

Comparative magnetotransport and T_c measurements on κ -(BEDT-TTF) $_2$ Cu(SCN) $_2$ under pressure

A-K Klehe^{1,3}, T Biggs¹, C A Kuntscher^{1,4}, A M Kini² and J A Schlueter²

¹ Clarendon Laboratory, Department of Physics, Oxford University, Parks Road, Oxford OX1 3PU, UK

² Materials Science Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439, USA

E-mail: a.klehe1@physics.ox.ac.uk

Abstract

We compare magnetotransport measurements under pressure on the organic superconductor κ -(BEDT-TTF) $_2$ Cu(SCN) $_2$ (BEDT-TTF \equiv bis(ethylene-dithio)tetrathiafulvalene) with different pressure media and discover that the results are pressure media dependent. This pressure medium dependence is thought to originate from the difference in thermal contraction between the very soft and highly anisotropic sample and the isotropically contracting, but solid pressure medium, thus resulting in non-hydrostatic pressure on the sample. However, comparison of pressure measurements with different media reveals a pressure medium independent correlation between the superconducting transition temperature, T_c , and the size of the quasi-two-dimensional Fermi surface pocket and thus the quasi-two-dimensional carrier density in κ -(BEDT-TTF) $_2$ Cu(SCN) $_2$. The observed pressure-induced increase in the quasi-two-dimensional carrier density can be interpreted as a transfer of carriers from quasi-one-dimensional Fermi surface sections, reminiscent of a mechanism in cuprate superconductors, where pressure is known to transfer carriers from the insulating charge reservoir layers into the conducting cuprate sheets.

1. Introduction

κ -(BEDT-TTF) $_2$ Cu(SCN) $_2$ (BEDT-TTF \equiv bis(ethylenedithio)tetrathiafulvalene) is one of the best characterized organic superconductors [1–3]. The resemblance of its pressure–temperature phase diagram [4] to the carrier density–temperature phase diagram in cuprate

³ Author to whom any correspondence should be addressed.

⁴ Permanent address: 1. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, D-70550 Stuttgart, Germany.

superconductors [5] has frequently been taken as evidence for similar interaction mechanisms governing superconductivity [6, 7]. The magnetic ordering, metallic behaviour and superconductivity in cuprate superconductors are determined by the carrier concentration in the conducting cuprate layers [5]. This carrier concentration can be regulated either through chemical doping [5] or pressure [8, 9]. In organic superconductors, however, evidence for a correlation between the carrier density and the superconducting transition temperature, T_c , has been lacking.

κ -(BEDT-TTF)₂Cu(SCN)₂ is a typical example of a quasi-two-dimensional superconductor. It 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 [1–3]. The resulting strong anisotropy is reflected in all physical properties of the material, including its electrical conductivity [1–3], its quasi-two-dimensional band structure [2, 3], its compressibility [10], its thermal contraction [11] and its uniaxial pressure dependence of T_c [12].

Also the Fermi surface of κ -(BEDT-TTF)₂Cu(SCN)₂ reflects the structural anisotropy: it consists of a quasi-two-dimensional (Q2D) Fermi surface pocket and two quasi-one-dimensional (Q1D) Fermi surface sections [1–3]. The Q2D Fermi surface pocket, also known as the α -pocket, gives rise to Shubnikov–de Haas oscillations of ~ 600 T at ambient pressure. At high magnetic fields and low temperatures, magnetic breakdown between the α -pocket and the Q1D Fermi surface sections can give rise to a semiclassical orbit, the β -orbit, which has the same cross sectional area as the Brillouin zone [2, 3]. Thus, this orbital size is a measure of the low temperature in-plane compressibility of the material. In conjunction with the size of the α -orbit, it also allows one to calculate the exact shape of the Q2D Fermi surface according to the effective dimer model [13]. The superconductivity in κ -(BEDT-TTF)₂Cu(SCN)₂ is thought to be strongly affected by the detailed warping of the Fermi surface [6, 14], with the pairing of the electrons in the Q2D band being mediated by an exchange of spin fluctuations within the Q1D band [14]. A highly critical parameter for the material is the ratio of the interdimer transfer integrals t_b/t_c [14], which has been predicted [14] and observed [15] to increase under pressure. This pressure-induced increase is currently considered to be the driving force of the different pressure-induced phase transitions observed in κ -(BEDT-TTF)₂Cu(SCN)₂ [14]. Recent studies [13, 15] have also 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)₂Cu(SCN)₂. The interlayer transfer integral, $t_{\perp} \approx 0.04$ meV, was found to be a factor $\leq 10^3$ smaller than that observed for the intralayer components, t_b and t_c [13], indicating that κ -(BEDT-TTF)₂Cu(SCN)₂ is predominantly Q2D in its electronic properties, even though there is a small degree of coherent interlayer transport [13]. The warping of the Fermi surface in the interplane direction is so small [13] that the Landau quantization is almost entirely determined by the field perpendicular to the two-dimensional planes, thus allowing an easy correction of the measured Shubnikov–de Haas frequencies in a tilted magnetic field [2]. Further evidence for the predominantly Q2D character of κ -(BEDT-TTF)₂Cu(SCN)₂ is the absence of ‘neck-and-belly’ frequencies in the de Haas–van Alphen effect [16] and the intrinsically broad width of the superconducting-to-normal transition in zero-field resistance measurements [17].

