Classification
Physics Abstracts
74.50 — 75.10N

Superconducting glass-phase diagram for ceramic $YBa_2Cu_3O_{7-\delta}$

Z. Kozioł, J. Piechota and H. Szymczak

Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warszawa, Poland

(*Reçu le 30 janvier 1989, révisé le 19 mai 1989, accepté le 19 juillet 1989*)

Résumé. — Nous avons mesuré la susceptibilité complexe de céramiques $YBa_2Cu_3O_{7-\delta}$ près de la transition supraconductrice en fonction des amplitudes du champ alternatif et d'un champ statique superposé. Du comportement du maximum de χ'', nous avons déduit une loi d'échelle universelle pour la dépendance en fonction du champ statique ainsi que la dépendance en température du courant Josephson intergrains et les champs critiques H_0 (où le champ est piégé par les jonctions) et H_J (où apparaît la supraconductivité en volume). Ces matériaux ont des propriétés qui rappellent celles des verres de spins et sont en accord qualitatif avec les prédictions de modèles pour les supraconducteurs désordonnés.

Abstract. — AC complex magnetic susceptibility of ceramic $YBa_2Cu_3O_{7-\delta}$ was measured close to the superconducting transition temperature, as a function of the amplitude of the alternating magnetic field and of a superimposed dc field. From the maxima of the imaginary part of susceptibility, the universal scaling of the dc field and the temperature dependence of the intergrain Josephson current were obtained, together with the critical fields : H_0, for trapping the flux by the junctions, and H_J for the occurrence of bulk superconductivity. The results suggest that these materials have spin-glass-like properties, and are qualitatively consistent with the predictions of models of disordered superconductors.

1. Introduction.

There is a large amount of work devoted to applying the ideas of spin-glass theory to granular classical (low T_C) superconductors [1, 2] and to high T_C superconductors [3]. We have investigated the properties of ceramic materials. The important feature of these ceramics is disorder. Superconducting grains of dimensions of the order of microns are separated by nonsuperconducting barriers. In the grains, the critical supercurrent can be as high as 10^8 A/cm^2, and the slope of the critical field H_{C2} is of the order of a Tesla per Kelvin near the transition temperature T_C. There is, however, another transition temperature T_J connected with the occurrence of bulk superconductivity in the sample, due to the interactions between the grains through Josephson links. At low temperatures, the interaction Hamiltonian is written in the form [2] :

$$H = \sum J_{ij} \cdot \cos \left(\Phi_i - \Phi_j - A_{ij} \right) \tag{1}$$

where Φ_i is the phase of the order parameter of the i-th grain with

$$A_{ij} = \frac{2\pi}{\Phi_0} \cdot \int \mathbb{A}_{ij} \, d\mathbb{L} \, , \quad J_{ij} = \frac{\hbar}{2e} I_{ij}$$

and

$$I_{ij} = \frac{\pi \cdot \Delta(T)}{2 \cdot e \cdot \rho \cdot \alpha} \tanh \left(\frac{\Delta(T)}{2kT} \right) \tag{2}$$

where I_{ij} is the maximum Josephson supercurrent between the i-th and j-th grains, $\Delta(T)$ is the temperature dependent energy gap, $\rho \cdot \alpha$ is the normal state resistance of the junction and α is the distance between the grains [4].

The Ambegaokar-Baratoff formula (2) can be applied to the case of superconductor-insulator-superconductor (S-I-S) structures [5]. Then, close to T_C, one obtains a linear temperature dependence of the critical supercurrent, I_C: $I_C \propto (T_C - T)$.

