SPIN-PAIR CORRELATION IN IRON BY NEUTRON DIFFRACTION TECHNIQUE

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The small angle critical scattering of neutrons in iron was measured for different temperatures of the sample and for different incoming neutron wavelengths. Predicted by Kociński's theory diffraction maxima in the differential cross-section were found. The temperature shift of the maximum of scattered intensity was also discussed in the light of the latest theories and experiments.

1. Introduction

1) The neutron scattering cross-section

As it is well known since the paper of van Hove [1], the differential cross-section for neutron magnetic critical scattering is the Fourier transform of the spin correlation function. In the static approximation it has the form

$$\frac{d\sigma}{d\Omega} \sim \int \langle \hat{S}_0^z(0)\hat{S}_r^z(0)\rangle_{T_0} e^{i\vec{\varkappa}\vec{r}} d\vec{r}. \tag{1}$$

According to an accepted physical model of the critical scattering phenomena one can get for a ferromagnet either the Ornstein-Zernicke type correlation function or the Kociński's one. The 0—Z correlation has, above the Curie point ($T_0 \geqslant T_c$) the following form

$$\langle \hat{S}_0^z(0)\hat{S}_r^z(0)\rangle_{T_0} \sim \frac{e^{-\varkappa_1 r}}{r} \tag{2}$$

where

$$\varkappa_1^2 \sim \frac{1}{\chi}$$

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χ — paramagnetic succeptibility, T_0 — temperature of the sample, $\varkappa_1^{-1}$ can be treated as the range of spin correlation, or the radius of a fluctuation.

In this case, the differential cross-section in the static approximation is proportional to

$$\frac{d\sigma}{d\Omega} \sim \frac{1}{\varkappa_1^2+\varkappa^2} \tag{3}$$

where the scattering vector, for small angles,

$$|\vec{\varkappa}| = |\vec{k}-\vec{k}_0| = 2\pi\theta/\lambda_0.$$

Kociński's theory [2] operates with two forms of the spin correlation, the Ornstein-Zernicke form and $|\sin \varkappa_2 r|/r$. The 0—Z form describes the correlation of spins in the fluctuations of low abnormality, while the $|\sin \varkappa_2 r|/r$ form correlation of spins in the regions of the system with highly abnormal fluctuations.

In the oscillating function *i.e.*

$$\langle \hat{S}_0^z(0)\hat{S}_r^z(0)\rangle_{T_0} \sim \frac{|\sin \varkappa_2 r|}{r} \tag{4}$$

the parameter $\varkappa_2$ is connected with the periodicity of the crystal lattice; for instance in iron $\varkappa_2 = \frac{5}{a}$, where a is the lattice constant.

In contrast with $\varkappa_1$, $\varkappa_2$ cannot be treated as the radius of fluctuation. In order to find this radius, or the correlation range, one has to discuss in detail the phenomena of formation and decay of the fluctuations. In conclusion one finds for the range of correlation the formula:

$$R = a\left[4\pi v\left(\frac{T_0-T}{T_0}\right)^2\right]^{-1/3} \tag{5}$$

where: v — number of atoms in an unit cell, T — temperature of the fluctuations.

Correspondingly, the differential cross-section for neutron magnetic critical scattering has in elastic approximation, and for $T_0 \geqslant T_c$ the following form

$$\frac{d\sigma}{d\Omega} \sim C_1 \frac{A_1^2}{\varkappa_1^2+\varkappa^2} + C_2 \frac{A_2^2}{\varkappa_2 \varkappa} \int_0^R \sin \varkappa r |\sin \varkappa_2 r| dr. \tag{6}$$

Where A_1 and A_2 denote the amplitudes of the corresponding correlations and C_1 and C_2 are the statistical weights of the occurrence of these correlations.

