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Magnetic coherence in the transition metal oxides

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Abstract

We review neutron scattering experiments which address the coherence of the ground states of four transition metal oxides. The states considered include some which, at the qualitative level, can be described classically and others which require a quantum mechanical understanding. In the former category are the antiferromagnetism of $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$ and the peculiar ground state of the Kagomé compound $\text{SrCr}_9\text{Ga}_{12-9p}\text{O}_{19}$, which is neither an antiferromagnet nor a spin glass. The latter category contains the Haldane quantum spin liquid, seen very clearly in the linear-chain compound Y_2BaNiO_5 , and the superconducting state of $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$.

Keywords: Kagomé lattice; Spin liquid; Transition metal oxide

1. Introduction

The transition metal oxides are known for displaying many different types of magnetic order, ranging from antiferromagnetism in the case of many insulating compounds to ferromagnetism in the metallic manganites. More exotic ground states, not involving magnetic order but with serious implications for the spins are also possible. The most popular of these states is the superconducting state,

as found in the high-transition temperature oxides of copper, where electrons of opposite spin are paired in a coherent fashion to yield the ground-state wave function. Less well-known states with non-trivial spin correlations are the quantum spin liquid Haldane state and the peculiar glassy ground state of geometrically frustrated magnets. In the present paper, we review briefly how we can observe the different magnetic coherence effects by measuring the magnetic fluctuations in transition metal oxides with different ground states. This is not an exhaustive survey. Instead, we describe one recently examined material in each category. Specifically, we begin by describing the spin waves

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in the bilayer antiferromagnet, $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$, which is also the parent of the most studied high- T_c superconductor, $\text{YBa}_2\text{Cu}_3\text{O}_7$. We then move to the seemingly classical glassy order of the frustrated layer compound SCGO. The last two sections are devoted to the quantum coherence in the $S = 1$ chain compound Y_2BaNiO_5 and the superconducting phase of $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$.

2. Spin waves in $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$

The fundamental constituents of this material are double layers of CuO_2 , with antiferromagnetic coupling between the copper ($S = \frac{1}{2}$) ions and their nearest in-plane and adjacent plane neighbors. The magnetic order is correspondingly simple and long-ranged, with each moment antiparallel to its neighbors [1]. Thus, at the qualitative level, $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$ is a classical antiferromagnet. The coherent excitations should then be harmonic spin waves, which are classified as acoustic and optic depending on whether neighboring moments are rotating in the same or opposite directions in adjacent layers. We have used inelastic neutron scattering [2] to measure the dispersion relation and amplitudes of the spin waves. The instrument chosen was the HET time-of-flight spectrometer at the ISIS pulsed spallation source of the Rutherford-Appleton Laboratory. Figs. 1(b)–(e) show data in the form of constant-energy transfer $\hbar\omega$ scans crossing the dispersion surface. For the smallest $\hbar\omega$ (frame(e)), there is a broad maximum, due to unresolved counterpropagating magnons, centered at $(0.5, 0.5, 1)$ (magnetic Bragg scattering in our tetragonal notation occurs at $(0.5, 0.5, 1)$). As $\hbar\omega$ rises and the dispersion surface opens up, there is a noticeable splitting in the maximum. Frame (a) shows the optic and acoustic dispersion curves deduced from all of our data, including some (not displayed) for lower $\hbar\omega$. These curves yield values for the in-plane and out-of-plane exchanges, which are 125 ± 5 and 11 ± 2 meV, respectively. In addition to obtaining the first complete dispersion relations for the parent antiferromagnet of YBaCu_3O_7 , we have been the first to establish the absolute

