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Magnetic Correlations in High T_c Oxides: Neutron Scattering Studies

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ABSTRACT

A review is given of current neutron scattering experiments on $La_{2-x}Sr_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$. Incommensurate magnetic excitations are observed in a high quality single crystal of $La_{1.85}Sr_{0.15}CuO_4$, which exhibits a sharp superconducting transition at 33 K. The study of magnetic correlations in the electron-superconductor $Nd_{2-x}Ce_xCuO_4$ is now being carried out.

I. INTRODUCTION

The magnetism in the layered high T_c superconductors has been extensively studied and inelastic neutron scattering has played a unique role in detailed studies of the magnetic excitations(1). Significant progress has been made in understanding the magnetic correlations in Sr-doped La_2CuO_4.(2-5) This is due to the remarkable success(6,7) attained in growing larger single crystals of these mixed oxides. High quality single crystals with volumes of 0.5 cc and larger are now available for inelastic neutron scattering.

Fig. 1 summarizes earlier measurements(3) of the correlation length ξ as a function of dopant concentration x in

$La_{2-x}Sr_xCuO_4$. The solid line represents a simple model calculation of the average distance between holes when the doping creates holes in O^{2-} randomly. The observed ξ approximately follows this line. One very important result not shown in this figure is the x-

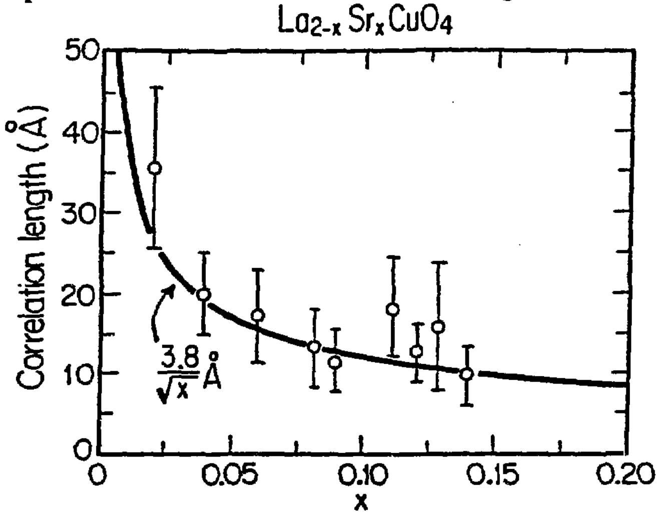

Fig. 1 Antiferromagnetic correlation length versus concentration in $La_{2-x}S_xCuO_4$. The solid line is the average separation between the holes introduced by the Sr doping. After Birgeneau et al.[3]

dependence of the average magnetic moment. The observed magnetic intensities are essentially independent of Sr concentration even into the superconducting range. Although tood single crystals were grown in this series, it was quite difficult to make T_c for these compounds near 40 K. Only very lately was this accomplished by the special growth technique of Tanaka and Kojima[7].

II. $La_{1.85}Sr_{0.15}CuO_4$

An exceptionally good single crystal of $La_{1.85}Sr_{0.15}CuO_4$ became available recently[7]. This crystal is labelled KOS-1 and Fig. 2 gives a direct comparison[5] between the parent (x = 0) crystal

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(NTT-2) and the superconducting (x = 0.15) crystal. These profiles reveal a few important characteristics. First, the integrated

