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# NMR and NQR studies of impurities in high-temperature superconducting cuprates

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The effect of impurities on the electronic and magnetic properties of high-temperature superconductors has proved to be highly successful in contributing to the understanding of the complex normal and superconducting states [1–4]. In particular, the substitution of non-magnetic Zn and magnetic Ni for Cu in the superconducting  $\text{CuO}_2$  planes leads to a rapid decline in  $T_c$  and localized behaviour near the impurities. However, there is no consensus concerning the mechanism for the impurity-induced suppression of superconductivity or the origin of the localized behaviour about the impurities. For this reason we have performed NMR and NQR measurements on  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_4\text{O}_8$ ,  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_4\text{O}_8$  and  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_{z/2}\text{Ni}_{z/2})_4\text{O}_8$ .

We show in the inset to Fig. 1 that non-magnetic Zn, magnetic Ni or both equally suppress  $T_c$ . Here we plot  $T_c$  against impurity fraction per Cu for  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_4\text{O}_8$  (open up triangles),  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_4\text{O}_8$  (open down triangles) and  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_{z/2}\text{Ni}_{z/2})_4\text{O}_8$  (filled circle). It can be seen that the data are well described by the Abrikosov–Gor'kov equation consistent with Ni and Zn being unitary

scatterers [2,5]. A similar equal suppression of  $T_c$  by Zn and Ni has also been observed in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . This can be contrasted with  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_3\text{O}_{7-\delta}$  and  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_3\text{O}_{7-\delta}$  where  $T_c$  is rapidly suppressed by Zn and not by Ni, which has led to Zn being interpreted in terms of superunitary scattering [6]. However, there is an evidence that in the case of  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_3\text{O}_{7-\delta}$ , Ni is not fully substituted onto the  $\text{CuO}_2$  planes [7].

The  $^{89}\text{Y}$  MAS NMR spectra from  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Zn}_{0.0175})_4\text{O}_8$  show two extra peaks which have been interpreted in terms of localized behaviour about the Zn impurity. This is apparent in Fig. 1 where we plot the  $^{89}\text{Y}$  NMR shift from the main line (open circles), first satellite (open down triangles) and second satellite (open up triangles). Also shown is the  $^{89}\text{Y}$  MAS NMR shift for pure  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (solid curve). We show by the dashed and dotted curves that the data can be fitted by the model of Mahajan et al. [8] where it is assumed that an additional Curie-like spin susceptibility arises from the four Cu sites that are nearest-neighbour to the Zn impurity. The  $^{17}\text{O}$  NMR spectra also display an evidence of localized behaviour near the Zn impurity. This can be contrasted with  $^{89}\text{Y}$  MAS NMR measurements on  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Ni}_{0.0175})_4\text{O}_8$  (filled diamonds) where we find the satellite peaks not clearly defined which may be due to the effect of the Ni magnetic

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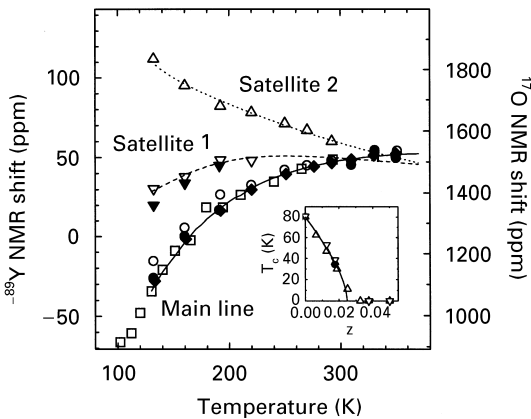


Fig. 1. Plot of the  $^{89}\text{Y}$  MAS NMR shift for the  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Zn}_{0.0175})_4\text{O}_8$  main peak (open circles), first satellite (open down triangles) and second satellite (open up triangles), the  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Ni}_{0.0175})_4\text{O}_8$  main peak (filled diamond), and the  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Zn}_{0.00875}\text{Ni}_{0.00875})_4\text{O}_8$  main peak (filled circles) and first satellite (filled down triangles). The solid curve is the  $^{89}\text{Y}$  MAS NMR shift for  $\text{YBa}_2\text{Cu}_4\text{O}_8$  and the dashed and dotted curves are for the model described in the text. Also included is the  $^{17}\text{O}$  NMR shift for  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Zn}_{0.0175})_4\text{O}_8$ . Inset:  $T_c$  against impurity fraction for  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_4\text{O}_8$  (open up triangles),  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_4\text{O}_8$  (open down triangles) and  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_{z/2}\text{Ni}_{z/2})_4\text{O}_8$  (filled circles). The solid curve is from the Abrikosov–Gorkov equation.

