

## Resistivity transitions in applied magnetic fields in epitaxial thin films of Fe- and Zn-doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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Epitaxial *c*-axis-oriented thin films of Fe- or Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  have been grown on (001)  $\text{LaAlO}_3$  substrates by pulsed excimer laser ablation. Adequate substitutional incorporation of Fe atoms required optimal choice of oxygen pressure during deposition as well as a postsynthesis annealing treatment at 550°C. Zn incorporation did not require such special processing conditions. The resistivity transitions for such high-quality well-characterized films were studied at different magnetic fields up to 5 T. Analysis of  $T_c$  depression and change in  $dH_{c2}/dT$  at  $T_c$  has been made in terms of the possible changes in the density of states at the Fermi level, contributions of the exchange field, magnetization field, and nonmagnetic scattering. Estimates of the activation energy [ $U_0(H)$ ] for vortex motion have also been made.

### INTRODUCTION

The systematics of resistivity transitions in applied external magnetic fields is a rich source of information about the basic aspects of superconductivity, especially the upper critical field and the vortex dynamics, in type-II superconductors.<sup>1,2</sup> The upper critical field yields an estimate of coherence length  $\xi$ , while the vortex dynamics focuses on the important issues of pinning and dimensionality. Several studies along these lines have been carried out on the different basic forms of high- $T_c$  oxide superconductors such as  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ,<sup>3,4</sup>  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,<sup>5-7</sup>  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $R = \text{Nd, Eu, Gd, Dy, Ho, Er, and Tm}$ ),<sup>8</sup>  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_{8+\delta}$ ,<sup>9,10</sup>  $\text{EuBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,<sup>11</sup>  $\text{Y}_2\text{Ba}_4\text{Cu}_8\text{O}_{16}$ ,<sup>12</sup>  $\text{Y}_{2-x}\text{Ba}_x\text{CuO}_{4-y}$ ,<sup>13</sup>  $\text{Y}_{2-x}\text{Ba}_x\text{CuO}_{y-x/2+\delta}$ ,<sup>14</sup> etc. However, their impurity-doped counterparts have not been studied so rigorously. The earlier reports on the effects of magnetic impurities on  $(dH_{c2}/dT)_{T_c}$  were by Crabree *et al.*<sup>15</sup> and Tarascon *et al.*<sup>16</sup> Crabree *et al.*<sup>15</sup> investigated the effect of replacement of La with Nd in the  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  system on the  $T_c$  as well as  $(dH_{c2}/dT)_{T_c}$ . Both these parameters showed a decrease with increasing doping concentration. By using the Werthamer, Helfand, and Hohenberg (WHH) equations these authors examined the possible contributions of the exchange field, magnetic polarization, nonmagnetic impurity scattering, and changes in the density of states at the Fermi level to the observed effects. They concluded that the observed changes can be

explained in terms of the removal of the van Hove singularity in the density of states near the Fermi level due to dopant incorporation. Tarascon *et al.*<sup>16</sup> studied the effect of Ni doping on  $(dH_{c2}/dT)_{T_c}$  in the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (polycrystalline) system. They found that Ni reduces  $T_c$  markedly whereas the value of  $(dH_{c2}/dT)_{T_c}$  remains about the same as that for undoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . More recently, Racah, Dai, and Deutscher<sup>17</sup> have discussed the comparison of  $H_{c2}$  and  $(dH_{c2}/dT)_{T_c}$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and the Pr-incorporated compound  $\text{Y}_{0.6}\text{Pr}_{0.4}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  in the oriented thin-film form. Their results and analyses reflect a more conventional three-dimensional (3D) behavior, absence of giant flux-creep effects, and considerably larger in-plane and out-of-plane coherence length in the Pr compound. They have argued that the  $4f$  wave functions of the Pr atoms lead to strong coupling between the CuO planes; thus removing the van Hove singularity responsible for the anomalously short coherence length in pure Y-Ba-Cu-O. It is clear that the attendant issues are interesting, yet open, and call for further studies. In fact, evaluation of impurity-induced perturbations on the behavior of high- $T_c$  superconductors in magnetic fields is an interesting way of understanding the basic phenomenon itself.

