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NEUTRON SCATTERING INVESTIGATIONS OF THE DYNAMICS OF CRITICAL STATE IN IRON

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BUDAPEST

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Abstract

The inelasticity of critical scattering has been investigated in iron with the use of pulsed monochromatic neutron beam at the IBR-1 reactor in Dubna. The measurement covered the temperature range $-2{,}5\cdot 10^{-3} \leq \tau \leq 6{,}5\cdot 10^{-3}$ where $\tau \equiv T/T_C - 1$ and the critical temperature was approached within $\tau < 5\cdot 10^{-4}$.

The slight temperature dependence of the inelasticity was confirmed for $q = 5{,}7\cdot 10^{-2}$. $Å^{-1}$, but the observed diffusion constant was found to be temperature independent and higher $(\mu = 16{,}5)$ than the value $(\mu = 11)$ reported earlier by other authors.

1. Introduction

Neutron scattering experiments offer one of the most powerful methods for the investigation of the critical magnetic state. Owing to its favourable magnetic and neutron scattering properties, iron has been extensively studied by this method.

The static properties of fluctuations i.e. the temperature dependence of the correlation range and the susceptibility of iron were measured by several authors [1] [2], [3], [4] and the correlation range and susceptibility were found to be $\xi(\tau) \sim \tau^{-0,68}$ and $\chi(\tau) \sim \tau^{-1,33}$; $\tau = T/T_c - 1$ respectively.

The dynamics of the magnetic fluctuations were first studied by Van Hove [5] who introduced a thermodynamical spin diffusion model. This model predicts a slowing down of the fluctuations near the Curie temperature. A similar result was obtained from several microscopic theories [6], [7], [8].

The cold neutron experiments of Jacrot, Konstantinovic, Parette and Cribier [9] showed, however, an unexpected broadening of the energy distribution even near the Curie point. Later, Passell, Blinowski, Brun and Nielsen [10] confirmed this observation and evaluated a temperature independent spin diffusion constant as

This contradiction of the experimental results with the theory suggests that some of the processes involved have not been taken into account in the thermodynamical fluctuation model. Marshall [11] and Brout [12] have pinted out that for $q\xi > 1$, where $\hbar q$ is the momentum transfer of neutrons in the scattering process, collective excitations driven by fluctuation of magnetization may occur even above the transition temperatures.

The spin-wave-like behaviour for relatively high momentum transfer above the Neel point was observed by Nathans, Menzinger and Pickart [12] in antiferromagnetic $RbMnF_3$. No similar effect was found, however, in metallic ferromagnets.

Considering that in the recent inelastic scattering experiments the closest approach to the Curie point was $\tau = 2 \cdot 10^{-3}$ [10], [11], it was thought of interest to investigate the temperature dependence of the inelasiticity in closer vicinity of T_c.

The results of the first experimental run are now reported.

2. Experimental

The spectrometer and the detection electronics as well as the methods of temperature stabilization, homogenization have been already described in an earlier report [13].

The moderated neutron beam of the pulsed reactor IBR-1 is monochromatized by a magnetite monocrystal. The first order (111) reflexion centred around the wavelength of 4,10 Å is used. A 24 min. Soller collimator located between the monocrystal and the detector determines the wavelength resolution of the system.

The overall moderator-to-detector distance is 16,10 m, while the sample-to-detector vacuum flight path is 6,40 m. The scattered, transmitted or incident neutrons are counted by detectors of 11x13 cm^2 surface. In the first set of experiments the detectors were located at fixed angle $\vartheta = 2{,}16^\circ$. The pulses are transferred to a multichannel analyzer with channel width $\theta = 16\ \mu sec$ and dead time $\tau = 25\ \mu sec$. The analyzer stores the counts of all the higher order reflexions of the monocrystal and the fast neutron burst from the reactor, too. The fast neutron burst determines the "O" mark of the time scale. A 0,5 cm thick, 10 cm long and 3,5 cm wide, zone-melted "Puron" iron sample of 99.99 % purity was used with a 3x3 cm^2 surface exposed to the neutrons. The temperature stability and homogeneity was found to be better than $0,2^o$ during a 100 hour operation. The ratio of the scattered to transmitted intensity showed a maximum at $T_C = 1041\ ^oK$. Measurements were performed in $0,5^o$ steps around the Curie point from $T-T_C = -1,5^o$ to $1,5^o$ and at temperatures $T_C - 2,5^o$ and $T_C + 6,5^o$.

