

## Magnetic penetration depth in Ni- and Zn-doped $\text{YBa}_2(\text{Cu}_{1-x}\text{M}_x)_3\text{O}_7$ films

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The penetration depth  $\lambda(T)$  in  $\text{YBa}_2(\text{Cu}_{1-x}\text{M}_x)_3\text{O}_7$  films, with  $M=\text{Ni}$  or  $\text{Zn}$  and nominal concentrations,  $0.02 \leq x \leq 0.06$ , is obtained from the mutual inductance of coaxial coils on opposite sides of the films. Both Ni and Zn increase  $\lambda(0)$  very rapidly, such that the superfluid density,  $n_s(0) \propto \lambda^{-2}(0)$ , decreases by a factor of 2 for each percent of dopant. The rapid increase in  $\lambda(0)$  implies that disorder fills in the superconducting density of states at low energy, so that  $N_S(0)$  is roughly 80–95 % of the normal-state density of states. An analytic  $d$ -wave theory, valid at  $T=0$ , finds that  $\lambda(0)$  increases rapidly with disorder, but not as rapidly as observed. It is striking that the dependence of  $\lambda(T/T_c)$  on  $T/T_c$  does not change significantly as  $x$  increases from 2% to 6%, although it is different from undoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films. An *ad hoc* phenomenological model finds that one should expect this result. Finally, the values of  $N_S(0)$  deduced from  $\lambda$  are somewhat larger than values deduced from specific-heat measurements on Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , which also indicate increasing gaplessness with doping.

### I. INTRODUCTION

The penetration depth  $\lambda(T)$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) has attracted much interest recently. High-quality crystals show a linear  $T$  dependence,  $\lambda(T) - \lambda(0) \propto T$ , below 40 K.<sup>1</sup> A comparison of  $\lambda(T)$  between these crystals and pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films, which likely have slightly more residual disorder than crystals, finds that  $\lambda(T)$  in films is similar in many ways to  $\lambda(T)$  in crystals.<sup>2</sup> Both the slightly larger  $\lambda(0)$  and the quadratic  $T$  dependence at low  $T$  in the films can be understood semiquantitatively within a theory of modifications to an intrinsically  $d$ -wave density of states induced by weak disorder.<sup>2,3</sup> Small ( $<0.4\%$ ) concentrations of Zn substituted for Cu cause a crossover from  $T$  to  $T^2$  at low  $T$ , again in agreement with  $d$ -wave predictions, but even 0.7% Ni substituted for Cu as no apparent effect, which contradicts the theory.<sup>1</sup> Other measurements<sup>4,5</sup> also suggest  $d$ -wave superconductivity in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . It is natural to measure the evolution of  $\lambda$  with higher levels of disorder, induced by various dopants, and to compare with models of disordered  $d$ -wave superconductivity.

### II. EXPERIMENTAL DETAILS AND RESULTS

We replaced  $x = 2\%$  to  $6\%$  of the Cu atoms with Ni or Zn in  $\text{YBa}_2(\text{Cu}_{1-x}\text{M}_x)_3\text{O}_{7-\delta}$ . Since Ni and Zn substitute primarily into the  $\text{CuO}_2$  planes, this corresponds to 3 to 9% of the plane Cu atoms. Ni and Zn produce the same scattering rate, proportional to  $x$ , within the  $\pm 30\%$  uncertainty in determining  $1/\tau(x)$  from infrared reflectance measurements.<sup>6</sup> Zn suppresses  $T_c$  about three times more rapidly than Ni:  $T_c$  (4% Zn)  $\approx 45$  K while  $T_c$  (6% Ni)  $\approx 65$  K.

Films were made with two techniques to check that results were independent of fabrication method, which then

suggests that  $\lambda$  is determined by the intrinsic properties of the material rather than grain boundaries, which are very sensitive to fabrication technique. The 2% and 6% Ni-doped films were made by codepositing Y,  $\text{BaF}_2$ , Cu, and dopant onto a  $\text{SrTiO}_3$  substrate at 300 K, then postannealing at 900°C in moist oxygen.<sup>7</sup> The 4% Ni film was made by laser ablation at OSU. The 2% and 4% Zn-doped films were made by laser ablation at Los Alamos. All films had diameters of about 12 mm, they were highly oriented with the  $c$  axis perpendicular to the substrate, and  $\theta$ – $2\theta$  x-ray measurements showed no second phases. Some film properties are given in Table I. Dopant concentrations are nominal values determined from the relative deposition rates of Cu and dopant or from the nominal concentration of dopant in the laser ablation target. Film resistivities grow smoothly with dopant;  $\rho(T, x)$  data for the Zn-doped films agree quite well with twinned Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals.<sup>7</sup> Transition temperatures agree with literature values for bulk materials with the same concentrations. Transition widths, estimated from  $\lambda(T)$ , are 1–1.5 K. These widths are always much smaller than  $T_c$ , so the films are quite homogeneous from this standpoint. A detailed examination of the fluctuations responsible for the transition width is beyond the scope of this paper. Note that an inhomogeneous doping of, for example,  $\pm 0.5\%$  in the Zn-doped films would yield a minimum 12 K range transition width, which is much larger than observed. We conclude that film quality is sufficient to justify a study of  $\lambda$ .

