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Anisotropic impurity scattering effects on T_c and H_{c2} in $YBa_2Cu_3O_x$

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We have studied the impurity effects on the superconducting transition temperature T_c and the upper critical field H_{c2} in electron irradiated $YBa_2Cu_3O_y$ with in-plane oxygen defects and $YBa_2(Cu_{1-x}Zn_x)_3O_y$. It is found that the effects of the same type of defects or impurities on T_c are the same regardless of the oxygen contents of the samples. Furthermore, T_c decreases slower in irradiated $YBa_2Cu_3O_y$ than in $YBa_2(Cu_{1-x}Zn_x)_3O_y$. This may be well explained by the model that the scattering due to in-plane oxygen defects is more anisotropic than that due to Zn impurities. The different behavior of the reduced slopes $(dH_{c2}/dT)_{T_c}/(dH_{c2}/dT)_{T_{c0}}$ in these two types of samples can also be understood in this context. [S0163-1829(99)08309-5]

The symmetry of the order parameter in high-temperature superconductors (HTSC's) has been actively debated. While there are now substantial evidences supporting the anisotropic pairing symmetry with line nodes and sign changes, which is consistent with *d*-wave symmetry,[1,2] this scenario still faces challenges for quantitative reconciliation between certain experiments and the theoretical models. One of these problems is T_c suppression rate due to nonmagnetic impurities in HTSC's. In *d*-wave superconductors, nonmagnetic impurity scattering may lead to pair breaking and extreme T_c suppression.[3–6] Furthermore, assuming that the change of the carrier concentration caused by impurities is negligible, most *d*-wave pair breaking models[3–6] lead to an approximately universal dependence of T_c suppression on the residual resistivity ρ_0 independent of the types of impurities and the details of the pairing mechanism. However, the observed T_c suppression rates $dT_c/d\rho_0$ due to different types of impurities or defects are not universal,[7–10] though they all qualitatively follow the predicted Abrikosov-Gorkov function.[11] Especially, the comprehensive studies of T_c suppression due to in-plane oxygen defects in $YBa_2Cu_3O_y$ (YBCO) lead to a T_c suppression rate which is at least twice slower than the simple *d*-wave pair breaking model predicts.[7,8,12,13] Several theoretical attempts have been made to solve these puzzles, which include: the magnetic correlation destruction by Zn impurities,[14] the possible hole depletion resulting from the oxygen displacement in irradiated samples,[15] and the effects of Van Hove singularity (VHS).[9] However, there exists a simple alternative model which takes the anisotropy of impurity scattering into account.[3,10] Very recently, Won and Maki have calculated the effects of impurities on the upper critical field H_{c2} based on this model.[16] The reduced slope of H_{c2} can be expressed in an analytical formula which shares the same parameters in the calculations of T_c suppression rate. Therefore, the model of anisotropic impurity scattering can be critically examined by studying the impurity effects on both T_c and $(dH_{c2}/dT)_{T_c}$.

The in-plane oxygen defects in YBCO were produced using 80 keV electron irradiation on YBCO thin films with thickness $d=50\,\mathrm{nm}$. The high quality $c_\perp$-oriented YBCO films we used were prepared on $LaAlO_3$ using the BaF_2 (Mankiewich) process. As-prepared films had critical temperature $T_c>90\,\mathrm{K}$ determined by the midpoint of the resistive transition. Details of preparation and characterization were described in Refs. 17 and 18. Several oxygenated films were annealed in He flow at 290 to 330 °C for proper periods of time to reduce the oxygen contents. The final oxygen contents of the samples were estimated by comparing T_c, resistivity ρ, and the Hall coefficient with literature data of YBCO thin films.[17,19] Details of irradiation process were described elsewhere.[7,8,20] Single crystals $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ with nominal $x=0$, 0.017, and 0.033 were grown from flux in an Y_2O_3 crucible.[21,22] These crystals were finally annealed in O_2 flow at 400 °C for seven days to ensure full oxygenation. The polycrystalline $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ ($x=0$ to 0.06) samples were prepared by the conventional solid-state reaction method. Magnetization measurements $M(T,H)$ were performed in a Quantum Design $MPMS_2$ superconducting quantum interference device magnetometer with the applied magnetic field H up to 5 T. The resistivity ρ were measured through the four-probe method in a cryogenic system equipped with a superconducting coil. For the measurements of single crystals and thin films, the direction of the applied magnetic field was parallel to the c axis. The accuracy of the thermometer was found to be better than 0.06 K in the range of experiments.

