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Spin wave propagation in the domain state of a random field magnet

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Abstract. Inelastic neutron scattering with high wave-vector resolution has characterized the propagation of transverse spin wave modes near the antiferromagnetic zone center in the metastable domain state of a random field Ising magnet. A well-defined, long wavelength excitation is observed despite the absence of long-range magnetic order. Direct comparisons with the spin wave dispersion in the long-range ordered antiferromagnetic state reveal no measurable effects from the domain structure. This result recalls analogous behavior in thermally disordered anisotropic spin chains but contrasts sharply with that of the phonon modes in relaxor ferroelectrics.

PACS. 75.30.Ds Spin waves – 75.50.Lk Spin glasses and other random magnets

Disorder in a condensed matter system can strongly influence mode propagation through the medium, providing a unique perspective on its microscopic behavior. To gain a more comprehensive understanding of such effects from disorder, we have performed a neutron scattering study of the long wavelength spin waves of the frozen domain state of a random field magnet. Despite the wealth of attention directed at the random field problem [1], little attention has been directed toward the spin wave dispersion in these systems. Our study has been motivated in part by experiments on the phonons in several relaxor ferroelectrics including $\mathrm{Pb(Zn_{1/3}Nb_{2/3})O_3}$ and $\mathrm{Pb(Mg_{1/3}Nb_{2/3})O_3}$ that have demonstrated interesting anomalies associated with the disorder in these systems [2–5]. Specifically, at wave vectors near 0.2 $\mathrm{Å^{-1}}$, the dispersion of the transverse optic branch appears to drop precipitously into the acoustic branch. This phenomenon has been identified with a sharply wave-vector dependent overdamping associated with the presence of polar nanoregions in the relaxors [2]. To test the generality of such behavior for systems with disorder, we have performed a search for analogous effects in the spin waves of the domain state of the random field magnet. In marked contrast to the phonons in the relaxors, we observe well-defined long wavelength excitations transverse to the ordering axis, despite the absence of long range order, and find no measurable effect on the spin waves from the domain structure. We identify the persistence of these modes as an apparently generic feature of disordered magnetic systems with anisotropy, as first realized in thermally disordered spin chains [6–8].

The experiments were performed on a diluted antiferromagnet, $\mathrm{Mn}_x\mathrm{Zn}_{1-x}\mathrm{F}_2$ with $x = 0.5$, cooled in a magnetic field parallel to the Ising axis. The physics of the diluted Ising antiferromagnet in a uniform field can be mapped directly on to that of a random field Ising magnet [9], providing a method for experimental realization of the random field system. $\mathrm{MnF_2}$ has a tetragonal rutile-type structure with lattice constants $a = 4.87$ Å and $c = 3.31$ Å. The spin interaction is primarily Heisenberg-like, and a weak dipolar interaction between the Mn moments accounts for the small Ising anisotropy that aligns the Mn spins along the c axis (*i.e.*, the tetragonal axis). Diluted $\mathrm{MnF_2}$ was chosen for this study because, as a weakly Ising antiferromagnet, its small spin gap makes the physics of interest easily accessible. We stress that, while the interesting phonon behavior in relaxors motivates our spin wave study on this system, the diluted antiferromagnet in field should not be considered a precise magnetic analog of disordered ferroelectrics. First, the antiferromagnet does not share the property of a conserved order parameter. In addition, the nature of the polar nanoregions in the relaxors as the source of random fields has recently come into question [10]. Nevertheless, in pursuing the general problem of the effects of disorder on the spin waves in a random field magnet, we view diluted $\mathrm{MnF_2}$ as the best candidate for experiments.

When cooled from the paramagnetic phase in the field, the diluted antiferromagnet forms a metastable state of

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frozen domains. Using high resolution neutron scattering, we have characterized this domain structure and the corresponding spin wave dispersion at wavelengths larger than the typical domain size. In addition, by cooling the antiferromagnet in zero field and then applying the field, we have produced a long-range ordered antiferromagnetic state in the system. Comparing the results obtained after cooling in field with the results of identical measurements taken after zero field cooling, we are able to determine directly the influence of the domain structure on the spin waves.

