THERMAL EXPANSION AND STEPSWISE SUPERCONDUCTING TRANSITION OF 
\( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{1-x}(\text{AuI}_2)_x \)

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The temperature dependence of the lattice parameters and
electrical resistivities of \( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{1-x}(\text{AuI}_2)_x \) were examined.
A stepwise resistivity drop similar to that of so-called "high-T\( c\)"
and "low-T\( c\)" transitions of \( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 \) was observed in \( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{1}(\text{AuI}_2)_{0.9} \). But the periodical lattice modulation
wave characteristic to \( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 \) system could not be observed down to
98 K. On the other hand, no superconducting behavior was obtained
down to 1.5 K in \( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{0.9}(\text{AuI}_2)_{0.1} \), whose X-ray
examination gave strong satellite reflections indicating
the development of the periodical lattice modulation. The anomalous
thermal contraction of the lattice constant \( b \) was observed in
\( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 \) and \( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{0.9}(\text{AuI}_2)_{0.1} \) around 190 K, which
may be attributable to the interaction between the ethylene groups and
the periodically distorted \( \text{I}_3 \) arrays.

Recently, much interest has been focused on the salient superconducting properties of an
isounit series of \( \beta-(\text{BEDT-TTF})_{2}\alpha \) (BEDT-TTF) (C\( _{n}\)H\( _{10}\)S\( _n \)) = bis(ethylenedithio)tetrathia-
fulvalene, \( \alpha=\text{I}_3, \text{AuI}_2, \text{IBr}_2 \). Compared with the first organic superconductor, (TMTSF)\( _2\)X(\( \text{X} = \text{CIO}_4, \text{PF}_6 , \ldots \)), the \( \beta-(\text{BEDT-TTF})_{2}\alpha \) systems have
relatively high transition temperatures(\( T_c \)), especially for the so-called "high-\( T_c \)" state of
\( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 (\text{I}_3 \approx 7 \text{ K}) \). The transition temperatures of the \( \beta-(\text{BEDT-TTF})_{2}\alpha \) systems seem to be correlated with the unit cell dimensions.
The coexistence of the high-\( T_c \) and low-\( T_c \) states has been regarded as a unique property of
\( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 \).\(^2\) The incommensurate lattice
modulation wave developed below 200 K\(^3\) is considered to play an important role in
determining the superconducting transition temperature of \( \beta-(\text{BEDT-TTF})_{2}\text{I}_3 \), which is
extremely sensitive to the condition of the resistivity measurements (pressure, heating (or
cooling) cycles, etc.,\( \ldots \)).\(^4\)

In order to contribute to a better understanding of the puzzling superconducting behaviors of the \( \beta-(\text{BEDT-TTF})_{2}\alpha \) systems, we have examined the temperature dependence of the
lattice constants and the electrical resistivity of \( \beta-(\text{BEDT-TTF})_{2}(\text{I}_3)_{1-x}(\text{AuI}_2)_x \).

EXPERIMENTAL

The crystals were prepared electrochemically. The tetrahydrofuran solution of the
mixed supporting electrolyte of \( \{(\text{n-C}_4\text{H}_9\text{Hg})_4\text{N}\}\text{I}_3 \) and \( \{(\text{n-C}_4\text{H}_9\text{Hg})_4\text{N}\}\text{AuI}_2 \) was used. The dominant
phase of the obtained crystals was usually that of well-known \( \alpha-(\text{BEDT-TTF})_{2}\text{I}_3 \).\(^5\) But when almost
pure electrolyte of \( \{(\text{n-C}_4\text{H}_9\text{Hg})_4\text{N}\}\text{AuI}_2 \) was used, the main product was \( \beta-(\text{BEDT-TTF})_{2}\text{AuI}_2 \).\(^6\) Besides
\( \alpha- \) and \( \beta- \) phases, the \( \beta- \) phase, the \( \theta- \) phase, whose superconducting properties have been
reported recently,\(^8\) and \( \gamma- \) phase\(^9\) were obtained. It should be noted that the crystal types could
be hardly identified from the shape of the crystal because the crystal habit was strongly
dependent on the condition of the preparation. For example, the crystals of \( \alpha- \) type salts were
obtained usually as thin plates but sometimes we obtained them as blocks or needles. The
stoichiometries (x-values) were determined by X-ray microanalyses (XMA) of S, I, Au and Br
atoms, which revealed an unexpected existence of very small amount of Br. \( (\text{n-C}_4\text{H}_9\text{Hg})_4\text{N}\text{AuI}_2 \) was prepared from \( \{(\text{n-C}_4\text{H}_9\text{Hg})_4\text{N}\}\text{AuBr}_2 \) and used
without recrystallization to avoid the decomposition. Therefore, the contamination of the
Br atoms is considered to be originated from
the impurity of \((n-C_4H_9)N_4)AuBr_2\) in \((n-C_4H_9)N_4)AuI_2\)

