Contrasting spin dynamics in Zn- and Ni-doped $NdBa_2Cu_3O_{6+y}$ single crystals from Cu nuclear quadrupole resonance: Evidence for correlations between antiferromagnetism and pseudogap effects

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We report ^{63,65}Cu nuclear quadrupole resonance (NQR) measurements on slightly underdoped $NdBa_2Cu_3O_{6+y}$ heavily doped by Ni and Zn impurities. Owing to the impurity doping, superconductivity is fully suppressed in both cases. The Ni strongly enhances magnetic correlations and induces a wipeout of the NQR signal comparable to that found in stripe ordered lanthanum cuprates. In contrast, the magnetism is suppressed in the Zn doped sample where no wipeout effect is observed and the nuclear spin relaxation rate is reduced. Our findings are in a striking correspondence with the different impact of Ni and Zn impurities on the charge pseudogap evidenced by recent optical data, uncovering thereby a close relationship between the magnetic correlations and pseudogap phenomena.

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INTRODUCTION

Magnetic (Ni) and nonmagnetic (Zn) substitution for copper in superconducting cuprates can help to provide an insight into the mechanism of high temperature superconductivity (HTSC) by studying the influence of these impurities on the electronic and magnetic properties of the CuO_2 planes. Adding a few percent of Ni or Zn is sufficient to suppress superconductivity in $La_{2-x}Sr_xCuO_4$ (LSCO).[1] Ni and Zn doping affects also the unusual normal state electronic properties of HTSCs such as the pseudogap (PG) state.[2] In particular, the observation of the magnetic moments induced by a nominally spinless Zn impurity in underdoped cuprates[3–10] has been one of the most spectacular manifestations of nontrivial correlations operating in the PG state. Recently, it was found[11] that higher amounts of Ni and Zn can be incorporated in $NdBa_2Cu_3O_{6+y}$ (NBCO), and therefore superconductivity is fully suppressed even at optimal doping. Measurements of the optical conductivity that probes the *charge* excitations in these compounds show that Ni and Zn have a profoundly different impact on the PG.[11] Large Zn doping suppresses the PG, whereas Ni enhances its energy scale. Such a drastic difference suggests that the nonmagnetic and magnetic doping may have opposite effects on the *spin* dynamics and antiferromagnetic (AF) correlations in the PG regime.

To elucidate the influence of the impurity doping on the spin dynamics, we have performed ^{63,65}Cu nuclear quadrupole resonance (NQR) measurements of single crystals of Ni and Zn doped NBCO. Since in addition to CuO_2 planes responsible for HTSC the crystal structure comprises CuO chains, we have estimated the distribution of the Ni and Zn ions between these two structural units by analyzing the Cu NQR line shape. The most striking result of our study is a Ni induced wipeout of the Cu NQR signal at low temperatures similar to that observed in stripe ordered and underdoped lanthanum cuprates.[12–16] In contrast, the Zn doping does not induce any wipeout, but a significantly larger concentration of the Zn in the planes compared to Ni. Here, the intensity, as well as the linewidth and shape of the spectra, is unchanged down to 4.2 K. Also, the spin lattice relaxation is reduced by the Zn in the whole temperature range. This difference in the spin dynamics is in a striking correspondence with the different charge dynamics measured by optical conductivity. Doping by Ni leads to a slowing of electronic spin fluctuations and a glassy magnetic order and enhances the energy scale of the charge PG. On the other hand, Zn doping does not induce such a quasistatic magnetic order, but reduces the charge PG and the spin lattice relaxation rate. Thus, our data point out an intimate interplay between the spin and charge excitations in the PG state of the cuprates and support theoretical predictions of the different effects of magnetic[17] and nonmagnetic[18,19] impurities on the properties of the strongly correlated CuO_2 planes in HTSCs.

