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

P. CARRETTA, A. RIGAMONTI, E. TODESCHINI

Dipartimento di Fisica "A. Volta" — INFM Unitá di Pavia Via Bassi 6, 27100 Pavia, Italy

and L. Malavasi

Dipartimento di Chimica Fisica, Via Taramelli 10, 27100 Pavia, Italy

(Received July 10, 2002)

¹³⁹La NQR spectra and relaxation and μ SR precessional frequencies in La₂Cu_{1-x}M_xO₄ (for M= Zn and Mg) are reported in order to study the effect of spin dilution in the planar quantum Heisenberg antiferromagnet (2DQHAF) La₂CuO₄. 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 ρ_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.

PACS numbers: 76.60.Es, 75.40.Gb, 75.10.Jm, 75.50.Ee

1. Introduction

The behavior of characteristic magnetic properties, such as spin stiffness ρ_s and in-plane correlation length ξ_{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 ¹³⁹La NQR relaxation in La₂Cu_{1-x}Zn_xO₄ 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 ρ_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

^{*} Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10-13, 2002.

that the percolation threshold, where no 3D long range order is developed, is approached. In this report we extend the ¹³⁹La NQR and μ SR measurements to La₂CuO₄ with Mg²⁺ (S = 0) for Cu²⁺ (S = 1/2) substitution to an extent $x \simeq 0.3$.

2. Experimental results

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

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

For $x \ge 0.25$ 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 $\text{La}_2\text{Cu}_{1-x}(\text{Mg},\text{Zn})_x\text{O}_4$. 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 σ model [8].

¹³⁹La 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 Cu²⁺ 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 La NQR spin-lattice relaxation rate in $La_2Cu_{0.7}Mg_{0.3}O_4$. 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 $|A_{\vec{q}}|^2 \simeq 10^6$ Gauss². 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:

$$\xi_{\rm 2D}/a = \frac{\hbar c_{\rm sw}}{16\pi k_{\rm B}\rho_{\rm s}} e^{\frac{2\pi\rho_{\rm s}(x)}{T}} \left[1 - 0.5 \frac{T}{2\pi\rho_{\rm s}(x)} \right].$$
 (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 ξ_{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, $x \ge 0.1$,



Fig. 3. Left: In-plane correlation length ξ_{2D}/a in La₂Cu_{1-x}(Mg,Zn)_xO₄ derived from nuclear relaxation rates (closed symbols) and from T_N values (open symbols). The solid lines show the behavior for ξ_{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 La₂Cu_{1-x}(Mg,Zn)_xO₄. The solid line is the behavior expected in the framework of a dilution-like model (see text).

 $\rho_{\rm s}(x)$ departs dramatically from the dilution like model behavior, where $\rho_{\rm s}(x) = 1.15 J_{\rm eff}(x)/2\pi \propto (1-x)^2$. However, while $\rho_{\rm s}(x)$ is strongly affected by the spin dilution and $T_{\rm N}(x)$ is drastically reduced, the transition to the ordered state still occurs when $\xi_{\rm 2D}$ reaches a value around 150 lattice steps, as in pure or lightly doped systems.

REFERENCES

- M. Corti, A. Rigamonti, F. Tabak, P. Carretta, F. Licci, L. Raffo, *Phys. Rev.* B52, 4226 (1995).
- [2] P. Carretta, T. Ciabattoni, A. Cuccoli, E. Mognaschi, A. Rigamonti, V. Tognetti, P. Verrucchi, *Phys. Rev. Lett.* 84, 366 (2000).
- [3] P. Carretta, A. Rigamonti, R. Sala, *Phys. Rev.* B55, 3734, (1997).
- [4] O.P. Vajk, P.K. Mang, M. Greven, P.M. Gehring, J.W. Lynn, Science 295, 1691 (2002).
- [5] A. Rigamonti, F. Borsa, P. Carretta, *Rep. Prog. Phys.* **61**, 1367 (1998).
- [6] K. Kato, S. Todo, K. Harada, N. Kawashima, S. Miyashita, H. Takayama, Phys. Rev. Lett. 84, 4204 (2000).
- [7] A.L. Chernyshev, Y.-C. Chen, A.H. Castro Neto, Phys. Rev. B65, 104407 (2002).
- [8] Y.-C. Chen, A.H. Castro Neto, *Phys. Rev.* B61, R3772 (2000).