$\lambda(T)$  was obtained from the mutual inductance  $M(T)$  of coaxial coils on opposite sides of the superconducting films.<sup>8,9</sup>  $1/\lambda^2(T)$  vs  $T$  is shown in Fig. 1; results for 4% Zn and 6% Ni are multiplied by factors of 2. The identical cylindrical coils had diameters of 2 mm and lengths of 2–3 mm. The gap between the coils was about 1.5 mm and  $M(100 \text{ K}) \approx 20\,000$  pH. Pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films,

TABLE I. Table of  $T_c$ ,  $\lambda(0)$  and disordered  $d$ -wave parameters for the doped  $\text{YBa}_2\text{Cu}_3\text{O}_7$  films used in this study.  $1/\tau(\hbar/k\tau)$  is the scattering rate determined from infrared measurements on these same films.  $T_{c0}$  and  $\lambda_{\text{pred}}(0)$  are obtained from the KPM  $d$ -wave theory,<sup>13</sup> as described in the text.

Dopant	Method	Thickness (Å) (±10%)	$T_c$ (±0.7 K)	$\lambda(0)$ (Å)	$1/\tau$	$T_{c0}$	$\lambda_{\text{pred}}(0)$ (Å)
2% Ni	Codep.	1000	79 K	2900±300	88 K	115 K	2400
4% Ni	Laser	1000	70 K	3920±60	185 K	150 K	3000
6% Ni	Codep.	3000	68 K	12440±60	273 K	190 K	3700
2% Zn	Laser	3200	62 K	3900±400	72 K	90 K	2400
4% Zn	Laser	2300	43 K	7800±800	144 K	110 K	3000

made by codeposition and postannealing, reduce  $M$  to about 30 pH at 4 K and yield  $1/\lambda^2(0)=43\pm 10\ \mu\text{m}^{-2}$ , for comparison with data on doped films in Fig. 1. The current in the primary coil was low enough to ensure linear response of the films, except possibly within 1 K of  $T_c$ . During the course of this investigation, the apparatus was repeatedly improved. The 2% Ni film data were taken with the earliest version and are the noisiest; the 2% and 4% Zn film data were taken with the second version; and the 4% and 6% Ni films were measured with both the second and the latest version. (The 2% Ni, 2% and 4% Zn films were destroyed in the course of other investigations and could not be remeasured.) With the latest apparatus, we can determine  $\lambda(0)$  within the error bars shown in Fig. 1 for the 4% and 6% Ni films.

