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Abstract

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ON THE MAGNETIC SCATTERING OF THERMAL NEUTRONS NEAR THE CURIE POINT OF A FERROMAGNET OR ANTI-FERROMAGNET

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The recently discovered [1,2] sharp increase in the magnetic scattering of neutrons by ferromagnets at temperatures close to the Curie temperature T_0 was qualitatively explained [3,4] by the presence of critical fluctuations of the magnetic moment $\vec{\mathfrak{M}}(\mathbf{r})$ near T_0. From this point of view, the effect under consideration is analogous to the anomalous scattering of x rays near the ordering point [5,6]. The thermodynamic theory of anomalous magnetic scattering is most simply constructed by considering the probability distribution of the Fourier components of $\vec{\mathfrak{M}}(\mathbf{r})$ and the time variation of these components.

1. A general expression for the cross section of magnetic scattering of unpolarized monochromatic neutrons, $d^2\sigma/dE\,d\Omega$, calculated per unit solid angle and per unit energy, can be obtained in the Born approximation on the basis of the calculation [7]. Carrying out a treatment analogous to that applied in [8] in calculating scattering by lattice vibrations, we obtain for a single crystal:

$$\frac{d^2\sigma}{dE\,d\Omega} = \frac{CV^2}{2\pi\hbar}\frac{k_2}{k_1}\sum_{i,j=1}^{3}\left(\delta_{ij} - \frac{q_{iq}j}{q^2}\right)\int_{-\infty}^{\infty}\mathfrak{M}_{iq}(0)\mathfrak{M}^*_{jq}(t)e^{i\omega t}\,dt. \tag{1}$$

Here $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the incident and scattered waves; $\mathbf{q} = \mathbf{k}_2 - \mathbf{k}_1$; i and j number the Cartesian coordinates; δ_{ij} is the δ-symbol; $C = 2\pi e^2\gamma^2/c^2\hbar^2$; $\gamma = 1.91$; $\omega = \Delta E/\hbar$, where ΔE is the decrease in the neutron energy upon scattering; V is the volume of the crystal; $\mathfrak{M}_{jq}(t) = V^{-1}\int_V \mathfrak{M}_j(\mathbf{r},t)\exp(i\mathbf{qr})\,d\tau$ is the Fourier component at time t.

$\vec{\mathfrak{M}}(\mathbf{r})$ can be represented as the product of the magnetization vector $\mathbf{M}(\mathbf{r})$ and a periodic function $\Psi(\mathbf{r})$, whose integral per unit volume is equal to unity. The magnetization may be decomposed into its mean value $\overline{\mathbf{M}}$ and a variable term $\Delta\mathbf{M}(\mathbf{r})$ arising as a result of fluctuations. The distribution of the magnetic moment $\overline{\mathbf{M}}\Psi(\mathbf{r})$ gives sharp lines (or spots) in the neutron diffraction pattern, which will not be considered here. Fluctuations of the magnetization lead to the appearance of a diffuse background. Here we restrict ourselves to investigating the background near the maxima in the neutron diffraction pattern corresponding to various reciprocal-lattice vectors $\mathbf{K}_n$, or to small scattering angles, since it is precisely in these cases that anomalous scattering should be observed and, at the same time, a phenomenological treatment is possible. In this case $\mathfrak{M}_{iq}(t) = M_{iq_n}(t) f_n$, where $\mathbf{q}_n = \mathbf{q} + 2\pi\mathbf{K}_n$, q_n is small, and f_n is the scattering factor corresponding to the maximum under consideration (for scattering at small angles $\mathbf{K}_0 = 0$, $f_n = 1$). Near T_0, the relaxation times—

cation become extremely large (cf. (9)). At the same time, the energy $\hbar\omega_l$ corresponding to the Larmor precession of the fluctuation of the magnetization about the magnetic field $\mathbf{H}$ does not exceed 10^{-5} eV. Therefore, except for the case of very small angles ($< \omega_l m/\hbar k_1^2 \sim 10^{-3}$), and also small angular deviations of the vectors $\mathbf{k}_2 - \mathbf{k}_1$ and $-2\pi\mathbf{K}_n$ ($< \omega_l m/2\pi\hbar K_n k_1$), one may neglect the change in q_n associated with the change of energy in the scattering. As a result, the integration of expression (1) over energy can be carried out by regarding only ω as a function of E:

$$\frac{d\sigma}{d\Omega} = CV^2|f_n|^2 \sum_{i,j=1}^{3} \left(\delta_{ij} - \frac{q_{iq}j}{q^2}\right) \overline{M_{iq_n} M^*_{jq_n}}. \tag{2}$$

Here, in the averaging, M_i and M_j are taken at the same instant of time.