The foremost reason for the relatively less effort on the study of  $H_{c2}$  and vortex dynamics in doped oxide superconductors appears to be related to the problem of getting the material of high crystalline quality. The polycrystalline samples on which most of the other dopant-

related studies have been performed are not very suitable for the suggested measurements because several extraneous factors tend to obscure the main information. Growth of single crystals with the desired uniform dopant incorporation is an involved problem and very few groups have attempted such studies. Growth of single-crystalline films with incorporated dopants is a third alternative which also has not been adequately examined and exploited. In fact, to the best of our knowledge, there have been hardly any systematic studies on dopant incorporation in high- $T_c$  oxides during film growth and the corresponding process parameter optimization. Recently, we addressed this issue in the context of pulsed laser deposition of doped oxides and could achieve process parameter optimization for epitaxial growth of Fe- and Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  on (001)  $\text{LaAlO}_3$  substrates.<sup>18</sup> We have used these high-quality process-optimized thin films in our studies of resistivity transitions in applied magnetic fields reported here.

There have been several studies in the past on the effect of different magnetic impurities on the transition temperature, oxygen stoichiometry, and structure of high- $T_c$  superconductors.<sup>19–30</sup> Amongst the different dopants used, Fe and Zn stand out for their several interesting implications. It is known that when individually doped the Fe and Zn atoms prefer to occupy the chain [Cu(1)] and the plane [Cu(2)] sites, respectively, in the Y-Ba-Cu-O matrix. Since the superconducting transport is suggested to occur primarily in the plane, it is not yet completely clear as to how a remote chain dopant, such as Fe, decreases  $T_c$ . It has been speculated that the influence of the chain dopant may operate via the bridging oxygen. It is also important to note that a diamagnetic impurity such as Zn supports an effective magnetic moment in the oxide superconducting systems via local perturbations related to the antiferromagnetic order. The effective moment of Fe, on the other hand, depends on whether it is in the high- or low-spin state as determined by the nature of oxygen bonding in its immediate neighborhood. It should be noted that the plane Cu(2) has the full complement of its oxygen neighbors while the chain Cu(1) does not. Thus the chain dopant has an opportunity to modify the local oxygen bonding; and indeed Fe does so and displays a valence of 3+. It also leads to a structural transformation from the ortho to the tetragonal structure above a certain concentration. Zn changes neither the oxygen content nor the structure in any significant way over the range of existence of the doped compound as a single-phase material. In the light of these interesting features we decided to examine and compare the cases of Fe- and Zn-doped Y-Ba-Cu-O in the context of changes in  $T_c$ ,  $(dH_{c2}/dT)_{T_c}$ , and  $U_0(H)$ , the activation energy for vortex motion.

## EXPERIMENT

In our experiments, the Fe- or Zn-doped Y-Ba-Cu-O bulk pellets prepared by standard ceramic techniques were used as target materials. High-purity powders of  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{CuO}$ ,  $\text{Fe}_2\text{O}_3$ , or  $\text{ZnO}$  (dopant) in the right proportions were thoroughly mixed and heated in air at

940°C for 24 h. Following the reaction, the powders were reground in each case, pelletized, and reheated in air at 940°C for 24 h followed by slow (1.5°C/min) cooling to room temperature. This procedure enabled us to obtain homogeneous single-phase samples as confirmed by x-ray diffraction data. Electrical properties of the doped bulk samples were measured using a standard four-probe method as well as ac susceptibility measurements. The data were found to be in agreement with those reported in the literature. Such pellets ( $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-\delta}$ ,  $M = \text{Fe, Zn}$ ) were used as targets for pulsed laser deposition (PLD). The results reported here are for the  $x = 0.2$  case. The details regarding the PLD process are described elsewhere.<sup>18</sup> Briefly, the target was mounted in a chamber capable of yielding a background pressure of  $10^{-7}$  Torr. The (001)  $\text{LaAlO}_3$  substrates were mounted at a distance of 5 cm from the target surface and were heated to a temperature of 800°C during deposition. In some cases, deposition at lower temperature was also attempted. After setting the oxygen pressure in the chamber to a desired fixed value, pulses from a KrF excimer laser (Lambda Physik model LPX 305, 248 nm, 20 ns) were fired at the target at a repetition rate of 10 Hz. The target was slowly rotated during deposition to avoid texturizing of its surface. The deposited films were examined by the techniques of ac susceptibility and four-circle x-ray diffraction (Siemens D-5000). In specific cases, the deposited films were annealed at a temperature in the range of 500°C to 650°C for 1 h in oxygen ambient at specific pressures.

