

A Stable Superconducting State at 8 K and Ambient Pressure in $\alpha_t - (\text{BEDT-TTF}^*)_2\text{I}_3$ ¹

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We report bulk superconductivity at 8 K and ambient pressure in crystals of $\alpha_t - (\text{BEDT-TTF})_2\text{I}_3$. In contrast to the earlier observed metastable superconducting state at 8 K in crystals of $\beta - (\text{BEDT-TTF})_2\text{I}_3$ here the superconducting state is stable and the crystals can be prepared by tempering $\alpha - (\text{BEDT-TTF})_2\text{I}_3$ above 70 °C for several days. ac-susceptibility measurements show that the observed superconducting state at 8 K is a bulk property of the crystals. Resistivity measurements indicate a sharp superconducting transition at 8 K with an onset temperature of about 9 K. The upper critical fields H_{c2} at 1.3 K lie between 3 and 11 T depending on the direction of the magnetic field with respect to the crystal axes. ESR- as well as NMR-measurements indicate a total transformation of the α -phase crystals into the new superconducting α_t -crystals after tempering.

Introduction

The discovery of superconductivity in $\beta - (\text{BEDT-TTF})_2\text{I}_3$ at $T_c = 1.3$ K and ambient pressure [1–3] has initiated an intensive study of the structural and physical properties of these crystals. It was found that under a pressure of about 1 kbar the superconducting

transition can be raised to 7.5 K [4, 5]. Volume superconductivity in $\beta - (\text{BEDT-TTF})_2\text{I}_3$ at ambient pressure and a $T_c = 8.1$ K was obtained after a particular pressure temperature cycling procedure [6, 7], but this superconducting state is only stable as long as the crystal temperature does not exceed 125 K, a temperature where a phase transition occurs which was observed first by thermopower measurements [8]. Recently it was shown, that this meta-stable 8.1 K-superconducting state can also be obtained by an electronic excitation with light at temperatures below 125 K [9].

The structural properties of the $\beta - (\text{BEDT-TTF})_2\text{I}_3$ -crystals are somewhat complicated. At room temperature the crystals are triclinic [10] and the structure is very similar to the Bechgaard-salts. Below

* BEDT-TTF = Bis(1,2-ethylenedithiolo)-tetrathiafulvalene

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19.5 K at ambient pressure an incommensurate structural modulation exists [11–13] and the origin of this modulation is connected with an anion-cation-interaction [13]. Below 125 K a commensurate superstructure with a unit cell volume of about 3 times as large as at room temperature was observed [14]. The basic structural change below 125 K with respect to the room temperature structure lies in a pronounced distortion of the triiodide chains and especially in a change of the linear and symmetric I₃-anions at room temperature into non-linear and asymmetric anions. Therefore it was assumed [14] that the symmetric linear structure of the I₃-anions at room temperature is stabilized down to low temperatures by the special temperature cycling procedure [6, 7] and this symmetric structure results in the high T_c superconducting transition at about 8 K. In fact recently this assumption could be proved by neutron-diffraction experiments [15, 16] and it was shown that no incommensurate modulated structure at 4.5 K exists [16]; furthermore it was shown that the terminal ethylene-groups of the BEDT-TTF-molecules in the stacks are also ordered in contrast to the low T_c superconducting structure where only one -C₂H₄-group is ordered while the other occupies the two possible conformations statistically.

Besides the β -phase of (BEDT-TTF)₂I₃ there exists also the so-called α -phase with identical stoichiometry which was actually discovered first [17–19]. Under ambient pressure the α -(BEDT-TTF)₂I₃ undergoes a metal-insulator phase transition at 135 K [17–19], which can be suppressed by an isotropic pressure of >12 kbar, but no sign of superconductivity could be observed [20]. It was claimed [21] that doping the α -phase crystals with iodine results in a metallic state below the insulator phase transition at 135 K followed by a transition to superconductivity at around 3.2 K. During investigations in our laboratory [22] similar results of resistivity measurements could be obtained as in [21] but microwave conductivity – [23] as well as ac-susceptibility measurements [22] showed that this metallic state is not a bulk effect of the crystals. Recently, Baram et al. [24] reported a structural transformation of α -phase crystals into β -phase crystals by tempering the crystals at a temperature of 70–100 °C for about 10 to 20 h. This is a quite surprising result because of essential differences in the structures of both phases but the structural transformation was confirmed by Weissenberg-pictures [24].

