Critical Depolarization of Neutrons Traversing a Ferromagnetic Body

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Depolarization of neutrons passing through a ferromagnet in the paramagnetic phase near the Curie point is analyzed theoretically. The depolarization is expressed in terms of the small-angle magnetic scattering cross section. Experimental data on depolarization, critical scattering and susceptibility data for nickel are presented. If parameters derived from scattering and susceptibility data are employed, the depolarization observed near T_c is found to exceed the theoretical value by more than an order of magnitude. This means that the scattering in the small-angle region is excessive compared to the ordinary critical scattering. It is suggested that this excessive scattering may signify that in reality the transition of nickel to the ferromagnetic state is a first-order phase transition but close to that of second order.

1. INTRODUCTION

RECENTLY there has been a more diligent study of the depolarization of neutrons passing through a ferromagnet near the Curie temperature (see the papers of Drabkin and co-workers[1-4] and also the paper by Rauch[5]). It was observed that when the temperature is lowered the polarization $P(T)$ of the transmitted beam changes rapidly, from a maximum value equal to the polarization of the incident neutrons P_0 to zero in a narrow temperature interval on the order of one degree near T_C. A study of the $P(T)$ curve has made it possible to increase by approximately one order of magnitude the accuracy with which the Curie temperature is determined, in comparison with other methods (the quantity $\tau = (T - T_C)/T_C$ can be determined from the $P(T)$ curve with an error on the order of several times 10^{-5}). It has turned out here that in the immediate vicinity of the Curie point the depolarization is quite large $(P(T \approx T_C) \sim (0.6 \times 0.7)P_0)$, and below T_C the sample turns out to be inhomogeneously magnetized. Finally, it was observed that in the paramagnetic phase $\ln[P(T)/P_0] \sim \lambda^2$, where λ is the neutron wavelength. The present paper is devoted to a theoretical analysis of the question of the depolarization in the paramagnetic phase, which is caused by magnetic scattering from critical fluctuations and is expressed in simple fashion in terms of the cross section of the magnetic scattering through angles that do not exceed the divergence angle of the incident beam. This was followed by an analysis of the extent to which the depolarization data agree with the present notions concerning second-order phase transitions (scaling theory). The theory has been compared with experiment using as an example nickel, for which there are at present many experimental data on the critical scattering[2,6], depolarization[1-3], and the magnetic susceptibility above T_C[7,8].

The main result of this comparison reduces to the following: whereas the data on the susceptibility and the critical scattering fit well within the framework of the concepts of similarity theory, data on the depolarization cannot be reconciled with these concepts if one uses only the parameters obtained from experiments on the susceptibility and critical scattering, for in this case the depolarization turns out to be smaller by more than one order of magnitude than the observed one. Therefore, on the basis of the connection between the depolarization and the cross section under the experimental conditions of[1-3], we conclude that there exists small-angle scattering in excess of the critical scattering. This "excess" scattering exists in a very narrow temperature interval ($\tau \sim 10^{-4}$) and in the region of very small momentum transfers ($q < 3 \times 10^{-3}\,\text{Å}^{-1}$), where no direct experiments on scattering have been performed as yet. One of the possible explanations of this excess scattering is the existence of unique "quasidomains," which are nuclei of a new phase, and which indicate that actually the transition to the ferromagnetic state is a first-order phase transition. However, further experimental and theoretical research is necessary to be able to make more exact statements.

2. CONNECTION BETWEEN DEPOLARIZATION AND SCATTERING

We consider a ferromagnet at temperatures higher than the Curie temperature, i.e., in the paramagnetic phase. The magnetization in this ferromagnet fluctuates. On the basis of the classical equation of motion of the polarization vector

$$\dot{\mathbf{P}} = 2\mu_n\hbar^{-1}[\mathbf{BP}], \qquad (1)^*$$

it is easy to verify that the rotation of the polarization vector due to one fluctuation is small if the condition $\mu_n\overline{B}R/v\hbar \ll 1$ is satisfied, where R is the radius of the fluctuation, $\overline{B}$ the average fluctuation in it, and v the neutron velocity. If we use for R the expression $R = \alpha\tau^{-2/3}$, which follows from similarity theory (a is a quantity on the order of the lattice constant), then this condition is in splendid agreement even at $\tau \sim 10^{-5}$ and at any reasonable values of $\overline{B}$ and $v \sim 10^4$–10^5 cm/sec. Consequently, the interaction of a neutron with such a fluctuation can be treated by perturbation theory. On the other hand, the interaction of a neutron with a fluctuation of radius R, by virtue of the un-

