Received by G71

NOV 07 1989

NEUTRON SCATTERING STUDIES OF MAGNETIC CORRELATIONS
IN THE LAYERED CUPRATES

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All of the copper oxide superconductors contain CuO_2 layers which, when insulating, exhibit strong, two-dimensional (2D) antiferromagnetic correlations due to the unusually large superexchange interaction between nearest-neighbor Cu^{2+} atoms. Magnetic correlations survive, although with a reduced correlation length, even when the layers are doped with holes. A short review of neutron scattering studies of magnetic correlations and excitations in the layered cuprates is presented.

By doping the CuO_2 planes with holes (or electrons), it is possible to transform a number of layered copper oxide compounds from antiferromagnetic insulators to metallic superconductors. There is clearly a close connection between magnetism and superconductivity in the cuprates, and it is of considerable interest to determine the extent to which superconductivity competes with or depends upon the magnetism. Neutron scattering is a powerful probe which has provided considerable insight into the nature of magnetic correlations in both the metallic and insulating phases of the cuprates. In this paper, we will give a brief review of the picture obtained from neutron scattering studies of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+y}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$.

The effect of doping on the magnetic correlations within a CuO_2 plane will depend to a large extent on the nature of the antiferromagnetic state which is found in the insulating phase. It is often assumed that the magnetic moments are localized on the Cu atoms, with a moment of $\sim 1 \mu_B$ per Cu and a Heisenberg coupling ($J\mathbf{S}_i \cdot \mathbf{S}_j$) between nearest-neighbor spins. Such a model can be tested in several ways. First of all, the intensity of magnetic Bragg diffraction peaks is proportional to the square of the staggered magnetization M_s , which in turn is equal to $g\mu_B\langle S_z \rangle$, where $\langle S_z \rangle$ is the average spin on a Cu site. Spin-wave theory¹ for the 2D, $S = \frac{1}{2}$, square-lattice Heisenberg model at $T = 0$ predicts that $\langle S_z \rangle = 0.30$, so that, taking $g \approx 2.2$, $M_s \approx 0.67 \mu_B$. Experimentally it is observed that both the staggered magnetization and the Néel temperature T_N decrease rapidly with doping of the planes; however, as T_N approaches its maximum, M_s approaches the value pre-

FIGURE 1

Staggered magnetization vs. Néel temperature obtained by neutron diffraction measurements. Data is from Ref. 2.

dicted assuming full local moments, as shown in Fig. 1 for $\text{La}_2\text{CuO}_{4+y}$ (Ref. 2). Similar behavior has been observed in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (Ref. 3 and 4).

The magnetic Bragg peak intensities are also proportional to the square of the magnetic form factor, which is the Fourier transform of the spin density in real space. The Q dependence of the antiferromagnetic form factor measured in a single crystal of $\text{La}_2\text{CuO}_{4+y}$ is in reasonable agreement with the Cu^{2+} form factor.⁵ Since by symmetry the spin density must be zero at the centers of the in-plane oxygen atoms, it is not clear to what

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FIGURE 2

Temperature dependence of the inverse correlation length in La_2CuO_4 . The data are from Ref. 12.

extent this result is or is not consistent with electronic structure calculations which indicate that the hole on each Cu atom contains a significant fraction of O 2p character. To get a measure of the spin density on the in-plane oxygen atoms, one can attempt to measure the ferromagnetic form factor induced by an applied magnetic field. Such a study at $T > T_N$ in a $\text{La}_2\text{CuO}_{4+y}$ crystal indicated no significant contribution from the oxygens⁶; however, the applied field was much smaller than the effective exchange field between Cu atoms, so that it is difficult to properly compare the experimental results with theoretical calculations.

The superexchange interaction between neighboring Cu atoms via the bridging oxygen atoms in the CuO_2 planes is extremely strong. As a result, the spin-wave velocity $c = \omega/q$ is quite large, making it difficult to resolve the in-plane spin-wave branches at small q . Recently Aeppli *et al.*⁷ succeeded in resolving the 2D spin waves using epithermal neutrons, obtaining a spin wave velocity $\hbar c = 0.85 \pm 0.03$ eV-Å. Taking into account quantum renormalization factors, that velocity corresponds to an exchange parameter of 135 meV, in good agreement with recent two-magnon Raman scattering measurements.⁸ Large 2D spin-wave velocities are also observed^{9,10} in antiferromagnetic $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$.

The 3D magnetic ordering temperature in La_2CuO_4 is limited by the weak coupling between planes. Because of the extremely large in-plane coupling, the 2D mag-

FIGURE 3

Antiferromagnetic correlation length vs. Sr concentration in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. The solid line represents the average separation between the holes introduced by Sr doping. After Ref. 14.

netic correlation length ξ remains large well above T_N . The correlation length can be determined by integrating over the inelastic magnetic scattering to obtain the Fourier transform of the instantaneous spin-spin correlation function. By taking advantage of the fact that the scattering from a 2D system is independent of the momentum transfer perpendicular to the plane, one can perform the energy integration using a special scattering geometry.¹¹ The inverse correlation length as a function of temperature is shown in Fig. 2, as measured in two different crystals.¹² From a theoretical analysis of the 2D, $S = \frac{1}{2}$ Heisenberg model, Chakravarty *et al.*¹³ have shown that the inverse correlation length should equal $(2/a) \exp(-2\pi\rho_s/k_B T)$, where $a = 3.8$ Å and $2\pi\rho_s = 0.576\hbar c/a$. The solid line in Fig. 2 corresponds to this formula with $\hbar c = 0.85$ eV-Å. The same type of temperature dependence is expected for a system of classical spins, but for $S = \frac{1}{2}$ the correlation length at a given temperature is greatly reduced due to quantum fluctuations.

