We observe significant discrepancies between the calculated and measured cross-sectional area of the Fermi surface for (BETS)NH<sub>4</sub> and (ET)NH<sub>4</sub>. It is unlikely that this is due to the sample orientation, because the discrepancy between the calculated and measured values would require the conduct-

TABLE I. The in-plane parameters of (BETS)NH<sub>4</sub> and (ET)NH<sub>4</sub> electron systems.

	$\alpha$ -(BETS) <sub>2</sub> NH <sub>4</sub> Hg(SCN) <sub>4</sub>		$\alpha$ -(ET) <sub>2</sub> NH <sub>4</sub> Hg(SCN) <sub>4</sub>	
	Calculated <sup>a</sup>	Expt.	Calculated <sup>a</sup>	Expt.
$F, T$	387 (9% <sup>b</sup> )	670 (15.6% <sup>b</sup> )	413 (9.4% <sup>b</sup> )	570 <sup>c</sup> (14% <sup>b</sup> )
$E_F$ , meV	109		76	
$m^*/m_e$ <sup>d</sup>		1.05 ± 0.05		2.7 <sup>c</sup> (dHvA) 2.5 <sup>e</sup> (SdH)
$m_c/m_e$ <sup>f</sup>				1.35 <sup>g</sup>
$m_b/m_e$ <sup>h</sup>	0.62		0.89	
$\tau$ , ps <sup>i</sup>		0.43		1.2 <sup>c</sup>
$v_F$ (10 <sup>7</sup> cm/s) <sup>j</sup>	1.6		0.56	
$l$ (10 <sup>-8</sup> cm) <sup>k</sup>		690		670

<sup>a</sup>The theoretical values are estimated from the band structure data (Ref. 16).

<sup>b</sup>Percent of the area of the first Brillouin zone.

<sup>c</sup>J. Woznitza, G. W. Grabtree, H. H. Wang, U. Geiser, J. M. Williams, and K. D. Carlson, Phys. Rev. B **45**, 3018 (1992).

<sup>d</sup>The effective mass determined from magnetoresistance or magnetization data.

<sup>e</sup>Reference 25.

<sup>f</sup>The cyclotron mass.

<sup>g</sup>Reference 22.

<sup>h</sup>The band mass  $m_b$  at the Fermi level was determined from the calculated dispersion data for the cylindrical part of the Fermi surface using the equation  $m_b = 2m_{a^*}m_{c^*}/(m_{a^*} + m_{c^*})$ , where  $m_{a^*,c^*} = (\hbar^2/\partial^2 E_{a^*,c^*}/\partial k_{a^*,c^*}^2)$  [T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982)].

<sup>i</sup>The electron scattering time  $\tau = \hbar/2\pi k_B T_D$ , where  $T_D$  is the Dingle temperature.

<sup>j</sup>The Fermi velocity  $v_F = (\hbar/m^*)(S/\pi)^{1/2}$ , where  $S$  is the cross-sectional area of the Fermi surface perpendicular to the magnetic field.

<sup>k</sup>The mean free path  $l = v_F \tau$ .

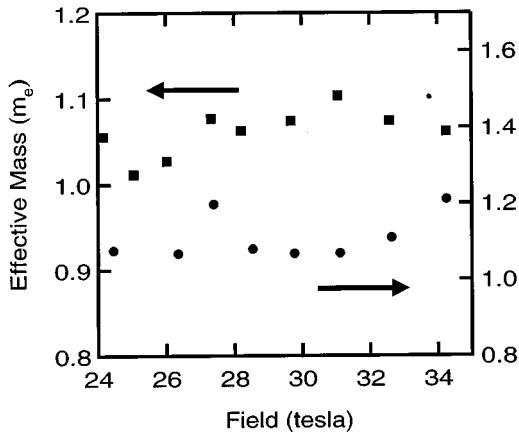


FIG. 5. Effective masses for the (BETS)NH<sub>4</sub> (squares) and (BETS)NH<sub>4</sub> (circles).

ing planes to be inclined at about 55° from the field direction. Moreover, we used a rotating platform with a precision of about ±2° to find the minimum in the SdH oscillation frequency, which corresponds to the conducting planes being perpendicular to the magnetic field.

