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Determination of the Electron Phonon Coupling and the Superconducting Gap in β-(BEDT-TTF)$_2$X Crystals (X = I_3, IAuI)

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We performed point-contact and tunneling measurements on the organic superconductors β-(BEDT-TTF)$_2I_3$ and β-(BEDT-TTF)$_2$IAuI, in the normal and superconducting states. The point-contact measurement in the normal state provides the Eliashberg function $\alpha^2(\omega)\,F(\omega)$. This function has maxima at 4 and 15 mV, as well as a sharp peak at 1 mV which seems to indicate a very soft phonon strongly coupled to the electrons. The measurements in the superconducting state provide the superconducting gap, which for the tunneling measurements has a value $2\Delta/k_B\,T_c \simeq 4$, just slightly above the BCS value.

1. Introduction

The electron-phonon coupling in organic metals is of considerable importance for the understanding of the superconducting as well as transport properties. In contrast to "ordinary" metals, there are not many experimental data concerning the question whether the electron-phonon coupling is responsible for the superconductivity, as well as the electrical resistivity at finite temperatures [1].

In normal metals, the electron-phonon coupling is described by the McMillan coupling constant $\alpha(\omega)$, where ω is the phonon frequency. The quantity $\alpha^2(\omega)\,F(\omega)$ where $F(\omega)$ is the phonon density of states can be determined experimentally as function of ω by tunneling measurements. Such measurements are most often carried out in the superconducting state (Giaever tunneling), however, they can also be carried out in the normal state, using point-contacts with a very small cross-sectional area, so that very large current densities can be obtained without heating. In that case, the $I-U$ characteristics are non-linear, and the second derivative $\mathrm{d}^2U/\mathrm{d}I^2$ yields the quantity $\alpha^2(\omega)\,F(\omega)$, just as in Giaever tunneling. This method has been used extensively in "normal" metals [2], including valence fluctuation compounds [3]. A preliminary experiment on the organic metal β-(BEDT-TTF)$_2I_3$ (**1**) was reported recently [4]. We report here data from more extensive measurements on this material both in the normal state, where $\alpha^2(\omega)\,F(\omega)$ is determined, and in the superconducting state, where the superconducting gap parameter is determined. We also report preliminary data on β-(BEDT-TTF)$_2$IAuI (**2**) from a tunneling experiment, in which the superconducting gap parameter is determined. We analyse the data using the Eliashberg strong-coupling theory of superconductivity, employing the Rainer-Bergmann computer programs. These results are in accord with the electron-phonon theory of superconductivity in organic metals [5].

2. Experimental and Results

In the previous work [4], a single crystal of β-(BEDT-TTF)$_2I_3$ (**1**) was pressed against a copper plate. In the present experiment, two crystals of **1** are pressed against each other, using a berylium-copper spring. In this method, the force pressing the two crystals together is much weaker than in the previous experiment, where a screw-device was used to push the crystal against the copper plate. Inspection of the crystals after the experiment, under a microscope, showed no appreciable damage to the crystal faces. The spring-loaded device was placed in a dilution refrigerator, and measurements of the $I-U$ curves were conducted in the range 1.6 K–80 mK. The $I-U$ characteristics were found to be much more stable than in the previous experiment.

In addition to this experiment, another experiment was performed on crystals of **2** using the tunneling method. Two crystals were brought close together within the dilution refrigerator, using a piezoelectric linear motor. At first the crystals were made to touch so that there was a slight cleavage of the surface, exposing a clean surface. After this procedure the crystals were slightly separated to perform the tunneling measurement. In this method, the crystals do not touch at all, and thus there is no stress whatsoever on the crystal faces during the experiment. Our experiments were all done in the ab-plane.

The dU/dI and d^2U/dI^2 curves of a crystal of **1** at 1.4 K (thus, in the normal state) are shown in Fig. 1 a, b. Note in particular the "clean" background at voltages above 25 mV. The d^2U/dI^2 curve has a very large, sharp maximum at 1 mV, a broad maximum around 15 mV, and a weak shoulder around about 4 mV.