An important criterion for this experiment is the ability to create domains under field cooling that are sufficiently small to resolve the spin wave dispersion at the appropriate wave vectors. Smaller domains are achieved with higher dilution [11,12], larger applied fields [12], and faster cooling rates. With too high dilution, however, the spin waves (in zero field) eventually become poorly defined [13]. In addition, the presence of a spin-flop transition in $Mn_xZn_{1-x}F_2$, with a spin-flop field that depends on x [12], limits the maximum field that one can apply while remaining in the random field Ising regime. Previous studies [11–13] indicate that $x = 0.5$ is the optimal compromise among these different concerns. The experiment was performed at the NIST Center for Neutron Research on the SPINS triple-axis spectrometer, with tight collimation (30'-10'-S-10'-20') to achieve high wave-vector resolution and a final neutron energy fixed at 3.5 meV. A single crystal of $Mn_{0.5}Zn_{0.5}F_2$, approximately 50 grams in size, was oriented with the c-axis (the Ising axis) parallel to the external field and perpendicular to the scattering plane. The crystal was strongly coupled to the bath of a helium flow cryostat, permitting rapid cooling ($>$ 20 K/minute) from the high temperature paramagnetic phase to 4.2 K, well below the Néel temperature in zero field, $T_N(H = 0) = 21$ K.

Figure 1 displays scattering profiles from elastic scans through the antiferromagnetic zone center (1, 0, 0) along the transverse direction $(1, q_K, 0)$ in an external field of $H = 2.3$ T. The open circles represent measurements taken after cooling the sample in zero field, so that the random field strength is zero through the Ising transition, and then raising the field (ZFC). The scattering follows a resolution-limited lineshape consistent with long-range antiferromagnetic order. The solid circles show the results from cooling in field (FC) and display a significantly broadened lineshape corresponding to the metastable domain state induced by the random fields. The scattering in the field-cooled measurement also shows a large overall enhancement in intensity, which we associate with the relief of extinction due to the domain structure. We model the short-ranged correlations of this field-cooled domain state with a Lorentzian-squared form,

$$I(\mathbf{q}, \omega = 0) = \frac{\sigma}{(1 + \mathbf{q}^2\xi^2)^2} \tag{1}$$

where $\mathbf{q}$ is the wave vector measured with respect to the antiferromagnetic zone center. The solid line in Figure 1 is the results of a fit with equation (1) convolved with the instrumental resolution function. This static neutron cross section is proportional to the Fourier transform of the real-space two spin correlation function. The transform of equation (1) corresponds to an exponential decay in the correlations between Ising spins,

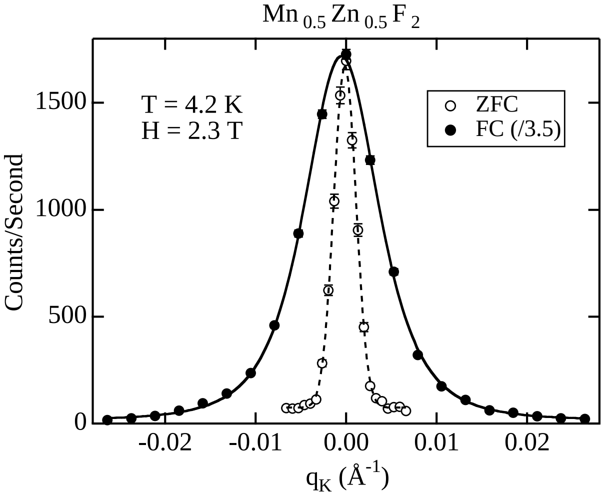

Fig. 1. Neutron scattering intensity at zero energy transfer scanning through the antiferromagnetic zone center (1, 0, 0) along the direction (0, q_K, 0) at $T = 4.2$ K and $H = 2.3$ T. The open circles are the results after zero field cooling and have a resolution-limited lineshape. The solid circles are the results after field cooling and have a lineshape reflecting the short-range ordered domain state. The field cooled intensities have been divided by 3.5 for comparison. The solid line is a fit to equation (1) convolved with the instrumental resolution.