The samples were patterned into  $400 \times 1600 \mu\text{m}^2$  four-probe bridge structure for transport measurements using laser patterning. Adequate care was taken to ensure good and stable electrical contacts. The dc four-probe transport method was used to measure the resistive transitions with and without the magnetic field. A Keithley 220 current source and a Keithley 182 digital voltmeter were used in the measurements. Nominally, a current of  $10 \mu\text{A}$  was used in the measurements and the shape of  $R$ - $T$  curves was found to remain unchanged with the current ranging from  $1 \mu\text{A}$  to 1 mA. The magnetic field was varied from 0 to 5 T. It was applied perpendicular to the ab plane and also perpendicular to the current, which was along the ab plane. The magnetic-field values quoted below are the nominal values with an error of less than 5%.

## RESULTS AND DISCUSSION

In Fig. 1(a) we show the dependence of  $T_c$  (peak of imaginary part of ac susceptibility) of Fe-doped films on the oxygen partial pressure during deposition. Also shown in the same figure with an arrow is the bulk  $T_c$  value (i.e., the value exhibited by the target used), which is expected to result in the film for full and homogeneous incorporation of Fe atoms at the Cu sites. The substrate temperature in all cases was 800°C. It may be first noted that when an oxygen pressure of 130 mTorr is used the Fe-doped films show a  $T_c$  of nearly 80 K. This is the pressure at which one gets high-quality undoped Y-Ba-Cu-O films with  $T_c$  of 91 K. Clearly, much of the Fe is not incorporated in the lattice substitutionally under this

growth condition. Use of a higher pressure improves the incorporation dramatically up to a pressure of about 325 mTorr bringing down the  $T_c$  to about  $58 \pm 3$  K. At still higher pressures the incorporation seems to worsen again.

It may be noted from the results of Fig. 1 that, even though the incorporation is maximum at an oxygen pressure of about 325 mTorr, it is still not complete because

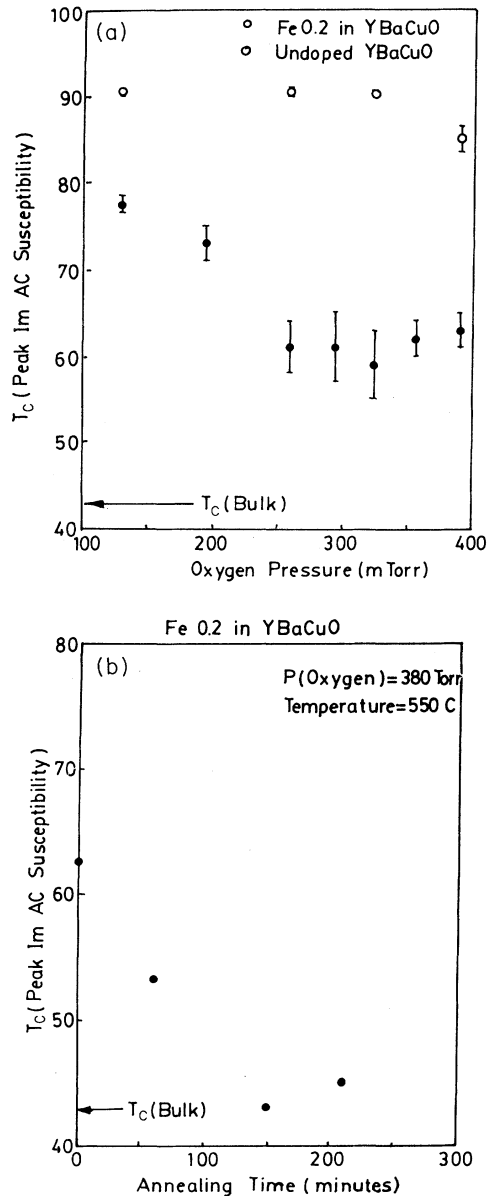


FIG. 1. (a) Dependence of  $T_c$  (peak imaginary ac susceptibility) of  $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_{7-8}$  ( $x=0.2$ ) and undoped Y-Ba-Cu-O films on the oxygen pressure during laser ablation. (b) Dependence of  $T_c$  (peak imaginary ac susceptibility) of  $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_{7-8}$  ( $x=0.2$ ) films deposited at oxygen pressure of 325 mTorr, on the duration of annealing at 550°C, at 380 Torr pressure of oxygen.

