PHYSICAL REVIEW B **78**, 104522 (2008)

Magnetization and Cu nuclear magnetic resonance study of $Sr_{0.9}La_{0.1}Cu_{1-x}Ni_xO_2$

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(Received 22 May 2008; published 22 September 2008)

We report the results from a Cu nuclear magnetic resonance (NMR) and magnetization study of the electron-doped high-temperature superconducting cuprate (HTSC) $Sr_{0.9}La_{0.1}Cu_{1-x}Ni_xO_2$. This compound shows a large suppression of superconductivity by Ni $[\Delta T_c/\Delta x = 18\ \text{K}/\%(\text{Ni})]$ that is comparable to that observed in underdoped and hole-doped HTSCs. We find small effective moments of $0.06\mu_B/\text{Cu}$ for the pure sample and $0.76\mu_B/\text{Ni}$ for the 1% Ni sample in the paramagnetic regime. The partial substitution of Ni for Cu causes the Cu NMR linewidth to increase and there is a further reduction in the Cu NMR intensity with decreasing temperature over and above that observed in the pure compound. This can be interpreted in terms of Ni inducing additional spin disorder and wipeout of the Cu NMR intensity for Cu sites near the Ni impurities.

DOI: 10.1103/PhysRevB.78.104522 PACS number(s): 74.72.−h, 74.25.Ha, 74.25.Nf

I. INTRODUCTION

It has recently been shown that the effect of Ni and Zn impurities on the superconducting transition temperature, T_c, in the electron-doped high-temperature superconducting cuprate (HTSC), $Sr_{0.9}La_{0.1}CuO_2$, is different from that observed in the hole-doped HTSCs, which may suggest a different superconducting order parameter.[1] In particular, T_c initially decreases by $\sim 18\ \text{K}/\%(\text{Ni})$ and it is barely affected by Zn impurities $[\Delta T_c/\Delta x \sim 1\ \text{K}/\%(\text{Zn})]$.[1] This can be contrasted with the effect of Ni and Zn on T_c in the hole-doped HTSCs, $YBa_2Cu_4O_8$,[2–5] and $La_{2-x}Sr_xCuO_4$ (Refs. 5 and 6) in which T_c is equally suppressed by Ni and Zn.[2] This has been interpreted in terms of strong scattering in the unitary limit for a superconductor with a *d*-wave superconducting order parameter.[7] In the case of another hole-doped HTSC, $YBa_2Cu_3O_{7-\delta}$, the superconducting transition temperature has been observed to decrease more rapidly for Zn substitution.[8] The initial rate of decrease in T_c by Zn in all these hole-doped HTSCs is $\sim 10\ \text{K}/\%(\text{Zn})$ on the overdoped side and rises to nearly $20\ \text{K}/\%(\text{Zn})$ on the underdoped side.[6] The increase in $\Delta T_c(p)/\Delta x$, where p is the hole concentration, for underdoped and hole-doped HTSCs has been attributed to the effect of the normal-state pseudogap in these materials.[6,9]

Zn substitution in the hole-doped HTSCs also leads to a Curie-type contribution to the susceptibility[10] and a temperature-dependent increase in the Cu nuclear magnetic resonance (NMR) linewidths, which has been interpreted in terms of a local moment (up to $\sim 1\mu_B/\text{Zn}$ where μ_B is the Bohr magneton).[11,12] It has been argued that Zn substitution in the hole-doped HTSCs leads to an induced local moment about the Zn impurity and a spin-density oscillation that is believed to account for the increase in the Cu and ^{89}Y NMR linewidths as well as the appearance of additional ^{89}Y NMR peaks with decreasing temperature.[11–14] The analysis of the Cu NMR linewidths relies on a nearly antiferromagnetic electron-spin response and hence the effect of Zn on the linewidths in this model is significantly affected by the antiferromagnetic correlation length. Ni substitution in the hole-doped HTSCs is more complex because the moment per Ni does not correspond to the value expected for a Ni impurity in the high spin $(S=1)$ state. Rather, similar to Zn in the hole-doped HTSCs, there must be considerable exchange between Ni and Cu to explain a measured moment that is as low as $0.6\mu_B/\text{Ni}$.[10]

