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Magnetic Correlations in Doped Transition Metal Oxides

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ABSTRACT

We review recent reactor- and spallation-source-based neutron scattering experiments on the magnetic fluctuations and order in a variety of doped transition metal oxides. In particular, data are shown for the NiO chain compound, $Y_{2-x}Ca_xBaNiO_5$, the two-dimensional cuprate superconductors $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$, and the classical three-dimensional "Mott-Hubbard" system $V_{2-y}O_3$.

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I. Introduction

For band metals and insulators, spin and charge fluctuations are different manifestations of the same electron-hole pair excitations, and therefore have similar spectra. In contrast, for charge-transfer and Mott-Hubbard insulators, spin fluctuations exist at essentially all energies down to zero, while the charge fluctuations exist only above a generally quite large (≥ 1 eV) gap energy. One of the most important challenges in the physics of solids is to understand if, and how, doping leads to a transition from the latter regime, where spin and charge excitations are of obviously different character, to the classical metal, where they are difficult to distinguish. In this paper, we review experiments which probe the magnetic sector in one-, two-, and three-dimensional oxides whose parents are Mott-Hubbard and charge-transfer insulators. We make no effort to survey or even provide references to the large body of theory on this subject because such theory is discussed in other chapters of this volume. The technique is neutron scattering, from which the cross-section is directly proportional to the Fourier transform in space and time of the two-spin correlation function. Other convenient features of neutron scattering include characteristic wavelengths of order interatomic spacings and an enormous range ($1 \mu\text{eV} - 0.5 \text{ eV}$) of accessible energy transfers. The technique is also a weak-coupling probe, which implies (i) that unless deliberate efforts are made (using grazing incidence or sponge-like samples with high surface to volume ratios), it has no surface sensitivity, and (ii) that a simple nonresonant Fermi's Golden Rule formula involving the precisely known magnetic dipole moment of the neutron, is sufficient not only to account for all data, but also to do so in absolute units familiar from bulk magnetometry. The only drawbacks of neutron spectroscopy are the scarcity of sources

and the scarcity of neutrons, relative to e.g. photons from lasers and synchrotrons, at the few sources which exist. The latter constraint means that unusually large single crystals (volumes $\geq 1 \text{ cm}^3$) need to be used and even then, long counting times are required to accumulate statistically meaningful data. We are looking forward to a new generation of sources (the U.S. Advanced [Reactor] Neutron Source and the European Spallation Source) and instruments which together should improve the efficiency of the technique by several orders of magnitude.

II. Three Dimensions, V_{2-y}O_3

Until the discovery of superconductivity in the layered cuprates, V_2O_3 and its doped derivatives constituted the most intensely studied "Mott-Hubbard" system.^(1,2) Figure 1 shows the phase diagram for V_{2-y}O_3 .⁽³⁾ Both pressure and vanadium deficiency (indistinguishable from oxygen excess) makes it possible to transform, via a first order transition, the antiferromagnetic insulator (AFI) into a metal. The spin waves and magnetic order in the AFI state were extensively characterized a long time ago.⁽²⁾ More recent experiments⁽³⁾ have searched for magnetic correlations in the metallic state with the surprising outcome that the low-temperature transport and thermodynamic anomalies⁽⁴⁾ which occur in this state, when it is stable to $T = 0$, are due to the onset of magnetic order which appears to be totally unrelated to the order in the AFI. Indeed, the ordering is incommensurate, with a propagation vector along the c-axis, in contrast to the commensurate $(1/2 \ 1/2 \ 0)$ (hexagonal notation) ordering vector for pure V_2O_3 . Searches⁽⁵⁾ for magnetic fluctuations — e.g. broadened spin waves — which might be inherited from the AFI failed, as did analogous surveys to find traces of the metallic

antiferromagnetism in the AFI phase. Another important result is that the order in the metal has all of the features of a small-moment spin-density wave derived from a Fermi surface instability. Figures 2 and 3 show the most dramatic evidence for this. In Fig. 2, the rise in staggered magnetization coincides with a Cr-like rise,⁽⁶⁾ rather than a spin-disorder scattering peak, in the resistivity at T_N . Figure 3 demonstrates that the dominant magnetic fluctuations are not conventional spin waves, but rather modes — most likely electron-hole pairs — with an $\omega \rightarrow 0$ length scale ($14 \pm 4 \text{ \AA}$) much smaller than that ($> 10^3 \text{ \AA}$) characterizing the static order. The slow variation⁽³⁾ of the incommensurate ordering vector with doping suggests that the nesting instability involves a large Fermi surface.