the expected  $T_c$  value of about 43 K is not reached. We attempted deposition at this pressure at different substrate temperatures, but it did not lead to any significant improvement in incorporation. Thus we attempted annealing of the deposited films. Indeed, it was observed that annealing of the Fe-doped films deposited at 325 mTorr at a temperature of 550°C under an oxygen pressure of 380 Torr for a short duration of a few hours completes the incorporation process and one achieves the desired  $T_c$  value [Fig. 1(b)]. We have observed that annealing at higher temperature of 650°C is also possible to achieve quicker incorporation. The details regarding our work on dopant incorporation are being reported separately.<sup>18</sup>

Interestingly, the difficulty in achieving homogeneous substitutional incorporation of Fe into epitaxial Y-Ba-Cu-O was not faced in the cases of Zn as a dopant. In fact, deposition at 130 mTorr oxygen with substrate temperature the same as that used for the deposition of pure Y-Ba-Cu-O yielded high-quality films with precisely the same  $T_c$  as the bulk target. We believe that these differences in dopant incorporation should depend on their surface chemistry and diffusion.<sup>18</sup> Also, the differences in the oxygen neighbors (vacancies) for chain and plane dopants could contribute to different intralayer segregation effects.

The results of the four-circle diffractometer analyses for the doped Y-Ba-Cu-O films are presented in Fig. 2.

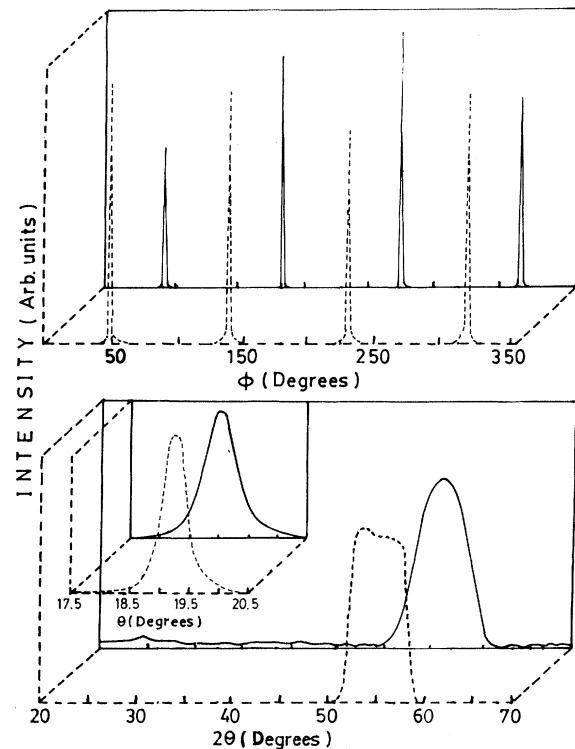


FIG. 2. Results of x-ray diffraction analysis [(a)  $\phi$  scan of (205) peak, (b)  $\chi$  scan of (102) peak, and (c) rocking curve] on  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-8}$  ( $M = \text{Fe or Zn}, x = 0.2$ ) films obtained under optimized process conditions.

The rocking-curve analysis typically shows full width at half maximum (FWHM)  $\sim 0.4$ – $0.6$  which is in the range of the values observed in the best Y-Ba-Cu-O films grown epitaxially on lattice-matched substrates. The films are thus *c*-axis oriented. Typical  $\phi$  scans for (205,102,103) sets of planes show four peaks (e.g., from 103, 301, 013, and 031 reflections) as expected for an orthorhombic *a*-*b* twinned system. The absence of any additional peaks in the  $\phi$  scan to within 2% of the main peak intensity (which is the detectability limit) indicates good in-plane alignment with the substrate *a*-*b* axes. Typical  $\chi$  scans for these films are also shown in Fig. 2. The absence of second peaks at complementary angles indicates that there is no *a*-axis-oriented fraction to within 2%. Thus the four-circle diffractometer analyses of our doped Y-Ba-Cu-O films show that these films are epitaxial, *c*-axis oriented, and are of good structural quality, comparable to the best-quality epitaxial undoped Y-Ba-Cu-O films grown on lattice-matched substrates.

