Nuclear quadrupole resonance study of magnetic and nonmagnetic impurities in $YBa_2Cu_4O_8$

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Cu nuclear quadrupole resonance has been used to investigate the effect of nonmagnetic Zn and magnetic Ni impurities on the normal-state properties in the underdoped $YBa_2Cu_4O_8$ superconducting cuprate. We find that, in contrast to measurements on optimally doped $YBa_2(Cu_{1-z}Zn_z)_3O_{7-\delta}$, there is no evidence of a complete local suppression of the antiferromagnetic correlations about the Zn impurity. There is also no evidence of Ni locally affecting the antiferromagnetic correlations. These results have important implications for superconductivity theories that assume a local suppression of the antiferromagnetic and superconducting correlations. [S0163-1829(99)13325-3]

INTRODUCTION

Although high temperature superconductors were first discovered over twelve years ago, controversy still exists concerning the effect of impurities in the superconducting and normal states.[1–8] An understanding of the pernicious effect of magnetic and nonmagnetic impurities is crucial for the development of theories to explain the origin of superconductivity in the high temperature superconducting cuprates.

One interpretation of the effect of nonmagnetic Zn is that Zn induces a local moment on the four nearest Cu sites. This interpretation was based on a ^{89}Y nuclear magnetic resonance (NMR) study of underdoped $YBa_2(Cu_{1-z}Zn_z)_3O_{7-\delta}$ (Ref. 9) and a muon spin rotation (μSR) study of underdoped $YBa_2(Cu_{0.96}Zn_{0.04})_3O_{7-\delta}$ samples with different δ and hence different hole concentrations, p.[2] However a recent systematic μSR study on $YBa_2(Cu_{1-z}Zn_z)_4O_8$ finds no evidence for a Zn-induced local moment over and above that which is present in the unsubstituted compound.[8]

An alternative interpretation of the effect of nonmagnetic Zn substitution is that Zn disrupts the antiferromagnetic correlations about the Zn impurity leading, within a spin fluctuation-induced-pairing model, to a reduction of the superconducting order parameter about the Zn impurity. Within this interpretation, Zn has been described as a ''superunitary'' scatterer while Ni is a Born scatterer.[5,10] This interpretation was motivated by the observation that Ni only moderately reduces T_c when substituted into $YBa_2Cu_3O_{7-\delta}$ compared with Zn substitution.[11] This picture was further supported by Ishida *et al.* who reported a nuclear quadrupole resonance (NQR) study of $YBa_2Cu_3O_7$ with Zn and Ni impurities where two ^{63}Cu spin lattice relaxation rates, $1/^{63}T$, were deduced from the $YBa_2(Cu_{1-z}Zn_z)_3O_7$ NQR data.[1] A plot of $^{63}T_1T$ from one component showed the bulk linear behavior attributed to a growing dynamical spin susceptibility about the antiferromagnetic wave vector. However, $^{63}T_1T$ from the other component was temperature independent which was attributed to a local suppression of the antiferromagnetic correlations about the Zn impurity. Only a single, $1/^{63}T_1$, was deduced from the $YBa_2(Cu_{1-z}Ni_z)_3O_7$ NQR data which was interpreted in terms of the Ni moment not disrupting the antiferromagnetic correlations. In fact the data indicated a small enhancement of the antiferromagnetic correlations by the Ni moment.

There are problems with both of the above interpretations. As already mentioned, there is no evidence from μSR measurements on $YBa_2(Cu_{1-z}Zn_z)_4O_8$ of an induced local moment.[8] Furthermore, recent μSR measurements[4] and the density-of-states-dependent depression of T_c by Zn (Ref. 12) both show that Zn is a simple unitary and not a superunitary scatterer.[4] Also, in contrast to Zn and Ni substitution in $YBa_2Cu_3O_{7-\delta}$, it is found that Zn and Ni equally suppress T_c in $YBa_2Cu_4O_8$ and $La_{2-x}Sr_xCuO_4$.[13] There is evidence that the smaller decrease in T_c for $YBa_2(Cu_{1-z}Ni_z)_3O_{7-\delta}$ when compared with $YBa_2(Cu_{1-z}Zn_z)_3O_{7-\delta}$ may originate from Ni not fully substituting onto the CuO_2 planes in $YBa_2Cu_3O_{7-\delta}$ (Refs. 11, 14, and 15) and in fact the relative degree of substitution on the chains and planes can be altered by annealing in different oxygen partial pressures.[16]

