# On the dynamical study of magnets by polarized neutron scattering

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The possibility of using energy-spectrum measurements of the polarization-dependent part of the neutron-scattering cross section to study the critical spin dynamics of amorphous ferromagnets and to elucidate the nature of their spin excitations is discussed.

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In recent years the inelastic scattering of polarized neutrons has become widely used for studying the spin dynamics of magnets.[1–5] As a rule, what is measured in the experiments is the energy spectrum of the difference in the neutron-scattering cross sections when the initial polarization $\mathbf{P}_0$ is directed parallel and antiparallel to the magnetization $\mathbf{M}$ of the sample. Experiments of this kind enable one to exclude the contribution to the scattering from the coherent and incoherent nuclear elastic scattering channels, and they have various other advantages as well.

In this paper we shall analyze the experimental situation of Refs. 1–5, discuss the results of these studies, and elucidate the nature of the information that can be extracted from measurements of this type.

Let us stress first of all that this information is not the same as the information obtained in conventional experiments on the inelastic scattering of unpolarized neutrons. In fact, the spin dynamics of magnetized samples is governed by three independent components of the spin-correlation tensor: two transverse components—the symmetric $K^{(1)}_{jj'}(t) = \langle S^x_j(0)S^x_{j'}(t)\rangle = \langle S^y_j(0)S^y_{j'}(t)\rangle$ and antisymmetric $K^{3}_{jj'}(t) = -i\langle S^y_j(0)S^x_{j'}(t)\rangle = i\langle S^x_j(0)S^y_{j'}(t)\rangle$—and the longitudinal component $K^{(2)}_{jj'}(t) = \langle S^z_j(0)S^z_{j'}(t)\rangle$. In ordered magnets at low temperatures the longitudinal component is small, and the other two are trivially interrelated. In the vicinity of $T_c$, however, as in the case of highly disordered magnets at arbitrary $T$, this is generally not true, and all three components of the tensor must be studied in the experiment. Experiments with unpolarized neutrons can provide information only on $K^{(1)}$ and $K^{(2)}$ (for more details see Ref. 6). At the same time, the difference in the cross sections for polarized neutrons is related to the antisymmetric component of the correlator

$$K^{(3)}_{\mathbf{q},\omega} = -\tfrac{1}{4}[\langle S^+S^-\rangle_{\mathbf{q},\omega} - \langle S^-S^+\rangle_{\mathbf{q},\omega}]$$

by the relation

$$\Delta_{\mathbf{q},\omega} = \frac{1}{2}\left\{\frac{d^2\sigma(P_0)}{d\Omega d\omega} - \frac{d^2\sigma(-P_0)}{d\Omega d\omega}\right\} = \frac{1}{2\pi}\frac{K'}{K}P_0\,\{\,2(r_0\gamma)^2F^2_{\mathbf{q}}(\mathbf{em})^2K^{(3)}_{\mathbf{q},\omega}$$

$$-\,(r_0\gamma)\frac{\bar{A}}{2\pi}F_{\mathbf{q}}\langle S(T)\rangle(\Phi_{\mathbf{q},\omega} + \Phi_{-\mathbf{q},-\omega})\,[\,1-(\mathbf{em})^2]\,\}. \tag{1}$$

Here $\mathbf{k}$ and $\mathbf{k}'$ are the momenta of the incident and scattered neutrons, $r_0$ is the classical radius of the electron, $\gamma \simeq -1.9$, $F_{\mathbf{q}}$ is the atomic spin form factor, $\mathbf{e} = \mathbf{q}q^{-1}$, $\mathbf{q} = \mathbf{k}' - \mathbf{k}$, $\mathbf{m} = \mathbf{M}M^{-1}$, $\omega = E' - E$ is the energy transfer, and $\bar{A}$ is the coherent nuclear scattering amplitude. In Eq. (1) the first term corresponds to magnetic scattering and the second term to its interference with the nuclear scattering; the corresponding structure factor in (1) is of the form

