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Reversible magnetization of radiation-disordered $YBa_2Cu_3O_{7-x}$ single crystals

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Single crystals of $YBa_2Cu_2O_7$ were exposed to fast neutron irradiation with fluences up to 10^{19} n/cm^2 ($E > 0.1$ MeV). Ac and dc magnetization measurements were performed to study the influence of irradiation-induced defects on the intrinsic and extrinsic parameters of superconductors. It has been shown that the observed increase of the anisotropy of the lower critical field in irradiated samples is due to the suppression of the Bean-Livingston surface barrier. Drastic changes of intrinsic parameters of $YBa_2Cu_3O_7$ single crystals are interpreted within the framework of existing theoretical models.

I. INTRODUCTION

The generation of defects by neutron irradiation has shown to be a useful method for the modification of magnetic and transport properties, both in the superconducting as well as in the normal state of high-T_c superconductors (see Ref. 1 for references). Among the various members of the high-T_c family of superconductors, $YBa_2Cu_3O_{7-x}$ with $T_c \simeq 90$ K stands out as a promising candidate for technical application, thus the effect of neutron irradiation has been the most frequently studied in this compound. It is now well established[2] that at the low fluence regime (up to $\simeq 4\times 10^{17}$ n/cm^2) the transition temperature is only slightly reduced in the $YBa_2Cu_3O_{7-x}$ compound. In this regime the radiation-induced changes observed in the magnetization or transport measurements are related first of all to changes in the defect structure responsible for flux pinning rather than to changes in electronic structure of superconductors. Higher fluences make the decrease of T_c more serious, to about 4 K/10^{18} n/cm^2 for fluences close to 1×10^{18} n/cm^2.[3] Further increase of fluence leads to a sharper drop of the critical temperature and in consequence to a transformation from the superconducting to the normal state at doses which correspond to 0.02–0.04 displacement per atom.[4] Generally, at higher fluences an amorphization process becomes dominant. It has been shown (see Ref. 3 and references therein) that the degradation of T_c under irradiation is accompanied by an essential change in the character of electrical resistivity in the normal state (it increases exponentially with the concentration of created defects). Therefore it was suggested that the degradation of T_c is due to localization effects arising from radiation-induced disorder. In Ref. 4 another approach to this problem has been developed. It has been supposed that the degradation of T_c under high fluence irradiation is due to some kind of destruction of the oxygen sites along the chains leading to the reduction of the electron transfer between the CuO_2 planes and the chains (while the stoichiometry of oxygen remains unchanged). It should be remembered that the supercurrent in $YBa_2Cu_3O_{7-x}$ compounds is carried in CuO_2 planes that are intercalated with layers of BaO and CuO. These intercalating layers, being not directly involved in the conductivity, act as charge reservoir (holes) to control the carrier density in CuO_2 planes. On the other hand, the CuO_2 planes develop an antiferromagnetic order in the insulating state when the hole doping from the reservoir is small but they become superconducting when the concentration of delocalized holes exceeds a few percent.[5] Since the oxygen atoms along the chains have vacant lattice sites nearby and are loosely bound (in comparison to oxygen atoms in CuO_2 plane) in the lattice, they are the dominant source of atomic displacement under irradiation. For low fluences the chain breaking has no effect on T_c. For higher fluences an atomic disorder in the charge reservoir increases (typically in the form of vacancies and interstitials in the oxygen sublattice) changing the density of states at the Fermi level and consequently decreasing T_c. It means that the local ordering along the chains leading to the charge transfer between the chains and planes plays a crucial role in determining the superconductivity of the $YBa_2Cu_3O_{7-x}$ system.

