CRITICAL MAGNETIC SCATTERING OF NEUTRONS IN ANTIFERROMAGNETS

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The cross section for critical magnetic scattering of unpolarized neutrons in a two sublattice, spin $s = \frac{1}{2}$, Heisenberg antiferromagnet, for temperatures above or at the Néel point, withouts external magnetic field has been calculated on the basis of the previously derived general cros section formula and the previously developped description of the spatial and temporal behaviour of thermodynamic fluctuations in antiferromagnetic order. The cross section has been derived in two forms connected with the Ornstein-Zernike and $|\sin \varkappa_2 r|/r$ spin correlations. In discussing these cross sections, emphasis has been laid upon the second one.

1. *Introduction*

The aim of this paper was the calculation of the cross section for the critical magnetic scattering of neutrons in two sublattice Heisenberg antiferromagnets on the basis of the general cross section formula (Kociński 1968) and the results of earlier papers (Kociński and Wojtczak 1968a, b) concerning the spatial and temporal behaviour of thermodynamic fluctuations in antiferromagnetic order. In the elastic approximation the cross section has been previously calculated by De Gennes (1958) and by Elliott and Marshall (1958) an the experimental papers have been interpreted in terms of these theories. The theoretical treatment of critical magnetic scattering previously developped (Kociński 1966, 1968) leads to two expressions for the cross section, one connected with the Ornstein-Zernike spin correlation and the other following from the $|\sin \varkappa_2 r|/r$ spin correlation. Mainly the latter one will be discussed in this paper. The inelasticity of scattering has been accounted for in the experimental paper by Antonini (1967), however the cross section formula he uses as basis for discussion of experimental data differs essentially, even in the Ornstein-Zernike

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spin correlation case (the only one this author considers), from ours, as far as inelasticity of scattering is concerned. The cross section formulae derived in this paper allow for an experimental verification of their angular dependence. The dependence on temperature has not been fully discussed since the number of fluctuations as a function of scattering system temperature remains unknown.

2. *Cross section for critical scattering*

Since the fluctuations in magnetic moment of each sublattice decay according to the diffusion equation (Kociński and Wojtczak 1968b), we get from the general expression for the cross section for the scattering of unpolarized neutrons in a two sublattice spin $s = 1/2$ antiferromagnet, above or at the Néel point, in zero external field (*vide* Kociński 1968, formula 37) the following cross section formula:

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \left(\frac{2ge^2}{m_0c^2}\right)^2 \frac{1}{3\pi\hbar v_{0m}} \frac{k}{k_0} |F(\vec{\varkappa})|^2 N_f N_1 z^{-1} \sum_{\varrho}^{z} (1-e^{i\vec{\varkappa}\vec{\varrho}})\times$$

$$\times \sum_{\tau_m} \frac{\Lambda|\vec{\varkappa}-2\pi\vec{\tau}_m|^2}{\Lambda^2|\vec{\varkappa}-2\pi\vec{\tau}_m|^4+\omega^2} \int_{V_1} d\vec{r}\, \langle \hat{S}_0^{\xi}(0)\hat{S}_r^{\xi}(0)\rangle_{T_0}\, e^{i(\vec{\varkappa}-2\pi\vec{\tau}_m)\cdot\vec{r}} \tag{1}$$

where N_f and N_1 denote the number of maximal fluctuations in antiferromagnetic order and the number of spins in a single fluctuation respectively, the vector $\vec{\varrho}$ connects a spin with its z nearest neighbors and V_1 denotes the volume of a single fluctuation, the remaining symbols having their usual meaning as in the previous paper (Kociński 1968). The spatial correlation in the integrand relates the spins of one sublattice only, and Λ is the diffusion coefficient for one sublattice.