Because of the controversy surrounding the effect of magnetic and nonmagnetic impurities, we have conducted Cu NQR measurements on $YBa_2(Cu_{1-z}Zn_z)_4O_8$ and $YBa_2(Cu_{1-z}Ni_z)_4O_8$. The parent $YBa_2Cu_4O_8$ unit cell contains two CuO_2 planes and two oxygen-stoichiometric CuO chains where two-dimensional (2D) superconductivity originates on the CuO_2 planes. This compound is also underdoped ($p=0.122$ compared with $p\sim0.16$ for optimally doped $YBa_2Cu_3O_{7-\delta}$ and $p\sim0.19$ for $YBa_2Cu_3O_7$) and the NMR Knight shift is similar to underdoped $YBa_2Cu_3O_{6.8}$.[17] A significant advantage of $YBa_2Cu_4O_8$ is that it is stoichiometric and does not suffer from the disorder effects seen in $YBa_2Cu_3O_{7-\delta}$ when oxygen is removed. As a consequence, the pure $YBa_2Cu_4O_8$ compound has a narrow NQR resonance when compared with $YBa_2Cu_3O_{7-\delta}$ at a similar hole

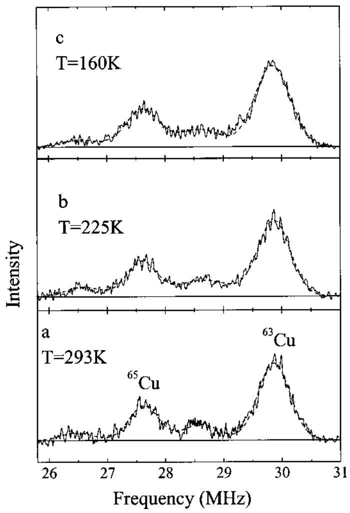

FIG. 1. Plot of the $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ ^{65}Cu NQR spectra from the CuO_2 planes at 293 K (a), 225 K (b), and 160 K (c). The dashed curves are fits to the data as described in the text.

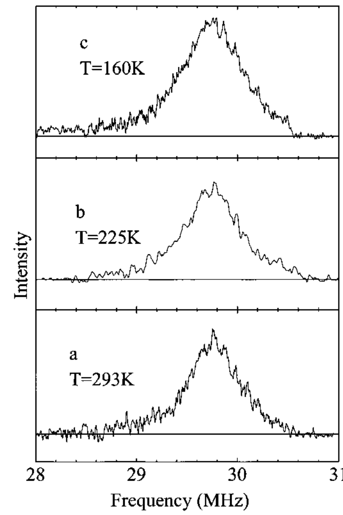

FIG. 2. Plot of the $YBa_2(Cu_{0.9813}Ni_{0.0187})_4O_8$ ^{65}Cu NQR spectra from the CuO_2 planes at 293 K (a), 225 K (b), and 160 K (c).

concentration. Unlike $YBa_2Cu_4O_8$, the $YBa_2Cu_3O_{7-\delta}$ Cu NQR spectra is complicated by a peak from Cu in the CuO chain that is close to the NQR peak from Cu in the CuO_2 planes. We show below that, while $^{63}T_1T$ is enhanced in $YBa_2(Cu_{1-z}Zn_z)_4O_8$, there is no evidence for a complete suppression of the antiferromagnetic correlations about the Zn impurity. By contrast $^{63}T_1T$ measurements on the $YBa_2(Cu_{1-z}Ni_z)_4O_8$ produce $^{63}T_1T$ values comparable to that observed in the parent compound indicating that magnetic Ni has no evident effect on the antiferromagnetic correlations.

EXPERIMENTAL DETAILS

The sample preparation and characterization of the $YBa_2(Cu_{1-z}Zn_z)_4O_8$ and $YBa_2(Cu_{1-z}Ni_z)_4O_8$ samples is described elsewhere.[13] NQR measurements were performed at temperatures from 150 to 350 K. The spectra were obtained using the 90°-τ-180° pulse sequence and the spin-lattice relaxation rates were measured using the inversion-recovery pulse sequence. The NQR spectra were obtained by scanning in discrete frequency steps and then adding the magnitude spectra.

