NQR AND $\mu$SR IN DILUTED TWO-DIMENSIONAL $S = 1/2$ HEISENBERG ANTIFERROMAGNETS*

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$^{139}$La NQR spectra and relaxation and $\mu$SR precessional frequencies in $\text{La}_2\text{Cu}_{1-x}\text{M}_x\text{O}_4$ (for $\text{M} = \text{Zn}$ and $\text{Mg}$) are reported in order to study the effect of spin dilution in the planar quantum Heisenberg antiferromagnet (2DQHAF) $\text{La}_2\text{CuO}_4$. The behavior of the spin stiffness $\rho_s(x)$ and of the in-plane correlation length $\xi_{2D}(x, T)$, of the sublattice magnetization and of the Néel temperature, for a dilution approaching the percolation threshold depart sizeably from the ones expected in dilution-like models. In spite of the marked reduction of $\rho_s$, the transition to the ordered state occurs at a temperature, where $\xi_{2D}(x, T_N)$ reaches a value close to the one in undoped 2DQHAF.

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1. Introduction

The behavior of characteristic magnetic properties, such as spin stiffness $\rho_s$ and in-plane correlation length $\xi_{2D}$, in pure as well as in charge- and/or spin disordered two-dimensional quantum Heisenberg antiferromagnets (2DQHAF) has recently attracted a great deal of interest. It has been proved [1–3] that $^{139}$La NQR relaxation in $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$ allows to derive the temperature and doping dependence of $\xi_{2D}(x, T)$. The main conclusions were that for $x \leq 0.1$ $\xi_{2D}$ follows the $T$-dependence expected in the renormalized classical (RC) regime, with $\rho_s$ and the spin-wave velocity $c_{sw}$ renormalized by quantum fluctuations, with respect to the mean field values. A simple dilution like model was found to account for most of the experimental findings [3]. Of particular interest is the range of strong dilution so

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that the percolation threshold, where no 3D long range order is developed, is approached. In this report we extend the $^{139}$La NQR and μSR measurements to La$_2$CuO$_4$ with Mg$^{2+}$ ($S = 0$) for Cu$^{2+}$ ($S = 1/2$) substitution to an extent $x \simeq 0.3$.

2. Experimental results

The $^{139}$La NQR spectra and μSR precessional frequencies were used to extract the Néel temperature $T_N(x)$ (Fig. 1(a)) and the sublattice magnetization, namely the expectation value $\langle \mu(x, T \to 0) \rangle$ (Fig. 1(b)) (for details on the procedure see Ref. [1]). For low dilution levels one has $(-1/T_N(0))/dT_N(x)/dx \approx 3.2$, as expected for an effective Hamiltonian

$$
{\mathcal{H}} = J_{\text{eff}}(x) \sum_{i,j} \vec{S}_i \cdot \vec{S}_j = J(1 - x)^2 \sum_{i,j} \vec{S}_i \cdot \vec{S}_j.
$$

For $x \geq 0.25$ a clear departure occurs, consistent with a percolation threshold at $x = 0.41$. The normalized sublattice magnetization $m = \langle \mu(x,0) \rangle/\langle \mu(0,0) \rangle$ (Fig. 1(b)) is compared to recent evaluations, carried out in the framework of different theoretical models. Only the behavior predicted for $m$ within spin wave theory seems to model the experimental findings. We point out that recent neutron scattering data, up to $x \simeq 0.42$ [4] qualitatively support our observation.

Fig. 1. (a) Néel temperature in La$_2$Cu$_{1-x}$(Mg,Zn)$_x$O$_4$. The dotted line is the behavior expected within the dilution model. (b) Sublattice magnetization as a function of spin dilution. The solid line was derived from quantum Monte Carlo simulations [6], the dashed line from spin-wave theory [7], the dotted line in the framework of an effective quantum non-linear σ model [8].
\(^{139}\)La NQR relaxation rate \(2W\), for \(x = 0.3\), is reported in Fig. 2. From the comparison of the recovery laws for the \(2\nu_Q\) and \(3\nu_Q\) resonance lines \([5]\) it has been proved that below \(T \approx 140\) K the relaxation process is of magnetic origin, i.e. driven by the time-dependence of the hyperfine magnetic field due to \(\text{Cu}^{2+}\) spins. The phonon contribution to the relaxation mechanism, which yields \(2W \propto T^2\), was subtracted to analyze the experimental data.

![Graph](image)

Fig. 2. Temperature dependence of \(^{139}\)La NQR spin-lattice relaxation rate in La\(_2\)Cu\(_{0.7}\)Mg\(_{0.3}\)O\(_4\). The solid line shows the phonon contribution to the relaxation.

### 3. Discussion and conclusions

The relaxation rate \(2W\) can be related to \(\xi_{2D}(x,T)\) along lines analogous to the ones already used \([2, 3]\) for pure and lightly doped 2DQHAF. The form factor relating \(2W\) to the generalized spin susceptibility was assumed \(|A_0|^2 \approx 10^6\) Gauss\(^2\). Thus, one can write \(2W \approx 3.3 \times 10^{-3}(\xi_{2D}/a)^2s^{-1}\) (\(a\) is the lattice step). The dynamical scaling exponent \(z\) was taken \(z = 1\), as for undoped or lightly doped 2DQHAF. The behavior of \(\xi_{2D}(x,T)\) (see Fig. 3) is close to the one expected in the RC regime:

\[
\xi_{2D}/a = \frac{\hbar e_{\text{SW}}}{16\pi k_B} \frac{e^{\frac{2\pi A_0(x)}{T}}}{T} \left[ 1 - 0.5 \frac{T}{2\pi \rho_h(x)} \right] \tag{2}
\]

In Fig. 3 the values \(\xi_{2D}(x,T_N(x))\) estimated from the mean-field argument (\(\xi_{2D}(x,T_N(x))/a^2J_\perp(1-x)^2 = T_N(x)\)) are also reported. It can be concluded that also for high dilution \(\xi_{2D}\) follows rather well Eq. (2), having left \(\rho_h(x)\) as an adjustable parameter. The values of \(\rho_h(x)\) are reported in Fig. 3. It is noted that in the strongly diluted regime, \(x \geq 0.1\),
\( \rho_0(x) \) departs dramatically from the dilution-like model behavior, where \( \rho_0(x) = 1.15 J_{\text{eff}}(x)/2\pi \propto (1 - x)^2 \). However, while \( \rho_0(x) \) is strongly affected by the spin dilution and \( T_N(x) \) is drastically reduced, the transition to the ordered state still occurs when \( \xi_{2D} \) reaches a value around 150 lattice steps, as in pure or lightly doped systems.

REFERENCES