Uniaxial pressure measurements [18] and calculations [19] have been performed on κ -(BEDT-TTF)₂Cu(SCN)₂, and indicated a strongly anisotropic pressure dependence of its physical properties. The observed increase of the Q2D Fermi surface area under pressure [15, 20, 21] as well as under uniaxial stress in the out-of-plane direction [18] is thought to originate from a pressure-induced intradimer sliding motion [19].

The value of T_c exhibits an inverse isotope effect upon deuteration of the ethylene groups of the organic molecule (BEDT-TTF). The origin of this effect is not understood,

but differences in Fermi surface warping [22] or internal, uniaxial lattice pressure effects [23] have been suggested. Isotope substitution on any of the other atoms in the organic molecule results in a small positive or no isotope effect [24]. The pressure dependence of T_c in κ -(BEDT-TTF)₂Cu(SCN)₂, however, is not affected by the isotope composition [25], as was determined in ac-susceptibility measurements where helium was used as a pressure medium for all samples. The latter result highlights that the difference in pressure dependence upon deuteration seen in [15] does not originate in the isotope composition, and that other experimental factors, such as the choice of pressure medium, might be relevant.

The main aim of this publication is to compare magnetoresistive measurements under pressure on κ -(BEDT-TTF)₂Cu(SCN)₂ using various pressure transmitting media, namely helium, Fluorinert [15] and petroleum spirit [20, 21]. Results obtained with Fluorinert and petroleum spirit differ from those with helium and with each other. This comparison highlights that the experimentally determined pressure is not a transferable parameter. When analysing the relationship between parameters determined simultaneously in each pressure experiment, we find that T_c exhibits a unique dependence on the Q2D carrier density in the material, independent of the pressure medium used. The increase of the Q2D carrier density can be understood as a charge carrier transfer from the Q1D sections of the Fermi surface to the Q2D hole pockets. Assuming that it is only the Q2D carriers that are superconducting [14], pressure applied to κ -(BEDT-TTF)₂Cu(SCN)₂ can be understood to increase the carrier concentration of the superconducting carriers; this is similar to the pressure-induced carrier increase observed in cuprate superconductors.

2. Experiment

Single crystals of κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ were grown by standard electrocrystallization techniques [24]. The sample of κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ investigated is from the same batch as that investigated in [25]. Magnetotransport measurements under pressure on the organic superconductor κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ were executed in a 15 T superconducting magnet from Oxford Instruments. The pressure cell used was a gas pressure cell from Unipress, attached to a Harwood Engineering compressor system with helium as the pressure medium. Above the melting curve of helium, the pressure in the cell is measured with a calibrated, temperature-compensated manganin gauge mounted in the pressure circuit at room temperature. The pressure in solid helium is calculated using the gas pressure measured and isochoric data [26]. During operation the pressure cell is permanently connected to a pressurized gas reservoir at room temperature via a CuBe capillary, thus reducing the pressure decrease in the cell during the cool down of the cell from room temperature to 1.5 K, the base temperature of our experiment.

The pressure cell is placed in the centre of the magnet bore inside a variable temperature insert. The temperature of the cell is regulated by varying the temperature of the variable temperature insert and by adjusting the power on a two-part heater placed directly on the outer cell body. Only the top part of the cell heater was heated during the transition of the pressure cell through the melting curve of helium, to artificially increase the temperature gradient over the pressure cell and thus to reduce strain in the sample [27].