In the case of spin correlation of the Ornstein-Zernike type

$$\langle \hat{S}_0^{\xi}(0)\hat{S}_r^{\xi}(0)\rangle_{T_0} = (\alpha \exp \varkappa_1\alpha/4) \exp(-\varkappa_1 r)/r \tag{2}$$

where the normalizing factor was determined on the condition that the magnetic moment at the lattice site $\vec{O}+\vec{\alpha}$ the nearest to the centre of the fluctuation, is equal to Bohr's magneton and where

$$\varkappa_1^2 = \frac{2\zeta\,[\eta^2-(\zeta-4)/\zeta]}{\delta^2\{k_BT_0/2J)\,(3-2\eta^2-\eta^4)-[\eta^2-(\zeta-4)/\zeta]\}} \tag{3a}$$

with $\delta = d, 2d$, (d is the crystal lattice constant) for $z = 8$, $\zeta = 6$ and $z = 6$, $\zeta = 12$ respectively (z is the number of nearest and ζ of next nearest neighbors in the crystal lattice) for weak nearest neighbor coupling[1], and

$$\varkappa_1^2 = 2zd^{-2}\left[\frac{(2J/k_BT_0)\,(\eta^{-2}-(z-4)/z)}{\eta^{-4}+2\eta^{-2}-3-2(1-\eta^{-2})^2\,(z-1)\,(zJ/k_BT_0)^{-1}}-1\right]^{-1} \tag{3b}$$

[1] The previous (Kociński and Wojtczak 1968a) statement that $\delta = \sqrt{2}\,d$ and z retained for the case $z = 6$, $\zeta = 12$ is erroneous.

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for strong nearest neighbor coupling, with $\eta = \exp(-J/k_B T_0)$ and d equal to the crystal lattice constant, in view of the fast convergence of the integral the integration over the volume V_1 of the fluctuation may be replaced by integration over whole space. We then get the formula

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \left(\frac{2ge^2}{m_0c^2}\right)^2 \frac{1}{3\hbar v_{0m}} \frac{k}{k_0} |F(\vec{\varkappa})|^2 N_f N_1 z^{-1} \sum_{\varrho}^{z} (1-e^{i\vec{\varkappa}\cdot\vec{\varrho}}) \times$$

$$\times \sum_{\tau_m} \frac{\Lambda|\vec{\varkappa}-2\pi\vec{\tau}_m|^2}{\Lambda^2|\vec{\varkappa}-2\pi\vec{\tau}_m|^4+\omega^2} \frac{\alpha \exp(\alpha\varkappa_1)}{\varkappa_1^2+|\vec{\varkappa}-2\pi\vec{\tau}_m|^2}. \tag{4}$$

It differs essentially from the conventional one (which has been calculated in the elastic approximation) by the presence of the factors N_f and N_1 instead of the number N of spins in the system. In the Smoluchowski approximation of treating the total scattering system we have $N_f N_1 = N$. The number of spins in a single fluctuation may be calculated in the term of the radius R of a spherical fluctuation in magnetic moment in one sublattice namely

$$N_1 = 2N_1' = \frac{8}{3}\pi\nu\left(\frac{R}{\delta}\right)^3 \tag{5}$$

where N_1' denotes the number of spin in the fluctuation belonging to one sublattice and where δ is the "sublattice constant" equal to the lattice constant d for the *b.c.c.* crystal lattice and to $2d$ for the *s.c.* crystal lattice and $\nu = 1{,}4$ for the *s.c.* and *f.c.c.* sublattices respectively. For the $O-Z$ correlation one usually defines $R = \varkappa_1^{-1}$. The temperature dependent factor N remains undetermined but it does not affect the angular dependence of the cross section.

In the case of the spin correlation

$$\langle \hat{S}_0^\xi(0)\, \hat{S}_r^\xi(0)\rangle_{T_0} = \frac{|\sin \varkappa_2 r|}{4\varkappa_2 r} \tag{6}$$

with:

$$\varkappa_2^2 = \frac{2\zeta[\eta^2+(7\zeta-4)/\zeta]}{\delta^2[\eta^2+(7\zeta-4)/\zeta-(k_BT_0/2J)(3-2\eta^2-\eta^4)]} \tag{7a}$$

where $\delta = d$, $2d$ for $z = 8$, $\zeta = 6$ and $z = 6$, $\zeta = 12$ respectively, for weak coupling between nearest neighbors, and

$$\varkappa_2^2 = 2zd^{-2}\left[1 - \frac{(2J/k_BT_0)(\eta^{-2}+(7z-4)/z)}{\eta^{-4}+2\eta^{-2}-3+2(1-\eta^{-2})^2(z-1)\,(zJ/k_BT_0)^{-1}}\right]^{-1} \tag{7b}$$

for strong coupling between nearest neighbors, with d and η as before. One has to know the radius R of the spherical fluctuation in magnetic moment in a single sublattice in order to calculate the spatial integral in (1). This radius (range of correlation) will be determined