The experimental effective mass for (BETS)NH<sub>4</sub> determined from the temperature dependence of the oscillations is about 1.05*m<sub>e</sub>* and is field independent up to 35 T (see Fig. 5). On the other hand, for (ET)NH<sub>4</sub>, the measured effective mass is significantly larger (see Table I). The calculated band mass for (BETS)NH<sub>4</sub> is also smaller than the band mass for (ET)NH<sub>4</sub>, which is a consequence of the larger overlap of the Se atoms.

The comparison of the effective masses of (BETS)K and (ET)K compounds gives similar results. The effective mass experimentally determined from the SdH oscillations in

(BETS)K is about 1.1*m<sub>e</sub>*. The effective mass for the (ET)K, determined at magnetic fields above the SDW phase, is about 2.5*m<sub>e</sub>*.

It was reported by Toyota *et al.*<sup>20</sup> that organic conductors with higher superconducting temperatures (*T<sub>c</sub>* > 7 K) tend to have higher effective masses (>2*m<sub>e</sub>*) and lower Dingle temperatures (<1 K). The authors ascribed the increase in the effective mass and the relaxation time  $\tau$  to renormalization due to electron-electron interactions. This hypothesis implies a correlation between the enhancement of electron-electron interactions and the enhancement of superconductivity and possibly other correlated electron effects in organic conductors. Furthermore, the mean free path *l* should be the same in systems with different electron-electron interactions strengths,<sup>20</sup> because the effective mass and the relaxation time are renormalized by the same factor  $l = v_F \tau \propto Z \tau / Z m_0 = l_0$ , where *Z* is the renormalization parameter,<sup>20</sup> see also the notes for Table I.

We found the effective masses for the BETS salts to be closer to the bare band masses than in the case of ET compounds. We also observe the absence of correlated electron effects such as superconductivity or SDW down to about 0.4 K in BETS. The fact that the mean free paths for BETS and ET compounds are very close (see Tables I and II), and the effective masses and transport properties in magnetic fields are different, seems to support the above hypothesis.<sup>20</sup>

We can assume that the measured effective mass is  $m^* = m_b(1 + \lambda)(1 + \alpha)$ , where *m<sub>b</sub>* is a bare band mass, and  $\lambda$  and  $\alpha$  characterize electron-phonon and electron-electron interactions. The parameter  $\lambda$  can be found from cyclotron-resonance experiments. Based on Kohn's theorem,<sup>21</sup> the resonant frequency for a parabolic band is independent of electron-electron interactions. The cyclotron resonance effective mass is then  $m_{cr} = m_b(1 + \lambda)$ . A recent experiment<sup>22</sup> gives  $m_{cr} \approx 1.35m_e$  for (ET)NH<sub>4</sub>. Given *m<sub>b</sub>*, we can then estimate  $\lambda \approx 0.52$  and  $\alpha \approx 0.85$ . Since the cyclotron-resonance experi-

TABLE II. The in-plane parameters of (BETS)K and (ET)K electron systems.

	$\alpha$ -(BETS) <sub>2</sub> KHg(SCN) <sub>4</sub>		$\alpha$ -(ET) <sub>2</sub> KHg(SCN) <sub>4</sub>	
	Calculated <sup>a</sup>	Expt.	Calculated <sup>a</sup>	Expt.
<i>F, T</i>	570 (13%)	684 (15.6%)	780 (19%) <sup>b</sup>	670 (16%) <sup>c</sup>
<i>E<sub>F</sub></i> , meV	185		122 <sup>b</sup> , 101 <sup>a</sup>	
<i>m<sup>*</sup>/m<sub>e</sub></i> <sup>d</sup>		1.1 ± 0.1		1.77 <sup>e</sup> (dHvA) 2.7 <sup>c</sup> (dHvA) 2.4 <sup>f</sup> (SdH) 0.94 <sup>f</sup>
<i>m<sub>c</sub>/m<sub>e</sub></i>				
<i>m<sub>b</sub>/m<sub>e</sub></i>	0.61		0.85	
$\tau$ , ps		0.32		5
<i>v<sub>F</sub></i> (10 <sup>7</sup> cm/s)	1.5		0.6	
<i>l</i> (10 <sup>-8</sup> cm)		480		300

<sup>a</sup>Using the data from Ref. 16.

