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Comparative pressure studies of the superconducting transition temperature in isotopically substituted samples of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$

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Abstract

We determined the pressure dependence of the superconducting transition temperature, T_c, of the organic superconductor κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ for three different isotopic compositions using helium as a pressure medium. These measurements demonstrated that, given identical measurement conditions, the pressure dependence of T_c is independent of the isotopic composition of the material. Assuming that these isotopically different materials have an identical unit cell compressibility, it is found that for all three materials T_c scales linearly with the quasi-two-dimensional unit cell area, and is thus inversely proportional to the quasi-two-dimensional carrier density in κ-(BEDT-TTF)$_2$Cu(SCN)$_2$.
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Keywords: Pressure; Organic molecular metal; Organic superconductor; Superconducting transition temperature; Susceptibility

1. Introduction

κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is one of the best known and most thoroughly investigated organic superconductors. Its Fermi-surface topology is well known [1,2], and its phase diagram with its closeness and co-existence [3] of an antiferromagnetic and a superconducting ground state suggests a superconducting pairing mechanism driven by antiferromagnetic fluctuations [3,4]. Even though κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is known to possess quite a strong electron–phonon coupling between some intramolecular phonons and its conduction electrons [5], this coupling does not appear to be the dominant cause for superconductivity [5,6].

κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is a strongly anisotropic material in which conducting layers of (BEDT-TTF)$_2^{+1}$ in the crystallographic *bc*-plane are separated by insulating layers of polymorphic Cu(SCN)$_2^{-1}$ [1,2,7]. The resulting strong anisotropy is reflected in all physical properties of the material, including its electrical conductivity [1,2,7], its compressibility [8] and its uniaxial

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pressure dependence of T_c [9]. κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$ is a typical example of a quasi-two-dimensional superconductor. However, recent studies [10,11] have discussed and investigated the importance of the out-of plane direction, the crystallographic a'-direction perpendicular to the bc-plane, for a more general understanding of the general physical properties of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$.

The interest in the inverse isotope effect upon deuteration of the organic molecule BEDT-TTF, where BEDT-TTF is bisethylenedithio-tetrathiafulvalene, received renewed interest, when Biggs et al. [11] found that the pressure dependence of its superconducting properties differed strongly from those of the protonated material [12]. The difference in the pressure dependence of T_c was assumed to be caused by the difference in pressure-induced changes in the Fermi-surface topology [11]. The negative isotopic effect was assumed to be due to small differences in the Fermi-surface topology caused by isotopic substitution [13]. Others had attributed the inverse isotope effect upon deuteration to internal, uniaxial lattice pressure effects [14]. The different pressure dependence of T_c upon deuteration [11] differed strongly from earlier measurements by Schirber et al. [15], who had investigated the structurally similar material κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl utilizing helium as a pressure medium and had found no variation of the pressure dependence with the isotopic composition of the organic superconductor. Systematic studies of the isotope effects in κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$ [16] found that the isotopic substitution of the ethylene carbon sites with ^{13}C and the sulphur sites with ^{34}S in addition to the deuteration of the ethylene sites (see Fig. 1), causes a combined isotope effect: an inverse isotope effect due to deuteration and a normal isotope effect due to the ^{13}C and ^{34}S substitution.

In the present paper we present comparative ac-susceptibility measurements of T_c under pressures of up to 5 kbar on three different isotopic compositions of the organic superconductor κ-(BEDT-TTF)$_2$Cu(SCN)$_2$; we find that this material exhibits a strong pressure dependence of T_c ($dT_c/dP \simeq -(3.93 \pm 0.26)$ K kbar^{-1}) which is independent of the isotopic composition. For all