*$[\mathbf{BP}] \equiv \mathbf{B} \times \mathbf{P}$.

certainty relation, leads to scattering of the neutron through an angle $\vartheta \sim (pR)^{-1}$, where $\hbar p$ is the neutron momentum. Therefore the interaction of the neutron with the ferromagnet above T_C should be regarded as scattering. Consequently the transmitted beam consists both of neutrons which have not experienced even a single scattering act, and of neutrons experiencing one or more such acts of scattering through angles smaller than or of the same order as the divergence of the incident beam, and the entire depolarization is due to the scattered neutrons.

In the absence of an external magnetic field and when it is certain that the energy transfer ω is small compared with the temperature, the scattering cross section is given by (see, for example, the paper of Collins et al.[9])

$$\frac{d\sigma}{d\Omega d\omega} = \frac{2}{3} r_0^2 \gamma_0^2 \frac{T}{\pi\omega} \operatorname{Im} G(\mathbf{q}, \omega),$$
$$G(\mathbf{q}, \omega) = i \int_0 dt\, e^{-i\omega t} \sum_{\mathbf{R}} e^{i\mathbf{qR}} \langle [S(\mathbf{R}, t), S(0,0)] \rangle. \tag{2}$$

The function $G(\mathbf{q}, 0)$ is proportional to the static magnetic susceptibility $\chi(\mathbf{q})$ and is connected with Im G by the dispersion integral

$$\chi(\mathbf{q}) = \frac{(g\mu)^2 N_0}{3T} G(\mathbf{q}, 0), \tag{3}$$

$$G(\mathbf{q}, 0) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \operatorname{Im} G(\mathbf{q}, \omega); \tag{4}$$

here N_0 is the density of the magnetic unit cells and g is the gyromagnetic ratio.

It should be noted that formula (2) is customarily used in the case of unpolarized neutrons. It is also valid, however, when the neutrons are polarized (see the paper by one of the authors[10]). The magnetic scattering of polarized neutrons is accompanied by a change in their polarization vector, which in the paramagnetic phase is determined by the equality[10]

$$\mathbf{P} = -\mathbf{e}(\mathbf{e}\mathbf{P}_0), \tag{5}$$

where $\mathbf{e} = \mathbf{q}/q$ is a unit vector in the direction of the momentum transfer.

Let the sample be sufficiently thin, so that the multiple scattering can be neglected. In this case the polarization of the transmitted neutrons can be represented in the form

$$\mathbf{P} = \frac{\mathbf{P}_0[1-(\sigma_1+\sigma_2)N_0L] + \bar{\mathbf{P}}\sigma_1 N_0 L}{1 - N_0 L \sigma_2} = \mathbf{P}_0 + (\bar{\mathbf{P}} - \mathbf{P}_0)\sigma_1 N_0 L, \tag{6}$$

where L is the thickness of the sample, σ_1 is the cross section for the scattering of neutrons in a narrow cone of angles corresponding to the transmitted beam, σ_2 is the cross section for scattering outside this cone, and $\bar{\mathbf{P}}$ is the average polarization of the neutrons scattered within the confines of the transmitted beam. It follows from (5) that

$$\bar{P}_\alpha = -\Delta_{\alpha\beta} P_{0\beta},$$
$$\Delta_{\alpha\beta} = \frac{1}{\sigma_1} \int d\Omega d\omega \frac{d\sigma}{d\Omega d\omega} e_\alpha e_\beta, \tag{7}$$

where the integral with respect to the angles is taken within the confines of the transmitted beam.