The resistance at zero voltage was 72 Ω; using the formula for the Sharvin resistance $R_s = 4p_F/(3ne^2\pi a^2)$ [2] with p_F being the fermi momentum and a the diameter of the contact, a value for the fermi wave vector $k_F = p_F/\hbar = 3\times10^7$ cm^{-1} [6], $n = 1.2\times10^{21}$ cm^{-3} (1/2 an electron per BEDT molecule), we obtain a value of $a = 78$ Å. Using this value in the formula $dR/dU = m/(ne a\hbar)\cdot\alpha^2F\,(m=m_e)$, we obtain a value of "$\alpha^2(\omega)\,F(\omega)$" as shown in Fig. 1 c. The α^2F-curve is calculated by averaging the positive and negative bias parts of $|d^2U/dI^2|$. Due to a slight asymmetry, the shoulder around 4 meV develops to a pronounced peak. This determination of the absolute value of α^2F assumes perfect surfaces, and a perfect contact. This is probably not the case. While in alkali metals it is possible to obtain such conditions, this is not the case in metals which are not free-electron like, and where lattice damage at the surface has a much larger effect on the electronic structure.

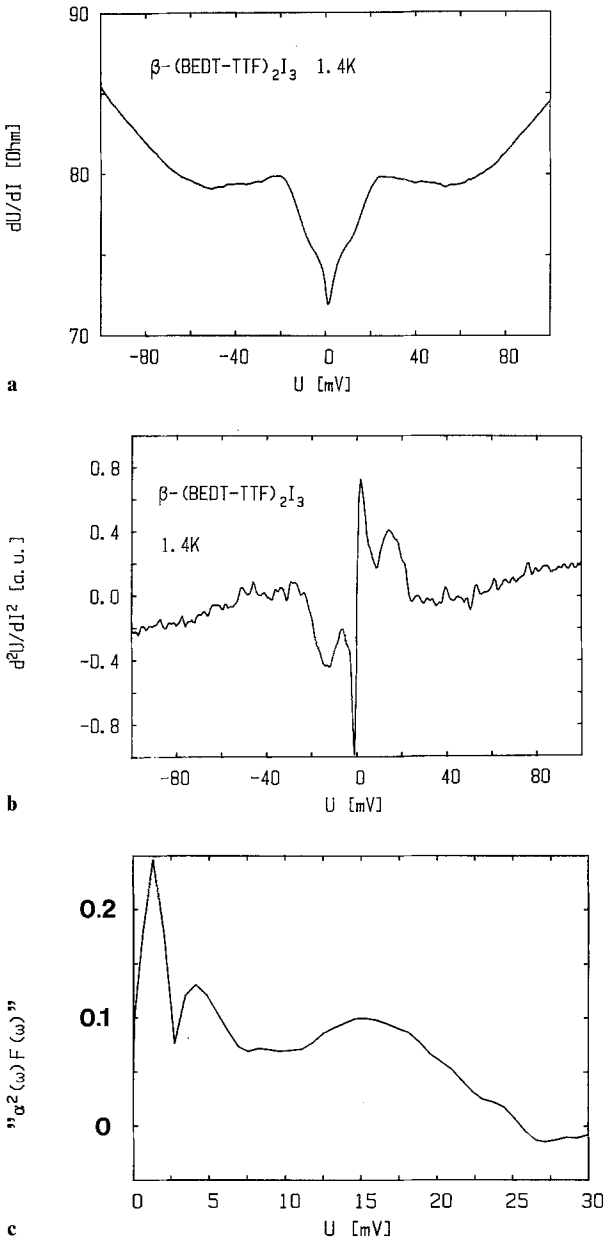

Fig. 1 a–c. Point-contact spectra of β-(BEDT-TTF)$_2I_3$ in the normal state. **a** dU/dI as function of U. **b** d^2U/dI^2 as function of U. **c** The Eliashberg function $\alpha^2(\omega)\,F(\omega)$. The scale is trustworthy to within a factor of 2

For this reason, we do not know how trustworthy the absolute scale of the quantity α^2F is, and we use quotations-marks for it. The absolute value of α^2F also depends on the assumption of a round, cylindrical Fermi surface as suggested in Ref. 6, and we do not know how reliable the electronic band structure reported there is. We note, however, that the integral $\lambda = 2\int\alpha^2(\omega)\,F(\omega)/\omega\,d\omega$ is found to be $\lambda = 1$, therefore we believe the absolute value of α^2F determined this way to be reliable within a factor of 2 or better.

