Nuclear quadrupole resonance study of $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ (R=Eu,Gd) and the effects of electronic doping

G. V. M. Williams
The MacDiarmid Institute, Industrial Research, P.O. Box 31310, Lower Hutt, New Zealand and the Leibniz Institute of Solid State and Materials Research (IFW), Dresden, Germany
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Cu nuclear quadrupole resonance measurements have been made on the single CuO_2 layer underdoped high temperature superconducting cuprate (HTSC) $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ (R=Eu,Gd). It is found that the nearly constant superconducting transition temperature for x=0.6 and 0.8 can be attributed to a similar hole concentration in the CuO_2 planes and the hole concentration is significantly lower for the nonsuperconducting x=1.0 sample. Similar to other HTSCs, the ^{63}Cu spin-lattice relaxation rate decreases with increasing hole concentration in the CuO_2 planes. There is evidence of a spatially inhomogeneous spin fluctuation spectrum that exists even for the superconducting samples. Similar inhomogeneities have been reported in some of the other HTSCs and, hence, the current results provide further evidence of inhomogeneities in the HTSCs.

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INTRODUCTION

The ruthanate-cuprates are proving to be particularly interesting because they exhibit superconductivity and magnetic order where the magnetic ordering temperature is above the superconducting transition temperature.[1–6] Nuclear magnetic resonance (NMR) studies have focused on $RuSr_2RCu_2O_8$ (R=Gd, Eu, Y) (Refs. 7–10) that displays low-field antiferromagnetic order with a small ferromagnetic component.[5,6,10–12] Even though the low-field ferromagnetic component is small, there is evidence that it leads to a spontaneous vortex phase in the superconducting state.[13] ^{63}Cu NMR measurements on $RuSr_2YCu_2O_8$ have shown that the spin dynamics in the CuO_2 layers are not affected by coupling to Ru moments and the temperature dependence of the spin-lattice relaxation rate has been interpreted in terms of the CuO_2 layers being similar to overdoped high temperature superconducting cuprates (HTSCs).[6] The understanding of $RuSr_2RCu_2O_8$ is complicated by studies that show a mixed Ru valence[7,14,15] in the RuO_2 layers even though the RuO_2 layers are conducting.[16] This may suggest phase separation in the RuO_2 layers that could lead to an inhomogeneous electronic state in the CuO_2 planes.

In many respects $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ (R=Eu,Gd) is a more interesting compound to study because it can be doped by changing the oxygen content or varying the Ce concentration[1,3,17] and, unlike $RuSr_2RCu_2O_8$, it has a fixed Ru valence near 5+ (Ref. 18 and 19) irrespective of the Ce concentration. Magnetization measurements show a ferromagnetic signal below temperatures of between 60 and 115 K, depending on the oxygen and Ce concentrations.[3,17] Remarkably the superconducting transition temperature, T_c, does not significantly change for Ce concentrations ranging from 0.4 to 0.8.[17] It has been suggested from the room temperature thermopower versus hole concentration correlation found in the hole-doped HTSCs (Refs. 20–22) that the average hole concentration in the CuO_2 layers is not changing for Ce doping in the range $0.4 <= x <= 0.8$.[17] Since the Ru valance does not change, then either Ce doping is being compensated for by a changing oxygen content or there is pair-breaking leading to T_c values that are lower than expected for the more hole-doped samples. The x=0.6 compound has a T_c of ~50 K but the transition into the bulk Meissner phase occurs at a much lower temperature of ~20 K, which has been attributed to a spontaneous vortex phase.[4] Our previous Cu nuclear quadrupole resonance (NQR) study on a nonsuperconducting x=1.0 sample provided evidence for an inhomogeneous electronic state in the CuO_2 layers.[23] It is not known if the same is true for superconducting $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$.

In this paper we report the results from Cu NQR measurements on the ruthenate-cuprates, $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$. The data are compared with those from measurements on other HTSCs.