EXPERIMENT

We used high quality single crystals of $NdBa_2(Cu,Ni,Zn)_3O_{6+y}$ with 11.5% Ni, 5% Zn, and 9.5% Zn, and a pure NBCO sample as a reference. All samples are slightly underdoped with $y \approx 0.9$. For details of the sample preparation, see Ref. 11. Midpoint T_c is 83 ± 4 K in the pure sample and 26 ± 5 K in the 5% Zn doped sample. Note that T_c in underdoped NBCO is lower than in underdoped $YBa_2Cu_3O_{6+y}$ (YBCO). Superconductivity is completely suppressed in the samples with 11.5% Ni and 9.5% Zn. A tuned NMR/NQR circuit with an almost T-independent low quality factor Q similar to that in Ref. 20 was used to minimize the influence of Q on the spectrum intensity $^{Cu}I_{NQR}$. The intensity was also corrected for the Boltzmann factor and for spin echo decay T_2. The spin lattice relaxation time T_1 was measured by inversion recovery of the longitudinal magnetization $M(t)$, which was fitted to the expression $M(t) = M(0)(1-2\exp[-(3t/T_1)^{\lambda}])$, where $\lambda \leq 1$.

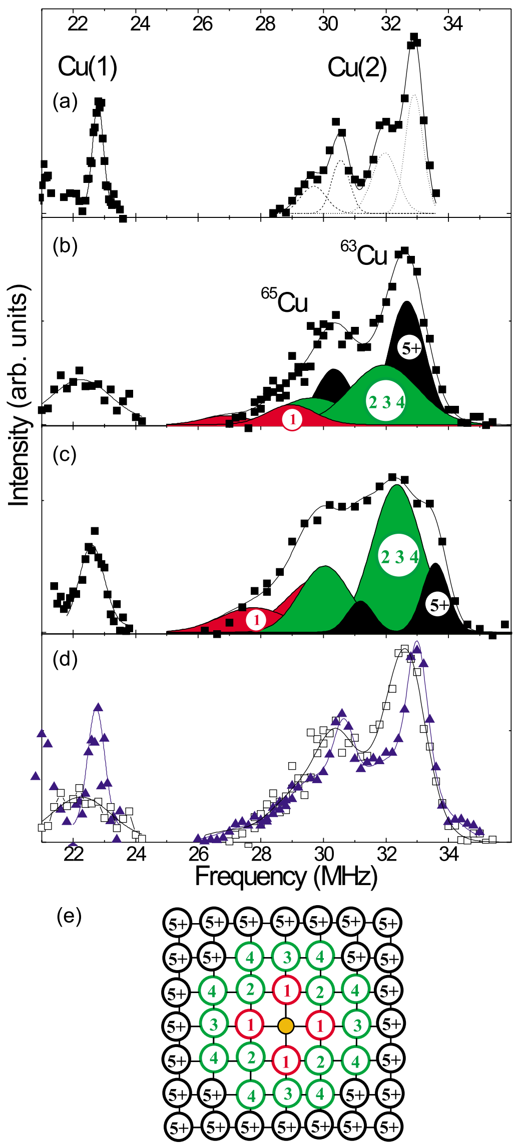

FIG. 1. (Color online) ^{63,65}Cu NQR spectra of NBCO for different dopings. Cu(1) denotes the signal from the chains, and Cu(2) the planar spectra. All spectra were taken at $T=200$ K. (a) pure NBCO, (b) 11.5% Ni doped sample, (c) 9.5% Zn doped sample, (d) comparison of the spectrum of the 11.5% Ni doped sample (□) with the spectrum of the 5% Zn doped sample (blue triangle), and (e) schematic picture of the Cu sites around an impurity site (gray circle). The fits are explained in the text.