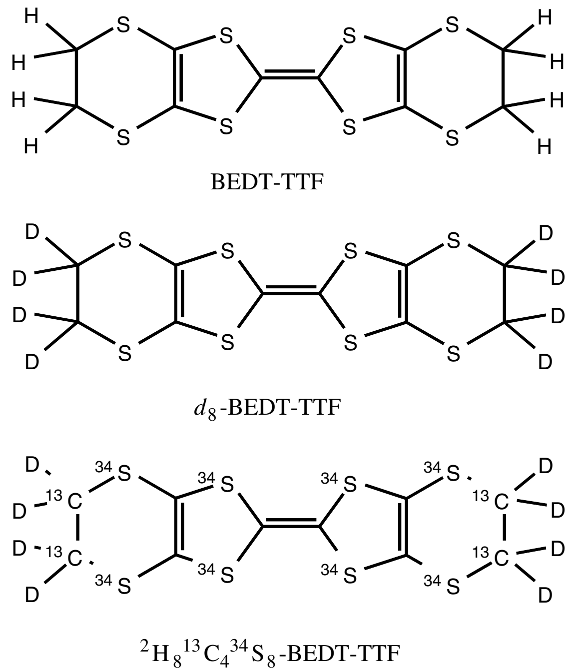

Fig. 1. The organic molecule BEDT-TTF in its protonated form (h_8) and the d_8 and $^2H_8^{13}C_4^{34}S_8$ molecules indicating the positions of the isotopic substitutions.

crystals the pressure dependence of T_c is found to be independent of the temperature of the pressure change. Furthermore, T_c is found to scale linearly with the pressure induced change in the in-plane area.

2. Experimental

Single crystals of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ with different isotopic composition were produced through standard electro-crystallization techniques [16]. Measurements were performed on three crystals of isotopically different composition of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$: one crystal (h_8) of $\sim 1.8 \times 0.76 \times 0.66$ mm^3 and $m = 1.20$ mg, where all the ethylene end-groups in the organic molecule were occupied by ^{1}H, one crystal (d_8) of $\sim 1.5 \times 1.3 \times 0.3$ mm^3 and $m = 0.89$ mg, with all those end-groups being occupied by ^{2}H (deuterium) and one crystal ($^2H_8^{13}C_4^{34}S_8$) of $\sim 1.5 \times 1.0 \times 0.45$ mm^3 and $m = 1.065$ mg, with additional isotopic substitutions on the ethylene carbon and sulphur sites (^{13}C and ^{34}S) (see Fig. 1). The crystals are shaped like thin plates, with the highly conducting *bc*-plane parallel to the large crystal face and orthogonal to the interplane direction. The h_8-crystal is from the same sample batch as the one investigated in [17]. The single crystalline character on the d_8 sample was verified in a backscattering Laue diffraction experiment [18].

The superconducting transition temperature, T_c, was measured as a function of pressure by ac-susceptibility in a helium gas pressure cell at Washington University in St. Louis. A Harwood Engineering gas pressure system with helium gas as a pressure medium was used for pressure generation. The pressure in the cell was monitored at all temperatures with a temperature-compensated Manganin gauge mounted inside the pressure circuit at room temperature. The crystals inside the pressure cell were mounted in a crystal holder, orienting each crystal with the ac-magnetic field approximately parallel to the in-plane direction [19]. The ac-susceptibility measurement system is described in detail elsewhere [20]. The measurements on all three crystals were performed independently, i.e. $T_c(P)$ for one crystal was measured, then the pressure was released, the cell opened and the crystal removed and the next crystal was inserted into the ac-coil system mounted inside the pressure cell. For all three samples the pressure in the cell was changed at a convenient temperature above the melting curve of helium, i.e. the pressure in the cell was changed at sample temperatures ranging from 9.5 K to room temperature.

Ambient pressure and selected high-pressure measurements were carried out using a two-stage closed cycle refrigerator (Balzers) to 7 K. The majority of the high-pressure measurements were executed using a Janis Varitemp liquid helium cryostat to 1.25 K. In all cases great care was taken when cooling the cell through the melting curve of helium. The temperature of the sample in the pressure cell is taken as the average of the temperatures measured simultaneously at the top and bottom of the cell with a set of calibrated Ge-thermometers. Care was taken to minimize the temperature difference between the top and bottom of the cell to less than 100 mK at all temperatures except near the melting temperature of helium.

3. Experimental results

3.1. Studies on κ-(h_8-BEDT-TTF)$_2$Cu(SCN)$_2$

Fig. 2 shows the real part of the ac-susceptibility at several pressures for κ-(h_8-BEDT-TTF)$_2$-Cu(SCN)$_2$, the protonated sample. The magnetic field is applied parallel to the highly conducting *bc*-plane. There are three principal observations: (i) the superconducting transition temperature, T_c, is decreasing with increasing pressure, (ii) the size of the superconducting transition, $\Delta\chi'$, is seen to increase with pressure and (iii) the superconducting transition is seen to become less broad with increasing pressure.