EXPERIMENTAL DETAILS

The preparation of the $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ samples is described elsewhere.[17] Samples were oxygen loaded at pressures of up to 100 bar. The oxygen content was changed by annealing at different temperatures and oxygen partial pressures followed by quenching. The superconducting transition temperature was measured from the resistance data using a four terminal technique as described elsewhere.[17] Magnetization measurements were made using a superconducting quantum interference device (SQUID) magnetometer. Room temperature thermopower measurements were made using the standard temperature gradient technique.

The description of the Cu NQR apparatus is described in a previous report.[24] The Cu spin-lattice relaxation rate, $1/T_1$, was measured using a Hahn echo sequence with a $\pi/2$ pulse width of 1 μs and the time between the $\pi/2$ pulse and the π pulse, τ, was 9–10 μs. The NQR spectra were obtained using the Hahn echo sequence and at discrete frequency steps. The intensity at each frequency was obtained by Fourier transforming the second half of the echo and then integrating the Fourier transformed spectra.

The Cu spin-lattice relaxation rate was obtained by fitting the spin-echo magnetization recovery to

$$M(\tau) = M_0\left\{1 - 2\exp\left[-\left(\frac{3\tau}{T_1}\right)^n\right]\right\}. \quad (1)$$

In this case τ is the time between the inversion pulse and the start of the spin-echo sequence. For homogeneous materials $n=1$, which is observed for temperatures near and above room temperature. For lower temperatures $n<1$. The departure from monoexponential behavior is due to a distribution of spin-lattice relaxation rates that can be parameterized by introducing the parameter n as done by other researchers.[23,25–29]

The spin-spin relaxation rate was obtained by fitting the spin-echo decay to[30]

$$M(\tau) = M_0\exp\left(-\frac{2\tau}{T_{2R}}\right)\exp\left(-\frac{1}{2}\frac{(2\tau)^2}{T_{2g}^2}\right). \quad (2)$$

The first factor is the Redfield contribution and the value found in $YBa_2Cu_4O_8$ is used in the current study, where $T_{2R}^{-1}=1.77T_{1,NQR}^{-1}$.[31] The second factor is a Gaussian decay function and it occurs when the fluctuating spins are in the same spectral window.[30,32,33] The Gaussian component disappears if there are large short-range inhomogeneities or if the excitation window is only a small fraction of the total spectral width. This can lead to $M(\tau)$ of the form $M(\tau) = M_0\exp(-2\tau/{}^{63}T_2^*)$.

RESULTS AND ANALYSES

The Cu NQR spectrum is plotted in Fig. 1 at 140 K from $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$ (open circles). This sample has a T_c of ~50 K as determined from the electrical resistance and the transition into the bulk Meissner phase commences at ~20 K as determined from magnetization measurements. It also displays magnetic order with a ferromagnetic component where the maximum slope occurs at $T_m \sim 70$ K. The two broad features in the Cu NQR spectrum are due to ^{65}Cu (lower peak) and ^{65}Cu (higher peak) where the different peak frequencies are due to different nuclear quadrupole moments. Similar Cu NQR spectra are obtained for temperatures above 100 K and there is no significant change in the spectral line shape.

The spectrum in Fig. 1 at 140 K was fitted to two Gaussians to account for the two Cu isotopes and the full width at half maximum (FWHM) for ^{63}Cu is 2.3 MHz. This is large but it is comparable to that observed in the single CuO_2 plane HTSC, $La_{2-x}Sr_xCuO_4$.[34–36] It is also similar to that reported in magnetic but nonsuperconducting $RuSr_2EuCeCu_2O_{10+\delta}$.[23] However, unlike $RuSr_2EuCeCu_2O_{10+\delta}$, we find that the variation in $1/{}^{63}T_1$ with frequency is small as can be seen in the inset to Fig. 1. For example, the maximum increase in $1/{}^{63}T_1$ at 140 K is ~16% when going from the high to the low frequency side of the $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$ spectrum. The corresponding increase at 300 K is only ~6% (Fig. 1 inset). This can be compared with a much larger change of ~70% at 163 K observed in $RuSr_2EuCeCu_2O_{10+\delta}$ over a similar frequency range.[23]