$$\langle S_z(r)S_z(0)\rangle \sim \mathrm{e}^{-r/\xi}. \tag{2}$$

The Lorentzian-squared form for the elastic cross section has been shown in previous studies of field-cooled $Mn_xZn_{1-x}F_2$ [11,12] and other random field Ising magnets, including $Co_xZn_{1-x}F_2$ [14] and $Rb_2Co_xMg_{1-x}F_4$ [15], to describe accurately the static short-range order in the field-cooled state. In addition, theoretical arguments have indicated that this form should be expected on general grounds [16–19]. Thus, the field-cooled state can be considered a collection of metastable domains whose size distribution produces the real-space correlation function given by equation (2). The correlation length extracted from the fit with equation (1), $\xi = 137 \pm 4$ Å, sets the characteristic length scale in the ab plane for the domains. Assuming that the domain structure is isotropic in units of the lattice spacings, as suggested by the crystal structure, the smaller lattice constant along the c-axis leads to a characteristic scale of 93 ± 3 Å in that direction.

To compare the spin wave behaviors in the long-range ordered and domain states, we have measured the scattering intensity as a function of energy transfer, ω, at $q = 0$ through the dispersion, as shown in Figure 2. Because of the finite experimental resolution, this measurement effectively integrates over the approximate wavevector ranges

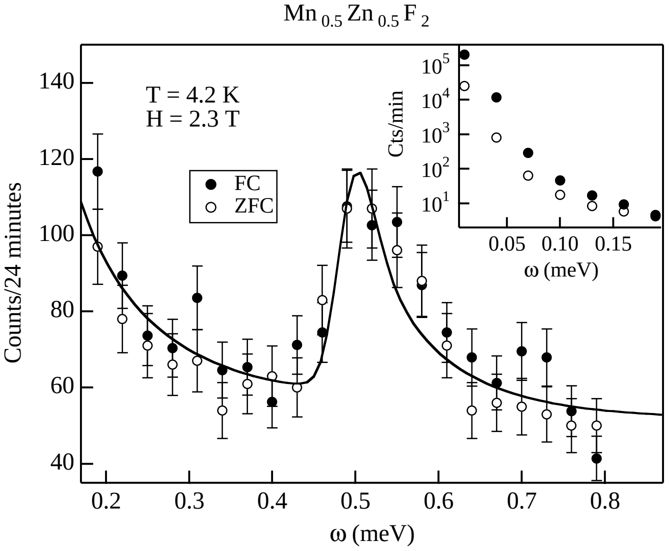

Fig. 2. Neutron scattering intensity as a function of energy transfer, ω, at the antiferromagnetic zone center (1, 0, 0) after field cooling (solid circles) and zero field cooling (open circles). The essentially identical profiles through the spin wave dispersion at $\omega \approx 0.52$ meV demonstrate that the long wavelength spin waves are insensitive to the domain structure. The solid line is the calculated result for the spin wave peak plus background based on the spectrometer resolution function and the known dispersion relation in $\mathrm{Mn_{0.5}Zn_{0.5}F_2}$ [13], assuming the that intrinsic width of the dispersion is zero. The enhanced intensity after field cooling at small ω, shown in the inset, reflects the enhanced Bragg scattering due to relief of extinction.

-0.004 Å$^{-1} < q_H < 0.004$ Å^{-1}, -0.003 Å$^{-1} < q_K < 0.003$ Å^{-1} within the scattering plane and -0.085 Å$^{-1} < q_L < 0.085$ Å^{-1} perpendicular to it [20,21]. Based on the phonon behavior observed in the relaxor ferroelectrics, we expect any change caused by the domain structure to affect the spin waves at wave vectors less than $q \approx 2\pi/\xi = 0.046$ Å^{-1} along H and K and $q \approx 2\pi/\xi = 0.068$ Å^{-1} along L. Thus, a redistribution of spectral weight to lower energy, like the dramatic overdamping seen in the relaxors, should be clearly visible in the scattering intensity in Figure 2. As the figure illustrates, the results for the long-range ordered antiferromagnet and domain state are essentially identical through the dispersion at $\omega = 0.5$ meV, indicating that the long wavelength spin waves are insensitive to the domain structure. The scattering at smaller energy transfers, shown in the inset to Figure 2, does display enhanced intensity in the domain state. However, we associate this enhancement with the stronger Bragg scattering observed for the domain state in Figure 1, extending away from $\omega = 0$ due to the finite experimental resolution, and not to any change in the spin wave behavior. In particular, any redistribution of spectral weight should be reflected in both enhanced scattering at small ω and reduced scattering at larger ω, which is excluded by the results in Figure 2. This dramatic contrast with the phonons in relaxors lends additional support to recent evidence questioning the accuracy of a random field picture for the relaxors [10].