2. $M_{iq_n} M^*_{iq_n}$ is calculated with the aid of the thermodynamic theory of fluctuations. In doing so it should be taken into account that $\partial^2\varphi/\partial M_i \partial M_j = \chi^{-1}_{ij}$, where $\varphi = \Phi/V$ is the thermodynamic potential per unit volume, and χ^{-1}_{ij} is the tensor inverse to the magnetic-susceptibility tensor χ_{ij}, and also the term connected with the inhomogeneities $\Delta\mathbf{M}(\mathbf{r})$,

$$\alpha_{ijkl} \frac{\partial M_i}{\partial x_k} \frac{\partial M_j}{\partial x_l},$$

in the expansion of φ, must be taken into account. Substituting the value found for $\overline{M_{iq_n} M^*_{iq_n}}$ in (2), we obtain

$$\frac{d\sigma}{d\Omega} = CV|f_n|^2 kT \left(\chi^{-1} + \alpha_{kl} q_{nk} q_{nl}\right)^{-1}_{ij}, \tag{3}$$

where -1 denotes the inverse matrix and summation is carried out over twice repeated indices. In particular, for sufficiently small q_n the last factor in (3) is equal to χ_{ij}. If only the change in the exchange part of the energy associated with the inhomogeneities $\mathbf{M}(\mathbf{r})$ is taken into account, then for cubic crystals $\alpha_{ijkl} q_{nk} q_{nl}$ may be replaced by $\alpha\delta_{ij} q_n^2$, where $\alpha \sim \chi^{-1}_\infty a_0^2$ (a_0 is the lattice constant, χ_∞ the magnetic susceptibility at $T \to \infty$).

Formula (3) makes it possible to determine the anomalous magnetic scattering by single-domain single crystals of arbitrary structure for arbitrary $\mathbf{H}$. In particular, for cubic crystals in which the equilibrium magnetization $\overline{\mathbf{M}}$ and $\mathbf{H}$ are parallel to the axis [001] (the OZ axis),

$$\frac{d\sigma}{d\Omega} = CV|f_n|^2 kT \left(\frac{1+\theta_z^2}{\chi_\perp^{-1} + \alpha q_n^2} - \frac{1-\theta_z^2}{\chi_\parallel^{-1} + \alpha q_n^2} \right), \tag{4}$$

where θ_z is the cosine of the angle between $\mathbf{q}$ and the axis OZ, $\chi_\parallel = \chi_{zz}$ and $\chi_\perp = \chi_{xx} = \chi_{yy}$. For $T > T_0$ in the absence of a field, $\chi_\parallel = \chi_\perp = \chi$ and

$$d\sigma/d\Omega = 2CV|f_n|^2 kT(\chi^{-1} + \alpha q_n^2)^{-1}.$$

Since at $T = T_0$ and $H = 0$, χ_{ij}^{-1} vanish, it follows from the formulas obtained that, near T_0, for small q_n, i.e., at small scattering angles φ or near the points on the neutron diagram corresponding to Laue reflection, sharply T-dependent, extremely intense diffuse scattering should be observed, whose maxima lie at $q_n = 0$. Since at small φ

$$q_n^2 = q_0^2 = (4\pi/\lambda)^2 \sin^2(\varphi/2),$$

there should in this case also be a sharp dependence of $d\sigma/d\Omega$ on φ and on the wavelength λ. Writing, for cubic crystals, φ in the form

$$\varphi = \frac{1}{2} f(M^2) + \frac{1}{2} K(M_x^2 M_y^2 + M_x^2 M_z^2 + M_y^2 M_z^2) - \mathbf{MH},$$

where K characterizes the magnetic anisotropy, we find that

$$\chi_{ij}^{-1} = f' \delta_{ij} + 2f'' \overline{M}_i \overline{M}_j + K(\overline{M}^2 - 3\overline{M}_i^2)\delta_{ij} + 2K\overline{M}_i \overline{M}_j.$$

Taking into account that $\partial\varphi/\partial M_i = 0$, we obtain for the case under consideration

for example, for $T > T_0$ and $H \neq 0$, $\chi_\parallel^{-1} = f' + 2f''\overline{M}^2 = \chi^{-1}(1 + 3f''\chi^3 H^2)$; $\chi_\perp^{-1} = f' + K\overline{M}^2 = \chi^{-1}[1 + (f'' + K)\chi^3 H^2]$. Therefore, for sufficiently small $T - T_0$ (large χ) and for large H, the field can appreciably reduce $d\sigma/d\Omega$. In this case $d\sigma/d\Omega$ will depend not only on the magnitude, but also on the direction of $\mathbf{q}_n$. $\chi_\parallel$ and $\chi_\perp$ differ especially sharply in the case when $T < T_0$ and $H = 0$ (then $f' = 0$ and $K \ll f''$). If one assumes that the presence of a domain structure does not affect the probabilities of the fluctuations ΔM, and neglects scattering at domain boundaries, then one can find $d\sigma/d\Omega$, taking into account that in the domains $\overline{\mathbf{M}}$ is oriented with equal probability in any of the six directions parallel to the edges of the cubic cells. Carrying out the averaging, we find that in this case $d\sigma/d\Omega$ can be determined from the formula for $T > T_0,\ H = 0$, if in it $(\chi^{-1} + \alpha q_n^2)^{-1}$ is replaced by ${}^2/_3(\chi_\perp^{-1} + \alpha q_n^2)^{-1} + {}^1/_3(\chi_\parallel^{-1} + \alpha q_n^2)^{-1}$.