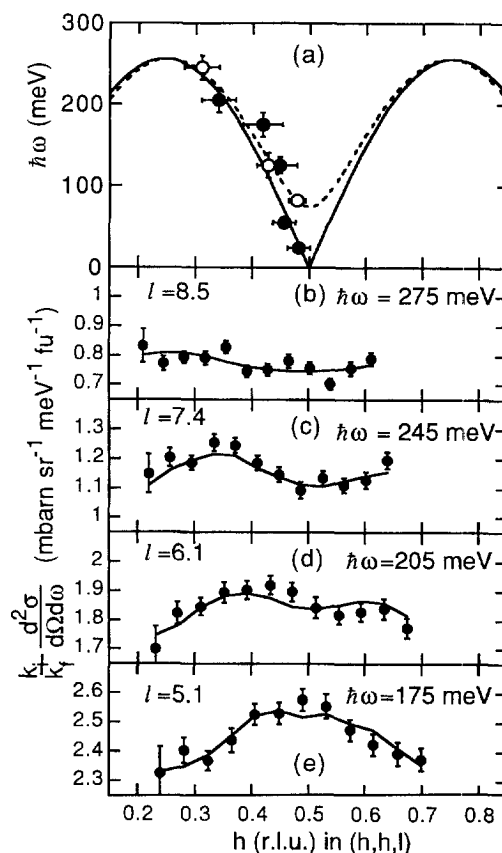


Fig. 1. (a) Spin wave dispersion relation for $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$. (b)–(e) Constant-energy cuts through spin waves; solid lines are from harmonic spin wave theory, corrected for instrumental resolution effects; from Ref. [2].

spin wave amplitudes in this material. Fig. 2 shows the frequency-dependent amplitudes for the optic and acoustic modes. Note the abrupt appearance of optic mode scattering at the gap frequency of 75 ± 5 meV. The solid lines are derived from linear spin wave theory, with the amplitude factor simply reduced by 60% from the classical value. We have obtained a similar result for the spin waves in the single-layer compound La_2CuO_4 [3]. Thus, while the classical description accounts qualitatively for the properties of these $S = \frac{1}{2}$ systems, zero point (quantum) fluctuations must be included to give a quantitative description.

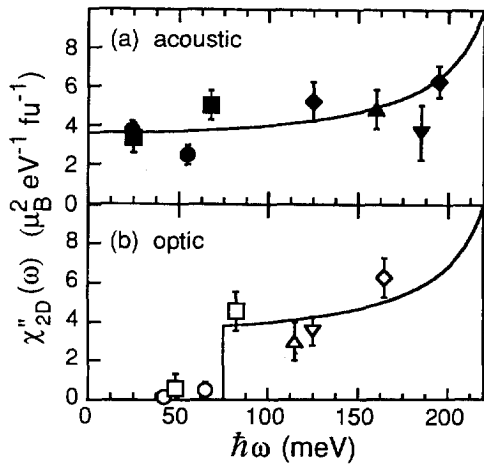


Fig. 2. Local magnetic susceptibility obtained as a function of energy transfer for spin waves in $\text{YBa}_2\text{Cu}_3\text{O}_{6.15}$. Solid lines are from harmonic spin wave theory; from Ref. [2].

3. Hidden classical order in a Kagomé compound

$\text{SrCr}_p\text{Ga}_{12-9p}\text{O}_{19}$ (SCGO) has been a very popular compound because its basic building blocks are geometrically frustrated Kagomé layers [4]. The bulk behavior is reminiscent of spin glasses, and there are no observable Bragg peaks indicative of long-range magnetic order at low temperatures. Instead, high-resolution inelastic neutron scattering [5], performed using the IRIS back-scattering spectrometer at ISIS, reveals a resolution-limited elastic peak, corresponding to spin correlations with lifetimes in excess of 0.3 ns. While this is precisely what one expects for conventional spin glasses, the momentum dependence of the zero-frequency structure factor $S(Q, \omega = 0)$ as well as the excitation spectrum take surprising forms. In particular, $S(Q, \omega = 0)$ appears to vanish as Q approaches zero. A structure factor which disappears in the long-wavelength limit is a well-known property of ordered antiferromagnets and not expected for randomly frozen spin glasses. For a system such as SCGO, which displays no magnetic Bragg peaks, it implies a magnetic coherence due to the (near) absence of spins not belonging to clusters which, when considered whole, are singlets. The observed magnetic dynamics (Fig. 3) provide further evidence for such a state. The local

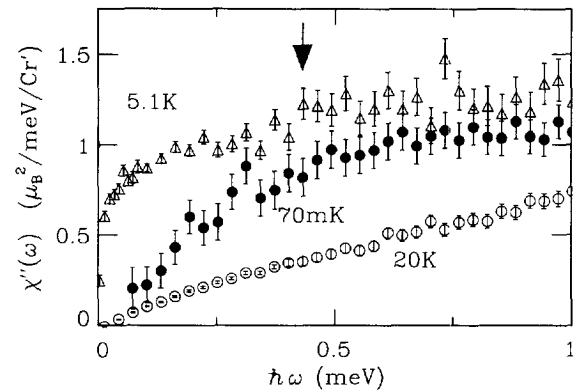


Fig. 3. Local susceptibility for Kagome compound $\text{SrCr}_p\text{Ga}_{12-9p}\text{O}_{19}$ ($p = 0.92(5)$); from Ref. [5].

susceptibility $\chi''(\omega)$ varies linearly with ω for $\hbar\omega < 0.4$ meV, in contrast to the ω -independence found for conventional spin glasses. Thus, the flat distribution of two-level systems generally thought to correspond, in the $\omega > 0$ limit, to “loose” spins is absent in this geometrically frustrated transition metal oxide with magnetic coherence, which places it between spin glasses and antiferromagnets.