$T_c(P)$ is defined by the intersection of the two tangentials in $\chi'(T)$, as demonstrated in Fig. 3 for the transition at $P = 1$ kbar. With the ac-magnetic field in the in-plane direction, the superconducting transition is seen to be relatively broad: the transition at $P = 1$ kbar with a $T_c = (5.82 \pm 0.03)$ K is still not fully completed at 1.5 K. The width of the transition is no reflection of the sample quality,

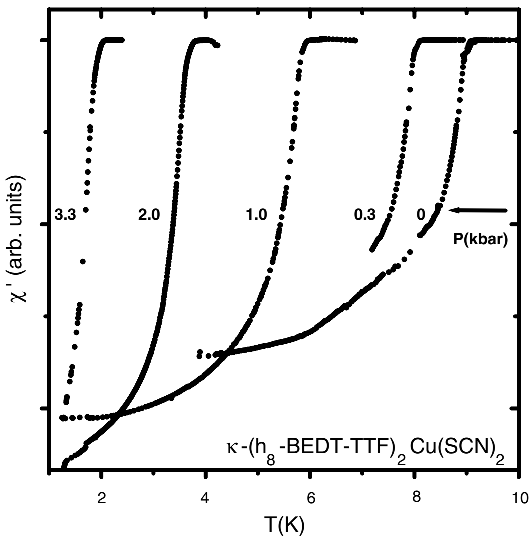

Fig. 2. Temperature dependence of the real part, χ', of the ac-susceptibility for κ-(h_8-BEDT-TTF)$_2$Cu(SCN)$_2$ with $H_{AC}^{(r.m.s)} = 1$ Oe and $\nu = 1.023$ kHz, $H_{AC}\|bc$-plane.

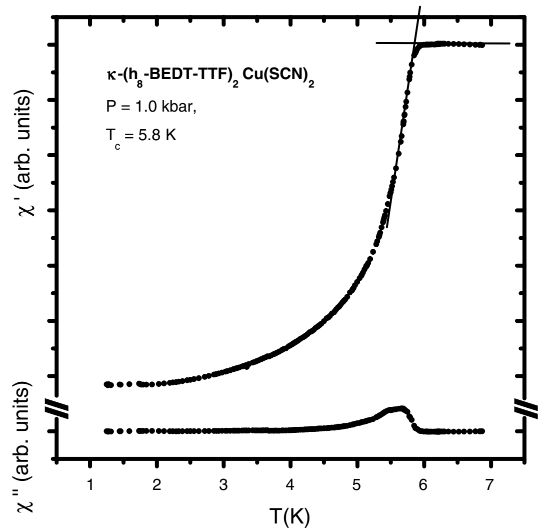

Fig. 3. Temperature dependence of the real $\chi'(T)$ and imaginary $\chi''(T)$ parts of the ac-susceptibility of κ-(h_8-BEDT-TTF)$_2$Cu(SCN)$_2$ at $P = 1$ kbar.

but is due to the temperature dependence of the interplane London penetration length, as will be discussed later.

The results for all our measurements on κ-(h_8-BEDT-TTF)$_2$Cu(SCN)$_2$ are compared to those in the literature [17,21] in Fig. 4. The numbers next to our data points indicate the order of measurement and the temperature at which the pressure was changed. It is clearly seen that the value of T_c is independent of the latter. As indicated in point '1', an initial pressure of $P = 4.1$ kbar applied at room temperature (RT) fully suppresses T_c below our base temperature of 1.25 K and no traces of superconductivity can be seen in the ac-susceptibility. The agreement of our data with those in the literature [17,21,22] obtained under similar pressure conditions, i.e. using helium gas pressure, is excellent.

A polynomial fit to the data in Fig. 3:

$$\begin{aligned} T_c(K) = {} & (9.17 \pm 0.02) - (4.08 \pm 0.09) \cdot P\ (\text{kbar}) \\ & + (0.79 \pm 0.07) \cdot P\ (\text{kbar})^2 \\ & - (0.07 \pm 0.01) \cdot P\ (\text{kbar})^3 \end{aligned} \quad (1)$$

agrees within its error bars with those in the literature [17,21].