The lattice constants of \(\beta-(BEDT-TTF)_2(I_3)_{1-x}(AuI_2)_x\) were measured on a Rigaku four-circle diffractometer between 300 K and 95 K. The lattice constants at room temperature are listed in Table 1 (see Fig. 1a and Ref. 10). The absolute values of the obtained lattice constants could not be considered to be accurate enough, but their temperature dependences appeared to be determined satisfactorily. They are given in Fig. 2. The X-ray oscillation photographs were taken at room temperature and 90 K for \(\beta-(BEDT-TTF)_2(I_3)_{0.9}(AuI_2)_0.1\) (Fig. 3) and for \(\beta-(BEDT-TTF)_2(I_3)_{0.1}(AuI_2)_0.9\). The electrical resistivities were measured over the temperature range of 300 K-1.5 K (Figs. 4 and 5).

Since the large crystal \((3 \times 2 \times 1.5 \text{ mm}^3)\) was obtained, X-ray diffraction experiments, XMA and electrical resistivity measurements of \(\beta-(BEDT-TTF)_2(I_3)_{0.9}(AuI_2)_0.1\) were made on the different portion of the same crystal. On the other hand, these experiments on \(\beta-(BEDT-TTF)_2(I_3)_{0.1}(AuI_2)_0.9\) were made on the different samples selected from the \(\beta\)-type crystals obtained in the same batch.

Table 1. The lattice parameters of \(\beta-(BEDT-TTF)_2(I_3)_{1-x}(AuI_2)_x\) at 298 K.\(^{10}\)

<table>
<thead>
<tr>
<th>(x)</th>
<th>0.0</th>
<th>0.1</th>
<th>0.9</th>
<th>1.0</th>
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<tr>
<td>(a(\AA))</td>
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<td>15.245</td>
<td>15.289</td>
<td>15.312</td>
</tr>
<tr>
<td>(b)</td>
<td>9.093</td>
<td>9.055</td>
<td>8.981</td>
<td>8.970</td>
</tr>
<tr>
<td>(c)</td>
<td>6.606</td>
<td>6.591</td>
<td>6.579</td>
<td>6.573</td>
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<tr>
<td>(\alpha(\degree))</td>
<td>109.89</td>
<td>109.86</td>
<td>110.60</td>
<td>110.75</td>
</tr>
<tr>
<td>(\beta)</td>
<td>95.60</td>
<td>95.57</td>
<td>95.96</td>
<td>96.15</td>
</tr>
<tr>
<td>(\gamma)</td>
<td>94.34</td>
<td>94.43</td>
<td>94.93</td>
<td>95.00</td>
</tr>
<tr>
<td>(V(\AA^3))</td>
<td>853.0</td>
<td>845.7</td>
<td>833.7</td>
<td>831.8</td>
</tr>
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</table>

RESULTS AND DISCUSSIONS

The anomalous temperature dependences of the lattice constants of \(\beta-(BEDT-TTF)_2I_3\) were observed for \(x, a, (\beta, \gamma)\) between 180 K and 210 K. As mentioned before, it has been reported that the incommensurate structural modulation is developed below 195 K.\(^{3}\) Therefore, these anomalies suggest that an onset of the periodical lattice distortion is accompanied by the anisotropic thermal contraction of the crystal lattice. Relatively smooth changes of \(a, \gamma\) and show that the molecular arrangement in \((010)\) is not affected greatly by the appearance of the modulation wave. Although the shortest \(I\cdots I\) contact (4.20 \(\AA\)) is that along [011] (the other \(I\cdots I\) contacts are longer than 4.50 \(\AA\) (see Fig. 1b)), the thermal contraction anomaly of the lattice spacing could be observed only in \(b\). The crystal structure of \(\beta-(BEDT-TTF)_2I_3\) shows that the magnitude of the lattice constant \(\gamma\) is determined by the "side-by-side contacts" of the \(BEDT-TTF\) molecules and that of \(a\) is determined by the cation-anion contacts along the long axis of the \(BEDT-TTF\) molecule. While the lattice spacing \(b\) is determined by the face-to-face and oblique contacts between \(BEDT-TTF\) molecules (see Fig. 1b). It is well-known that the ethylene groups of \(BEDT-TTF\) have a strong tendency for being disordered. The large thermal ellipsoids of one of the two ethylene groups (Fig. 1) suggest a positional disorder (or large thermal motion). Unlike other \(\beta\)-type salts such as \(\beta-(BEDT-TTF)_2AuI_2\) and \(\beta-(BEDT-TTF)_2IBr_2\), \(\beta-(BEDT-TTF)_2I_3\) appear to have a considerable degree of positional disorder even at low temperature, because the characteristic close \(I\cdots I\) contacts pointed out by Williams et al.\(^{3}\) will give rise to the freezing of the disordered conformation. In fact, Williams et al. have shown that each ethylene group takes one of the two possible conformations at low temperatures\(^{3}\) and that the existence of incommensurate lattice distortion appears to introduce "partially" ordering of the ethylene group.\(^{12}\) Thus, the disorderness remains to exist at low temperatures.