III. Two Dimensions — $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$

These compounds differ significantly from V_{2-y}O_3 in that the antiferromagnetic transitions of the insulating parents are second order, and that upon doping, the magnetic fluctuations are generally peaked in the vicinity of the magnetic ordering vector ((π, π) in square lattice notation) for the insulators. Figure 4 summarizes how the magnetic response⁽⁷⁾ in $Q-\omega$ space evolves with x in $\text{La}_{2-x}(\text{Sr}, \text{Ba})_x\text{CuO}_4$. The pure compound displays a cone, emanating from (π, π) , of long-lived spin waves which are replaced first (near the metal-insulator transition) by over-damped commensurate fluctuations, and subsequently by Cr-like incommensurate fluctuations. Of course, at high ω , the incommensurate peaks merge and the scattering profile is indistinguishable from a single broad peak centered at (π, π) . Figure 5 shows $\hbar\omega = 50 \text{ meV}$ data from the MARI time-of-flight instrument at ISIS which clearly illustrates this point for $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$.

For low and intermediate x , Fig. 4(b) gives a fair description of the magnetic response⁽⁸⁾ in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. However, to date, no peaks with incommensurate in-plane coordinates have been found in any part of the phase diagram, including that for $x \approx 1$ where the best superconducting properties are found. Indeed, a peculiar resonance^(8,10) at 41 meV and peaked at (π, π) is the most apparent feature of the magnetic response $\chi''(Q, \omega)$ for material with a 90K transition temperature. The evidence that the peak is magnetic is the polarized neutron scattering data⁽¹⁰⁾ reproduced in Fig. 6, which also shows the astonishing growth which it undergoes on reducing T below T_c . While it grows at (π, π) , it also narrows, as established in constant-energy scans, shown in Fig. 7, in the basal plane. We are not aware of a theory for the 41 meV peak and its evolution below T_c .

IV. One Dimension — $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$

We conclude this survey with a brief discussion of the topic about which there is both the least experimental data and the highest chance for theoretical progress, namely one-dimensional systems. $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$ is the only one-dimensional transition metal oxide (TMO) for which transport and x-ray absorption⁽¹¹⁾ measurements have shown that carriers are introduced onto the TMO chains by off-chain donor atoms (Ca), just as they are introduced onto the copper oxide planes by off-plane donors in the high-temperature superconductors. However, what differentiates this system from the doped cuprates and V_{2-y}O_3 is that the parent compound, YBa_2NiO_5 , actually has a disordered Haldane rather than an ordered Neel ground state.⁽¹²⁾ Thus, $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$ represents the first gapped spin liquid, of any dimension, into which charges can be introduced.

Figure 8 shows how off-chain doping with Ca introduces magnetic states⁽¹¹⁾ with energies below the ~ 8 meV Haldane gap, and also demonstrates that such states are not introduced to the same extent by on-chain substitution of nonmagnetic Zn for the ($S = 1$) Ni atoms. The amplitude of the states within the gap is considerably larger than what one would associate with the $S = 1/2$ holes inserted onto the chains by the donors. This means that the charges are dressed by a large disturbance in the spin background. Presumably, this is also the case in the higher-dimensional doped transition metal oxides. However, the advantage of the 1D spin liquid for making the dressing effect explicit is that the parent compound has a gapped spin spectrum.

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FIGURE CAPTIONS

- Fig. 1: Phase diagram for $V_{2-y}O_3$. From ref. [3].
- Fig. 2: Temperature dependence of staggered magnetization and excess resistivity in metallic spin density wave phase. From ref. [3].
- Fig. 3: Constant energy scans for metallic $V_{2-y}O_3$ with $y=0.027(3)$ and $T=1.5K$. From ref. [3].
- Fig. 4: Evolution of $\chi''(Q,W)$ across the $La_{2-x}(Sr,Ba)_x CuO_4$ series. From ref. [7].
- Fig. 5: Constant energy scan through (π,π) point for $La_{1.86}Sr_{0.14}CuO_4$. Data taken using MARI instrument at ISIS. From ref. [13].
- Fig. 6: Polarized and unpolarized neutron scattering spectra for $YBa_2Cu_3O_{6+x}$ superconductor with $x \approx 1$. From ref. [10].
- Fig. 7: Constant energy scans showing that 41 meV peak narrows considerably on going below the $T_c = 90K$ of $YBa_2Cu_3O_{6+x}$ with $x \approx 1$. From ref. [10].
- Fig. 8: Spectra of magnetic excitations in the pure, Ca-doped, and Zn-diluted linear chain compound $Y_{2-x}Ca_xBaNi_{1-y}Zn_yO_5$. From ref. [11].

FIG. 1

FIG. 2

Intensity (Counts pr. min.)

FIG. 3

(a)

(b)

(c)

FIG. 4

FIG. 5

FIG. 6

FIG. 8

FIG. 7