Most of recent works on neutron damage in $YBa_2Cu_2O_{7-x}$ superconductors have been done in the low fluence regime. An important result of these investigations is the report of enhanced critical current density in the $YBa_2Cu_3O_{7-x}$ single crystals upon neutron irradiation.[6,7] At low neutron fluence, the enhancement of critical current density is attributed to the creation of defects acting as pinning centers.[8,9]

To obtain deeper insight into the irradiation-induced changes in superconducting and magnetic properties of high-T_c superconductors we have performed ac and dc magnetic-susceptibility measurements of neutron-irradiated $YBa_2Cu_3O_{7-x}$ single crystals. The work was especially concentrated on the magnetization measurements in the reversible part of $M(H)$ dependences providing information on the values of the penetration depth and on the low magnetic-field regime in the vicinity of the lower critical field.

II. EXPERIMENT

Single crystals of $YBa_2Cu_3O_{7-x}$ were grown by the crystallization from melt.[3] The samples had the form of

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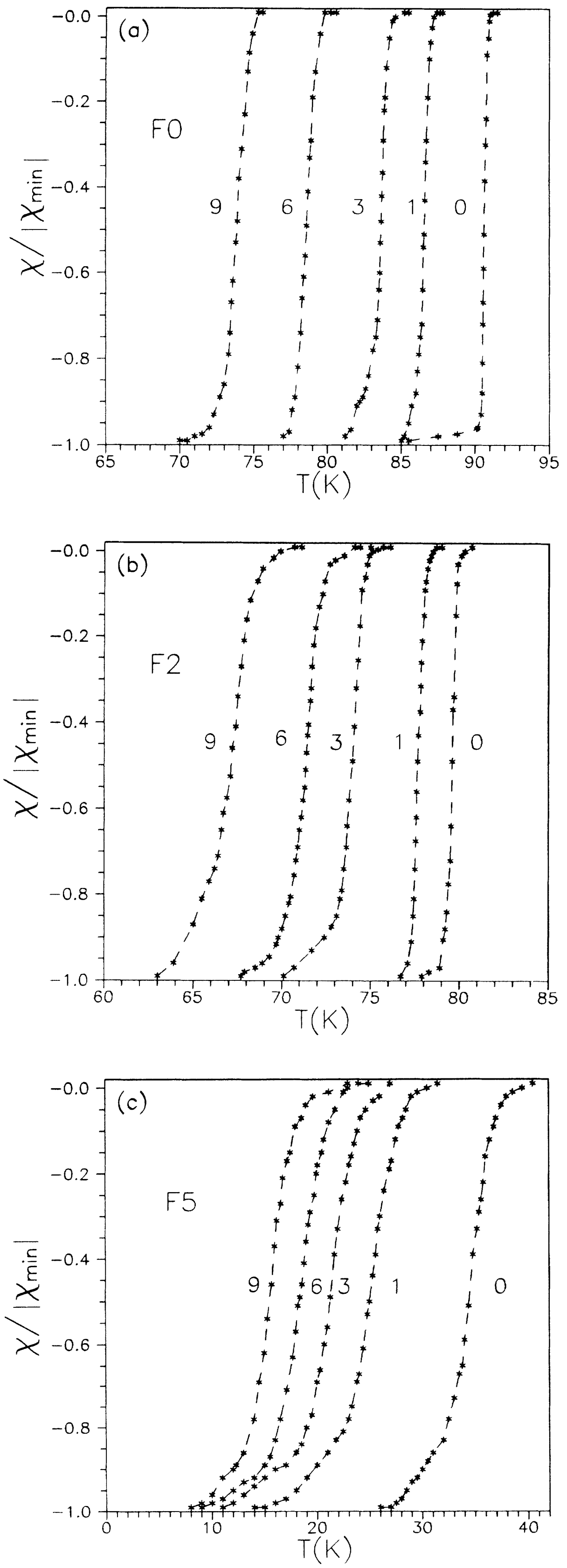


FIG. 1. The temperature dependences of ac susceptibility, $\chi(T)$, in constant magnetic fields ($H \| c$) up to 9 T; (a) for reference sample F0; (b) and (c) for irradiated samples F2 and F5, respectively.

platelets with the c axis perpendicular to the surface and with typical sizes of the order of $2\times2\times0.05$ mm^3. The samples were irradiated at 80 K by fast neutrons with fluences of 2×10^{18} n/cm^2 (sample F2), 5×10^{18} n/cm^2 (sample F5), and 1×10^{19} n/cm^2 (sample F10).