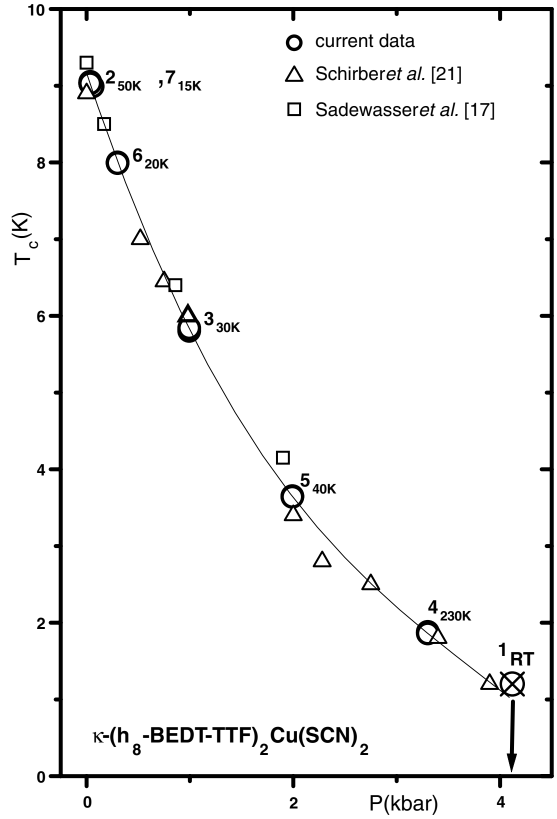

Fig. 4. Dependence of T_c on hydrostatic pressure for κ-(h_8-BEDT-TTF)$_2$Cu(SCN)$_2$ using helium gas as a pressure medium. The ○ indicate our data, the □ and △ the literature data from [17,21], respectively. The numbers give the order of measurement, their subscript indicates the temperature of the pressure change. The solid line is a polynomial fit to the current data.

3.2. Studies on *κ-(d_8-BEDT-TTF)$_2$Cu(SCN)$_2$ and κ-($^2H_8^{13}C_4^{34}S_8$-BEDT-TTF)$_2$Cu(SCN)$_2$*

Fig. 5 shows the temperature dependence at several selected pressures of the real and imaginary parts of the ac-susceptibility for κ-(d_8-BEDT-TTF)$_2$Cu(SCN)$_2$ and κ-($^2H_8^{13}C_4^{34}S_8$-BEDT-TTF)$_2$-Cu(SCN)$_2$, respectively. Again for both crystals the size of $\Delta\chi'$ increases slightly with increasing pressure while the width of the superconducting transition decreases.

The superconducting transition temperature decreases with increasing pressure (Fig. 6). Again, no dependence of $T_c(P)$ on the temperature of the