In contradiction to van Hove's formula, this cross-section does not change monotonically with temperature or scattering angle, in particular there appear side maxima. The conditions for the scattering angles at which these maxima appear, for a constant temperature, follow from the form of the integral in the cross-section, and are given by

$$\Theta_n^{\max} = \frac{1}{2}(\Theta_n^{\min}+\Theta_{n+1}^{\min}) \tag{7}$$

where

$$\Theta_n^{\min} = n \cdot \frac{\lambda_0}{R} \qquad (n - \text{integer}) \tag{8}$$

determines the angle of the n-th minimum, and λ_0 is the incoming neutrons wavelength.

Since the two terms in the cross-section have a qualitatively different dependence on temperature or scattering angle, it might seem that the detection of their role in describing the real situation in the spin system should not be difficult.

2) Experimental conditions according to Kociński's theory

Neutron magnetic critical scattering was the subject of interest of many investigators. Riste, Jacrot, Shull, Gersh, Passel and others have interpreted their results in terms of

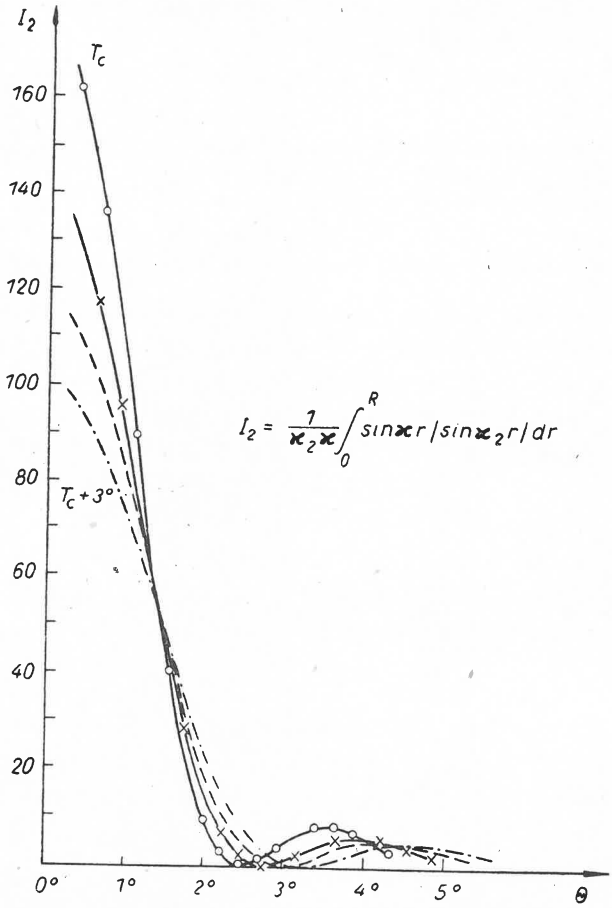

Fig. 1. Example of Kociński's theoretical cross-section for iron for given neutron wavelength. Curves represent this part of the cross-section which is connected with the $|\sin \varkappa_2 r|/r$ correlation only

van Hove's theory only. So one can come to the conclusion that these experiments forejudged finally the whole question.

However, as deduced from Kociński's theoretical cross-section for iron (see Fig. 1), a temperature change in sample of 1°C causes a neutron intensity change in the first side maximum of about 40%. So, to detect this maximum, a very high temperature stability of the sample has to be applied. The intensity of neutrons critically scattered falls very rapidly with the scattering angle. To get these maxima in a region in which the intensity can be well observed, one has to use incoming neutrons of small wavelengths; in case of iron about 1 Å (see formula (7)). As shown in Fig. 1 the half-width of the side maxima is of about several tens of min. of arc. Therefore collimators used during the experiment should give the final divergence of the neutron beam not more then several min. of arc.

The angular position of the side maximum is also determined by the temperature of the sample (see Fig. 1). The intensity in the peak will reach its maximum for $T_0 = T_c$. For temperatures $T_0 > T_c$ this intensity becomes much lower, and the whole side maximum moves to larger scattering angles. So the choice of the proper sample temperature is also of great importance, to say nothing about the temperature gradient over the sample.

Summarizing — the finding of the maxima in the measured intensity depends on the proper selection of the experimental parameters.

