Control of electronic properties of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K, NH$_4$) by the uniaxial strain method

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Systematic uniaxial strain studies of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K,NH$_4$) have been carried out. By controlling the in-plane lattice parameters a and c independently, we find that both compounds have essentially the same phase diagram in electronic properties, although, under the hydrostatic pressures, they show quite different phase diagrams. The K salt shows the superconductivity like NH$_4$ salt under the c-axis uniaxial strain, and the latter undergoes a transition to the density-wave state like the former under the a-axis uniaxial strain. These changes in electronic structures are verified by measuring the angle-dependent magnetoresistance. The electronic properties of these compounds can be systematically understood as a function of c/a, the ratio of the in-plane lattice parameters.

I. INTRODUCTION

An isostructural series of compound α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K,Rb,Tl,NH$_4$), where BEDT-TTF denotes bis(ethylenedithio)tertrathiafulvalene, has attracted much attention due to the variety of intriguing electronic properties. This system has a highly two-dimensional electronic structure, since donor molecular conducting planes are sandwiched by thick insulating anion layers. The chevronlike donor molecular arrangement within the conducting plane leads to a unique electronic structure. The Fermi surface is composed of both a two-dimensional (2D) cylinder and a pair of one-dimensional (1D) sheets as predicted by the tight-binding band structure calculation.[1,2]

Among them α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K,Rb,Tl) shows a transition to density-wave (DW) states at about 10 K.[3–5] The temperature dependence of the resistivity of these compounds shows a steplike anomaly at the transition temperature T_{DW}, and the angle-dependent magnetoresistance oscillation (AMRO) pattern dramatically changes below T_{DW}, suggesting the reconstruction of the Fermi surface.[6] It is generally considered that this transition originates in the nesting instability of the one-dimensional part of the Fermi surface.[7] Under the hydrostatic pressure the DW state is suppressed and the normal metallic properties are observed. In spite of a number of experimental and theoretical studies, the exact nature of the ground state remains to be discussed. On the other hand, the NH$_4$ salt shows superconductivity below about 1 K at ambient pressure.[8] The superconductivity was suppressed under the hydrostatic pressure, and the normal metallic state is stabilized without the transition to the DW state. Although these compounds are isostructural and their lattice parameters are almost the same, only the NH$_4$ salt shows superconductivity at ambient pressure.[9] It is still an open question what causes the difference in their electrical properties.

In order to examine the role of anisotropy in the different electronic properties and to search for novel electronic states, we performed a systematic uniaxial strain study on the isostructural compounds of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K,NH$_4$) having different low-temperature ground states. By controlling the in-plane lattice parameters, we found that both compounds have essentially the same phase diagram in electronic properties. It is possible to understand the electronic properties of this series of compounds as a function of c/a, the ratio of the in-plane lattice parameters in the conducting plane.

II. EXPERIMENT

The basic idea of the uniaxial strain method is to compress a sample in a hard cylinder as suppressing the expansion perpendicular to the applied force due to Poisson effects. Details of the experimental technique have been described in our previous paper.[10] We used a clamp-type Be-Cu pressure cell for this purpose. A sample crystal to be studied was embedded in organic materials such as the frozen-oil or epoxy resin. We used the Demnum S-20 as the oil and the Stycast 1266 as the epoxy resin. The sample composite was inserted in the pressure cell and then an external force was applied to a piston put on the sample composite. The cell cylinder hinders Poisson's effect when the cell is hard enough compared to the sample composite. For the frozen-oil method, the pressure cell was first cooled down to low temperatures so as to freeze the oil. Then the external force was applied at 20 K. The applied pressure denoted below indicates the pressure applied on the cross-sectional area of the rod of the sample composite. The maximum pressure applied was 10 kbar, which creates the strain of about -4%, although the exact value of the strain is uncertain at the present stage.[11] The lowest temperature reached was 0.6 K.

Single crystals of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ ($M=$K,NH$_4$) were prepared by standard electrochemical oxidation method. The typical size of investigated single crystals was $0.5\times0.5\times0.1$ mm^3. Their crystallographic axes were
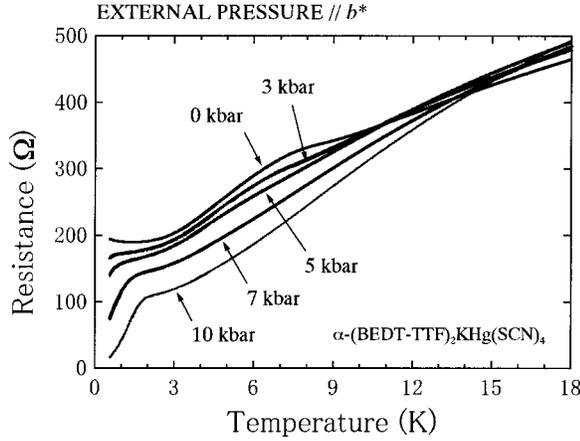
FIG. 1. The temperature dependence of the out-of plane resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under the uniaxial strain along the b^* axis, normal to the conducting ac plane. The external pressure was applied at 20 K using the frozen-oil method.