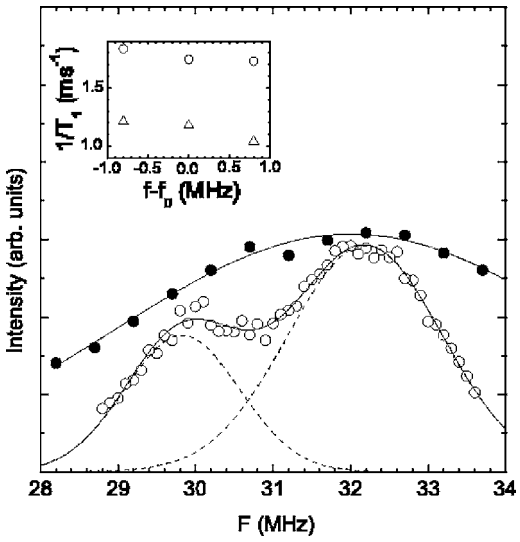

FIG. 1. Plot of the ^{63}Cu NQR spectrum from $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$ at 140 K (open circles). The solid curve is a two Gaussian fit to the data. The contributions from ^{65}Cu and ^{63}Cu are also shown (dashed curves). Also shown is the ^{63}Cu NQR spectrum from $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$ at 9 K (filled circles). The solid curve is a two Gaussian fit to the data. Inset: Plot of $1/{}^{63}T_1$ at 292 K (open circles) and 140 K (open up triangles) against the difference in frequency from the peak frequency.

It should be noted that $1/{}^{63}T_1$ measured on the low frequency side of the ^{63}Cu peak will also contain a contribution from the ^{65}Cu nuclei. This can increase the measured $1/T_1$ because the gryomagetic ratio, γ_n, is larger for ^{65}Cu when compared with ^{63}Cu. For metallic and magnetic systems, $1/{}^{65}T_1=({}^{65}\gamma_n/{}^{63}\gamma_n)^2(1/{}^{63}T_1)$, and hence, $1/{}^{65}T_1$ is 1.15 times greater than $1/{}^{63}T_1$. As can be seen in Fig. 1, $1/T_1$ will only be affected by a contribution from ^{65}Cu nuclei on the low frequency side of the ^{63}Cu peak. However, most of the small change in $1/T_1$ arises on the high frequency side of the ^{63}Cu peak and there is no significant change in the NQR line shape for temperatures above 100 K. Thus, the small changes in $1/{}^{63}T_1$ with frequency can not be attributed to a contribution from ^{65}Cu nuclei.

A variation in $1/{}^{63}T_1$ with frequency has previously been reported from Cu NQR measurements on $La_{2-x}Sr_xCuO_4$ and it was interpreted in terms of spatially inhomogeneous spin fluctuations due to a spatially inhomogeneous hole concentration, p, where the length scale of the charge inhomogeneity is ~3 nm.[36] Attributing the variation in $1/{}^{63}T_1$ across the NQR peak to a variation in the hole concentration relies on the experimental observation that $1/{}^{63}T_1$ systematically decreases with increasing p (Ref. 37) and the ^{63}Cu NQR frequency, ν_Q, increases with increasing p.[37,38] Thus, within the interpretation of Singer, Hunt, and Imai,[34] the lower frequency part of the Cu NQR spectrum arises from regions in the CuO_2 planes with a low p and the higher frequency part of the Cu NQR spectrum arises from regions in the CuO_2 planes with a higher p. As we show later, $1/{}^{63}T_1$ at the peak NQR frequency is also lower for a higher p in $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$. Consequently, the frequency variation in $1/{}^{63}T_1$ for $x=1.0$ and $x=0.6$ could be interpreted in

terms of the hole concentration being spatially inhomogeneous for both samples. The frequency variation is large for $x=1.0$ and small for $x=0.6$, which could imply that the inhomogeneities are larger for $x=1.0$.

