OBSERVATION OF CRITICAL FLUCTUATIONS USING MÖSSBAUER SPECTROSCOPY

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Critical fluctuations in the neighborhood of a magnetic phase transition can be observed using Mössbauer spectroscopy if the spin fluctuation rate of the atom containing the Mössbauer nucleus slows down sufficiently at the critical temperature. We have observed such a critical slowing down of the spin fluctuations near the Néel temperature of the antiferromagnet $K_2FeO_4$. Consider an iron-57 atom with no orbital angular momentum, total electronic spin $S=1$, and non-degenerate atomic eigenstates $S_z=1, 0, -1$. Assume, for simplicity, that the hyperfine interaction can be represented by an effective magnetic field in the $z$-direction at the nuclear site, and that this magnetic field takes on the values $-H$, 0, and $+H$ when $S_z=1, 0, -1$, respectively. If the atom undergoes transitions among these eigenstates due to relaxation in the solid, we must determine the magnetic field that the nucleus experiences.

Consider the cases of slow, fast, and intermediate relaxation shown in figure 1. By the time-energy uncertainty principle, the time required for a nucleus to measure a hyperfine magnetic field is approximately the Larmor period of the nucleus in that field, which for iron-57 is typically $10^{-9}$ sec. If the atomic relaxation is slow, and an atom spends many Larmor periods in each eigenstate, then a Mössbauer nucleus experiences a magnetic field $-H$, 0, or $+H$ depending on the state of the atom containing the nucleus. In this case, the Mössbauer spectrum is the Boltzmann-weighted sum of three spectra characteristic of these three fields. An example is shown in the upper curve in figure 1. The four outer peaks are from nuclei which experience magnetic fields $H$ and $-H$, and the large central peak is from nuclei of atoms in the state $S_z=0$, i.e., $H=0$.

If the atomic relaxation rate is fast compared to the Larmor frequency, then the nucleus cannot respond to the individual fields $H$, 0, and $-H$, and all nuclei experience the same, single valued, hyperfine magnetic field which is the Boltzmann-weighted average of these three values. A typical spectrum is shown as the middle curve in figure 1. Note that the central peak in the slow relaxation spectrum is completely absent under conditions of fast relaxation because the magnetic field of zero has lost its individual identity. Furthermore, the overall width of the fast relaxation spectrum is less than that of the slow relaxation spectrum.

If the atomic relaxation rate is comparable to the nuclear Larmor frequency, then the hyperfine magnetic field experienced by the nucleus is not well defined. This gives rise to a "relaxation
The iron atom in \( \text{K}_2\text{Fe}_4 \) is in a nominal 6+ charge state, and the two remaining d-electrons are coupled with parallel spins and no orbital angular momentum to yield \( J=S=1 \). The points in Fig. 2 show our experimental Mössbauer transmission spectra of polycrystalline \( \text{K}_2\text{Fe}_4 \) at 3.6K and 3.55K. Although these spectra exhibit some relaxation broadening, a comparison with Figure 1 shows that the 3.6K spectrum is characteristic of \( S=1 \) slow relaxation, and the 3.55K spectrum is characteristic of fast relaxation. Note that the central peak which is prominent at 3.6K is missing at 3.55K, and that the overall width of the spectrum at 3.6K is greater than at 3.55K. This slow atomic relaxation at 3.6K may be due to the proposed critical slowing down of the iron atom spin fluctuations at the Néel temperature.

We have used the stochastic relaxation theory of line shape of Clauser and Blume to fit our experimental Mössbauer spectra of \( \text{K}_2\text{Fe}_4 \) between 0.16K and 4.2K. The details of our calculation will be published elsewhere. Two of these theoretical spectra are shown as solid curves in Figure 2. The atomic relaxation rate and microscopic magnetic ordering parameter derived from the calculation are shown in Table 1. It can be seen that the relaxation rate drops by almost three orders of magnitude at the Néel temperature, in agreement with the above discussion. Furthermore, the microscopic magnetic ordering parameter can be seen to follow a magnetization-type curve from low temperatures up to the Néel temperature of 3.6K. Above 3.6K, the Mössbauer spectra of the two samples we have studied differ. We are currently attempting to resolve this discrepancy.

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