Precise knowledge of the spin susceptibility $\chi'(q, \omega)$ of the cuprates is essential for understanding their unusual normal state properties. The imaginary part, $\chi'(q, \omega)$, can be probed either by inelastic neutron scattering (INS) or in the low frequency limit by NMR measurements of the spin-lattice relaxation rate $1/T_1$. In contrast, one knows little about the real part of the susceptibility, $\chi(q)$, since information can, so far, only be extracted from the NMR observation of the Gaussian component of the transverse relaxation time, $T_{2G}$, of planar Cu. In particular, the analysis of INS and NMR experiments has not yet led to a consensus on the shape of $\chi(q, \omega)$ in momentum space and the temperature ($T$) dependence of the antiferromagnetic correlation length, $\xi$. To this end, we present new numerical simulations in this issue based on experiments by Bobroff et al. Our principal conclusions are that $\xi$ in YBa$_2$Cu$_3$O$_{6+\delta}$ is $T$-dependent and that using Ni impurities in YBa$_2$(Cu$_{1-x}$Ni$_{x}$)$_3$O$_{6+\delta}$ provides a completely consistent description of the data, whereas the Gaussian form can be ruled out.

Bobroff et al. recently presented a novel approach to the measurement of $\chi'(q)$ using Ni impurities in YBa$_2$(Cu$_{1-x}$Ni$_{x}$)$_3$O$_{6+\delta}$. These impurities induce a spin polarization at the planar Cu sites via $\chi'(q)$. The hyperfine coupling between Cu and O induces a spatially varying polarization and an additional broadening

$$\Delta \nu_{\text{imp}} = \Delta \nu - \Delta \nu_0 = \alpha f(\xi)/T$$

of the planar $^{17}$O NMR, where $\Delta \nu$ and $\Delta \nu_0$ are the total and $x = 0$ line width, respectively. In Eq. (1), $\alpha$ is the overall amplitude of $\chi'(q)$ and $f(\xi)$ characterizes the dependency of $\Delta \nu$ on $\xi$ ($\alpha = 4\pi \chi^*$ in the notation of Ref. [9] and [4]). Finally, the factor $1/T$ is caused by the Curie behavior of the Ni impurities in YBa$_2$Cu$_3$O$_{6+\delta}$ with effective moment $p_{\text{eff}} \approx 1.9\mu_B$ ($1.59\mu_B$) for $\delta = 0.6$ ($\delta = 1$). Bobroff et al. found that $T\Delta \nu(T)$ strongly depends on temperature and the Ni concentration $x$ in the sample. Furthermore, they observed a much stronger broadening in the underdoped, $\delta = 0.6$, sample than in the overdoped one with $\delta = 1$. Performing numerical simulations of the NMR line shape by assuming a Gaussian form for $\chi'(q)$, they found that $f(\xi)$ is basically constant for all physically reasonable values of $\xi$. Combining these results with $T_{2G}$ data by Takigawa, they concluded that $\xi$ is $T$-independent for the underdoped samples. On the other hand, in every scenario of cuprate superconductors in which the anomalous low-energy behavior is driven by spin fluctuations one would expect the correlation length $\xi$ to be $T$-dependent (for recent reviews, see: [22, 23]). Thus their result has important implications about the mechanism of superconductivity. We recently pointed out, that our simulations using a Lorentzian form of $\chi'(q)$ yield a different result and are actually compatible with a $T$-dependent $\xi$.

Before going into the details of our calculations, it is important to notice that the fact that $\xi$ must be $T$-dependent can be deduced even without a detailed model from the very experimental data by Bobroff et al. for $\Delta \nu(T)$ and Takigawa for $T_{2G}$. To show this, we need to recognize that we can always express $T_{2G}$ as a product of $\alpha$ and a function of $\xi$, namely

$$T_{2G}^{-1} = \alpha g(\xi).$$

(2)

We can then eliminate $\alpha$ by forming the product

$$T \Delta \nu_{\text{imp}} T_{2G} = \frac{f(\xi)}{g(\xi)}$$

(3)

which depends solely on $\xi$. In Fig. 1, we plot the product $T\Delta \nu(T)$ as a function of $T$. We see that this product is strongly $T$-dependent, dropping by more than a factor of 2 between 100 K and 200 K. Therefore $\xi$ must have a substantial $T$-dependence.

To have a more quantitative insight into the $T$-dependence of $T\Delta \nu(T)$ of Ref. [9], we must go into details. We present in the following a theoretical analysis of the $^{17}$O line shape using a method first applied by Bobroff et al., to simulate their experimental data.