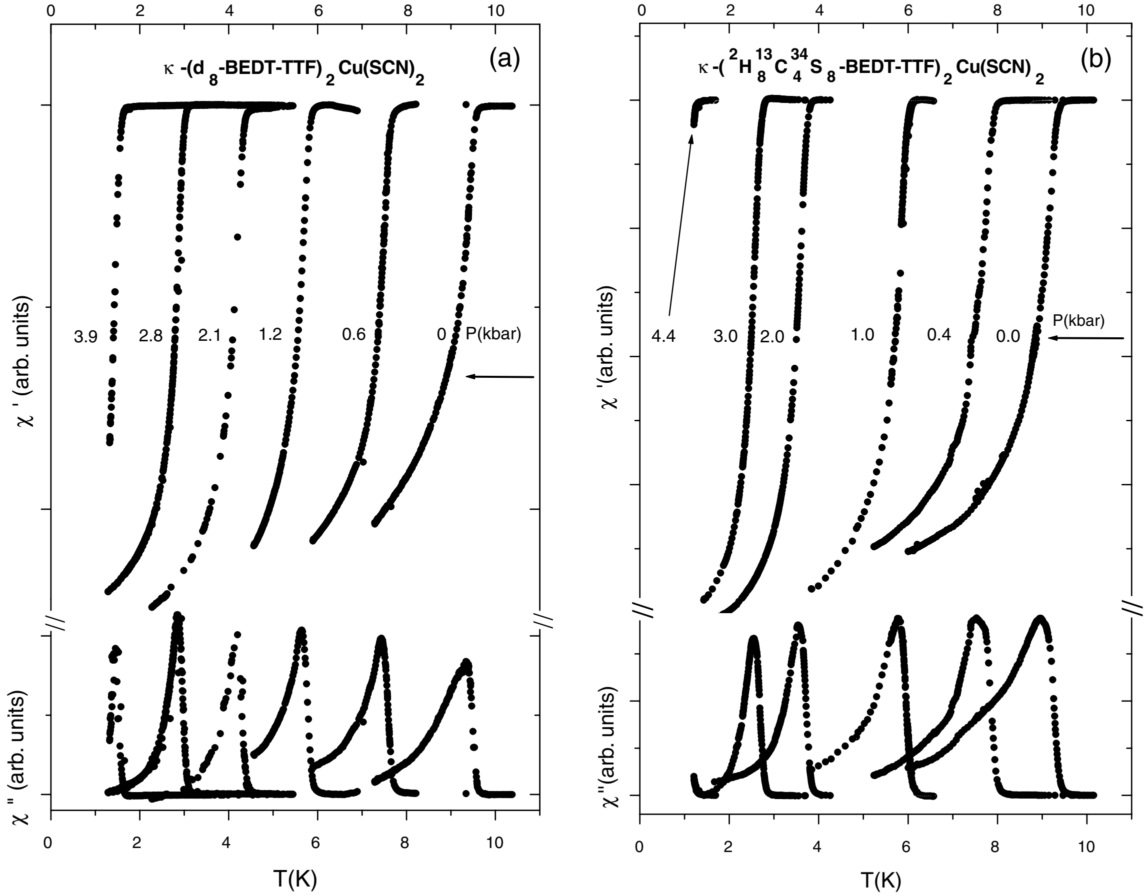

Fig. 5. Temperature dependence of the real χ' and imaginary χ'' parts of the ac-susceptibility of (a) κ-(d_8-BEDT-TTF)$_2$Cu(SCN)$_2$ ($m = 0.89$ g) and (b) κ-($^2H_8^{13}C_4^{34}S_8$-BEDT-TTF)$_2$Cu(SCN)$_2$ ($m = 1.066$ mg) at 6 representative pressures, respectively. $H_{AC}^{(r.m.s)} = 1$ Oe and $\nu = 1.023$ kHz with $H_{AC} \| bc$-plane.

pressure change was seen for either sample. A polynomial fit to T_c vs. pressure (kbar) on the d_8-crystal yields:

$$\begin{aligned} T_c(K) = {} & (9.52 \pm 0.06) - (3.88 \pm 0.19) \cdot P \text{ (kbar)} \\ & + (0.76 \pm 0.11) \cdot P \text{ (kbar)}^2 \\ & - (0.07 \pm 0.02) \cdot P \text{ (kbar)}^3, \end{aligned} \tag{2}$$

and for the ($^2H_8^{13}C_4^{34}S_8$)-crystal yields:

$$\begin{aligned} T_c(K) = {} & (9.40 \pm 0.06) - (3.82 \pm 0.15) \cdot P \text{ (kbar)} \\ & + (0.68 \pm 0.09) \cdot P \text{ (kbar)}^2 \\ & - (0.05 \pm 0.01) \cdot P \text{ (kbar)}^3. \end{aligned} \tag{3}$$

4. Discussion

Isotopic substitution in the organic molecule BEDT-TTF has the effect of changing the ambient pressure value of T_c [16]. Those ambient pressure values of T_c agreed well with the ones quoted in the literature [16]. Our current measurements as well as those from Ref. [16] are inductive measurements, and can thus be compared directly. Inductive values for T_c are known to be lower than those frequently quoted for the resistive onset due to the difference between the volume and filamental character of the transition probed using the two different measurement techniques.