It is currently not known how Ni impurities affect the NMR spectra in the electron-doped HTSCs. For this reason we performed magnetization and Cu NMR measurements on $Sr_{0.9}La_{0.1}CuO_2$ and $Sr_{0.9}La_{0.1}Cu_{0.99}Ni_{0.01}O_2$ and report the results in this paper. Unlike the other electron-doped HTSCs based on $R_{2-x}Ce_xCuO_4$ (R=Nd, Pr, and Sm), the constituent atoms in our unsubstituted compound ($Sr_{0.9}La_{0.1}CuO_2$) do not possess intrinsic magnetic moments that can mask the estimate of the magnetic moment created by Ni substitution. We will show below that the magnetization data can be interpreted in terms of an effective moment per Ni that is within the range of values reported in the hole-doped HTSCs and there is a temperature-dependent reduction in the ^{63}Cu NMR intensity as well an increase in the ^{63}Cu NMR linewidth.

II. EXPERIMENTAL DETAILS

The samples for this study were prepared using a high-pressure synthesis method described elsewhere.[1] All samples were *c* axis aligned in a resin using a magnetic field of 11.71 T. The alignment was confirmed by x-ray diffraction. T_c was determined using a superconducting quantum interference device (SQUID) magnetometer in an applied magnetic field of 2 mT. T_c were found to be 43 K for the pure compound and 25 K for 1% Ni substitution.[1] A full Meissner fraction was found in all samples.[1] The normal-state magnetization was measured at 1 T with a SQUID magnetometer.

The Cu NMR measurements were performed in magnetic fields of 9.2 and 11.71 T. The Cu NMR spectra were taken

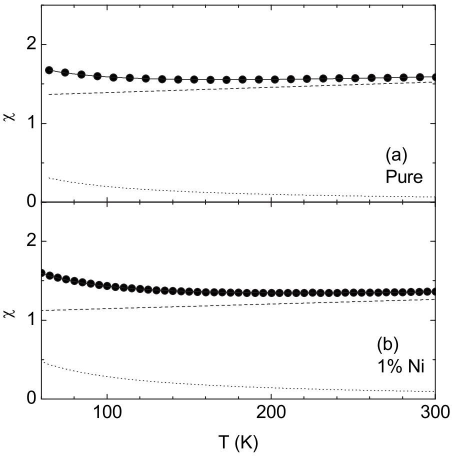

FIG. 1. Plot of χ against temperature at 1 T for polycrystalline (a) $Sr_{0.9}La_{0.1}CuO_2$ and (b) $Sr_{0.9}La_{0.1}Cu_{0.99}Ni_{0.01}O_2$. Also shown is a fit to Eq. (1) (solid curves) and the Curie (dotted curves) and linear (dashed curves) terms.

using a Hahn echo sequence at discrete frequency steps with π pulse widths from 2.8 to 13 μs and a time, τ, between the $\pi/2$ and π pulses of 12–20 μs. The echo was integrated at each frequency step to plot the spectra. The spectra were corrected for the small Q of the coil and the Boltzmann factor. The experimentally observed spin-echo intensity, $M(\tau)$, was extrapolated back to $\tau=0$ to correct for the temperature dependence of the spin-echo decay. In the hole-doped HTSCs, $M(\tau)=M_0\exp(-2\tau/T_{2R})\exp(-2\tau^2/T_{2g}^2)$, where the first factor is the Redfield correction arising from spin-lattice relaxation and the second Gaussian factor is due to electron-mediated indirect nuclear-spin-spin coupling between neighboring Cu nuclei.[15–17] This factor reduces to $\exp(-2\tau/T_2)$ if the Gaussian approximation to the coupling breaks down. Such a simple exponential law is observed in $Sr_{0.9}La_{0.1}CuO_2$ (Ref. 18) and $Pr_{1.85}Ce_{0.15}CuO_4$ (Ref. 19) below 300 K. Therefore, the magnetization at $\tau=0$ was estimated by fitting the spin-echo intensity to $M(\tau)=M_0\exp(-2\tau/T_2^*)$, where $1/T_2^*=1/T_{2R}+1/T_2$.