In the case of granular superconductivity, a crossover to the Ginzburg-Landau dependence, $I_C \propto (T_C - T)^{3/2}$, should be observed in the vicinity of T_C, due to the suppression of the order parameter through the interaction between the grains [4]. The Ginzburg-Landau theory should, however, apply away from the fluctuation regime, which is believed to be wide in the high T_C oxide superconductors [6]. The goal of our work is to investigate the critical region properties measuring the field and temperature dependence of the critical Josephson supercurrent. Below T_J, the process of freezing of the quantum phases of the order parameter of the grains begins. The Hamiltonian (1) is then not adequate to describe the critical region properties, but some features described by (1) are still valid. A magnetic field introduces frustration into the system. This means that below the phase-locking temperature T_J the magnetic flux can be frozen in the sample. Frustration is the mechanism leading to the occurrence of pinning energies. Experimentally, a remanence of the magnetisation, time- and history-dependent effects, are observed in these superconductors for fields well below the first critical field for the grains [7]. Due to this, we can speak about the existence of a critical state in low fields, and at temperatures below T_J. Further, we apply Bean's critical state theory, in the version developed by Goldfarb [8], to obtain the critical intergrain supercurrent from results on ac susceptibility. This method was used by other authors [9], and in our earlier work [10]. In the case of a regular network of exactly identical grains, the Hamiltonian of the system is periodic in the field, and all measurable properties, such as T_J, should be also periodic in the field [2]. Because of the frustration caused by disorder and the magnetic field, this periodicity is destroyed, and T_J becomes smoothly decreasing with the applied field [2, 11]. As large fluctuations of the order parameter exist above T_J, critical supercurrents cannot be measured at these temperatures. $T_J(H)$ is the border line between the normal state and bulk superconductivity, and can be determined from the occurrence of a small supercurrent in the sample. Results on $I_C(T, H)$ from the ac method are compared with a Fraunhofer diffraction-like dependence, giving values of the characteristic field for flux trapping by the intergrain junctions, $H_0(T)$, and the Peterson and Ekin [12] scaling dependence of $I_C(H/H_0)$.

2. Experimental details.

Ceramic samples 3 mm in diameter and 5 mm long were obtained by the standard solid-state reaction method [13]. An ac bridge with two identical coils and AT resistors (with a very small temperature coefficient of their resistances) was immersed in a glass cryostat, with the sample placed in the middle of one of the coils. We were able to cause slow temperature runs (about 1 K per min) of the sample. The temperature was monitored with a relative accuracy of about 0.1 K, using a calibrated carbon resistor. The complex magnetic susceptibility was measured at 7 kHz using two lock-ins. The experimental data were collected by a microcomputer connected through an interface, working with voltmeters in IEC-625 standard. It would not be possible in practice to obtain the results presented here without such automation because of the large number of $\chi(T)$ traces needed for the analysis. The background signal from the bridge was compensated a few degrees above the transition temperature, and the phase angle of the susceptibility was adjusted at a low temperature in the case of a zero dc field and at small amplitude of the ac field. The measurements were done with the ac and dc fields parallel to the axis of the sample.

3. Results.

3.1 CHARACTERISTIC FEATURES OF THE AC SUSCEPTIBILITY. — The temperature dependences of susceptibility $\chi'(T)$ (real part) and $\chi''(T)$ (imaginary part) for different amplitudes and different dc fields have already been published many times, and we shall only summarise the main typical features of $\chi(T; H_{ac}; H_{dc})$ [7] :

i) In good samples at low T, $\chi'(T) \simeq -1/4\pi$ and does not depend on the temperature, while $\chi''(T) \simeq 0$.

ii) With increasing T, $\chi''(T)$ goes through a maximum at $T = T^*$, and, under certain conditions, a second maximum at a higher temperature, connected with the onset of intragrain superconductivity.

For $T = T^*(H_{ac}, H_{dc})$, the ac field penetrates exactly to the middle of the sample and, for cylindrical infinitely long samples, $\chi''(T^*) \simeq 0.212/4\pi$, and for T^*, the critical supercurrent density can be calculated from the relation [8] :

$$I_C = 10\, H_{ac}/4\,\pi r \tag{3}$$

(H_{ac} in Oe, I_C in A/cm^2, r in cm). The validity of this formula is based on the assumption of the existence of the critical state, and on I_C being independent of the field.

iii) Below a certain value of H_{ac}, of the order of 0.05 Oe in our experiments, independent of the dc field, one is not able to see any difference between the traces of $\chi(T)$ for different H_{ac}. We will call T_J the temperature of the $\chi''(T)$ maximum for such low H_{ac}. Below this temperature, a small macroscopic supercurrent begins to flow in the bulk sample.

iv) One does not measure the derivative of dc magnetisation in ac susceptibility measurements. It is easy to obtain ac susceptibility close to $-1/4\pi$ in superimposed dc fields, while the slope dM/dH in the same fields would give much lower values of χ. This means that the response of the sample to a small disturbance (field) variable in time is the nonequilibrium one that has been discussed in models of superconducting glasses [2].