To deduce  $\lambda$  from  $M$ , the London equation  $\mathbf{J}_s = -\mathbf{A}/(\mu_0\lambda^2)$  and Maxwell's equation  $\nabla \times (\nabla \times \mathbf{A}) = \mu_0(\mathbf{J}_s + \mathbf{J}_p)$  were solved self-consistently for  $\mathbf{J}_s$  in the film as a function of  $\lambda$ , given the current density  $\mathbf{J}_p$  in the primary coil. Given  $\mathbf{J}_p$  and  $\mathbf{J}_s(r)$ , it is straightforward to determine the flux linking the secondary coil, and hence  $M[\lambda(T)]$ . Because the 12 mm diameter films are relatively large compared with the 2 mm diameter coils, we solve for an infinite film and subtract from  $M$  the contribution,  $M'$ , from magnetic flux that goes around the film.  $M'$  can be measured by putting an  $\approx 12$  mm diameter circle of  $\approx 0.150$  mm Pb foil in place of the sample film. Residual uncertainty in  $M'$  of  $\pm 2$  pH for the latest apparatus yields a  $\pm 0.05\ \mu\text{m}$  uncertainty in  $\lambda^2(0)/d$ , ( $d$  = film thickness). Error bars in Fig. 1 represent the uncertainty in  $\lambda^{-2}(0)$  from the uncertainty in  $M'$ ; there is an additional  $\pm 10\%$  uncertainty in  $\lambda^2(T)$  from the  $\pm 10\%$  uncertainty in  $d$ . Since  $\lambda^2(0)/d$  ranges up to several  $\mu\text{m}$ , a  $\pm 0.05\ \mu\text{m}$  uncertainty is small. In contrast, in the first and second versions of the apparatus the uncertainty in  $\lambda^2(0)/d$  was  $\pm 3\ \mu\text{m}$ , and an *ad hoc* procedure was needed to determine  $M'$ . The choice of  $M'$  determined  $\lambda(0)$ ; nearer to  $T_c$ ,  $M$  is much larger than  $M'$ , so the experimental  $\lambda(T)$  here is insensitive to the choice of  $M'$ . Since  $M'$  influences the dependence of  $\lambda$  on  $T$ , the procedure used commonly with several experimental techniques that suffer from this problem is to choose  $M'$  so that  $\lambda(T)/\lambda(0)$  is close to a target function of  $T/T_c$ . The 4% Ni and 6% Ni data, which were taken with the latest apparatus, which does not suffer from this problem, provide the target function for the earlier data. Specifically,  $1/\lambda^2(T)$  just below  $T_c$  in the 4% and 6% Ni films extrapolates to  $T=0$  at about twice the experimental value of

$1/\lambda^2(0)$ , so we chose  $M'$  for the 2% Ni, 2% Zn, and 4% Zn films to do the same.

### III. DISCUSSION

At the level of detail appropriate here, all of the  $1/\lambda^2$  curves in Fig. 1 have the same shape, namely,  $1/\lambda^2(T/T_c)$  decreases monotonically and has a negative curvature everywhere.  $1/\lambda^2$  generally flattens off as  $T/T_c \rightarrow 0$ . Doping induces no dramatic changes in the dependence of  $\lambda^2(0)/\lambda^2(T/T_c)$  on  $T/T_c$ . The magnitude of  $\lambda$  is another story. The superfluid densities at  $T=0$ ,  $n_s(0) \propto 1/\lambda^2(0)$ , for 2% Ni, 4% Ni, and 6% Ni are lower than for pure YBCO films by factors of 3, 6, and 60, respectively. For 2% and 4% Zn films,  $n_s(0)$  is lower by factors of 5 and 25, respectively. Within the  $\pm 0.005$  uncertainty in  $x$  and the uncertainties in  $\lambda^2(0)$ , these numbers represent a factor of 2 reduction in  $n_s(0)$  for each percent of dopant, for both Ni and Zn.

The strong decrease in  $n_s(0)$  with a relative minor decrease in  $T_c$  stands in stark contrast to the correlation of  $T_c$  with  $n_s(0)$  found by Uemura *et al.*<sup>10</sup> for a variety of high- $T_c$  superconductors, including Pr-doped and oxygen-depleted  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , suggesting that substitutions for Cu behave differently than Pr doping and oxygen depletion. It appears that Ni and Zn act primarily to

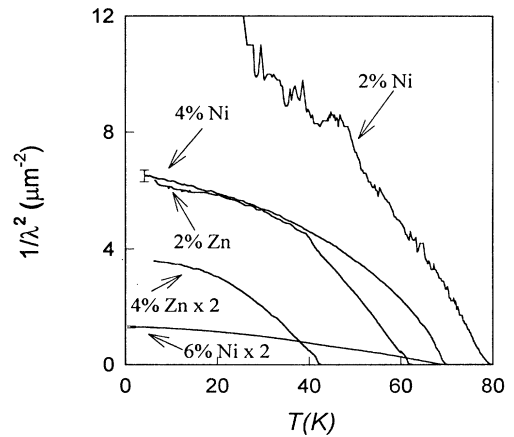


FIG. 1. Experimental  $1/\lambda^2(T)$  for Ni- and Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films. Data for the 4% Zn and 6% Ni data are multiplied by 2 for clarity. The error bars at  $T=4$  K give the uncertainty in  $1/\lambda^2(0)$  for the 4% Ni and 6% Ni films, as discussed in the text.

induce disorder, while Pr doping and O depletion act primarily to affect carrier density. Since the decrease in  $n_s$  occurs for both laser ablated and codeposited films, it seems unlikely that it is associated with film microstructure, and more likely that it is an intrinsic effect. We note that infrared measurements on irradiated crystals of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Ref. 11) and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Ref. 12) show a dramatic decrease in  $n_s(0)$  with irradiation, which generates point disorder but not grain boundaries.

