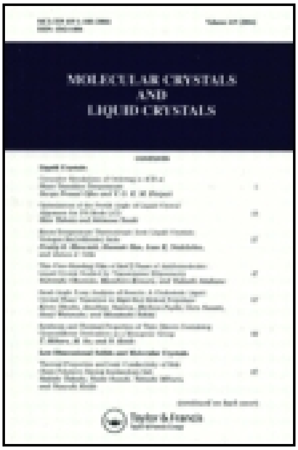

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Filamentary Superconducting-Normal-Metal Composites

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Version of record first published: 14 Oct 2011.

To cite this article: C. M. Bastuscheck, R. A. Buhrman, J. D. Kulick & J. C. Scott (1982): Filamentary Superconducting-Normal-Metal Composites, Molecular Crystals and Liquid Crystals, 81:1, 155-161

To link to this article: http://dx.doi.org/10.1080/00268948208072562

Mol. Cryst. Liq. Cryst., 1982, Vol. 81, pp. 155–161
0026-8941/82/8101-0155$06.50/0

(Proceedings of the International Conference on Low-Dimensional Conductors, Boulder, Colorado, August 1981)

FILAMENTARY SUPERCONDUCTING-NORMAL-METAL COMPOSITES

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Submitted for publication September 3, 1981

We describe the properties of a composite material consisting of superconducting filaments separated by normal metal barriers. The coupling between superconducting elements, and hence the diamagnetic response, is by proximity effect diffusion through the normal material and increases as the temperature is lowered. This behavior is contrasted with that of Josephson tunnelling in anisotropic structures which has been shown to lead to a decoupling transition. The diamagnetism and critical field behavior of $(SN)_x$, $TaSe_3$, and $NbSe_3$ are compared with the predictions of the model.

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I. INTRODUCTION

The superconducting properties of many highly anisotropic metals have been found to be anomalous[1-3] and have, up to now, defined fully quantitative explanation. In this paper, we describe a model which accounts in detail for the magnitude and temperature dependence of the diamagnetism of superconducting $(SN)_x$,[2] both pristine and brominated. The same model is applied to $NbSe_3$[1,3] and $TaSe_3$[1,3] giving semiquantitative agreement with the data.

II. THE MODEL

The essence of the description which we propose is that the materials are composed of superconducting fibers sheathed and separated by normal metal. Coupling between superconducting elements occurs via S-N-S tunnelling and therefore increases as the temperature is lowered, leading to progressively greater flux exclusion.

Specifically, consider a sample in the form of a cylinder of radius R, composed of many microscopic fibers of radius r. Let the typical distance between fibers, i.e., the thickness of normal metal, be L. The diamagnetic susceptibility, χ has two components: χ_i due to flux exclusion from individual fibers and χ_c due to shielding currents which flow from fiber to fiber. For fields applied parallel to the fibers $\chi_i=-(1/32\pi)(r/\lambda)^2$ where the penetration depth $\lambda >> r$. If even a small supercurrent can flow from one superconducting element to another, χ_c can easily dominate over χ_i.

The contribution χ_c is conveniently described in terms of the effective penetration depth λ_e, which was introduced by Deutscher and Entin-Wohlman:[4]

$$\lambda_e^{-2} = \frac{2\mu_0 eL}{\hbar} J_c \tag{1}$$

where J_c is the critical current density for supercurrent flow between adjacent elements. For T near T_c in a S-N-S junction, it is well established that J_c varies as $(1-T/T_c)^2$ and that when $T << T_c$ and $L/\xi_N >> 1$[6]

$$J_c \simeq \left(\frac{\pi\Delta^2}{2ekT}\right)\left(\frac{L}{\xi_N}\right) e^{-L/\xi_N}. \tag{2}$$

Here, ξ_N is the normal metal coherence length given in the dirty limit by $\xi_N=(\hbar v_F \Lambda/6\pi kT)^{1/2}$, Δ is the superconducting gap, v_F is the