RESULTS AND DISCUSSION

Figure 1 shows ^{63,65}Cu NQR spectra of the chains and planes of NBCO for different dopings at $T=200$ K. The resonance frequency ν_{NQR} of the planar Cu(2) in pure NBCO is blueshifted in comparison to YBCO by ~1.5 MHz owing to the larger ionic radius of the Nd compared to the Y,[21] but the linewidth and shape corresponds well to that in YBCO.[9] In contrast to the rare earth (RE) elements, Zn and Ni doping on the Cu site affects ν_{NQR} only slightly, but changes significantly the linewidth and shape. Therefore, Ni broadens the chain signal more strongly than Zn, and Zn broadens the planar signal more strongly than Ni. This difference may occur since Ni preferably occupies the chain Cu site in YBCO whereas Zn substitutes mainly for the Cu in the planes.[11] The only small change in ν_{NQR} indicates that the hole concentration of the planes does not change by Ni or Zn doping.[22] To estimate the impurity content in the CuO_2 planes, we have fitted the spectra by three lines for each Cu isotope using a model similar to that used by Itoh *et al.*[9] (see also Ref. 23). The black shaded line in Fig. 1 corresponds to those Cu sites that are fifth and farther nearest neighbors to the impurity (5+NNs).[24] The light gray (online green) shaded line originates from the Cu sites that are closer to an impurity, namely, the 4NNs to 2NNs. Such a line has also been found before in Zn doped YBCO and $YBa_2Cu_4O_8$,[7–9,25] but not for the Ni doped samples, probably due to a smaller Ni content in the planes (see below). Finally, the dark gray (red online) shaded line corresponds to the 1NNs Cu sites to the impurity. Such a line has been suspected by Itoh *et al.*[9] but could not be verified in the spectra. It has been assumed that this line as well as the missing line in case of Ni doping[7,8] are wiped out. These lines are, however, not wiped out in our spectra. We have then numerically calculated the number of particular NN Cu sites around an impurity and compared them with the intensity of the respective lines of the fits. From that, we obtain ~4% Ni and ~9.5% Zn impurities in the CuO_2 planes. Since there are two CuO_2 layers and one chain layer in the unit cell, we calculate for the chains an impurity content of 9.5% Zn and 26.5% Ni. That the Ni prefers the chain sites is evident from a much stronger broadening of the chain spectrum in this case compared to the Zn doped sample. Moreover, we compare in Fig. 1(d) a spectrum of a sample with 5% Zn with the spectrum of the Ni doped sample. Clearly, the planar Cu(2) spectra of both samples are very similar, whereas the chain Cu(1) signal of the Zn sample is only weakly broadened, which evidences a preferential planar site occupancy by Zn. Despite the controversial discussion about the Cu site assignment in the CuO_2 planes around a defect,[26–28] our data support a model that distinguishes different Cu sites close to an impurity by Cu NQR. Also, the frequency dependence of the relaxation rate T_1^{-1} supports this model (see below).

Our most obvious observation that proves the different effect of Zn and Ni doping on the electronic properties of the CuO_2 planes is a wipeout of the Cu NQR signal in the Ni doped sample below ~50 K, whereas the Cu NQR intensity $^{Cu}I_{NQR}$ of the Zn doped sample is constant down to the lowest temperature.[29] Also, with ν_{NQR}, the linewidth and shape of the spectra of the Zn doped sample exhibit almost no T dependence, whereas ν_{NQR} of the Ni sample increases when the wipeout sets in, possibly because those Cu which are close to the Ni are wiped out first. The $^{Cu}I_{NQR}(T)$ dependencies are shown in Fig. 2 and compared with those obtained for an oriented powder of $La_{1.67}Eu_{0.2}Sr_{0.13}CuO_4$ (LESCO) and a single crystal of $La_{1.875}Ba_{0.125}CuO_4$ (LBCO). The onset of the wipeout in the Ni doped NBCO occurs at a slightly lower T in comparison to LESCO and LBCO. In lanthanum cuprates, the wipeout of the Cu signal is well known, and has been interpreted as the stripe order parameter and as evidence for a glassy, inhomogeneous freezing of Cu-spin fluctuations.[12–15] However, for almost optimally doped