In Figs. 3(a) and 3(b) are shown the resistance-temperature curves for the epitaxial Fe- and Zn-doped Y-Ba-Cu-O films in magnetic fields up to 5 T; the field being parallel to the *c* axis which is normal to the film plane. First, it may be noted that in the cases of Fe-doped film the onset temperature is in the vicinity of 70 K while that in the Zn-doped film is close to 55 K. The

90–10% transition widths are about 12 and 3.5 K in the case of Fe- and Zn-doped films, respectively. These results are in complete agreement with the data reported in the literature on these systems for the dopant concentrations used. The zero-resistance temperature is about the same (i.e.,  $\sim 45$  K) in the case of both Fe- and Zn-doped films for the studied case of  $x=0.2$  in  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-\delta}$ , in the absence of magnetic field. This, of course, is not true at all concentrations. In fact, at lower dopant concentrations the  $T_c$  reduction is much more significant in the Zn-doped system as compared to the Fe-doped one. At concentrations higher than about  $x=0.2$ ,  $T_c$  depression is more rapid in the Fe-doped case as compared to the Zn-doped one. Beyond  $x=0.3$ , the Zn-doped system does not yield a single-phase compound, hence a fruitful comparison is not possible. It has been suggested that at lower concentration all the Fe dopants occupy the Cu(1) chain site and their impact on superconductivity is less severe. As the concentration increases beyond  $x=0.2$  the concentration of Fe atoms in Cu(2) plane sites increases and  $T_c$  drops faster. Yet the characteristic differences in the nature of the transitions in zero magnetic field exhibited by Fe- and Zn-doped Y-Ba-Cu-O (which have been reported by others<sup>19–21</sup> as well) remain noteworthy and, as discussed later, probably relate to the site selectivity of dopants in the layered compound with specific functional features associated with the individual layers. We shall return to this issue in the course of discussion regarding vortex dynamics and dimensionality.

It may be observed from Figs. 3(a) and 3(b) that in the presence of a magnetic field the resistivity transition in the case of Fe-doped film broadens to a greater extent compared to that for the Zn doped film. In fact, in a magnetic field of 5 T the zero-resistance temperature for the Fe-doped film is close to 20 K as against 38 K for the Zn-doped film. In order to examine the effect of dopants on  $(dH_{c2}/dT)_{T_c}$ , the  $H$  vs  $T$  curves were plotted using the data of Fig. 3 with 90% and 50% of the resistance value at the onset (onset is defined as the lowest field-independent resistance value) as the criteria for selection of points. These curves are shown in Fig. 4. The  $(dH_{c2}/dT)_{T_c}$  with 90% criterion has values of  $-1.265$  and  $-2.546$  T/K for Fe and Zn films, respectively. The corresponding values with the 50% criterion are  $-0.452$  and  $-1.0$  T/K. Clearly, the decrease in  $(dH_{c2}/dT)_{T_c}$  is much more significant in the Fe-doped film as compared to the Zn-doped one.

Crabtree *et al.*<sup>15</sup> have analyzed the combined effect of magnetic impurity incorporation on  $T_c$  and  $(dH_{c2}/dT)_{T_c}$  within the dirty-limit pair-breaking formula of de Gennes and Maki. In this formulation, the depression of  $T_c$  is given by

$$\ln(T/T_c) = \psi(\frac{1}{2}) - \psi(\frac{1}{2}[1 + \rho T_c/T]), \quad (1)$$

where  $T_c$  is the transition temperature in the absence of any pair-breaking agent,  $T$  is the transition temperature in the presence of pair breaking,  $\rho$  is the pair-breaking parameter, and  $\psi$  is the digamma function.

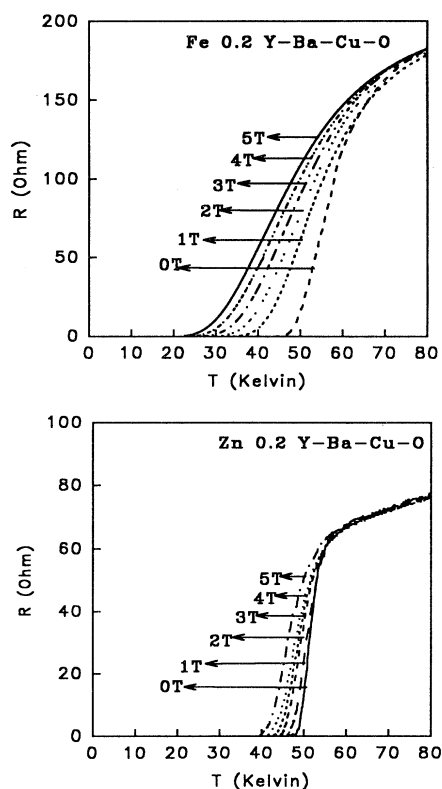


FIG. 3. Resistance vs temperature curves for  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-\delta}$  ( $M=\text{Fe}$  or  $\text{Zn}$ ,  $x=0.2$ ) films at different magnetic fields up to 5 T; the field being parallel to the *c* axis which is normal to the film plane.