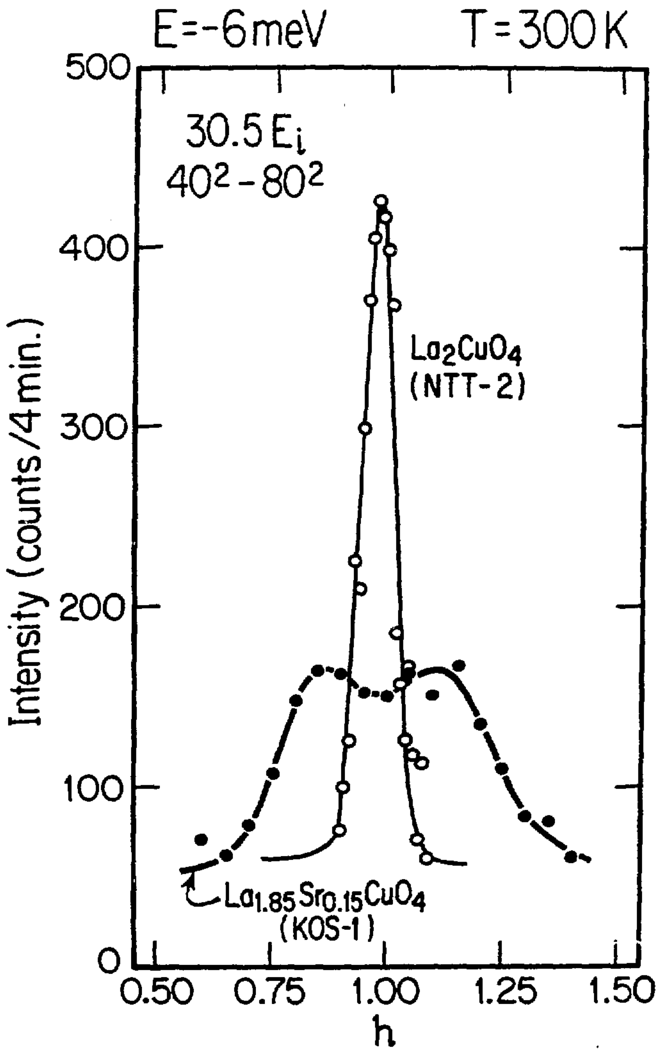

Fig. 2 Direct comparison between the magnetic excitations of La_2CuO_4 and $La_{1.85}Sr_{0.15}CuO_4$ [Ref. 5]

magnetic intensities at constant energy transfer are nearly identical, though a considerable broadening is noted for x = 0.15. Second, the modulated nature of this excitation, as previously reported[3,8], is clearly demonstrated. These double peaks are observed[4,5,8] only in superconducting concentrations.

Figure 3 shows the most significant results obtained so far in this series of experiments. It depicts the integrated intensities as functions of temperature for scans at 6 meV and 12 meV. The

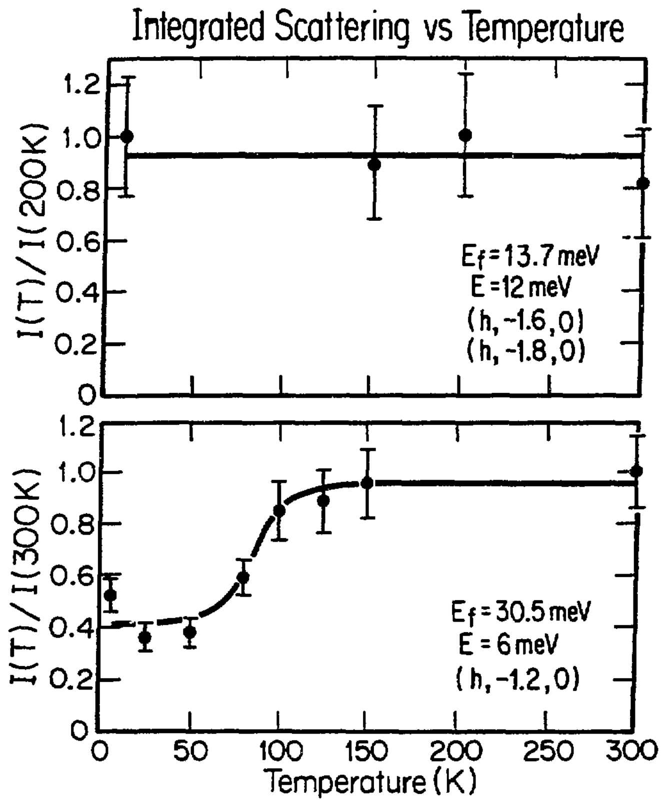

Fig. 3 Temperature dependence of normalized integrated intensities at energies 6 meV and 12 meV for $La_{1.85}Sr_{0.15}CuO_4$ (Ref. 5). Solid lines are guides to the eye.

intensity at 12 meV is independent of temperature, as is the case for doped compounds in the non-superconducting range(4). However, in KOS-1 there is a dramatic dimmunition in the intensity between 150K and 50K at 6 meV. Similar behavior was observed at