identified by x-ray diffraction measurements. The electrical resistance measurements under the uniaxial strain were performed by the conventional four-probe dc method. Four gold wires are attached on both sides of crystal surface (ac plane) with carbon paste as electrodes. We used the frozen-oil method to systematically study the effect of the uniaxial strain on the title compounds except for the AMRO study, where the epoxy method was employed. The out-of plane resistance of α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ (M=K,NH$_4$) was measured under the uniaxial strain created along the a, b^*, and c axes independently.

III. EXPERIMENTAL RESULTS

A. Effect of the b^*-axis uniaxial strain

Figure 1 shows the temperature dependence of the out-of plane resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under the uniaxial strain along the b^* axis, normal to the conducting ac plane. The pressure was applied at 20 K. Under the zero strain the steplike anomaly of the resistance was observed around 8 K, indicating the onset of the transition to the DW state. The DW state was suppressed by the b^*-axis strain and its transition temperature T_{DW}, which is defined as the temperature at which the derivative dR/dT shows a minimum, decreasing with an increase of the applied pressure. Above 7 kbar (strain of about -2.8%) we observed no sign of resistance anomaly associated with the DW transition.

The suppression of the DW state is attributable to the suppression of the nesting instability of the Fermi surface due to the increase in the warping of the 1D part of the Fermi surface. The external force parallel to the b^* axis causes not only the increase in the interplane bandwidth but also the change in the intraplane band structure, possibly leading to the increase in the Fermi surface warping. It is to be noted that the change in the intraplane band structure is possible because the donor molecular long axis is not parallel to the b^* axis. Rotations of molecules might be caused by the b^*-axis strain.
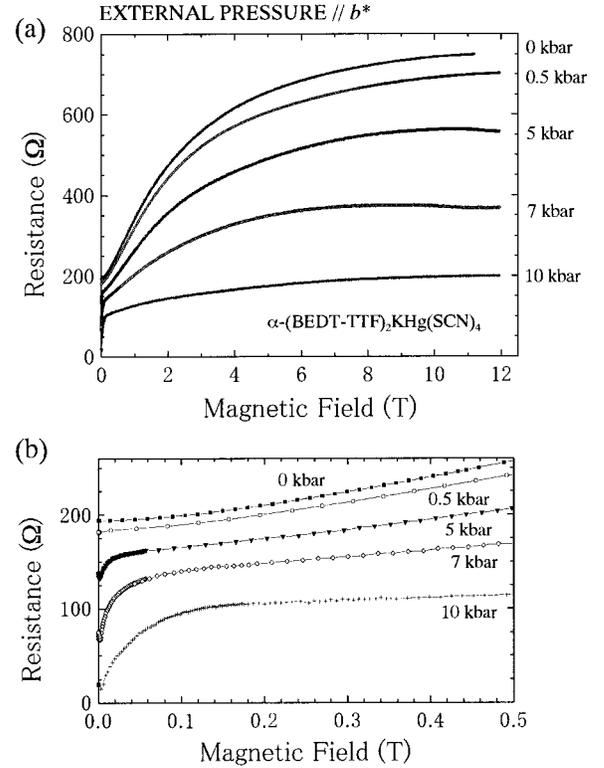

FIG. 2. (a) The magnetic-field dependence of the out-of plane magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under the b^*-axis uniaxial strain at 0.6 K. The magnetic field was applied along the b^* axis. (b) The magnetoresistance at low magnetic field.

We also found a sharp drop in the low-temperature resistance under high pressures above 5 kbar (strain of about -2%), suggesting the onset of the superconducting transition. Figure 2(a) shows the magnetic-field dependence of the out-of plane magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under the b^*-axis uniaxial strain. The large magnetoresistance characteristic of the DW state was observed at ambient pressure.[12] As the pressure was increased, the magnetoresistance became small, followed by a sharp drop of the low-field magnetoresistance above 5 kbar, indicating the superconducting critical field behavior as shown in Fig. 2(b). Both the superconducting critical field H_C and the transition temperature T_C continued to increase with increasing external pressure. At the maximum pressure of 10 kbar the magnetoresistance shows the almost sublinear magnetic field dependence, suggesting the resurrection of the original Fermi surface as predicted by the band calculation and observed in the NH$_4$ compound. We note that no Shubnikov–de Haas (SdH) oscillations were observed up to 12 T in the present crystal, presumably due to the poor quality of the crystal.

