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## The « irreversibility line » of $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ : a possible breakdown of an intrinsic proximity effect

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**Résumé.** — La ligne d'« irréversibilité » de l'aimantation  $H^*(T)$  d'un composé granulaire supraconducteur  $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  ayant une température critique  $T_c$  de 110 K a été étudiée. A basses températures, on observe  $H^* \sim e^{-T/T_0}$  et près de  $T_c$  le comportement universel  $H^* \sim \left(1 - \frac{T}{T_c}\right)^{3/2}$ . L'aimantation réversible obéit, en fonction du champ magnétique  $H$ , aux lois

calculées pour le réseau de vortex d'un supraconducteur idéal quand  $H_{c1} \ll H \ll H_{c2}$ . L'aimantation décroît linéairement avec  $T$ , jusqu'à des températures très inférieures à  $T_c$ . On interprète la loi exponentielle de basses températures par l'existence d'un champ critique  $H^*$  détruisant la supraconductivité induite par effet de proximité. On se représente la structure comme un super-réseau fait de feuillets alternativement normaux et supraconducteurs. Les feuillets normaux pourraient être constitués des plans SrO-BiO-BiO-SrO, les feuillets supraconducteurs étant associés aux 3 plans CuO. La forte dépendance du courant critique avec le champ magnétique s'expliquerait donc par l'affaiblissement de l'énergie de corrélation des vortex à la transition de phase quand la supraconductivité induite dans les feuillets normaux est détruite par le champ magnétique.

**Abstract.** — The study of the magnetization « irreversibility line » of a  $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  compound with a critical temperature  $T_c$  of 110 K is presented. At low temperatures, we observe  $H^* \sim e^{-T/T_0}$  and close to  $T_c$ ,  $H^* \sim \left(1 - \frac{T}{T_c}\right)^{3/2}$ . The reversible magnetization as a function of the magnetic field  $H$  obeys the laws calculated for the Abrikosov flux lattice of an ideal

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superconductor when  $H_{c1} \ll H \ll H_{c2}$ . Even far from  $T_c$ , magnetization decreases linearly with  $T$ . The exponential behavior of  $H^*(T)$  can be interpreted by the existence of a breakdown field in which a proximity effect induced superconductivity is destroyed. The structure may be seen as a superlattice composed of laminae alternatively normal and superconducting. The normal laminae could be the SrO-BiO-BiO-SrO layers, the superconducting ones being the 3 CuO layers. The strong dependence of the critical current with magnetic field is explained by the weakening of the correlation energy of vortices at the phase transition corresponding to the breakdown by the magnetic field of an induced superconductivity in the normal laminae.

### Introduction.

Müller, Takashige and Bednorz [10] have first reported that the « irreversibility line » of  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_{4-y}$  follows a simple law :

$$H^* = H_0 \left( 1 - \frac{T}{T_c} \right)^{3/2} \quad (1)$$

analogous to the Almeida-Thouless line of spin glasses. The line separates the region in the  $(H, T)$  plane in which the magnetization  $M(H, T)$  is reversible from the region in which  $M$  depends on time and on the previous path in the  $(H, T)$  plane. Préjean and Souletie [2] have argued, by comparing these results to spin glass properties, that the irreversibility line cannot be a phase transition line and that the hysteresis of these systems may not be fundamentally different from that of type II superconductors with pinning. Yeshurun and Malozemoff [3] have shown that a similar line exists in the YBaCuO system. They have tentatively explained this behaviour in terms of flux creep and predicted a similar thermal variation (see Eq. (1)). The activation energy should have the following form :

$$U_0 = \beta H_c^2 \xi \frac{\phi_0}{B} \quad (2)$$

which leads to equation (1) for the irreversibility line.  $\xi$  is the coherence length,  $B$  is the flux density,  $\frac{\phi_0}{B}$  is approximately the square of the distance between vortices,  $\phi_0$  is the flux quantum and the parameter  $\beta$  is a numerical factor of the order of 1 when the fluxons are strongly correlated because their separation distance is much smaller than the penetration depth  $\lambda$ . Tinkham [4] has extended this explanation to account for the broadening of the resistive transition under magnetic field without invoking compound inhomogeneity. Deutscher and Müller [5] have argued that the activation energy  $U_0$  which must be overcome to allow vortex motion, is strongly weakened by the small coherence length of these high  $T_c$  materials. Then, the relaxation of the magnetization has to be fast at high temperatures. Gammel *et al.* [6] have claimed that mechanical measurements give evidence for flux-line lattice melting in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_8$  single-crystals. A transition line  $H^*(T)$  seems to exist which is about the same for fields along the  $c$ -axis and in the  $(ab)$  plane.  $H^*$  decreases linearly with temperature. The pinning effect only exists at low temperatures. Civale *et al.* [7] have also shown that the reversible state is strongly correlated to superconductivity percolation in a sintered material.  $\text{La}_{1.8}\text{Sr}_{0.2}\text{CuO}_{4-y}$  contains regions of strongly reduced superconductivity surrounding superconducting islands. The reversible flux magnetization near  $T_c$  is a consequence of flux expulsion from the non-interacting superconducting islands. This result raises the problem of the existence of intrinsic