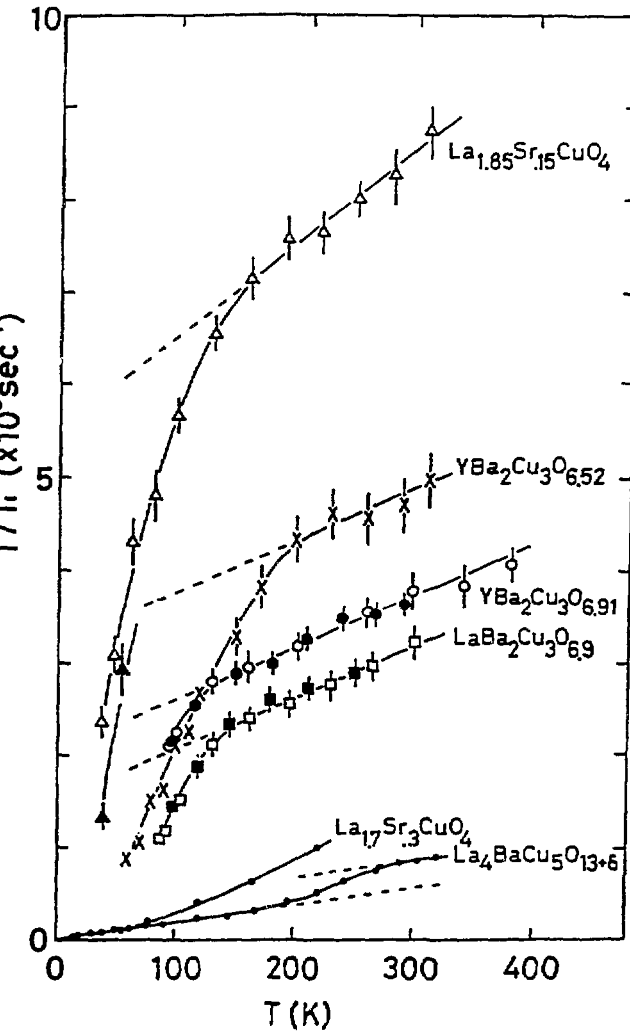

Fig. 4 The normal state behavior of $1/T_1$ of ^{63}Cu for various high T_c and related oxides. Figure 4 is taken from the recent abstract by Yasuoka et al.(9).

3 meV and 9 meV. For T_c = 33 K (KOS-1) the BCS gap $2\Delta = 3.5\ k_B T_c$ is 10 meV. The data shown in Fig. 3 are suggestive of the opening of a gap in the Cu^{2+} spin-excitation spectrum with an energy gap comparable to the BCS gap value.

We note that the apparent gap develops well above T_c. This is quite unexpected. This precursor for superconductivity must be revealed, if it is true, in other physical properties. We are delighted to see the NQR results of the relaxation time T_1, very recently reported by Yasuoka et al(9). Fig. 4 shows $1/T_1$ of ^{63}Cu for several high T_c superconductors including $La_{1.85}Sr_{0.15}CuO_4$. This compound shows a clear deviation from the high temperature behavior of aT+b around 150 K! They also noted that the constant b is closely related to the inverse of T_c.

III. $Nd_{2-x}Ce_xCuO_4$

A new class of high T_c superconductors, $Nd_{2-x}Ce_xCuO_4$, has been recently discovered(10). These materials have the unique property of a negative Hall coefficient in the normal state, suggesting electron rather than hole conduction as in $La_{2-x}Sr_xCuO_4$. A second important difference between these two compounds concerns the crystal structure shown in Fig. 5. In Nd_2CuO_4 each Cu ion is surrounded by only four oxygen atoms, located in the CuO_2 plane; this contrasts with the CuO_6 octahedral structure in $La_{2-x}Sr_xCuO_4$. Further, this new system contains Nd^{3+} ions which may couple magnetically to the Cu^{2+}. Therefore it is essential to elucidate the role of the Nd moments in the overall magnetic properties, in particular their effect on the strongly correlated CuO_2 layers.

Static magnetic order in the electron-superconductors $Nd_{2-x}Ce_xCuO_4$ has been studied by muon spin rotation(11) and by neutron scattering(12,13). Nd_2CuO_4 shows well defined magnetic ordering at 255 K as shown in Figs. 5 and 6. The same spin arrangement is observed in Pr_2CuO_4 with T_N = 270 K, and it shows only one magnetic transition[14]. In Nd_2CuO_4, on the other hand, additional magnetic transitions occur at 80 K and 30 K, where the Cu^{2+} spins apparently re-orient twice. Then below 30 K, on cooling, the Nd^{3+} moments start to couple with the Cu^2 spins. Details of this behavior are now being examined.