RESULTS AND ANALYSIS

We show in Fig. 1 that there are four peaks in the $YBa_2(Cu_{1-z}Zn_z)_4O_8$ NQR spectra. Here we plot the $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ NQR spectra at (a) 293 K, (b) 225 K, and (c) 160 K. We show by the dashed curves that the NQR spectra can be fitted with four Gaussians over this frequency range. By comparison with previous studies we attribute the two main peaks at 29.86 MHz and 27.64 MHz to ^{63}Cu and ^{65}Cu in the CuO_2 planes, respectively.[18] We note that the NQR spectra from Cu in the CuO ribbons is found at a much lower frequency (~18 MHz).[18] The other two peaks may originate from Cu that are nearest neighbor to the Zn impurity. We note that satellite nearest-neighbor peaks have already been observed in ^{17}O and ^{89}Y NMR spectra from underdoped $YBa_2(Cu_{1-z}Zn_z)_3O_{7-\delta}$ (Ref. 9) and $YBa_2(Cu_{1-z}Zn_z)_4O_8$.[13] The integrated intensity of the ^{63}Cu satellite peak is consistent with this interpretation, namely that this peak arises from Cu that is nearest-neighbor to the Zn impurity. We find that the integrated intensity ratio of the ^{63}Cu satellite peak to that of both the ^{63}Cu satellite and ^{63}Cu main peak is 0.19. This is close to the expected integrated intensity of $8z(1-2z)^4=0.17$.

We show below that the lower frequency of the satellite can be interpreted in terms of charge localized on Cu nearest neighbor to the Zn impurity. We first note that the NQR frequency, v_Q, is related to the electric field gradient by

$$v_Q=\frac{eQV_{zz}}{2h}\sqrt{1+\tfrac{1}{3}\eta^2}, \tag{1}$$

where eQ is the nuclear electric quadrupole moment, V_{zz} is the maximum principle component of the electric-field-gradient tensor and η is the asymmetry parameter given by $\eta=(V_{xx}-V_{yy})/V_{zz}$ with $|V_{zz}|\geqslant|V_{yy}|\geqslant|V_{xx}|$. The asymmetry parameter is zero within error for Cu on the CuO_2 plane site in the pure compound[18] indicating that the lower v_Q cannot originate from a change in η. Consequently, the lower v_Q must originate from a reduced principle electric field gradient on the Cu site nearest neighbor to the Zn impurity. The electric field gradient is usually analyzed as originating from the sum of a valance part and a lattice part. However, in the superconducting cuprates, the lattice part is much less than the valence part and hence the electric field gradient reduces to a sum over the partially filled Cu orbitals.[19] From systematic NQR measurements on $YBa_2Cu_3O_{7-\delta}$ (Ref. 20) it is found that $dv_Q/dp=41$ MHz and hence the lower satellite NQR frequency corresponds to a change in hole concentration on Cu sites that are nearest neighbor to the Zn impurity, Δp_{nn}, of -0.030. While the origin of this localized charge is not clear, we show later that some of this localized decrease in hole concentration could be due to an increase in hole concentration from sites in the bulk that are not nearest neighbor to the Zn impurities.

It is apparent in Fig. 2 that no extra lower-frequency ^{63}Cu peak from the CuO_2 planes is observed when Ni is substituted onto the $YBa_2Cu_4O_8$ CuO_2 planes. Here we show Cu

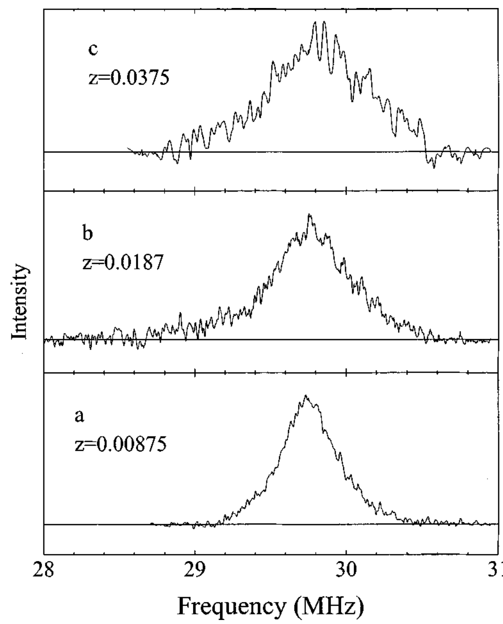

FIG. 3. Plot of the $YBa_2(Cu_{1-z}Ni_z)_4O_8$ ^{63}Cu NQR spectra from the CuO_2 planes at 293 K for $z=0.00875$ (a), $z=0.0187$ (b), and $z=0.0375$ (c).