The correlation length is reduced when holes are added to the planes by doping La_2CuO_4 with either Sr or O. The variation of ξ vs. Sr content in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ as determined by neutron scattering is shown in Fig. 3. A typical length scale for the hole-doping is the average hole separation, $a/\sqrt{x}$, and the correlation length is observed to closely follow this curve. The picture of antiferromagnetically coupled moments in which the correlation length decreases due to O 2p holes implic-

FIGURE 4

Scans along the 2D scattering rod at a constant energy transfer of 6 meV measured in a single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{6.3}$. From Ref. 9.

itly assumes that the Cu moments themselves are not changed by the doping. At small x , muon spin rotation experiments have shown that this is indeed the case.¹⁵ In metallic samples of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ no direct measurement of the moments has been possible. To get a measure of the moments with neutron scattering, one would have to properly integrate the inelastic scattering up to the maximum excitation energies, which are presumably at least tenths of eV, and such a measurement would be exceedingly difficult.

It would also be desirable to measure the magnetic correlation length in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. Unfortunately, it is not possible to use the special scattering geometry which performs the energy integration because the inelastic scattering from the CuO_2 layers is not independent of the momentum transfer perpendicular to the planes. The Cu atoms in the CuO_2 layers do not form a Bravais lattice, and hence the spin waves are split into acoustic and optic modes, with each mode having a sinusoidal structure factor. The coupling between nearest-neighbor layers is sufficiently strong that one observes only the acoustic mode at energies up to at least 60 meV,^{9,10} so that a scan at a low energy along the 2D scattering rod is strongly modulated by the acoustic mode structure factor, as illustrated in Fig. 4. The

FIGURE 5

Direct comparison between the magnetic excitations of La_2CuO_4 and $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. From Ref. 16.

observation of the modulation at temperatures above T_N indicates that the bilayers remain correlated in the absence of long-range order.

If magnetic excitations are directly connected with electron-pairing in superconducting samples, then one might expect to see some anomalous temperature dependence of the inelastic scattering at low temperatures. Measurements of inelastic scattering in metallic cuprate samples is difficult because, relative to antiferromagnetic samples, the intensity at a given energy is spread out over a much wider range in reciprocal space due to the short correlation length. Very large and good quality crystals are required for such work. Figure 5 compares scans across the 2D scattering rod at an energy transfer of 6 meV for a superconducting crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ ($T_c = 33$ K) and an antiferromagnetic La_2CuO_4 crystal.¹⁶ Although the peak-to-background ratio is reduced in the doped crystal, the integrated intensity is essentially unchanged. An interesting feature is the development of incommensurate peaks, which so far have been observed only in superconducting crystals.¹⁷

Figure 6 shows the temperature dependence of the integrated intensity measured at energy transfers of 6

FIGURE 6

Temperature dependence of normalized integrated intensities at energy transfers of 6 and 12 meV for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. Solid lines are guides to the eye. From Ref. 16.

and 12 meV in the $T_c = 33$ K crystal.¹⁶ In crystals with $T_c \sim 0$ the intensity of low energy scattering is essentially independent of temperature,¹⁷ and the same behavior is observed at 12 meV in the superconducting crystal. In contrast, a sharp decrease in intensity is found at 6 meV between 150 and 50 K. Similar behavior was also observed at 3 and 9 meV. If the intensity actually went to zero at low temperature, it would suggest that a gap had opened up in the low-energy magnetic excitation spectrum. That the intensity decrease occurs at temperatures well above T_c is quite surprising; however, recent spin-lattice relaxation rate data from Cu NQR measurements by Imai *et al.*¹⁸ indicate a decrease in low-frequency spin fluctuations occurring in a similar temperature range.

Experiments are underway in search of magnetic excitations in superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, and we speculate that they will eventually be observed. A recent study¹⁹ has shown that strong 2D antiferromagnetic correlations are also present above T_N in Nd_2CuO_4 ; it will be quite interesting to learn how these

correlations evolve on doping the compound with Ce. Clearly, neutron scattering will continue to play an important role in investigations of magnetism in the layered cuprates.

ACKNOWLEDGEMENTS

Much of the work discussed here has been carried out at Brookhaven in collaboration with many scientists, including R. J. Birgeneau, Y. Endoh, P. Gehring, Y. Hidaka, B. Keimer, H. Kojima, M. Sato, T. R. Thurston, and K. Yamada. The major part of this work was supported by the U.S.-Japan Cooperative Neutron Scattering Program. Research at Brookhaven National Laboratory is supported by the Division of Materials Sciences, U.S. Department of Energy, under Contract No. DE-AC02-76CH00016.

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