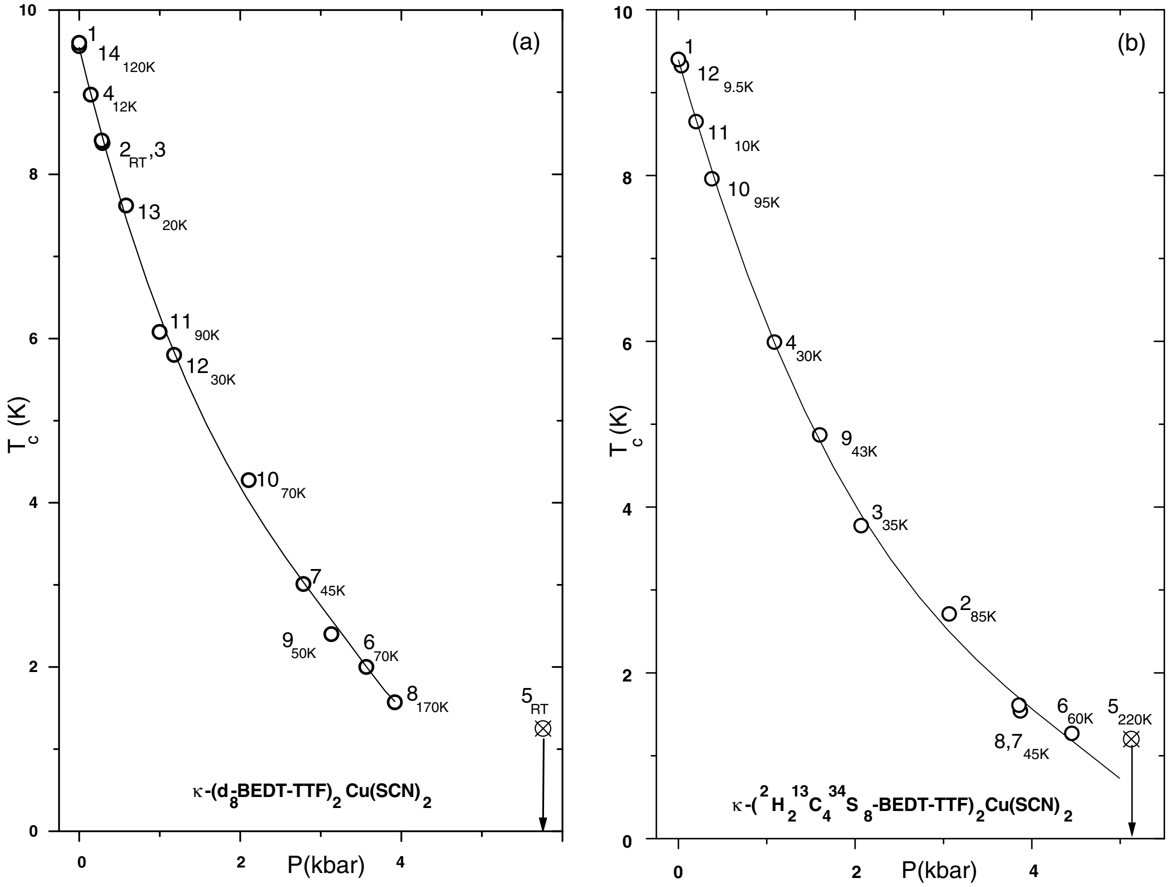

Fig. 6. Dependence of T_c on hydrostatic pressure for (a) κ-(d_8-BEDT-TTF)$_2$Cu(SCN)$_2$ and (b) κ-($^2H_8^{13}C_4^{34}S_8$-BEDT-TTF)$_2$Cu(SCN)$_2$ using helium gas as a pressure medium. The numbers give the order of measurement, their subscript indicate the temperature of the pressure change. The solid line is a polynomial fit to the current data. For both samples point '5' indicates the suppression of superconductivity below $T \simeq 1.25$ K.

In all three samples the size of the superconducting transition, $\Delta\chi'$, is increasing with increasing pressure. This is thought to be a direct result of a pressure induced reduction in the large interplane London penetration length, ($\lambda_a(0)_{P=0\,\mathrm{kbar}} = 40$ μm [23]), which is a significant fraction ($\geqslant 10\%$) of the interplane sample dimension; the London penetration length, λ_i, is a measure of the distance over which an external magnetic field is expelled from the interior of a superconductor. The sample is thus not exhibiting perfect diamagnetism over a circumference of the order of λ_i and thus in those parts not contributing fully to the diamagnetic signal measured by $\Delta\chi'$. With λ_a being a significant fraction of the interplane sample dimension, the volume fraction of the sample not exhibiting perfect diamagnetism is significant and the signal size is visibly reduced. A decrease in λ_a with increasing pressure is resulting in a larger portion of the sample displaying perfect diamagnetism and thus a larger signal size in $\Delta\chi'$.

The broadness of the transition for all three samples does not reflect on the sample quality but is a direct result of the large interplane London penetration length and its temperature dependence. The sharpening of the transition under pressure can also be directly linked to a decrease of λ with increasing pressure.