Our experimental results raise numerous questions. Are the suppressions in  $n_s(0)$  and  $T_c$  accounted for by  $d$ -wave theories, given the scattering rate from IR measurements? How does the superconducting density of states,  $N_S(E)$ , evolve with doping? Are dramatic reductions in  $n_s(0)$  consistent with the relatively minor changes in the dependence of  $\lambda/\lambda(0)$  on  $T/T_c$ ?

Before getting into specific models, we want to discuss conclusions one can draw from the data, independent of model. The decrease in  $n_s(0)$  could signify changes in the density  $n$  and effective mass  $m^*$  of electrons, i.e., the plasma frequency  $\omega_p^2 = ne^2/m^*\epsilon_0$  (mks units). Since doping is less than 9% of planar Cu sites, one might expect  $\omega_p^2$  to change by no more than about 9%. Infrared measurements<sup>6</sup> on these same films support this expectation. The decrease in  $n_s(0)$  could signify that disorder pushes spectral weight [i.e., area in  $\sigma_1(\omega)$  vs  $\omega$ ] to frequencies much higher than the “gap” frequency, so there is less conductivity available at low frequencies to condense into the  $\delta$  function at  $\omega=0$ . This is the primary mechanism for reduction of  $n_s$  with disorder in  $s$ -wave models. For the most heavily doped films, 4% Zn and 6% Ni,  $1/\tau$  (Table I) is about  $4kT_c/\hbar$ , so that roughly half of the spectral weight lies above the gap frequency. This would account for a factor of about 2 reduction in  $n_s$ , which is much smaller than the observed reductions of 25 (4% Zn) and 60 (6% Ni). The reduction in  $n_s$  must be due almost entirely to modifications of the superconducting density of states. Quite rigorously, the large reduction in  $n_s$  means that the area missing from the real part of the optical conductivity,  $\sigma_{1S}(\omega)$  vs  $\omega$ , in the superconducting state is reduced by the same large factor.  $\sigma_{1S}(\omega)$  in the doped films must be very close to  $\sigma_{1N}(\omega)$ ; within uncertainty, infrared measurements find no difference at all. In other words, disorder activates photon absorption processes that are forbidden in the undoped material. The obvious physical interpretation is that disorder moves the superconducting density of states  $N_S(E)$  toward the normal-state density of states  $N_N(E)$  by filling in  $N_S$  at low energies. Then the number of quasiparticles available to absorb photons, and the density of final states for quasiparticles to scatter into are only slightly smaller than in the normal state. The precise dependence of  $N_S(E)$  on  $E$  for a particular level of disorder is model dependent, but the fact that  $N_S(E)$  is not much different from  $N_N(E)$  is not model dependent.

It is interesting to consider our results within models of disordered  $d$ -wave superconductivity.<sup>3,13</sup> In these models, the intrinsic anisotropy of the order parameter combined with strongly scattering disorder leads to creation of an “impurity band” in  $N_S$  centered at  $E=0$ , which

dramatically decreases  $n_s$  and  $T_c$ . For very small concentrations of impurities, the impurity band changes the dependence of  $\lambda$  on  $T$  from linear to quadratic below a cutoff temperature proportional to the width of the impurity band. This crossover may have been observed in Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals (but not in Ni-doped crystals),<sup>1</sup> and in very thin  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films.<sup>2</sup> Based on theory and these measurements, disorder in the present samples is much too large to expect to observe the crossover.