3. Data evaluation

Since the counting rate corrections and the choice of the energy and angular resolution functions play a decisive role in the evaluation of the parameters of inelasticity, the procedure used for the evaluation of the experimental data will be described in some detail.

3.1 Analyzer dead time corrections

In the case of pulsed reactors the counting rate obtained during a reactor pulse may be so high as to result in an appreciable counting loss, for analyzers with conventional dead time. Thus, it was necessary to work out a suitable method of dead time correction. The dead time corrections were found to be especially important in the measurement of incident beam distribution.

Assuming the neutron counts to have a Poissonian distribution the number of counts in the i-th channel with correction for dead time is given by the formula

$$C_i = N \ln Y_i$$

where N is the number of reactor pulses and Y_i is given by the equation

$$Y_i = (N - m_{i-1} - m_{i-2} - m_{i-2} Y_i^K)/(N - m_i - m_{i-1}) \ ; \quad K = (\tau - \theta)/\tau$$

where m_i is the measured number of counts in the i-th channel.

The validity of the correction was checked by comparison of the corrected spectrum data with those measured at a small counting rate.

3.2 Background corrections

It was observed that a major fraction of the background of the scattered beam is due to diffuse incoherent scattering from the collimator plates. Whenever the background intensity originates from the "forward" direction /i.e. from neutrons transmitted by the sample/ the intensity has to be corrected by use of the formula

$$n_k = N_k - B_k f_k$$

where n_k and N_k are the corrected and measured intensities B_k is the background in the k-th channel, respectively, f_k is the /temperature dependent/ transmission factor.

3.3 Determination of the input beam parameters

Assuming a Γ -type time distribution of the neutrons which have left the moderator, the probability of a neutron count in the i-th channel is

$$P(t_j) \sim \sum_{j=0}^{i} t_j^{\alpha} \exp\left\{-t_j/\tau\right\} \exp\left\{-\left[(t_i - t_j - t_o)/\rho t_o\right]^2\right\} f(t_i - t_j). \qquad /1/$$

Here t_j is the time at which the neutron leaves the moderator, $t_i - t_j = c \cdot \lambda$, $t_o/c = \lambda_o$ is the maximum of the Gaussian wavelength distribution, and $f(t_i - t_j)$ is the correction for detector efficiency. The parameters α, τ, ρ and t_o as determined by least square fit are $\alpha = 0{,}69 \pm 0{,}07$; $\tau = 133 \pm 10$ μ sec, $\rho = 8{,}7 \pm 0{,}7 \cdot 10^{-3}$; $t_o = 16420 \pm 16$ μsec. The measured distribution and the best fit are shown in Fig. 2.

3.4 Correction for angular resolution

Owing to the finite dimensions of the source, sample, detector and collimator, the measured effect is proportional to the value of the scattering cross section averaged over all possible angles permitted by the geometry. The geometry of a system determines a weight function $A(\vartheta)$ of the scattering angles. Thus, the average cross section is given by

$$\overline{\sigma(\vartheta)} = \int \sigma(\vartheta) A(\vartheta) d\vartheta \approx \sum_{k=1}^{m} A(\vartheta_k) \sigma(\vartheta_k) \Delta\vartheta$$

The distribution function $A(\vartheta)$ was evaluated by Monte Carlo method for the geometry described in § 2. It proved to be sufficient to sum up till m=10. Fig. 2a shows the distribution function $A(\vartheta)$. In Fig. 2b the cross section $\sigma(\vartheta_{av})$ is compared with the averaged $\sigma(\vartheta) = \sum \sigma(\vartheta_k) A(\vartheta_k)$ as a function of the wavelength plotted on time scale. Since the intensity of each element of the scattered beam is proportional to ϑ^2 and its half-width on the wavelength scale is $\mu\vartheta^2$, it is not surprising that the averaged cross section is narrower than the cross section evaluated for the average angle. In our case the difference in half-width is about 30 %.