Figure 3 shows the temperature dependence of the out-of plane resistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the b^*-axis uniaxial strain. In contrast to α-(BEDT-TTF)$_2$KHg(SCN)$_4$, α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ showed a superconducting transition at 1.5 K at ambient pressure. We found that the b^*-axis uniaxial strain gradually increases the T_C. At the maximum applied pressure of 10
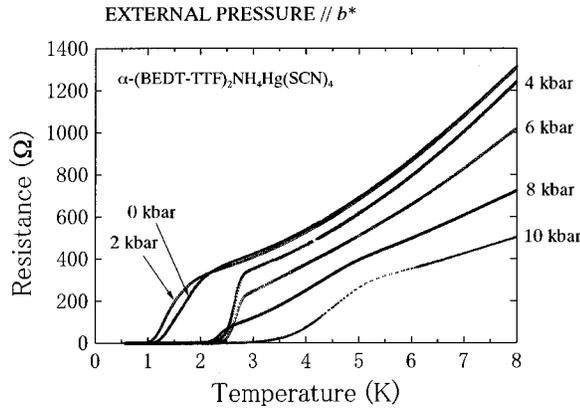
FIG. 3. The temperature dependence of the out-of-plane resistance of α-$(BEDT\text{-}TTF)_2NH_4Hg(SCN)_4$ under the uniaxial strain along the b^* axis, normal to the conducting ac plane. The external pressure was applied at 20 K using the frozen-oil method.

kbar, the T_C reached 4.5 K, which is three times higher than that at ambient pressure. Figure 4 shows the magnetic-field dependence of the magnetoresistance under the b^*-axis uniaxial strain. In this compound, the SdH oscillation superimposed on the smooth background magnetoresistance was clearly observed. Although the oscillation amplitude decreased rapidly with increasing strain, the SdH oscillation was still observable at the maximum applied pressure of 10 kbar (strain of about -4%). Figure 5(a) shows the pressure-dependent Fourier-transform (FT) spectra, and the pressure dependence of the (SdH) frequency is given in Fig. 5(b). Under the zero strain we observed the single fundamental frequency of 572 ± 26 T, which is in good agreement with the previous SdH and de Haas–van Alphen (dHvA) measurements.[13,14] This oscillation is ascribed to the 2D part of the Fermi surface.

We found that the SdH frequency is almost independent of pressure or slightly increased at the rate of $+3$ T/kbar with the b^*-axis uniaxial strain. It has been reported that the SdH frequency of this compound increases at the rate of $+17$
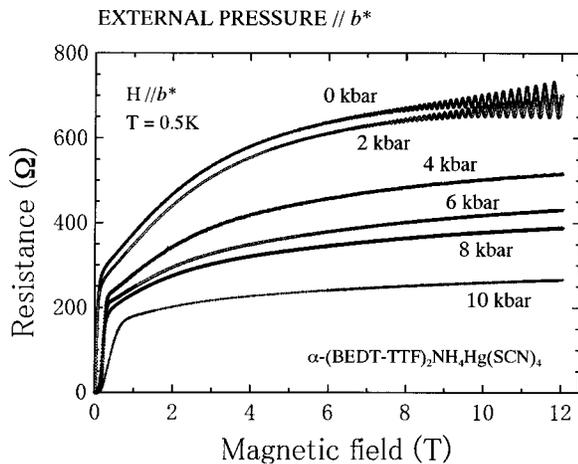

FIG. 4. The magnetic-field dependence of the magnetoresistance of α-$(BEDT\text{-}TTF)_2NH_4Hg(SCN)_4$ under the b^*-axis uniaxial strain at 0.6 K. The magnetic field was applied along the b^* axis.
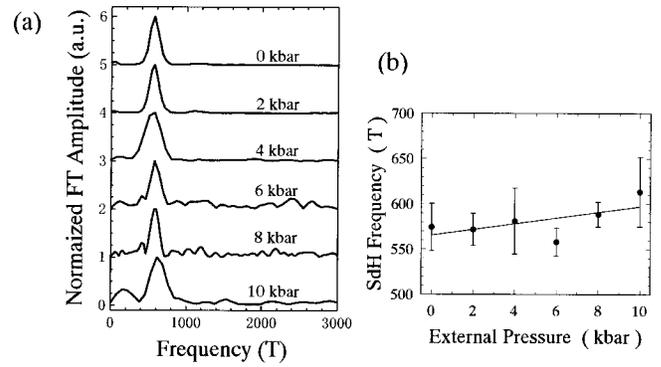

FIG. 5. The pressure dependence of the (a) Fourier-transform (FT) spectra and the (b) SdH frequency. The solid line is a guide for the eye.