Kim, Preosti, and Muzikar<sup>13</sup> (KPM) present a  $d$ -wave calculation of  $n_s(0)$  and  $T_c$  which extends the Hirschfeld and Goldenfeld<sup>3</sup> theory to high concentrations of both weakly and strongly scattering impurities. The parameters of the KPM theory are the scattering rate  $1/\tau$ , the strength of scattering from each impurity which ranges from “Born” to “unitarity,” a hypothetical transition temperature  $T_{c0}$  of the “pure” material, and the  $T_c$  of the disordered material. We assume unitarity scattering in the following. With  $1/\tau$  from infrared measurements and  $T_{c0}$  fixed at 90 K, KPM predict  $T_c$  suppressions about the same as observed for 2% Zn, a little larger than for 4% Zn, and much larger than observed for Ni-doped films. Indeed, with  $T_{c0}$  fixed, KPM predict  $T_c=0$ , and therefore  $n_s=0$ , for 4% and 6% Ni. To compare KPM with  $n_s(0)$  vs  $1/\tau$ , we assume that Ni and probably Zn affect the mechanism for superconductivity and hence  $T_{c0}$ . For example, if we take  $T_{c0}$  of the 4% Zn film to be 110 K, then  $\hbar/2\tau kT_{c0}=0.65$ , and KPM predict  $T_c=0.38T_{c0}=43$  K, which matches the measured  $T_c$ . Given  $\hbar/2\tau kT_{c0}=0.65$ , KPM find  $n_s(0)$  reduced by a factor of 4, which is well below the observed reduction of 25. The assumed values of  $T_{c0}$  and the predicted values of  $\lambda(0)$  found by the same procedure are given in Table I. Overall, given our assumption about the dependence of  $T_{c0}$  on disorder, KPM underestimate the reduction in  $n_s(0)$ . The discrepancy grows from roughly a factor of 2 at 2% Zn and Ni to a factor of 6 for 4% Zn and 10 for 6% Ni. However, considering the simplicity of the theory, the extreme sensitivity to disorder and the experimental uncertainty in  $1/\tau$ , even a factor of 10 discrepancy is not too discouraging. The values of  $T_{c0}$  larger than the 90 K value for the pure material may imply that Ni (and to a lesser extent Zn) increases the effectiveness of the pairing mechanism in addition to increasing the scattering rate.

Next, we want to get an idea of how  $N_S(E)$  evolves as  $x$  increases, and to address the question of whether the  $T$  dependence of  $\lambda(T)/\lambda(0)$  is consistent with a strong filling-in of the superconducting density of states at low energies. We must construct an *ad hoc* phenomenological model which relates a disorder-broadened  $N_S(E)$  to  $\lambda(T)$ , since there is no tractable analytic theory available. We base our model on  $d$ -wave theories, largely because we can then compare our numerical results for  $\lambda(0)$  vs  $1/\tau$  with the analytic KPM theory as a check on the model.

Our *ad hoc* model begins with a generic  $d$ -wave density of states  $N_{s,p}(E)$  for pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  generated by summing all of the BCS densities of states corresponding

to the  $d_{x^2-y^2}$  gap function,  $\Delta_k = \Delta_0[\cos^2(k_x) - \cos^2(k_y)]$ , over a cylindrical Fermi surface. This density of states is a universal function of  $E/\Delta_0$ . We then find the order parameter  $\Delta_0(T)$  that results in a calculated  $\lambda_p(T)$  in perfect agreement with the data of Hardy *et al.*<sup>1</sup> on pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals, except within about 3 K of  $T_c$  where 3D-XY fluctuations affect  $\lambda(T)$ .<sup>14</sup>  $\lambda_p(T)/\lambda_p(0)$  is related to  $\Delta_0(T)$  through<sup>15</sup>

$$\begin{aligned} \lambda_p^2(0)/\lambda_p^2(T) = & 1 - (\beta/\pi)\Delta_0 \\ & \times \int_{-\infty}^{\infty} dx \operatorname{sech}^2(\beta\Delta_0 x/2) \\ & \times [\theta(x-1)K(x^{-1}) \\ & + \theta(1-x)xK(x)], \quad (1) \end{aligned}$$

where  $\theta$  is a step function,  $K$  is a complete elliptic integral, and  $\beta = 1/kT$ . The resulting  $\Delta_0(T)$  is shown in Fig. 2.  $\Delta_0(0)$  is  $(3.0 \pm 0.1)kT_c$  and  $\Delta_0(T)/\Delta_0(0)$  is similar to BCS, i.e., it is flat at low temperatures and approaches  $T_c$  as  $(2.25)(1-T/T_c)^{1/2}$ , with a coefficient only 30% larger than the BCS value of 1.74.

We assume that  $\lambda(T)$  changes with doping because  $N_S(E)$  fills in at low energies. To model changes in  $N_S(E)$  with disorder, we broaden the density of states  $N_{s,k}(E)$  corresponding to each  $\mathbf{k}$  direction with the relation,<sup>16</sup>

$$N_{s,k}(E, T) = \operatorname{Re}\{(E + i/\tau)/[(E + i/\tau)^2 - \Delta_k(T)^2]^{1/2}\}, \quad (2)$$

and then sum over all directions in  $\mathbf{k}$  space. We assume that  $\Delta_0(0)/kT_c = 3$ , even as doping suppresses  $T_c$ , and that the dependence of  $\Delta_0(T)/\Delta_0(0)$  on  $T/T_c$  is also unaffected by doping. This broadening algorithm has the important property that it conserves states. It also has the undesirable property that the peak in  $N_S(E)$  occurs at  $E > \Delta_0$  when disorder is present. Therefore, after calculating  $N_S(E)$  with a particular disorder parameter,  $\hbar/\tau\Delta_0$ , we rescale the energy axis so that the peak in  $N_S(E, T)$  occurs at  $\Delta_0(T)$ . The densities of states at  $T=0$  corresponding to the various samples are shown in Fig. 3. As  $T$  increases,  $\Delta_0(T)$  decreases so disorder effects are stronger. The peak in  $N_S(E)$  drops below  $\Delta_0(0)$  and