The pair-breaking parameter which describes the effect of an applied field  $H$  is (in the limit of  $\lambda_{\text{SO}} \gg 1$ )

$$\rho = AH[1 + \alpha^2 AH / \lambda_{\text{SO}}] \quad (2)$$

with  $A = 2ev_f^2\tau / 6\pi k_B T_c$ ,  $\alpha = 3\hbar / 2mv_f^2\tau$ , and  $\lambda_{\text{SO}} = 2\hbar / 3\pi k_B T_c \tau_{\text{SO}}$ . Here  $\alpha$  is the Maki parameter,  $v_f$  is the Fermi velocity,  $\tau$  is the transport scattering lifetime, and  $\tau_{\text{SO}}$  is the spin-orbit scattering lifetime. The first and second terms in the above equations describe the pair breaking due to orbital motion and spin polarization of electrons, respectively, in the applied magnetic field.

The pair-breaking parameter corresponding to magnetic impurities is given by

$$\rho_{\text{AG}} = x(g-1)^2 N(0)J(J+1)\Gamma^2 / 8k_B T_{c0}, \quad (3)$$

where  $x$  is the concentration of the impurity,  $T_{c0} = T_c(x=0)$ ,  $g$  is the Landé  $g$  factor,  $N(0)$  is the conduction-electron density of states at the Fermi level,  $J$  is the total angular momentum of the magnetic impurity, and  $\Gamma$  is the exchange coupling between the local mo-

ment and the conduction electrons. The contributions to  $\rho$  in Eqs. (2) and (3) add up in Eq. (1). In the presence of magnetic impurities, the magnetizing field  $M$  of the sample contributes to  $H$  leading to the substitution of  $H + M$  in the place of  $H$  in Eq. (2). An exchange field  $H_J$  acting on the spin of the conduction electrons through its exchange coupling to the local moment also adds to  $H$ . This field is given by

$$H_J = (g-1)\Gamma M(H, T) / gg_e N\mu_B^2, \quad (4)$$

where  $N$  is the number of magnetic ions and  $g$  is the conduction-electron  $g$  factor.

The solution of Eq. (1) in the presence of all the pair-breaking mechanisms,  $H_{c2}$ , can be written in terms of the solution for the orbital critical field  $H_{c2}^*$  [solution of Eq. (1) with the contribution of the spin-orbit term only]:

$$H_{c2}(T) = H_{c2}^*(T) - 4\pi M(H_{c2}, T) - 3.56\rho_{\text{AG}}H_{c2}^*(0)$$

$$- 0.22\alpha[H_{c2}(T) + H_J(H_{c2}, T)]^2 / \lambda_{\text{SO}}T_{c0}. \quad (5)$$

All the terms in this equation are dependent on temperature except that involving  $\rho_{\text{AG}}$ . This term, thus, only lowers the critical field without altering its shape. Differentiation of Eq. (4) with respect to  $T$  gives

$$(dH_{c2}/dT)_{T_c} = (dH_{c2}^*/dT)_{T_c} [1/(1+\chi)]. \quad (6)$$

The terms in the exchange field are zero at  $T_c$  and therefore do not contribute to the initial slope. The magnetic susceptibility decreases the initial slope because the polarization of the impurity moments increases the effective field acting on the orbital motion of the electrons.  $dH_{c2}^*/dT$  as obtained from Eq. (1) with the spin-orbit contribution only, equals  $-3.81k_B / ev_f^2\tau$ .

Interestingly, the effects of various magnetic and nonmagnetic mechanisms on  $T_c$  and  $(dH_{c2}/dT)_{T_c}$  are identifiably distinct. Thus the magnetization field and nonmagnetic scattering lead to decrease and increase, respectively, of  $(dH_{c2}/dT)_{T_c}$  without affecting  $T_c$ . The exchange field, on the other hand, decreases  $T_c$  without affecting  $(dH_{c2}/dT)_{T_c}$ . The increase or decrease in the density of states at the Fermi level leads to increase or decrease of both  $T_c$  and  $(dH_{c2}/dT)_{T_c}$ . The physical reasons for these differences have been discussed in more detail by Crabtree *et al.*<sup>15</sup> In our case, both Fe and Zn dopants decrease  $T_c$  substantially; however, the decrease in  $(dH_{c2}/dT)_{T_c}$  is significantly higher in the case of Fe-doped films as compared to the Zn-doped ones. Thus the influence of Fe appears to operate via decrease in the density of states at the Fermi level while that of Zn acts via the exchange field. Notably, the Ni dopant,<sup>16</sup> which also happens to occupy the Cu(2) site in Y-Ba-Cu-O as does Zn, is also known to suppress  $T_c$  without significant change in  $(dH_{c2}/dT)_{T_c}$ . Both Ni and Zn are known to support magnetic moments in the Cu(2) substitutional site and, since these sites are in the planes which are considered responsible for primary transport of the superconducting electrons, the dominance of the exchange