If the incident beam has a circular cross section, then the tensor $\Delta_{\alpha\beta}$ can be written in the form

$$\Delta_{\alpha\beta} = A\delta_{\alpha\beta} - Bn_\alpha n_\beta, \tag{8}$$

where $\mathbf{n}$ is a unit vector in the direction of the incident beam. From (7) and (8) we obtain

$$A = B + \frac{1}{\sigma_1} \int d\Omega d\omega (\mathbf{en})^2 \frac{d\sigma}{d\Omega d\omega},$$
$$3A - B = 1. \tag{9}$$

If the scattering is also elastic, then the vectors e and n are perpendicular and $A = B = \frac{1}{2}$. It follows from (8) that

$$\bar{\mathbf{P}} = -A\mathbf{P}_0 + B\mathbf{n}(\mathbf{P}_0\mathbf{n}). \tag{10}$$

Let us expand all the polarization vectors into parts parallel and perpendicular to the incident beam; we then obtain from (6), (9), and (10)

$$P_{\parallel} = P_{0\parallel}[1 - 2(1-A)\sigma_1 N_0 L],$$
$$P_{\perp} = P_{0\perp}[1 - (1+A)\sigma_1 N_0 L]. \tag{11}$$

These formulas describe the polarization of neutrons passing through a thin sample. If the cross section σ_1 does not depend on the maximum divergence angle ϑ_m of the incident beam (the entire scattering is through angles that are small in comparison with ϑ_m), then the polarization of neutrons passing through a thick sample is determined by the formulas

$$P_{\parallel} = P_{0\parallel} \exp[-2(1-A)\sigma_1 N_0 L],$$
$$P_{\perp} = P_{0\perp} \exp[-(1+A)\sigma_1 N_0 L]. \tag{12}$$

For pure elastic scattering $A = \frac{1}{2}$ and the factor preceding $\sigma_1 N_0 L$ in (11) and (12) is transformed into unity for $P_{\parallel}$ and into $\frac{3}{2}$ for $P_{\perp}$.[1)]

In concluding this section, we note the following. Formulas (11) and (12) are valid not only in the paramagnetic phase, but also below T_C, if the ferromagnet is broken up into sufficiently small domains and there is no average magnetization. In particular, in this case formulas (12) with $A = \frac{1}{2}$ (the scattering is obviously elastic) are well applicable provided only $\vartheta_m \gg 1/pR_d$, where R_d is the dimension of the domain. The standard methods yield for the cross section for scattering by

[1)]The question of depolarization in the paramagnetic phase on a thin sample was considered earlier by the authors[11] on the basis of macroscopic considerations. It should be noted, however, that the final formula given there does not take into account the difference between $P_{\parallel}$ and $P_{\perp}$, and is therefore accurate only to order of magnitude. In addition, the connection between the depolarization and the scattering was not fully explained in[11]. Our present treatment of this connection refines the considerations given in[11] concerning broad and narrow beams, and in addition, it follows from them that to calculate the observed depolarization it is not necessary to add up the contributions due to the rotation of the polarization vector and to scattering, since the two are equivalent. The depolarization by spin waves was therefore actually calculated in[11] by two different but equivalent methods. The same applies also to Toperverg's work[12].

the domains

$$\frac{d\sigma_d}{d\Omega} = r_0^2\gamma_0^2 S^2 \left(\frac{M(T)}{M(0)}\right)^2 N_0 \int d\mathbf{R} e^{i\mathbf{qR}}\Phi(\mathbf{R}),$$
$$S^2[M(T)/M(0)]^2\Phi(\mathbf{R}) = \langle \mathbf{S}(\mathbf{R})\mathbf{S}(0)\rangle, \qquad (13)$$

where M(T) is the spontaneous magnetization at the temperature T.

The second of these formulas is a definition of $\Phi(\mathbf{R})$—the average shape of the domain. In the case under consideration the effective scattering angles $\vartheta \sim 1/pR_d$ are small and the formulas for the depolarization contain the total scattering cross section. It can be easily calculated by integrating first with respect to the scattering angles, and then with respect to R:

$$\sigma_d = r_0^2\gamma_0^2 N_0 \frac{(4\pi S)^2}{3p^2}\left(\frac{M(T)}{M(0)}\right)^2 R_p = \frac{(2\mu_n B(T))^2}{3v^2 N_0 \hbar^2} R_p, \qquad (14)$$