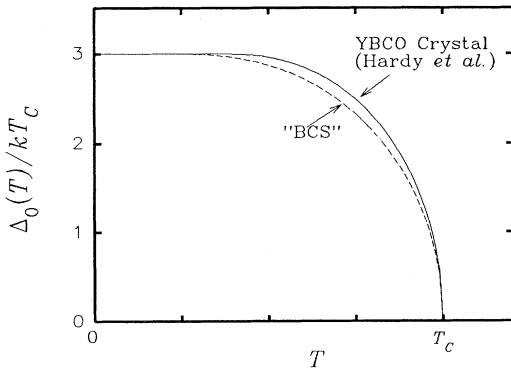


FIG. 2.  $\Delta_0(T/T_c)$  used to fit  $\lambda(T/T_c)$  for pure  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals from Hardy *et al.* (Ref. 1). The BCS temperature dependence, scaled to  $\Delta_0(0) = 3kT_c$ , is shown for comparison.

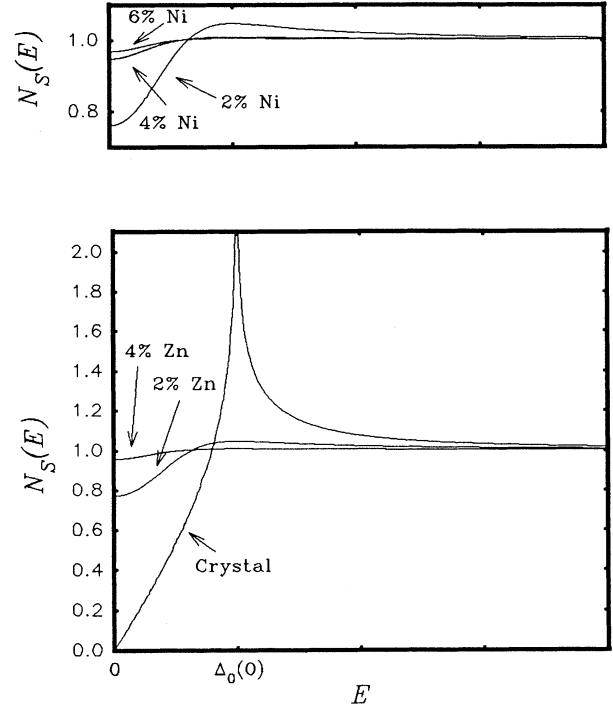


FIG. 3. Disorder-broadened  $d$ -wave densities of states deduced from our phenomenological model, with parameters corresponding to the Ni- and Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films. The pure  $d_{x^2-y^2}$  density of states used to fit the crystal data of Hardy *et al.* (Ref. 1) is shown also.

structure in  $N_S$  disappears rapidly. The rapid broadening of  $N_S$  as  $T$  increases could, in principle, lead to qualitative changes in the dependence of  $\lambda(T)$  on  $T$  as disorder increases. In the following, we find that this is not the case.

We determine  $\lambda(T)$  by calculating the area missing from  $\sigma_{1s}(\omega)$  vs  $\omega$ .  $\sigma_{1s}(\omega)$  is proportional to the rate at which photons with energy  $\hbar\omega$  are absorbed by quasiparticles. It is an integral over initial-state energy of the product of the densities of initial and final states, the appropriate Fermi functions and the Drude conductivity,  $\sigma_{1D}(\omega) = \omega_p^2 \tau \epsilon_0 / (1 + \omega^2 \tau^2)$ .<sup>6,17</sup> Coherence factors do not play a significant role in the strongly disordered state, although they do contribute to the pure (and weakly disordered<sup>3</sup>)  $d$ -wave result [Eq. (1)]. In our model

$$\begin{aligned} \frac{\lambda_0^2(0)}{\lambda^2(T)} = & \frac{2\tau}{\pi} \int_0^{\infty} \frac{d\omega}{1 + \omega^2 \tau^2} \frac{1}{\hbar\omega} \\ & \times \int_{-\infty}^{\infty} [1 - N_s(E)N_s(E + \hbar\omega)] \\ & \times [f(E) - f(E + \hbar\omega)] dE \quad (3) \end{aligned}$$

with the densities of states in Fig. 3. We fix  $\lambda_0(0) = 1500 \text{ \AA}$ , which corresponds to assuming that  $\omega_p = c/\lambda_0(0)$  is unaffected by doping.  $1/\tau$  and  $\omega_p$  are assumed independent of  $T$ . The results of the calculation are shown in Fig. 4.