Two approximations implied in the above calculations have to be pointed out. First, the beam incident on the Soller collimator is taken to be isotropic. This approximation is reasonable since the angular divergence of the beam before the monocrystal is greater than that of the collimator. Second, λ_i is assumed to be independent of ϑ , since the mosaic spread of the monocrystal is not greater than that of the collimator.

3.5 Distribution function of scattered neutrons

Supposing that the inelastic scattering is due to the diffusive motion of spins, the cross section for small angle scattering near T_C is given as

$$\sigma(\lambda_i,\lambda_S;\vartheta,K_1,\mu)\Delta\lambda_S=\mathrm{const.}\lambda_i z x^3\left[(K_1\lambda_i/2\pi)^2+zx\right]^{-1}\left[\mu^2 z^2+(x-x^{-1})^2\right]^{-1}\Delta\lambda_S$$

where the following notations are used.

$x=\lambda_i/\lambda_S$, λ_i being the incident, λ_S the scattered wavelength

$$z=x+x^{-1}-2\cos\vartheta \quad ; \quad \mu=\Lambda 2m/\hbar$$

is the spin diffusion coefficient.

Considering that $\lambda_o=ct_o$; $\lambda_i=c(t_i-t_j)$ and $\lambda_S=(t_\ell-t_j-\ell_R t_i)/(1-\ell_R)$ where ℓ_R is the "reduced flight path"; $\ell_R=\ell/L$, ℓ being the sample-to-moderator, and L the moderator-to-detector distance, the experimental curve was fitted to the distribution function

$$R(t_\ell)\sim\sum_{j=0}^{i}\left[t_j^{\alpha}\exp\left\{-t_j/\tau\right\}\sum_i\exp\left\{-\left[(t_i-t_j-t_o)/\varrho t_o\right]^2\right\}\times\right.$$

$$\left.\times\sum_k A_k\sigma(\vartheta_k;t_i,t_j;t_\ell,K_1,\mu)\cdot f(t_i,t_j,t_\ell)\right] \quad . \qquad /2/$$

Since in the neighbourhood of the Curie point K_1^2 does not play any important part in the evaluation of the diffusion parameter, its values were replaced by those obtained from angular distribution measurements as

$$K_1^2(T_C)=0 \quad , \quad K_1^2(T_C-2{,}5^\circ)=4\cdot 10^{-4} \quad , \quad K_1^2(T_C+6{,}5^\circ)=5{,}7\cdot 10^{-4}$$

All calculated and measured distributions were normalized to unit area.

4. Results and discussion

Before evaluating the diffusion parameters by the method described in the previous section the distribution measured at T_C was compared with that measured at $T_C-2{,}5^\circ$ and at $T+6{,}5^\circ$ using Smirnov's statistical test [14] .

The distributions measured above, and below the Curie point, were found to be broader as compared with that measured at T_C . The reliability of the disagreement with the Curie point curve was higher than 90 %. Then, the normalized distribution /2/ was fitted to the experimental curves making use of K_1^2 values listed at the end of the previous section. The calculations yielded a temperature - independent diffusion constant:

$$\mu = 16{,}5$$

The best fit and the experimental curve for T_C are shown in Fig. 3.