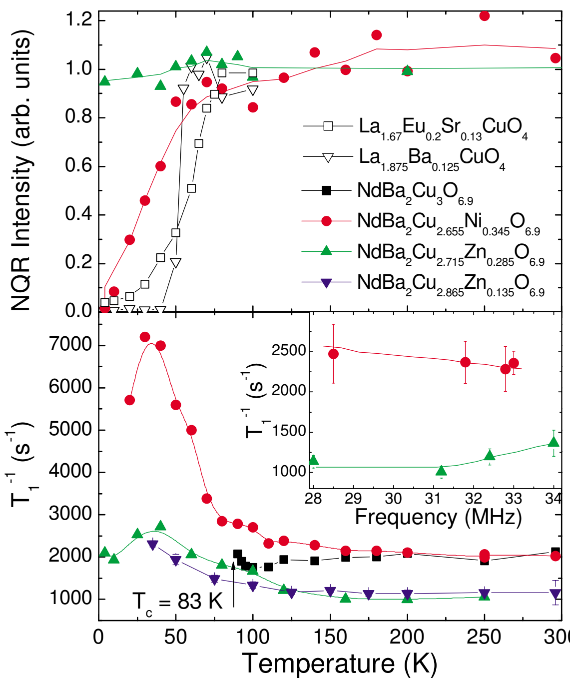

FIG. 2. (Color online) Upper panel: ${}^{Cu}I_{NQR}$ vs T in NBCO in comparison to lanthanum cuprates. Lower panel: $T_1^{-1}(T)$ dependence in NBCO. Note its strong enhancement at low Ts for the Ni, and the reduction in the whole T range for both the 5% and 9.5% Zn doped NBCO. Inset: frequency dependence of T_1^{-1} at 150 K for the 9.5% Zn and the Ni doped sample. Data at 28 and 28.5 MHz were multiplied by $({}^{63}\gamma_N/{}^{63}\gamma_N)^2$. Lines are guides to the eye.

*R*BCO (*R*=RE or Y) the situation is different. Here, a "… significant loss of the Cu NMR intensity…" has been mentioned in Ref. 30 for Zn doped YBCO, but that partial wipeout was not further quantified. This finding disagrees with our data, but to our knowledge no further wipeout of the Cu NQR intensity has been reported for almost optimally doped *R*BCO (*R*=RE or Y) since then.

In NQR, the wipeout occurs when the Cu electronic spin fluctuation frequency τ^{-1} decreases with T and approaches ν_{NQR}. At this point, the field h_0 at the nuclear site caused by the fluctuating electronic spins enhances the nuclear spin relaxation rate $T_1^{-1}=\gamma_N^2 h_0^2\tau/(1+4\pi^2\nu_{NQR}^2\tau^2)$ so that the NQR response relaxes before it can be measured.[31] Here, γ_N is the gyromagnetic ratio. The slowing down of spin fluctuations with decreasing T in Ni doped NBCO is evidenced by the strong enhancement of T_1^{-1} (Fig. 2) concomitant with the wipeout. Remarkably, Zn doping has an entirely different effect: T_1^{-1} is reduced even compared to the pure NBCO at high T and increases only slightly in the low-T regime. The latter is probably related to the fluctuating Nd moments whose susceptibility increases with decreasing temperature.[32]

In the inset of Fig. 2, the relaxation rate T_1^{-1} is plotted as a function of frequency, which in our model is related to the distance from the impurity site (see Fig. 1). For the Zn doped sample, T_1^{-1} is significantly smaller compared to the Ni doped sample everywhere in the shown frequency range. Furthermore, albeit with a large uncertainty, one may notice that in the Zn doped case T_1^{-1} tends to decrease with decreasing frequency, i.e., by approaching the impurity site, whereas it tends to increase slightly in the Ni doped case. Such a tendency further demonstrates the different effects of Ni and Zn on the spin relaxation and additionally supports the model of the site assignment in the CuO_2 planes.

The magnetization decay $M(t)$ is single exponential ($\lambda \approx 1$) at high temperatures for the Ni doped and the pure NBCO. Remarkably, we find $0.5 \leqslant \lambda \leqslant 1$ for the Zn doped sample, and below ~70 K for the Ni doped sample, implying a distribution of the rates T_1^{-1} (see Ref. 33). Such a distribution is not unexpected for magnetic defects, as has been recently analyzed in more detail.[34,35] Supporting evidence for the different spin dynamics in Ni and Zn doped NBCO comes from μSR measurements of this compound.[11] It reveals static magnetic moments on the time scale of μSR in the case of Ni doping but not for Zn doping. This is also a feature of the stripe order in lanthanum cuprates.[36] In particular, the weakening of the spin correlations by large Zn doping was found in a μSR study of LSCO.[37]

The distinction of NQR data between the Zn- and Ni-substituted NBCO samples is remarkable. Given that a *small* amount of both Zn and Ni impurities induces magnetic moments and enhances AF correlations in cuprates,[3–9] their opposite impact on low-energy spin dynamics of CuO_2 planes that we observe here is highly surprising. Unexpected suppression of AF correlations by *large* Zn doping—in contrast to the Ni case—suggests that a picture of Zn induced magnetic moments and associated enhancement of AF is no longer applicable at a high concentration of Zn impurities. The reason is that while the Ni ion introduces its own local spin $S=1$ coupled antiferromagnetically to the neighboring Cu spins, the Zn induced magnetic moment has a completely different microscopic origin.[17–19] Namely, it can be viewed as a spatially extended bound state near the Fermi energy, originating from the many-body response of a correlated Cu-spin background on a "missing-spin" defect. Because of their highly nonlocal, hence fragile, nature, low-energy resonances induced by different Zn ions should strongly overlap and may eventually disappear at larger Zn doping because of the destructive interference. In this limit, nonmagnetic Zn impurities would mainly act to suppress low-energy spin collective modes via the dilution and/or disorder effects[38] amplified at the presence of charge carriers. Apparently, this is what we see here as a decrease of NQR relaxation rates. On the contrary, Ni induced moments are robust because of their local origin and thus have a positive impact on AF at all the doping levels.[39]