Comparison of the ambient pressure Shubnikov–de Haas frequency [2] to that measured on the crystal mounted inside the pressure cell revealed that the latter was aligned with its out-of-plane axis $\sim 21^\circ$ off the magnetic field direction and corrections to the measured Shubnikov–de Haas frequencies were made accordingly [2]. The resistance of the crystal was measured in the interplane direction with a current of $I = 9 \mu\text{A}$ at a frequency of 33 Hz. The minimum temperature accessible in these measurements was 1.5 K and the maximum magnetic field

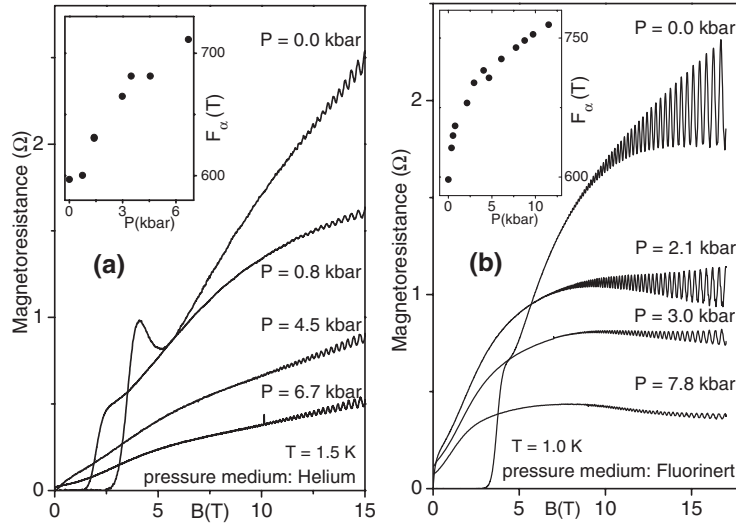


Figure 1. (a) The magnetoresistance, exhibiting quantum oscillations, of κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ at $T = 1.5$ K at selected pressures with helium as a pressure medium. At $T = 1.5$ K, the superconductivity at zero field is not suppressed until the pressure exceeds 4.5 kbar. The inset shows the increase of the α -frequency, $F_{\alpha}(T)$, with pressure. (b) The magnetoresistance of the same material, but using Fluorinert as a pressure medium [15]. In this pressure medium, the superconductivity is suppressed below $T = 1.0$ K at pressures exceeding 2.1 kbar. The inset shows again the increase of $F_{\alpha}(T)$ with pressure.

was 15 T. The temperature of the sample was determined with a RuO thermometer thermally anchored to the top of the pressure cell.

Details of the measurements on κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ with Fluorinert as a pressure medium can be found in [15]. The corresponding measurements on κ -(h₈-BEDT-TTF)₂Cu(SCN)₂ with petroleum spirit are described in detail in [20, 21].

3. Experimental results

Figure 1(a) shows the magnetoresistance of κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ at selected pressures using helium as a pressure medium. Clearly visible are quantum oscillations associated with the α -frequency. The pressure dependence of these oscillations is illustrated in the inset of figure 1(a). As is known from several previous measurements [15, 21] the size of the Q2D Fermi surface pocket increases with increasing pressure. The relatively high temperatures ($T \geq 1.5$ K) and the relatively small magnetic fields ($B \leq 15$ T) used in these measurements prevented us from observing the β -frequency (see section 1). At low pressures one can also see the well-known peak in the magnetoresistance, thought to originate from the motion of flux lines in the magnetic field [2]. Superconductivity in this measurement is seen to prevail above 1.5 K at pressures up to 4.5 kbar.