Thus, for a given $\mathbf{q}_n$, $d\sigma/d\Omega$ decreases from its maximum value at $T = T_0$ proportionally to χ^{-1} for $T > T_0$, and proportionally to ${}^1/_3(2\chi_\perp^{-1} + \chi_\parallel^{-1})$ for $T < T_0$.

3. In the case of solid solutions, owing to the fact that the fluctuations ΔM and the fluctuations of concentration c, generally speaking, are not statistically independent (cf. (6)), in (4) the quantities $\partial^2\varphi/\partial M_i^2 = \chi_{ii}^{-1}$ should be replaced by

$$\frac{\partial^2\varphi}{\partial M_i^2} - \left(\frac{\partial^2\varphi}{\partial M_i \partial c}\right)^2 \Bigg/ \frac{\partial^2\varphi}{\partial c^2}.$$

   Near T_0,

$$\frac{\partial^2\varphi}{\partial M_z \partial c} = \chi_\parallel^{-1}\frac{\partial \overline{M}}{\partial T}\frac{dT_0}{dc}; \qquad \frac{\partial^2\varphi}{\partial M_x \partial c} = \frac{\partial^2\varphi}{\partial M_y \partial c} = 0$$

   and

$$\frac{\partial^2\varphi}{\partial c^2} = N\left(\frac{\partial\mu_A}{\partial c} - \frac{\partial\mu_B}{\partial c}\right),$$

   where N is the number of atoms per unit volume; μ_A and μ_B are the chemical potentials of atoms A and B (for weak solutions $\partial^2\varphi/\partial c^2 = NkTc^{-1}$). Therefore, in the case of solid solutions in (4) one should replace $\chi_\parallel^{-1}$ by

$$\chi_\parallel^{-1} - \chi_\parallel^{-2}\left(\frac{\partial M}{\partial T}\right)^2\left(\frac{dT_0}{dc}\right)^2 N^{-1}\left(\frac{\partial\mu_A}{\partial c} - \frac{\partial\mu_B}{\partial c}\right)^{-1}$$

   (and replace $|f_n|^2$ by $|\bar{f}_n|^2$), which leads to an increase in the intensity of magnetic scattering (for the same $\chi_\parallel, \chi_\perp, \alpha$ and $\bar{f}_n \approx f_n$) in comparison with a one-component crystal. For $T > T_0$ and $H = 0$ one may still use formula (4).

4. In the case $T > T_0,\ H = \overline{M} = 0$, the Larmor precession of the quantity ΔM does not occur, and the relaxation of the different components of the vector $\mathbf{M}_x$ may be considered separately. This makes it possible simply to find the distribution of the scattered neutrons over energies. Near T_0, in this case

$$\frac{dM_i}{dt} = -\gamma_1\frac{\partial^2\varphi}{\partial M_i^2}(M_i - \overline{M}_i) + \gamma_2\frac{\partial^2\varphi}{\partial M_i^2}\nabla^2 M_i,$$

   where the kinetic coefficients γ_1 and γ_2 may be regarded as independent of T (cf. (9)). As follows from the theory of time-dependent thermodynamic fluctuations ((10), § 17), if

$$dM_{i\mathbf{x}}/dt = -hM_{i\mathbf{x}},$$

then

$$\int_{-\infty}^{\infty} \overline{M_{i\mathbf{x}}(0)M^*_{i\mathbf{x}}(t)}\exp(i\omega t)\,dt = \frac{2h}{\omega^2+h^2}\,|\overline{M_{i\mathbf{x}}}|^2.$$

Substituting this relation and the expression for $\overline{M_{i\mathbf{x}}M^*_{j\mathbf{x}}}$ into (1), we find that for cubic crystals at $T > T_0,\ H = 0$

$$\frac{d^2\sigma}{dE\,d\Omega} = \frac{2C}{\pi\hbar}V|f_n|^2\frac{k_2}{k_1}kT\frac{1}{\chi^{-1}+\alpha q_n^2}\frac{\chi^{-1}(\gamma_1+\gamma_2 q_n^2)}{\chi^{-2}(\gamma_1+\gamma_2 q_n^2)^2+\omega^2}. \qquad (5)$$

It follows from (5) that near T_0 the mean change in the neutron energy upon scattering is very small and (since $\gamma_1 a_0^2/\gamma_2$ is small) depends substantially on the scattering angle. We note that in the presence of a field, because of the Larmor rotation of the fluctuation ΔM, a sharp resonance maximum will appear in the distribution of the scattered neutrons over energies, corresponding to the transfer energy $\hbar\omega_n$. A considerably larger change in energy ($\sim 10^{-2}$ eV) may be expected in the scattering of slow neutrons by ferrimagnets, owing to interaction with the optical branch of ferrimagnons.