$$\Phi_{\mathbf{q},\omega} = \frac{N}{V}\int d\mathbf{r}_{lj}\,dt\,W(\mathbf{r}_{lj})\langle e^{-i\mathbf{q}\mathbf{r}_{lj}(t)}\rangle e^{i\omega t}, \tag{2}$$

here $\mathbf{r}_{lj} = \mathbf{R}_l(0) - \mathbf{R}_j(t)$, where $\mathbf{R}_j$ and $\mathbf{R}_l$ are the coordinates of the nuclei of the magnetic and nonmagnetic atoms, respectively, and $W(\mathbf{r}_{lj})$ is the nuclear density distribution function. The correlator $K^{(3)}_{\mathbf{q},\omega}$ is given by the formulas

$$K^{(3)}_{\mathbf{q},\omega} = -\frac{1}{4}[K^{+-}_{\mathbf{q},\omega} - K^{-+}_{\mathbf{q},\omega}] = -n(-\omega)\,\mathrm{Im}\,G^{(3)}_{\mathbf{q},\omega},$$

$$K^{\alpha\beta}_{\mathbf{q},\omega} = \frac{N}{V}\int d\mathbf{r}_{jj'}\,dt\,W(\mathbf{r}_{jj'})\langle S^{\alpha}_j(0)\,S^{\beta}_{j'}(t)\,e^{-i\mathbf{q}\mathbf{r}_{jj'}}\rangle e^{i\omega t}, \tag{3}$$

where $n(\omega) = [\exp(\omega)/T) - 1]^{-1}$, and the function $\mathrm{Im}G^{(3)}_{\mathbf{q},\omega} = -\frac{1}{4}\mathrm{Im}[G^{+-}_{\mathbf{q},\omega} - G^{-+}_{\mathbf{q},\omega}]$ is even in $\omega$ ($G^{\alpha\beta}$ are the spin Green's functions). Therefore, for $\omega \ll T$ the function $K^{(3)}_{\mathbf{q},\omega}$ is odd.

In Ref. 1 a study was made of the quantity $\Delta_{\mathbf{q},\omega}$ during small-angle critical scattering in iron under an applied magnetic field. Under the conditions of this experiment the second term in Eq. (1) can be ignored, since $\langle S(t)\rangle$ is small in a weak field near $T_c$, while the correlator (3), on the other hand, is large. Here the quantity $K^{(3)}_{\mathbf{q},\omega}$ can be extracted directly from the experimental data. In weak fields above $T_c$ the data of Ref. 1 are particularly interesting, since according to Ref. 7 in this case $\Delta_{\mathbf{q},\omega}$ is proportional to the ternary dynamical spin correlator. To the best of the author's knowledge, no direct measurements of the dynamics of the ternary correlators had previously been made, and the experiment of Ref. 1 was the first of its kind.

Unfortunately, for arbitrary values of $\mathbf{q}$ and $\omega$, the form of the ternary Green's function $G^{(3)}_{\mathbf{q},\omega}$, like that of the binary function $G_{\mathbf{q},\omega}$, is unknown. The data of Ref. 1 were processed with the aid of the formula

$$\operatorname{Im} G^{(3)}_{\mathbf{q},\,\omega} = g\,\mu H T_c\, q^{3/2}\, \gamma_3\,(q/\kappa)\,\frac{\omega}{\Gamma} G_0\,(\kappa)\operatorname{Im} G^2_{\mathbf{q},\,\omega}\,, \tag{4}$$