Since we supposed that the dc field changes the critical supercurrent in the sample, and because the measured response to the ac field is connected with the value of this supercurrent, we should calculate the field dependence of critical currents from the χ_{ac} measurements in dc

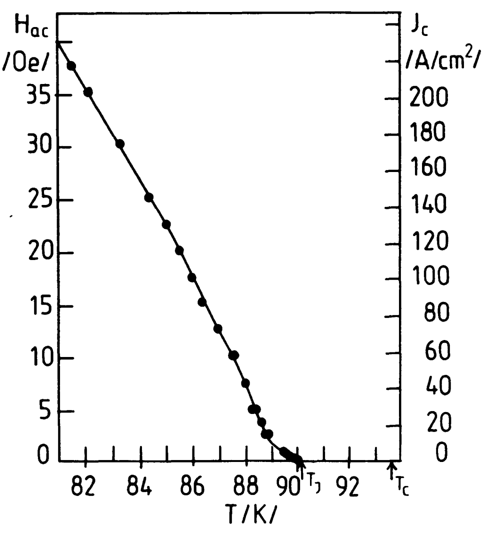


Fig. 1. — Temperatures of the maxima in the imaginary part of the ac susceptibility for fields H_{ac} (left vertical scale) measured in zero external dc field. According to equation (3) this curve corresponds to the temperature dependence of the critical supercurrent of the sample (right vertical scale). The critical temperatures T_C and T_J are marked by arrows.

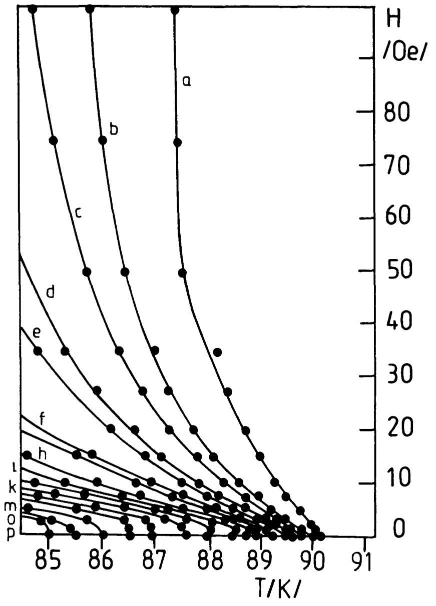


Fig. 2. — Temperatures of the maxima in the ac susceptibility as a function of the dc applied field (vertical scale), obtained for different amplitudes of the ac field. In the figure, a) to p) correspond to : 0.025, 0.25, 0.5, 0.75, 1.0, 1.75, 2.5, 5, 7.5, 10, 12.5, 15, 17.5, 20, and 22.5, respectively, in Oe. The curve for $H_{ac} = 0.025$ Oe is the $H_J(T)$ dependence.

fields. On the basis of this assumption, we will perform measurements of $T^*(H_{dc})$ for different H_{ac} and will apply the analysis used in the case of a zero external dc field to obtain the field dependence of the critical Josephson supercurrent. Since $\chi(T)$ depends on the history of the sample [7] all the data presented were obtained under the same field-cooling conditions. The reason for this is also the fact that, during cooling in the field, the magnetisation of the sample is much lower than in the case of zero-field cooling, which allows us to assume with an accuracy sufficient for our later discussion that the internal dc field is equal to the applied dc field.