In contrast to the ambient pressure value, the pressure dependence of T_c does not seem to be affected by the isotopic substitution. Within their error bars, Eqs. (1)–(3) parameterize the same pressure dependence of T_c and agree well with earlier T_c–P measurements where helium was used as a pressure medium [17,21,22]. The results, however, differ strongly from other earlier measurements [11,12] which indicated that the deuterated sample has a much stronger pressure dependence than the protonated material. The latter conclusions were based on the comparison of magnetotransport measurements on d_8- and h_8-crystals, where the different crystals were exposed to two different quasi-hydrostatic pressure media, i.e. petroleum spirit and Fluorinert, respectively. Using the identical pressure medium, i.e. helium, for the pressure-measurements on all crystals as well as identical measurement conditions in the measurements presented here suggests that the earlier results [11,12] were affected by external factors and did not fully reflect the intrinsic properties of the material under investigation.

κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is a highly anisotropic material with respect to all of its physical properties. Special attention should be paid to its compressibility [8] as well as its uniaxial dT_c/dP_i [9]. These make this material extremely sensitive to even very small non-hydrostatic effects. In addition, κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ is a very soft material [8] and can thus be most easily strained if the pressure conditions are not fully hydrostatic. In measurements of C_{60}-superconductors, helium was seen to partially penetrate the structure [24]. Our present measurements cannot rule out that something similar is happening in κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$. However, penetration of the sample with helium should to some degree depend on the density of helium surrounding the sample, and thus on the temperature at which the pressure was changed. Our current result, that $T_c(P)$ is totally independent of the temperature of the pressure change, thus makes it unlikely that helium is penetrating the sample upon applying pressure.

The fact that the measurements presented in this paper agree in every aspect with much older, independent results [17,21,22], indicates that the use of helium as a pressure medium allows one to obtain reproducible and transferable results, i.e. helium thus seems to be highly suitable as a pressure medium for a material as soft and anisotropic as the organic superconductors.

$T_c(P)$ for all three isotopic compositions investigated here does not depend linearly on pressure as claimed before [11,12,22] but exhibits a concave slope (see Figs. 4 and 6). Such a pressure dependence of T_c is not uncommon, i.e. it has also been observed in the strongly anisotropic, but electronically three-dimensional superconductor MgB_2 [25]. In MgB_2 a linear dependence could be restored upon plotting T_c against the relative unit cell volume V/V_0 [25].

Structural data under pressure for κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ are restricted to room temperature for an h_8 sample with Fluorinert as a pressure medium [8]. Given the present result that within the error of measurement the pressure dependence of T_c is identical for all three isotopic compositions investigated, it seems fair to assume that the isotopic composition does not affect the compressibility of the material. Thus using the data from [8] for all isotopic compositions and the fact that the lattice parameters for all three materials are essentially the same [26], the dependence of T_c on the unit cell parameters can be analyzed using the present $T_c(P)$ data and the structure data from Ref. [8].

It is interesting to note that in κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ the concave pressure dependence of T_c is replaced by a linear appearance when T_c is plotted against the two-dimensional unit cell size in the *bc*-plane (Fig. 7). (Plotting T_c against the unit cell volume results in a convex curvature and is not shown here.) We will now discuss whether there might be a reasonable explanation for this linearity: the linear volume dependence of T_c in an anisotropic, but truly three-dimensional superconductor, i.e. MgB_2, appears to be replaced by the equivalent behaviour in our quasi-two-dimensional superconducting material, i.e. the size of the two-dimensional unit cell. MgB_2 is recognized as a truly BCS-type superconductor [27] and the linear dependence of T_c on the unit cell volume can be traced to the logarithmic volume dependence of its electron–phonon coupling parameter, λ [25,28,29]. This volume dependence of λ in MgB_2 is a direct