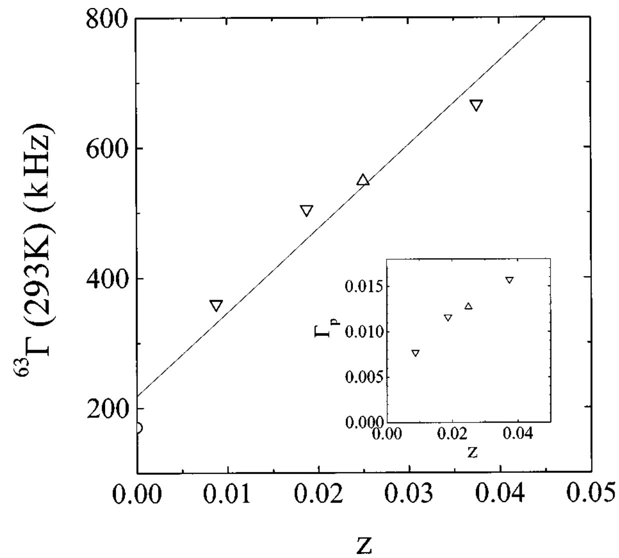

FIG. 4. Plot of the $YBa_2Cu_4O_8$ (open circle), $YBa_2(Cu_{1-z}Zn_z)_4O_8$ (open up triangles) and $YBa_2(Cu_{1-z}Ni_z)_4O_8$ (open down triangles) ^{63}Cu NQR linewidth, $^{63}\Gamma$, from the main peak at 293 K against impurity concentration z. The line is a guide to the eye. Inset: plot of the hole concentration FWHM, Γ_p, against z assuming that the increase in ^{63}Cu NQR linewidths was solely attributable to a distribution of hole concentrations.

NQR spectra from $YBa_2(Cu_{0.9813}Ni_{0.0187})_4O_8$ at (a) 293 K, (b) 225 K, and (c) 160 K. The absence of a peak near the Ni impurity is most likely due to the Ni moment locally wiping out the NQR signal. This would be consistent with our previous interpretation of ^{17}O and ^{89}Y NMR data where the Ni moment locally wipes out the signal from both ^{17}O and ^{89}Y sites.[13]

It can be seen in Fig. 3 that the $YBa_2(Cu_{1-z}Ni_z)_4O_8$ ^{63}Cu NQR spectra systematically broaden with increasing Ni content. Here we show $YBa_2(Cu_{1-z}Ni_z)_4O_8$ ^{63}Cu NQR spectra at 293 K for (a) $z=0.00875$, (b) $z=0.0187$, and (c) $z=0.0375$. This is quantified in Fig. 4 where we plot the resultant NQR linewidth against impurity concentration (open down triangles) and is consistent with the increasing Ni concentration leading to an increase in the second moment of the electric-field gradient. Also shown in Fig. 4 is the main line ^{63}Cu NQR linewidth for the $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ sample (open up triangle) where it can be seen that the Ni- and Zn-induced increase in NQR linewidths are comparable. If the increase in linewidth was purely related to either an increase in hole-concentration disorder or a charge-density oscillation about the Ni or Zn impurities then we can use the v_Q and p relation above to determine the corresponding full width at half maximum (FWHM) of the hole-concentration distribution. We show in the inset to Fig. 4 that attributing the increase in Cu NQR linewidth to a distribution of hole concentrations would require a hole concentration FWHM of less than 0.016.