The solid line in Figure 2 is the calculated result for the spin wave peak plus background based on the spectrometer resolution function and the previously measured dispersion relation in $\mathrm{Mn_{0.5}Zn_{0.5}F_2}$ [13], assuming the that intrinsic width of the dispersion is zero. The only free parameters entering the calculation are the overall amplitude of the peak and the form of the sloping background, $A\omega^{-2} + B$. The strongly asymmetric lineshape for the calculated result derives from the non-zero slope of the dispersion relation away from the zone center and the relatively coarse wave-vector resolution perpendicular to the scattering plane [21]. Any small discrepancies between the calculation and measurements likely result from uncertainties in this contribution to the resolution and in the dispersion relation, implying that the long wavelength spin waves in both field cooled and Néel states are long-lived. Furthermore, direct comparisons of the amplitudes and widths of the measured FC and ZFC peaks indicate that they are the same to within 5%, setting a small upper bound on any possible broadening specific to the field cooled domain state.

This clear insensitivity of the long wavelength spin waves to the static disorder of the FC domain structure raises two interesting issues that merit further study. The first concerns the effect on short wavelength spin waves. A comparison between the spin wave dispersions in the FC and ZFC states at wave vectors close the antiferromagnetic zone edge would address this question. As shown in reference [13], the spin wave dispersion at wave vectors away from the zone center in a diluted Ising antiferromagnet becomes increasingly diffuse due to the geometric disorder of the dilution. While this effect could severely complicate comparisons between FC and ZFC behaviors, it also indicates that short wave length spin waves are strongly affected by disorder. The second issue for further study concerns the temperature dependence of the spin wave dispersion, particularly in the transition region. The interplay of the critical behavior and the disorder-induced slow dynamics in the random field Ising magnet has presented serious challenges to a comprehensive description of the transition [22–25]. A characterization of the evolution with temperature of the spin wave dispersion near the zone center on approach to the transition would provide a new perspective on this issue.

The energy gap of approximately 0.52 meV for the excitation in Figure 2 matches that expected from the Heisenberg exchange field and dipolar anisotropy of $\mathrm{Mn_{0.5}Zn_{0.5}F_2}$, approximately 0.27 meV [13], and the Zeeman energy from the precession around the applied field, 0.25 meV for a 2.3 T field. This excitation, the gapped mode of the Ising state, corresponds to long wavelength oscillations in the spin components transverse to the easy axis. Because of the experimental resolution, the peak in Figure 2 represents contributions from oscillations over a range of wavelengths, most of which far exceed the correlation length for magnetic order in the domain state. The existence of these modes, despite the disorder, bears a striking similarity to the behavior in thermally disordered spin chains with easy plane anisotropy. As first predicted by Villain [6], well-defined long wavelength transverse modes have been observed in a number of one-dimensional anisotropic spin systems including $\mathrm{CsNiF_3}$ [7] and $\mathrm{(CD_3)_4NMnCl_3}$ [8]. The measurement of such excitations in the domain state of a three-dimensional random field Ising magnet indicates that these long-lived transverse modes occur more generally than previously realized, irrespective of the nature of the anisotropy, the system's spatial dimension, or the type of disorder. Indeed, studies of spin glasses appear to support this picture that these modes are generic to anisotropic disordered magnets. While spin waves in canonical Heisenberg spin glasses [26] are strongly overdamped, well-defined long wavelength excitations have been observed in Ising spin glasses [27]. Theoretical insight into the apparently universal nature of these transverse modes in disordered anisotropic magnets would be invaluable.

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