5. In the same way one may consider neutron scattering near the Curie point of an antiferromagnet. In this case, for $T < T_0$, a periodic distribution of magnetic moments arises spontaneously (with a symmetry different from the symmetry of the lattice), which on the average gives a magnetic moment equal to zero. For simplicity let us suppose that this distribution can be characterized by a single macroscopic vector parameter $\vec{\lambda}$, which vanishes for $T > T_0$. Anomalously large fluctuations of $\vec{\lambda}$ near the Curie point lead to extremely intense neutron scattering near the "superstructure"lines (or spots), for which the vectors $\mathbf{K}_n$ correspond to a symmetry lower than the symmetry of the lattice, the symmetry $\overline{\mathfrak{M}}(\mathbf{r})$. The intensity of such scattering is determined by formulas (3) and (4), if in them $|f_n|^2$ is replaced by a certain constant $|\varphi_n|^2$, and χ_{ij} by $\partial^2\varphi/\partial\lambda_i\partial\lambda_j$ ($\partial^2\varphi/\partial\lambda_i\partial\lambda_j = 0$ for $T = T_0$). The intensity of the "superstructure" lines in this case is proportional to $\vec{\lambda}^2$. When a magnetic field is applied in a cubic crystal along an axis of type [100] (along the OX axis), $\vec{\lambda}$ is established perpendicular to the field (along the OZ axis). In contrast to a ferromagnet, in an antiferromagnet in the presence of a field the second-order phase transition is preserved. Since for $T < T_0$ the fluctuations $\vec{\lambda}$ and $\mathbf{M}$ are not statistically independent, in the expressions for $\overline{|\lambda_{zx}|^2}$ and for $d\sigma/d\Omega$ at $H \neq 0$ one must replace $\partial^2\varphi/\partial\lambda_z^2$ by

$$\frac{\partial^2\varphi}{\partial\lambda_z^2} - \left(\frac{\partial^2\varphi}{\partial\lambda_z\partial M}\right)^2\Big/\frac{\partial^2\varphi}{\partial M^2} = \frac{\partial^2\varphi}{\partial\lambda_z^2} - \chi^{-1}\left(\frac{\partial^2\varphi}{\partial\lambda_z^2}\right)^2\left(\frac{\partial\lambda}{\partial T}\right)^2\left(\frac{dT_0}{dH}\right)^2.$$

For $H \neq 0$ the quantities $\partial^2\varphi/\partial\lambda_i^2$ also change; in this case $\partial^2\varphi/\partial\lambda_x^2 \neq \partial^2\varphi/\partial\lambda_y^2$, which will change the angular distribution of the scattering.

6. Owing to magnetostriction, fluctuations of M^2 (or λ^2) give rise to fluctuations of the density ρ and, in addition to magnetic scattering, also lead to additional nuclear scattering. To estimate this effect, let us suppose that the relation between $\Delta\rho$ and ΔM^2 is the same as the relation between the corresponding macroscopic quantities, and let us not take into account fluctuations of the short-range magnetic order. Then the Fourier component of the fluctuation of ρ is equal to $\rho_{\mathbf{q}} = \rho \left(\frac{\partial \overline{M^2}}{\partial T}\right)^{-1} \delta\beta \, (\Delta M^2)_{\mathbf{q}}$, where $\delta\beta$ is the abrupt change of the volume-expansion coefficient for $T < T_0$ near T_0. It is easy to show that the change in the nuclear scattering of neutrons associated with fluctuations of ρ at small scattering angles is equal to

$$\Delta \frac{d\sigma_{\mathrm{n}}}{d\Omega} \frac{m^2 V}{\pi^2 \hbar^4} N^2 |\overline{A}|^2 kT \overline{M^2} \left(\frac{\partial \overline{M^2}}{\partial T}\right)^{-2} (\chi_{\parallel}^{-1} + \alpha q_n^2)^{-1} (\delta\beta)^2,$$

where m is the neutron mass, $\overline{A}$ is the nuclear scattering factor averaged over isotopes. Usually this scattering is smaller than scattering by thermal vibrations.

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Note: Figure translations are in progress. See original paper for figures.