The most surprising fact after the structural transformation was that these crystals show a sharp decrease in resistivity at around 8 K indicating a superconducting transition at this temperature. Baram et al. [24] did not report on resistivity measurements

with a simultaneously applied magnetic field and these authors could not tell whether or not the reported drop in resistivity and the eventual superconducting transition is a bulk effect in these crystals. A further open question is what the differences in the structures between the normal grown β -(BEDT-TTF)₂I₃ and the tempered α_r -(BEDT-TTF)₂I₃-crystals are. (In the following we will call the tempered α -phase crystals α_r -(BEDT-TTF)₂I₃). This is an important question since β -(BEDT-TTF)₂I₃ becomes superconducting under normal conditions at 1.3 K while in α_r -(BEDT-TTF)₂I₃ this transition seems to be at around 8 K where as the β -(BEDT-TTF)₂I₃ shows superconductivity only after special treatment (temperature pressure cycling or electronic excitation by light).

In order to obtain answers to this question we carried out measurements of the temperature dependence of the resistivity and ac-susceptibility, both with and without applying a magnetic field, furthermore ESR-, NMR-, resonance Raman-, and thermopower investigations on α_r -(BEDT-TTF)₂I₃ as well as on β_r -(BEDT-TTF)₂I₃ crystals (in the following we call β -phase crystals which were tempered above 75 °C for several days β_r -(BEDT-TTF)₂I₃). We will see that the α_r -(BEDT-TTF)₂I₃ crystals show bulk superconductivity at 8 K while the transformation of β_r -(BEDT-TTF)₂I₃ into the high temperature superconducting phase by tempering takes much longer but seems to be possible as well.

Experimental

α - and β -(BEDT-TTF)₂I₃ crystals were grown from a THF-solution by electro-chemical methods as described earlier [19]. The crystals were tempered in air for 4–6 days at temperatures between 75 and 95 °C. α -phase crystals tempered at 75 °C kept their shapes much better than those tempered at higher temperatures. In all cases ESR-linewidth measurements before and after tempering indicated already a structural transition for the α -crystals. Before tempering the α -phase-crystals showed the usual 80–120 Gauss linewidth at room temperature [8, 25], while after tempering the linewidth was in the order of 20 Gauss, the same value as usually observed for β -phase crystals [8, 25]. In β - and β_r -crystals no change in ESR-linewidth could be observed.

dc and ac-conductivity-measurements were performed with the usual four point-method on α_r -phase crystals of typical dimensions of $3 \times 1 \times 0.2 \text{ mm}^3$ while the β_r -crystals were smaller about $1.5 \times 0.5 \times 0.5 \text{ mm}^3$. Thermopower and Raman-investigations were car-

ried out as described earlier [26, 9]. ^{13}C -NMR spectra were recorded at room temperature at 68 MHz. The samples were powdered and mixed with fine quartz sand (ratio 2:1). Magic angle spinning (MAS) with a spinning frequency between 4–5 KHz as well as proton decoupling were used. The spectra were excited simply with ^{13}C - 90° pulses rather than with proton carbon cross polarization, since, due to the short relaxation rates, high repetition rates could be used. Cross polarization for all carbons was only effective in the case of BEDT-TTF while for the organic metals only the $-\text{C}_2\text{H}_4$ -carbons were affected.

Results

Figure 1 shows the temperature dependence (logarithmic scale) of the resistivity of α -, β - and α_t -crystals; while at room temperature the typical resistivity for α -crystals is around 0.005–0.01 Ohm-cm in α_t -crystals this resistivity value is about one order of magnitude higher. In contrast to the α -phase crystals which show the typical metal insulator transition around 135 K [18, 19] the α_t -crystals show a metallic behaviour down to low temperatures and around 8 K a sharp superconducting transition occurs while the β -crystals become superconducting only around 1.3 K.