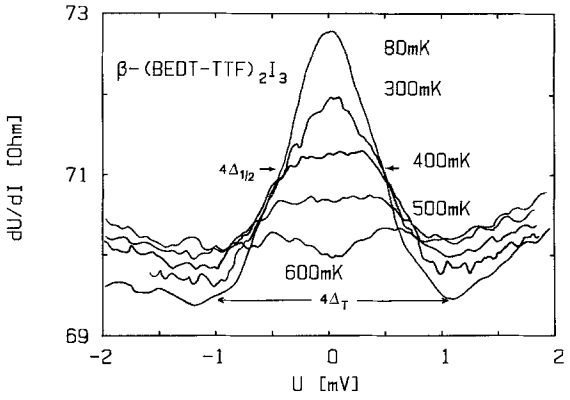

Fig. 2. Tunneling data in the superconducting state of β-(BEDT-TTF)$_2I_3$ at various temperatures

The data for temperatures below T_c (~ 1.3 K) for a crystal of **1** are shown in Fig. 2. There is a maximum in the resistivity at $U=0$, and the dU/dI vs. U curve is bell-shaped. We indicate the half-width of this curve by $2\Delta_{1/2}$, and the half-width at the bottom of this curve (where the first derivative dU/dI has a minimum) by $2\Delta_T$, since in the BCS theory Δ_T should give the superconducting gap. The maximum in the resistivity at $U=0$ is seen to decay rapidly with temperature around $T\simeq 0.6$ K. In this temperature range, there is a *minimum* at $U=0$, characteristic of samples with some paramagnetic impurities [7].

The dI/dU vs. U curves for crystals of **2** measured by the tunneling method are shown in Figs. 3a–c, for various temperatures. Figure 3a and 3b show superconductor-superconductor tunneling and Fig. 3c superconductor to normal metal tunneling. The shape of the curve is somewhat steeper, and closer to the "theoretical" curves given by BCS-theory for the superconductor-superconductor tunneling and by the BTK-theory [8] for the superconductor to normal metal tunneling. On a few crystals of *2* some hysteresis in the dI/dU-curves is observed (Figs. 3b and 3c). This hysteresis may be due to cracks formed during the cleavage procedure.

The values of $2\Delta_{1/2}$, $2\Delta_T$ at the lowest temperatures (~ 80 mK) are given in Table 1. It is seen that for the point-contact data, $2\Delta_{1/2}$ is close to the BCS value (for $T_c=1.3$ K) while $2\Delta_T$ is about a factor of 2 larger. For the tunneling experiment on **2** the difference between the values of $\Delta_{1/2}$ and Δ_T is somewhat smaller. $2\Delta_T/kT_c$ is approximately 3.8–4.2 i.e. about 15% larger than the BCS weak coupling value. These results deviate from the previous data of Hawley et al. [9] who reported that $2\Delta_T$ is about 5 times larger than the BCS value for tunneling in the ab-plane; however, their data for tunneling in the c^*-direction give a value closer to the BCS-value. In our tunneling