The disagreement between the diffusion constant evaluated from the present measurement and the values reported by other authors can be attributed most probably to the different procedures of data evaluation, particularly to the fact that no corrections for angular resolution have been made in the earlier calculations of the diffusion constant. It is apparent from Fig. 2a that without correction for angular resolution, we would have $\mu = 13$, a value which is close to the recently reported $\mu = 11$ [10] . Of course other reasons, like difference in sample purity, may also explain some of the disagreement.

Finally, it is of interest to note that in this experiment $q\xi > 2{,}5$, thus the temperature independent diffusion constant is related to processes occurring within a region of a radius smaller than the correlation length. With the present experimental technique it would be difficult to get any information about processes for which $q\xi < 1$.

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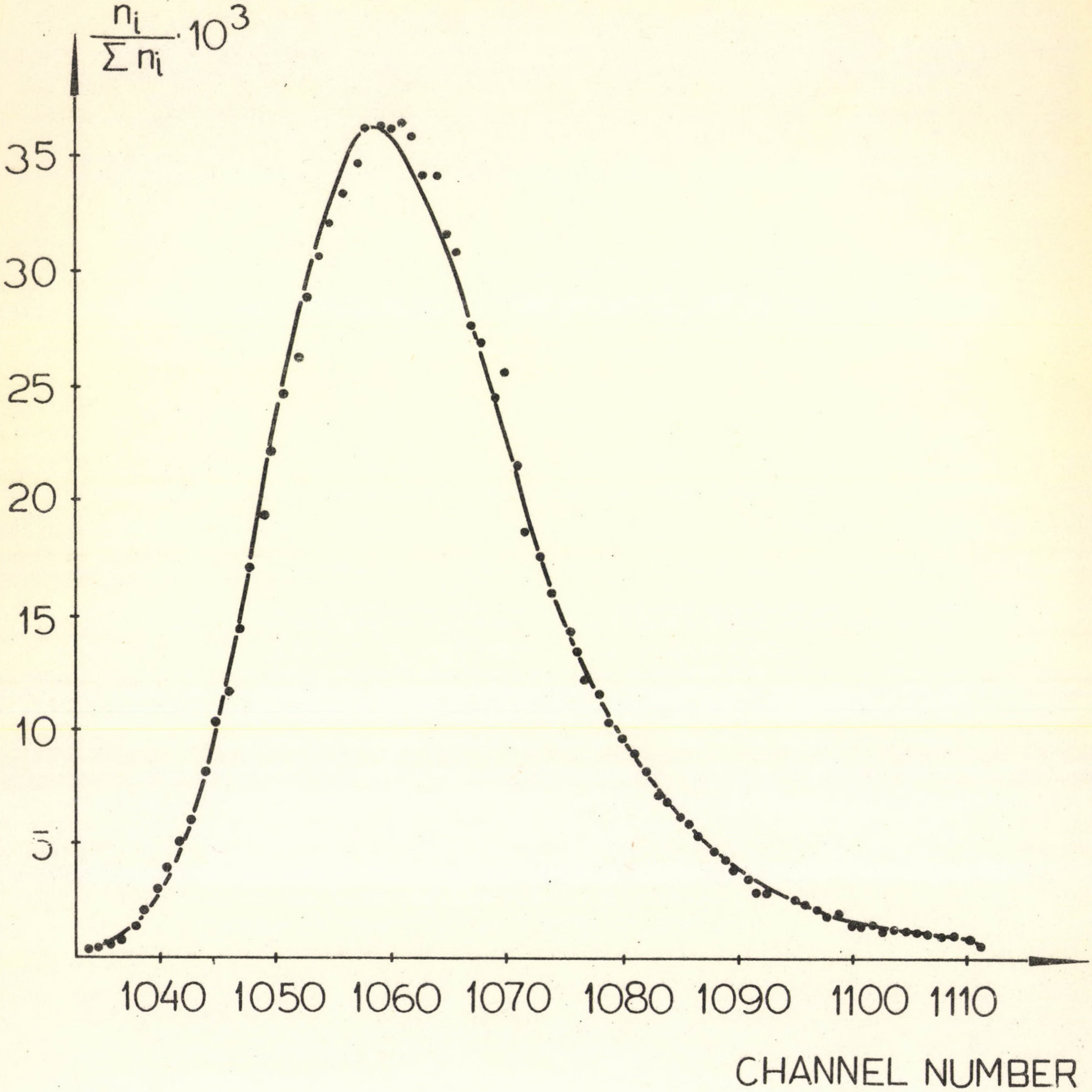