where $B(T) = 4\pi M(T)$ is the induction in the domain, μ_n is the magnetic moment of the neutron, and R_p is defined by

$$R_p = \frac{p^2}{8\pi^2}\int d\Omega d\mathbf{R}\Phi(\mathbf{R})e^{i(\mathbf{p}-\mathbf{p}')\mathbf{R}} = \frac{p}{2\pi}\int \frac{d\mathbf{R}}{R}\Phi(\mathbf{R})e^{i\mathbf{pR}}\sin pR$$
$$= \frac{1}{2\pi}\int dR \int d\varphi \Phi(R, 0, \varphi). \qquad (15)$$

In this expression the z axis is directed along the beam; in the derivation we took into account the fact that $\Phi(R, 0, \varphi) = \Phi(R, \pi, \varphi + \pi)$, and that $\sin^2 pR$ can be replaced by $1/2$, since the oscillations are fast. Formulas (12) together with (14) and (15) refine the results obtained by Halpern and Holstein[13] who, like ourselves[11], failed to note that $P_\parallel$ and $P_\perp$ are depolarized differently.

3. CROSS SECTION WITHIN THE FRAMEWORK OF SCALING THEORY

At the present time the universally recognized theory of second-order phase transitions is scaling theory (see, for example, the papers of Kadanoff et al.[14], A. A. Migdal[15], Polyakov[16,17], and Halpern and Hohenberg[18]). According to the general formula of dynamic scaling[17]

$$G(\mathbf{q}, \omega) = \frac{1}{T_c\tau^\gamma}\varphi\left(\frac{qa}{\tau^\nu}, \frac{\omega}{T_c\tau^{\nu z}}\right). \qquad (16)$$

Here γ is the critical index of the static susceptibility; it is assumed at present that $\gamma \approx 4/3$, a is a quantity of the order of the lattice constant, and ν is the critical index of the correlation radius, for which a value close to $2/3$ is customarily used. Finally, z is the critical index of the dynamic theory, which according to[18] is equal to $5/2$. The factor $1/T_c$ is separated in (16) from considerations of dimensionality so that φ is a dimensionless function of its arguments.

The momentum transfer in scattering through a fixed angle ϑ is a function of the transferred energy ω, and if $\omega \ll E$, it can be represented in the form[10]

$$q = p[\vartheta^2 + (\omega/2E)^2]^{1/2}. \qquad (17)$$

We shall now show that the dependence of q on ω can be neglected under the conditions of the experiments on depolarization, i.e., that the scattering is quasielastic. It follows from (16) that the characteristic transferred energy is given by

$$\Omega(q) \sim T_c(qa)^\nu f(qa\tau^{-\nu}). \qquad (18)$$

It is shown in[18] that $f(x) \sim x^{-1/2}$ if $x \ll 1$ and $f(x) = \text{const}$ if $x \gg 1$. Since a is of the order of the lattice constant, $qa \ll 1$ in both cases and therefore $\Omega(q) \ll T_c(qa)^2$ (we recall that $z \approx 5/2$). Starting from this inequality, we obtain the following quasi-elasticity condition:

$$\vartheta \lesssim E/T_c(pa)^2. \qquad (19)$$

Usually $E/T_c \gtrsim 10^{-1}$ and $(pa)^2 \lesssim 5$, so that the dependence on ω can be neglected in (17), at any rate for angles smaller than 1°. In the experiment of Drabkin's group[1-4], $\vartheta < 3$–10 min, i.e., the condition (19) is certainly satisfied.

Drabkin, et al.[1] estimated the degree of scattering inelasticity from the polarization of neutrons scattered through an angle on the order of 10 min, and have shown experimentally that in this temperature region, where there is critical depolarization, the scattering is inelastic. Taking this circumstance into account, we obtain on the basis of (2)

$$d\sigma/d\Omega = {}^2/_3 r_0^2\gamma_0^2 T_c G(p\vartheta, 0),$$
$$T_c G(p\vartheta, 0) = \tau^{-\gamma}\varphi(p\vartheta a\tau^{-\nu}), \qquad (20)$$
$$\sigma_1 = \frac{4\pi r_0^2\gamma_0^2}{3\tau^\gamma}\int_0^{\vartheta_m}\vartheta\varphi\left(\frac{p\vartheta a}{\tau^\nu}\right)d\vartheta.$$

If $p\vartheta_m a\tau^{-\nu}$ is small, the integral in the last formula (2) can be easily obtained and we get

$$\sigma_1 = {}^2/_3\pi r_0^2\gamma_0^2\tau^{-\gamma}\varphi(0)\vartheta_m^2, \qquad (21)$$

where the constant $\varphi(0)$ is directly determined, by virtue of (3), from experiments on magnetic susceptibility.