inhomogeneities in superconducting high  $T_c$  materials. In a more recent work, Safar *et al.* [8] have confirmed, with vibrating reed measurements, that something drastic is happening in the flux lattice below the irreversibility line. Nevertheless, the irreversibility line appears to follow equation (1) close to  $T_c$ .

A theoretical work of Tachiki and Takahashi [9] has shown that the layer structure itself strongly pins vortices. The superconducting order parameter is modulated along the c-axis of the crystals with the period of the lattice constant. This modulation acts as natural pinning centers. Then, the existence of a phase transition which should produce a flux-lattice melting appears to be an open question [6]. If the layer structure [9] itself is at the origin of the strong pinning observed in all these materials at low temperatures, we could expect that the flux-lattice melting, if it exists, must be due to the disappearance of this lattice pinning energy at a certain field level.

The purpose of this paper is to determine the « irreversibility line » of a  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  granular material. This formula has been established some time after the discovery of superconductivity in bismuth compounds [10, 12]. The material we used, has been prepared at the CRTBT-Grenoble following a known procedure [13].

We report a systematic study of the magnetization and of the a.c. susceptibility in a superimposed d.c. field. We find, for the first time that, far from  $T_c$ , the « irreversibility line » follows an exponential decrease with temperature and that the reversible magnetization exists on a large scale of fields and temperatures far from  $H_{c2}$  and  $T_c$ . We show how these new results can be naturally interpreted by the existence in the layered structure of normal zones composed of BiO-SrO planes in which superconductivity is induced by a proximity effect [14] with superconducting Cu-O layered blocks. This new interpretation is consistent with the suggestion of Tachiki *et al.* [9]. The breakdown field for the proximity effect weakens the correlation between vortices and consequently the intrinsic pinning energy of the structure. The reversible magnetization obeys the known laws for an Abrikosov lattice [15] of interacting vortices. *Consequently, interacting vortices are always present beyond the breakdown field.*

The flux creep regime occurs near  $T_c$  in fields lower than the breakdown field. Equation (1) is then recovered. There is a crossover regime in the temperature range where the « irreversibility line » meets the breakdown field line.

### Experimental results.

The « 2223 » phase is prepared by solid state reaction of a mixture of  $\text{Bi}_2\text{O}_3$ ,  $\text{PbO}$ ,  $\text{CuO}$ ,  $\text{SrCO}_3$  and  $\text{CaCO}_3$ . The initial composition is  $\text{Bi}_2\text{Pb}_{0.6}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ . An excess of bismuth oxide is used. The powder is calcinated three times in air at  $800^\circ\text{C}$  during 24 hours. Then, the powder is pressed into a pellet and heated in air at  $860^\circ\text{C}$  for 200 hours and slowly cooled down ( $50^\circ\text{C/h}$ ) to room temperature. The X-ray diffraction pattern of figure 1 shows the presence of a major phase corresponding to the  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  lattice. The weak extra peaks correspond to the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  and  $\text{Ca}_2\text{PbO}_4$  phases. A bar-shape specimen of about  $1 \times 1 \times 8 \text{ mm}^3$  is used for the magnetization measurements.

The d.c. magnetometer has a sensitivity in low fields of about  $10^{-6}$  e.m.u. We used an extraction method to produce the signal in the pick-up coils. The time dependence of the voltage which is induced by the detection coil is measured with a superconducting chopper. The signal is integrated to obtain the magnetic moment. The typical time for stabilizing the temperature is 10 min. Each point of measurement needs several extractions to determine the corresponding magnetization. From point to point there is an interval of about 15 min.