Figure 2 shows the low temperature part of the resistivity for two typical α_t -crystals. The onset temperature for superconductivity lies for both α_t -crystals around 9 K. However, the superconducting transition temperature evaluated by the center of the resistive transition amounts to 8 K. By 6.5 K the superconducting transition is complete and the resistivity is 0 within the experimental possibilities. The suppression of the superconducting state by a magnetic field of about 6 T along the c^* -axis at a temperature of 1.3 K for sample II is also shown in Fig. 2.

The temperature dependence of the thermopower of α - and α_t -crystals is shown in Fig. 3. It can be seen that the phase transition at 135 K observed in the α -phase crystals does not appear in the α_t -crystals. In addition the phase transition at 125 K as it was seen for β -phase crystals [8] could not be observed.

Figure 4 demonstrates the result of an ac-susceptibility measurement on the α_t - $(\text{BEDT-TTF})_2\text{I}_3$ -crystal II. There is clear evidence of an onset of dia-magnetic shielding below 6.5 K, the temperature where according to the resistivity data the superconducting transition is already complete. A similar lower transition temperature in the ac-susceptibility was already observed before for β - $(\text{BEDT-TTF})_2\text{I}_3$ -crystals [27]. The signal which still increases on cooling to 1.3 K is about 70% of that expected for a perfect super-

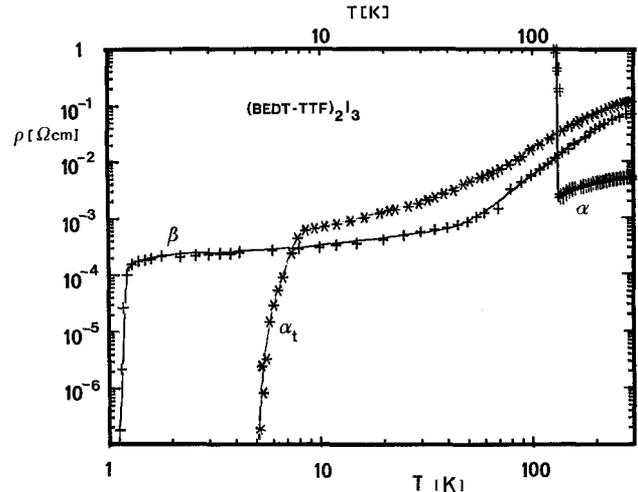


Fig. 1. Resistivity versus temperature (logarithmic scale) for α - $(\text{BEDT-TTF})_2\text{I}_3$ (+) (metal insulator phase transition at 135 K), β - $(\text{BEDT-TTF})_2\text{I}_3$ (+) and α_t - $(\text{BEDT-TTF})_2\text{I}_3$ (*)

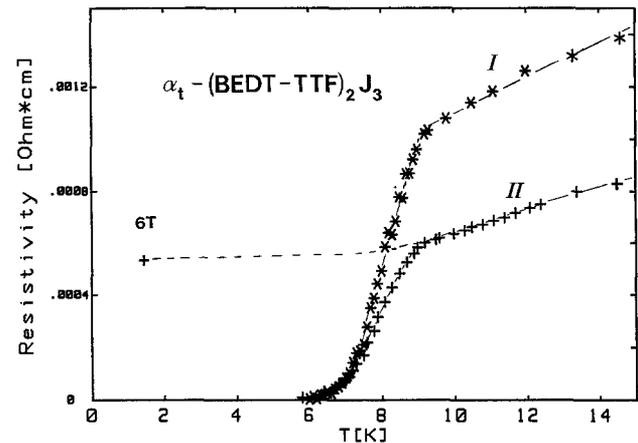


Fig. 2. Resistivity of two α_t - $(\text{BEDT-TTF})_2\text{I}_3$ -crystals (I and II) in the temperature region between 0 and 15 K. The suppression of superconductivity in sample II by applying a magnetic field of 6 T at 1.3 K parallel to the c^* -axis is shown as well