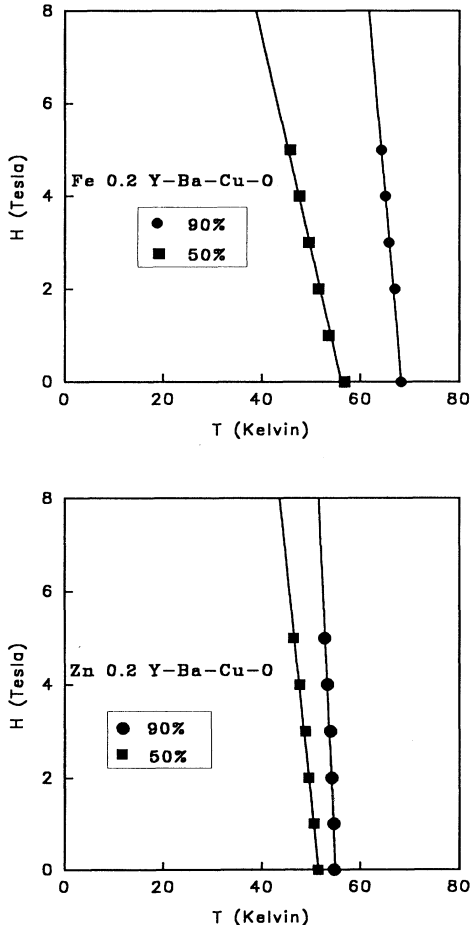


FIG. 4.  $H_c$  vs  $T$  data for  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}$  ( $M = \text{Fe}$  or  $\text{Zn}$ ,  $x = 0.2$ ) films for 90% and 50% values of the resistance at the onset.

field in the case of these dopants is understandable. Fe on the other hand is a remote dopant with reference to the CuO plane system. Also, by virtue of their tendency to acquire  $3+$  states in the Cu(1) site, the Fe atoms bring in additional oxygen atoms and thus can have a greater influence on the chain bonds, modifying thereby the band structure of the coupled chain-plane system. Zn (or Ni) does not modify oxygen stoichiometry in any notable way.

Since the present study is restricted to the measurements at fields up to 5 T only, it is clearly not appropriate to discuss the  $H_{c2}$  values obtained by extrapolation over several decades in quantitative terms. In fact, there is hardly any established picture regarding the nature of the  $H_{c2}$  vs  $T$  dependence in the case of the high- $T_c$  superconductors over the entire temperature range down to  $T=0$ . We are in the process of exploring the possibility of obtaining such information by performing studies on thin films with higher dopant concentrations which could yield substantially lower  $H_{c2}$  values at  $T=0$ , allowing the practicality of the intended measurements. Such information could allow inference about the in-plane coherence