In comparison, in figure 1(b) the magnetoresistance under pressure of κ -(d₈-BEDT-TTF)₂Cu(SCN)₂ with Fluorinert as a pressure medium is shown. The quantum oscillations visible in these measurements are a combination of those originating from the Q2D α -pocket and the magnetic breakdown β -orbit. The inset in figure 1(b) shows the development of the α -frequency with pressure. Details of those measurements can be found in [15]; they show that with Fluorinert as a pressure medium, T_c is suppressed below $T = 1$ K at pressures

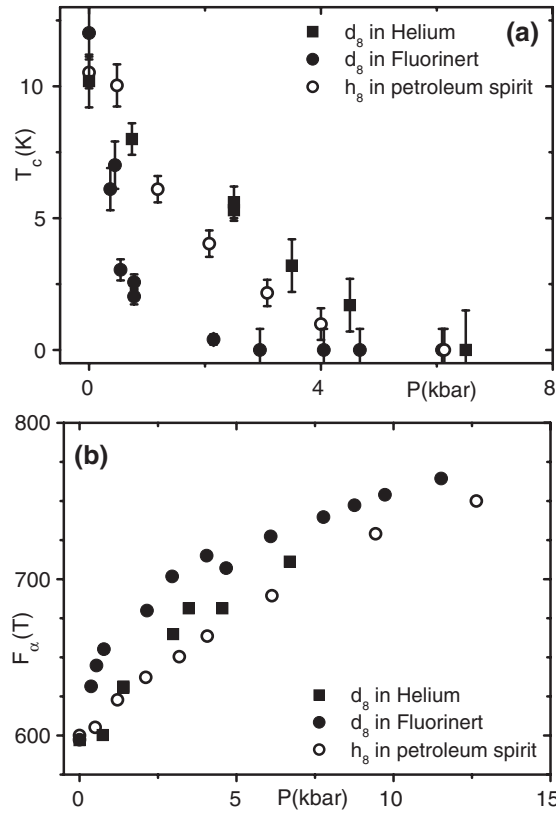


Figure 2. (a) T_c and (b) the α -frequency, $F_\alpha(T)$, as a function of pressure for κ -(d_8 -BEDT-TTF) $_2$ Cu(SCN) $_2$ using helium (■) and using Fluorinert (●) [15] and for κ -(h_8 -BEDT-TTF) $_2$ Cu(SCN) $_2$ using petroleum spirit (○) [21] as a pressure medium.

exceeding 2.1 kbar. This value is much smaller than that obtained from our measurements with helium as a pressure medium.

4. Discussion

4.1. The effect of the pressure medium

Isotopic substitution does not affect the pressure dependence of T_c [25]. Thus, the magnetoresistance data under pressure on κ -(h_8 -BEDT-TTF) $_2$ Cu(SCN) $_2$ [20, 21] using petroleum spirit as a pressure medium are included in the comparison seen in figure 2. Figure 2 compares measurements made with three different pressure media: helium, Fluorinert and petroleum spirit. The three measurement sets indicate three different pressure dependences and only agree in their general trends: (i) T_c is decreasing and (ii) the α -frequency, $F_\alpha(T)$, is increasing with increasing pressure. Having ruled out the possibility that the isotope composition [25] or the source of the sample [15] could affect the pressure dependence, one has to conclude that the pressure medium has to be responsible for the difference in pressure dependence observed in κ -(BEDT-TTF) $_2$ Cu(SCN) $_2$. No evidence for shear stresses on the sample could be observed in any of those measurements, i.e. the superconducting transitions did not broaden under pressure nor could a reduction of the amplitude of quantum oscillations

as a function of pressure history be observed. There was also no evidence in any of those measurements of any sample deterioration after the pressure measurements, which could have indicated a chemical reaction between the sample and pressure medium.

The pressure media were selected because of their quasi-hydrostatic and hydrostatic properties, but all will have solidified at the temperatures at which most T_c and quantum oscillation measurements were executed. Thus, in most measurements, the sample investigated is submerged in a pressurized solid during the measurement. The pressure felt by the sample depends on the difference in the thermal contractions of the pressure cell, the pressure medium and the sample itself [27]. κ -(BEDT-TTF)₂Cu(SCN)₂ is a very soft [10] material with a highly anisotropic thermal contraction [11]. The difference in its anisotropic thermal contraction from that of the uniformly contracting pressure medium results in non-hydrostatic stresses on the sample upon cooling. Evidence for this effect of non-hydrostatic, pressure medium-induced stresses on a soft sample with an anisotropic compressibility [28] are well documented for zinc [27, 29]. Thus, similar effects of a frozen pressure medium on the soft and highly anisotropic [10] κ -(BEDT-TTF)₂Cu(SCN)₂ are not surprising.