4. A one-dimensional spin liquid

The materials described so far (in Ref. [2, 3]) can be understood in classical terms. We turn now to a system whose ground state has no obvious classical antecedent and is best described as a quantum spin liquid. Y_2BaNiO_5 is a compound whose most prominent feature is NiO chains. The Ni ions carry spin $S = 1$, and the superexchange via the oxygen ions intervening between neighboring Ni ions is antiferromagnetic. The resulting ground state is the Haldane state, a singlet separated by a gap from a continuum of triplets [6]. Our data, collected at the NIST reactor, show that the continuum has a well-defined dispersion, consistent with earlier work. What is especially important is that the excited states have the long-range coherence which characterize the Bragg peaks and spin waves of ordinary magnets, even though the instantaneous two-spin correlation function is short-ranged, as for

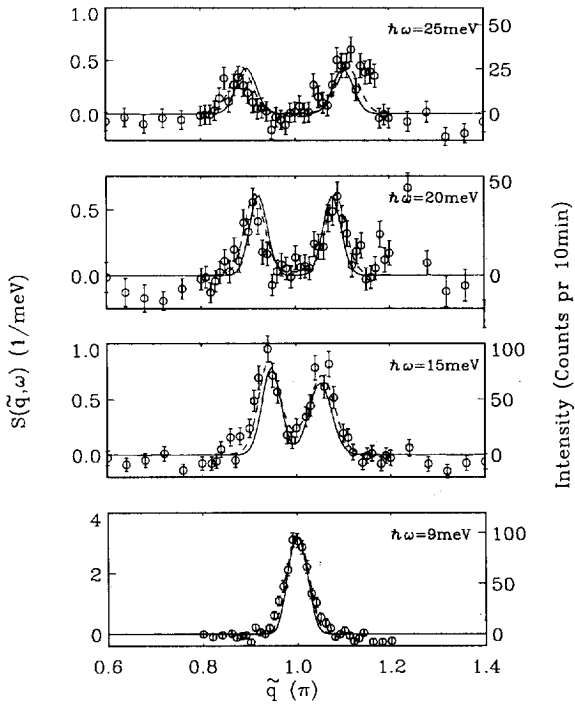


Fig. 4. Constant-energy scans through Haldane continuum in Y_2BaNiO_5 ; from Ref. [7].

a proper liquid. Fig. 4 illustrates this coherence very graphically, in the form of constant-energy scans through the Haldane continuum. Of greatest significance is the sharp nature of the peaks derived from counterpropagating excited states. The corresponding (quantum) coherence length is in excess of 100 \AA . We conclude that the phase coherence length of the Haldane state in this material is itself in excess of 100 \AA .

5. Magnetic spectroscopy of the superconducting state of $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$

$\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ is one of the simplest high- T_c superconductors, in that the Cu ions occur in essentially isolated CuO_2 planes. The band structure and Fermi surface are correspondingly simple, which should ease the understanding of the magnetic correlations in both the normal and superconducting states. The major features in reciprocal space are

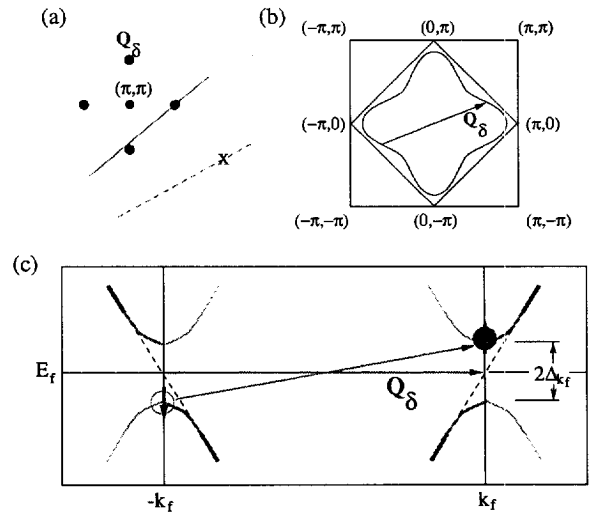


Fig. 5. Schematic, explained in text, of the magnetic response in the normal and superconducting states of $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$; from Ref. [10].

