Coexistence of Ising cluster excitations and intracluster excitations in diluted Ising magnets

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To elucidate magnetic excitations in diluted Ising magnets over the entire magnetic concentration, we performed neutron inelastic-scattering experiments on two-dimensional diluted Ising magnets $Rb_2Co_cMg_{1-c}F_4$, ($c=0.1,0.2,0.3,0.4,0.5,0.58$), whose magnetic concentration was below the percolation threshold. We found that Ising cluster excitations and intracluster excitations coexist over the entire concentration region, and that the scattering intensities from these different origins change with the concentration.

Magnetic excitations in diluted Ising antiferromagnets have so far been elucidated in systems whos magnetic concentration is beyond or close to the percolation concentration. A typical result was obtained in two-dimensional antiferromagnets, $Rb_2Co_cMg_{1-c}F_4$ ($c=0.89$, 0.70, and 0.58), using a neutron inelastic scattering technique.[1,2] In Ising systems with nonmagnetic impurities, the excitation energies split into z dispersionless branches, z being the coordination number. The excitation energies $[2zIS, 2(z-1)IS, 2(z-2)IS, \ldots, 2IS]$ correspond to the Ising energies required for a transition from the $S^z=\pm S$ to the $S^z=\pm(S-1)$ state, where I is a diagonal part of the exchange constants. In diluted magnets there are z different local configurations in which the number of magnetic neighbors is different, thus producing z different molecular field energies. These are known as so-called *Ising cluster excitation*. In this way we now have an understanding of the spin dynamics in diluted Ising antiferromagnets with high magnetic concentration.

On the other hand, in highly diluted systems whose magnetic concentration is far below the percolation threshold, we can easily imagine what happens in the inelastic neutron scattering spectrum, though no data are available yet. In highly diluted magnets, only small magnetic clusters exist which comprise exchange-coupled two magnetic atoms (2-spin cluster), three magnetic atoms (3-spin cluster), and so forth. The population probability of these clusters depends, of course, on the overall magnetic concentration (c). In these paramagnetic systems we can determine *intracluster excitation* energies which are exactly solvable by diagonalizing the Hamiltonians.

Upon increasing the magnetic atoms, more complex and larger clusters appear in the systems; eventually the energy spectrum might seem to become more complex due to contributions from many clusters with different constituent magnetic atoms. However, upon further increasing the number of magnetic atoms up to the precolation concentration, we obtain only four Ising cluster energies in a square lattice system. We must answer the following question: how does the energy spectrum change the magnetic concentration in diluted Ising magnets below the percolation threshold?

In order to resolve the magnetic excitations over the entire concentration region in a diluted Ising magnet, we have performed inelastic neutron scattering experiments on two-dimensional diluted using magnets, $Rb_2Co_cMg_{1-c}F_4$, whose magnetic concentrations (c) were 0.1, 0.2, 0.3, 0.4, 0.5, and 0.58; the percolation concentration of the square lattice is 0.593. Neutron scattering experiments are performed on a LAM-D spectrometer installed at the pulsed spallation neutron source at National Laboratory for High Energy Physics (KENS/KEK), Tsukuba.[3] The LAM-D spectrometer has large analyzer mirrors made of pyrolytic graphite with a fixed outgoing neutron energy of 4.62 meV. Since the typical energy resolution is about 3% of the incident neutron energy, the energy resolution becomes worse with increasing transferred energy. As is evinced in previous triple-axis experiments concerning $Rb_2Co_{0.58}Mg_{0.42}F_4$,[2] magnetic scattering from systems with a concentration near to the percolation threshold is incoherent. We could thus integrate the scattered intensities over four analyzer-detector systems set at different scattering angles. The measurements were performed on polycrystalline samples with a volume of about 2 cm^3, both at 25 K and room temperature. The room-temperature experiments were performed in order to differentiate the magnetic contributions from phonons. The concentration fluctuations within samples with higher concentrations than the percolation threshold had been estimated by measurements of the transition temperature. The estimated variations of the Co^{2+} concentration were within 1%. We can give no reason for the present materials having bigger concentration fluctuations than 1%. The typical time required to accumulate inelastic data was about 4 h on the average.