To simulate the $^{17}$O line shape numerically, we distribute Ni impurities on a $(100 \times 100)$ lattice with concentration $\frac{1}{2}$ randomly at positions $r_j$ on a two dimensional lattice. We consider the Ni impurities as foreign
atoms embedded in the pure material, which is characterized by a non-local spin-susceptibility \( \chi'(q) \). In the following, we consider two different forms of the spin susceptibility. We used the data from Ref. [8] and [14].

The coupling constant \( J \) is an unknown parameter of the theory and will be estimated below. Furthermore, we will assume like Bobroff et al. that the Ni impurities do not change the magnetic correlation length or the magnitude of the spin susceptibility.

For the NMR experiments we consider an external magnetic field \( B_0 \) along the \( z \)-direction. The Ni spins have a non-zero average value obeying \( \langle S_i^z \rangle = C_{\text{Curie}}B_0/T \) with Curie constant \( C_{\text{Curie}} = \mu_0/(2\sqrt{3}k_B) \).

Adopting a mean field picture, the induced polarization for the electron spins at the Cu sites \( r_i \) is given by

\[
\langle s_i^z \rangle = \frac{J}{(g\mu_B)^2} \sum_j \chi'(r_i - r_j) \langle S_j^z \rangle .
\]

Here, \( \chi'(r) \) is the real space Fourier transform of \( \chi'(q) \). In the following we consider two different forms of the spin susceptibility [17]. For the commensurate case, there is only one peak, whereas in the incommensurate case, one has to sum over four peaks. The Gaussian form of \( \chi'(q) \) is given by

\[
\chi'_G(q) = \alpha \xi^2 \exp \left(-\frac{(q - Q)^2}{\xi^2}\right) \tag{6}
\]

and the Lorentzian form by

\[
\chi'_L(q) = \alpha \xi^2/(1 + (q - Q)^2\xi^2) \tag{7}
\]

Since the question whether there exist incommensurate peaks in YBa\(_2\)Cu\(_3\)O\(_{6+x}\) has not been settled yet, we will consider below both cases, a commensurate wavevector \( Q = (\pm \pi, \pm \pi) \), and an incommensurate one with \( Q = \delta_i(\pm \pi, \pm \pi) \). The calculation of the real space Fourier transform finally yields

\[
\chi'_G(r) = \frac{\alpha}{4\pi} F(Q) \exp \left(-\frac{r^2}{4\xi^2}\right),
\]

\[
\chi'_L(r) = \frac{\alpha}{4\pi} F(Q)K_0 \left(\frac{r}{\xi}\right), \tag{8}
\]

where \( K_0 \) is the modified Bessel function, and \( F(Q) = \cos(Q_x r_x) \cos(Q_y r_y) \).

Having determined the Ni induced Cu spin polarization \( \langle s_i^z \rangle \), it is straightforward to investigate the \(^{17}\)O NMR line shape, determined by the coupling of the \( I = \frac{3}{2} \) nuclear spins \(^{17}I \) to the Cu electron spins \( s_i \) with spatially varying mean value \( \langle s_i^z \rangle \). The hyperfine Hamiltonian is

\[
H_{hf} = h^2 \gamma_n \gamma_e \sum_{i,j} \alpha_s^{i,j} s_i \cdot {^{17}I} \, i , \tag{9}
\]

where \( \gamma_n, \gamma_e \) are the gyromagnetic ratios for the \(^{17}\)O nucleus and the electron, respectively. The hyperfine coupling constants \( C_{i,j} \) is dominated by a nearest neighbor hyperfine coupling \( C \approx 3.3T/\mu_B \) [18]. However, it was recently argued that a next-nearest neighbor hyperfine coupling \( C' \approx 0.25C \) is relevant for the explanation of the spin-lattice relaxation rate [13] in La\(_{2-x}\)Sr\(_x\)Cu\(_3\)O\(_y\). We will therefore also consider its effects on the \(^{17}\)O NMR line.