An alternative interpretation of the frequency variation of $1/^{63}T_1$ in $La_{2-x}Sr_xCuO_4$ has recently been provided by Itoh *et al.*[39] It was suggested that the hole concentration is homogeneous and there exists a random distribution of "impurity relaxation centers" with an induced charge density oscillation. It was speculated that the lower frequency part of the Cu NQR spectrum arises from Cu sites near the "impurity relaxation centers" and "staggered moments" close to the "impurity relaxation centers" lead to an enhanced Cu spin-lattice relaxation rate. The middle and higher frequency parts of the spectrum are associated with Cu sites that are an intermediate distance from the impurity relaxation centers and in the bulk respectively. While there is no evidence of impurity relaxation centers and staggered moments in $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$, there is insufficient evidence to discount this possibility. However, the frequency variation in $1/^{63}T_1$ observed in $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ does indicate that the spin fluctuation spectrum is spatially inhomogeneous.

For temperatures between ~30 and ~90 K there is a large decrease in the NQR intensity that arises from the magnetization fluctuations in the RuO_2 planes and, hence, it was not possible to measure the Cu NQR spectrum in this region. At low temperatures the NQR intensity is recovered and the NQR spectrum is significantly broadened as can be seen in Fig. 1 (filled circles, 9 K). The FWHM was estimated by fitting the NQR spectrum to two Gaussians to account for the ^{65}Cu and ^{63}Cu isotopes. The resultant ^{63}Cu FWHM is ~6.7 MHz at 9 K, which is significantly greater than that above 100 K.

$RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ is unusual amongst the HTSCs because the variation in T_c, for samples oxygen-loaded under the same conditions, is small for a change in x from 0.8 to 0.4.[17] This corresponds to a change of 0.2 in the doped holes per Cu if all the doped holes appear in the CuO_2 planes. There are number of possibilities for the similar T_c values that include, (a) charge compensation by a decrease in the oxygen content, or (b) pair-breaking and, hence, a lower T_c value for the more hole doped samples. To investigate the origin of the anomalously similar T_c values, the Cu NQR spectra from $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ samples was measured at room temperature for different oxygen contents and with different Ce fractions.

In Fig. 2 ν_Q is plotted against the room temperature thermopower, S (300 K), for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10+\delta}$ with different oxygen contents. It has been found that there is a correlation between S (300 K) and p in the HTSCs, where increasing thermopower implies a lower p.[20–22] Thus, the data in Fig. 2 are consistent with ν_Q increasing with increasing average p, assuming a similar S (300 K) vs p correlation for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10+\delta}$. Using the S (300 K) vs p data found for a number of HTSCs (Ref. 22) it is possible to plot the data in Fig. 2 against p. The result can be seen in the left bottom inset to Fig. 2. Also shown is a linear fit to the data where the coefficient is 33 MHz/doped hole per Cu. This