3) The temperature shift of the maximum of scattered intensity

Theories developed after van Hove [1] identified always the temperature of the maximum of the intensity of critically scattered neutrons with the ferromagnetic Curie point.

According to the recent theory by Fisher and Burford [3] the maximum should be shifted on the temperature scale proportionally to the scattering vector in the power 1.55, starting from zero scattering angle. One the other hand Kociński [4], [5] points out that such a shift does not start from zero angle but appears for some larger value of the scattering vector $\varkappa$ and incrases with $\varkappa$ in a power higher than 3/2.

The essential difference between former theories based on the O—Z type spin pair correlation function and Kociński's work lies in the basic approach to the problem. The final consequences of these different treatments required hence an experimental verification.

2. Experiment

1) Sample and furnace

The policrystalline Fe sample was cut from an Armco block and had approximate dimensions $5 \times 2 \times 0.5$ cm. The sample mounted on a ceramic holder was heated by a coil wound on a ceramic tube 17 cm long. The temperature of the sample was measured by two thermocouples, one mounted inside the sample and the second at its bottom. The temperature

was controlled by an electronic controller and was stable to within ± 0.1 °C. The temperature gradient over the sample is believed to have been within the same limit since otherwise no effect could have been observed.

2) Neutron spectrometer

A triple axis spectrometer was used for the small angle measurements. To reduce the neutron background two Zn crystals in the (002) reflection were applied to work as a double crystal spectrometer (see Fig. 2). A Soller collimator with 13 min. of arc. horizontal divergence was placed in front of the BF_3 counter.

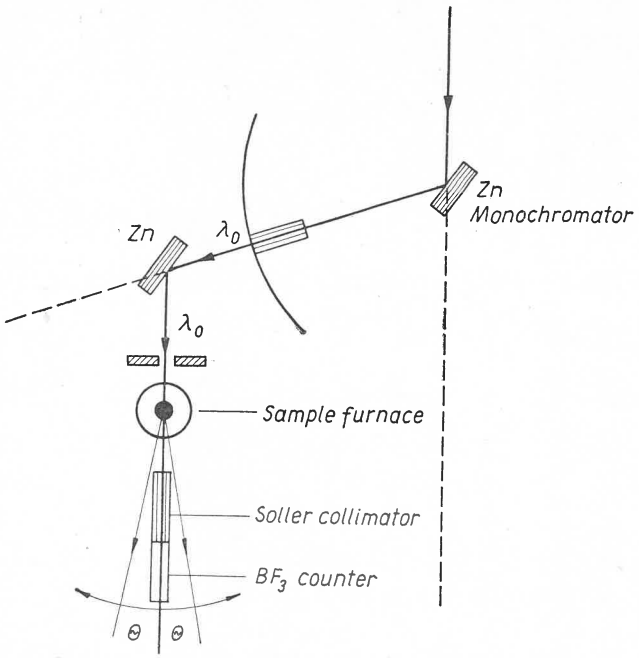

Fig. 2. Experimental set-up used for small-angle neutron magnetic critical scattering measurements

3) Determination of the critical temperature

As it was mentioned, the angular position of the side maximum is determined by the temperature of the sample. The intensity in the peak will reach its maximum for $T_0 = T_c$. For temperatures $T_0 > T_c$ this intensity becomes much lower and the whole maximum moves to a larger scattering angle (see Fig. 1). Hence only temperature differences from T_c were of importance, and the aim was therefore to determine the thermocouple voltage corresponding to the critical state of the sample. But as shown in Fig. 3 there exists a "temperature shift" of the maximum of intensity. Therefore it was assumed that the Curie temperature is equal to the temperature of these main maxima for which the "temperature shift" does not begin to appear.