III. RESULTS AND ANALYSIS

The uniform electronic spin susceptibility χ was measured at 1 T and plotted in Fig. 1 for the pure and 1% Ni samples. There is a Curie-type upturn in χ for the Ni doped sample and the pure sample. The appearance of a Curie-type increase in χ for the pure sample could arise from a small impurity phase below the detection limit of the x-ray diffraction measurements ($<\sim 1\%$) or a small moment per Cu that has been suggested from the analysis of the Cu NMR linewidth data.[18] There is also a small linear increase in χ at high temperatures and hence the data above T_c were fitted to

$$\chi = \frac{N}{V}\frac{\mu_0 P_{\mathrm{eff}}^2 \mu_B^2}{3k_B T} + \chi_1 T + \chi_0, \tag{1}$$

where N/V is the number of impurities per unit volume, P_{eff} is the effective moment per Bohr magneton, k_B is Boltzmann's constant, and χ_1 and χ_0 are constants. The first term is the Curie term. The third term includes the temperature-independent parts to the spin susceptibility (van Vleck term, the Landau term, and the core term).

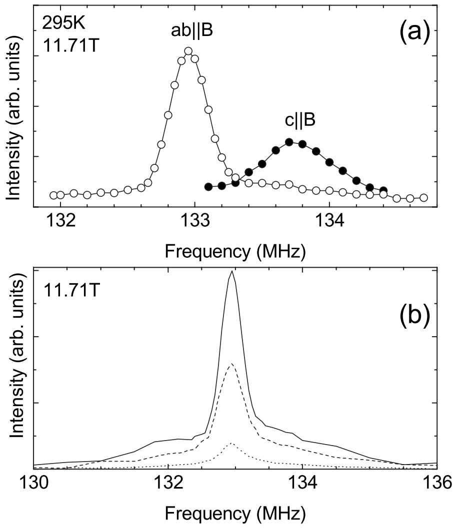

FIG. 2. (a) Plot of the ^{63}Cu NMR spectra at 295 K and 11.71 T for $ab\|B$ (open circles) and $c\|B$ (filled circles). (b) Plot of the ^{63}Cu NMR spectra from c-axis aligned $Sr_{0.9}La_{0.1}Cu_{0.99}Ni_{0.01}O_2$ with $ab\|B$ and for temperatures of 295 K (solid curve), 150 K (dashed curve), and 50 K (dotted curve). The Boltzmann, Q, and T_2^* corrections have been made.

We show by the solid curve in Fig. 1(a) that Eq. (1) can describe the χ data for the pure sample (solid curve). Also shown is the contribution of the Curie (dotted curve) and $\chi_1 T+\chi_0$ (dashed curve) terms. From the Curie fit it is possible to deduce a P_{eff} of 0.06 per Cu site. However, it is not known if such a small P_{eff} is intrinsic or extrinsic. The $\chi_1 T+\chi_0$ term could be due to a small increase in the density of states (DOS) for energies away from the Fermi energy that will lead to a small increase in the Pauli-spin susceptibility with increasing temperature. A much larger temperature-dependent increase in χ occurs in the underdoped hole-doped HTSCs and it has been attributed to a normal-state pseudogap.[20]

The χ data from the 1% Ni sample were also fitted to Eq. (1) and the resultant best fit is plotted in Fig. 1(b) (solid curve). The contributions from the Curie (dotted curve) and $\chi_1 T+\chi_0$ (dashed curve) terms are also plotted. The small linear increase in $\chi_1 T+\chi_0$ is similar to that seen in the pure sample although the absolute values of $\chi_1 T+\chi_0$ are lower. If the Curie term only arises from the Ni moments then it corresponds to $P_{\mathrm{eff}}=0.76/\mathrm{Ni}$. This reduces to $P_{\mathrm{eff}}=0.40/\mathrm{Ni}$ if the small $P_{\mathrm{eff}}=0.06/\mathrm{Cu}$ found in the pure compound is assumed for the remaining Cu sites.[18]

P_{eff} for the Ni substituted sample is significantly less than that expected for the high spin configuration ($S=1$ and $P_{\mathrm{eff}}=2.83$) or even for $S=1/2$ ($P_{\mathrm{eff}}=1.73$). Rather, similar to measurements on the hole-doped HTSCs,[10] P_{eff} is small and shows that there is considerable hybridization between the Ni 3d and the oxygen 2p orbitals.