Independent measurements in different pressure cells by different groups [15, 30] with Fluorinert as a pressure medium give the same pressure dependence of T_c . This indicates that the non-hydrostatic pressure conditions caused by the difference in thermal contraction between a very soft, anisotropic sample and the pressure medium can be reproducible. Also measurements of dT_c/dP on κ -(BEDT-TTF)₂Cu(SCN)₂ using helium as a pressure medium give, within the error bars, identical results, as can be seen by comparing the results obtained in [25] and those of the present measurements, but they are different from those obtained with the other pressure media (see figure 2).

Helium is generally considered to be the most hydrostatic and the softest pressure medium, even when it is solidified under pressure [26, 27]. At $P = 1$ kbar, solid helium has a bulk modulus, $B_{\text{solid helium}} \sim 6$ kbar [26], compared to an estimated $B_{\text{sample}} \sim 123$ kbar [10] for κ -(BEDT-TTF)₂Cu(SCN)₂. One could thus argue that those measurements with helium as a pressure medium reflect the most accurate hydrostatic pressure dependence of κ -(BEDT-TTF)₂Cu(SCN)₂. Helium, on the other hand, has been known to penetrate the structure of some materials upon use as a pressure medium [31]. Recent experiments [25], however, indicated that helium penetration into the structure of κ -(BEDT-TTF)₂Cu(SCN)₂ under pressure is unlikely, due to the insensitivity of T_c to the temperature of the pressure change, even though the possibility of intercalation cannot be excluded.

In summary, it appears from figure 2 and the above discussion that the experimentally determined pressure cannot be taken as a reliable parameter for describing the physical properties of κ -(BEDT-TTF)₂Cu(SCN)₂. This is most probably due to the fact that fully hydrostatic conditions of the pressure medium at low temperatures cannot be guaranteed.

4.2. The pressure medium independent correlation between T_c and the quasi-two-dimensional carrier density, n_{Q2D}

Common to all measurements in figure 2 is the simultaneous determination of T_c and F_α under pressure. Figure 3 shows the correlation between these two parameters. Even though each of the measurements in figure 2 exhibits its own, individual pressure dependence of T_c and F_α , these parameters demonstrate a strong correlation to each other, seemingly independent of the pressure medium used and thus independent of the possible degree of shear on the sample. Thus, the strong correlation between the bulk property of superconductivity, as indicated by T_c , and the two-dimensional Fermi surface parameter, F_α , suggests that superconductivity in this organic superconductor is fully determined by processes in the

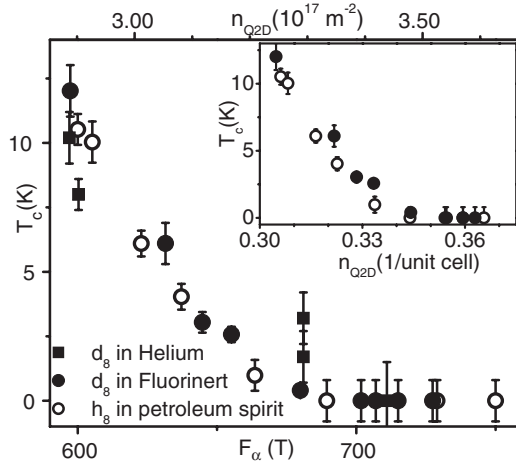


Figure 3. T_c and F_α from figure 2, indicating their correlation. The correlation is independent of the individual pressure dependence observed, and thus of the individual pressure scale. The inset shows T_c against the quasi-two-dimensional carrier density per unit cell, n_{Q2D} (1/unit cell).

quasi-two-dimensional conducting planes. Hence, warping effects of the Fermi surface in the interplane direction [15, 22] cannot be of major significance for the superconducting properties in κ -(BEDT-TTF)₂Cu(SCN)₂. A similar lack of dependence of the superconducting properties on the interlayer parameters was indicated in [25].

Figure 3 might partially explain the inverse isotope effect observed in κ -(d₈-BEDT-TTF)₂Cu(SCN)₂; the quantum oscillation frequency for the Q2D Fermi surface pocket is $F_\alpha = 600 \pm 1$ T for h₈ and $F_\alpha = 597 \pm 1$ T for d₈ samples, which although very close were consistently smaller for the d₈ sample [15, 32]. According to figure 3, a smaller F_α leads to a higher T_c , as is observed upon deuteration of the organic BEDT-TTF molecule. The origin of the decrease in F_α upon deuteration, however, is unknown. In addition, κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl with $T_c \approx 12.7$ K has a Q2D Fermi surface pocket with $F_\alpha \approx 577$ T [33]. This is again in agreement with the tendency for smaller quasi-two-dimensional Fermi surface areas to result in a higher T_c .