In Figs. 1(a) and 1(b), the energy spectras taken at 25 K are depicted. In the data for the $c=0.58$ sample we can see four Ising cluster energies at 7.0, 13.6, 19.9, and 26.0 meV, in accord with previous triple-axis experiments on a sample with the same concentration.[2] On the other hand, in the sample with the lowest magnetic concentration

($c=0.10$), intracluster transition energies of 3.1 and 7.8 meV from the 2-spin clusters at 13.0 meV from the 3-spin clusters are well resolved, as is expected. These transition energies can easily be obtained by solving the 4×4 and 8×8 matrices of the exchange-coupled Hamiltonians. It should be noted here that the diagonal part of the exchange constants used in the calculations is $I=7.0$ meV and that the off-diagonal part is $J=3.9$ meV, slightly different from those of a pure material, Rb_2CoF_4 ($I=7.73$ meV and $J=4.27$ meV).[4] The modified exchange constant of I is closely in accord with the observed lowest Ising cluster energy of 7.0 meV. With an increasing number of magnetic atoms, the intensities form intracluster excitations increase; they then decrease, since the population probability of the 2-spin and 3-spin clusters increases and takes a maximum at $c\sim0.2$ and $c\sim0.3$, respectively, and then decreases. From a simple calculation we can see that the population probability of the 2-spin clusters exceeds that of the other clusters over the entire concentration region. Therefore, the intensities from 2-spin clusters dominate those from other clusters.

Next, we carefully consider the concentration variation of the scattering intensities, particularly in the energy region between 2 and 9 meV. Among the three peaks in this region, two peaks at 3.1 and 7.8 meV are form intracluster excitations of the 2-spin clusters and one peak at 7.0 meV is from the lowest Ising cluster excitation. The intensity from this Ising cluster excitation can be observed even at a low magnetic concentration of $c=0.20$, and increases with increasing magnetic concentration. On the other hand, the intensities of the intracluster excitations at 7.8 meV relative to those at 7.0 meV decreases with increasing c, and eventually become less at around $c=0.55$. The signals from intracluster excitations in 2-spin and 3-spin clusters can be clearly seen at 3.1, 7.8, and 13.0 meV, even in a highly concentrated system with $c=0.58$. These signals were missed in the previous triple-axis experiments[2] due to the low counting statistics and low energy resolution.

At first glance, these results are surprising. Two kinds of magnetic excitations with the different physical origins coexist over the full concentration region. At lower c, i.e., $c=0.20$, the Ising cluster energy can clearly be observed at 7.0 meV, even though the 2-spin clusters are dominant in this concentration region.

An interpretation of the findings involving mixed signals from two different origins is as follows. The intracluster energies of small magnetic clusters are exactly calculated by using the corresponding Hamiltonians and, therefore, arise from quantum-mechanical origin; oppo-