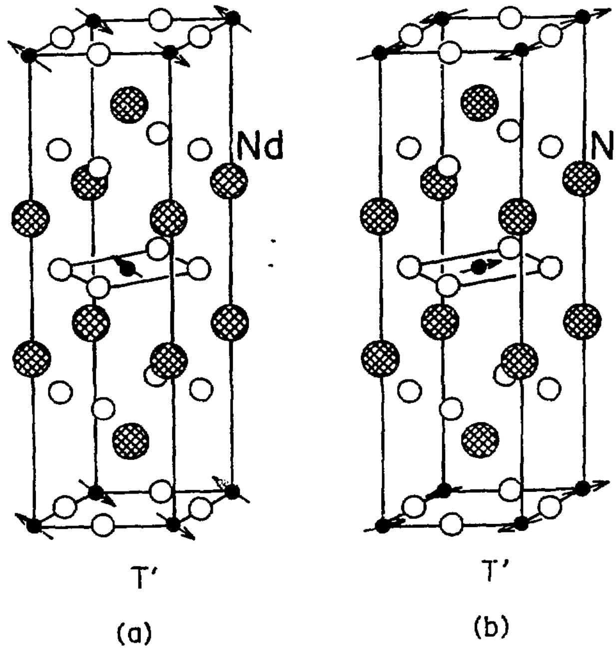

Fig. 5 Crystal structure and magnetic structures of the Cu^{2+} spins in Nd_2CuO_4. After Endoh et al[12]. (a) Above 80K to T_N, (b) between 80 and 30K.

On structural grounds alone, we anticipated the strong 2D magnetic coupling of CuO_2 layers in Nd_2CuO_4. As shown in the upper part of Fig. 6, the typical quasielastic 2D scattering peak is observed at $\zeta = 0.33$, the point at which the outgoing neutron wave vector k_f is exactly along c*. As described previously[1], this phenomena provides direct evidence for rapidly fluctuating 2D spin correlations.

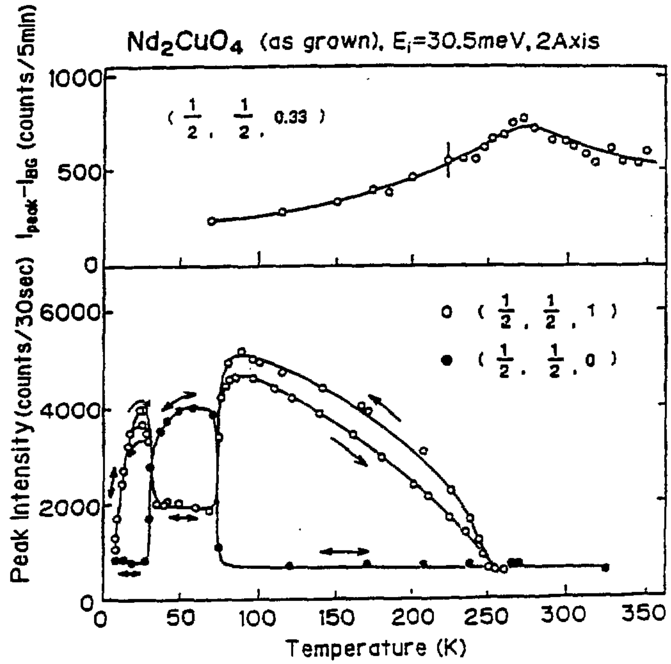

Fig. 6 Temperature evolution of the peak intensities of the 3D antiferromagnetic Bragg reflections and 2D rod. After Endoh et al[12].

The magnetic phase diagram of Ce^{3+}-doped compounds is not yet established. The doping creates additional electrons, and not oxygen holes, which may have strong frustrating effects on the Cu^{2+} ordering. Thus we expect that the Néel state will persist over a much wider range of Ce doping x than in the case of Sr doping. However, this picture has not yet been confirmed by either muon[11] or neutron experiments.

