SPIN CORRELATIONS AND IMPURITIES IN A ONE-DIMENSIONAL QUANTUM SPIN LIQUID

Though all solids contain electrons with spin-induced magnetic moments few materials will actually cling to your refrigerator door. The quantum theory of atoms explains that matter is generally non-magnetic because electron spins form non-magnetic (singlet-) states in the filled electronic shells of individual atoms. Here we discuss a new class of materials wherein singlet formation takes place between rather than within atoms to yield a macroscopic spin-less "molecule". We explore the magnetism of Y_2BaNiO_5, in which magnetic Ni^{2+} atoms interact antiferromagnetically (AFM) through intervening O^{2-} atoms to form spin chains.

Inelastic magnetic neutron scattering is a powerful probe of spin chains. The open circles in Fig. 1 show the low temperature equal time spin correlation function versus wave-vector transfer along the chain. In contrast to Néel AFM's that develop Bragg peaks when long range order develops at a second order phase transition, Y_2BaNiO_5 has no magnetic phase transition and our snap-shot of the spin configuration reveals short-range AFM order with a dynamic correlation length $\xi = 4.3(6)$ lattice spacings. Still, this is not a thermally disordered paramagnet. The solid symbols in Fig. 1 show that at fixed energy transfer there are sharp peaks in the wave-vector dependence of the dynamic correlation function that allows us to put a lower limit of 50 lattice spacings on the coherence length for magnetic excitations.

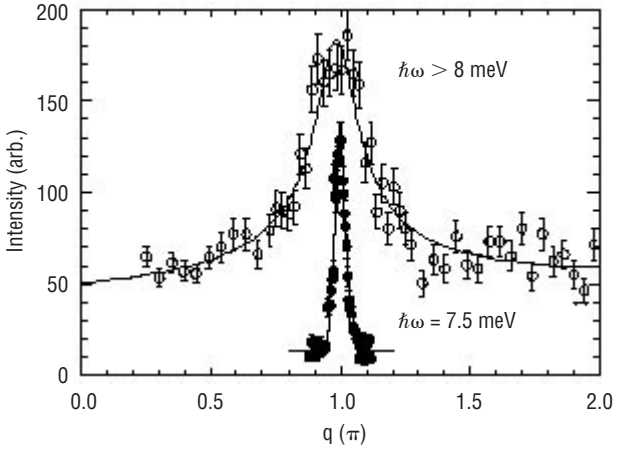

FIGURE 1. Neutron scattering from Y_2BaNiO_5 at T=10 K. Open symbols show energy-integrated data probing equal-time correlations. Solid symbols show a constant-$\hbar\omega$ scan at the gap energy.

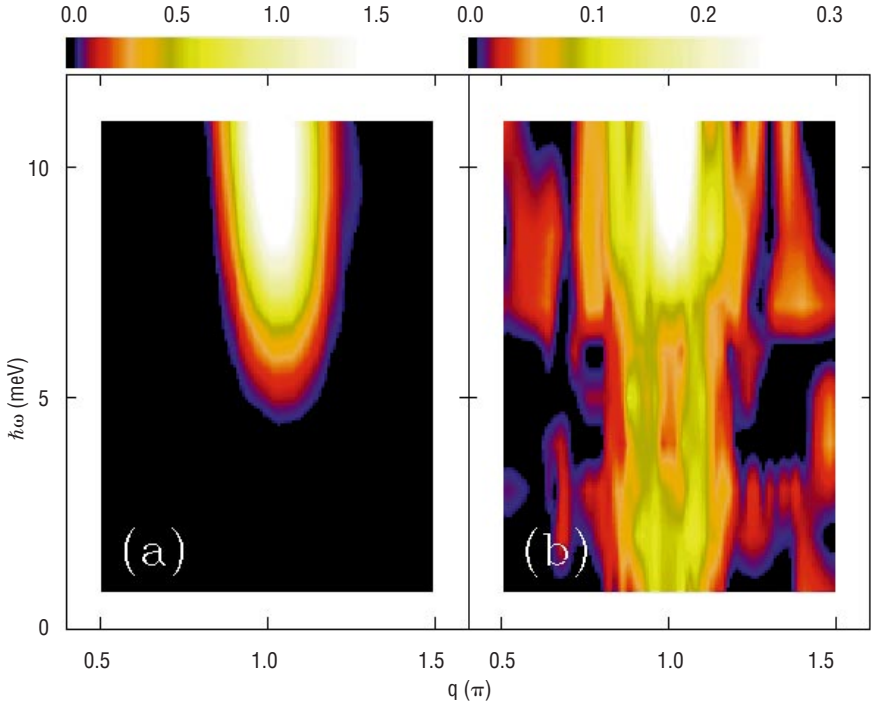

FIGURE 2. Contour map of low energy magnetic neutron scattering at T=10 K from (a) pure Y_2BaNiO_5 and (b) $Y_{1.905}Ca_{0.095}BaNiO_5$. The MARI spectrometer at the ISIS facility in the UK was used for (a) while BT2 and SPINS at the NIST were used for (b).