Considering that the degree of coherent transport in the interplane direction in κ -(BEDT-TTF)₂Cu(SCN)₂ is very small [13], we shall in the following treat it as a two-dimensional, 2D, electronic system: in a 2D electronic system, simple Fermi statistics correlates the frequency of the 2D Fermi surface pocket, F_α , with the carrier density, n_{Q2D} , of that pocket:

$$n_{Q2D} \text{ (m}^{-2}\text{)} = \frac{A_{FS}}{2\pi^2} \quad (1)$$

with

$$A_{FS} = \frac{2\pi e}{\hbar} F_\alpha.$$

The observed correlation between T_c and F_α is thus equivalent to a correlation between T_c and the Q2D carrier density, n_{Q2D} . n_{Q2D} which has been calculated according to equation (1) is plotted as the top x -axis in figure 3: the effect of pressure is to increase the number of carriers in the Q2D hole pockets in κ -(BEDT-TTF)₂Cu(SCN)₂. For the measurements in Fluorinert [15] and petroleum spirit [20] the in-plane compression of the unit cell at low temperatures has been determined from the pressure dependence of the β -frequency to be $\sim 4\%$ GPa⁻¹. This

compares to an overall increase in F_α or n_{Q2D} (m^{-2}) of $\sim 30\%$ in the same pressure region, indicating that n_{Q2D} (m^{-2}) increases beyond what would be expected from the compression of the Brillouin zone alone. Given the in-plane compressibility information [15, 20], n_{Q2D} (m^{-2}) for the Fluorinert and petroleum spirit measurements is converted to the carrier density per unit cell, n_{Q2D} (1/unit cell). It appears from the inset of figure 3 that n_{Q2D} (1/unit cell) increased from ~ 0.30 (holes/unit cell) to ~ 0.34 (holes/unit cell) when T_c was suppressed. Thus, in the temperature–pressure phase diagram of κ -(BEDT-TTF)₂Cu(SCN)₂ one can think of pressure as a driving force for increasing the Q2D carrier density, n_{Q2D} (m^{-2}). However, whereas experimentally determined pressure is in general not a suitable parameter for describing the superconducting state of the system (see figure 2), the carrier density, n_{Q2D} , obtained from F_α and the in-plane compressibility, predicts T_c extremely well.

κ -(BEDT-TTF)₂Cu(SCN)₂ has two holes per unit cell [1] due to the transfer of two electrons from the four (BEDT-TTF) molecules per unit cell to the polymorphic Cu(SCN)₂ layer. At the small pressures used in our experiments, this overall carrier density per unit cell has to be considered independent of pressure. In the conducting organic layer in κ -(BEDT-TTF)₂Cu(SCN)₂, these holes are distributed between the Q2D Fermi surface pockets and the Q1D Fermi surface sheets:

$$\begin{aligned} n_{\text{total}}(\text{1/unit cell}) &\equiv 2 \text{ holes/unit cell} \\ &= n_{Q2D}(\text{1/unit cell}) + n_{Q1D}(\text{1/unit cell}). \end{aligned} \quad (2)$$

According to equation (2) the observed increase in n_{Q2D} is equivalent to an identical decrease in n_{Q1D} . Thus, the increase in n_{Q2D} can be understood as a simple, pressure-induced charge transfer of holes from the quasi-one-dimensional Fermi surface planes to the quasi-two-dimensional Fermi surface pockets in κ -(BEDT-TTF)₂Cu(SCN)₂. On the basis of our experiments, it is not possible to decide whether it is the increase in n_{Q2D} or the decrease in n_{Q1D} that is the relevant parameter for the suppression of T_c .

This mechanism bears a resemblance to the mechanism observed in cuprate superconductors where the effect of pressure is to transfer holes from the insulating charge reservoir layers into the Q2D, conducting CuO₂ layers [8]. Thus, in organic superconductors, similar to the case for cuprate superconductors, pressure has the effect of increasing the Q2D carrier density by transferring holes from other parts of the Fermi surface to those bands that support superconductivity [14].