The a.c. susceptibility is measured at 9 Hz using a mutual inductance coil working at 4 K, followed by a low noise amplifier and a lock-in detector. The best sensitivity can be as low as

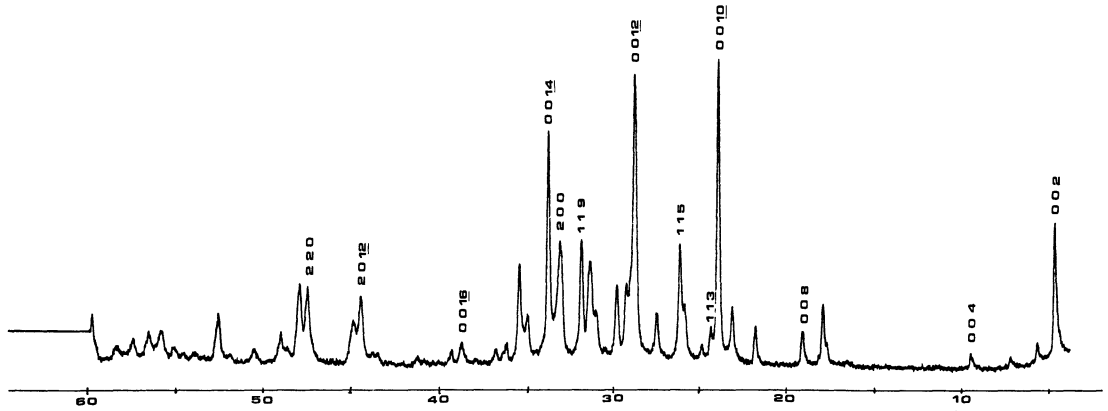


Fig. 1. — The X-ray pattern of a powder of a  $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  compound is presented. The spectrum corresponds to the « 2223 » phase with some extra peaks belonging to  $\text{Ca}_2\text{PbO}_4$  and to  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  minor phases.

$10^{-10}$  e.m.u. with an a.c. field amplitude of 1 Oe. The specimen can be removed from the cell, the mutual inductance coils staying at 4 K. The d.c. field is produced by a superconductive coil surrounding the detection coils.

In figure 2, the zero field-cooled and field-cooled susceptibilities measured in a 10 Oe magnetic field, applied at 4 K, are plotted *versus* the temperature. A relatively sharp transition is observed below 110 K. The  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  phase gives a contribution of about 10 % to the Meissner effect observed below 75 K. The zero field-cooled susceptibility is larger than  $-1/4 \pi$  as expected for independent superconducting grains ( $\chi = \frac{-3}{8 \pi}$  for spherical grains). The slope  $dM/dH$  is larger in very low fields because superconducting percolation between the grains of a low density material leads to a large susceptibility per unit mass. In the insert of figure 2, the magnetization measured at 4 K is plotted *versus* the field ( $0 < H < 10$  Oe). We observe a crossover in a 4 Oe field which corresponds to the breakdown of the superconducting paths between the grains.

The « irreversibility line » is determined by measuring the zero field-cooled and the field-cooled magnetizations, with applied fields at 4 K ranging from 10 Oe to 50 kOe. This procedure shows the occurrence of a strong flux trapping below a certain temperature  $T^*$  in an applied field  $H$ . In figure 3, the magnetization measured in 10 kOe, 2 kOe and 500 Oe is plotted *versus* the temperature. The reversible magnetization above  $T^*$  decreases linearly with  $T$  up to a temperature very close to  $T_c$ . Hence  $\left(-\frac{dH_{c2}}{dT}\right)_{T_c}$  is very large even though  $-\frac{dH^*}{dT}$  appears to be very small around  $T_c$ . The large increase of the zero field-cooled magnetization below  $T^*$  could correspond to a slow relaxation in the flux creep phenomenon or to the appearance of a large pinning energy which stabilizes the flux-line lattice. In figure 3, the increase of the diamagnetism below  $T^*$  appears to be very abrupt. It suggests a phase transition from a normal to a superconducting state for some parts of the sample.

In figure 4, the zero field-cooled and field-cooled magnetizations are plotted on a different magnetization scale. Note the large irreversibility at 4 K corresponding to high critical currents.

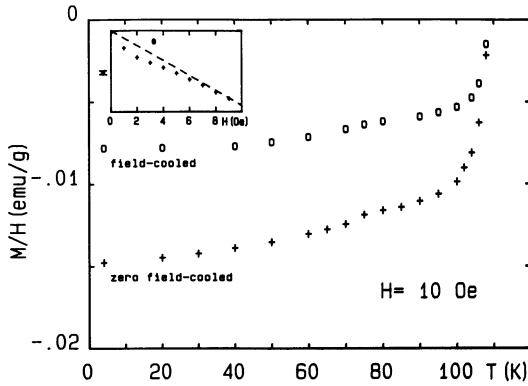


Fig. 2.

