Feasibility of studying the dynamics of three-spin correlations in ferromagnets above T_c by the method of pseudorandom modulation of neutron polarization

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(Submitted 15 March 1983)
Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 9, 432–435 (5 May 1983)

The energy spectra of triple-spin correlations in Fe are observed experimentally above T_c in a weak magnetic field. The data obtained agree with the predictions of the hypothesis of dynamic similarity for triple-spin Green's function $G^{(3)}(\omega)$.

PACS numbers: 75.25. + z, 75.50.Bb

Triple-spin correlations in iron above T_c were studied in Refs. 1 and 2. The technique used in these investigations is based on measuring the asymmetry of critical scattering of polarized neutrons, which follows from the general expression for the scattering intensity $I(Q)$ (Ref. 3) in a magnet in a weak magnetic field $\mathbf{H}$:

$$I(q) = I_0 \int \frac{d\omega}{\omega}\frac{k'}{k} \mathrm{Im}\,\{G^{(2)}(q,\omega) + g\mu H(\mathbf{eh})(\mathbf{eP}_0)\,G^{(3)}(q,\omega)\}\ , \tag{1}$$

where $G^{(2)}$ and $G^{(3)}$ are the pair and triple Green's functions with $H=0$, $\mathbf{e}=\mathbf{q}q^{-1}$, $\mathbf{q}=\mathbf{k}''-\mathbf{k}$, $\omega = E'-E$, $\mathbf{k},\mathbf{k}'$, E and E' are initial and final values of the neutron momenta and energies, $\mathbf{h}=\mathbf{H}H^{-1}$, $\mathbf{P}_0$ is the initial polarization of the beam. The scattering asymmetry is caused by the term with $G^{(3)}$, depending on the sign of $\mathbf{P}_0$ and the angle ϕ between $\mathbf{e}$ and $\mathbf{h}(\|\mathbf{P}_0)$ integrated over ω with fixed $k\theta$, where θ is the scattering angle. The asymmetry $P=(I_+ - I_-)/(I_+ + I_-)$, where $I_+ = I(q, +P_0)$, and $I_- = I(q, -P_0)$, was observed experimentally in Ref. 4 and explained theoretically in Ref. 3. The dependence of P on θ, ϕ, H and $\tau = (T-T_c)/T_c$ near T_c was investigated in Ref. 1, while the temperature behavior of $\Delta I = I_+ - I_-$ was studied in a wide range τ in Ref. 2. The results of the experiments agree with the theory in Ref. 3, but the integral dependences of ΔI can be interpreted only under asymptotic conditions with respect to the parameters H, τ, $k\theta$, when the integral (1) over ω can be calculated. The energy dependence of $G^{(3)}(\omega)$ is more informative. However, because the spin-dependent term in (1) is small, it is difficult to study $G^{(3)}(\omega)$ using the usual spectrometers. At the same time, it follows from expression (1) that the effect sought, which is related to $G^{(3)}$, is given by the difference between the scattering intensities with polarization P_0 and $-P_0$:

$$I_3(q,\omega) = I_+(q,\omega) - I_-(q,\omega) = 2I_0\,\frac{1}{\omega}\frac{k'}{k}(\mathbf{eh})^2\,\mathrm{Im}\,G^{(3)}(q,\omega)\,. \tag{2}$$

In this case, we can use the method of pseudorandom modulation of polarization (PRMP), proposed in Ref. 5, which is sensitive to the spin-dependent part of the scattering cross section. Such an experiment was performed for an iron specimen. Below we present our results.

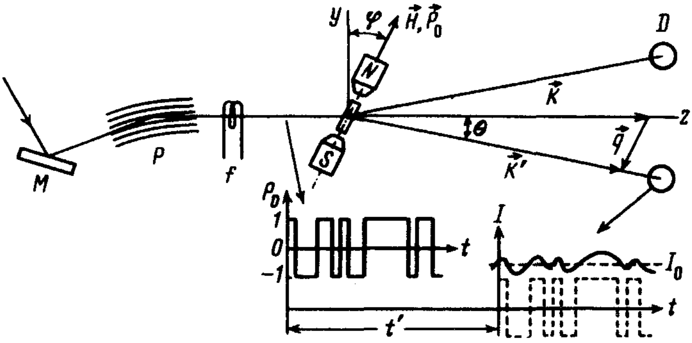

FIG. 1. Diagram of experiment with PRMP.