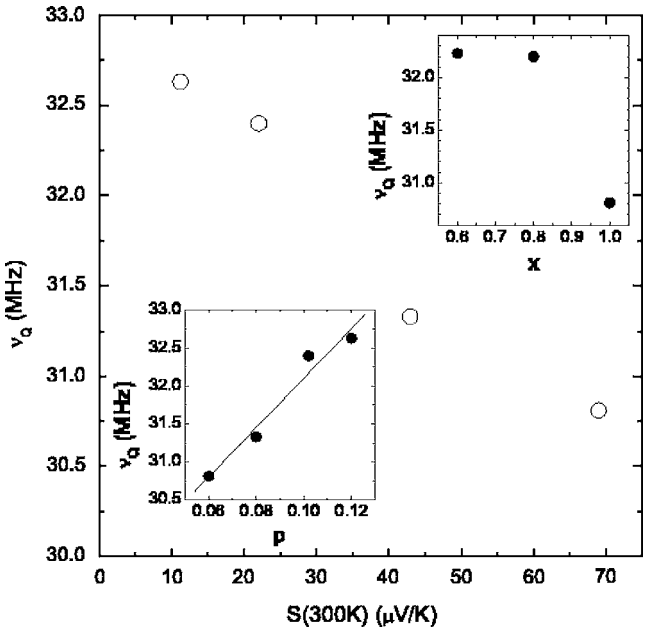

FIG. 2. Plot of the ^{63}Cu NQR frequency at 292 K against the room temperature thermopower for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10+\delta}$. Bottom left inset: Plot of the ^{63}Cu NQR frequency against p for $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10+\delta}$. Top right inset: Plot of the ^{63}Cu NQR frequency against the Ce fraction for $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ oxygen loaded at 100 bar.

coefficient is within the range found in the hole-doped HTSCs.[37,38] The correlation between ν_Q and p in the hole-doped HTSCs can be understood by first noting that ν_Q is directly proportional to the electric field gradient at the Cu nucleus. It is believed that the Cu electric field gradient is dominated by large positive and negative terms arising from the Cu $3d$ orbitals and virtual hoping from the neighboring oxygen ions to the unoccupied Cu $4p$ orbitals.[38,40] The net difference between the positive and negative terms systematically increases with increasing hole concentration.

It can be seen in the right upper inset to Fig. 2 that the $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ samples with 0.6 Ce and 0.8 Ce, which have comparable T_c values, also have nearly the same ν_Q, while the nonsuperconducting sample has a much lower ν_Q. Therefore, the current results show that the hole concentration in the CuO_2 planes is nearly the same for $x=0.6$ and 0.8 and it is greatly reduced for the nonsuperconducting sample. It has previously been shown that the doped holes do not appear in the RuO_2 plane[18,19] and, hence, it is reasonable to assume that the changing Ce fraction is offset by a changing oxygen content.

The temperature-dependent spin dynamics were probed by ^{63}Cu spin-lattice relaxation measurements and the resultant $1/^{63}T_1T$ data are plotted in Fig. 3 (filled circles) for $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ with $x=0.6$ along with previous data from Cu NQR measurements on a nonsuperconducting sample with $x=1.0$ (filled squares[23]). The $1/^{63}T_1$ data were obtained by fitting the spin-echo magnetization recovery to Eq. (1) and the resultant n values are plotted in the inset to Fig. 3 for $x=0.6$ and $x=1.0$. Both samples show a decrease in n for temperatures below 300 K and the values of n are

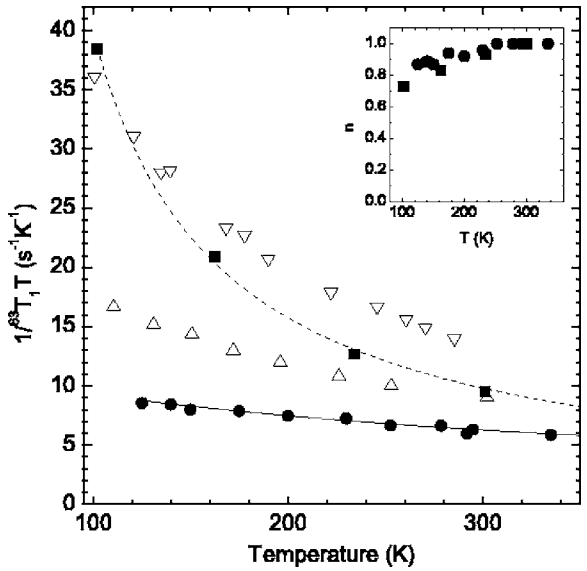

FIG. 3. Plot of $1/^{63}T_1T$ from $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ with $x=1$ [filled squares (see Ref. 23)] and $x=0.6$ (filled circles) at the peak frequency. Also shown is $1/^{63}T_1T$ from $La_{2-x}Sr_xCuO_4$ with $x=0.075$ [open down triangles (see Ref. 37)] and $x=0.24$ [open up triangles (see Ref. 37)] at the NQR peak frequency. The dotted and dashed curves are fits to the data as described in the text. Inset: Plot of the n parameter used to fit the $1/^{63}T_1$ magnetization data for $x=1$ (filled squares) and $x=0.6$ (filled circles) at the peak frequency.

