PRESSURE DEPENDENCE OF THE EXCHANGE INTERACTION IN La$_2$CuO$_4$

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We report the results of Raman scattering measurements which determine the pressure dependence of the in-plane exchange coupling constant $J$ in magnetic La$_2$CuO$_4$ for pressures between 1 bar and 100 kbar. We find that pressure increases the Neel temperature $T_N$ much more rapidly than $J$. This result together with the theory for a quasi two-dimensional magnetic system then suggests that the interplanar exchange coupling decreases with pressure.

1. INTRODUCTION

Raman scattering studies of the magnetic excitations of both the oxide superconductors and their insulating parent compounds have played a central role in elucidating the importance of spin fluctuations in high temperature superconductors(1). At the same time, both the superconductivity(2) and the magnetism(3) are remarkably sensitive to pressure, although the fundamental mechanism for this remains unclear. Although Raman scattering has been used to study the pressure dependence of the phonons in several of these materials(4), it has not previously been used to study the effects of pressure on the spin fluctuations. We present here the first report of the pressure dependence of the two-magnon excitation spectrum of magnetic La$_2$CuO$_4$, directly probed by Raman scattering.

2. EXPERIMENTAL TECHNIQUES

The light scattering experiments were performed on a mechanically polished single crystal of La$_2$CuO$_4$, grown from CuO flux. The as-grown crystal was annealed in nitrogen at 650°C for ten hours, a procedure which reduces the oxygen doping level, yielding an antiferromagnet with a measured Neel temperature of 305 K. The sample was mounted in the (110) orientation in a specially designed high pressure diamond anvil cell, using a stainless steel gasket and krypton as a pressure transmitting medium(5). The pressure was determined by the frequency shift of the luminescence of a small ruby chip placed near the sample. The Raman scattered light was analyzed with a CCD camera and a SPEX triple spectrometer(6). Laser power was kept below 7 mW to minimize sample heating.

3. EXPERIMENTAL RESULTS

The room temperature Raman spectra of B$_{1g}$ symmetry for our oriented single crystal sample of La$_2$CuO$_4$ at pressures ranging from 1 bar to 100 kbar and laser wavelength of 4579 Å are graphed in Fig. 1. The data have been corrected for the wavelength dependent response of the collection optics, spectrometer, and detector. A linear fluorescence background has been subtracted. We focus our attention on the broad peak centered near ~ 3200 cm$^{-1}$ which has previously been identified as two-magnon scattering in a Neel ground state(1). From these data, we have determined the pressure dependence of the two-magnon peak frequency $\omega_p$. The peak frequency $\omega_p$ is related(7) to the Cu superexchange interaction $J$ in a two-dimensional quantum antiferromagnet by the proportionality $\omega_p=3.18 J$. The pressure dependence of $J$ extracted from this relationship is plotted in Fig. 2, demonstrating that pressure increases $J$ monotonically in La$_2$CuO$_4$. The break in $J$ near 60 kbar may reflect the pres-

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sure at which the tetragonal to orthorhombic phase transition passes through room temperature(8).

4. DISCUSSION

As a result of a residual interlayer exchange interaction $J_\perp$, an ensemble of two dimensional magnetic sheets having an in-plane correlation length $\xi$ and staggered magnetization $M^*$ may order in three dimensions if the condition $T_N \approx J_\perp M^*\xi^2$ is met (9). Reference (9) suggests that for a choice of parameters consistent with neutron scattering experiments at 1 bar, both $\xi$ and $M$ increase with the in-plane exchange coupling $J$. Assuming that the renormalization constants describing the effects of the two-dimensional fluctuations are independent of pressure, the pressure dependence of $T_N$ is given by $T_N \propto J_\perp(P) J^2(P) \exp(1.4 J(P)/k_B T)$.

With this expression in mind, it is instructive to compare the pressure dependence of $T_N$ from neutron scattering experiments$^3$, plotted in Fig. 2, with that of $J(P)$, determined by the present Raman scattering experiment. Note that $T_N$ has been multiplied by a factor of 3.0 in order to make it equal to $J(P)$ at 1 bar. It is clear that while $T_N$ is more sensitive to pressure than $J(P)$, the theoretical expression predicts a much stronger dependence of $T_N(P)$ on $J(P)$ than is observed. This discrepancy suggests that pressure rapidly decreases $J_\perp$. Alternatively, the renormalization factors describing the effects of the two-dimensional fluctuations may be pressure dependent. More detailed investigation of the two-magnon lineshapes, which may shed light on this point, are underway.

5. CONCLUSIONS

We have presented here the first direct measurement of the pressure dependence of the in-plane exchange interaction $J(P)$ in magnetic La$_2$CuO$_4$. We find that pressure increases $J(P)$, but apparently either magnetically decouples the CuO$_2$ planes or fundamentally affects the two-dimensional fluctuations.

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