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in the next section in a way analogous to the ferromagnetic case. The cross section takes now the form

$$\frac{d^2\sigma}{d\Omega d\varepsilon} = \left(\frac{2ge^2}{m_0c^2}\right)^2 \frac{1}{3\hbar v_{0m}} \frac{k}{k_0} |F(\vec{\varkappa})|^2 N_f N_1 z^{-1} \sum_{\varrho}^{z} (1-e^{i\vec{\varkappa}\cdot\vec{\varrho}}) \times$$

$$\times \sum_{\tau_m} \frac{A|\vec{\varkappa}-2\pi\vec{\tau}_m|}{A^2|\vec{\varkappa}-2\pi\tau_m|^4+\omega^2} \frac{1}{\varkappa_2} \int_0^R |\sin \varkappa_2 r| \sin(|\vec{\varkappa}-2\pi\vec{\tau}_m| r)\, dr \tag{8}$$

with the factor N_1 determined by (5). The integral in (8) may be calculated in the form

$$\int_0^R |\sin \varkappa_2 r| \sin qr\, dr = (-1)^{\mathcal{M}} \left\{ \frac{1}{2} \left[\frac{\sin(\varkappa_2-q)R}{\varkappa_2-q} - \frac{\sin(\varkappa_2+q)R}{\varkappa_2+q} \right] + \right.$$

$$+ \sin\left[\frac{\pi}{2}(\mathcal{M}-s+2)\left(1+\frac{q}{\varkappa_2}\right)\right] \cos\left[\frac{\pi}{2}(\mathcal{M}+s-1)\left(1+\frac{q}{\varkappa_2}\right)\right] \times$$

$$\times \left((\varkappa_2+q)\cos\left[\frac{\pi}{2}\left(1+\frac{q}{\varkappa_2}\right)\right]\right)^{-1} - \sin\left[\frac{\pi}{2}(\mathcal{M}-s+2)\left(1-\frac{q}{\varkappa_2}\right)\right] \times$$

$$\left. \times \cos\left[\frac{\pi}{2}(\mathcal{M}+s-1)\left(1-\frac{q}{\varkappa_2}\right)\right] \left((\varkappa_2-q)\cos\left[\frac{\pi}{2}\left(1-\frac{q}{\varkappa_2}\right)\right]\right)^{-1} \right\} \tag{9}$$

where

$$\mathcal{M} = E(\varkappa_2 R/\pi) \tag{10}$$

is the "Entier" function of the argument $\varkappa_2 R/\pi$, and $s = 1, 2$ for $\mathcal{M}$ odd or even respectively, and $q = |\vec{\varkappa}-2\pi\vec{\tau}_m|$.

Diffraction minima represent the characteristic feature of this cross section, as in ferromagnets. The conditions for the angles at which these minima appear follow from the form of the spatial integral in the cross section, and are given by

$$R(T_0) = 2nr_0(q) = mr_0(\varkappa_2), \qquad n, m \text{ integers} \tag{11}$$

where $r_0(q)$ and $r_0(\varkappa_2)$ denote the positions of the first zeros of the functions $\sin qr$ and $|\sin \varkappa_2 r|$ respectively. Since $\varkappa_2$ is determined by the formulas (8a, b) the condition $R(T_0) = mr_0(\varkappa_2)$ cannot be fulfilled for every $R(T_0)$. However, it is of secondary importance if only $r_0(q) \gg r_0(\varkappa_2)$. On the other hand the condition $R(T_0) = 2nr_0(q)$ may always be fulfilled by a suitable choice of q. With the geometry of experiment as by Antonini (1967) we have

$$q = 2k_0 \sin\theta_B \cdot \delta\theta \tag{12}$$

where $2\theta_B$ is the angle between $\vec{k}_0$ and $\vec{k}$ and $\delta\theta$ is the angle of cristal rotation from the Bragg scattering position and $k_0 = 2\pi/\lambda_0$ is the incident wave vector length. From (11) and (12) we find for the angles at which the minima occur

$$(\delta\theta)_n = n\,\frac{\lambda_0}{2R\sin\theta_B}\,, \qquad n \text{ integer} \tag{13}$$

$$R = m\pi\varkappa_2^{-1}, \qquad m \text{ integer} \tag{14}$$

and the latter condition for R may be omitted for $r_0(q) \gg r_0(\varkappa_2)$ as already stated.