lower for $x=1.0$. The changes in n are small for $x=0.6$ and it reduces from 1 at 250 K to ~0.9 at 125 K. This small change in n is unlike to arise from a contribution from ^{65}Cu nuclei that increases with decreasing temperature because there is no significant change in the NQR spectrum for temperatures above 100 K. It may be due to spatially inhomogeneous spin fluctuations that are known to lead to an n less than 1.

Both the $x=1.0$ and $x=0.6$ samples show an increasing $1/^{63}T_1T$ with decreasing temperature that is also observed in the HTSCs as can be seen in Fig. 3 for $La_{1.925}Sr_{0.075}CuO_4$ (open down triangles[37]) and $La_{1.76}Sr_{0.24}CuO_4$ (open up triangles[37]). For the HTSCs it has been argued that the Curie-Weiss-like increase in $1/^{63}T_1T$ with decreasing temperature arises from antiferromagnetic spin fluctuations where the antiferromagnetic spin fluctuation spectrum as probed by $1/^{63}T_1T$ increases with decreasing temperature.[37,41,42]

The effect of spin fluctuations on $1/^{63}T_1T$ for metallic and magnetic systems can be understood by considering the magnetic relaxation rate which can be written as[43]

$$(T_1T)^{-1} = \frac{1}{2}\hbar\, k_B \gamma_n^2 \sum_{\mathbf{q}} |A(\mathbf{q})|^2 \frac{\chi''(\mathbf{q},\omega_0)}{\hbar\,\omega_0}, \qquad (3)$$

where $\chi''(\mathbf{q},\omega_0)$ is the imaginary part of the dynamical spin susceptibility at the NQR angular frequency, ω_0, and $|A(\mathbf{q})|$ is the form-factor containing the hyperfine coupling constants. Using the spin Hamiltonian derived for the HTSCs,[44] the dynamical spin susceptibility of Millis, Monien, and Pines (MMP),[42] and the assumed temperature-dependence of the antiferromagnetic correlation length, it is possible to produce $1/^{63}T_1T$ of the form $1/^{63}T_1T=b_1/(T+\vartheta)$, which is the temperature-dependence displayed by the data.

We show in Fig. 3 that $1/^{63}T_1T$ from the $x=0.6$ and $x=1.0$ samples can also be fitted to $1/^{63}T_1T=b_1/(T+\vartheta)$. The resultant (b_1,ϑ) values are ($2610\ s^{-1}$, -34 K) for $x=0.6$, which can be compared with ($3850\ s^{-1}$, 314 K) found in $x=1.0$. The $1/^{63}T_1T$ absolute values are also comparable to those in the HTSCs,[37] which suggests that $1/^{63}T_1T$ over the measured temperature range is dominated by antiferromagnetic spin fluctuations in the CuO_2 planes. The significantly lower $1/^{63}T_1T$ found in the superconducting 0.6 Ce sample when compared with the nonsuperconducting 1.0 Ce sample shows that, similar to the HTSCs, $1/^{63}T_1T$ decreases with increasing hole concentration.