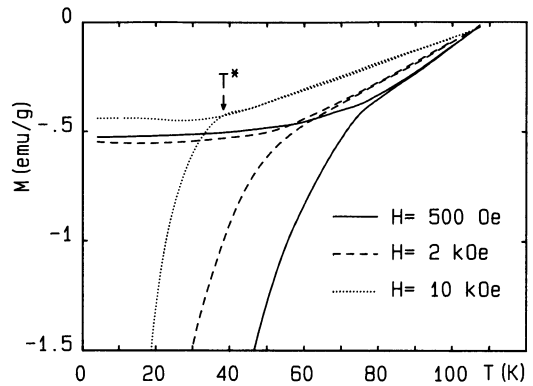


Fig. 3.

Fig. 2. — The zero field-cooled susceptibility (+) and the field-cooled susceptibility (O) of  $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  are plotted *versus* temperature. The applied field is 10 Oe. In the insert the zero field-cooled measured at 4 K is plotted *versus* the field for  $0 < H < 10$  Oe.

Fig. 3. — The zero field-cooled and the field-cooled magnetizations are measured *versus* temperature in different applied fields ( $H = 500$  Oe, 2 000 Oe, 10 000 Oe). Note the linear variation of magnetization *versus* temperature in the reversible region and the abrupt change of the slope at  $T^*$ . The error on magnetization near  $T_c$  does not permit to evaluate the field dependence of  $T_c$ .

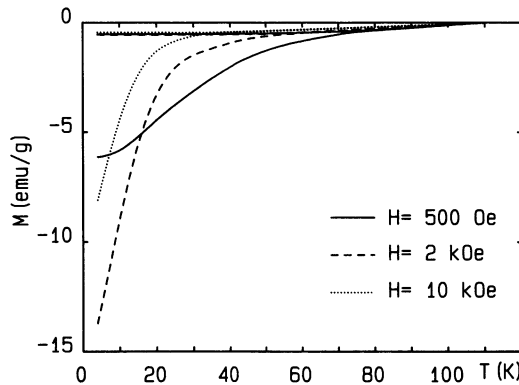


Fig. 4. — The zero field-cooled and the field-cooled magnetizations are plotted *versus* temperature from 4 K to 110 K in different applied fields ( $H = 500$  Oe, 2 000 Oe, 10 000 Oe). Note the large irreversibility at low temperatures corresponding to high critical currents.

As shown in figure 5a, the « irreversibility line »  $H^*(T)$  can be fitted to an exponential form :

$$H^* = 147\,000 e^{-T/13.3} \tag{3.1}$$

$T$  being in Kelvin and  $H^*$  in Oersteds.

This equation is not obeyed close to  $T_c$  (it would lead to a finite breakdown field at  $T_c$ ). Figure 5b shows that  $H^*$  varies as  $(1 - T/T_c)^{3/2}$  close to  $T_c$  as already observed in  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_{4-\delta}$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The exponential behavior covers a large range of temperatures followed by an abrupt fall leading to the universal behavior  $H \sim (1 - T/T_c)^{3/2}$ .