In the model of *d*-wave pair breaking with nonmagnetic anisotropic impurity scattering,[3,10]

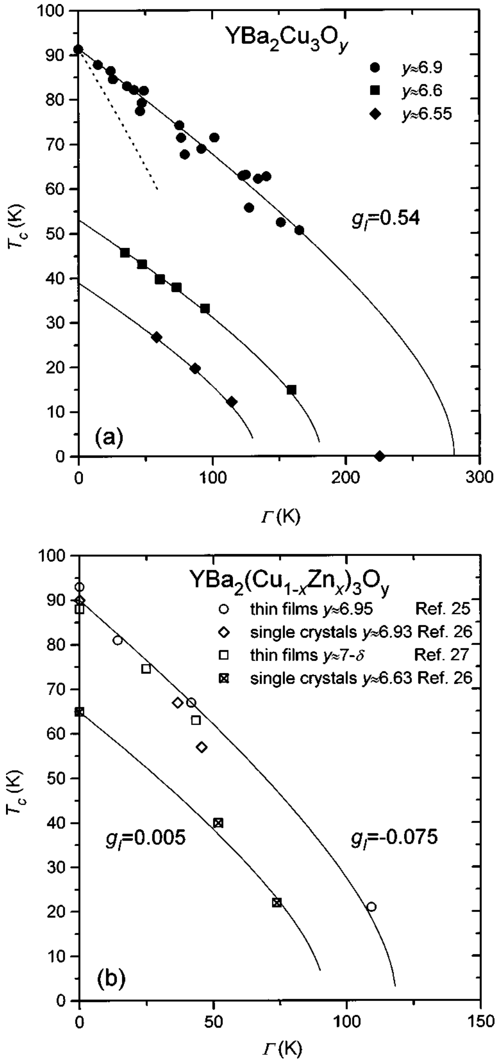

FIG. 1. (a) T_c vs $\Gamma\propto\tau_{\rm imp}^{-1}$ for irradiated YBCO. The solid lines are from Eq. (1) with $g_I=0.54$. The dotted line is with $g_I=0$. (b) T_c vs Γ for YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_y$. The solid lines are from Eq. (1) with $g_I=-0.075$ and 0.005 for oxygenated and oxygen deficient samples, respectively. $\lambda_{\rm tr}$ is fixed to be 0.2 for all lines.

$$\ln\frac{T_{c0}}{T_c}=\psi\left[\frac{1}{2}+(1-g_I)\frac{\tau_{\rm imp}^{-1}}{4\pi T_c}\right]-\psi\left(\frac{1}{2}\right), \tag{1}$$

where ψ is the digamma function, T_{c0} is the initial T_c of the sample, $\tau_{\rm imp}^{-1}$ is the isotropic component of impurity scattering rate. In this model, the impurity scattering potential is given by $|w(\mathbf{k}-\mathbf{k}')|=|w_0|^2+|w_1|^2 f(\mathbf{k})f(\mathbf{k}')$, where $|w_0|(|w_1|)$ is the isotropic (anisotropic) scattering amplitude and $f(\mathbf{k})$ is the momentum-dependent anisotropic function. $g_I=|w_1|^2/|w_0|^2$ is the ratio of anisotropic to isotropic scattering. Note that we have set $\hbar=k_B=1$. For $g_I=0$, the isotropic scattering case is reverted. In Eq. (1) the assumption is made that the anisotropic scattering is *d* wave in nature. However, it is found that even with other small components such as *s* wave, as long as *d*-wave scattering dominates, the conclusion of this paper will not be altered.