Using a mean field description of this hyperfine coupling by replacing \( s_i \) by \( \langle s_i^z \rangle \) of Eq. [4], we finally obtain for the shift of the resonance at a given \(^{17}\)O site \( r_i \)

\[
\nu_i = \frac{A}{T} \sum_{j} C_{i,j} \chi'(r_i - r_j) . \tag{10}
\]

Here, the sum over \( i \) runs over the Cu spin sites, coupled to the \(^{17}\)O nuclear spin, whereas the \( j \)-summation goes over all Ni-sites. Furthermore, the constant prefactor \( A \) is given by \( \frac{2\gamma_n \gamma_e}{J} h C_{\text{Curie}}B_0/(g\mu_B)^2 \). Note, \( \nu_l \) as given in Eq. [4] is the shift of the \(^{17}\)O resonance with respect to the case without Ni impurities.

To obtain the \(^{17}\)O NMR line shape, we create a histogram \( I_o(\nu) = \sum_i \delta(\nu - \nu_i) \) counting the number of nuclei with shift \( \nu \). Since we want to compare the resulting distribution with the experimental data where
the line has a finite width even in the absence of impurities, we convolute \( I_0(\nu) \) with a Gaussian distribution \( \exp \left( -\nu^2/(2\sigma^2) \right) / \sqrt{2\pi}\sigma^2 \), yielding the lineshape \( I(\nu) \). By comparison with the experiments of Ref. 8 we expect that \( \Delta \nu_0 = \sqrt{2}\log 2\sigma \) should be of the order of the high temperature (i.e. \( \xi < 1 \)) Ni-impurity induced linewidth. In the following calculation we therefore choose \( \sigma = 20\text{kHz} \) for both the Lorentzian and Gaussian \( \chi(\mathbf{q}) \). Finally, we define the resulting \( \Delta \nu \) by half the width of the peak at half maximum. In Fig. 3 we present the lineshape of the \(^{17}\text{O} \) NMR signal, calculated with the Lorentzian form \( \chi_L(\mathbf{q}) \) for two different values of \( \xi \). We clearly observe that the line becomes broader as we increase \( \xi \). From a comparison of Eq. (11) with the experimentally measured broadening we can extract the value of the interaction \( J \) in Eq. (8). For \( C' = 0 \) and \( \xi(200\text{K}) = 4 \) (\( \xi(200\text{K}) = 3 \)) we obtain \( J \approx 25\text{meV} \) (43 meV). These values are accompanied by some uncertainties, but enable us to estimate the effects of a Cu spin mediated Ni-Ni spin (RKKY-type) interaction. We find within a self consistent mean field treatment of this interaction that the effect of the Ni-Ni interaction changes \( \Delta \nu \) only within a few percent, consistent with the fact that no significant deviation from a Curie law was found in susceptibility measurements 5,6.

In Fig. 3 we present a comparison of \( \Delta \nu(\xi) \) for the Gaussian \( \chi_G(\mathbf{q}) \) (open diamonds) and the Lorentzian \( \chi_L(\mathbf{q}) \) (filled squares). Here we chose a Ni concentration of \( x = 2\% \), \( C' = 0 \), and \( Q = \delta_i(\pm\pi, \pm\pi) \) to be incommensurate with \( \delta_i = 0.94 \) 4. We also compute \( \Delta \nu \) for the commensurate case, and find that in general \( \Delta \nu \) decreases. However, since the incommensurability, \( 1-\delta_i \), in YBa\(_2\)Cu\(_3\)O\(_{6.6}\), if present at all, is rather small, differences are negligible for \( \xi < 8 \). In Fig. 3 we clearly see that the effect of \( \chi_G(\mathbf{q}) \) and \( \chi_L(\mathbf{q}) \) on the behavior of the line width is qualitatively different. In agreement with the results by Bobroff et al. we find using \( \chi_G(\mathbf{q}) \) that \( \Delta \nu \) is basically independent of \( \xi \) for all physically reasonable values \( 2 < \xi < 5 \). The Lorentzian form \( \chi_L(\mathbf{q}) \), however, yields a much stronger increase in \( \Delta \nu \) between \( \xi = 2 \) and \( \xi = 5 \) than the Gaussian. This result immediately implies that a temperature dependent \( \xi \) is clearly compatible with the experimental results by Bobroff et al. Furthermore, we find that the function \( f(\xi) \) of Eq. (8) behaves like \( f(\xi) \sim \xi^{3/2} \) in the Lorentzian case and \( f(\xi) \sim \text{const.} \) in the Gaussian case. This qualitatively different behavior of \( \Delta \nu(\xi) \) for \( \chi_G(\mathbf{q}) \) and \( \chi_L(\mathbf{q}) \) makes this experiment extremely sensitive to details of the momentum dependence of \( \chi(\mathbf{q}) \).