T/kbar under the hydrostatic pressure,[15] and decreases at the rate of -40 T/kbar under the b^*-axis uniaxial *stress*.[16] Since the fundamental frequency is proportional to the cross-sectional area of the 2D Fermi surface and, therefore, to the first approximation, is proportional to the size of the first Brillouin zone, the different pressure dependence of the fundamental frequency is consistent with the different nature of the applied pressures. Hydrostatic pressure compresses the unit cell, leading to the increase in the area of the first Brillouin zone. On the other hand, the b^*-axis uniaxial *stress* expands the in-plane lattice parameters due to Poisson effects. Therefore the area of the first Brillouin zone decreases. The much smaller dependence of the SdH frequency on pressure in the present study suggests that the strain created in the crystal has a uniaxial nature. This ascertains that Poisson effects is basically suppressed. The small dependence on pressure is ascribed to either the incomplete suppression of Poisson effects or some changes in the intraplane band structure or both.

Campos *et al.* have found that the uniaxial stress applied perpendicular to the conducting plane induces and enhances the superconductivity in the K compound and NH_4 compound, respectively.[16] They also found a systematic trend in the increase of T_C with the real space expansion in the ac plane. In the present uniaxial *strain* study, the expansion in the ac plane is negligible. Nevertheless, the superconductivity was also enhanced. Therefore, we have to consider other factors to account for the large enhancement of the superconductivity under the uniaxial strain/stress in these compounds. The increase in the interplane bandwidth and/or the modification of intraplane band structure should be carefully considered for more quantitative discussions.

B. Effect of the in-plane c-axis and a-axis uniaxial strain

Drastic changes in the electronic properties were observed under the uniaxial strain within the conducting planes. Electronic properties of these compounds are largely dominated by the direction of the in-plane uniaxial strain.

Figure 6(a) shows the temperature dependence of the out-of plane resistance of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$ under the uniaxial strain along the c axis. The DW state was gradually suppressed with increasing strain and the superconduc-
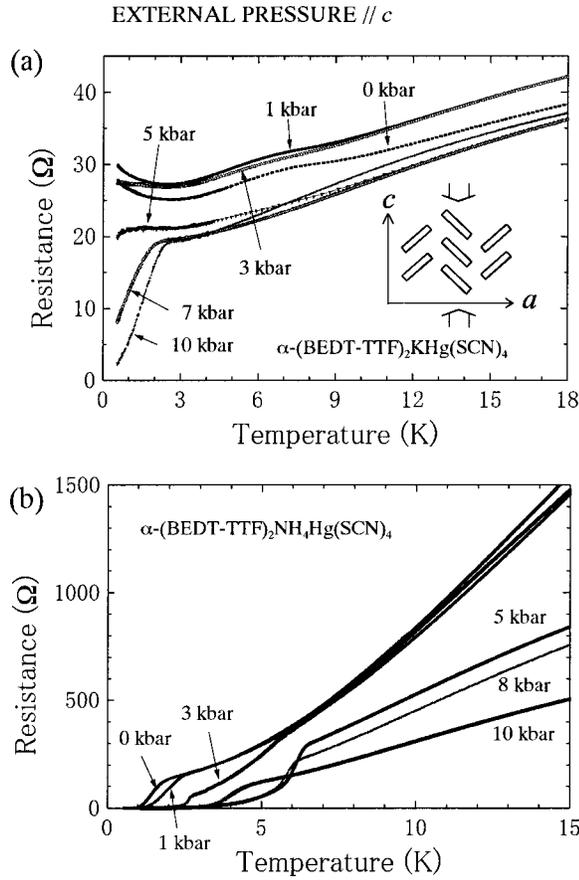
EXTERNAL PRESSURE // c

EXTERNAL PRESSURE // a

FIG. 6. The temperature dependence of the out-of-plane resistance of (a) α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and (b) α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the uniaxial strain along the c axis. The external pressure was applied at 20 K using the frozen-oil method. The inset in (a) shows the donor layer structure viewed along the molecular long axis. The arrows denote the direction of the strain.

FIG. 7. The temperature dependence of the out-of-plane resistance of (a) α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and (b) α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the uniaxial strain along the a axis. The external pressure was applied at 20 K using the frozen-oil method. The inset in (a) shows the donor layer structure viewed along the molecular long axis. The arrows denote the direction of the strain.

tivity was induced under the large strain. Although T_C continued to increase up to the maximum pressure of 10 kbar (strain of about -4%), the resistance never reached zero within the temperature range investigated, presumably due to the low T_C and the broad transition temperature width. The latter suggests the existence of the inhomogeneous strain within the crystal. Overall features under the c-axis uniaxial strain were similar to those under the b^*-axis uniaxial strain.