incommensurate peaks (see Fig. 5(a) and Ref. [8]) near the (π, π) point at which the magnetic superlattice peaks appear in the insulating parent, La_2CuO_4 . It is very natural to understand these peaks as arising from transitions across the Fermi surface, as Fig. 5(b) indicates. The interpretation in terms of electron-hole pairs is directly verified by the fact that at low frequencies [9], the entry into the superconducting state much reduces the incommensurate peaks, an effect which follows immediately because superconductivity inserts a gap into the electron-hole pair continuum, at least for most values of the momentum Q (see Fig. 5(c)). From the magnetic point of view, we are therefore faced with a $\chi''(Q, \omega)$ with a (possibly) Q -dependent gap function, somewhat analogous to what is seen for a Y_2BaNiO_5 . If the total moment sum rule is to be obeyed, there should be some additional spectral weight above the gap to compensate for the loss of spectral weight below the gap. In a recent experiment at Risø [10], we have found the additional weight. In particular, Fig. 6 shows the temperature dependence of the scattering at the incommensurate peak position for several $\hbar\omega$. At $6.1 \text{ meV} < 2\Delta = 3.5k_B T_c$ ($T_c = 35 \text{ K}$), the growth in normal state intensity is reversed by

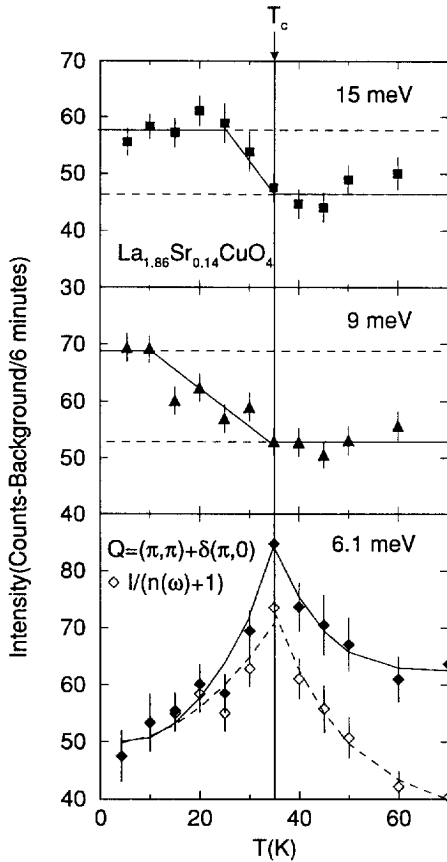


Fig. 6. Temperature dependence of magnetic response measured at incommensurate point (see Fig. 5(a)) for various energy transfers; from Ref. [10].

entry into the superconducting state. On the other hand, for $9 \text{ meV} \approx 2\Delta$ and $15 \text{ meV} > 2\Delta$, the signal actually begins to grow for $T < T_c$. A very natural question, especially in light of the long-range quantum coherence of the excitations above the Haldane state, is to ask about the coherence of the states added because of the appearance of the BCS condensate. Fig. 7 provides a very remarkable answer, namely that the additional states have resolution-limited coherence near threshold, i.e., for $\hbar\omega \cong 2\Delta$. This implies a coherence length in excess of 50 \AA , which is longer than all other magnetic and superconducting length scales, with the exception of the superconducting phase coherence length, for $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$.

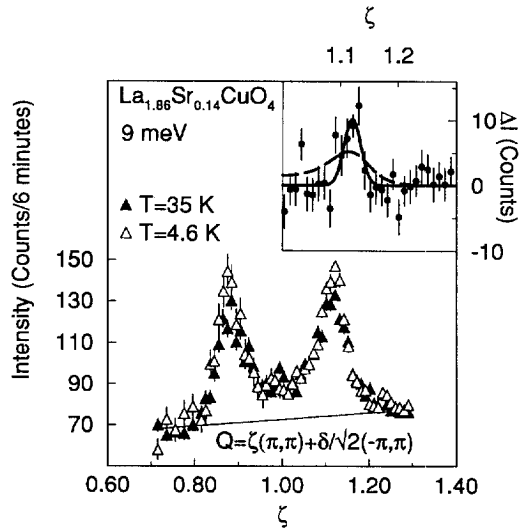


Fig. 7. Magnetic response measured as a function of momentum for energy transfer fixed at 9 meV at $T = T_c = 35 \text{ K}$ and low temperature. Inset shows difference between high- and low-temperature results; from Ref. [10].

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