The ^{63}Cu NMR spectra from the Ni substituted sample are plotted in Fig. 2(a) for the ab plane parallel to the applied magnetic field, $ab\|B$ (open circles), and for the c axis parallel to the applied magnetic field, $c \| B$ (filled circles), at 295 K. The peaks arise from the central $1/2 \leftrightarrow -1/2$ transition of the ^{63}Cu NMR nucleus. Since ^{63}Cu has a nuclear quadrupole moment, two additional satellite peaks from the $3/2 \leftrightarrow 1/2$ and $-1/2 \leftrightarrow -3/2$ transitions are expected at frequencies ν_Q above and below that of the $1/2 \leftrightarrow -1/2$ transition, where ν_Q is the nuclear quadrupole frequency. However, similar to the pure compound,[18,21] we find no well-resolved satellite transitions, which in the pure compound has been attributed to charge disorder in the CuO$_2$ layers.[18] Consequently, the satellite transitions are smeared out and lead to weak and broad additional intensity that extends to ± 3 MHz.

It is apparent in Fig. 2(a) that the ^{63}Cu NMR linewidth is larger for $c \| B$ than for $ab \| B$. Anomalously larger $c \| B$ linewidths have previously been reported in the pure compound where the linewidth for $c \| B$ is about three times that found for $ab \| B$.[18,21] It has been suggested that this could be due to a small antiferromagnetic spin-density oscillation on the Cu sites that is nearly temperature independent in the 50–300 K temperature range.[18] A similar effect was previously reported from Cu NMR measurements on YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_{6.7}$.[12] For the 1% Ni substituted sample we find that the linewidth at room temperature is significantly greater than that found in the pure sample where the ^{63}Cu NMR linewidth is 110 kHz for $ab \| B$. This suggests that Ni induces an additional antiferromagnetic spin-density oscillation.

We show in Fig. 2(b) that there is a temperature-dependent loss of ^{63}Cu NMR intensity. Here we plot some of the ^{63}Cu NMR spectra at 250 K (solid curve), 150 K (dashed curve), and 50 K (dotted curve). The spectra have been corrected for Q, T_2^*, and the Boltzmann factor. A temperature-dependent loss of the ^{63}Cu NMR intensity is also observed in the pure compound,[18] the electron-doped HTSC Pr$_{2-x}$Ce$_x$CuO$_4$,[19] as well as in hole-doped HTSCs.[22–31] It has been argued that this could arise from a slowing down of the spin fluctuations where a slowing down results in an increase in $1/{}^{63}T_1$ and the Cu NMR signal rapidly decreases as the correlation time of the spin fluctuations approaches the Larmor period so that $^{63}T_1$ and hence T_2 become smaller than the time between the $\pi/2$ and π pulses.[22]

This is a reasonable interpretation for the hole-doped HTSCs because there is clear evidence of antiferromagnetic spin fluctuations as well as dynamic inhomogeneities that have been interpreted in terms of fluctuating hole-rich and hole-poor regions.[32–34] It can be seen in Fig. 3(a) that the loss in ^{63}Cu NMR intensity for the 1% Ni sample is not signaled by a large change in T_2^* for $ab \| B$ at 11.71 T (filled circles) or 9.2 T (crosses). Rather, T_2^* is only a weak function of temperature and the values are comparable to those in the pure compound (open circles). This suggests that similar to the pure compound,[18] the electron-doped HTSC Pr$_{2-x}$Ce$_x$CuO$_4$,[19] and the hole-doped HTSCs,[22,27] the slowing down is spatially inhomogeneous.

The temperature dependence of the ^{63}Cu NMR intensity normalized to the intensity at room temperature, $I(T)/I(295\ \mathrm{K})$, is plotted in Fig. 3(b) for the pure compound at 9.2 T (open circles) and the 1% Ni sample at 9.2 T (crosses) and 11.71 T (filled circles). It is apparent that there is an additional loss of Cu NMR intensity for the 1% Ni sample where $(I_{\mathrm{Ni}}(T)/I_{\mathrm{Ni}}(295\ \mathrm{K})) = F(T)(I_{\mathrm{pure}}(T)/I_{\mathrm{pure}}(295\ \mathrm{K}))$ and $F(T)$ represents the additional loss in NMR intensity. To parametrize the data over the experimental temperature range, we take $F(T) = 1 - b_1 \times \exp(-T/b_2)$ where b_1 and b_2 are parameters. This function has the convenience that it is large at low temperatures and zero at high temperatures. We show in Fig. 3(c) the temperature dependence of $F(T)$ obtained from the experimental data in Fig. 3(b). The additional Ni-induced loss of ^{63}Cu NMR intensity is very large at low temperatures. This could be due to Ni pinning more of the spin fluctuations that are already slowing down with decreasing temperature or there is possibly a wipeout of the ^{63}Cu NMR intensity from Cu sites near the Ni site similar to that reported in the hole-doped HTSC NdBa$_2$(Cu$_{1-x}$Ni$_x$)$_3$O$_{7-\delta}$.[35] We note that wipeout of the ^{63}Cu NMR signal has also been reported near the Zn impurity in YBa$_2$Cu$_3$O$_{7-\delta}$ and YBa$_2$(Cu$_{1-x}$Zn$_x$)$_4$O$_8$.[12,36]