The interpretation that $1/^{63}T_1T$ above 100 K is dominated by antiferromagnetic spin fluctuations in the CuO_2 planes rather than magnetic fluctuations from the RuO_2 planes is consistent with a simple estimate of the magnitude of the Ru moment contribution to $1/^{63}T_1T$ via the direct dipole interaction. The effect of dipole relaxation can be estimated in a manner similar to that done for Gd in $GdBa_2Cu_3O_7$ (Ref. 45) and Pr in $Y_{1-y}Pr_yBa_2Cu_3O_{7-\delta}$ (Ref. 46) and $Pr_{2-y}Ce_yCuO_4$.[23] As shown by Reyes *et al.*[46] the direct dipole interaction can lead to a Ru moment contribution to $1/^{63}T_1$ for temperatures far above the magnetic ordering temperature that can be written as

$$1/^{63}T_{1,\mathrm{Pr}} \sim {}^{63}\gamma_n^2 \mu_{eff}^2 \langle r^{-6}\rangle/\omega_{ex}, \qquad (4)$$

where ${}^{63}\gamma_n$ denotes the ^{63}Cu nuclear gyromagnetic ratio, $\mu_{,eff}$ the Ru effective moment, ω_{ex} the Ru-Ru exchange frequency, and r is the distance between Cu and Ru where the brackets indicate a spatial average. ω_{ex} can be estimated from the magnetic ordering temperature.[45] The high temperature contribution of Ru to $1/^{63}T_1$ from the dipole interaction is estimated to be of the order of 4 s^{-1} from Eq. (4), and using the magnetic ordering temperature of 60 K, the known atomic positions,[47] and the measured Ru effective moment.[48] This is significantly less than the measured value of $1800\ s^{-1}$ at room temperature for $x=0.6$.

It is more difficult to estimate the magnitude of any contribution to $1/^{63}T_1T$ from transferred hyperfine coupling from Ru to Cu in the paramagnetic temperature regime. However, this might be expected to be small because the transferred hyperfine coupling occurs via a number of different orbitals. Furthermore, the Ru moment spin fluctuations in the NMR frequency range might be expected to be similar for samples with $x=0.6$ and $x=1.0$ because experimental studies show that $\mu_{,eff}$ is similar for both doping levels.[48] If the Ru-Cu transferred hyperfine coupling constants are also similar, then this should lead to values of $1/^{63}T_1T$ in the Curie-Weiss temperature regime that are comparable for $x=1.0$ and 0.6. However, it is apparent in Fig. 3 that this is not the case.

Additional information about the spin dynamics in the CuO_2 planes can be obtained from the spin-spin relaxation rate. At 140 K the spin-echo intensity can be fitted to Eq. (2), with a Gaussian decay. This is apparent in Fig. 4 (filled circles, top and right axis), where the spin-echo intensity is

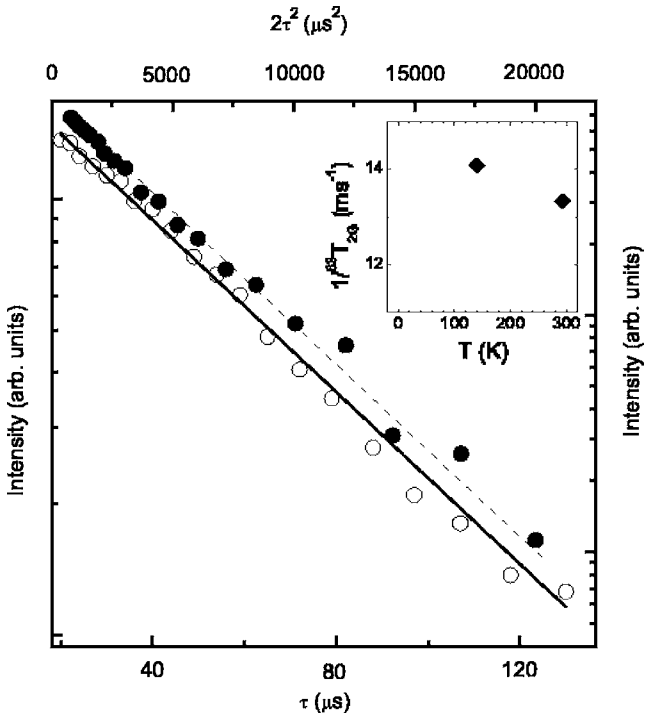

FIG. 4. Plot of the log of the inversion recovery magnetization from $RuSr_2Eu_{1.4}Ce_{0.6}Cu_2O_{10+\delta}$ against τ at 9 K (right and bottom axes, open circles) and against $2\tau^2$ at 140 K (left and top axis, filled circles). Inset: Plot of $1/^{63}T_{2G}$ against temperature.