X-Ray crystal structure analyses of the \(BEDT-TTF\) compounds have shown that thermal
Figure 2.
Figure 2. Temperature dependence of the lattice constants of $\beta$-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$.

Figure 3. X-Ray oscillation photographs around c of $\beta$-(BEDT-TTF)$_2$(I$_3$)$_{0.9}$(AuI$_2$)$_{0.1}$. Strong satellite reflections are observable at 98 K.
(a) 298 K  (b) 98 K

Figure 4. Resistivities of $\beta$-(BEDT-TTF)$_2$(I$_3$)$_{0.9}$ (AuI$_2$)$_{0.1}$.
motion of the terminal ethylene groups tends to be extremely large, as mentioned above. This is due to the steric effect of the ethylene substituent, which makes it difficult for BEDT-TTF to take stable molecular conformation in the crystal. In many BEDT-TTF compounds, one of the two ethylene groups has very large thermal parameters indicating its positional disorder (and/or thermal motion). Usually, this disorder becomes small with decreasing temperature. The room-temperature and low-temperature crystal structure analyses of α-(BEDT-TTF)$_2$PF$_6$ show that the disordered conformation of ethylene group transforms to ordered one above 100 K. The room-temperature structure of α-(BEDT-TTF)$_2$PF$_6$ has a disordered conformation of the ethylene group quite similar to that of β-(BEDT-TTF)$_2$I$_3$. But the structure analysis at 99 K gave the completely ordered conformation (see Fig. 6). Probably similar conformational change of ethylene group will occur in the β-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$ systems, if there is no strong I-...H interaction.

The disorder of ethylene group is a very important factor to determine the packing of the BEDT-TTF molecules along the direction perpendicular to the molecular plane. The freezing of the disorder will lead to the thermal contraction anomaly in the (100) plane. In most of BEDT-TTF compounds, the intermolecular contact is closest along the transverse direction. In β-(BEDT-TTF)$_2$I$_3$, the BEDT-TTF molecules are arranged along c with the close S...S contacts (3.60 Å). So that, it is natural that the lattice constant c was not affected by the onset of the lattice modulation wave (see Fig. 2). In contrast, the freezing of the thermal motion of the ethylene group appears to affect mostly the temperature dependence of the lattice constants b and a.

Owing to the small number of the experimental data, the thermal contraction anomalies were not so clear in β-(BEDT-TTF)$_2$(I$_3$)$_{0.9}$(AuI$_2$)$_{0.1}$. Nevertheless, anomalous temperature dependence of b, a and y appeared to be obtained around 190 K. General features of the temperature dependence of the lattice
suggesting that $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ does not show superconducting transition down to 1.2 K.

The electrical resistivities of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ were measured on two crystals selected from the same batch (the chemical compositions of these crystals are assumed to be equal to the composition of the crystal of the same batch analyzed by XMA). The temperature dependence of the resistivities showed the sample dependence. As shown in Fig. 5, one sample showed stepwise resistivity drops at 4.5 K and 2.1 K ($\rho$(5 K)/$\rho$(300 K)=30). At the first step the resistivity decreases to 20% of the initial resistivity. Then, the resistivity becomes zero at the second step. This anomalous resistivity behavior was reproducibly observed in the cooling and warming cycles (300 K $\rightarrow$ 1.5 K $\rightarrow$ 13 K $\rightarrow$ 1.5 K $\rightarrow$ 300 K).

The temperature of the first anomaly (4.5 K) is close to the superconducting transition temperature of the pure $\beta$-(BED-TTF)$_2$Au$_{1.5}$ whose transition temperature is $5\,K - 3.9\,K$ (determined by the penetration depth measurement).