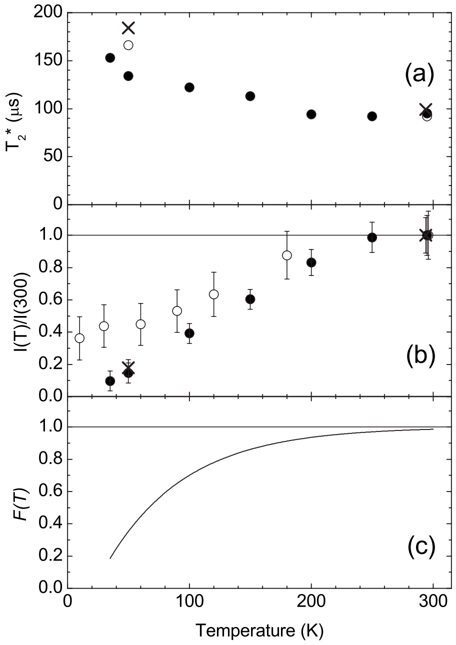

FIG. 3. (a) Plot of the ^{63}Cu NMR T_2^* against temperature for Sr$_{0.9}$La$_{0.1}$Cu$_{0.99}$Ni$_{0.01}$O$_2$ (filled circles) and Sr$_{0.9}$La$_{0.1}$CuO$_2$ (open circles) with $ab \| B$ and at 11.71 T. Also shown are data from measurements on the 1% Ni sample at 9.2 T (crosses). (b) Plot of the ^{63}Cu NMR integrated intensity from Sr$_{0.9}$La$_{0.1}$Cu$_{0.99}$Ni$_{0.01}$O$_2$ with $ab \| B$ at 11.71 T (filled circles) and 9.2 T (crosses). Also shown is the integrated intensity measured in the pure compound (open circles) and for $c \| B$ at 9.2 T (Ref. 15). The data have been corrected for T_2^*, Q, and the Boltzmann factor. (c) Plot of the additional loss in ^{63}Cu NMR intensity from Sr$_{0.9}$La$_{0.1}$Cu$_{0.99}$Ni$_{0.01}$O$_2$ when compared with Sr$_{0.9}$La$_{0.1}$CuO$_2$, $F(T)$, against temperature.

The ^{63}Cu NMR linewidth of the central $1/2 \leftrightarrow -1/2$ transition in the 1% Ni sample is larger at lower temperatures. This can be seen in Fig. 4 where the ^{63}Cu NMR spectra at 9.2 T are plotted for the 1% Ni sample at 294 K in Fig. 4(a) and at 50 K in Fig. 4(b). The resultant full widths at half maximum (FWHMs) are 206 kHz at 294 K and 540 kHz at 50 K. This can be compared with FWHMs of 110 kHz at 295 K in the pure compound that increases slightly to 130 kHz as the temperature is reduced to 50 K. The significantly greater

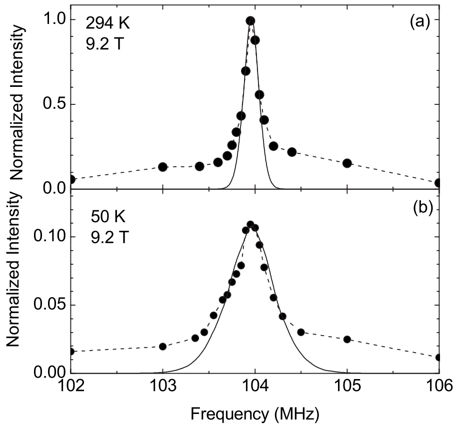

FIG. 4. Plot of the normalized ^{63}Cu NMR spectra for $Sr_{0.9}La_{0.1}Cu_{0.99}Ni_{0.01}O_2$ at (a) 294 K (filled circles) and (b) 50 K (filled circles) for $ab \| B$ and in an applied magnetic field of 9.2 T. The data were normalized to the peak intensity at 294 K after the Boltzmann, Q, and T_2^* corrections were made. Also shown are modeled $1/2 \leftrightarrow -1/2$ ^{63}Cu NMR spectra (solid curves) using the Ni-induced spin-density oscillation model described in the text with $\xi = 4.5$, $r_{\mathrm{wipeout}} = 5.5$ at 50 K, and $r_{\mathrm{wipeout}} = 0.5$ at 294 K.