In case of α-(BEDT-TTF)$_2$NH$_4$(SCN)$_4$), the effect of the c-axis uniaxial strain was dramatic. The superconducting transition temperature T_C initially increased up to about 6 K, which is four times higher than the one at ambient pressure, by the application of the c-axis uniaxial strain of about -2% (external pressure of 5 kbar), as shown in Fig. 6(b). At the higher applied pressures above 5 kbar, T_C turned to decrease with further increasing the pressure. These results in both compounds suggest that the c-axis compression stabilizes the superconducting state. It has been reported that among the series of the isostructural compounds α-(BEDT-TTF)$_2 M$Hg(XCN)$_4$ (M=K,Rb,NH$_4$; X=S,Se), only the superconducting NH$_4$Hg(SCN)$_4$ salt shows a significant decrease in the ratio of the in-plane lattice constant c/a
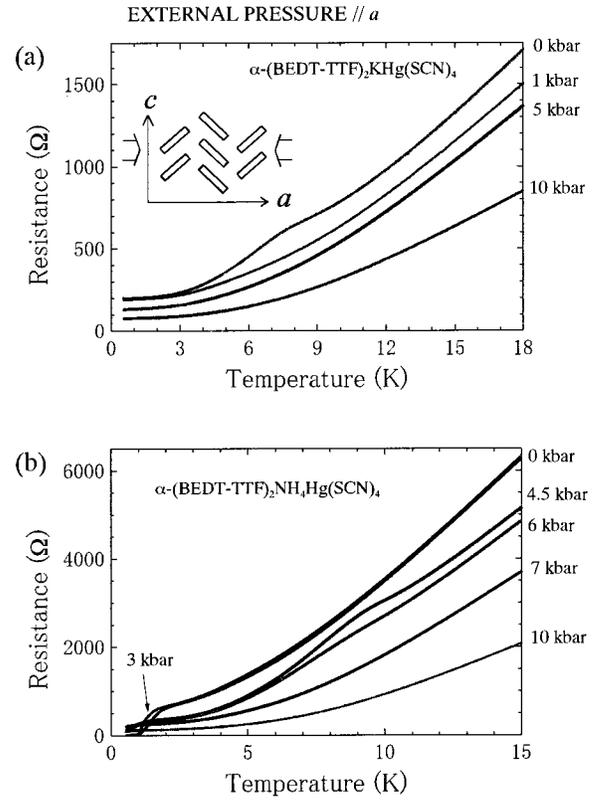
below about 200 K at ambient pressure.[17] We found, in accordance with the anisotropic thermal contraction, the reduction in c plays a dominant role in the appearance of the superconductivity.

The effect of the in-plane a-axis uniaxial strain was completely different from that of the in-plane c-axis uniaxial strain. Figure 7(a) depicts the temperature dependence of the out-of plane resistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under the a-axis uniaxial strain. The DW state was entirely suppressed under the external pressure of 1 kbar (strain of about -0.4%), and the normal metallic behavior was observed down to 0.6 K and up to 10 kbar. Under the a-axis strain the critical pressure to suppress the DW state was less than 1 kbar, which is so small compared to that under the hydrostatic pressure, 5 kbar. It implies that the hydrostatic pressure effectively compresses the sample along the a axis, the highest conducting axis.

As shown in Fig. 7(b), the superconducting transition temperature T_c is lowered by the in-plane a-axis uniaxial strain in α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ and at above 4.5 kbar (strain of about -1.8%) the resistance does not reach zero down to 0.6 K. At the high external pressures above 7 kbar (strain of about -2.8%), this compound showed the normal metallic temperature dependence of the resistance down to 0.6 K, while in the intermediate external pressure
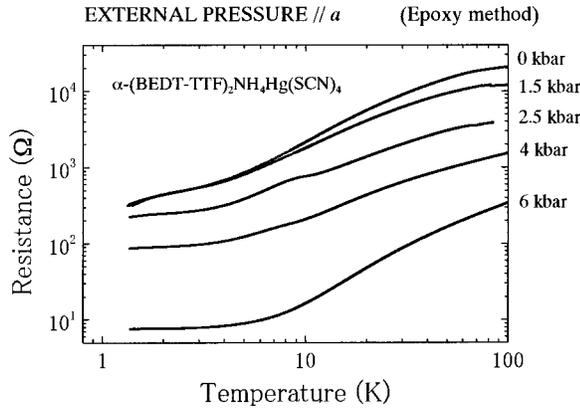
FIG. 8. The temperature dependence of the out-of-plane resistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the a-axis uniaxial strain. The external pressure was applied at room temperature using the epoxy-crystal method.

range, 4–6 kbar (strain of about -1.6% to -1.8%), the step-like anomalies were observed at about 10 K in the temperature dependence of the resistance. These anomalies appeared at the intermediate pressures are reminiscent of the DW transition observed in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. In order to investigate this low-temperature state more precisely, we performed the AMRO study of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the a-axis uniaxial strain. We employed the epoxy method to uniaxially compress the crystalline sample, since it is feasible for the rotation of the sample in the magnetic field because of the absence of the force-transmitting system from outside.