Both Ni and Zn also lead to a small increase in the main-line ^{63}Cu NQR frequency as can be seen in Fig. 5 where we plot the ^{63}Cu NQR frequency for $YBa_2(Cu_{1-z}Ni_z)_4O_8$ (open up triangles) and $YBa_2(Cu_{1-z}Zn_z)_4O_8$ (open down triangles) at 293 K against impurity concentration. If the increases were due to a change in hole concentration then we can use dv_Q/dp mentioned above to deduce that, at the critical Ni and Zn impurity concentration required to reduce T_c to zero ($z=0.0313$), the corresponding increase in hole concentration on the Cu sites that are not nearest neighbor to the Zn impurity, Δp_{bulk}, is 0.0022 for Ni and 0.0043 for Zn. Using the empirical relation between T_c and p (Ref. 21) we deduce that, in the absence of Ni and Zn scattering-induced pair-breaking, these small increases in hole concentration would actually correspond to small increases in T_c of ~1.3 K for Ni and ~2.1 K for Zn. It is easy to show by charge conservation that if the increase in hole concentration for Cu sites that are not nearest neighbor to the Zn impurity is due to a localized decrease in hole concentration about the Zn impurity site then the localized charge on the Cu sites nearest neighbor to the Zn impurity can be written as $\Delta p_{nn}=-(1-2z)\Delta p_{\mathrm{bulk}}/8z$ for small z. Using $\Delta p_{\mathrm{bulk}}=0.0037$ for the $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ sample, as deduced from the change in frequency of the main Cu NQR peak, we find that, by charge conservation, $\Delta p_{nn}=-0.018$ which is less than

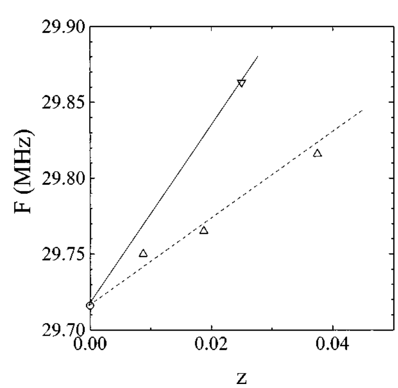

FIG. 5. Plot of the $YBa_2(Cu_{1-z}Zn_z)_4O_8$ (open up triangles) and $YBa_2(Cu_{1-z}Ni_z)_4O_8$ (open down triangles) ^{63}Cu NQR frequency at 293 K against impurity concentration z. The lines are guides to the eye.

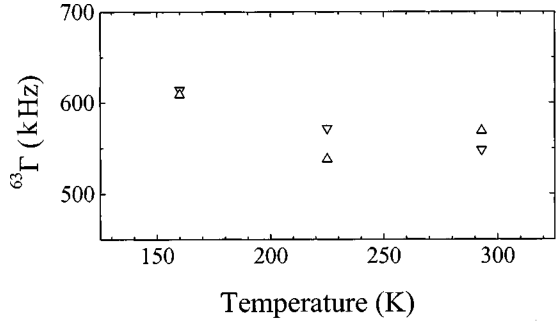

FIG. 6. Plot of the $YBa_2(Cu_{1-z}Zn_z)_4O_8$ (open up triangles) and $YBa_2(Cu_{1-z}Ni_z)_4O_8$ (open down triangles) ^{63}Cu NQR linewidths against temperature.

$\Delta p_{nn} = -0.030$ as determined from the Cu satellite peak frequency. Within this simple model it is possible that the extra Δp_{nn} of -0.012 could arise from the Zn defect level. Interestingly, a similar analysis for the Ni substituted samples yields a possible localized change in hole concentration of $\Delta p_{nn} > -0.0074$ for the 3.75% sample.

We show in Fig. 6 that the substitution of Ni and Zn results in a temperature-dependent NQR linewidth. Here we plot the $YBa_2(Cu_{0.9813}Ni_{0.0187})_4O_8$ (open up triangles) and $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ (open down triangles) ^{63}Cu NQR linewidths against temperature. Both Ni and Zn substitution result in similar small increases in NQR linewidth.