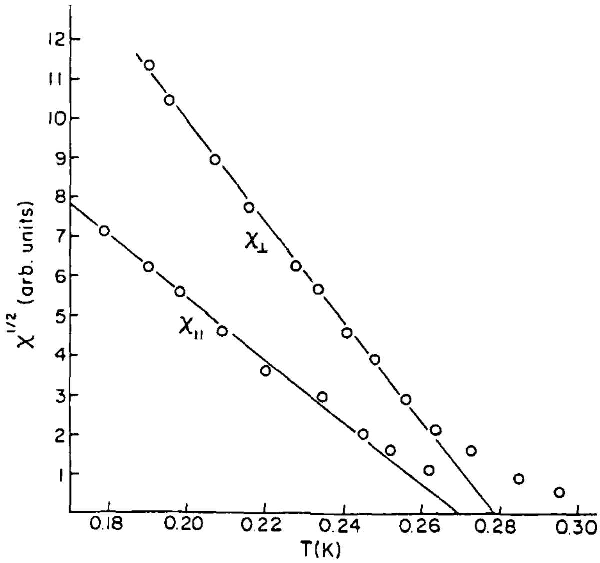

FIGURE 1 The susceptibility of $(SN)_x$.

Fermi velocity and Λ is the mean free path. Hence, we obtain for $T<<T_c$, $L/\xi_N>>1$ the result $J_c \sim \exp(-T^{1/2})$. The susceptibility χ_c is found in the usual way:

$$\chi_c = -\frac{1}{4\pi}\left[1-\frac{\lambda_e}{R}\,\frac{I_1(R/\lambda_e)}{I_0(R/\lambda_e)}\right] \tag{3a}$$

$$\simeq -\frac{1}{4\pi}\ ;\ R>>\lambda_e \tag{3b}$$

$$\simeq -\left(\frac{1}{32\pi}\right)(R/\lambda_e)^2\ ;\ R<<\lambda_e \tag{3c}$$

where I_n is the nth order Bessel function of imaginary argument. The important result is that for weak flux exclusion, Eqs. (1) and (3c) imply that $\chi_c \sim J_c$. Therefore, near T_c we expect

$$\chi_c^{1/2} \sim (1 - T/T_c)\ . \tag{4a}$$

and at lower temperature (provided $R<<\lambda_e$ still holds)

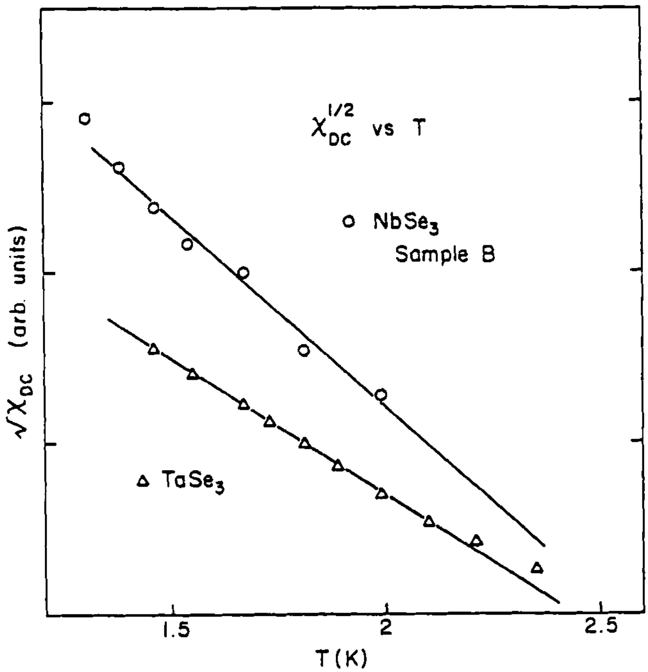

FIGURE 2 The susceptibility of $NbSe_3$ and $TaSe_3$ near T_c. Vertical scales are not the same for the two materials.

$$\chi_c \sim \exp(-T^{1/2}) . \qquad (4b)$$

Results of a complete analysis are given elsewhere.[7]

III. $(SN)_x$ AND BROMINATED $(SN)_x$

Susceptibility data[8] for $(SN)_x$ are shown in Fig. 1, plotted according to Eq. (4a). Apart from deviations very close to T_c which are attributed to variations in the transition temperature of the fibers, χ is seen to follow accurately a $(1-T/T_c)^2$ behavior near T_c. The extrapolation of the linear portion of $\chi^{1/2}$ versus T indicates a