Figure 8 shows the temperature dependence of the out-of-plane resistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the a-axis uniaxial strain by use of the epoxy method. The pressure was applied at room temperature. The resistance anomaly at 10 K was clearly observed at the external pressure of 2.5 kbar, which corresponds to about 5 kbar in the frozen-oil method,[10] and it disappeared above 6 kbar. Overall feature in the epoxy method is essentially the same as that in the frozen-oil method.

Figure 9 shows the angle-dependent magnetoresistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ on the magnetic field direction under the external pressure of 2.5 kbar along the a axis. The sample crystal was rotated around the crystallographic a axis and the c' axis, normal to the ab^* plane, in the magnetic field. The field angles ω and χ were measured from the ab^* and the b^*c' plane, respectively. Magnetoresistance was collected as a function of χ (or ω) for different offset angle $\omega_{\rm offset}$ (or $\chi_{\rm offset}$). We note that the angle χ (or ω) is regarded as the polar angle θ only when the other one ω (or χ) is just equal to zero. In such conditions, the positions of the dips in the magnetoresistance were found to be periodic in $\tan\theta$ ($\theta=\chi$ or ω) without the phase shift between $\theta>0$ and $\theta<0$, as shown in the inset of Fig. 10. These features are characteristic of the AMRO due to the quasi-one-dimensional Fermi surface. We also found the large anisotropy in the AMRO depending on the offset angle $\omega_{\rm offset}$. It means that the AMRO pattern strongly depends on the direction of the azimuthal angle ϕ, which is measured from the c
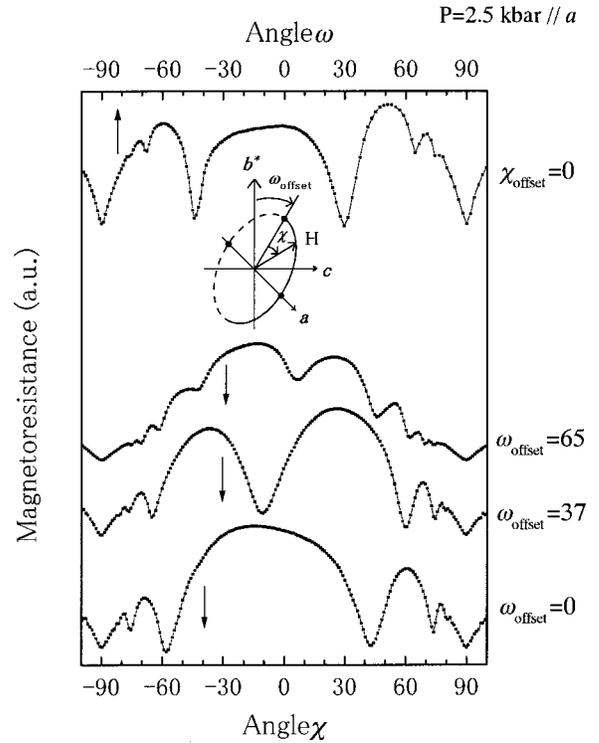

FIG. 9. The angle-dependent magnetoresistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ on the magnetic field direction at 1.5 K under the external pressure of 2.5 kbar along the a axis. The field angles ω and χ were measured from the ab^* plane and the b^*c' plane, respectively.
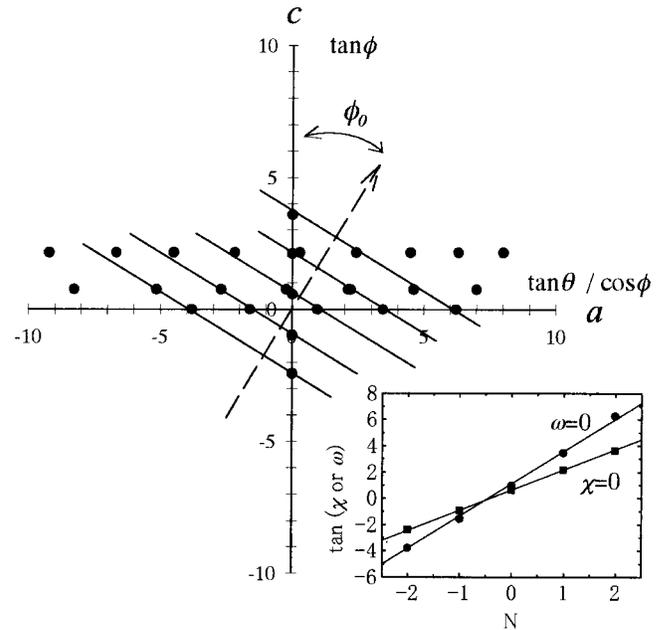