The alternating-current (ac) magnetic-susceptibility measurements were done with field h_{ac} less than 1 Oe parallel to the c axis and with external magnetic field H up to 9 T. The frequency of the ac field was taken to be 10 kHz. The results of measurements for F0 (unirradiated reference sample), F2, and F5 samples are presented in Figs. 1(a), 1(b), and 1(c), respectively.

All the dc magnetization measurements were done with a superconducting quantum interference device magnetometer, equipped with a superconducting 5-T magnet, at temperatures between 4.2 and 300 K and in the magnetic field parallel to the c axis. At first, the relations of the magnetization on the temperature were investigated in the low magnetic field (5 Oe) in zero-field-cooled (ZFC) and field-cooled (FC) regimes. In Fig. 2 one can see that ZFC susceptibility, corrected for the demagnetizing field, even for irradiated samples is close to that of an ideal diamagnet. The FC signals are much less than the corresponding ZFC ones. This incomplete Meissner effect has, at least, two explanations.[10] One is based on flux-pinning effects,[11] while the other is related to the mesoscopic defect structure of high-T_c materials.[12] Since the value of the Meissner effect is practically the same for F0, F2, and F5 samples and does not depend on defects introduced during irradiation it seems reasonable to suppose that the mechanism proposed in Ref. 12 is responsible for the observed effects. The obtained values of magnetization T_c onset correspond well to the values of T_c determined using the ac technique and are equal to 34.3, 79.6, and 90.6 K for F5, F2, and F0 samples, respectively.

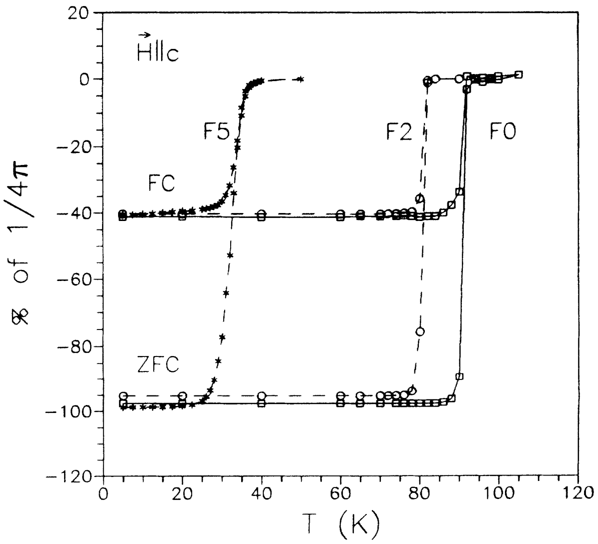


FIG. 2. The dc susceptibility versus temperature for reference (F0) and irradiated samples (F2,F5), in zero-field-cooling (ZFC) and field-cooling (FC) regimes. The values of susceptibility were corrected for the demagnetizing field and related to the susceptibility of an ideal diamagnet.

It was stated that the sample F10 was not superconducting at temperature down to 4.2 K.

The normal-state dc magnetic susceptibility ($\chi \equiv M/H$) was determined in the range of temperature from T_c to room temperature in relatively strong magnetic fields (in the range 5–40 kOe) because of small magnetic moments related to low sample sizes. It is worth stressing that the values of M/H were field independent sufficiently far from appropriate T_c. The data show (see Fig. 3) that, except the reference sample, for all the irradiated samples (F2, F5, and F10) the Curie-Weiss contribution

$$\chi = A + C/(T+\theta) ,$$

where $\theta = 7.5$ K, becomes really significant.