<sup>b</sup>T. Mori, S. Tanaka, K. Oshima, M. Oshima, and G. Saito, J. Phys. Soc. Jpn. **59**, 2624 (1990).

<sup>c</sup>Reference 17.

<sup>d</sup>The effective mass for the (ET)K is given for the high-field phase because the Fermi surface inside the low-field phase is possibly different from the one at room temperature.

<sup>e</sup>Reference 2.

<sup>f</sup>Reference 25.

ments have not been done with (BETS)NH<sub>4</sub> or (BETS)K samples, we can only roughly estimate  $\lambda$  for the selenium-based compounds assuming that the change in  $\lambda$  between BETS and ET comes only from the different atomic masses of S and Se. Using the results of the BCS theory<sup>23</sup> [ $\exp(1/\lambda) \propto M^{0.5}$ ], where  $M$  is the ion mass, and the value of  $\lambda$  for (ET)NH<sub>4</sub>, we find  $\lambda \approx 0.41$  and  $\alpha \approx 0.21$  for (BETS)NH<sub>4</sub>.

We can estimate the effective Coulomb correlation energy  $U_{\text{eff}}$ , for example, for (ET)NH<sub>4</sub> and (BETS)NH<sub>4</sub> salts by employing the work of Brinkman and Rice,<sup>24</sup> as was done previously by Singleton *et al.*<sup>25</sup> for (ET)<sub>2</sub>Cu(NCS)<sub>2</sub>. Using the results of the band-structure calculations,<sup>16</sup> we find  $U_{\text{eff}} \approx 0.21$  eV for (ET)NH<sub>4</sub> and  $U_{\text{eff}} \approx 0.18$  eV for (BETS)NH<sub>4</sub>. Assuming that the kinetic energy of the electrons  $E_{\text{kin}} \approx E_F$ , we find that the ratio  $U_{\text{eff}}/E_{\text{kin}}$  is about 2.8 for (ET)NH<sub>4</sub> and about 1.6 for (BETS)NH<sub>4</sub>. If our estimation is correct, then we can qualitatively understand the drastic difference between BETS and ET compounds as coming from  $U_{\text{eff}}$  being much closer to  $E_{\text{kin}}$  in BETS than in ET.

The information we extracted from the SdH oscillations and the preceding discussion is concerned with the 2D portion of the Fermi surface (FS). There also is an influence of the 1D part of the FS on, in particular, the formation of the correlated electron phases. Band-structure calculations<sup>16</sup> predict a warping for the open parts of the FS for BETS compounds to be larger than for ET. Larger warping decreases the temperature of the SDW transition and at some value of the transfer integral perpendicular to the most conducting direction, the transition is completely removed.<sup>26</sup> The fact

that our experimental data do not show any indication of the SDW transition supports the band structure results.<sup>16</sup>

## SUMMARY

We have presented an observation of Shubnikov-de Haas oscillations in the organic conductors  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> and  $\alpha$ -(BETS)<sub>2</sub>KHg(SCN)<sub>4</sub>. Both compounds show behavior in magnetic fields that is different from the isostructural ET materials. Unlike its ET analog,  $\alpha$ -(BETS)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub> does not become superconducting at temperatures down to 0.4 K. The magnetoresistance of  $\alpha$ -(BETS)<sub>2</sub>KHg(SCN)<sub>4</sub> does not show the behavior characteristic of  $\alpha$ -(ET)<sub>2</sub>KHg(SCN)<sub>4</sub>. The probable reason for the difference is the larger bandwidth of the BETS compounds which results in a smaller effective Coulomb correlation energy.

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