It seems that there is a one-to-one correspondence between our finding that the spectral weight of low-energy magnetic excitations in heavily Ni (Zn) doped cuprates is enhanced (suppressed) and that of Ref. 11 reporting an increase (decrease) of the optical pseudogap by Ni (Zn) doping. A straightforward implication of this comparison is that quasistatic (as probed here by NQR) AF correlations are essential for the understanding of a celebrated pseudogap phenomenon in cuprates. Clearly, an enhancement of both PG and AF by Ni doping would be difficult to rationalize solely in terms of binding of spins into singlet pairs, as this would lead to a reduction of the nuclear spin lattice relaxation rate in contradiction to what is observed. Therefore, both AF correlations and pairing effects should be considered on equal footing, which remains a challenge for theory.

One may speculate that similar to the RE doped LSCO,[40] the renormalized classical regime with a reduced spin stiffness can be recovered in the Ni doped NBCO in spite of a substantial concentration of holes in the planes. In this scenario, the frequency of the spin fluctuations may decrease exponentially with decreasing temperature, causing the wipeout of the NQR signal, as found in the RE doped LSCO[12–14] and widely discussed in connection with the stripe physics, i.e., spatial modulations of the spin density in the CuO_2 planes. Structural distortions induced by RE codoping of LSCO result in the pinning of dynamic stripes and suppression of superconductivity.[40] These findings in RE doped LSCO are in an apparent similarity with our data that show a strongly enhanced nuclear relaxation rate T_1^{-1} (slow electron spin dynamics) in the heavily Ni doped NBCO. One can speculate therefore on a possible relevance of the stripe scenario to the physics of NBCO.

Regardless of a particular model, our Cu NQR results on the different effects of the Zn and Ni doping on the spin dynamics in NBCO suggest it as a generic property of the HTSC cuprates. The slowing down of the electronic spin fluctuations should have a direct implication on the dynamics in the charge sector. Indeed, the recovery of the antiferromagnetism owing to the strong interaction between the Ni impurities enhances the tendency of the hole localization and thus inhibits the charge dynamics. For this reason, the PG energy scale is pushed up to much higher energies as compared with the pure NBCO, as it has recently been observed.[11]

SUMMARY

In summary, our Cu NQR study of the heavily Ni doped NBCO reveals a complete wipeout of the NQR intensity concomitant with the strong enhancement of the nuclear relaxation rate T_1^{-1}. In a striking contrast, the Zn doping yields a reduction of T_1^{-1} and does not cause a wipeout, although the impurity content of Zn in the planes is much higher. The data enlighten an entirely different effect of the magnetic and nonmagnetic dopings on the spin dynamics and correlations in the CuO_2 planes that is in a remarkable correspondence with the different impacts of the Ni and Zn impurities on the charge excitations probed by optical measurements. In particular, the Cu NQR results provide strong experimental evidence that magnetic correlations play a vital role in the charge PG phenomenon in cuprates.

ACKNOWLEDGMENTS

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cies that were taken from Ref. 21, and then also the frequency was released. Hence, finally, the fit was completed with free unconstrained parameters.

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[32] Indeed, in heavily Zn doped *R*BCO with nonmagnetic Eu^{3+} at the *R* site, our recent T_1^{-1} data do not reveal this low-T upturn.

[33] As has been discussed, e.g., in Refs. 12 and 16, the formula expressing T_1^{-1} in terms of the electronic spin fluctuation frequency τ^{-1}, which is used in the present work, is applicable for a qualitative analysis even if there is a distribution of τ^{-1}. Such a distribution affects the width of the peak in the T dependence of T_1^{-1} but not its position.

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