It is apparent in Fig. 7 that, unlike $YBa_2(Cu_{1-z}Zn_z)_3O_{7-\delta}$ (Ref. 1), Zn substitution in $YBa_2Cu_4O_8$ does not lead to a dramatic decrease in the antiferromagnetic correlations about the Zn impurity. Here we plot $^{63}T_1T$ from the main peak in the $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ ^{63}Cu NQR spectra (open down triangles). The ^{63}Cu NQR $^{63}T_1$ values are multiplied by 3 to correspond with $^{63}T_1$ as deduced by NMR measurements. Also shown is $^{63}T_1T$ for the pure material as reported by Curro *et al.* (open circles)[22] and Corey *et al.* (filled circles).[23] The 2.5% impurity concentration was chosen because it leads to a mean impurity spacing ($4.5a$ where a is the average lattice parameter in the ab plane) that is less than twice the antiferromagnetic correlation length [$\sim 8a$ at 150 K (Ref. 22)] and hence if Zn did locally suppress the antiferromagnetic correlations it should result in only one $^{63}T_1$ component. This removes the inherent difficulty in fitting two exponentials to the data required for low Zn concentrations where one exponential component corresponds to Cu sites within the antiferromagnetic correlation length of the Zn impurity and the other component corresponds to Cu sites further away from the Zn impurity.[1] The data in Fig. 7 can be understood by noting that $YBa_2Cu_4O_8$ $^{63}T_1T$ data have previously been interpreted in terms of a growing dynamical spin susceptibility about the antiferromagnetic wave vector, $\mathbf{Q} = (\pi, \pi)$, with decreasing temperature.[24] This interpretation can be understood by noting that $1/^{63}T_1T$ can, quite generally, be expressed as,[25]

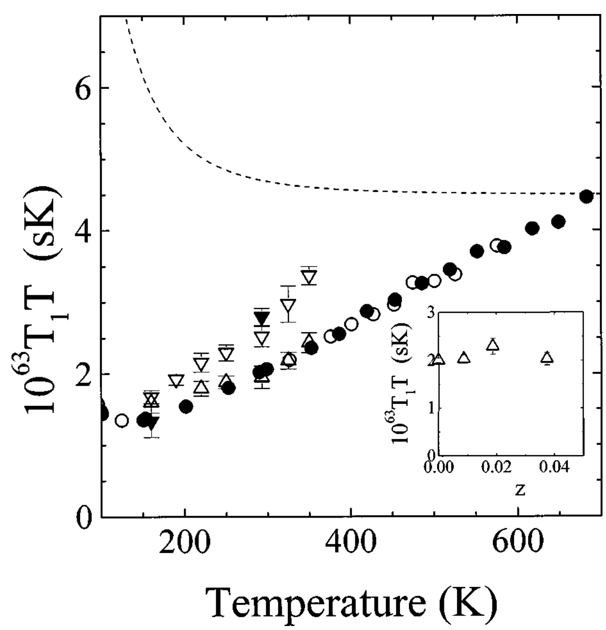

FIG. 7. Plot of $^{63}T_1T$ from the main NQR peak against temperature for $YBa_2(Cu_{0.975}Zn_{0.025})_4O_8$ (open down triangles), $YBa_2(Cu_{0.9813}Ni_{0.0187})_4O_8$ (open up triangles) and the pure compound [open circles (Ref. 22), filled circles (Ref. 23)]. Also included is $^{63}T_1T$ from the $YBa_2(Cu_{0.9813}Zn_{0.0187})_4O_8$ NQR satellite peak (filled down triangles). The dashed line is the minimum $^{63}T_1T$ expected in the absence of antiferromagnetic correlations. Inset: plot of $^{63}T_1T$ from $YBa_2(Cu_{1-z}Ni_z)_4O_8$ against impurity concentration at 293 K.

$$\frac{1}{^{63}T_1T} = \frac{k_B}{2\mu_B^2\hbar^2}\sum_q |A(\mathbf{q})|^2 \frac{\chi''(\mathbf{q},\omega)}{\omega}, \tag{2}$$

where μ_B is the Bohr magneton, $\chi''(\mathbf{q},\omega)$ is the imaginary part of the dynamical spin susceptibility and $A(\mathbf{q}) = \Sigma\, ^{63}A_j \exp(i\mathbf{q}\cdot\mathbf{r})$ is the form factor with A_j being the hyperfine coupling constants. The effect of antiferromagnetic correlations can be accounted for by the phenomenological dynamical spin susceptibility developed by Millis, Monien, and Pines,[24] $\chi(\mathbf{q},\omega)$, which can be expressed as,

$$\chi(\mathbf{q},\omega) = \frac{\chi_s(T)}{1 - i\omega/\Gamma_0} + \frac{\beta\xi^2}{[1 - (\mathbf{Q}-\mathbf{q})^2\xi^2 - i\omega/\omega_{sf}]} \tag{3}$$