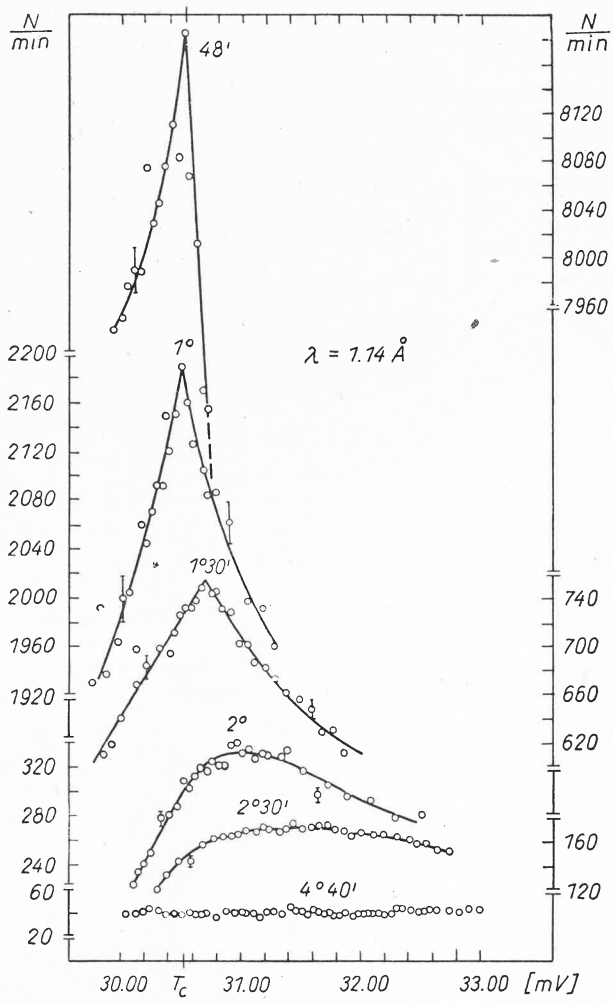

Fig. 3. Intensity of neutrons scattered by iron in the critical temperature range for different scattering angles. Curie temperature corresponds to the 30.53 mV thermocouple voltage

3. *Experimental data*

1) Small angle measurements

Small angle critical magnetic scattering of neutrons in iron have been determined for different temperatures of the sample and for different wavelengths of the incoming neutrons, as earlier reported in [6]. The obtained results are shown in Fig. 4 and Fig. 5. The temperature of the sample was kept with a stability better than ± 0.1°C. The circles representing the neutron intensity cover the statistical error of the counts. The angular position of the side maxima, according to Kociński's prediction, changes with the temperature of the sample as well as with the neutron wavelength. For the temperature of the sample $T_0 = T_c$