Guangyong Xu
Department of Physics and Astronomy
Johns Hopkins University
Baltimore, MD 21218

G. Aeppli, P. Bischer, and M. Treacy
NEC Research Institute
4 Independence Way
Princeton, NJ 08540

C. Broholm
Department of Physics and Astronomy
Johns Hopkins University
Baltimore, MD 21218
and
NIST Center for Neutron Research
National Institute of Standards and Technology
Gaitherburg, MD 20899

J. F. DiTusa
Department of Physics
Louisiana State University
Baton Rouge, LA 70803

C. D. Frost
ISIS Facility
Rutherford Appleton Laboratory
Chilton, Didcot, Oxon OX11 0QX, UK

T. Ito and K. Oka
Electrotechnical Laboratory
Tsukuba 305, Japan

H. Takagi
The Institute of Solid State Physics
University of Tokyo, Roppongi
Tokyo 106 Japan

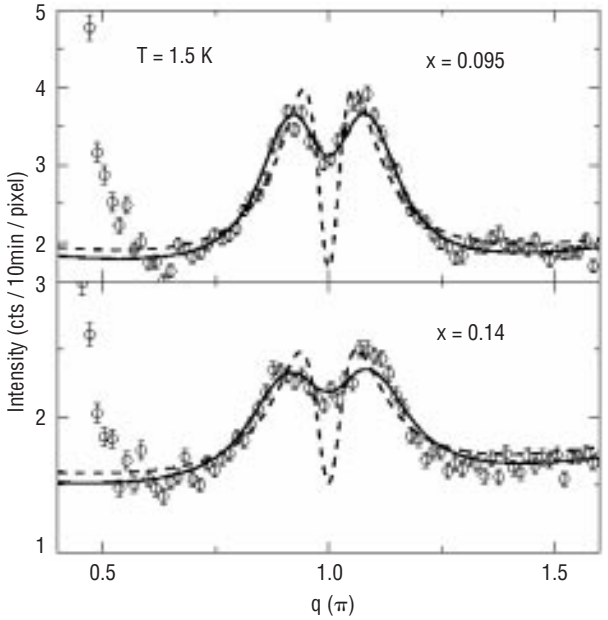

FIGURE 3. Q-dependence of energy integrated intensity from $Y_{2-x}Ca_xBaNiO_5$ with x=0.095(5) and x=0.14(1). The dashed line is a single impurity model. The solid line is a random impurity model.

Figure 2 (a) shows another important feature of the AFM spin-1 chain: There is a gap in the excitation spectrum separating the ground state from excited states. The energy gap is the cost of creating a magnetic wave packet on the spin chain.

There are interesting effects of substituting Ca^{2+} for Y^{3+}. The extra hole occupies the π-orbital on the super-exchange mediating oxygen site and leads to a ferromagnetic (FM) impurity bond in the otherwise AFM spin chain. Figure 2(b) shows that such doping yields new sub-gap excitations. The extra magnetic scattering takes the form of a double ridge versus energy, which indicates that we are dealing with slow fluctuations of a rigid composite object. The wave-vector dependence provides valuable but ambiguous information about the real space structure of this object. A natural first interpretation would be that the holes have ordered to yield a new incommensurate periodicity. To explore this possibility Fig. 3 shows a comparison of high statistics $\hbar\omega$-integrated data for samples with Ca concentrations differing by almost 50%. The absence of a significant shift in the peak positions rules out hole ordering.

We propose instead that the double peak in Fig. 3 is the magnetic form factor of a hole in a quantum spin liquid. Consider first the dilute and static hole limit of a single FM impurity bond on the spin chain. The energy of the macroscopic spin singlet has increased and a degenerate magnetizable state is now the ground state. If we assume that the spin disturbance associated with the impurity bond decays away from the impurity then we can show that the corresponding form-factor has the general features observed in the scattering data. Denote by M(q) the form factor for the spin density which develops to the right of an impurity bond. The corresponding form-factor for the disturbance to the left of the impurity is the complex conjugate, M*(q). The form factor for the combined object becomes $F_{\pm}(q) = M(q)e^{iq/2} \pm M^*(q)e^{-iq/2}$. When the chain end spin degrees of freedom are antiferromagnetically combined, corresponding to the negative sign in this equation, $|F_-(q)|^2$ becomes the conventional structure factor for the quantum spin liquid in Fig. 1. However, when the chain ends are stitched together ferromagnetically as we should expect for FM impurity bonds we have $F_+(q) = 2\mathrm{Re}\{M(q)e^{iq/2}\}$. This function vanishes for $q = (2n+1)\,\pi$ because M(q) is real for $q = n\pi$ and the result is a notch at $q = n\pi$ as shown by the dashed lines in Fig. 3. To account for the finite intensity between the peaks we need to consider the finite density of impurity bonds. Neighboring holes arranged at random break inversion symmetry about individual holes and this brings back intensity at $q = \pi$. The solid lines in Fig. 3 correspond to an analytical expression for the scattering from a distribution of uncorrelated asymmetric impurities with exponentially decaying spin densities. The fact that the impurity scattering is distributed over a range of energies in Fig. 2(b) indicates that the holes are moving or more likely that neighboring bound states interact.

The significance of all this is that we have directly measured the spin wave function associated with a bond reversing hole impurity in a quantum spin liquid. Because our raw data are so similar to the scattering data from doped copper oxide superconductors our results suggest that hole form-factor effects may also be important for interpreting those data.

REFERENCES

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