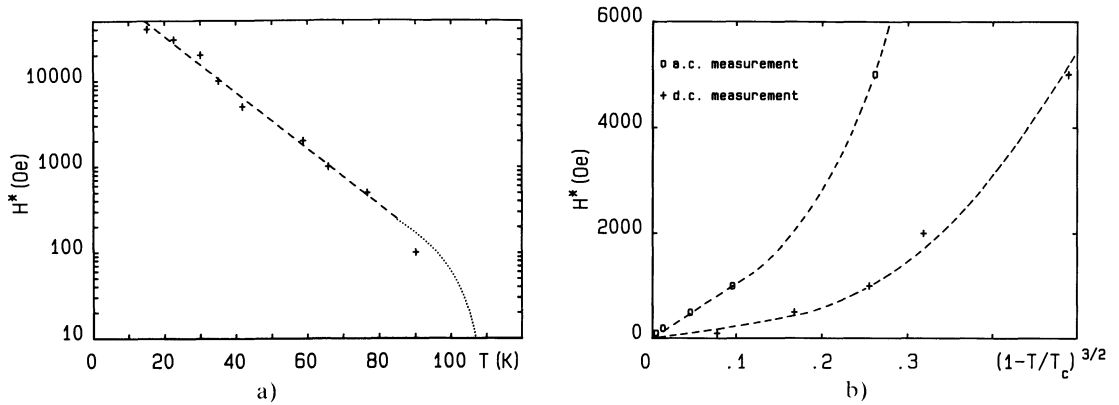


Fig. 5. — (a) The data (+) corresponds to the temperature  $T^*$  below which a strong increase of the zero field magnetization is observed in different applied fields  $H$ . Note the exponential decrease of  $H^*$  versus  $T$ .  $H^* = 147\,000 e^{-T/13.3}$ . Near  $T_c$ ,  $H^*$  has been extrapolated with a  $\left(1 - \frac{T}{T_c}\right)^{3/2}$  law (dotted line). (b) The field and the temperature in which the irreversibility of the d.c. magnetization occurs are plotted in the following diagram  $H^*$  versus  $\left(1 - \frac{T}{T_c}\right)^{3/2}$ . The crosses (+) correspond to the temperature below which a large increase of the zero field-cooled occurs. The circles (0) correspond to the temperature in which the a.c. diamagnetic susceptibility in a constant field disappears. Note that at high temperatures  $H^* \sim \left(1 - \frac{T}{T_c}\right)^{3/2}$ . Note also the time dependence of the irreversibility line.

In figure 5b, the large influence of the time of measurement on the « irreversibility line » near  $T_c$  is shown. For an a.c. measurement at a frequency of 9 Hz we have :

$$H^* \approx 10\,400 \left(1 - T/T_c\right)^{3/2}. \quad (3.2)$$

This equation is determined using the field in which the a.c. susceptibility goes to zero (see Fig. 6).

In figure 6, the a.c. susceptibility measured at a frequency of 9 Hz is plotted versus temperature. It has been measured in an a.c. field amplitude of 0.1 Oe in different superimposed d.c. fields varying from 0 to 9 kOe. The d.c. zero field-cooled and field-cooled magnetizations are also plotted in the same figure. The experimental points in the d.c. magnetization are taken each 15 min. Diamagnetism still exists when the diamagnetic a.c. susceptibility has completely disappeared. For reversible superconductor in the mixed state and below  $T_c$ , a positive a.c. susceptibility is even expected which is too small here to be measurable. Magnetization appears to be fully reversible. The pinning centers have in no way any effect on vortices in a large range of fields and temperatures. No critical current exists in any part of the sample, whatever the orientations of the crystals of this sintered material are. Thus, the pinning energy appears as to be abruptly weakened above a certain field applied along the  $c$ -axis as well as in the  $(ab)$  plane ;

In figure 7, the field dependence of the magnetization normalized by  $(1 - T/T_c)$  is plotted on a semi-logarithmic scale at  $T = 77, 40$  and  $30$  K. The equilibrium magnetization  $M_{eq}$  below  $H^*$  is calculated from the hysteresis cycle.  $M_{eq}$  is assumed to be equal to :

$$M_{eq} = \frac{M \uparrow (H) + M \downarrow (H)}{2}$$

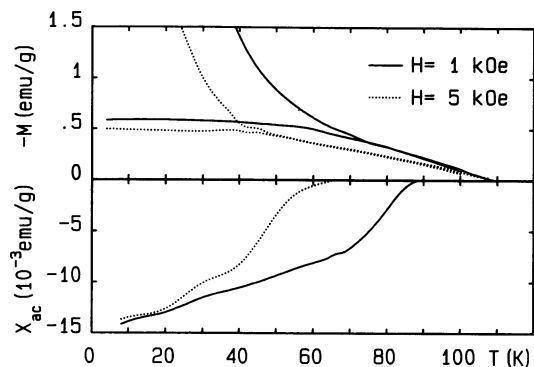


Fig. 6.

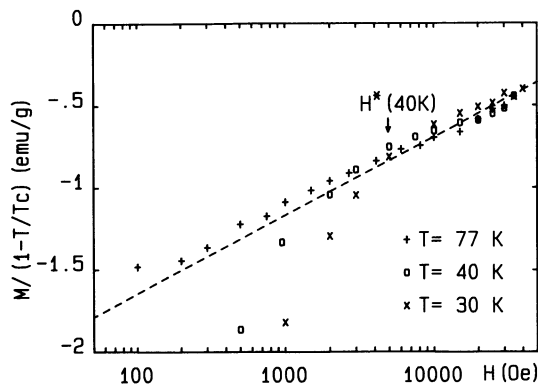


Fig. 7.

Fig. 6. — The a.c. susceptibility and d.c. magnetization are plotted at high temperatures. Note that the a.c. susceptibility has completely disappeared while the sample is still diamagnetic.

Fig. 7. — The magnetization in the reversible state is scaled by  $1 / \left(1 - \frac{T}{T_c}\right)$  versus the applied field. Above  $H^*$ , the reversible magnetization linearly varies with  $\ln H$ . Below  $H^*$ , the equilibrium magnetization deviates from the universal law (Eq. (4.2)) at 30 K and 40 K. The dotted line has for equation  $M / \left(1 - \frac{T}{T_c}\right) = 0.207 (\ln H - 12.6)$ .