FIG. 10. The positions of the dips in the magnetoresistance plotted in polar coordinates for $X=\tan\theta\sin\phi$ and $Y=\tan\theta\sin\phi$, which corresponds to $X=\tan\chi$ and $Y=\tan\omega/\cos\chi$, respectively. The inset shows the positions of the dips in the magnetoresistance against $\tan\theta$.
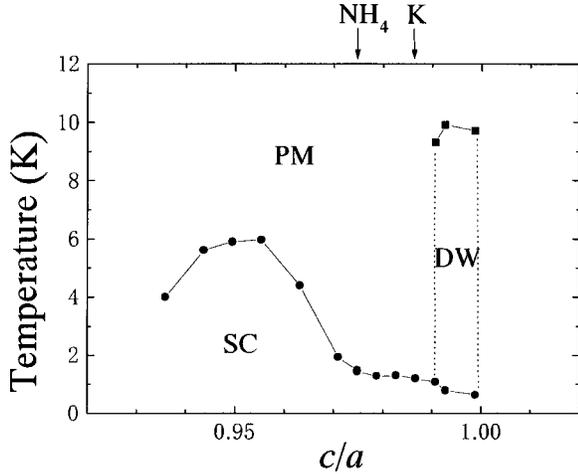
FIG. 11. The phase diagram of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$. The strain at 10 kbar is estimated to be about -4% in the frozen-oil method, although the exact value is uncertain at the present stage. The arrow indicates the value of c/a in the NH$_4$ and K compounds at ambient pressure.

axis. In Fig. 10 the positions of the dips were plotted in polar coordinates for $X = \tan\theta\sin\phi$ and $Y = \tan\theta\sin\phi$, which corresponds to $X = \tan\chi$ and $Y = \tan\omega/\cos\chi$, respectively. One observes the regularity in the positions of the dips. The solid lines in Fig. 10 can be described as

$$\tan\theta\sin(\phi + \phi_0) = 1.30N + 0.5,$$

where $\phi_0 = 31°$ and $N = -2, -1, 0, 1, 2$. The periodicity of the dip position against the polar angle has the minimum at $\phi = \phi_0$ and has the $1/\sin(\phi + \phi_0)$ dependence. Hence, these periodic dips in the AMRO are ascribed to the quasi-one-dimensional Fermi surface running parallel to the direction tilted with $\phi_0 = 31°$ from the c axis. This reconstructed Fermi surface of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ under the a-axis uniaxial strain was found to be very similar to that of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at ambient pressure.[18,19] Therefore we conclude that the DW state, just like the K salt, is induced in the NH$_4$ salt under the a-axis uniaxial strain.

IV. DISCUSSION

From these systematic uniaxial strain studies, we derived the phase diagram of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ as shown in Fig. 11. Here, the strain at 10 kbar is estimated to be about -4%, although the exact value is uncertain at the present stage.[11] The phase diagram of the K salt is essentially the same as that of the NH$_4$ salt. Their electronic properties were found to be largely dominated by the direction of the uniaxial strain created in the conducting plane. The low-temperature ground state of these isostructural compounds varies from the metallic state through the density-wave states to the superconducting state with decreasing the ratio of the in-plane lattice constant c/a.

It should be mentioned that the in-plane uniaxial *stress* study of the K salt was carried out by Brooks *et al.*[20] recently. They observed the destruction of the DW for both in-plane stress directions but no superconducting transitions.

The suppression of the DW by the a-axis stress was moderate compared to that by the c-axis stress. These results are in contrast to those of the present uniaxial *strain* study. In their method the Poisson effects are present. This makes it difficult to discriminate between the effects of the compression of the sample parallel to the stress and those of the expansion along the perpendicular direction. In contrast, the present uniaxial strain method excludes the Poisson effects. Therefore, our method is preferable for the discussion on the relationship between the changes in electronic properties and that of the crystal structure.