The lower critical field, H_{c1}, was determined using the dc magnetization technique. Generally, H_{c1} for type-II superconductors corresponds to the magnetic field at which the virgin magnetization versus field curve deviates from linearity. In practice, the detection of this field value is difficult, because the strong local demagnetizing effects at the corners of the sample make possible some flux penetration even at fields much below H_{c1}. Therefore for the magnetic field near H_{c1} no sudden flux penetration occurs. It has been demonstrated using various experimental techniques (e.g., a miniature Hall probe,[13] visual observations[14]) that the measured H_{c1} is much larger in the sample before electron or neutron irradiation. It demonstrates the existence of the Bean-Livingston barrier to flux penetration which has also significant influence on the experimentally determined H_{c1} data. For all the mentioned reasons we will limit ourselves to a qualitative estimation of the effect of neutron irradiation on H_{c1}. We have observed that

(i) irradiation strongly decreases the values of H_{c1} (in agreement with previous observations[13,14]);

(ii) irradiation strongly increases anisotropy of H_{c1} (from about 4 for F0 to about 50 for F5). This result is in contrast with the observed decrease of the anisotropy of electrical resistivity and of the upper critical field upon neutron irradiation.[15]

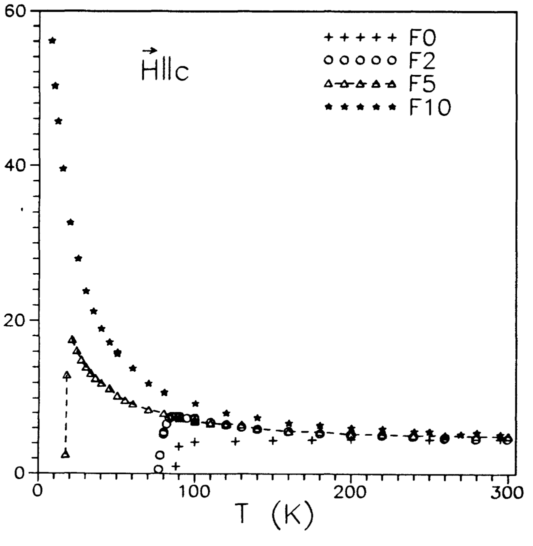


FIG. 3. The temperature dependences M/H for reference F0 and irradiated F2, F5, and F10 samples.

To explain the increase of H_{c1} anisotropy one should take into account that according to theoretical calculations[16,17] the Bean-Livingston barrier weakly depends on the direction of the external magnetic field. It means that the existence of the surface barrier leads to a "hidden" anisotropy which is disclosed due to irradiation. Therefore intrinsic anisotropy of H_{c1} is much higher than that observed in unirradiated samples. The mechanism responsible for high intrinsic anisotropy of H_{c1} is related to the anisotropy of the effective mass. According to Ref. 18 for the quasi-two-dimensional superconductors the lower critical fields $H_{c1}^{\parallel}$ and $H_{c1}^{\perp}$ for external fields parallel and perpendicular to the plane, respectively, are given by

$$H_{c1}^{\parallel} = [\Phi_0/(4\pi\lambda_x\lambda_z)]\ln(\lambda_x/s) , \tag{1}$$

$$H_{c1}^{\perp} = [\Phi_0/(4\pi\lambda_x^2)]\ln(\lambda_x/\xi_z) . \tag{2}$$

Here, λ_x refers to the penetration depth in the x (in-plane) direction, while λ_z is the value of the penetration depth in the $c(z)$ direction. ξ_z is the coherence length in the c direction, Φ_0 is the flux quantum ($=h/2e$), and s is the distance between CuO_2 planes.