Figure 3 demonstrates very clearly that there is a strong dependence between T_c and n_{Q2D} , which in turn is intimately related to n_{Q1D} . The sum of those two parameters is always equal to two holes per in-plane unit cell area. Thus, it is the in-plane pressure that seems to be relevant for the superconducting properties in κ -(BEDT-TTF)₂Cu(SCN)₂ and not the bulk pressure, as would be indicated by the pressure gauge. Such a dependence of the superconducting properties on the compression of the in-plane unit cell size has been suggested by recent ac-susceptibility measurements under pressure [25].

Louati *et al* [14] considered the ratio of the transfer integrals t_b/t_c to be the most critical parameter for κ -(BEDT-TTF)₂X salts. This ratio of transfer integrals has been calculated for the Fluorinert and petroleum spirit measurements [15], and is plotted against T_c in figure 4. The two measurements exhibit similar trends in the dependence of T_c on this ratio, but the exact dependence is seen to be affected by the pressure medium (see figure 4). Also the correlation between T_c and the effective mass is known to exhibit a dependence on the pressure medium [15]. In contrast, no dependence on the pressure medium could be seen in the correlation between T_c and n_{Q2D} (see figure 3), indicating that the superconducting carrier density could be a major determining factor for the superconducting properties of the organic superconductor κ -(BEDT-TTF)₂Cu(SCN)₂.

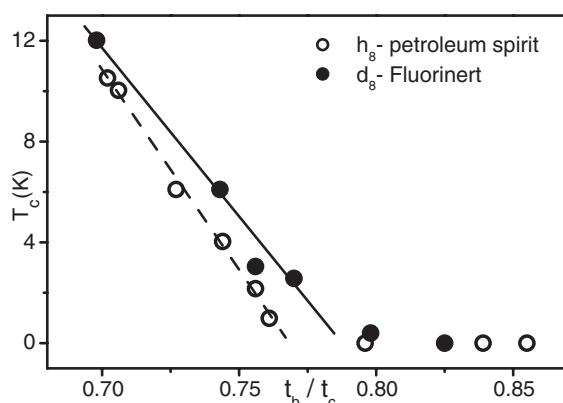


Figure 4. T_c as a function of the ratio of the in-plane transfer integrals, t_b/t_c [15]. The two measurements follow similar, but pressure medium dependent trends. This dependence is in contrast to the pressure medium independent correlation between T_c and n_{Q2D} (see figure 3).

5. Conclusion

We compared different magnetotransport measurements under pressure on the organic superconductor κ -(BEDT-TTF)₂Cu(SCN)₂ and found that pressure in general is not a reliable parameter for this system. This is thought to be due to the strong anisotropy of all physical properties in κ -(BEDT-TTF)₂Cu(SCN)₂ and possible departure from fully hydrostatic conditions when the sample is cooled in the frozen pressure medium. However, a strong correlation between the superconducting transition temperature, T_c , and the carrier density of the Q2D Fermi surface, n_{Q2D} , can be observed (see figure 3). This correlation is independent of the pressure medium used.

Pressure can be understood to increase n_{Q2D} by transferring carriers from the Q1D sections of the Fermi surface to the Q2D sections. Thus the effect of pressure on an organic superconductor resembles that observed in cuprate superconductors: in both materials pressure causes the transfer of holes from non-superconducting sections of the Fermi surface to the superconducting sections.

Acknowledgments

Research at the Clarendon Laboratory, Oxford University, is supported by the EPSRC, Grant No GR/R16075/01. Research at Argonne National Laboratory is sponsored by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences under contract W-31-109-ENG-38. CAK thanks the Deutsche Forschungsgemeinschaft for financial support. Special thanks are due to J Annett for stimulating discussions. The authors thank P A Goddard, S Blundell and W Hayes for their assistance in the preparation of this manuscript.