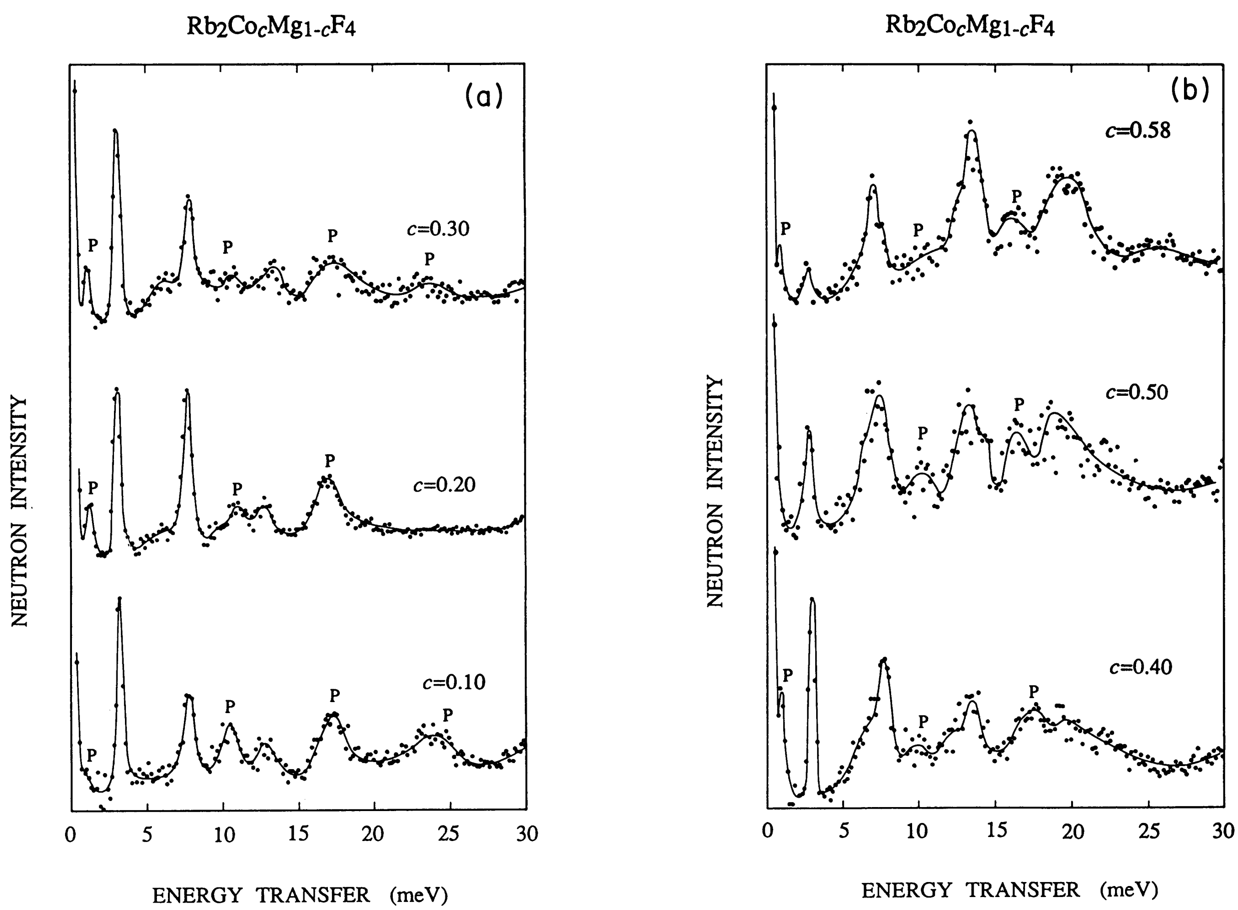

FIG. 1. Inelastic neutron scattering at 25 K from $Rb_2Co_cMg_{1-c}F_4$. (a) $c=0.1$, 0.2, and 0.3. (b) $c=0.4$, 0.5, and 0.58. The experiments were performed with a fixed outgoing neutron energy of 4.62 meV. The solid lines are guides to the eye. P denotes the phonon peaks identified from the room-temperature measurements.

sitely, the Ising cluster energies are from statistical or thermal effects, like spin-wave excitations. The latter has a strong temperature dependence and at low temperature, where the thermal correlation length can exceed the interatomic distance, Ising cluster excitation can occur even in 2-spin clusters. At 25 K this scenario can be realized and the well-defined Ising cluster energies were observed, even in such a low-concentration material.

From these observations it is obvious that we now have complete information concerning magnetic excitations in diluted Ising magnets over the entire concentration region. Both Ising cluster excitations and intracluster excitations coexist over the entire concentration region. Only the intensities from two different origins change with the concentration. Those from Ising cluster excitations increase and those from intracluster excitations decrease with increasing magnetic concentration. Although the mixed inelastic signals from two origins, one quantum mechanical and another statistical, were at first glance surprising, the latter excitation can occur at low temperatures where a thermal correlations between spin develops.

In conclusion, we have clarified magnetic excitations in a diluted Ising magnet over the entire magnetic concentration region. The nature of this process is much simpler than we had imagined. Our findings involve the coexistence of mixed excitations with different origins, even in a small magnetic cluster comprising only two magnetic atoms.

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