Fig. 1. Incident beam spectrum

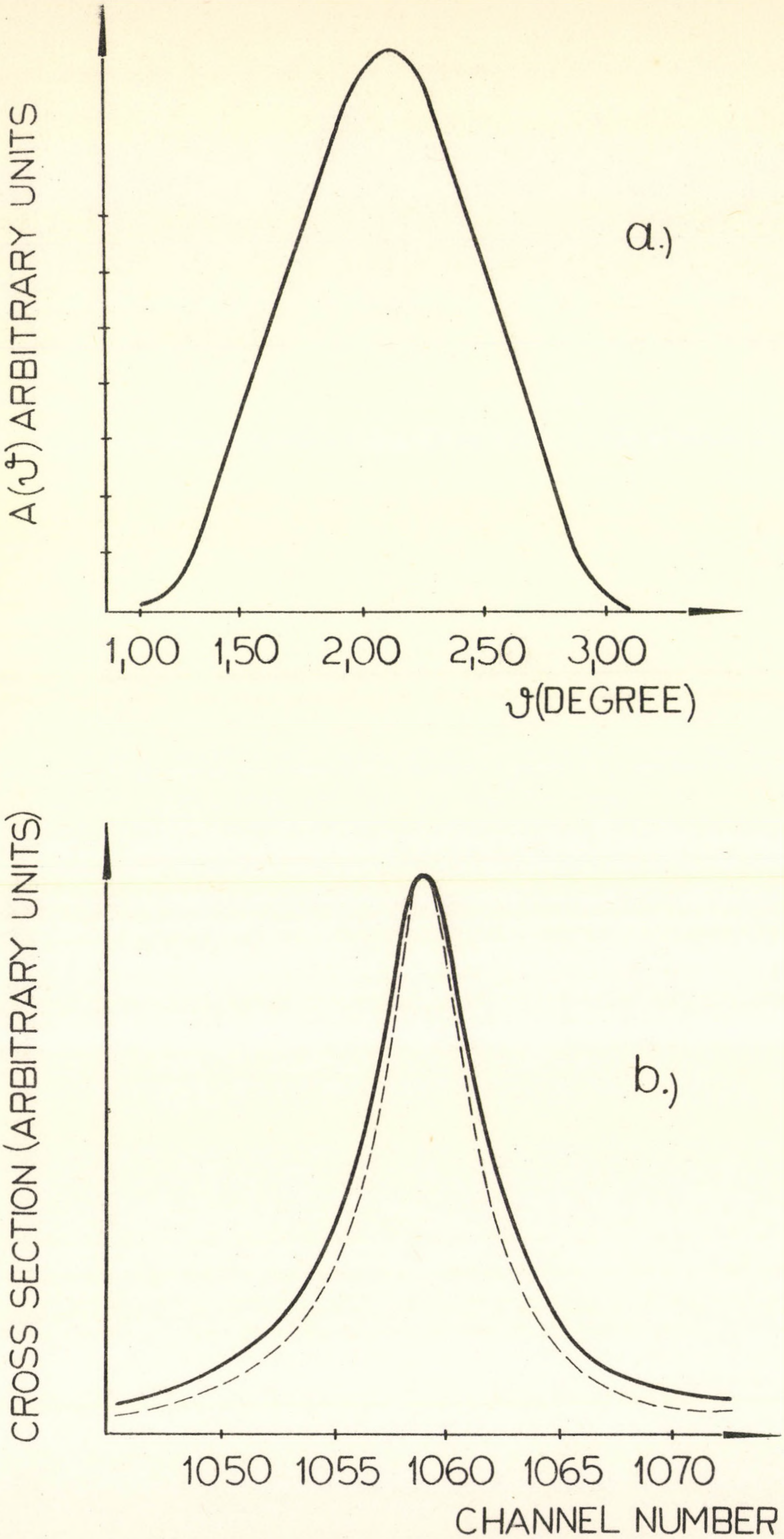

Fig. 2. a./ The $A(\vartheta)$ distribution function

b./ The scattering cross section: without angular integration /full line/ and with angular integration /dotted line/