References

- [1] Ishiguro T, Yamaji K and Saito G 1998 *Organic Superconductors* (Berlin: Springer)
- [2] Singleton J 2000 *Rep. Prog. Phys.* **63** 1111
- [3] Singleton J and Mielke C 2002 *Contemp. Phys.* **43** 63
- [4] Lefebvre S, Wzietek P, Brown S, Bourbonnais C, Jérôme D, Mézière C, Fourmigué M and Batail P 2000 *Phys. Rev. Lett.* **85** 5420

- [5] Presland M R, Tallon J L, Buckley R G, Liu R S and Flower N D 1991 *Physica C* **176** 95
- [6] Schmalian J 1998 *Phys. Rev. Lett.* **81** 4232
- [7] McKenzie R 1997 *Science* **278** 820
- [8] Neumeier J J and Zimmermann H A 1993 *Phys. Rev. B* **47** 8385
- [9] Goldschmidt D, Klehe A-K, Schilling J S and Eckstein Y 1996 *Phys. Rev. B* **53** 14631
- [10] Rahal M, Chasseau D, Gaultier J, Ducasse L, Kurmoo M and Day P 1997 *Acta Crystallogr. B* **53** 159
- [11] Schultz A J, Beno M A, Geiser U, Wang H H, Kini A M, Williams J M and Whangbo M H 1991 *J. Solid State Chem.* **94** 352
- [12] Müller J, Lang M, Steglich F, Schlueter J A, Kini A M, Geiser U, Mohtasham J, Winter R W, Gard G L, Sasaki T and Toyota N 2000 *Phys. Rev. B* **61** 11739
- [13] Singleton J, Goddard P A, Ardavan A, Harrison N, Blundell S, Schlueter J A and Kini A M 2002 *Phys. Rev. Lett.* **88** 037001
- [14] Louati R, Charfi-Kaddour S, Ali A B, Bennaceur R and Heritier M 2000 *Phys. Rev. B* **62** 5957
- [15] Biggs T, Klehe A-K, Singleton J, Bakker D, Symington J, Goddard P A, Ardavan A, Hayes W, Schlueter J A, Sasaki T and Kurmoo M 2002 *J. Phys.: Condens. Matter* **14** L495
- [16] Sasaki T, Biberacher W, Neumaier K, Hehn W, Andres K and Fukase T 1998 *Phys. Rev. B* **57** 10889
- [17] Singleton J, Harrison N, Mielke C, Schlueter J A and Kini A M 2003 *Synth. Met.* **133/134** 241
- [18] Campos C E, Brooks J S, van Bentum P J M, Perenboom J A A J, Klepper S J, Sandhu P, Tokumoto M, Kinoshita T, Kinoshita N, Tanaka Y and Anzai H 1995 *Physica B* **211** 293
- [19] Campos C E, Sandhu P S, Brooks J S and Ziman T 1996 *Phys. Rev. B* **53** 12725
- [20] Caulfield J, Lubczynski W, Lee W, Singleton J, Pratt F L, Hayes W, Kurmoo M and Day P 1995 *Synth. Met.* **70** 815
- [21] Caulfield J, Lubczynski W, Pratt F L, Singleton J, Ko D Y K, Hayes W, Kurmoo M and Day P 1994 *J. Phys.: Condens. Matter* **6** 2911
- [22] Narduzzo A, Edwards R S, Ardavan A and Singleton J 2003 *Synth. Met.* **133** 129
- [23] Schlueter J A, Kini A M, Ward B H, Geiser U, Wang H H, Mohtasham J, Winter R W and Gard G L 2001 *Physica C* **351** 261
- [24] Kini A M, Carlson K D, Wang H H, Schlueter J A, Dudek J D, Sirchio S A, Geiser U, Lykke K R and Williams J M 1996 *Physica C* **264** 81
- [25] Klehe A-K, Tomita T, Schilling J S, Kini A M and Schlueter J A 2004 *Physica C* **402** 17
- [26] Spain I L and Segall S 1971 *Cryogenics* **11** 26
- [27] Schirber J E 1970 *Cryogenics* **10** 418
- [28] Steinle-Neumann G, Stixrude L and Cohen R E 2001 *Phys. Rev. B* **63** 054103
- [29] Takemura K 1999 *Phys. Rev. B* **60** 6171
- [30] Martin C, Agosta C C, Tozer S W, Radovan H A, Kinoshita T and Tokumoto M 2003 *Preprint cond-mat/0303315*
- [31] Schirber J E, Kwei G H, Jorgensen J D, Hitterman R L and Morosin B 1995 *Phys. Rev. B* **51** 12014
- [32] Singleton J 2002 private communication
- [33] Kartsovnik M V, Biberacher W, Andres K and Kunshch N D 1995 *JETP Lett.* **62** 905