$M \uparrow (H)$  is taken on the ascending branch, and  $M \downarrow (H)$  on the descending branch of the hysteresis cycle. The reversible magnetization decreases linearly with  $(\ln H)$  and is proportional to  $\left(1 - \frac{T}{T_c}\right)$  over a large range of temperatures. The equation :

$$M = 0.183 \left(1 - \frac{T}{T_c}\right) (\ln H - 13.0) \tag{4.1}$$

is followed within an error bar of about 10 % in all temperature range,  $M$  being in e.m.u./g and  $H$  in Oersteds. At 30 and 40 K, an additional contribution to the equilibrium magnetization is added below  $H^*$  to equation (4.1). This contribution, which is negligible at 77 K progressively increases when  $H$  decreases or when  $T$  decreases.

In figure 8, the reversible part of different  $M(T)$  curves in constant field have been used to plot  $M / \left(1 - \frac{T}{T_c}\right)$  versus  $(\ln H)$ . The data noted « 0 K » are in fact extrapolated from the reversible part of the  $M(T)$  curves (see insert). Magnetization follows the law :

$$M = 0.207 \left(1 - \frac{T}{T_c}\right) (\ln H - 12.6) . \tag{4.2}$$

Equation (4.2) is a better representation in low fields ( $H < 10$  kOe) ; some corrections are needed because of the existence of a demagnetizing field and also because the applied field is not far from the first critical field  $H_{c1}$  (see Eq. (7)).



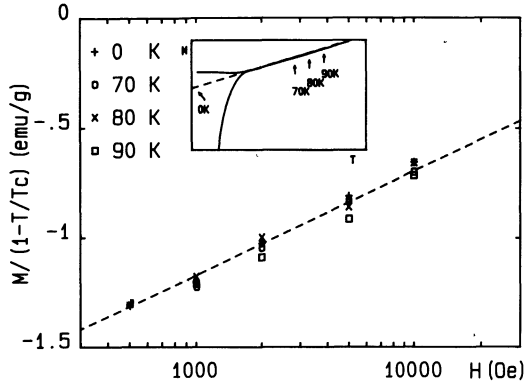


Fig. 8. — The reversible magnetization has been measured as a function of temperature in a constant applied field  $H$ . The magnetization  $M$  is divided by  $\left(1 - \frac{T}{T_c}\right)$  and plotted versus  $\ln H$ . The points noted (+) are in fact extrapolated at zero Kelvin as indicated in the insert. The influence of the variation of  $\ln \frac{\phi_0}{4 \pi \xi^2}$  appears to be very small in equation (7). Equation (4.2) is the best representation of the temperature region for which  $\ln \frac{\phi_0}{4 \pi \xi^2}$  is roughly temperature independent.

**Interpretation.**

The main experimental observation in this paper is the exponential decrease with temperature of the irreversibility line (see Eq. (3)). It could correspond to the disappearance of a large fraction of the pinning energy in a certain applied field. A large part of the correlation energy of vortices would be destroyed. This proposal has nothing to do with an explanation based on thermally activated processes because the breakdown field  $H^*$  ( $H^* \ll H_{c2}$ ) exists even at low temperatures when the quantity  $\frac{U_0}{kT}$  becomes very large. Tachiki *et al.* [9] have recently proposed that the strong vortex pinning observed in YBaCuO compounds could be intrinsic. The layered structure itself would provide strong pinning, the CuO<sub>2</sub> layers and neighboring ones being strongly superconducting and the layers with Cu-O chains and neighboring ones weakly superconducting. The vortices would be stabilized in the weakly superconducting layers. This mechanism would also work in Tl and Bi compounds which have a similar layered structure.

The layered structure is actually more pronounced in Bi and Tl compounds because the CuO chains do not exist. The block SrO-BiO-BiO-SrO is more weakly superconductive, as indicated by the larger anisotropy of superconductivity in these compounds [18]. We suggest that this weak superconductivity could be destroyed in a relatively small field leading to a super-lattice of superconducting blocks composed of 3 CuO layers separated by normal blocks of about 12.16 Å thick. At low fields, superconductivity would be induced in the « normal » blocks by the proximity effect from the neighboring CuO layer blocks. In the proximity effect [14], the breakdown field varies exponentially with temperature following the relation :

$$H_b = H_0 \exp \frac{-d_N 2 \pi k_B T}{\hbar V_N} \tag{5}$$

where  $d_N$  is the thickness of the normal block (limited by a free surface parallel to the applied field),

$V_N$  is the Fermi velocity in the normal part and

$H_b$  is calculated in the « clean » limit.