The penetration depths λ_i and coherence lengths ξ_i can be written in terms of the effective masses:

$$\lambda_i = \lambda(M_i/M)^{1/2}, \quad \xi_i = \xi/(M_i/M)^{1/2} , \tag{3}$$

$$\lambda = (\lambda_1\lambda_2\lambda_3)^{1/3}, \quad \xi = (\xi_1\xi_2\xi_3)^{1/3}, \quad M = (M_1M_2M_3)^{1/3} . \tag{4}$$

Since the effective mass tensor is highly anisotropic [$M_c/M_a = 26$ for $YBa_2Cu_3O_{7-x}$ (Ref. 19)] one should expect high anisotropy of the intrinsic value of lower critical fields in accordance with expressions (1) and (2).

Magnetic dc and ac measurements have been also used to determine the positions of irreversibility lines, $H_{irr}(T)$. It is well known that a criterion for the determination of the "true" irreversibility line is not universal and depends on the experimental technique used to determine this transition. In our case the values of the irreversibility temperature $T_{irr}(H)$ were determined from ac measurements as the midpoint of the diamagnetic transition according to procedure proposed in Ref. 20. In most cases these values are close to the positions of onset (H_{irr}, T) of the hysteresis in magnetization curves $M(H)$. The obtained values are presented in Figs. 4(a), 4(b), and 4(c) in the form of log-log plots of H_{irr} vs $(1-T_{irr}/T_c)$. The irreversibility lines fit the relation

$$H = H_0(1-T_{irr}/T_c)^n \tag{5}$$

with the values of n equal to 1.6, 1.2, and 3.4 for samples F0, F2, and F5, respectively. The respective values obtained from dc magnetization data are very similar being equal to 1.2, 1.3, and 3.4. The exponent n is a model-dependent parameter. The giant flux-creep model[21] predicts $n = \frac{3}{2}$, a vortex-glass formation theory leads to

$n=\frac{4}{3}$,[22] while for a melting transition of vortex lattice the value $n=2$ is predicted.[23,24]

Presented data raise the intriguing question of the effect of defects on the irreversibility line. It has been demonstrated (Ref. 25, for example), that the irreversibility line upon proton irradiation was lowered, and that the decrease in $T_{\rm irr}$ scales with the decrease in T_c. Our results demonstrate clearly that upon strong neutron irradiation the irreversibility temperature decreases but the reduction in $T_{\rm irr}$ does not scale with reduction in T_c. It is particularly noticeable in the case of sample F5. Another interesting problem concerns the effect of the irradiation on the exponent n. Our data suggest that for samples F0 and F2 the irreversibility line is described in terms of the flux-creep model. Much more difficult to explain is an unexpected high value of n obtained for the sample F5. It seems that only the model developed by Sergeenkov[26] gives the base to understand the high value of exponent n in highly defected samples. This model, although developed for deoxygenated superconductor, seems to be directly applicable to our case, since it is based on two assumptions:

(i) defects may act both as pinning centers as well as weak links;

(ii) the sample contains phases with different magnetic susceptibility.

According to Ref. 26, in samples with a large number of dislocations, the crossover $n=\frac{3}{2}\Longrightarrow n=4$ with increasing field should appear. Such an effect was observed in our studies.

According to the standard Ginzburg-Landau theory, the magnetization depends on the logarithm of the magnetic field H in the range of intermediate magnetic fields $H_{c1}\ll H\ll H_{c2}$.[27] This behavior is well obeyed both in conventional and in high-T_c superconductors. This result has recently been generalized to include the effect of vortex fluctuations in the Josephson-coupled layered superconductors.[28,29] The fluctuations give an extra term to the field relation of magnetization $M(H)$. In the case of $H\parallel c$ the magnetization is described by the expression

$$M(H,T)=\frac{\Phi_0}{32\pi^2\lambda_{ab}^2}\left[\ln\frac{\eta H_{c2}}{eH}-g\ln\frac{gH_{c2}}{\alpha\sqrt{e}H}\right], \qquad (6)$$

where $\lambda_{ab}(T)$ is the in-plane penetration depth, α and η are constants of the order of unity, and

$$g=\frac{32\pi^2k_B}{\Phi_0^2 s}T\lambda_{ab}^2(T)\ . \qquad (7)$$

The slope $\partial M/\partial\ln H$ does not depend on the numerical constants η and α (which cannot be determined within the theory) and can be expressed as follows:

$$\frac{\partial M}{\partial\ln H}=\frac{\Phi_0}{32\pi^2\lambda_{ab}^2(T)}[1-g(T)]\ . \qquad (8)$$