where $\chi_s(T)$ is the static spin susceptibility, Γ_0 is the temperature-independent effective bandwidth, ξ is the antiferromagnetic correlation length and ω_{sf} is the paramagnon frequency. The second term, which is peaked near the antiferromagnetic wave vector, accounts for the antiferromagnetic correlations. Using Eqs. (2) and (3) and the Mila-Rice Hamiltonian[26] it can be deduced that $^{63}T_1T$ is strongly weighted by the peak in $\chi''(\mathbf{q},\omega)$ about $\mathbf{Q} = (\pi,\pi)$. Within this model, $\chi''(\mathbf{q},\omega)$ about $\mathbf{Q} = (\pi,\pi)$ increases with decreasing temperature and hence $^{63}T_1T$ decreases with decreasing temperature in unsubstituted $YBa_2Cu_4O_8$ as can be seen in Fig. 7.

In the absence of antiferromagnetic correlations, the second term in Eq. (3) is zero. Thus using Eqs. (2) and (3) and the Mila-Rice Hamiltonian it is possible to show that $^{63}T_1T$ reduces to $^{63}T_1T \propto 1/\chi_s(T)$. Consequently a lower limit for $^{63}T_1T$ in the absence of antiferromagnetic correlations can be obtained and is shown in Fig. 7 by the dashed curve. Here we use $\chi_s(T)$ as deduced from ^{89}Y NMR Knight shift measurements on $YBa_2Cu_4O_8$.[27] It can be seen in Fig. 7 that the low temperature $^{63}T_1T$ values for the Zn substituted sample are far below the lower limit expected if Zn locally suppresses the antiferromagnetic correlations indicating that, in $YBa_2Cu_4O_8$, there is no significant local suppression of the antiferromagnetic correlations by Zn impurities. This can be contrasted with ^{63}Cu NQR $^{63}T_1T$ data from $YBa_2(Cu_{1-z}Zn_z)_3O_7$ $(p\sim 0.19)$ (Ref. 1) where two $^{63}T_1T$ components were deduced from the data below 300 K leading to the interpretation of a local suppression of the antiferromagnetic correlations.

Further evidence for the absence of a local suppression of the antiferromagnetic correlations by Zn is apparent in Fig. 7 where we include $^{63}T_1T$ from the satellite peak (solid down triangles). It can be seen that $^{63}T_1T$ from Cu sites which are nearest neighbor to the Zn impurity is close to that from Cu sites which are not nearest neighbor to the Zn impurity.

Finally, we show in Fig. 7 that Ni substitution has a negligible effect on $^{63}T_1T$. Here we plot $^{63}T_1T$ data from $YBa_2(Cu_{0.9813}Ni_{0.0187})_4O_8$ (open up triangles) where it can be seen that $^{63}T_1T$ is close to that from the pure compound. We show in the inset in Fig. 7 that $^{63}T_1T$ is also independent of Ni content for Ni concentrations up to 3.75%.

CONCLUSION

In conclusion, we have shown that there is no evidence from NQR measurements on $YBa_2(Cu_{1-z}Zn_z)_4O_8$ for a local suppression of the antiferromagnetic correlations within a radius ξ from the nonmagnetic Zn impurity. This is in contrast to the interpretation of Cu NQR measurements on $YBa_2(Cu_{1-z}Zn_z)_3O_7$ where the data was analyzed in terms of a local suppression of the antiferromagnetic correlations. Furthermore, we find that the Cu sites nearest neighbor to the Zn impurity have an NQR frequency that is lower than that from Cu sites further away from the Zn impurity. We interpret this in terms of a locally reduced hole concentration. The effect of magnetic Ni in $YBa_2(Cu_{1-z}Ni_z)_4O_8$ is to increase the quadrupole broadening but $^{63}T_1T$ remains close to that of the pure compound indicating that there is no significant change in the dynamic spin susceptibility or a local suppression (or enhancement) of the antiferromagnetic correlations. These results indicate that, within the nearly antiferromagnetic Fermi liquid model, the Zn impurity in $YBa_2(Cu_{1-z}Zn_z)_4O_8$ cannot be described as a superunitary scatterer as there is no evidence of a local suppression of the antiferromagnetic correlations.

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