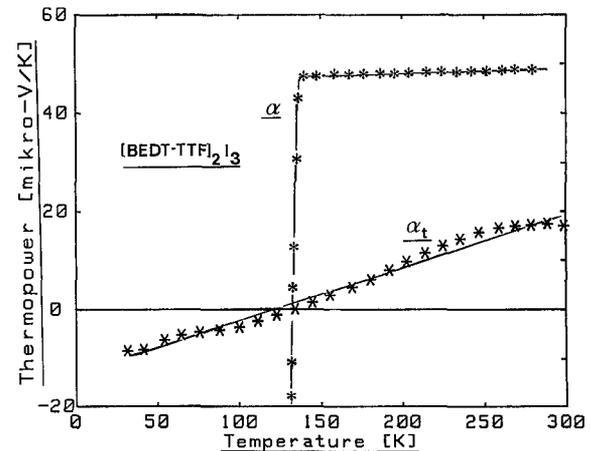


Fig. 3. Temperature dependence of the thermopower for α - and α_t - $(\text{BEDT-TTF})_2\text{I}_3$ -crystals

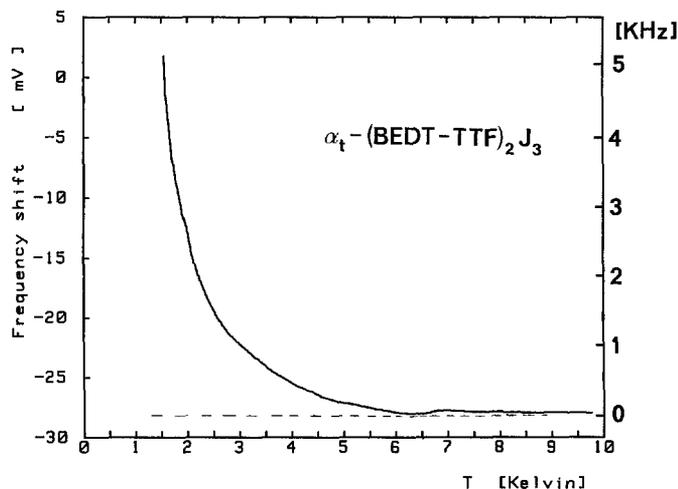


Fig. 4. Increase in resonance frequency of a LC-circuit (3.3 MHz) due to exclusion of the rf-field by diamagnetic shielding currents (ac-susceptibility) in α_t -(BEDT-TTF) $_2$ I $_3$ (sample II of Fig. 2) by lowering the temperature

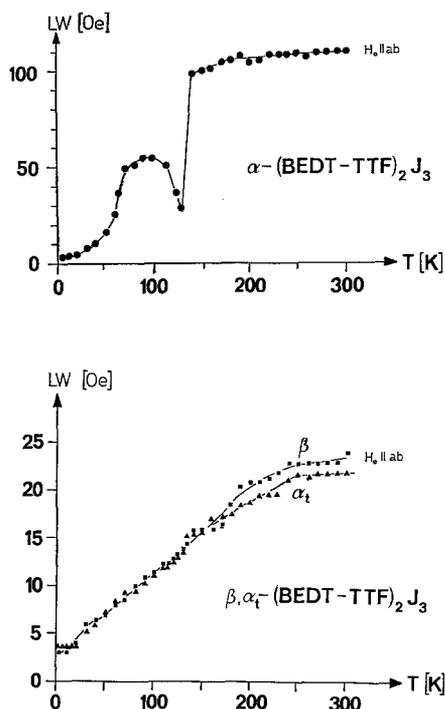


Fig. 5. Temperature dependence of the ESR-linewidth for α -(BEDT-TTF) $_2$ I $_3$ (top) as well as for β - (■) and α_t -(BEDT-TTF) $_2$ I $_3$ (▲) (bottom)-crystals

conductor and indicates an inhomogeneous T_c -distribution in the sample.

Figure 5 shows the temperature dependence of the ESR-linewidth obtained from an α -, β - and an α_t -(BEDT-TTF) $_2$ I $_3$ -crystal for the magnetic field perpendicular to the c^* -direction. The clear phase transition observed from the ESR-linewidth for α -crystals