which is valid for $\omega \ll \Gamma_{\mathbf{q},\kappa}$, where $\Gamma$ is the characteristic energy of the critical fluctuations, $\gamma_3$ is a homogeneous function of $q/\kappa$, and $\kappa$ is the inverse correlation length. It can be shown that this formula is applicable over the entire range of $\omega$ if $q \gg q_i = a^{-1}(2E/T_c ka)^{2/3}$ (see Ref. 8). Only the condition $q \gtrsim q_i$ was satisfied in Ref. 1. At the same time, however, the value of $\Gamma$ obtained in Ref. 1 agrees to good accuracy with the results of other, independent measurements. Evidently, this fact more or less justifies the use of Eq. (4). Another factor which probably contributed to the success of this approach was the circumstance that the bulk of the data in Ref. 1 was concentrated in the quasielastic region $\omega < 2E\theta$, where $\theta$ is the scattering angle. The spectra of $\Delta_{\mathbf{q},\omega}$ slightly above $T_c$ were also measured in Ref. 1. Here the results are curious in that they display sharp peaks in $\Delta_{\mathbf{q},\omega}$ at certain $\omega \simeq \omega_0$, although the measurements were made in the region $q \sim \kappa$, where spin waves should be not very well defined. By changing the angle $\psi$ between $\mathbf{m}$ and $\mathbf{k}$, the authors of Ref. 1 were able to almost completely suppress the contribution of one of the scattering processes—either that involving energy gain or that with energy loss. This means that at the values $\psi = \psi_0$ that were found, we have $\mathbf{e}(\omega_0)\mathbf{m}(\psi_0) = 0$. Thus, according to Ref. 1, the measurement of the angles $\psi_0$ at which $\Delta_{\mathbf{q},\omega}(\psi_0) = 0$ can serve as the basis of a method of determining $\omega_0$ under conditions of insufficient energy resolution. We note that, as was shown in Ref. 4 and 5, the energy resolution is a fundamentally important question in measurements of $\Delta_{\mathbf{q},\omega}$. In particular, the results of Refs. 4 and 5 cast doubt upon the existence of a "roton-like" branch of spin excitations in amorphous magnets. The observation of such a branch in the vicinity of the first peak in the structure factor was reported in Ref. 3. It was pointed out in Refs. 4 and 5, however, that the observed $\Delta_{\mathbf{q},\omega}$ spectra were not odd in $\omega$, as had been expected, and were nonzero at $\omega = 0$. In Ref. 4, these effects were explained essentially by the contribution to $\Delta_{\mathbf{q},\omega}$ from multiple-scattering processes. Without ruling out the influence of such processes, which can easily be diminished by changing the thickness of the samples, let us give some other possibly more important, reasons for these effects. In the experiments of Refs. 3–5, the maximum value of $\Delta_{\mathbf{q},\omega}$ generally amounted to a few percent of the scattering cross section (the scattering was primarily nuclear, as the measurements were made at large values of $q$). In order for the contribution to $\Delta_{\mathbf{q},\omega}$ from the second term in (1) to be adequately suppressed, it is necessary that $\mathbf{e}$ and $\mathbf{m}$ be parallel to within one percent or better. When $\omega$ is scanned at constant $q$, however, the direction of $\mathbf{e}$ changes, and so one must also with the same accuracy change the direction of $\mathbf{m}$, i.e., one must rotate the magnetizing field together with the sample. This procedure, while extremely complicated from a technical standpoint, still does not guarantee that $\Delta_{q,\omega}$ will not be contaminated by magnetic—vibrational scattering on account of the finite energy resolution $\delta$. The contribution to $\Delta_{q,\omega}$ from this scattering mechanism has both even and odd (in $\omega$) components, which under ideal conditions are of the order of $(\delta/E)^2$ and $\omega/E(\delta/E)^2$, respectively, times the scattering cross section. If $\mathbf{e}(\omega)$ and $\mathbf{m}$ are not strictly parallel, both components increase with the angle between $\mathbf{e}$ and $\mathbf{m}$, so that the odd contribution can become comparable to the effect of the magnetic scattering and even imitate it. In the experiments of Refs. 1 and 2 these questions are all less important, since the inelastic nuclear scattering at small angles is highly suppressed. The spectra of $\Delta_{q,\omega}$ observed in Refs. 1 and 2 are therefore to good accuracy odd in $\omega$.

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