Formula (8) is used in this paper to determine the effect of irradiation on the value of the penetration depth λ_{ab}. In order to analyze the $M(H)$ curve using Eq. (8) the paramagnetic contribution of the normal phase should be subtracted from measured $M(H,T)$ dependences. Such a procedure was successfully applied to sample F2 irradiated with a fluence of 2×10^{18} n/cm^2. But in the case of sample F5, paramagnetic contribution prevails and the accuracy of the above procedure is rather low. For this reason we decided to perform a detailed analysis of the experimental data only for samples F0 and F2.

In Figs. 5(a) and 5(b), there are presented the field dependences of the magnetization M, both for increasing and decreasing field, at various temperatures, for the reference sample F0 and for irradiated sample F2 (after subtraction of the paramagnetic contribution), respectively. These data have been used to evaluate the experimental slopes $\partial M/\partial\ln H$ for $H>H_{\rm irr}$ and to determine $\lambda_{ab}(T)$ using Eq. (8). Since the unit cell of YBa$_2$Cu$_3$O$_7$ contains two pairs of closely spaced ($\simeq3.2$ Å) CuO$_2$ lay-

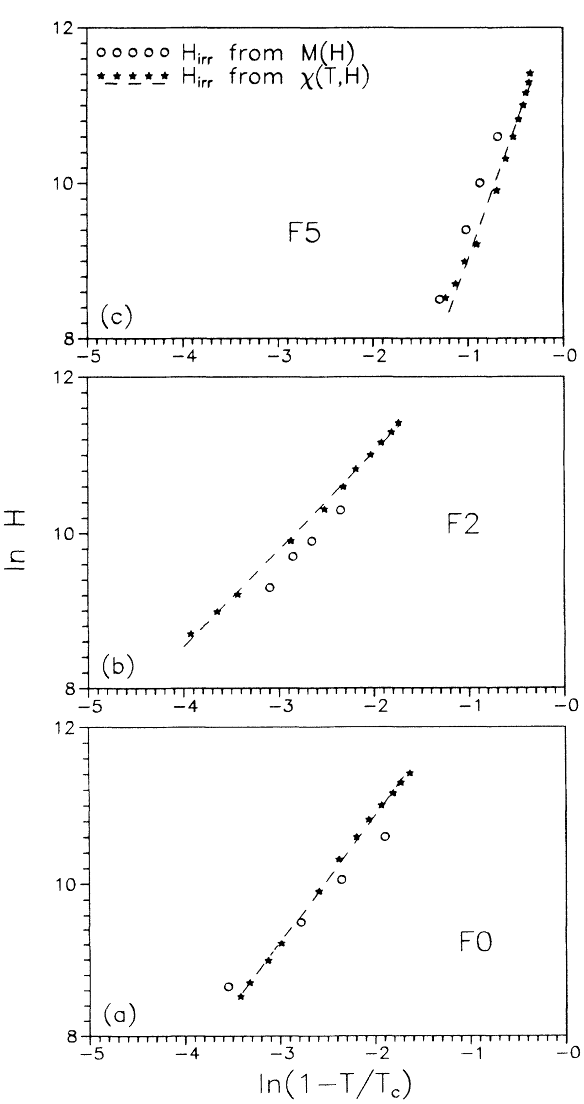
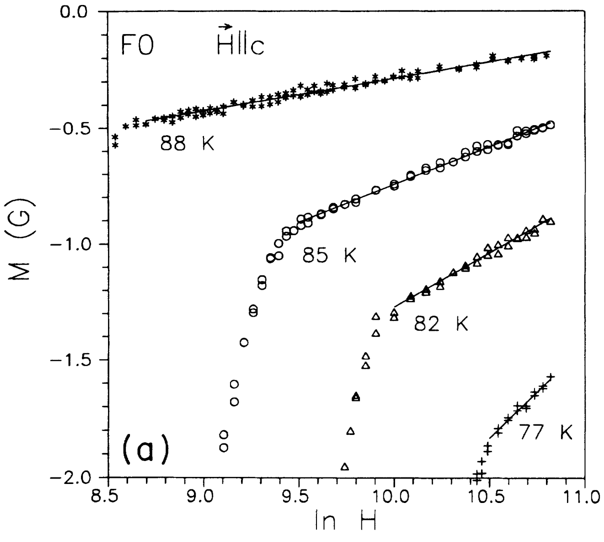