Equation (5) is valid in the low temperature limit where the transition at  $H_b$  is first order.

The field  $H_0$  is then well approximated by  $H_0 = \frac{3.8 \phi_0}{2 \pi \xi_N \lambda(0)}$ , where  $\lambda(0)$  is the London

penetration depth at the interface. Although the domain of validity of equation (5) does not cover the geometry at hand here (stack of layers, different orientations of the field), it does contain the basic physics involved, i.e. the ability of a normal layer to carry Meissner currents up to a certain field.

If we take  $d_N = 12.16 \times 10^{-8}$  cm we obtain  $V_N = 1.33 \times 10^6$  cm/s as compared to  $V_F = 1.92 \times 10^7$  cm/s in CuO blocks. The latter value is deduced from relation (6), using  $\xi_0 = 24 \text{ \AA}$  (as experimentally deduced from equation (7) and discussed later) :

$$\xi_0 = 0.18 \frac{\hbar V_F}{k T_c} \quad (6)$$

$V_N$  is an order of magnitude smaller than  $V_F$ .

The coherence length at 110 K in the boundary between the normal and the superconducting blocks would be equal to  $\xi_{0N} = 1.66 \text{ \AA}$  using relation (6). This value is roughly equal to Cu and O atom spacing. The coherence lengths  $\xi_c$  and  $\xi_{ab}$  of YBaCuO are 3 and 15  $\text{\AA}$  respectively as recently demonstrated by Welp *et al.* [16]. The values  $\xi_c = 1.66 \text{ \AA}$  and  $\xi_{ab} = 24 \pm 4 \text{ \AA}$  are possible in  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  because the anisotropy of the critical field is much larger in Bi compounds [18]. It is thus tempting to identify  $\xi_{0N}$  and  $\xi_c$ .

The disappearance of critical currents above  $H^*$  as demonstrated by the absence of diamagnetism in the a.c. susceptibility measurements shows that the sample is in a flux flow regime or in a melted flux-lattice as suggested by Gammel *et al.* [6]. The proximity effect interpretation gives a justification for the existence of a phase transition at low temperatures. Nevertheless, an Abrikosov lattice seems to be always present because reversible magnetization obeys the law of magnetization of interacting vortices (Eq. (7)). The flux-lattice melting corresponds to a strong weakening of the vortex correlation energy. A flux flow phenomenon is observed. We suggest that it occurs in the CuO stacks, with no interstack correlation above  $H^*$ . The vortex lattice remains ordered in the individual CuO stacks, but there is no 3D ordering.

The strong intrinsic vortex pinning energy is destroyed at the breakdown field, in which screening currents in the normal blocks are quenched. The critical current which can be very high at low temperatures ( $2 \times 10^7$  Ampères/cm<sup>2</sup> below 40 K as recently shown on textured thin films), is indeed easily destroyed in a small field of the order of 5 kOe [19].

Another important observation is the extended range of fields and temperatures where reversible magnetization can be measured. In a classical superconductor, magnetization is reversible only over a small range of fields and temperatures. It is much easier to observe it in high  $T_c$  superconductors because the energy barrier  $U_0$  is weakened by the small coherence length [5]. For  $H_{c1} \ll H \ll H_{c2}$  and extreme high  $\kappa$  classical superconductors with an ideal flux-line lattice, the equilibrium magnetization is given by [15] :

$$4 \pi M = \frac{\phi_0}{8 \pi \lambda^2} \left[ \ln (H - H_{c1}) - \ln \frac{\phi_0}{4 \pi \xi^2} \right] \quad (7)$$

$\ln(H - H_{c1})$  can be replaced by  $\ln H$  far from  $H_{c1}$ . The different parameters can be calculated by comparing equation (7) to equation (4.2). The quantity  $\ln \frac{\phi_0}{4 \pi \xi^2}$  can be determined from the second term in parenthesis in equations (4.2) and (7); this leads to a value of  $\xi = 24 \pm 4 \text{ \AA}$ . Equation (7) can be roughly represented by equation (4.2) because  $\ln \frac{\phi_0}{4 \pi \xi^2}$  is slowly varying with temperature. In addition equation (4.2) is fitting well the experimental points shown in figure 7 above 5 kOe at low temperatures.

The penetration depth  $\lambda$  can be roughly determined introducing a correction factor of 2 to account for the uniaxial magnetization distribution in all the directions of space. The superconducting volume has to be corrected because the sample contains some normal minor phases which can be evaluated to 20 %.