The experiment was arranged on a D-7 setup of the high-flux reactor at the Laue-Langevin Institute (Grenoble, France). The neutron beam (Fig. 1), reflected from the monochromator (M) consisting of pyrolytic graphite, was polarized by a multislit neutron guide (P) consisting of supermirrors and passed through the rapid flipper (F),[6] which modulated the polarization $\mathbf{P}_0(t)$ according to a pseudo-random sequence with period L. The neutrons were scattered by the specimen magnetized by a field $\mathbf{H}$ in the scattering plane at an angle ϕ to the Y axis. The detectors (D) recorded the flight-time spectra $I(t)$ (Fig. 1). The specimen served as the polarization analyzer and the modulation $I(t)$ arose due to the difference $\Delta I(t')$ between the cross sections for scattering of neutrons with polarization P_0 and $-P_0$. The spectrum $\Delta I(t')$ was extracted from $I(t)$ by calculating the cross correlation function

$$K(t') = \frac{1}{L}\int_0^L I(t)P(t-t')dt,$$

equal within a constant to a convolution of the sought-for spectrum $\Delta I(t')$ and the instrumental function $\Phi(t)$ (Ref. 5):

$$K(t') = \frac{1}{2}\int \Phi(t-t')\,\Delta I(t)\,dt + \text{const}. \qquad (3)$$

Then, the flight time spectra $K(t')$ were transformed into the dependence

$$K(\omega) \propto I_3(\omega) \propto \operatorname{Im} G^{(3)}(\omega).$$

Typical spectra $K(\omega)$, obtained in the experiment, are shown in Fig. 2. It is evident that $K(\omega)$ is an odd function of ω, as follows from the theory in Ref. 3 for scattering by three spin correlations. In this case, neutrons with positive spin state ($I+$) lose energy, while neutrons with a negative ($I-$) spin state acquire energy. At $\phi = 0$ the integral values $I+$ and $I-$ are equal and the integral asymmetry P does not arise ($P = 0$, Fig. 2a). For $\phi \neq 0$ scattering with loss and acquisition of energy have different probabilities due to the angular factor $(\mathbf{eh})(\mathbf{eP}_0)$ (Fig. 2b and c). This is evident from the difference in the areas under the curves $K(\omega)$ for $\omega < 0$ and $\omega > 0$, which is what leads to the appearance of a nonzero integral asymmetry effect $P \propto \int K(\omega)d\omega$, observed in Ref. 4. The sign of the asymmetry P differs for scattering at angles θ and $-\theta$ (Figs. 2b and 2c). All curves in Fig. 2 completely confirm the results in Refs. 3 and 1.

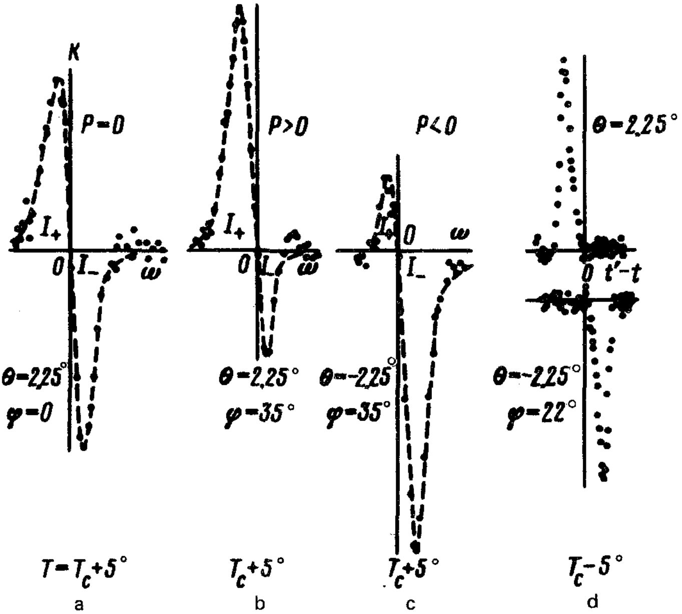

FIG. 2. Cross correlation function $K(\omega)$ (a, b, and c) and $K(t'-t)$ (d).