FIG. 4. The log-log dependence of $H_{\rm irr}$ obtained from ac and dc magnetic measurements versus $1-T/T_c$; (a) for reference sample F0; (b) and (c) for irradiated samples F 2and F5, respectively. $H_{\rm irr}$ is expressed in Oe.



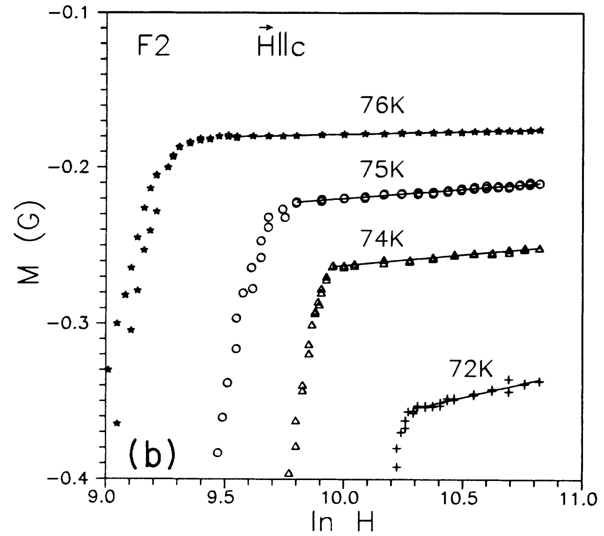


FIG. 5. The field dependences of the magnetization M, both for increasing and decreasing field, at various temperatures; (a) for reference sample F0; (b) for irradiated sample F2. Magnetic field H is expressed in Oe.

ers and the distance between these layers is equal to 11.4 Å, this last value was used for s in the further calculations. The results of those calculations, the values of $1/\lambda^2$ versus T/T_c, are shown in Fig. 6. It is seen that irradiation caused significant change in values and temperature behavior of λ_{ab}.

III. DISCUSSION

It is now generally accepted that most of the available experimental data concerning physical properties of cuprate superconductors could be described in terms of the charge-transfer model.[30] Although this model can also explain the radiation-induced effects,[4] the alternative point of view, based on the localized carrier model, was also proposed (see Ref. 3 for details and references) in order to interpret properties of neutron-irradiated superconductors. There are at least two kinds of experiments: Hall-effect studies[3,31] and measurements of the crystal-field parameters by neutron spectroscopy[32] which seem to confirm the possibility of application of the localized carrier model to the interpretation of radiation-induced effects in $YBa_2Cu_3O_x$ superconductors.