Figure 1(a) shows T_c vs Γ for electron irradiated YBCO. $\Gamma\equiv\rho_0/\alpha$ is experimentally determined from the transport data, where $\alpha\equiv d\rho/dT$ is the slope of the *T*-linear region. The quantity Γ is proportional to $\tau_{\rm imp}^{-1}$,[8,9] and it can be shown that $\Gamma=\tau_{\rm imp}^{-1}/2\pi\lambda_{\rm tr}$, where $\lambda_{\rm tr}$ is the transport coupling constant.[4,7,8] The purpose of defining Γ is to replace the plasma frequency used in Ref. 4 with $\lambda_{\rm tr}$ as the convenient fitting parameter. It turns out that the experimental data are in qualitative agreement with *d*-wave pair breaking theory. If one assumes the isotropic scattering ($g_I=0$) for in-plane oxygen defects and takes $\lambda_{\rm tr}$ as the only fitting parameter, all three solid curves in Fig. 1(a) share the same value of $\lambda_{\rm tr}$, which is consistent with the results of Ref. 23, and suggests that the T_c suppression effect of in-plane oxygen defects is the same regardless of the oxygen contents. However, under the assumption of isotropic scattering, the resultant parameter $\lambda_{\rm tr}$ is 0.092 in Fig. 1(a), much smaller than the widely believed value of 0.2 to 0.4 for $\lambda_{\rm tr}$ in YBCO.[4,24] The dotted line in Fig. 1(a) is the theoretical prediction with $g_I=0$ and $\lambda_{\rm tr}=0.2$.

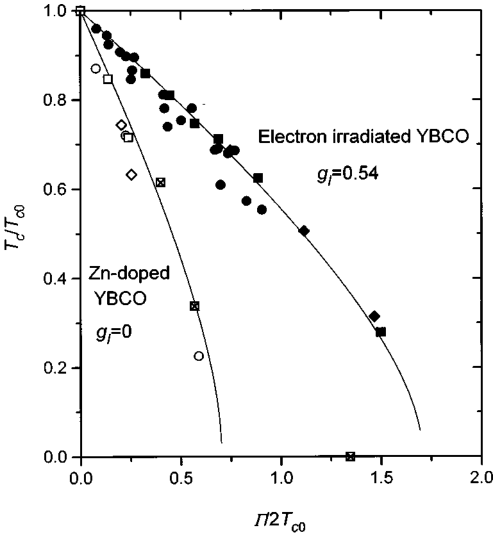

FIG. 2. T_c/T_{c0} vs $\Gamma/2T_{c0}$. The two solid lines are the results of Eq. (1) with $\lambda_{\rm tr}=0.2$ and $g_I=0.54$ and 0 for electron irradiated and Zn-doped YBCO, respectively. All data symbols follow the definitions in Fig. 1.

As for the case of YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_y$, T_c is suppressed much faster by impurity scattering. In Fig. 1(b), data of YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_y$ from Refs. 25–28 are plotted in the same way as in Fig. 1(a). In contrast to YBCO with in-plane oxygen defects, satisfactory agreements with Eq. (1) can be made by taking $\lambda_{\rm tr}=0.215$ for oxygenated samples and $\lambda_{\rm tr}=0.199$ for oxygen deficient ones. Both values fall into the acceptable range for $\lambda_{\rm tr}$.