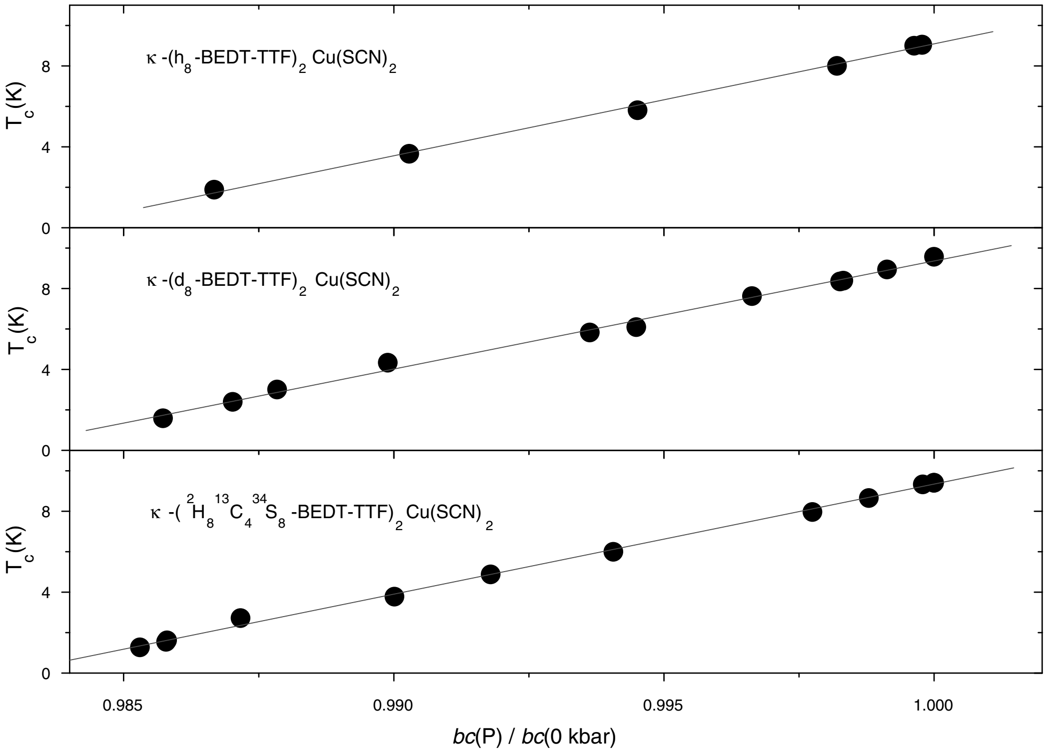

Fig. 7. Dependence of T_c on the renormalized size of the in-plane unit cell.

result of the volume dependence of the electron density, $N(E_F)$ [25,28,29]. κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$, on the other hand, is considered to be an example of a d-wave superconductor mediated by antiferromagnetic fluctuations [3] and even though there is strong electron–phonon coupling present in the material, the latter interaction is not thought to be the dominant cause of superconductivity [5]. For both coupling mechanisms, however, the electron density, $N(E_F)$, is a relevant parameter for the superconducting transition temperature, and a possible candidate for the underlying cause for a similar linear dependence of T_c on the quasi-n-dimensional unit cell volume.

This very simple relationship between T_c and the two-dimensional unit cell area is very surprising, as it suggests that the out-of plane direction is not highly significant for the superconducting properties of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$. Such a suggested behaviour contradicts thermal expansion measurements [9] that indicated that the uniaxial pressure dependence dT_c/dP_i is largest for the out-of plane direction. More experimental evidence in support of or against the simple relationship found in our current investigation is needed.

κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ has two holes per unit cell [7]. At the small pressures used in our experiments, this overall carrier density per unit cell can be considered independent of pressure. The size of the in-plane unit cell area is thus inversely proportional to the carrier density per unit area. Thus a linear dependence of T_c on the unit cell area is simply indicating that T_c is inversely proportional to the carrier density in the material. The increase in the quasi-two-dimensional carrier density with increasing pressure is not unexpected, as it is well known that κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ becomes more metallic under pressure while superconductivity is being suppressed [1,2,12]. It is also well known that especially the in-plane conductivity increases strongly under pressure ($\sigma_0(P) = \sigma_0 \times (1 + 8 \times P \text{ (GPa)})$) [6]. In this paper we reported the pressure-induced decrease of the London penetration length ($\lambda_i \sim (m_s/N_s)^{1/2}$), where m_s is the mass of the superconducting carriers and N_s the superconducting carrier density. Such a decrease of λ is fully consistent with a pressure-induced increase of the superconducting carrier density, N_s. Considering these facts, it is not surprising to discover that T_c is decreasing with increasing carrier density in the material.

5. Summary

Ac-susceptibility measurements under pressure, with helium as a pressure medium, of different isotopic compositions of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ demonstrated that, given identical measurement conditions, the pressure dependence of the superconducting transition temperature, T_c, is independent of the isotopic composition of κ-(BEDT-TTF)$_2$Cu(SCN)$_2$. The measured concave pressure dependence of T_c results in a linear dependence of T_c on the size of the in-plane two-dimensional unit cell size, or a simple inverse correlation between T_c and the carrier density in this organic superconductor.

Acknowledgements

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