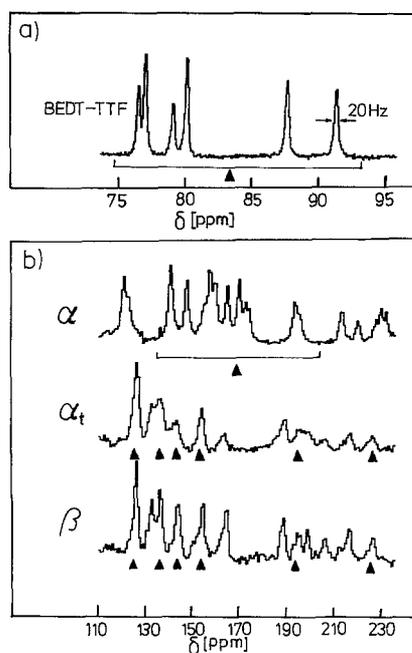
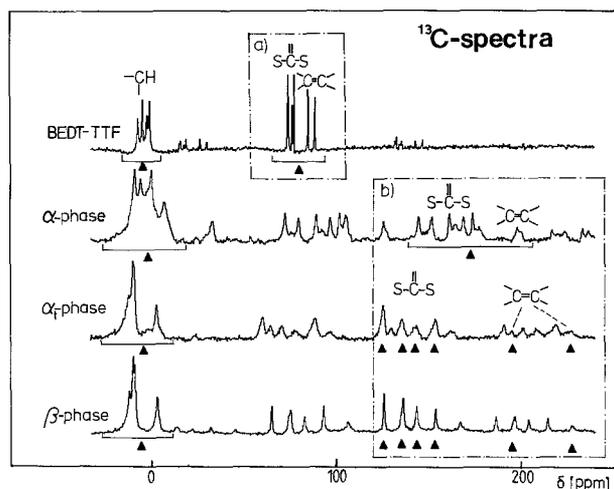


Fig. 6. Top: MAS ^{13}C -NMR spectra at 300 K of the neat donor material BEDT-TTF and of the organic metals α -, β - and α_t -(BEDT-TTF) $_2$ I $_3$. The center bands are marked by \blacktriangle . Chemical shifts/Knight shifts are referenced to TMS. Bottom: Blow up of the framed parts of the spectra from top for slightly modified recording conditions (spinner frequencies). Note that all 6 lines of the corner and center carbons of BEDT-TTF are fully resolved

disappears after tempering and the temperature dependence of the ESR-linewidth for the α_t -crystals is very similar to the one observed for β -(BEDT-TTF) $_2$ I $_3$ -crystals.

The susceptibility of the α_t -crystals obtained from the ESR-data is more or less temperature-independent similarly as found earlier for β -(BEDT-TTF) $_2$ I $_3$ -crystals [25], but there exists a certain difference below 8 K because, due to the superconducting transition the amplitude of the ESR-signal decreases rapidly while the linewidth is con-

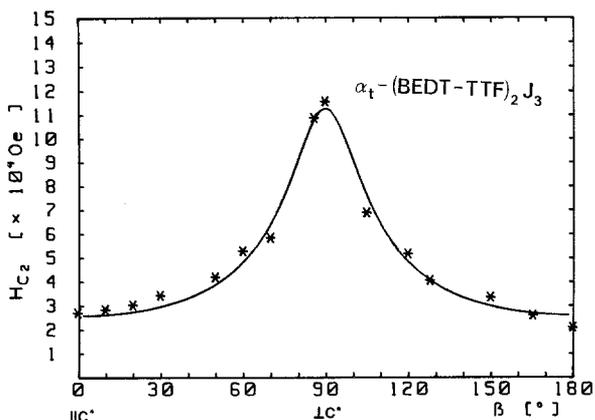


Fig. 7. Upper critical fields H_{c2} at a temperature of 1.3 K for α_t -(BEDT-TTF) $_2$ I $_3$ as evaluated by the mid-transition of the resistivity curve for several magnetic field directions

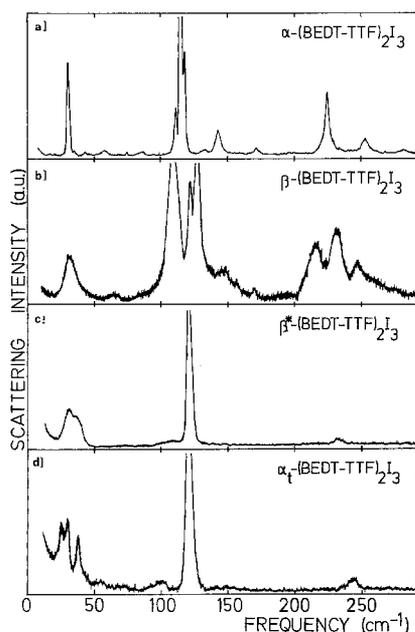


Fig. 8a-d. Low energetic parts of the resonance Raman-spectra at 2 K of a α -(BEDT-TTF) $_2$ I $_3$ b β -(BEDT-TTF) $_2$ I $_3$ (low temperature superconducting (1.3 K phase) c β' -(BEDT-TTF) $_2$ I $_3$ (high temperature superconducting (8 K) phase, as prepared by optical excitation [9]. d α_t -(BEDT-TTF) $_2$ I $_3$. (Excitation wave length 4880 Å, 10 mW)