One might interpret these experimental results as the manifestation of anisotropic impurity scattering to reconcile the difficulties of the different T_c suppression rates in irradiated YBCO and YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_y$, and the discrepancy between experiments and the theory in the case of irradiated YBCO. In this scenario, taking $\lambda_{\rm tr}=0.2$, one would obtain $g_I=0.54$ for in-plane oxygen defects in irradiated YBCO, and $g_I=-0.075$ and 0.005 for oxygenated and oxygen deficient YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_y$ (see Fig. 1). To scale the experimental results, the plot of T_c/T_{c0} vs $\Gamma/2T_{c0}$ is shown in Fig. 2 for the data in Fig. 1. As expected, all the data for the same types of impurities or defects fall onto the scaling lines. Furthermore, it is clearly seen that the points divide into two groups so that Zn impurities are really distinguished from in-plane oxygen defects. The two solid lines are the predictions from Eq. (1) with $g_I = 0.54$ and 0, respectively. The results shown in Fig. 2 suggest that impurity scattering due to in-plane oxygen defects is much more anisotropic than that due to Zn impurities, and that the same type of impurities possesses one single value of g_I regardless of oxygen contents in YBCO, as one would expect. Therefore, the puzzle of T_c suppression by impurity scattering in the d-wave scenario may be well explained in the context of anisotropic impurity scattering.

The anisotropy implied by $g_I = 0.54$ for in-plane oxygen defects is certainly unusual, and deserves further verification. Very recently, Won and Maki[16] have proposed for d-wave superconductors an expression for impurity scattering effects on the reduced slope of H_{c2} as

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c} \Bigg/ \left(\frac{dH_{c2}}{dT}\right)_{T_{c0}} = \left(\frac{T_c}{T_{c0}}\right)\frac{a(0)}{a(v)}\left[1-\frac{v}{2}\psi'\left(\frac{1}{2}+\frac{v}{2}\right)\right]. \tag{2}$$

For $g_I = 0$, $a(v) = (-1/4)\psi''(1/2 + v/2)$, and for $g_I > 0$

$$a(v) = \frac{1}{(v_0 - v)}\left\{\psi'\left(\frac{1}{2}+\frac{v}{2}\right) - \frac{2}{(v_0 - v)}\left[\psi\left(\frac{1}{2}+\frac{v_0}{2}\right) - \psi\left(\frac{1}{2}+\frac{v}{2}\right)\right]\right\}.$$

Here ψ' and ψ'' are the first and second derivative of ψ. $v_0 \equiv \tau_{\mathrm{imp}}^{-1}/2\pi T_c$ and $v \equiv (1-g_I)v_0$.

Since both Eqs. (1) and (2) contain the same parameters $\tau_{\mathrm{imp}}^{-1}(\lambda_{\mathrm{tr}})$ and g_I, it is of interest whether predictions of two different physical quantities T_c and the reduced slope are consistent with experiments *simultaneously with the same parameters* λ_{tr} *and* g_I *in both equations*. The measurements of H_{c2} near T_c for irradiated samples have been carried out. $T_c(H)$ was determined by the midpoint of the resistive transition, and $(dH_{c2}/dT)_{T_c}$ was obtained from data with $1 < H < 5.2$ T. The results are shown in Fig. 3, where the reduced slope $(dH_{c2}/dT)_{T_c}/(dH_{c2}/dT)_{T_{c0}}$ is used to compare with the theory. Similar preliminary results were reported in Ref. 29. Here only quality data from the same batch of films were presented. The data of the reduced slope from polycrystalline $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}}$ determined by the same method are also shown in Fig. 3, together with data from $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_y}$ thin film taken from Ref. 27. For the latter, $(dH_{c2}/dT)_{T_c}$ were estimated with H between 5 and 10 T parallel to the c axis. To compare with the reduced slope obtained from the resistive measurements, following Ref. 30, we have measured the dc magnetization $M(T,H)$ and used the linear temperature dependence of M near T_c to determine $T_c(H)$ [and thus $(dH_{c2}/dT)_{T_c}$] of single crystals $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}}$ with nominal $x = 0$, 0.017, and 0.033 ($T_c = 92.4$, 65.5, and 45.5 K respectively). For $x = 0$ sample, $(dH_{c2}/dT)_{T_c} = -2.0$ T/K consistent with that in Ref. 30. The reduced slopes $(dH_{c2}/dT)_{T_c}/(dH_{c2}/dT)_{T_{c0}}$ of the three single crystals are plotted in Fig. 3, too. It is shown that the reduced slopes from magnetization measurements are close to those from resistive ones. Γ/Γ_c of single crystalline and polycrystalline $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_y}$ was determined by comparing T_c with the best fit in Fig. 1(b) and Γ_c is the value of Γ with $T_c = 0$. With increasing pair breaking strength, the reduce slopes of irradiated samples decrease significantly slower than those of Zn-doped ones. In the context of anisotropic impurity scattering, there is *no fitting parameter* in Eq. (2) once λ_{tr} and g_I are determined from T_c suppression using Eq. (1). With the same parameters used in Fig. 2, the solid and dotted lines in Fig. 3 are the predictions of Eq. (2) for in-plane oxygen defects and Zn impurities, respectively. The consistency between the theory and experiments is remarkable, especially with no free parameter to adjust. There are other models for the impurity scattering effects on the reduced slope such as the Ginzburg-Landau expansion calculations[31] or the double pair breaking model.[32,33] However, neither is able to explain the results of both irradiated and Zn-doped YBCO simultaneously.