The cross section implies the dependence of the temperature at which the absolute maximum of critical scattering occurs on the scattering angle. The position of this maximum on the temperature scale moves towards higher temperatures with increasing the angle of rotation from the Bragg position. Therefore for larger angles of rotation from the Bragg position the temperature at which the absolute maximum of scattering occurs, may differ considerably from the Néel temperature. This effect has been already described in the case of ferromagnets (Kociński 1966) and subsequently detected in nickel by Stump and Maier (1967).

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3. The range of correlation R

In complete analogy with the ferromagnetic case (Kociński 1966) we shall assume that thermodynamic fluctuations in antiferromagnetic order are conditioned by fluctuations in temperture in the crystal lattice. The fluctuation in antiferromagnetic order occuppies the same region in space as the fluctuation in temperature. The range of correlation R between spins in one sublattice is identified with the most probable radius of the spherical fluctuation in temperature. Repeating the argument of our previous paper (Kociński 1966) we find for the range R the relation

$$R = \delta\left[4\pi\nu\left(1 - \frac{T}{T_0}\right)^2\right]^{-\frac{1}{3}} \tag{15}$$

where δ and ν have the same meaning as in (5) while T denotes the average fluctuation temperature and T_0 the system temperature. In analogy to the ferromagnetic situation the fluctuation temperature T is expected to remain in the immediate lower vicinity of T_N and to be almost independent on T_0. In the ferromagnetic case this prevision is supported by comparison with experiment, (Kociński and Mrygoń 1967). The value of T may be determined on the basis of experimental data as it has been done in the latter paper. The influence of the fluctuation temperature on the values of the parameters $\varkappa_2$ and Λ may be disregarded as in the ferromagnetic case. One can take approximately $\varkappa_2(T_0) = \varkappa_2(T_N)$, and $\Lambda(T_0) = \Lambda(T_N)$ for any value of T_0.

4 Comparison with experimental data

The experiments have been so far interpreted in the terms of the Ornstein-Zernike correlation. In particular the results for the magnitude of $\varkappa_1$ arrived at by Cooper and Nathans (1966) and Antonini (1967) show good agreement with those previously calculated by us which may be found on the basis of formulas (3a, b). Similar agreement between experimental and calculated values exist in the ferromagnetic case (Kociński 1965). Since there are reasons to think that the $|\sin \varkappa_2 r|/r$ correlation gives a better dersciption of the fluctuation phenomena than the $O-Z$ correlation, the question of a possible relation between the two correlation formulas arises.

Antonini (1967) found the dependence of the half width of the scattered spectrum on the square of the cristal missetting angle. His result is reproduced with the new cross tsection (8).

REFERENCES

Antonini M., *J. Phys. Chem. Solids*, **28**, 11 (1967).

Cooper M. J. and Nathans R., *J. Appl. Phys.*, **37**, 1041 (1966).

De Gennes P. G., *J. Phys. Chem. Solids*, **6**, 43 (1958).

Elliott R. J. and Marshall W., *Rev. Mod. Phys.*, **30**, 75 (1958).

Kociński J., *J. Phys. Chem. Solids*, **26**, 895 (1965); *Acta Phys. Polon.* **30**, 591 (1966), **32**, 13 (1968).

Kociński J. and Wojtczak L., *International Congress on Magnetism*, Boston 1967; *J. Appl. Phys.* **39**, 618 (1968a); *Acta Phys. Polon.* **34**, 1009 (1968b).

Kociński J. and Mrygoń B., *Phys. Letters*, **25A**, 600 (1967).

Stump N. and Maier G., *Phys. Letters*, **24A**, 625 (1967).

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