We can compare the calculated increase in  $\lambda(0)$  with results found above for the KPM theory. The compar-

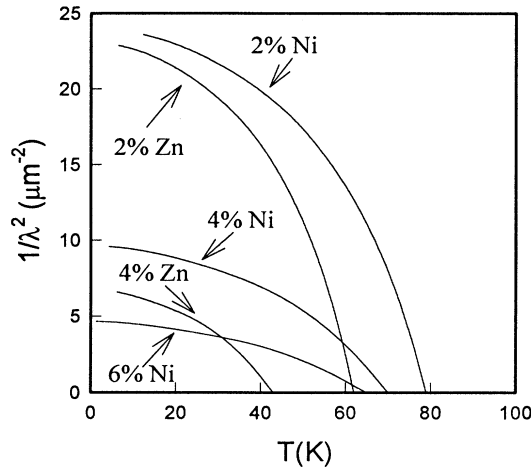


FIG. 4. Calculations of  $1/\lambda^2(T)$  from our model, Eq. (3), for various Ni and Zn concentrations, given the parameters in Table I.

ison is admittedly rough, since the KPM theory provides a wide range of predictions for  $n_s$  depending on the assumed dependence of  $T_{c0}$  on disorder, but nevertheless, for 4% Zn, for example, our calculation yields a sixfold reduction in  $n_s$  where the KPM theory as applied above, finds a fourfold reduction. Thus, our simple model is not obviously implausible. Next, we examine the calculated shape of  $\lambda^2(0)/\lambda^2(T)$  vs  $T/T_c$ , Fig. 4, and we find that it is not strongly affected by doping. To emphasize this point, Fig. 5 shows the surprisingly good agreement with the 4% Ni film, when  $1/\tau$  used in the calculation is 30% larger than the value in Table I, which is within the experimental uncertainty in  $1/\tau$ . Near  $T_c$ , the model correctly reproduces the mean-field linear  $T$  dependence. The discrepancy near  $T_c$  in Fig. 5 likely involves non-mean-field fluctuations<sup>14</sup> not included in the model. Similar agreement can be found for all except the 6% Ni sample, which requires a  $1/\tau$  much larger than found in infrared measurements. These comparisons with  $d$ -wave theories and models indicate that  $d$ -wave theories likely will be able to account for our experimental results. Of course, they do not rule out other models.

Having generated approximations to  $N_s(E=0)/N(0)$  for each doping level, we can compare with the specific heat measurements of Loram *et al.*<sup>18</sup> on Zn-doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  bulk sintered samples, which also indicate a gapless density of states. The normal-state density of states ratio,  $N_n(0)/N_{n,p}(0)$ , is about unity for Zn concentrations up to 10%, which we assumed in our model when we assumed  $\lambda_0(0)=1500 \text{ \AA}$  for all films. More importantly, 3% Zn increases  $N_s(0)$  to about 60% of  $N_n(0)$ , a significant filling in of the gap. We estimate that  $N_s(0)$  is about 80% of  $N_n(0)$  at 2% Zn. This apparently larger effect could be explained by uncertainties in the interpretations of the two experiments or by larger disorder nucleated by dopants in films, which cannot be annealed as thoroughly as bulk samples. In the end, both measurements indicate a substantial filling-in of the density of states at low energy at small doping levels.