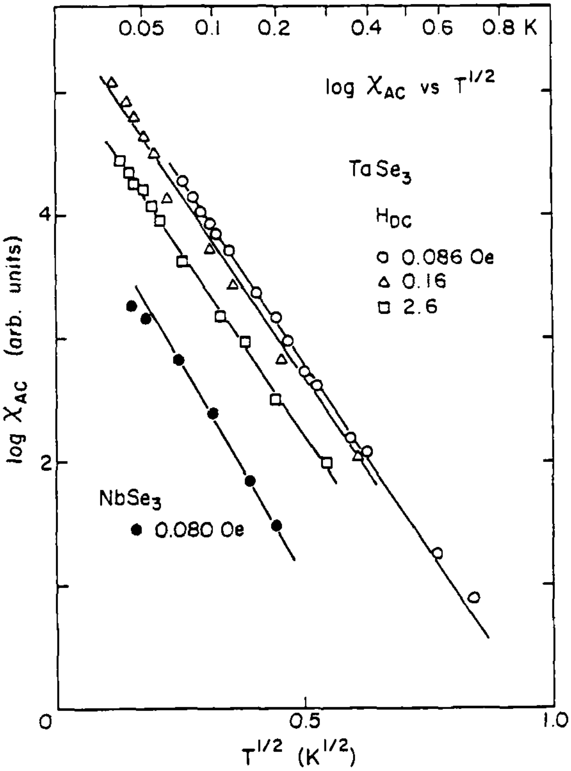

FIGURE 3 The susceptibility of $NbSe_3$ and $TaSe_3$ for $T<<T_c$.

transition temperature $T_c \simeq 0.27K$, close to the value obtained from the resistive transition.[2] The slight difference between the values of T_c obtained from the $\chi_{\parallel}$ and $\chi_{\perp}$ data is explained by a variation in T_c of the fibers and by the different paths taken by shielding currents in the two orientations. Thus, $\chi_{\parallel}$ represents current paths with lower average T_c than $\chi_{\perp}$. A fuller analysis, using Eq. (3a) yields a satisfactory fit over the entire temperature range of measurement, for $L/\xi_N \sim 1$.

Electron microscopy reveals fibers of diameter $r \lesssim 100$Å surrounded by relatively disordered material of approximately 10Å thickness. Then $L = \xi_N$ corresponds to a mean free path $\Lambda \approx 1$Å.

A similar description can be applied to brominated $(SN)_x$. The data can be fit[7] using larger values of R/λ_e, implying a stronger coupling between fibers, consistent with the interpretation of H_{c2} measurements.

IV. $NbSe_3$ AND $TaSe_3$

There has been some controversy concerning the existence of superconductivity in $NbSe_3$. In a previous publication,[9] we reported weak flux exclusion and flux trapping below 2K. We and other workers[3] have reported resistive anomalies at different temperatures, (but never zero-resistance) or in some cases[10] none at all.

The superconductivity of $TaSe_3$ is better established[11] with resistivity measurements showing a transition temperature T_c=2.5K. However, several samples are reported to give nonzero resistance.[12]

Here, we give a reevaluation of previous data, in light of the model described above. In Fig. 2, we show data taken near T_c on both materials, plotted as $\chi^{1/2}$ versus T. The agreement with Eq. (4a) is moderately good. At lower temperature, a plot of $\log\chi$ versus $T^{1/2}$ (Fig. 3) confirms the behavior predicted in Eq. (4b).

A problem exists with the application of the filamentary-composite model: namely, the identification of the superconducting fibers, especially for $NbSe_3$. The above measurements were made on polycrystalline samples consisting of needle-like crystallites of order $2\times10\times100$ μm and it is tempting to assume that the crystallites themselves are the superconducting elements. However, single crystal resistivity measurements[9] show that $NbSe_3$ is not a bulk superconductor. The filamentary must be attributed to microscopic inhomogeneities within the crystals themselves, beyond the resolution of electron microscopy. Possible origins include local strain fields around dislocations and impurities, inclusion of additional selenium as an intercalant or in the form of other Nb-Se phases,[13] and the domain structure[14] asssociated with CDWS.

V. CONCLUSIONS

We have presented a model which gives a good account of the superconducting diamagnetism of $(SN)_x$, and which is consistent with the data on $NbSe_3$ and $TaSe_3$. Superconducting filaments are separated by normal material through which shielding current must pass by S-N-S tunnelling. The nature of the proximity effect supercurrent leads naturally to increasing flux exclusion at lower temperatures. This is in contrast to the decoupling phenomenon described by Turkevich and Klemm[15] for Josephson tunnelling between filaments.

The same model easily accounts for the upward curvature observed in $H_{c2}^{\parallel}(\tau)$,[9,16] since thinner fibers, in contact with normal metal, have lower transition temperatures and since thinner fibers also have larger parallel critical fields.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation through Grants No. DMR77-09879 and DMR79-09177, and by the NSF-MRL program, Grant No. DMR76-81083.

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