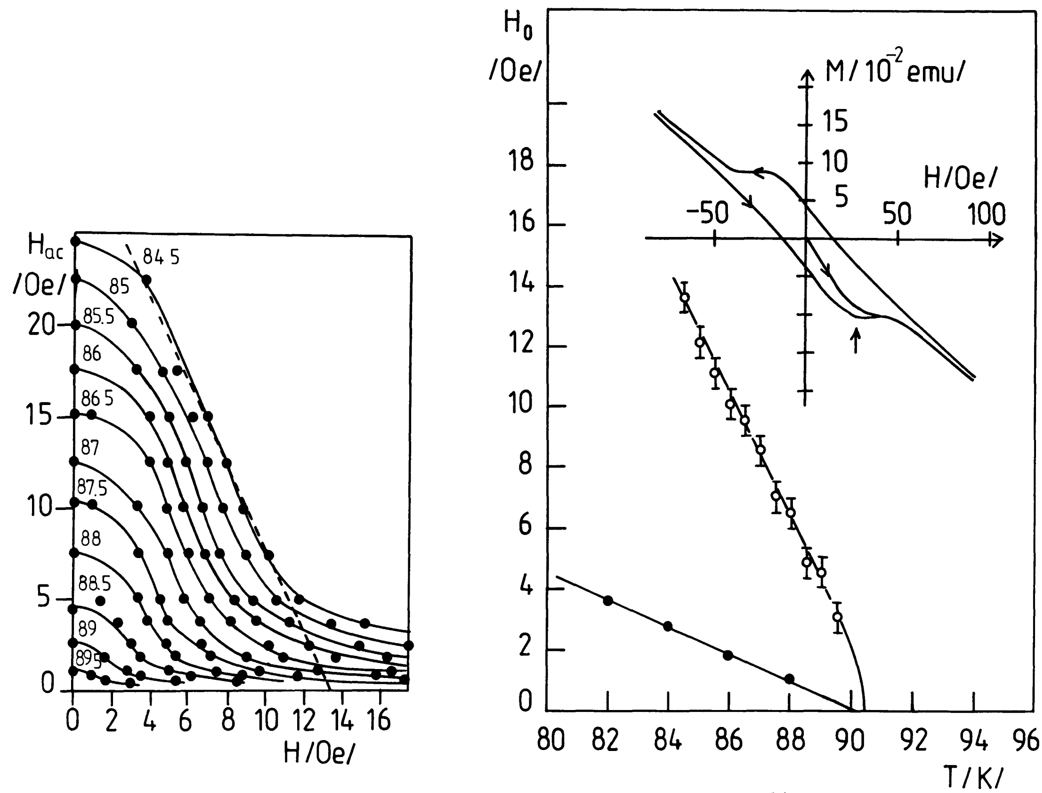
3.2 BULK CRITICAL JOSEPHSON SUPERCURRENTS. — Figure 1 presents the amplitude of the ac field ($H_{dc} = 0$) as a function of the temperatures T^* of the $\chi''(T)$ maxima, i.e., it is $I_C(T)$ according to (3). The characteristic temperatures of the sample, T_C and T_J, are shown by arrows. These results were discussed [10], together with the results for a series of $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ samples with different iron contents. Measurements of the temperatures of the $\chi''(T)$ maxima were performed for dc fields of up to 100 Oe, and the results for temperatures about 84 K are presented in figure 2 on the H-T plane. The lines connect the points obtained for fixed amplitudes of the ac fields, for fields from 0.025 Oe to 22.5 Oe, and are drawn only to guide the eye. The curve obtained for 0.025 Oe can be assumed to be the border curve on the H-T plane for the occurrence of bulk superconductivity, and we will call it the curve of critical field for the Josephson supercurrent, $H_J(T)$. As was already said, one would obtain the same curve for H_{ac} lower than about 0.05 Oe.

4. Discussion.

4.1 CRITICAL FIELDS. — Measurements of transport critical currents as a function of the dc field [14, 15] have been discussed on the basis of a Fraunhofer diffraction-like dependence :

$$I_C(T; H) = I_C(T; 0) \left| (\sin(\pi \,.\, H/H_0))/(\pi \,.\, H/H_0) \right| . \tag{4}$$