IV. Concluding Remarks

There is another very important class of high T_c oxides, $YBa_2Cu_3O_{6+x}$, and these have also been extensively studied by neutron scattering[15-18]. In this review, we limit ourselves to a brief remark. The phase diagram is well established for this system and is shown in Fig. 7 by smooth curves. Magnetic excitations

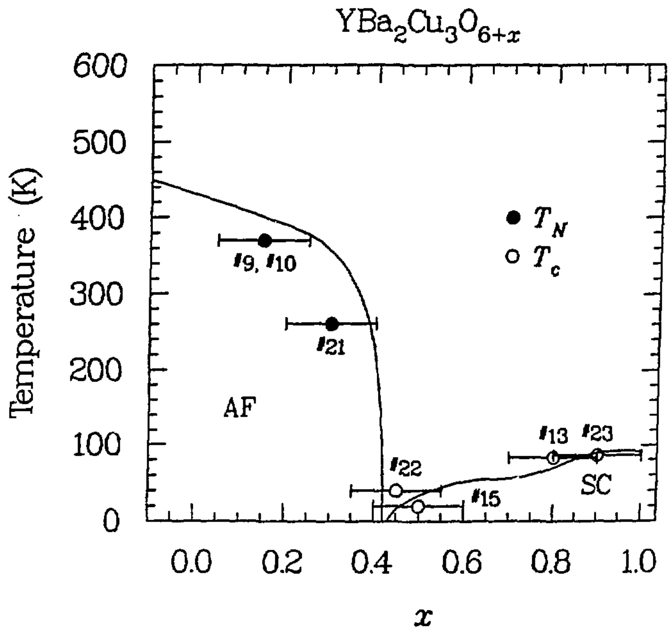

Fig. 7 $YBa_2Cu_3O_{6+x}$ phase diagram. Antiferromagnetic (AF) and superconducting (SC) phases, studied by Tranquada et al[18], are shown by sample numbers with horizontal bars.

in the tetragonal phase ($x \leq 0.4$) are now well characterized and they are very similar to the 2D correlations observed in La_2CuO_4. However, magnetic scattering for $x = 0.6 - 1.0$ has not been

6. Hidaka, Y. et al, Jpn. J. Appl. Phys. 26, L377 (1987), J. Cryst. Growth 85, 581 (1987).

7. Tanaka, I. and Kojima, H., Nature 337, 21 (1989).

8. Yoshizawa, H. et al., J. Phys. Soc. Japan 57, 3686 (1988).

9. Yasuoka, H., Imai, T. and Shimizu, T. Abstract, IBM Japan International Symposium on Strong Correlation and Superconductivity, May 21, 1989, Mt. Fujii, Japan. Imai, I. et al., Phys. Rev. B. (to be published).

10. Tokura, Y., Takagi, H. and Uchida, S., Nature 337, 345 (1989). Takagi, H., Uchida, S. and Tokura, Y., Phys. Rev. Lett. 62, 1197 (1989).

11. Luke, G. M. et al. Nature 338, 49 (1989).

12. Endoh, Y. et al. Phys. Rev. Lett. (submitted).

13. Akimitsa, J., private communication, May 1989.

14. Cox, D. E. et al., preprint May 1989.

15. Sato, M. et al. Phys. Rev. Lett. 61, 1317 (1988).

16. Rossat-Mignod, J. et al., J. Phys. (Paris) Colloq. 49, C8-2119 (1988), Jurgens, M. J. et al., Physica B 156 & 157, 846 (1989).

17. Brickel et al. Europhys. Lett. 4, 1189 (1987).

18. Tranquada, J. M. et al. Phys. Rev. B (to be published).

observed [17,18] in an energy range below 20 meV. What are the implications of these negative results?

One possiblity is that there are no magnetic moments present in the superconducting, orthorhombic phase of $YBa_2Cu_3O_{6+x}$. However, there is mounting experimental evidence against such a conclusion. We prefer to speculate that we have somehow missed the proper window for observing inelastic neutron scattering in (Q, ω, T) space. Further work is required to characterize properly the magnetic fluctuations in orthorhombic $YBa_2Cu_3O_{6+x}$ and any relationship they may have to superconductivity.

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

REFERENCES

1. Birgeneau, R. J., and Shirane, G., "Physical Properties of High Temperature Superconductors", D. M. Ginzberg, editor, p. 151 (World Scientific Publishing, 1989) and references therein.

2. Shirane, G. et al. Phys. Rev. Lett. 59, 1613 (1987). Endoh, Y. et al. Phys. Rev. B 37, 7443 (1988).

3. Birgeneau, R. J. et al. Phys. Rev. B 38, 6614 (1988) and B 39, 2868 (1989).

4. Thurston, T. R. et al., Phys. Rev. B. (to be published).

5. Shirane, G. et al. Phys. Rev. Lett. (to be published).