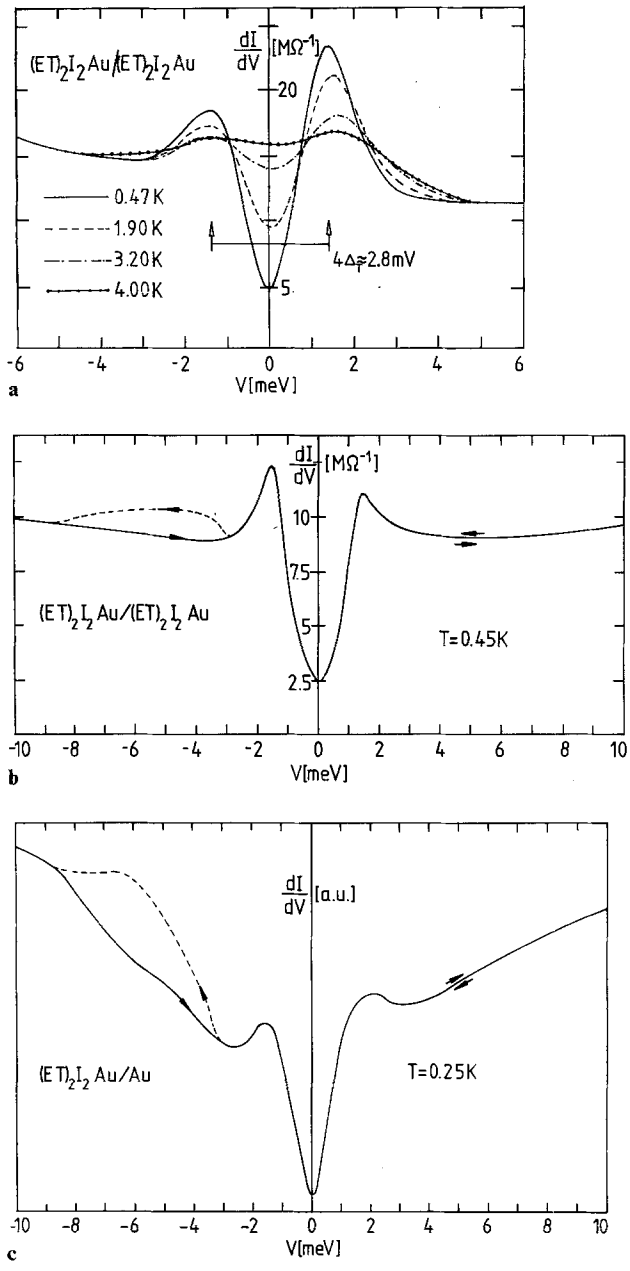

Fig. 3a, b. Tunneling data in the superconducting state of β-(BEDT-TTF)$_2$IAuI at various temperatures. **c** β-(BEDT-TTF)$_2$IAuI to Au tunneling

experiments we slightly cleaved the crystals just prior to the measurement, therefore we are not absolutely sure about the orientation of the surfaces between which the tunneling actually takes place.

In the superconductor-to-superconductor experiment on crystals of **2**, following the tunneling measurement the crystals were brought together by the piezoelectric drive and a point-contact was formed. $I-U$ curves were measured near $U=0$. A steep slope dI/dU was observed. This is probably an indication

Table 1. The tunneling gap for the various experimental techniques. $4\Delta_T$ is defined as the distance between points where the second derivative is maximum (or minimum); $4\Delta_{1/2}$ is defined as the distance between points where the second derivative has half its maximum value. The tunneling between the crystals of **2** and the gold needle may be 'standard' normal-to-superconductor tunneling, described by the BTK theory [8] and the measured gap is Δ. But it is possible that when the needle is brought in contact with the crystal, a tiny part of it splits off and sticks to the needle, so that the actual tunneling is between two superconducting crystallites, and the measured gap is 2Δ. Therefore in the table the values of $2\Delta/k_B T_c$ for both alternatives are given. The BCS value is $2\Delta/k_B T_c = 3.5$

Sample	T	$2\Delta_T$ (mV)	$2\Delta_{1/2}$ (mV)	T_c (K)	$\frac{2\Delta_T}{k_B T_c}$	$\frac{2\Delta_{1/2}}{k_B T_c}$
β-(BEDT-TTF)$_2I_3$ – β-(BEDT-TTF)$_2I_3$ (point contact)	0.08 K	1	0.42	1.35	8	3.6
β-(BEDT-TTF)$_2$IAuI – β-(BEDT-TTF)$_2$IAuI (tunneling)	0.45 K	1.4	0.7	4.1	4	2
β-(BEDT-TTF)$_2$IAuI – Au (assuming s.c.-s.c. tunneling)	0.25 K	1.75	0.6	4.1	5	1.7
β-(BEDT-TTF)$_2$IAuI – Au (assuming s.c.-normal tunneling)	0.25 K	3.5	1.2	4.1	10	3.4

of a Josephson effect, with a small series resistance which may be due to a small normal region at the extremity of the sample, or perhaps a proximity effect.

In the point-contact experiments on the crystals of **1**, where the contact was established by a mechanical screw device, a minimum resistance was observed at $U = 0$. However, the reduction in resistance is much smaller than in the previous case.