stant, indicating a decrease in susceptibility at these low temperatures.

Figure 6 (top and bottom) summarizes the results of the ^{13}C -NMR solid state investigations. On top the ^{13}C -spectrum of the neat donor material BEDT-TTF and the spectra of α -, α_t - and β -crystals are presented. On the bottom a blow up of the framed parts of the full spectra from the top for slightly modified recording conditions are shown. The Knight shifts present in these spectra shall be discussed elsewhere [28]. Here we are concerned only with the structural

information contained in these data. It can be clearly seen that the spectra of α - and α_t -crystals differ markedly – as well in the region of the terminal $-\text{C}_2\text{H}_4$ -groups as in the corner ($S-C-S$) and center ($C=C$) carbons – while the spectra of the α_t - and the β -crystals are virtually identical. This indicates – since a large amount of α_t -crystals (≈ 0.3 g) was used for these NMR-measurements – that by tempering the α -crystals above 70°C for several days all crystals were converted into the new α_t -phase.

The upper critical fields H_{c2} at a temperature of 1.3 K for α_t -crystals as evaluated by the mid-transition of the resistivity curve for several magnetic field directions are presented in Fig. 7. The value of H_{c2} for the direction of the magnetic field perpendicular to the c^* -direction is about 11 T.

Figure 8a-d shows the low energetic parts of the resonance Raman-spectra of α -, β -(in the low T_c -phase) as well as in the high T_c -phase (c), both from Ref. 9 and α_t -(BEDT-TTF) $_2$ I $_3$ (d) at a temperature of 2 K. It can be seen that the symmetric stretching mode of the I_3 -anions around 120 cm^{-1} in the α_t -crystal (d) is identical to the high temperature T_c -phase of β -(BEDT-TTF) $_2$ I $_3$ (c) in contrast to the split mode of the low temperature T_c -phase (b), indicating the linear and symmetric I_3 -anions and the higher order present in the α_t -crystals with respect to the low T_c -phase of β -crystals [9]. In α_t -crystals three phonon modes between 30 and 50 cm^{-1} are somewhat better resolved.

Discussion

All results presented above indicate clearly that tempering of α -(BEDT-TTF) $_2$ I $_3$ crystals at temperatures above 70°C during several days leads to a structural phase transition. In fact, it is possible to observe the phase transition under a polarisation microscope in a thin α -crystal at a temperature of about 90°C optically. The ^{13}C -NMR data show that the transition is complete and they indicate also that the new structure is more or less identical with that of β -(BEDT-TTF) $_2$ I $_3$. This is further indicated by ESR-linewidth measurements. Nevertheless, the resistivity data at low temperatures show that the structure is related to the high temperature superconducting phase ($T_c=8\text{ K}$) of the β -crystals.

The tempered α_t -(BEDT-TTF) $_2$ I $_3$ -crystals are organic metals down to low temperatures (Fig. 1) in contrast to the α -phase crystals. The disappearance of the metal insulator phase transition in the α_t -crystals can be seen in the resistivity curve, in the thermopower data as well as in the ESR-line-width measurements.

The α_r -(BEDT-TTF)₂I₃-crystals become superconducting at 8 K and the superconductivity can be suppressed by applying a high magnetic field. The critical fields H_{c2} are similar to those observed in the high T_c -phase of β -(BEDT-TTF)₂I₃ after the special temperature pressure cycle [6, 7]. This superconducting state at 8 K is stable and fully reproducible for several temperature cycles. Furthermore, the ac-susceptibility measurements show that the superconductivity is a bulk effect of the crystals affecting at least 70% of the total volume. They show further that the transition is rather broad and still not complete at a temperature of 1.3 K.