Because of large disorder, this equation should be averaged over the junction lengths and orientations, and such averaging will smear out the structure in $I_C(T; H)$ [12]. In (4), H_0 plays the role of the critical field of the Josephson junction for trapping the flux by the junction itself. For some fixed temperatures, we plotted straight vertical lines in figure 2, and, in this way, obtained a set of field dependences of I_C for these temperatures (Fig. 3a). We see a strong suppression of the current in fields of the order of a few Oe. Now, for each temperature in figure 3a, we draw a straight line with a slope corresponding to the maximum slope of the $I_C(H)$ dependence, as illustrated by the broken curve for $T = 84.5$ K. The dependence of H_0 on temperature is surprisingly close to linear (Fig. 3b), although the zero value of H_0 is obtained by extrapolation at a temperature a little higher than $T_J(0)$. Kwak *et al.* [14] report, from transport measurements, a small negative curvature of H_0 with decreasing temperature (measurements of Ref. [14] were done in a wider temperature range). Our values of H_0 are consistent with these reports, but are in disagreement with our dc measurements on a Foner magnetometer (the full circles in Fig. 3b). For different temperatures, we have plotted small hysteresis loops of magnetisation (inset of Fig. 3b) and, for all these loops, we applied the same procedure to obtain the lower characteristic dc field (it is not obvious where is the critical field H_0 in such a loop ; in the inset in Fig. 3b, this field is shown by the arrow). The fields obtained are very close to being linear with T in the vicinity of T_C, although they are about five times smaller than those obtained from the ac susceptibility procedure — we believe this to be connected mainly with the time dependence of the properties measured.



a) b)

Fig. 3. — a) dc field dependences of the critical supercurrents (in units of H_{ac} amplitude, Eq. (3)) for temperatures from 84.5 to 89.5 K (matched in the Fig.), obtained by the method described in the text. From extrapolations of the steepest portions of the data to zero value of H_{ac}, as illustrated by the broken line for $T = 84.5$ K, values of the junction critical field $H_0(T)$ were obtained. These fields are represented by open circles in figure b, and the fit to them, represented by the solid line, is discussed in the text. The inset in b shows a small hysteresis loop of the dc magnetisation at $T = 4.2$ K, and the method of obtaining the characteristic dc field for the loop marked by the arrow. These fields (the full circles in Fig. b) are compared with the H_0 measured by the ac method.

The field H_0 depends on the geometrical dimensions of the junctions and, for large junctions (width $L > \lambda_J$), H_0 is given by :

$$H_0 = \Phi_0 / \pi s \lambda_J \tag{5}$$

where Φ_0 is the flux quantum, $s = \alpha + 2\lambda_g$, with λ_g being the field penetration depth into the grains and λ_J the penetration depth into the junction. λ_J depends on the intergrain critical current :

$$\lambda_J = [\Phi_0 c / 8 \pi^2 s I_C(T)]^{1/2} . \tag{6}$$

Since λ_g is high in these materials and the separation between the grains is small, we can assume that $s = 2\lambda_g$. Now, we have :

$$H_0 = 2(\Phi_0 / c\lambda_g)^{1/2} (I_C)^{1/2} ,$$

where we have used (6).

The key to understanding the temperature dependence of I_C and H_0 is the observation that the ac method measures the nonequilibrium properties of the system. In each experiment, the measurements give an « average over the time » of some quantum-mechanical « observables ». The classical equation for the tunnelling current density in a single junction is : $I = I_C \sin \delta$, where δ is the quantum phase difference across the junction. In our case, above T_J, fluctuations lead to $\delta = 0$ and, consequently, to $I = 0$. Below, T_J, some average over time $\delta \neq 0$ is measured by determination of a finite supercurrent. It is easy to see that T_J should be frequency dependent, as has been obtained in [16]. This leads us to the conclusion that H_0 should be dependent in a non-standard way on the temperature through the divergence of λ_J at T_J rather than at T_C :

$$H_0 \propto [(T_J - T)^{\alpha} / (T_C - T)^{-1/2}]^{1/2}, \tag{7}$$

where we have assumed, quite arbitrarily, that $I_C \propto (T_J - T)^{\alpha}$.

In figure 3a, the solid line was drawn using equation (7) with the following parameter values : $T_C = 93.7$ K, $T_J = 90.2$ K and $\alpha = 1.25$. If the BCS dependence of $\lambda_g(T)$ is taken, then a good agreement with the measured values of I_C is obtained with $\lambda_g(0)$ of about 0.15 μm, a value close to those obtained in other ways [17]. λ_J is then about 5 μm at $T = 84.5$ K.