The coefficient 0.207 in equation (4.2) has then to be taken equal to about 0.52, giving  $\lambda = 1\,500 \text{ \AA}$ . The  $\xi_{ab}$  and  $\lambda_{ab}^c$  ( $\lambda$  in the  $(a, b)$  plane with  $H$  applied along  $c$  axis) values of  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  and  $\text{YBaCuO}$  are similar. The reversible magnetization of our polycrystalline sample is dominated by the contribution of the  $c$ -axis magnetization, because the  $c$  axis component of the magnetization is large even if the field is far from the  $c$  axis. In the superconducting superlattice model that we have developed, the reversible magnetization measured along the  $(ab)$  plane is expected to be negligible because the penetration depth is much larger than the thickness ( $6.4 \text{ \AA}$ ) of  $\text{CuO}$  layered blocks.

The linear decrease of the magnetization of the  $\text{CuO}$  layers with temperature is due to the temperature dependence of the penetration depth :

$$1/\lambda^2 = \frac{1}{\lambda_0^2} (1 - T/T_c). \quad (8)$$

This law is expected to be followed near  $T_c$  in weak coupling superconductors. Krusin-Elbaum *et al.* [20] have observed that it works very close to  $T_c$  in  $\text{YBaCuO}$ . Our magnetization measurements show that it is obeyed further away from  $T_c$  than expected.

A linear decrease of the magnetization has also been recently observed in  $\text{YBaCuO}$  crystals for a narrow range of temperatures close to  $T_c$  [16]. We note that an interpretation of our data figure 7 in terms of equation (7) and equation (8) implies that  $\ln H_{c2}$  is temperature independent.

A cross-over separates the two regimes defined by equation (1) and equation (3). The cross-over temperature depends on the time of the measurement. This could indicate a large change of the activation energy  $U_0$  with temperature instead of a phase transition.

Alternatively, in the proximity effect interpretation, a first order transition is expected at the breakdown field at low temperatures [14]. The appearance of additional magnetization below  $H^*$  shows the existence of a new contribution to the Gibbs free energy  $G$  because  $\left( \frac{\delta G}{\delta H} \right)_T = -M$ , suggesting a phase transition. The additional magnetization appears to be negligible at high temperatures and to increase strongly at low temperatures. There is no appreciable discontinuity in the magnetization above 20 K at  $H = H^*$ . Flux jumps are observed below 20 K which might hide any observation of a possible first order transition.

## Conclusions.

The  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  phase has been prepared with a relatively abrupt transition in the Meissner effect at  $T_c \simeq 110 \text{ K}$ . The « irreversibility line » has been studied for the first time in

this material. Near  $T_c$ , the law  $H^* \sim \left(1 - \frac{T}{T_c}\right)^{3/2}$  is followed ; it appears as being universal [1, 3, 7]. At lower temperatures, a new phenomenon dominates :  $H^*$  is exponentially decreasing with  $T$ ,  $H^* \sim e^{-T/T_0}$ .

The reversible magnetization of interacting vortices in an Abrikosov lattice can be analyzed using the classical calculations for high  $\kappa$  ideal superconductors in intermediate magnetic fields  $H_{c1} \ll H \ll H_{c2}$ . The reversible magnetization  $M$  decreases very slowly with increasing field because the leading term is  $-\ln(H - H_{c1})$ . Main superconducting parameters such as  $\xi$  and  $\lambda$  of the ideal superconductor can be determined from the reversible magnetization curve.  $M$  decreases linearly with temperature even far from  $T_c$ . This behavior is attributed to the thermal variation of  $\lambda(T) = \lambda_0 \left(1 - \frac{T}{T_c}\right)^{-1/2}$ .

The exponential decrease of the « irreversibility » line can be interpreted as due to the existence of a breakdown field of proximity effect induced superconductivity. The Cu-O layered blocks would be the superconducting entities separated by normal blocks such as the SrO-BiO-BiO-SrO layered blocks. The thickness of these normal blocks is compatible with the experimental exponential decrease of the breakdown field.

The layered structure of bismuth compounds leads not only to a quasi two-dimensional situation [21], but to a superlattice of layered blocks alternatively superconducting and normal above a breakdown field of induced superconductivity. At the breakdown field, superconductivity is quenching in the normal layers, inducing a strong weakening of the activation energy  $U_0$  that must be overcome to allow vortex motion. This transition is not a complete melting of the Abrikosov lattice because at  $H > H^*$  the reversible magnetization  $M(H)$  follows the laws expected for an ideal Abrikosov lattice.

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