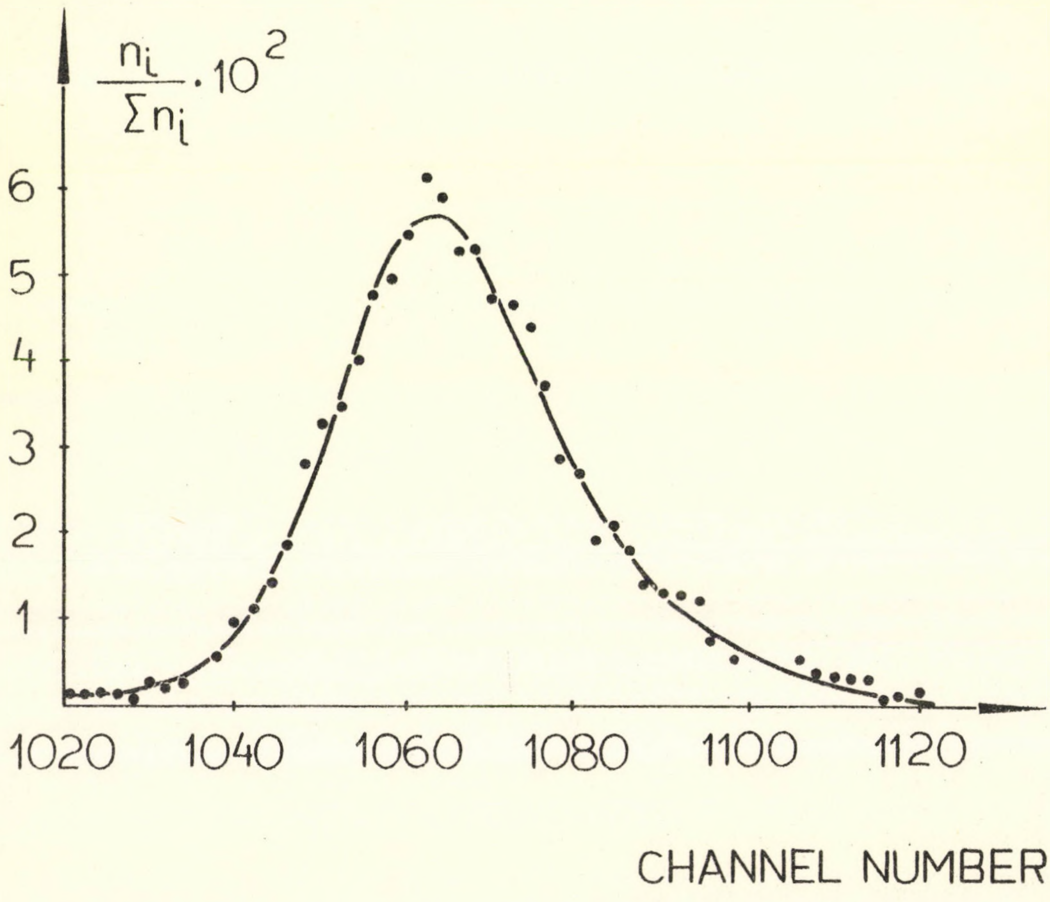

Fig. 3. Scattered neutron distribution: measured counts and the best fit curve at $T = T_C$

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