Finally, we note that measurements on  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

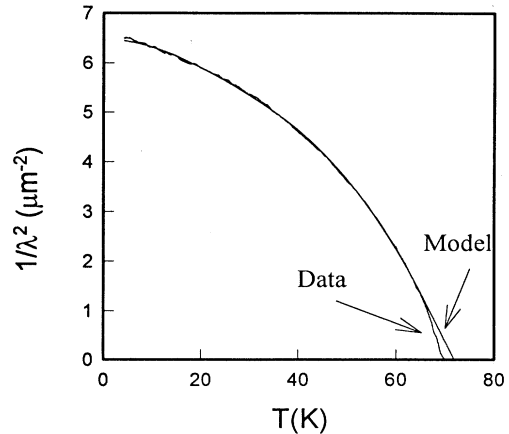


FIG. 5.  $\lambda(T)$  for the 4% Ni film and the two-parameter ( $T_c, 1/\tau$ ) fit from a disordered  $d$ -wave model showing good agreement in both the magnitude and  $T$  dependence of  $\lambda$ .  $1/\tau=240 \text{ K}$  is the best-fit scattering rate, 30% larger than the value in Table I.

crystals doped with much smaller concentrations of Ni (up to 0.75%) and Zn (up to 0.31%) find that Ni and Zn have different effects on the low-temperature behavior of  $\lambda(T)-\lambda(0)$ , at least at small concentrations.<sup>1</sup> 0.75% Ni does not significantly change the low-temperature linear term,  $\lambda(T)-\lambda(0) \propto T$ , while 0.15% Zn is sufficient to make the low-temperature term quadratic, i.e.,  $\lambda(T)-\lambda(0) \propto T^2$ . In apparent contrast, we find that Zn and Ni have similar effects. The apparent contradiction may not be real, since these authors measured the difference  $\lambda(T)-\lambda(0)$ , but not the absolute value of  $\lambda(0)$ , so a direct comparison with our results is not possible.

#### IV. CONCLUSION

We conclude that the evolution of  $\lambda$  with strong disorder is consistent with expectations for  $d$ -wave superconductors, namely, severe gaplessness at only a few percent dopant, although a realistic microscopic theory is needed. Other theories may account for the data as well. We look forward to theoretical development. The dependence of  $\lambda$  on  $T$  is well accounted for by a simple model with a disorder-broadened superconducting density of states, implying that the  $T$  dependencies of other quantities like the scattering rate, plasma frequency, etc., are of minor importance. Experimentally, we need new measurements that are sufficiently accurate to discern the differences between Ni and Zn dopants in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , and we need data on these same dopants in other cupric oxide superconductors for comparison.

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- <sup>1</sup>W. N. Hardy *et al.*, Phys. Rev. Lett. **70**, 3999 (1993); D. A. Bonn *et al.*, Phys. Rev. B **50**, 4051 (1994).
- <sup>2</sup>J. Y. Lee *et al.*, Phys. Rev. B **50**, 3337 (1994).
- <sup>3</sup>P. J. Hirschfeld and N. Goldenfeld, Phys. Rev. B **48**, 4219 (1993).
- <sup>4</sup>J. A. Martindale *et al.*, Phys. Rev. B **47**, 9155 (1993).
- <sup>5</sup>D. A. Wollman *et al.*, Phys. Rev. Lett. **71**, 2134 (1993).
- <sup>6</sup>M. J. Sumner, J.-T. Kim, and T. R. Lemberger, Phys. Rev. B **47**, 12 248 (1993); J.-T. Kim *et al.*, *ibid.* **49**, 15 970 (1994).
- <sup>7</sup>J.-T. Kim *et al.*, J. Appl. Phys. **72**, 803 (1992).
- <sup>8</sup>A. T. Fiory and A. F. Hebard, Appl. Phys. Lett. **52**, 2165 (1988).
- <sup>9</sup>B. Jeanneret *et al.*, Appl. Phys. Lett. **55**, 2336 (1989).
- <sup>10</sup>Y. J. Uemura *et al.*, Phys. Rev. Lett. **66**, 2665 (1991).
- <sup>11</sup>D. Mandrus *et al.*, Phys. Rev. Lett. **70**, 2629 (1993).
- <sup>12</sup>D. N. Basov *et al.*, Phys. Rev. B **49**, 12 165 (1994).
- <sup>13</sup>H. Kim, G. Preosti, and P. Muzikar, Phys. Rev. B **49**, 3544 (1994).
- <sup>14</sup>S. Kamal *et al.*, Phys. Rev. Lett. **73**, 1845 (1994).
- <sup>15</sup>H. Won and K. Maki, Phys. Rev. B **49**, 1397 (1994).
- <sup>16</sup>R. C. Dynes *et al.*, Phys. Rev. Lett. **53**, 2437 (1984).
- <sup>17</sup>J.-T. Kim *et al.*, J. Phys. Chem. Solids **54**, 1335 (1993).
- <sup>18</sup>J. W. Loram, K.A. Mirza, and P. F. Freeman, Physica C **171**, 243 (1990).