3. Discussion

a) The "$\alpha^2 F$" Curve

The d^2U/dI^2 vs. U-curve in Fig. 1 has a very sharp and high peak at 1 meV; it has a broad maximum around 15 meV, and a shoulder around 4 meV. If we use the "standard" formula for the Sharvin resistance, the maximum value of $\alpha^2 F$ at 1 meV is approximately 0.2, and the McMillan $\lambda = 2\int \omega^{-1}\alpha^2(\omega) F(\omega)\, d\omega$ is approximately 1. Use of these formulas assumes ideal contacts – the material being in perfect crystalline state even at the point of contact. This is probably not the case; there must be an appreciable pressure in the region of the contact, and the electronic properties of this material are strongly pressure dependent. The value $a = 78$ Å obtained from the formula for the Sharvin resistance probably gives the radius of good electrical contact only. There may be mechanical contact between damaged surfaces, with poor electrical contact, over much larger areas in addition (a stress of order 1 kbar, with a force of order 10 dynes, would require a mechanical contact over an area with a radius of order 5000 Å). Thus, the large, and reasonable value of "$\alpha^2 F$" and of λ is somewhat surprising.

The maxima at 4 meV and 15 meV correlate well with lines observed in Raman spectra [10]. There is a phonon line around 30 cm^{-1}, due to an external mode (translation or libration or a mixture of the two). There are strong Raman lines at 107, 120 and 126 cm^{-1} [10b], due to an $I-I$ stretching mode. The Raman lines are sharp, while the peak around 15 meV in the "$\alpha^2 F$" curve is very broad. Therefore, we do not know yet whether the two are related. In any case, the cation external modes are expected to extend over this frequency range as is the case in the other organic materials [11].

We see no contribution to $a^2 F$ above 26 meV; this is the region of the low-lying *internal* modes, i.e. bond twisting and bending. Thus, it appears that these modes do not couple strongly with the electrons at least not with the ordinary linear coupling. This is in accord with the data of resistivity under pressure of organic metals of the TTF-TCNQ family. [12].

The surprising result of the present experiment is the observation of the soft phonon at 1 meV. Such soft phonons have not been observed before in organic metals [13]. In BEDT-TTF_2I_3 there is a preliminary indication from Raman data of a line at 7 cm^{-1} [14]. This soft phonon was not observed in the previous run on β-(BEDT-TTF)$_2I_3$ [4], where a crystal was pressed against a copper plate by a screw device. In that experiment the force on the crystal is appreciably higher, therefore it is not surprising that this extra soft mode is not observed there.

This soft phonon may be related to the low $T_c = 1.3$ K which is investigated in the present work and might be shifted to higher frequencies in the high $T_c = 8$ K phase which is stabilized under pressure [15]. From the Eliashberg theory, a phonon that is so soft

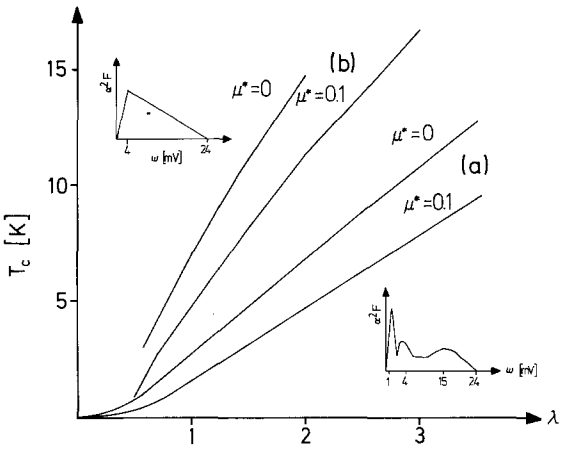

Fig. 4. T_c calculated from Eliashberg theory as function of λ for the α^2F function of Fig. 1c (shown as 'a') as well as for the α^2F function of Ref. 4 (shown as 'b'), for two values of the Coulomb repulsion μ^*. The calculations were performed by D. Rainer

(i.e. 1 meV), does not contribute much to T_c, as long as the electron phonon coupling constant $\alpha(\omega)$ remains constant as ω decreases [16]. Thus, it is possible that this soft phonon marks the low-T_c phase, while in the high-T_c phase this phonon is harder and thus contributes to T_c.