All these results together with the fact that Baram et al. [24] stated that the tempered α_r -crystals show in Weissenberg-pictures the structure of β -phase-crystals indicate that the α_r -crystal structure is identical with the one of the high T_c superconducting phase of β -(BEDT-TTF)₂I₃. Therefore in the following we shall consider the differences between the structures of the normal β -phase (at 300 K), the low T_c β -phase (below 125 K) and the high T_c - β -phase (after a special pressure temperature cycle below 125 K) as well as the question why the structural transition of the α -phase obtained by tempering results in this high T_c -superconducting β -phase.

In the room temperature structure of the β -phase the I₃-anions are linear and symmetric; there exists one kind of donor stacks in which the terminal-C₂H₄-groups of the BEDT-TTF-molecules are ordered and have all the same positions in the stacks on one side of the molecules while they are disordered and occupy statistically both possible positions on the other side [15, 16]. In the low T_c β -phase below 125 K there exists a pronounced distortion of the I₃-chains [14]. The I₃-anions are nonlinear and asymmetric due to an anion-cation interaction [13] and a freezing of the ethylene group flipping, which can be inferred from NMR-relaxation measurements [28].

In the high T_c superconducting β -phase obtained by the special pressure temperature cycling procedure [6, 7] the I₃-anions are again linear and symmetric but in addition in the BEDT-TTF-donor stacks all the terminal C₂H₄-groups on both sides are fully ordered as was shown recently by neutron diffraction experiments [15, 16].

The unit cell of the α -(BEDT-TTF)₂I₃-crystals at room temperature is about twice as large as the unit cell of the β -phase (1717 Å³ versus 849 Å³). In the α -phase crystals at room temperature the I₃-anions are linear and the -C₂H₄-groups of the BEDT-TTF donor molecules in both crystallographically non-equivalent stacks are totally ordered [19], as in the high T_c -superconducting β -phase. In addition NMR

relaxation measurements [28] indicate that the ethylene-groups do not flip rapidly. Therefore, we may assume that during tempering the α -crystals at temperatures of about 75 °C the structural transition results in a β -phase structure where all the terminal -C₂H₄-groups are again ordered. This structure would be the same as the one of the high T_c -superconducting β -phase structure but now it exists already at room temperature and ambient pressure. This might be the reason that α_r -(BEDT-TTF)-crystals become superconducting at 8 K and ambient pressure. Further structural investigations are necessary and in fact already under progress to test these conclusions.

The final question which arises is why the β -phase crystals do not stay in the high temperature superconducting structure above 125 K after being prepared by the special temperature pressure cycle. We assume in the moment that due to impurities and some disorder in the anion channels as well as due to the cation-anion interaction [13] and the fast movement of the ethylene-groups at temperatures above 125 K the structural change is induced.

Therefore, we tried by tempering β -(BEDT-TTF)₂I₃-crystals to convert those into their high temperature superconducting phase. Those crystals (which we call in the following β_r -crystals) seem to convert much slower than the α -phase crystals but after tempering for twelve days at 85 °C a clear indication was found that these crystals can also be transformed into the high temperature superconducting phase. While the resistivity curve at low temperature of β_r -crystals is very similar to those of the α_r -crystals in Fig. 2, the ac-susceptibility measurements indicate a superconducting transition below 4.5 K and that at 1.3 K at least 50% of the material has become superconducting. This transition temperature of 4.5 K is below the value for α_r -crystals but clearly above the 1 K of β -phase-crystals in their low temperature superconducting phase. Therefore, it seems that the structural transformation into the high T_c -superconducting phase – meaning an ordering of all terminal -C₂H₄-groups in the β -phase-crystals at about 85 °C – can be obtained for β -phase-crystals by tempering as well, but it might be that this ordering takes much longer than the phase transition in the α -phase-crystals at 75 °C. Further investigations to clarify this point are under progress.

In conclusion we can state, that the α_r -(BEDT-TTF)₂I₃-crystals are organic metals with a stable 8 K superconducting transition at ambient pressure. Superconductivity in the α_r -crystals is a bulk effect and the upper critical field H_{c2} is 3 T for a magnetic field parallel c^* and about 11 T for a magnetic field perpendicular to c^* .

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