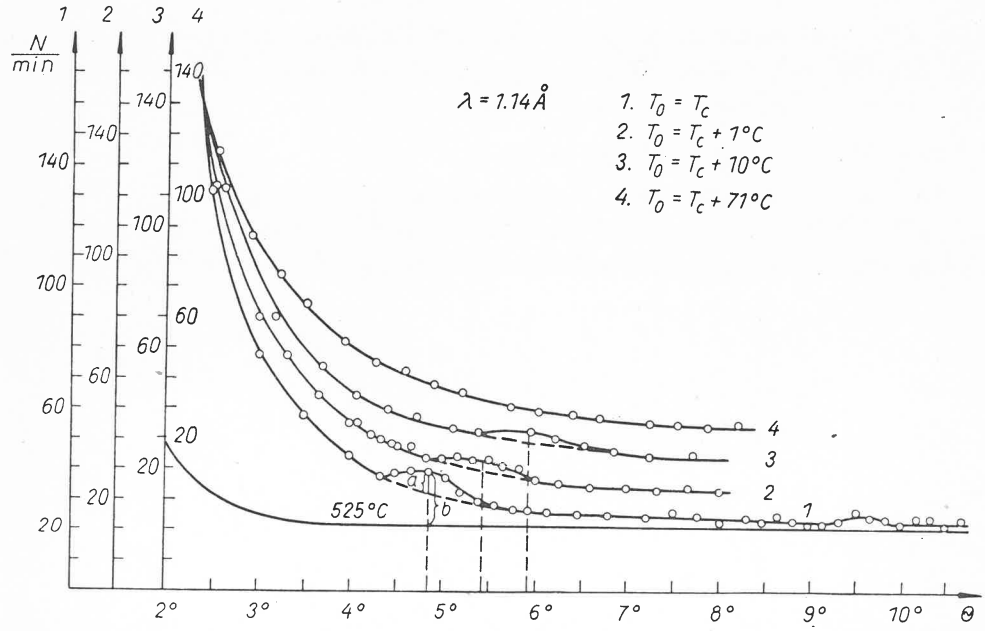

Fig. 4. Small-angle critical magnetic scattering of neutrons in iron for different temperatures. Sample temperature stability ±0.1 °C. The circles cover the statistical error of the counts. The height a of the first side maximum for sample temperature $T_0 = T_c$ is of about 40% of the whole height b measured from the background level obtained for $T_0 = 525$ °C

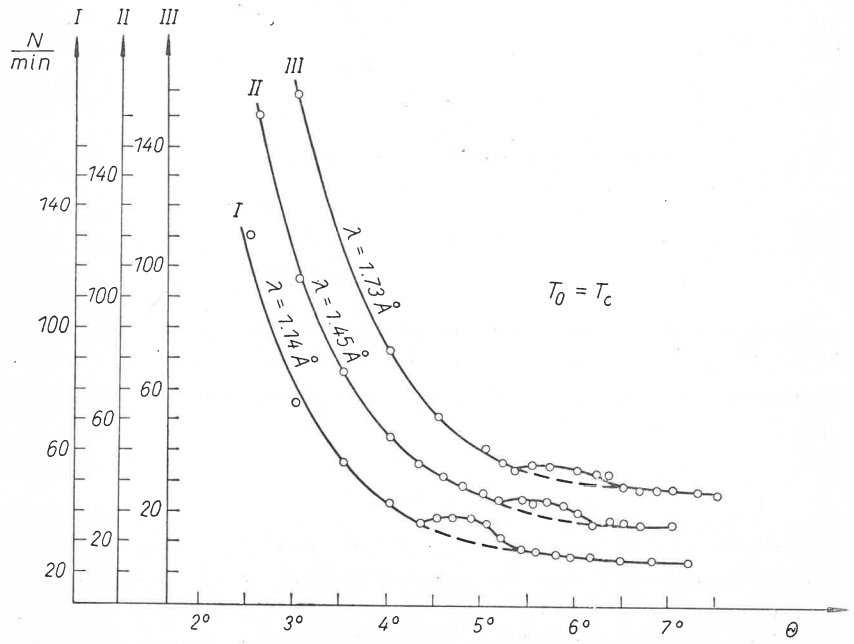

Fig. 5. Small-angle critical magnetic scattering of neutrons at the Curie point for different wavelengths. The angular position of the side maximum moves with the wavelength. Sample temperature stability better than ±0.1 °C. The circles cover the statistical error of the counts. No background subtracted

a trace of the 2-*nd* maximum can be observed (see Fig. 4). On the same curve the height of the first side peak is about 40% of the parameter b. It should be also mentioned that for all curves no background was subtracted.

2) "Temperature shift" determination

Fig. 6 shows the dependence obtained for $\Delta T = T_m - T_c$ *versus* $\varkappa = 2\pi\Theta/\lambda_0$ and reported earlier in [7]. Neutrons of three different wavelengths $\lambda_0 = 0.92$ Å; 1.29 Å and 1.53 Å were used. The temperature shift begins at the larger scattering angle Θ with increasing