In this connection, it should be recalled that the superconducting transition temperature of $\beta$-(BED-TTF)$_2$Au$_{1.5}$ determined by the Meissner effect measurements is much lower (3.2 K).

The stepwise resistivity drops remain of the so-called high-$T_c$ and low-$T_c$ phenomena in $\beta$-(BED-TTF)$_2$I$_3$, where similar stepwise resistivity drop has been reported (ca. 7 K and 1.8 K). The high-temperature anomaly at 4.5 K of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ will correspond to the high-$T_c$ anomaly of $\beta$-(BED-TTF)$_2$I$_3$ and the superconducting transition at 2.1 K will correspond to the low-$T_c$ transition. Tokumoto et al. have pointed out that the superconducting transition of $\beta$-type salt with linear anion (except I$_3$) is the high-$T_c$ transition, whose transition temperature decreases with decreasing anion size. From this point of view, the transition temperature (ca. 4.5 K) of $\beta$-(BED-TTF)$_2$Au$_{1.5}$ is that of "high-$T_c$ state", which is consistent with the idea that the 4.5 K anomaly corresponds to the high-$T_c$ anomaly.

The other sample showed only a slight anomaly around 4 K and its resistivity became zero at 2.1 K. That is, the high temperature anomaly has a strong sample dependence, which seems to be analogous to the case of the "high-$T_c$ state" of $\beta$-(BED-TTF)$_2$I$_3$.

The 4.5 K and 2.1 K anomalies of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ suggest that the "high-$T_c$ transition" becomes incomplete by the introduction of the disorder in the anion sites and superconducting transition temperature is depressed to 2.1 K.

As mentioned before, in the case of $\beta$-(BED-TTF)$_2$I$_3$, the lattice distortion takes place at the temperature higher than the temperature where ethylene conformation tends to be ordered and the conformation of ethylene group will be frozen by the close contact with the modulated I$_3$ arrangement. Contrary to this, in the system without the periodical modulation of the anion sites, the ethylene conformation will transform to the ordered one above 100 K to form a three-dimensionally ordered arrangement of BED-TTF. Then the effect of the lattice constant resembles that of $\beta$-(BED-TTF)$_2$I$_3$. The X-ray oscillation photographs at 98 K gave strong satellite reflections showing that the lattice modulation wave similar to that observed in $\beta$-(BED-TTF)$_2$I$_3$ appears also in $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ at low temperatures (Fig. 3). The temperature dependence of the lattice constants of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ resembles that of $\beta$-(BED-TTF)$_2$Au$_{1.5}$ (Fig. 2). The anomalies of $b$ and $y$ observed in $\beta$-(BED-TTF)$_2$I$_3$ were not observed in these two systems. The temperature variations of $a$, $c$, and $b$ are smaller and those of $b$ and $y$ are larger than those of the corresponding lattice constants of $\beta$-(BED-TTF)$_2$I$_3$. No satellite reflections suggesting the appearance of the lattice modulation waves could be observed down to 98 K, which is consistent with smooth temperature dependence of the lattice parameters of this system.

The room-temperature resistivities of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ systems are of the same order of $\beta$-(BED-TTF)$_2$I$_3$. But unlike $\beta$-(BED-TTF)$_2$I$_3$, the resistivity measurements of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.74}$Au$_{2.26}$ gave no indication of the superconducting behavior down to 1.5 K ($\rho$(5 K)/$\rho$(300 K) $\sim$ 2000) (Fig. 4). Then, we examined the resistivity of the crystals with different composition of $\beta$-(BED-TTF)$_2$(I$_3$)$_{0.4}$Au$_{2.6}$ ($\rho$(5 K)/$\rho$(300 K) $\sim$ 2000). Again, any indication of the superconductivity could not be obtained. These results seem to be consistent with the recent report by Amberger et al.
modulation might differ from the effect in B-(BEDT-TTF)$_2$13 even if the lattice distortion waves could develop at very low temperatures. Thus, the reduction of the superconducting temperature from 4.5 K to 2.1 K by the introduction of the 10% I$_3$ in the pure AuI$_2$ system may not be attributable to the effect of the lattice modulation wave. Considering that the superconducting behavior of the organic solid is very sensitive to the disorder effect, some kinds of disorder effects will be important for the depression of the superconducting transition temperature of B-(BEDT-TTF)$_2$190.1 (AuI$_2$)$_{0.9}$. Unlike other B-(BEDT-TTF)$_2$X systems with symmetrical linear anion X$_3$I$_2$Br$_2$, B-(BEDT-TTF)$_2$I$_3$Br$_2$ with the orientational disorder of the I$_2$Br does not show superconducting transition down to 0.5 K, 2.1 K.