In Fig. 4 we plot curves of T_c vs. λ for the observed a^2F curve, according to the Eliashberg theory using the Bergmann-Rainer computer programs. To vary λ, and T_c, we change the scale of the α^2F curve without changing its shape. We plot curves for values of the effective Coulomb coupling constant $\mu^*=0.1$ (characteristic of non-transition metals) and $\mu^*=0$ (the electron on a molecule may be more delocalized than on an atom, giving rise to a smaller Coulomb term). We also plot curves for the α^2F curve observed in the previous, preliminary, run. It is seen that the value of T_c for a given value of λ (or the value of λ for a given T_c) are very different. This reflects the fact that the soft phonon contributes much to λ, but not to T_c. Thus, in the ambient-pressure phase, we may have a value of λ of about 1, corresponding to $T_c \simeq 1.3$ K, (for $\mu^*=0.1$, curve a) while in the high-T_c phase, a value of λ of about 1.5 corresponds to $T_c \simeq 8$ K (with the same value of μ^*, curve b). Note that if we stay on the *same* curve, such a large change in T_c implies a very large change in λ (from $\lambda \simeq 1$ to $\lambda \simeq 3$ on curve a, and from $\lambda \simeq 0.5$ to $\lambda \simeq 1.5$ on curve b). The resistivity of the ambient-pressure phase, at ambient temperature, is somewhat higher than that of the high-pressure phase (by about 30–40%); this suggests a higher value of λ; however, there is a significant quadratic (T^2) contribution to the resistivity, which seems to be much stronger in the ambient-pressure phase. This T^2 term does not contribute neither to T_c nor to λ [17]. Also, the soft-phonon implies very strong anharmonic effects at ambient temperature, therefore we cannot deduce a value of λ from the ambient-temperature resistivity.

In any case, qualitatively, hardening the soft phonon by going to the high-pressure phase, *increases* its contribution to T_c and *decreases* its contribution to the electrical resistivity.

Structures similar to the 1 meV-peak were observed also in valence-fluctuation and heavy-Fermion compounds [18], a strongly energy-dependent density of states might be the reason there.

Here, in the β-(BEDT-TTF)$_2I_3$ system a narrow Van-Hove singularity in the density of states is possible [19], but there is no reason why it should be exactly at the Fermi level. Therefore, we believe that the observed peak at 1 meV is due to a soft phonon, in accordance to indication from Raman data [14].

b) Pressure-Dependence of T_c

The pressure-dependence of T_c in BEDT-TTF$_2I_3$ is very strong [20]. The value of $\mathrm{d}\ln T_c/\mathrm{d}\ln V \simeq 13$ is significantly higher than in Pb, for example. From the curves of Fig. 4 we can estimate the theoretical pressure dependence. For curve *a*, with $\mu^*=0$, $\mathrm{d}\ln T_c/\mathrm{d}\ln\lambda \simeq 1.16$ around $T_c=8$ K; while for curve *b*, $\mathrm{d}\ln T_c/\mathrm{d}\ln\lambda \simeq 1.9$ around $T_c=1.3$ K. Taking $\mathrm{d}\ln\lambda/\mathrm{d}\ln\omega=2$, then for a Grüneisen constant $\gamma=-\mathrm{d}\ln\omega/\mathrm{d}\ln V$ of 4 to 6, characteristic of such molecular crystals [11], we obtain a contribution of $\mathrm{d}\ln T_c/\mathrm{d}\ln V=9.3$ for curve a at $T_c=8$ K, for the small value of $\gamma=4$. For curve b, at $T_c=1.3$ K, and the large value of $\gamma=6$ (because of the soft phonon), we obtain $\mathrm{d}\ln T_c/\mathrm{d}\ln V=23$. Thus, the phonon contribution accounts for most of the observed pressure-variation of T_c, even for curve a and a very conservative estimate of γ, in accord with the electron-phonon theory of superconductivity of organic metals [5]. There is also a contribution from the pressure-dependence of the electronic density of states. Using the observed value of the pressure-variation of the spin susceptibility [21], and assuming $\mathrm{d}\ln N(E_F)/\mathrm{d}P=\mathrm{d}\ln\chi_s/\mathrm{d}P=-3\%$/kbar, and the observed compressibility [22] of $\mathrm{d}\ln V/\mathrm{d}P=-1\%$/kbar, we obtain a contribution of $\mathrm{d}\ln T_c/\mathrm{d}\ln V=3.5$ from this source. Thus, together we can account quite well for the observed value of $\mathrm{d}\ln T_c/\mathrm{d}\ln V=13$. If the matrix element $\langle I^2\rangle$ (where $\lambda=\langle I^2\rangle N(\varepsilon_F)/(M\cdot\omega^2)$, M=mass of the molecule) would *increase* under pressure, as it is the case for a simple LCAO band, this would reduce $\mathrm{d}\ln T_c/\mathrm{d}\ln V$ and introduce some discrepancy. There is a proposed LCAO band structure for BEDT-TTF$_2I_3$ [6]. However, in inorganic non-transition metals, which are

not well described by the LCAO approximation, $\langle I^2 \rangle$ *decreases* with pressure. A preliminary band calculation [19] indicates that BEDT-TTF$_2$I$_3$ cannot be described by a simple LCAO band, and is closer to a nearly free electron description, like ordinary non-transition metals. Therefore, it seems that the pressure-dependence of T_c can be accounted for by the conventional BCS-Eliashberg theory.