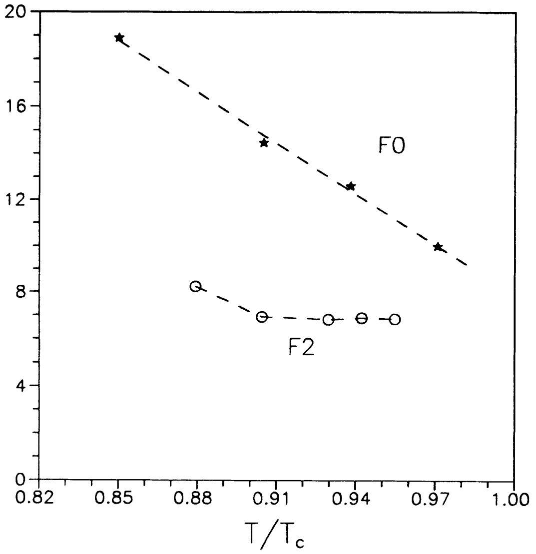


FIG. 6. The dependence of λ_{ab}^{-2} on reduced temperature T/T_c for reference sample F0 and for irradiated sample F2.

According to Ref. 31, the Hall carrier concentration, n_H, at low temperatures (about 100 K) does not change upon neutron irradiation. But it was argued in Ref. 4 that n_H was calculated from the standard formula $R_H=1/n_H ec$, which is valid only in the free-electron limit. Moreover, the free-electron formula cannot be accepted because the carrier concentration thus obtained varies strongly with temperature, in favor with the two-band model. It is suggested[33] that the radiation-induced disorder can result in a broadening of the narrow band postulated in the two-band model. In any case it is clear that the analysis of the Hall coefficient measurements cannot be done without detailed calculations of the electronic structure of the cuprate superconductors. Simple free-electron calculation of R_H makes the estimation of n_H questionable.

Neutron-scattering studies of the crystal-field splitting of the Er^{3+} $4f$ shell in the $ErBa_2Cu_3O_x$ disordered with fast neutron irradiation indicate[32] that irradiation does not affect the crystal-field parameters. This result seems to be, at first glance, in disagreement with the charge-transfer model. However, one should take into account that the crystal-field parameters are, in fact, not too sensitive to small charge redistribution. To calculate these parameters one needs to consider properly the modified charge redistribution as well as the fact that the $4f$ wave functions are also modified by this redistribution.

As shown in Ref. 34 the penetration depth for a magnetic field perpendicular to the Josephson-coupled layers in the clean limit is equal to the London penetration depth

$$\lambda_{ab}^2=\frac{mc^2}{4\pi e^2 n_s}\ , \tag{9}$$

where n_s is the carrier density in the planes. The observed increase of λ_{ab} under strong neutron irradiation could point to the decrease of the carrier density in CuO_2 planes. It may confirm the concept of charge transfer between "charge reservoir" layers and the electronically active CuO_2 layers.[30] On the other hand, in the dirty limit when $\lambda_{ab} \ll l$ (l is a carrier mean free path), the penetration depth increases in comparison to the clean limit:

$$(\lambda_{ab}^{\mathrm{dirty}})^2=\lambda_{ab}^2(1+\xi/l)\ . \tag{10}$$

Therefore the decrease of l after irradiation could also lead to an increase of λ.

The strong increase of λ_{ab} upon irradiation is also in agreement with the localization model. According to Ref. 35 the penetration depth is related to the conductivity σ:

$$\lambda^{-2}\approx\sigma/(\sigma+\sigma_c)\ , \tag{11}$$

where $\sigma_c=450\ \Omega^{-1}\mathrm{cm}^{-1}$.[36] Since σ decreases upon irradiation[32] one should expect, according to (11), a considerable increase of λ.

IV. CONCLUSIONS

We have shown that neutron irradiation changes considerably both intrinsic (T_c,λ_{ab}) and extrinsic (irreversibility line, surface barrier) properties of high-T_c superconductors. The observed increase of the lower critical field anisotropy due to irradiation has been described as a consequence of the surface barrier suppression. We have found that the flux-creep model can be used in order to explain the behavior of the irreversibility line after strong irradiation. We have suggested also that strong irradiation (with a dose of about 5×10^{18} n/cm^2) leads to the creation of weak links and a phase separation in the irradiated superconductors. To explain the defect-induced changes in intrinsic properties of superconductors we have used the model of charge transfer between CuO chains and CuO_2 planes.

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