In α-(BEDT-TTF)$_2$MHg(SCN)$_4$ ($M =$ K,NH$_4$) the intercolumnar ($\|a$) interaction is stronger than the intracolumnar ($\|c$) one within the conducting donor layer, leading to the one-dimensional open Fermi surface perpendicular to the a axis. Therefore, naively speaking, the reduction of a, the lattice constant along the intercolumnar direction, makes the system more one-dimensional, and the reduction of the c, the lattice constant along the intercolumnar direction, makes the system more two-dimensional. This is consistent with the suppression of the DW state in the K salt by the c-axis uniaxial strain and the inducement of the DW state in the NH$_4$ salt by the a-axis uniaxial strain. Due to the anisotropic thermal contraction, the electronic structure of the NH$_4$ salt is considered to be more 2D than that of the K salt at ambient pressure, leading to the absence of the DW transition in the former salt. The present study strongly suggests that the small difference in the c/a, that is to say, the in-plane anisotropy, is the origin of the difference in their electronic properties at low temperatures.

However, it remains an open question why the DW state is suppressed by further a-axis uniaxial strain and there appears to be no superconductivity in the temperature range we studied. The above consideration is too naive to discuss the topology of the Fermi surface, since the donor molecules have a triangular network and do not stack uniformly within a column. Therefore, we need the exact arrangement of the molecules to precisely discuss the electronic structure and the topology of the Fermi surface under the uniaxial strain. In addition, we should consider other effects such as the reduction in the density of states to account for the above results. It has been theoretically pointed out that the increase in the bandwidth along the 1D direction reduces the density of states at the Fermi level, leading to the suppression of the SDW state in the quasi-1D organic conductors, (TMTSF)$_2$X, where TMTSF is tetramethyltetraselenafulvalene.[21] A similar explanation is considered to be possible in the present system.

Another point to be noted is the inducement and the enhancement of the superconductivity in the K salt and NH$_4$ salt, respectively, by the reduction of c and also the inter-plane distance. In the highly anisotropic materials such as (TMTSF)$_2$X, the hydrostatic pressure sometimes removes the nesting instability of the Fermi surface and induces the superconductivity. In these cases, however, the superconducting transition temperature T_C usually starts to decrease with further increasing pressure. The suppression of the superconductivity is generally ascribed to the decrease in the density of states, because the bandwidth is usually increased by the hydrostatic pressure due to the decrease in the unit-

cell volume. In the present uniaxial strain study the unit-cell volume is also reduced; however, the T_C continued to increase to some extent. Therefore, the increase in T_C cannot be explained by the simple cell volume effect. It is to be noted that the uniaxial strain possibly causes not only the translations of molecule parallel to the external force but also some changes in the molecular orientation. In the α-type BEDT-TTF salts, both the in-plane a and c axes are tilted from donor molecular plane and are also not perpendicular to that. Therefore, the compression along the a and c axes possibly causes the changes in the dihedral angle θ_d between neighboring molecules. As is suggested by Mori *et al.*, the overlap integral of the two inclined BEDT-TTF molecules strongly depends on θ_d.[22] The c-axis uniaxial strain may increase θ_d, leading to the decrease in the intercolumnar transfer integral in the present compounds. Such change in θ_d may increase the T_C, because, naively speaking, the bandwidth is largely dominated by the intercolumnar transfer integrals.

Moreover, we cannot rule out a possible change in the molecular dimerization under the uniaxial strain. In the α-type BEDT-TTF salts, donor molecules do not stack uniformly within a column and there exists a weak dimerization. The degree of the dimerization is the largest in NH_4 salt among the series of the present compounds,[23] and it might be changed by the uniaxial strain. The strong dimerization separates the bands and, therefore, reduces the overlap between the two bands that across the Fermi level, leading to the increase in the density of states at the Fermi level in the 1D part of the band, while that in the 2D part of the band unchanged. The former may account for the increase in the T_C. In order to discuss the band structure, the geometry of the Fermi surface, the molecular dimerization, the density of states at the Fermi level and the role of the Coulomb interaction, further investigations such as AMRO and the x-ray diffraction are needed.

V. CONCLUSION

We succeeded to artificially control the electronic properties of α-(BEDT-TTF)$_2 M$Hg(SCN)$_4$ ($M =$K, NH$_4$) by the uniaxial strain method. Both compounds showed very similar electronic properties to each other by controlling the in-plane lattice parameters a and c independently. The reduction in c/a enhanced the superconductivity, while the increase in c/a stabilized the metallic state. The density-wave state appeared between them. The electronic properties of these compounds can be systematically understood as a function of c/a, the ratio of the in-plane lattice parameters. Because of the anisotropic thermal contraction, the c/a of the NH$_4$ salt is smaller than that of the others with $M =$K,Rb,Tl at low temperatures at ambient pressure. This accounts for the reason why only the NH$_4$ salt shows the superconductivity, while the others undergo the transition to the density-wave state at ambient pressure. For more quantitative studies, however, we have to clarify the exact arrangement of atoms or molecules under uniaxial strain. We are preparing an x-ray diffraction apparatus for this purpose.

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