Note however, that we *cannot* use the McMillan formula for T_c, which predicts saturation at large values of λ. The McMillan formula describes well "normal" phonon distributions, i.e. without soft phonons, for values of λ around 1 or lower. This is not the case here. There is an extension to very large values of λ by Allen and Dynes [23], which predicts that T_c increases like $\lambda^{1/2}$. (i.e. $\mathrm{d} \ln T_c / \mathrm{d} \ln \lambda = 1/2$). Here, T_c is seen (Fig. 4) to increase approximately linearly with λ, or even more steeply. The Allen-Dynes theory may perhaps apply at still larger values of λ.

c) *The Superconducting Gap*

In the superconducting state, the resistivity has a maximum at zero bias. This is characteristic of $S-N-S$ junctions, or even junctions with an insulating layer. The normal (or insulating) region may however be very thin, and not have a marked effect on the $I-U$ characteristics at temperatures above T_c.

As seen from Fig. 2 and Table 1 for the point contact method and crystals of **1** the width of the curve at half intensity, $2\Delta_{1/2}$, corresponds quite well to the BCS value. However, the width of the curve at the bottom, $2\Delta_T$, is about a factor of 2 larger. In contrast for the tunneling method and crystals of **2** the value of $2\Delta_T$ corresponds quite well to the BCS-theory while Hawley et al. [9] also for crystals of **2** reported a value 5 times larger than the BCS value, and attributed it to a soft phonon mode.

From theoretical considerations a soft phonon contributes to the gap at $T=0$, but does not contribute much to T_c. Thus, the value of $\Delta/k_B T_c$ is increased. We used the observed $\alpha^2 F$ curve, and the Eliashberg theory to calculate the enhancement of $\Delta/k_B T_c$ over the BCS (weak coupling) value using the Bergmann-Rainer computer program. We calculate an enhancement of about 10% for $T_c = 1.3$ K, and an enhancement of about 50% for $T_c = 8$ K. Since the temperature-dependence of the amplitude of the signal corresponds at most to $T_c = 1.3$ K, the enhancement of $\Delta/k_B T_c$ by the soft-phonon modes seems to be much too weak to account for the large value of Δ_T, for the point contact measurements for crystals of **1** as well as the data reported by Hawley et al. [9].

An alternative explanation is in terms of an anisotropic gap. The BTK theory applies to a "dirty" superconductor, with an isotropic gap. Here, the large resistance ratio (one thousand) indicates a mean-free-path of the order of 0.1 micron, while the correlation length $\xi \simeq \hbar v_F / \Delta$ is of the order of a few thousand Angstrom for $T_c = 1.3$ K, and even less for higher values of T_c. Therefore, we may have a "clean" superconductor, with an anisotropic gap. Preliminary band calculations [19] indicate that the fermi surface has several separate sheets, which obviously may possess different superconducting gaps. In such a situation, the average gap may correspond to the BCS value, and the maximum value of the gap may be considerably higher. Perhaps Δ_T corresponds to this maximum value, while $\Delta_{1/2}$ corresponds to the average gap.

For such a case, the crystallographic orientation of the contacts may be very important. Hawley et al. [9] already pointed out a difference between tunneling in the *c*-direction and in the ab plane. Our contacts are in the ab plane, however, there may be a considerable anisotropy in this plane as well. The preliminary band structure calculation [19] indicates a considerable anisotropy in the ab plane. More experimental data for contacts with different orientations in the ab plane are needed. In any case, we did not observe major inconsistencies with the predictions of electron-phonon theory of superconductivity. In particular our tunneling data on β-(BEDT-TTF)$_2$IAuI (*2*) are in very good accord with the conventional electron phonon theory of superconductivity.

We wish to thank Prof. Rainer for letting us make use of his computer program for the calculation of T_c, λ, and Δ.

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