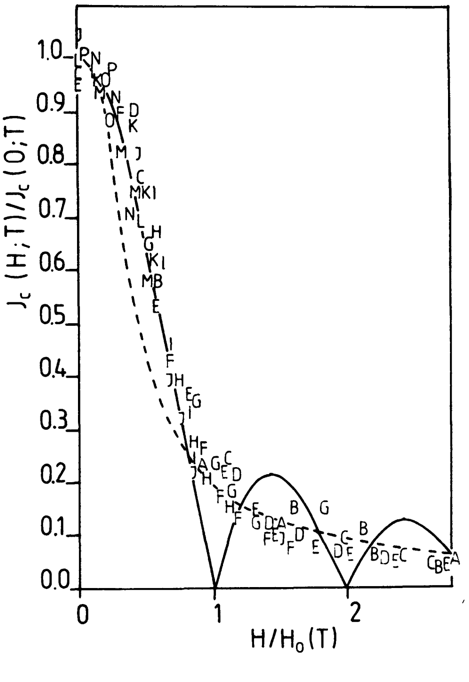


Fig. 4. — Universal scaling of the field dependence of the Josephson supercurrent obtained from the data of figure 2 (see the text). The symbols from A to P refer to the values of the H_{ac} amplitudes in the order of figure 2. For comparison, a Fraunhofer diffraction-like dependence is drawn by the solid line, and the dependence of Peterson and Ekin [12], calculated after applying the specific averaging procedure on the junction weak-link properties (see Ref. [12] for the details), is shown by the broken line.

4.2 UNIVERSAL SCALING OF $I_C(T, H)$. — As a result of our determination of $I_C(T, 0)$ and $H_0(T)$, we can try to plot a universal temperature independent scaling of $I_C(H)$ (Fig. 4) :

$$I_C(T, H)/I_C(T, 0) = f(H/H_0(T)),$$

where we are using only the experimental points of figure 2. For comparison, the solid line shows the Fraunhofer diffraction-like dependence of the Josephson supercurrent (Eq. (4)). Quantitative agreement between our field dependence of I_C and the measurements and calculations of Petersen and Ekin [12] (represented by the broken curve in Fig. 4) is obtained. It was one of our aims to show this good accordance between transport critical current measurements and the data derived from susceptibility measurements, bearing in mind all the approximations involved by using the latter method.

4.3 CRITICAL STATE OF THE JOSEPHSON SUPERCONDUCTIVITY AND THE GLASS TRANSITION. — The critical lines $T_J(H)$ for the occurrence of bulk superconductivity was shown in the paper of Barbara *et al.* [18], where susceptibility measurements were reported for ceramic and single-crystal samples. These authors suggested a spin-glass-like origin of this critical line. There also exists a marked similarity of transport results [19] for the field dependence of the critical temperature for appearance of a very small critical current. The frequency dependence of $T_J(H)$ has been shown to scale in the same way as in the spin-glass transition [16]. The border line between localised and delocalised superconductivity obtained by us is qualitatively similar to that calculated by Aksenov and Sergeenkov [11] for a spin glass transition in disordered superconductors.

Lastly, Muller *et al.* [20] reported measurements of flux penetration into ceramic samples in low fields, and successfully applied the critical state model of Josephson superconductivity. Their work justifies our findings of I_C from the measurements of losses. We are thus proposing an alternative view of the origin of the pinning forces in these materials. We suggest that spin-glass like mechanism, caused by the frustration induced by disorder and the magnetic field, is leading to the occurrence of frozen flux of the magnetic field. Then there exist metastable states and energy barriers to overcome for flux motion. These barriers can be higher than those for the mixed state of Josephson superconductivity without frustration. The interplay between these two mechanisms of the bounding in critical current measurements is not clear to us.