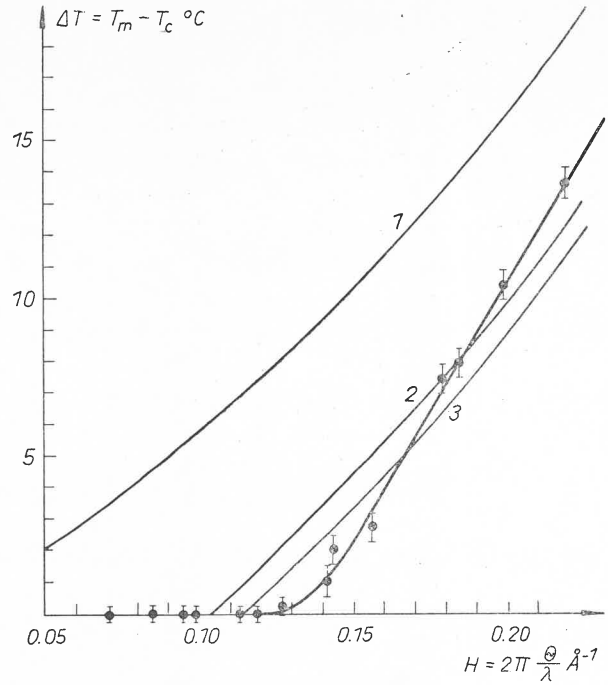

Fig. 6. Experimental points show the dependence of $\Delta T = \Delta T_m - T_c$ *versus* scattering vector $\varkappa$. Theoretica curves *1*, *2*, *3* are calculated according to [5] for $n = 0$

value of λ_0. Fig. 6 indicates that the shift begins to appear at $\varkappa \cong 0.125$ Å^{-1}. No shift was detected for smaller $\varkappa$. The theoretical curves 1, 2, 3 are calculated from the formula (6) given in [5] with $n = 0$. They correspond to three temperatures of fluctuation $T = T_c$; $T_c - 6$°C and $T_c - 7$°C respectively.

4. Discussion

The temperature shift has been first reported for nickel by Stump and Maier [8] who suggested to interpret this result as the effect predicted by Kociński [4]. Bally *et al.* [9] have observed the shift for Fe and Co and compared their results with Fisher's theory [3]. Our measurements (Fig. 6) indicate that the shift begins to appear at $\varkappa \cong 0.125$ Å^{-1}, which is in good agreement with Kociński's calculation, namely $\varkappa = 0.112$ Å^{-1}. The increase of the observed shift is faster than the calculated one, because the formula (6)

given in Ref. [5] is derived from an expression for the cross-section which does not include the temperature dependence of the number of spins inside a fluctuation N_1, as well as the density of fluctuations N_f introduced in Ref. [10]. One may expect that taking into account both these parameters N_1 and N_f will give a better agreement between experiment and theory. However, one more reason can cause the faster increase of the "temperature shift", namely crystalline imperfection. Stump and Maier [11] have found in nickel a strong dependence between the shift of maximum intensity and the dislocation density, however, they did not prove that there is no shift for a perfect crystal, since this would require larger scattering vector values than those applied by them.

The aim of this work consisted also in verifying Kociński's prediction, that the angular position of the side maximum should move towards larger Θ's with increasing temperature and should be proportional to the neutron wavelength for constant temperature of the sample. The verification of this prediction is important since it points out in a straightforward way the role of the $|\sin \varkappa_2 r|/r$ correlation independently on the particular theoretical formula for the range of correlation R.

The small angle scattering at the critical temperature was measured for three neutron wavelengths, namely 1.14 Å; 1.45 Å and 1.73 Å (see Fig. 5). With increasing λ_0 values the angular position of the maximum moves towards larger Θ regions in qualitative agreement with the theory. The intensity in the peak decreases with the increase of the wavelength or the temperature. However the angular distance between the maxima for two different wavelength is smaller than it follows from the formula

$$\Theta^{\max}(\lambda_2) = \Theta^{\max}(\lambda_1)\,\frac{\lambda_2}{\lambda_1} \tag{9}$$

mplied by Eqs (7) and (8). The reason for these discrepancies is sought in the effect of nelasticity unaccounted for, as well as in the fact the experimental curves represents a superposition of two terms of the cross-section, one connected with the O—Z correlation and the other with the $|\sin \varkappa_2 r|/r$.

A quantitative interpretation of the obtained results will be possible after having developed the forms of both correlation functions for small r, for which the assumption of spherical symmetry is questionable [12].

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