moment. Interestingly, the  $^{89}\text{Y}$  MAS NMR spectra from  $\text{YBa}_2(\text{Cu}_{0.9825}\text{Zn}_{0.00875}\text{Ni}_{0.00875})_4\text{O}_8$  also display additional satellite peaks where the shifts are plotted in Fig. 1 (filled circles and filled down triangles).

The Cu NQR spectra from  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_4\text{O}_8$  also display an extra peak which we interpret in terms of localized charge about the Zn impurity. A similar peak is not observed in the Cu NQR spectra from  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_4\text{O}_8$  consistent with the above interpretation where the Ni moment washes out the signal near the Ni impurity [9].

We show in Fig. 2 that there is no evidence of a local suppression of the antiferromagnetic spin fluctuations about the Zn or Ni impurities in contrast to similar measurements on  $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_3\text{O}_{7-\delta}$  [6]. Here we plot  $^{63}\text{T}_1\text{T}$ , where  $^{63}\text{T}_1$  is the  $^{63}\text{Cu}$  spin-lattice relaxation time for  $\text{YBa}_2(\text{Cu}_{0.975}\text{Zn}_{0.025})_4\text{O}_8$  (open down triangles),  $\text{YBa}_2(\text{Cu}_{0.9813}\text{Ni}_{0.0187})_4\text{O}_8$  (open up triangles) and for the  $\text{YBa}_2(\text{Cu}_{0.9813}\text{Zn}_{0.0187})_4\text{O}_8$  satellite peak (filled down triangle). It can be seen that the  $\text{YBa}_2(\text{Cu}_{0.9813}\text{Ni}_{0.0187})_4\text{O}_8$  data closely follow those

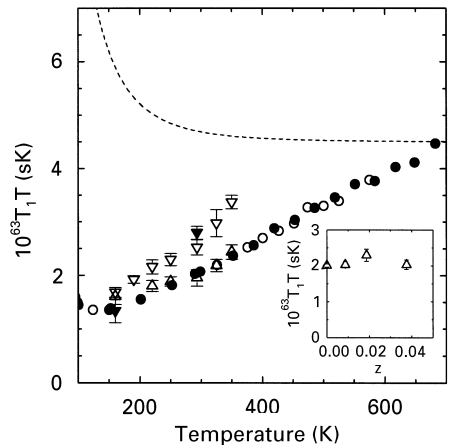


Fig. 2. Plot of  $^{63}\text{T}_1\text{T}$  from the main NQR peak against temperature for  $\text{YBa}_2(\text{Cu}_{0.975}\text{Zn}_{0.025})_4\text{O}_8$  (open down triangles),  $\text{YBa}_2(\text{Cu}_{0.9813}\text{Ni}_{0.0187})_4\text{O}_8$  (open up triangles), and the pure superconductor (open circles and filled circles [10,11]). Also included is  $^{63}\text{T}_1\text{T}$  from the  $\text{YBa}_2(\text{Cu}_{0.9813}\text{Zn}_{0.0187})_4\text{O}_8$  NQR satellite peak (filled down triangles). The dashed curve is the minimum  $^{63}\text{T}_1\text{T}$  expected in the absence of antiferromagnetic correlations. Inset: Plot of  $^{63}\text{T}_1\text{T}$  from  $\text{YBa}_2(\text{Cu}_{1-z}\text{Ni}_z)_4\text{O}_8$  against impurity fraction at 293 K.

from  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (open and filled circles). While the  $\text{YBa}_2(\text{Cu}_{0.975}\text{Zn}_{0.025})_4\text{O}_8$  data are above those for  $\text{YBa}_2\text{Cu}_4\text{O}_8$ , they are much lower than the dashed curve expected if Zn locally suppressed the antiferromagnetic correlations within an antiferromagnetic correlation length of the Zn impurity. This dashed curve gives the minimum  $^{63}\text{T}_1\text{T}$  expected in the absence of antiferromagnetic correlations.

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