Similarly, the superconducting transition temperature of the ClO$_4$-ReO$_4$ alloys of TMTSF (TMTST)$_2$(ClO$_4$)$_x$-ReO$_4$ decreases with increasing the ReO$_4$ content and the superconducting transition disappears when the ReO$_4$ content is larger than 5%. In this connection, it should be noted that the possibility of the reduction of the superconducting transition temperature of B-(BEDT-TTF)$_2$13 by the disorder of ethylene substituent has been recently suggested also by Williams et al.

There are two types of the disorder in B-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$. One is the conformational disorder of ethylene group coupled with the periodical lattice distortion wave. The conduction band is formed by the intermolecular overlapping of the highest occupied molecular orbital (HOMO) of BEDT-TTF. This disorder will produce the randomness of the intermolecular transfer integrals of HOMO. The other is the disorder in the anion sites, which will produce a random potential on BED-TTF molecules. The pure I$_3$ salt has the first type disorder and B-(BEDT-TTF)$_2$(I$_3$)$_{0.1}$(AuI$_2$)$_{0.9}$ has the second type disorder. Since the lattice distortion waves develop in B-(BEDT-TTF)$_2$(I$_3$)$_{0.1}$ (AuI$_2$)$_{0.9}$, both types of the disorders will coexist in this system, which did not show superconducting behavior down to 1.5 K.

In conclusion, we have observed the thermal expansion anomaly of B-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$ (x=0.1) around 190 K. indicating that the development of the lattice distortion wave is accompanied by the anisotropic thermal contraction of the crystal lattice. The introduction of the 10% I$_3$ in the pure AuI$_2$ salt depresses the superconducting transition temperature from ca. 4.5 K to 2.1 K. Our observation of the stepwise resistivity drop of B-(BEDT-TTF)$_2$(I$_3$)$_{0.1}$(AuI$_2$)$_{0.9}$ suggests that the coexistence of two superconducting anomalies is not a unique property of B-(BEDT-TTF)$_2$13. The examination of the homogeneity of the impurity distribution will be important to make clear the origin of the coexistence of the two resistivity anomalies. There are two types of the disorder in B-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$. One is the conformational disorder of the terminal ethylene groups of BEDT-TTF and the other is the disorder in the anion sites. Examination of the effects of the disorders will be important to understand the superconducting nature of these salts.

Magnetic studies of B-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$ are now in progress.

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References

10. Since B-(BEDT-TTF)$_2$(I$_3$)$_{1-x}$(AuI$_2$)$_x$ belongs to triclinic system, the unit vectors a, b and c are not identified uniquely. The unit cell adopted in this work is that reported by Kobayashi et al. (T. Mori, A. Kobayashi, Y. Sasaki, H. Kobayashi, G. Saito, and H. Inokuchi, Chem. Lett., 1984, 957 (see Fig. 1)) and is not identical to those of Shibaeva et al. (Ref. 7) and Williams et al. (P. C. W. Leung T. J. Emge, A. J. Schultz, M. A. Beno, K. D. Carlson, H. H. Wang, M. A. Firestone, J. M. Williams, Solid State Commun., 57, 93 (1986)).
11. Taking into account of a trace amount of Br revealed by XMA, the stoichiometries of the mixed-anion systems examined in this work are B-(BEDT-TTF)$_2$(I$_3$)$_{0.1}$(AuI$_2$)$_{0.9}$ (AuBr$_2$)$_{0.02}$ and B-(BEDT-TTF)$_2$(I$_3$)$_{0.1}$(AuI$_2$)$_{0.9}$ (AuBr$_2$)$_{0.04}$. But small amount of AuBr$_2$ is omitted in this paper.
13. H. Kobayashi et al., to be published.
14. The crystal structure of α-(BEDT-TTF)$_2$PF$_6$ resembles to that of B-(BEDT-TTF)$_2$13 and


21. Our crystal structure analysis of $\beta$-BEDT-TTF$2I_2Br$ at room temperature suggested the anisotropic and large thermal motion of the ethylene group. But the degree of the anisotropy and the magnitude of the thermal parameters are smaller than those of $\beta$-BEDT-TTF$2I_3$ (H. Kobayashi, R. Kato, A. Kobayashi, G. Saito, M. Tokumoto, H. Anzai, and T. Ishiguro, Chem. Lett., 1985, 1293). Since the molecular conformation of BEDT-TTF is described to be completely ordered at low temperature (Ref. 12), only orientational disorder of the non-centrosymmetric $I_2Br$ anion is considered to remain at low temperature.