5. Conclusions.

We have used ac complex magnetic susceptibility measurements to determine the dependence of the critical Josephson intergrain supercurrent on the temperature, close to the superconducting transition temperature, from the losses $\chi''(T)$. We made measurements of critical currents in different dc fields. The results are consistent with predictions of $I_C(H)$ for Josephson junctions, smeared out by large disorder in the sample. We obtained the temperature dependence of the critical field for trapping the flux by the junctions, and the temperature dependence of the critical field for occurrence of bulk Josephson superconductivity. All the results presented here strongly support the existence of a field-dependent superconducting glass state in the HTCS, connected with intergrain weak links. It was suggested that this spin-glass-like mechanism is responsible for flux pinning and the similarities to classical critical state behavior. The properties measured are frequency dependent, and more systematic studies of this effect will be published [16].

We obtained similar results for a $(Bi_{0.7}Pb_{0.3})_2Sr_2Ca_2Cu_3O_{10}$ sample [21].

This work was supported by IF PAN under contract RPBP 01.9.

References

[1] JOHN S., LUBENSKY T. C., *Phys. Rev.* **B 34** (1986) 4815.
[2] SHIH W. Y., EBNER C., STROUD D., *Phys. Rev.* **B 30** (1984) 134.
[3] FISHER M. P. A., *Phys. Rev. Lett.* **62** (1989) 1415 ;
GREGORY S., ROGERS C. T., VENKATESAN T., WU X. D., INAM A., DUTTA B., *Phys. Rev. Lett.* **62** (1989) 1548.
[4] CLEM J. R., BUMBLE B., RAIDER S. I., GALLAGHER W. J., SHIH Y. C., *Phys. Rev.* **B 35** (1987) 6637 ;
CLEM J. R., *Physica* **C 153-155** (1988) 50.
[5] AMBEGAOKAR V., BARATOFF A., *Phys. Rev. Lett.* **10** (1963) 486 ; and **11** (1963) 104.
[6] KAPITULNIK A., BEASLEY M. R., CASTELLANI C., SI CASTRO C., *Phys. Rev.* **B 37** (1988) 537.
[7] PIECHOTA J., KOZIOL Z., PAJACZKOWSKA A., SZYMCZAK H., *Phys. Status Solidi (a)* **113** (1989) 151.
[8] BEAN C. P., *Rev. Mod. Phys.* **36** (1964) 31 ;
GOLDFARB R. B., CLARK A. F., *IEEE Trans. Magn.* **MAG-21** (1985) 332.
[9] POLAK M., HANIC F., HLASNIK I., MAJOROS M., CHOVANEC F., HORVATH I., KREMPSKY L., KOTTMAN P., KEDROVA M., GALIKOVA L., *Physica* **C 156** (1988) 79 ;
GOMORY F., LOBOTKA P., *Sol. State Commun.* **66** (1988) 645.
[10] PIECHOTA J., KOZIOL Z., to be published.
[11] AKSENOV V. L., SERGEENKOV S. A., *Physica* **C 156** (1988) 18.
[12] PETERSON R. L., EKIN J. W., *Phys. Rev. B* **37** (1988) 9848.
[13] WIŚNIEWSKI A., BARAN M., PRZYSŁUPSKI P., SZYMCZAK H., PAJACZKOWSKA A., PYTEL B., PYTEL K., *Sol. State Commun.* **65** (1988) 577.
[14] KWAK J. F., VENTURINI E. L., NIGREY P. J., GINLEY D. S., *Phys. rev.* **B 37** (1988) 9749.
[15] ZHAO Y., SUN S., ZHANG H., SU Z., CHEN Z., ZHANG Q., *Sol. State Commun.* **66** (1988) 35.
[16] KOZIOŁ Z., *Physica C* **159** (1989) 281.
[17] COOPER J. R., CHU C. T., ZHOU L. W., DUNN B., GRUNER G., *Phys. Rev.* **B 37** (1988) 638.
[18] BARBARA B., KHODER A. F., COUACH M., HENRY J. Y., *Europhys. Lett.* **6** (1988) 621.
[19] DUBSON M. A., HERBERT S. T., CALABRESE J. J., HARRIS D. C., PATTON B. P., GARLAND J. C., *Phys. Rev. Lett.* **60** (1988) 1061.
[20] MULLER K. H., MACFARLANE J. C., DRIVER R., *Physica* **C 158** (1989) 69.
[21] KOZIOŁ Z., unpublished.