Next we discuss the \( \xi \) dependence of \( \Delta \nu \) for different values of the Ni concentration \( x \). We present our results for a Ni concentration of \( x = 0.5\%, 2\% \) and \( 4\% \) and for \( C' = 0.25C \) in Fig. 4. In agreement with the experimental results we find that \( \Delta \nu \) for a given \( \xi \) increases with \( x \). We believe that the results in Figs. 3 and 4 also provide an explanation for the different behavior of \( \Delta \nu \) in the underdoped (YBa\(_2\)Cu\(_3\)O\(_{6.6}\)) and overdoped (YBa\(_2\)Cu\(_3\)O\(_{7}\)) samples. Bobroff et al. obtained that for the overdoped sample the variation of \( \Delta \nu \) with \( T \) is much weaker than for the underdoped sample. As far as \( \chi(\mathbf{q}) \) is concerned, the main difference between these two regimes consist in the value of \( \xi \), namely \( \xi = 1.2 \) for the overdoped and \( \xi = 2.4 \) for the underdoped sample. We see from Figs. 3 and 4 that the \( \xi \) variation of \( \Delta \nu \) for the overdoped sample is much weaker than for the underdoped one, in agreement with the experimental results.

Finally, we can use our numerical results to investigate in more detail the consequences of the \( T \)-dependence of
\[ \Delta \chi^\prime(\mathbf{q}) \propto \xi \] for \( \mathbf{q} = (\pi, \pi) \) and the other results for \( f(\xi) \), it follows from Eq. 3 for the Gaussian case \( f(\xi) \propto \text{const.} \) that \( T \Delta \chi^\prime T_{2G} \propto \xi^{-1} \), i.e. \( \xi \) has to increase with increasing \( T \). This result seems to be unphysical and thus strongly suggests that the Gaussian form \( \chi^\prime(\mathbf{q}) \) is inappropriate for the description of the spin susceptibility. On the other hand, for the Lorentzian case, \( f(\xi) \propto \xi^{3/2} \) and it follows \( T \Delta \chi^\prime T_{2G} \propto \xi^{1/2} \), i.e. \( \xi \) decreases as \( T \) increases, as we would expect. One can also solve Eqs. 1 and 2 to obtain \( \alpha \) as a function of \( T \). However, our result possess error bars which are quite large. The conclusion that \( \alpha \) is independent of \( T \) is acceptable within those errorbars, however, a weak \( T \) dependence cannot be excluded.

It is important to contrast our findings with the observations of INS experiments. In \( \text{YBa}_2\text{Cu}_3\text{O}_{6+\delta} \), INS observes a \( T \)-independent broad peak around \((\pi, \pi)\), above \( T_c \), resembling a Gaussian form of \( \chi^\prime(\mathbf{q}, \omega) \). However, strong indications for incommensurate peaks with Lorentzian like shape in \( \text{YBa}_2\text{Cu}_3\text{O}_{6+\delta} \) suggest that the broad structure around \((\pi, \pi)\) is only a superposition of incommensurate peaks. Its width is therefore determined by the merely \( T \)-independent incommensuration instead of \( \xi^{-1} \). This is consistent with the recent analysis by Pines that the overall magnitude of \( \chi^\prime(\mathbf{q}, \omega) \) in \( \text{YBa}_2\text{Cu}_3\text{O}_{6+\delta} \), as obtained from NMR experiments, necessitates a considerable improvement of the experimental resolution of INS experiments to resolve the incommensurate peaks in the normal state.

In conclusion we obtain from the analysis of the \( ^{17}\text{O} \) NMR data by Bobroff et al. and the \( T_{2G} \) data by Takigawa that the correlation length \( \xi \) must have a substantial temperature dependence. A detailed analysis shows that the Gaussian form \( \chi^\prime(\mathbf{q}) \) of the spin susceptibility can be excluded as an appropriate description of the spin dynamics in the doped cuprates. A more correct description is provided by a Lorentzian-type form \( \chi^\prime(\mathbf{q}) \), which is fully compatible with the experimental data and a temperature dependent \( \xi \). Though the resolution of the experiment does not allow us yet to determine the precise \( T \)-dependence of \( \xi \), our analysis shows that \( \xi \) considerably decreases with increasing temperature.

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**FIG. 4.** \( \Delta \nu(\xi)/\Delta \nu_0 \) for \( C' = 0.25C \) and three different Ni concentrations: \( x = 0.5\% \) (squares), \( x = 0.5\% \) (triangles) and \( x = 0.5\% \) (diamonds).