On the other hand, if $p\vartheta_m a\tau^{-\nu} > 1$, then to calculate σ_1 it is necessary to know more accurately the structure of the function φ. In accordance with present-day concepts, $\varphi(x)$ differs very little from the corresponding expression in the Ornstein-Zernike theory: the measure of this difference is the so-called Fisher parameter $\eta = 2 - \gamma/\nu$, and is apparently on the order of 10^{-1}–10^{-2} (see, for example,[19]). We therefore carry out all the estimates on the basis of the Ornstein-Zernike theory, after which we shall indicate briefly the consequences ensuing from the fact that η is finite.

Thus, we assume that $\gamma = 2\nu$ and that

$$T_c G(q, 0) = S(S+1)\left(\frac{a}{r}\right)^2 \frac{1}{\tau^{2\nu}[1 + (pa\vartheta/\tau^\nu)^2]}. \qquad (22)$$

This leads to the following formula for σ_1:

$$\sigma_1 = \frac{2}{3}\pi r_0^2\gamma_0^2 S(S+1)\frac{1}{(pr)^2}\ln\left[1+\left(\frac{pa\vartheta_m}{\tau^\nu}\right)^2\right]. \quad (23)$$

Thus, the cross section σ_1 depends on two parameters, r and a, which generally speaking should be of the order of the lattice constant.

So far, the critical depolarization has been investigated in greatest detail in nickel[1-3]. The critical scattering[2,6] and the magnetic susceptibility[7,8] of nickel were also investigated, so that we can carry out a detailed comparison of theory and experiment. Drabkin et al.[1-3] obtained the following main result concerning the depolarization in the paramagnetic phase: the quantity $P_\perp/P_{0\perp}$ decreases monotonically with decreasing τ, and becomes equal to $(0.6-0.7)P_0$ at a distance $\tau \approx 10^{-4}$ from the Curie point; it can therefore be estimated with sufficient accuracy from formula (11) for a thin sample (formulas (12) are not valid, since σ_1 depends on ϑ_m). In these experiments the sample thickness was 0.5 cm, the neutron wavelength $\lambda \approx 4$ Å, and $\vartheta_m \sim (3-10)$ min. For nickel, $N_0 = 9\times 10^{22}$ cm^{-3} (we define the unit cell in such a way that it contains only one atom, and the cross section necessary for the depolarization to be experimentally observable is $\sigma_1 = 3.5-4.5$ b.

The parameters in (23) can be determined from the data on the magnetic susceptibility and the critical scattering[7,8]. It follows from them that the magnetic susceptibility can be represented in the form

$$\chi(0) \approx 4.2\cdot 10^{-5}\,\tau^{-4/3} \quad (24)$$

and therefore, starting from formulas (3), (22), and (24), and assuming that $S = 1/2$ and $g = 2$, we find that $r/a = 1.4$.

A more complicated and less unambiguous procedure must be used to find the second parameter. Minkiewicz et al.[6] give for critical scattering a value τ^ν/a at $T = T_c + 52°$, which satisfies the inelastic-scattering data in best fashion if these data are described by the well known Van Hove formula, from which it follows that $a = 1.7$ Å. An independent determination of a from the experimental data for $\chi^{-1}(q)$, given in[6], leads to $a = (0.8-0.9)$ Å.