plotted at 140 K against $2\tau^2$ for $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$. As mentioned earlier, a Guassian decay is observed for electron mediated spin-spin coupling between neighboring Cu copper spins having the same Lamor frequency. Thus, any short-range charge or spin inhomogeneities are not sufficient to significantly shift the Cu NQR frequencies for the nearest neighbor Cu sites. Although the measured $1/^{63}T_{2G}$ values are within the range observed in other HTSCs,[31,49,50] it should be noted that the $\pi/2$ pulse width of 1 μs is insufficient to uniformly flip all of the nuclear spins. This is known to lead to a measured $1/^{63}T_{2G}$ that is less that the actual value. Unfortunately, this was unavoidable because it was not possible to perform the measurements with a $\pi/2$ pulse width of less than 1 μs.

It can be seen in the Fig. 4 inset that $1/^{63}T_{2G}$ from $RuSr_2Eu_{1.6}Ce_{0.4}Cu_2O_{10+\delta}$ shows a small increase with decreasing temperature for temperatures at, and above, 140 K. Since the spectral line shape did not change over the same temperature range, it is unlikely that this small increase is due to a decrease in the fraction of flipped nuclei. In fact a similar initial increase in $1/^{63}T_{2G}$ with decreasing temperature has been observed in other HTSCs. It has been accounted for within the MMP model by $1/^{63}T_{2G}$ probing the real part of the dynamical spin susceptibility near the antiferromagnetic wavevector and a antiferromagnetic correlation length that increases with decreasing temperature.[37]

At low temperatures the spin-echo intensity is no longer Gaussian, as can be seen in Fig. 4 (open circles, left and bottom axis). Here the spin-echo intensity is plotted at 9 K and the solid line is a best fit to $M(\tau)=M_0\exp(-2\tau/^{63}T_2^*)$. The absence of a Gaussian component could indicate that the nearest neighbor Cu spins have a different Larmor frequency arising from very short range charge or spin inhomogeneities in the CuO_2 plane. However, the absence of a Gaussian component could also be attributed to the excitation window being much smaller that the spectral width at 9 K. The second possibility is more likely because the ^{63}Cu NQR linewidth is $\sim$3 times greater than that above 100 K and there is significant overlap between the ^{63}Cu and ^{65}Cu lines.

CONCLUSION

In conclusion, $RuSr_2R_{2-x}Ce_xCu_2O_{10+\delta}$ samples with $x=0.8$ and $x=0.6$ annealed under the same conditions and with similar T_c values also have similar hole concentrations in the CuO_2 planes. Thus, the negligible change in T_c is not anomalous because the additional holes introduced by Ce doping do not appear in the CuO_2 planes. As suggested in an earlier study, Ce doping is possibly compensated for by a changing oxygen content at least for $x=0.8$ and $x=0.6$. Similar to $La_{2-x}Sr_xCuO_4$, it is found that $1/^{63}T_1T$ from $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ decreases with increasing hole concentration and, hence, the spin fluctuation spectrum changes with hole concentration. From $1/^{63}T_1T$ measurements, the spin fluctuation spectrum is found to be spatially inhomogeneous for the superconducting $x=0.6$ sample, which has also been observed in the nonsuperconducting $x=1.0$ sample. However, the inhomogeneities are much smaller for $x=0.6$ when compared with $x=1.0$. The appearance of a spatially inhomogeneous spin fluctuation spectrum in the $RuSr_2Eu_{2-x}Ce_xCu_2O_{10+\delta}$ HTSC is not unique because similar inhomogeneities were found in $La_{2-x}Sr_xCuO_4$. Measurements on other families of HTSCs are required to determine if inhomogeneities are intrinsic to the HTSCs.

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