increase in the FWHM found in the 1% Ni sample at 50 K could arise from a Ni-induced antiferromagnetic spin-density oscillation about the Ni impurity, similar to that found in Zn-substituted hole-doped HTSCs.[11,12]

The effect of an antiferromagnetic spin-density oscillation about the Ni site can be illustrated using the model of Walstedt *et al.*[11] that was developed to account for the Cu NMR linewidth in $YBa_2Cu_{3-x}Zn_xO_{7-d}$. We assume that Ni induces a spin-density oscillation that has the same spatial dependence as that assumed to occur about the Zn induced moment. This can be written as[11]

$$\langle s\rangle(i,j) = \gamma(-1)^{i+j} \sum \exp[-(i^2 + j^2)/(4\xi^2)], \qquad (2)$$

where ξ is the antiferromagnetic correlation length divided by the ab-plane lattice parameter, a. In the model of Walstedt *et al.*,[11] $\gamma = -J\chi_0\beta^{1/2}\langle S_L\rangle/(16\pi\mu_B^2)$, where J is the exchange energy, χ_0 is the host-band uniform susceptibility, β is a scale parameter, and $\langle S_L\rangle = (P_{\mathrm{eff}}^2 B)/(g\mu_B 3k_B T)$. The spectra were modeled by placing Ni randomly on a 400×400 two-dimensional (2D) grid to represent a CuO_2 plane and then adding the induced spin-density oscillation. The hyperfine field at each Cu site, $h(i,j)$, was calculated using the model applied to the hole-doped HTSCs where $h(i,j) = A\langle s\rangle(i,j) + B\Sigma\langle s\rangle(i+\delta i, j+\delta j)$ and $\delta i, \delta j = -1, 1$.[11] For simplicity we assume that the hyperfine constants are the same as those reported in the hole-doped HTSCs where $A = 4.8$ T and $B = 8$ T for $ab \| B$ and $A = -33$ T for $c \| B$.[37] The ^{63}Cu NMR spectra were modeled using Gaussians and an intrinsic linewidth that is equivalent to 100 kHz. Since J is not known, we modeled the data with $\gamma = a_1/T$, where a_1 was varied to model the data. This simple model requires that ξ is large so that the ^{63}Cu NMR spectra do not show large peaks from nearest-neighbor Cu sites. For this reason we used $\xi = 4.5$. It is also possible to include wipeout of the ^{63}Cu NMR intensity near the Ni site using a simple model with complete wipeout within a radius, r_{wipeout}. The resultant simulations of the central $1/2 \leftrightarrow -1/2$ transitions are shown in Fig. 4 (solid curves) where we used $r_{\mathrm{wipeout}} = 0.5$ at 294 K and $r_{\mathrm{wipeout}} = 5.5$ at 50 K. It can be seen that this model can account for the large ^{63}Cu NMR linewidths at low temperature.

IV. CONCLUSION

In conclusion, we have shown that the Ni moment in $Sr_{0.9}La_{0.1}Cu_{0.99}Ni_{0.01}O_2$ is small when compared with the expected value for an isolated Ni ion. This has also been reported in some of the hole-doped HTSCs. We find a temperature-dependent loss of the ^{63}Cu NMR signal for the 1% Ni sample that is greater than that observed in the pure compound. This could arise from an additional temperature-dependent pinning of the dynamic inhomogeneities by Ni. The ^{63}Cu NMR linewidth is also significantly larger in the sample containing 1% Ni and increases with decreasing temperature. It is possible that this is due to an induced spin-density oscillation about the Ni sites as well as wipeout of the ^{63}Cu NMR signal for Cu sites near the Ni sites.

ACKNOWLEDGMENTS

Funding support was provided by the New Zealand Marsden Fund (G.V.M.W.), the New Zealand Foundation for Research Science and Technology (G.V.M.W.), and the Ministry of Science and Technology of Korea through the Creative Research Initiative Program (C.U.J., Min-Seok Park, and S.-I.L.).

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