The experiments on critical depolarization[1-3] revealed a neutron-beam attenuation corresponding to an approximate cross section of 2 b for scattering through angles than ϑ_m.[2)] If we use for this cross section the formula

$$\sigma_2 \approx {}^4/_3\pi r_0^2\gamma_0^2 S(S+1)(pr)^{-2}\ln(1/\vartheta_m), \quad (25)$$

then we arrive at a value $a \approx 0.7$, which is in surprisingly good agreement with the value a obtained from the $\chi^{-1}(q)$ curves. The values of the parameters $a = 1.7$ and 0.8 at $\tau = 10^{-4}$ and $\lambda = 4$ Å lead to cross sections σ_1 equal to 0.09 and 0.24 b. Thus, in either case the cross section is too small to explain the observed depolarization, and a cross section 15 times larger is necessary for this purpose in the most favorable case.

Thus, experiments on critical depolarization cannot be explained in natural fashion by starting from data on large-angle scattering. In the region of very small momentum transfers ($q < 3\times 10^{-3}$ Å^{-1}) and in a narrow temperature interval ($\tau \sim 10^{-4}$) the scattering apparently exceeds the usual critical scattering. It should be noted here that the usual critical scattering is well described by the formulas of scaling theory[6,9]. Thus, the parameter a, calculated from the data of[6] at $\tau = 2\times 10^{-2}$, is in splendid agreement with the value of a estimated from the total cross section σ_2 at $\tau = 10^{-4}$. It should also be noted that according to (22) we have $d\sigma/d\Omega \sim \vartheta^{-2}$ at sufficiently large angles (small τ). It is precisely such a dependence of the intensity of the critical-scattering peak which was observed by Drabkin et al.[2] [3)]

It must be emphasized that the excess scattering lies in that region of q and τ where direct experiments on scattering have not yet been carried out (in[6], for example, $q_{min} = 0.02$ Å^{-1}), and therefore the experiments on depolarization do not contradict other experiments.

The nature of the "excess" scattering is still unclear. One of the possible explanations is the formation of nuclei of a new phase, namely regions with homogeneous magnetization (peculiar "quasi-domains"). Such regions can lead, by virtue of (14), to a large depolarization, owing to the large values of R_p, even at very small magnetization. At the same time, in order for them not to contribute to the static susceptibility (which is proportional to the cross section at $\vartheta = 0$), it is necessary by virtue of (13) and (20) to satisfy the inequality

$$\tau^{-\gamma} \gg S^2[M(T)/M(0)]^2 R_d^3. \quad (26)$$

The appearance of such quasi-domains would actually mean that the transition to the ferromagnetic state is a first-order phase transition close to a second-order transition. Such transitions have been intensively discussed in the literature lately (see, for example, the paper of Larkin and Pikin[20]). However, it is still too early to draw definite conclusions.

So far we have used formula (22) to describe the scattering, i.e., we assumed the Fisher parameter η to be equal to zero. It is easy, by using (20), to write down the cross section in the general case when $P\vartheta_m a\tau^{-\nu} \gg 1$, if we use the asymptotic formula $\varphi(x) = \varphi_1 x^{-\gamma/\nu}$ [15,16]. After simple calculations, recognizing that $\eta = 2-\gamma/\nu$, we obtain

$$\sigma_1 = \frac{4}{3}\pi r_0^2\gamma_0^2\frac{1}{(pa)^2}\left\{\frac{\varphi_1}{\eta}[(\vartheta_m pa)^\eta - \tau^{\nu\eta}] + \tau^{\nu\eta}C\right\}$$
$$C = \int_0^1 dx\,x\varphi(x) + \int_1^\infty dx\,x\left[\varphi(x) - \frac{\varphi_1}{x^{\gamma/\nu}}\right] \sim 1. \quad (27)$$

Formulas (23) and (27) should coincide at $pa\nu_m/\tau^\nu \gg 1$ and $\eta \to 0$ from which it follows that $\varphi_1 \approx (a/r)^2 S(S+1)$, and consequently allowance for finite η cannot increase the cross section.

[2)]We are grateful to G. M. Drabkin and A. I. Okorokov for supplying this figure, which was omitted from their brief communication[3].

[3)]Figure 4 of[2] shows the wrong intensity scale for the angles $\vartheta = 6.8'$ and $\vartheta = 10.2'$. In both cases the actual intensity must be increased threefold. We are grateful to G. M. Drabkin and A. I. Okorokov for pointing this out to us.

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53