

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

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$^{139}\text{La}$  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  $\xi_{2\text{D}}(x, T)$ , 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  $\xi_{2\text{D}}(x, T_N)$  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  $\xi_{2\text{D}}$ , 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  $^{139}\text{La}$  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  $\xi_{2\text{D}}(x, T)$ . The main conclusions were that for  $x \leq 0.1$   $\xi_{2\text{D}}$  follows the  $T$ -dependence expected in the renormalized classical (RC) regime, with  $\rho_s$  and the spin-wave velocity  $c_{\text{sw}}$  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  $^{139}\text{La}$  NQR and  $\mu\text{SR}$  measurements to  $\text{La}_2\text{CuO}_4$  with  $\text{Mg}^{2+}$  ( $S = 0$ ) for  $\text{Cu}^{2+}$  ( $S = 1/2$ ) substitution to an extent  $x \simeq 0.3$ .

## 2. Experimental results

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

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

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.

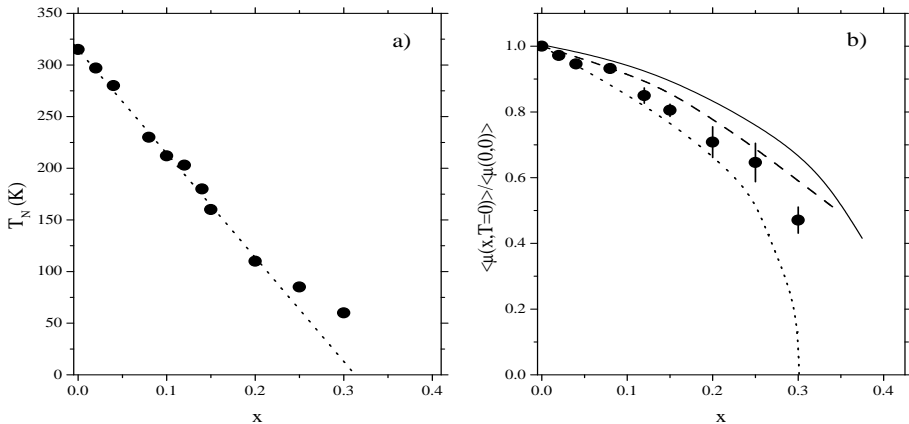


Fig. 1. (a) Néel temperature in  $\text{La}_2\text{Cu}_{1-x}(\text{Mg,Zn})_x\text{O}_4$ . The dotted line is the behavior expected within the dilution model. (b) Sublattice magnetization as a function of spin dilution. The solid line was derived from quantum Monte Carlo simulations [6], the dashed line from spin-wave theory [7], the dotted line in the framework of an effective quantum non-linear  $\sigma$  model [8].

$^{139}\text{La}$  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  $T \simeq 140$  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.

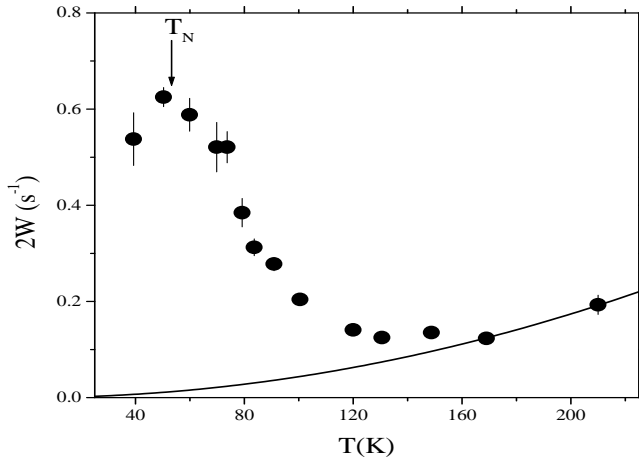


Fig. 2. Temperature dependence of  $^{139}\text{La}$  NQR spin-lattice relaxation rate in  $\text{La}_2\text{Cu}_{0.7}\text{Mg}_{0.3}\text{O}_4$ . The solid line shows the phonon contribution to the relaxation.

### 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  $|A_{\vec{q}}|^2 \simeq 10^6 \text{ Gauss}^2$ . Thus, one can write  $2W \simeq 3.3 \times 10^{-3} (\xi_{2D}/a)^z s^{-1}$  ( $a$  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:

$$\xi_{2D}/a = \frac{\hbar c_{\text{sw}}}{16\pi k_B \rho_s} e^{\frac{2\pi\rho_s(x)}{T}} \left[ 1 - 0.5 \frac{T}{2\pi\rho_s(x)} \right]. \quad (2)$$

In Fig. 3 the values  $\xi_{2D}(x, T_N(x))$  estimated from the mean-field argument  $(\xi_{2D}(x, T_N(x))/a)^2 J_{\perp} (1-x)^2 = T_N(x)$  are also reported. It can be concluded that also for high dilution  $\xi_{2D}$  follows rather well Eq. (2), having left  $\rho_s(x)$  as an adjustable parameter. The values of  $\rho_s(x)$  are reported in Fig. 3. It is noted that in the strongly diluted regime,  $x \geq 0.1$ ,

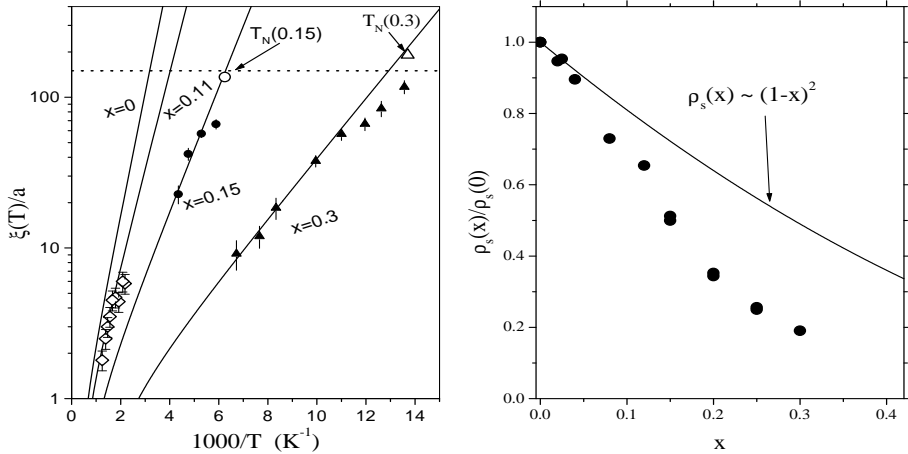


Fig. 3. Left: In-plane correlation length  $\xi_{2D}/a$  in  $\text{La}_2\text{Cu}_{1-x}(\text{Mg,Zn})_x\text{O}_4$  derived from nuclear relaxation rates (closed symbols) and from  $T_N$  values (open symbols). The solid lines show the behavior for  $\xi_{2D}$  in the RC regime. The horizontal dotted line shows the value of  $\xi_{2D}(x, T \rightarrow T_N)$ . Right: Spin-stiffness  $\rho_s(x)$  in  $\text{La}_2\text{Cu}_{1-x}(\text{Mg,Zn})_x\text{O}_4$ . The solid line is the behavior expected in the framework of a dilution-like model (see text).

$\rho_s(x)$  departs dramatically from the dilution like model behavior, where  $\rho_s(x) = 1.15 J_{\text{eff}}(x)/2\pi \propto (1-x)^2$ . However, while  $\rho_s(x)$  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.

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