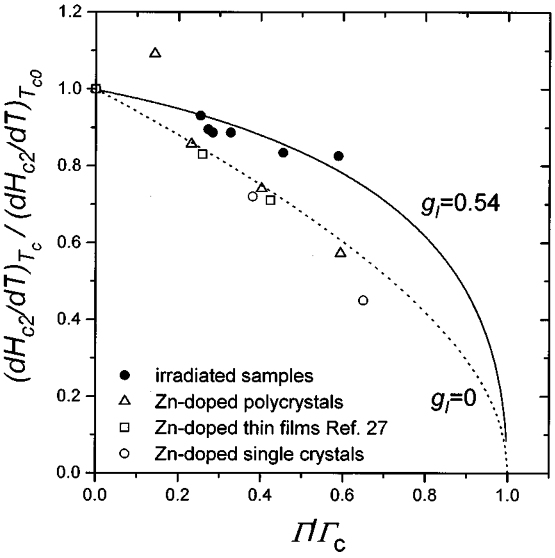

FIG. 3. The reduced slope of H_c vs Γ/Γ_c for electron irradiated and Zn-doped samples. The solid and dotted lines are the predictions of Eq. (2) with $g_I = 0.54$ and 0, respectively.

Why these two types of impurities differ so dramatically in their anisotropic character is not well understood. The effective charge Q of a impurity can be argued to be $Q \leqslant 1$ for Zn and ≈ 2 for oxygen defects.[7] According to the Friedel sum rule, it implies that s wave would dominate at scattering off Zn impurities while p- and/or d-wave scattering may dominate in the case of oxygen defects.[7] Therefore, the assumption of a large d-wave scattering component in Eq. (1) is plausible.

$(dH_{c2}/dT)_{T_c}$ of polycrystalline $\mathrm{YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}}$ determined by the resistive transition represent the value of a crystallographic average. Previously, the reduced slopes $(dH_{c2}/dT)_{T_c}/(dH_{c2}/dT)_{T_{c0}}$ of polycrystalline samples were found to be very close to those of single crystals and thin films with H parallel to the c axis in Pr-doped YBCO.[32–34] It appears to be so in Zn-doped YBCO, as the reduced slopes of both polycrystalline and thin film samples follow the same trend in Fig 3.

In summary, we have presented compelling evidences that the model of anisotropic impurity scattering in anisotropic superconductors can well describe the impurity effects on T_c suppression and the reduced slope of H_{c2} in YBCO. Our results verify a long-assumed isotropic scattering for Zn impurities in YBCO.[35] On the other hand, for in-plane oxygen defects, the effects from the anisotropic part of impurity scattering may not be negligible. Further comprehensive data of T_c suppression and the reduced slopes with other types of impurities (like Ni) would be desirable to lead to the final conclusion about this issue.

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