According to Ref. 3, Im $G^{(3)}$ can be written in the form

$$\operatorname{Im} G^{(3)}(q,\omega)=\operatorname{Im}[G^{(2)}(q,\omega)]^2\,G^{(2)}(0,0)\,\gamma_3(q,\omega,0,0), \tag{4}$$

where $\gamma_3=q^{3/2}\psi$ is the three-particle interaction vertex, while ψ according to the hypothesis of dynamic similarity, is a homogeneous function of arguments q/κ and $\omega/\Gamma(q)$, where κ is the inverse correlation radius, $\Gamma(q)=Cq^{5/2}f(\kappa/q)$ is the characteristic energy fluctuation, and $f(\kappa/q)$ is the similarity function. In the limit $\omega\ll\Gamma(q)$ the vertex γ_3 has the form $\gamma_3(q,\omega)=(\omega/\Gamma)q^{3/2}\gamma_0(\kappa/q)$. Separating from $G^{(2)}(q,\omega)$, a ω-dependent part in the form of a Lorentz function $G^{(2)}(q,\omega)=G^{(2)}(q,0)i\Gamma/(\omega+i\Gamma)$, Eq. (2) can be written in the form

$$I_3(\omega)=A(q,\kappa)\frac{k'}{k}\gamma_0(\kappa/q)[\mathbf{e}(\omega)\,\mathbf{h}]^2\frac{\omega\Gamma^2}{(\omega^2+\Gamma^2)^2}, \tag{5}$$

which permits determining the characteristic energy Γ by comparing (5) with the experimental curve $K(\omega)$. The results of fitting the experimental data for iron with $\tau=3\times10^{-2}$ and $k\theta=1.05\times10^{-1}$ Å^{-1} using Eq. (5) in the quasielastic approximation $q\approx\kappa\theta$ with allowance for the instrumental function are shown in Fig. 3 (curve 1). The function $I_3(\omega)$ with parameters obtained from a fit using the least-squares method is shown in Fig. 3 (curve 2). The value $\Gamma=240\pm20\,\mu$eV was obtained for the characteristic energy ($Cf(\kappa/q)=74$ (5) meV·Å$^{-5/2}$). This agrees well with previously obtained data[7] for iron under the same conditions ($\tau\approx3\times10^{-2}$, $\kappa\theta=1\times10^{-1}$ Å^{-1}) while studying inelastic scattering by pair correlations ($\Gamma_{G^{(2)}}\approx230$ meV), which confirms the validity of the hypothesis of dynamic similarity for three-spin Green's function $G^{(3)}$.

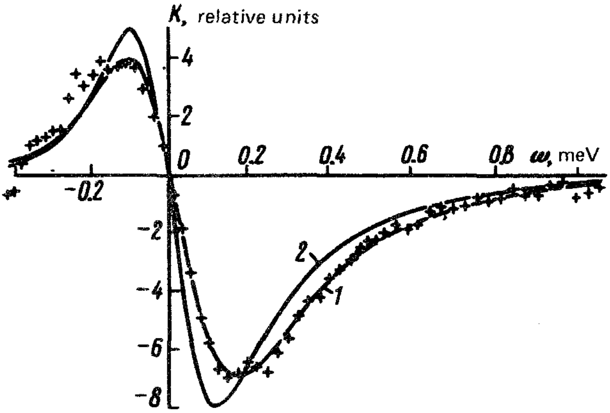

FIG. 3. (1) Experimental values of $K(\omega)$ fitted using the least-squares method according to Eq. (5) and with the instrumental function; (2) $I_3(\omega)$ with parameters obtained from the fit in 1.

Thus, we have observed for the first time the spectra of triple-spin correlations, while the PRMP methods permitted separating reliably the spin-dependent inelastic scattering and eliminating the disadvantages of the integral method noted above. The PRMP method is also effective in studying spin dynamics below T_c. The spectra of inelastic scattering by spin waves in the ferromagnetic phase obtained by us are shown in Fig. 2d. In contrast to analogous works[4] on the investigation of spin waves, in the geometry with $\phi \neq 0$ used by us, it is possible to separate completely the processes involving absorption (upper curve) and emission (lower curve) of a spin wave, which are manifested in the scattering at angles θ and $-\theta$, respectively, thereby eliminating uncertainties at $\omega = 0$ (Ref. 9) (this is described in greater detail in Ref. 10).

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Translated by M. E. Alferieff
Edited by S. J. Amoretty