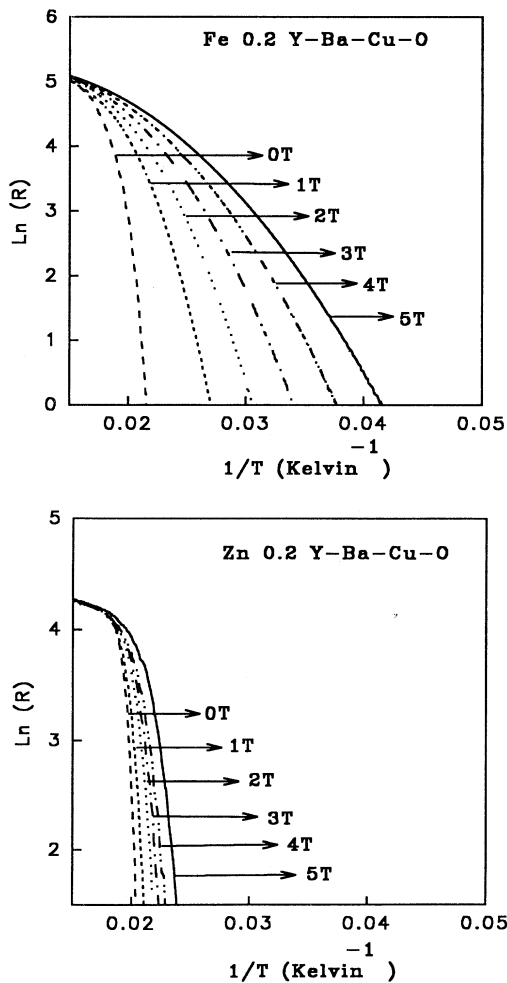


FIG. 5.  $\ln R$  vs  $1/T$  data in the transition region for  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-\delta}$  ( $M=\text{Fe}$  or  $\text{Zn}$ ,  $x=0.2$ ) films.

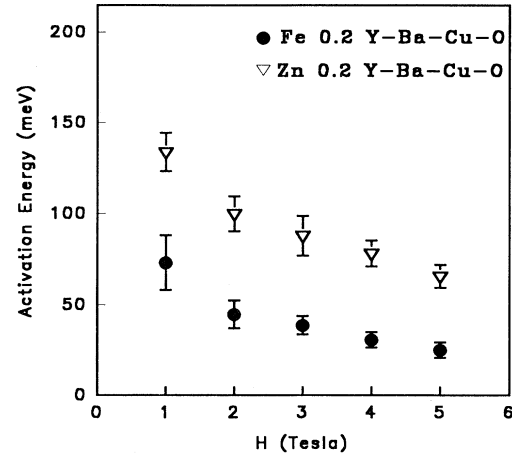


FIG. 6. Dependence of  $U_0$  (the activation energy for vortex motion) on  $H$  for  $\text{YBa}_2\text{Cu}_{3-x}\text{M}_x\text{O}_{7-\delta}$  ( $M=\text{Fe}$  or  $\text{Zn}$ ,  $x=0.2$ ) films.

length ( $\xi_{ab}$ ) in the case of the doped films.

Finally, in Fig. 5 we show the dependence of  $\ln(R)$  on  $1/T$  in the transition region for the Fe- and Zn-doped films. It can be seen that the behavior reflects the presence of activated processes, though with a distribution of activation energy values. Similar behavior has been noted by Palstra *et al.*<sup>9,10</sup> Because of the absence of a well-defined linear region<sup>12</sup> and the possibility of the presence of other physical mechanisms, it is perhaps not appropriate to attach precise quantitative significance to the values of activation energy that can be extracted from these data. Nonetheless, in order to get an idea about the qualitative differences between the dynamics of vortices in the Fe- and Zn-doped cases, we obtained the values of  $U_0$  for the two cases at different values of the magnetic field  $H$  by performing sectional linear fits over the interval from 0 to 2 of  $\ln(R)$ . The average of these values for a given value of  $H$  is shown as a point in Fig. 6, with the bar indicating the distribution. It may be seen that the values of  $U_0$  are lower at all values of the magnetic field in Fe-doped films as compared to the Zn-doped ones. The lower values of  $U_0$  reflect a weaker interplanar coupling strength, as suggested in the work of Palstra *et al.*<sup>9,10</sup> In fact, in his work on two-dimensional vortices in a stack of superconducting films Clem<sup>31</sup> has suggested that application of a magnetic field perpendicular to the layers should strongly suppress the electromagnetic coupling of the 2D pancake vortices in adjacent layers and reduce the temperature at which their decoupling occurs. Since Fe atoms are known to occupy the Cu(1) chain sites, it is possible that they reduce the order parameter in the regions between the CuO bi-planes probably leading to a 2D pancake-type vortex structure in the magnetic field. Zn being in the plane sites may not lead to a similar situation. Further studies are clearly required to sort out these interesting issues.

## CONCLUSION

The broadening of resistivity transitions in thin epitaxial  $c$ -axis-oriented films of Fe- and Zn-doped Y-Ba-Cu-O

in applied magnetic fields up to 5 T is studied. It is argued that the effects of the exchange field dominate in the case of Zn-doped films while the changes in the density of states at the Fermi level have a maximum influence on the superconductivity in the Fe-doped films. The specific locations of the dopants, viz., chain versus plane, appear to be responsible for these differences. The values of  $U_0(H)$  are also significantly lower for Fe-doped films than those for the Zn-doped films at higher fields.

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