

# Three-point dynamic correlations of magnetization fluctuations in ferromagnets, and possibilities of studying them by means of polarized neutrons

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(Submitted 24 March 1978)

*Zh. Eksp. Teor. Fiz.* **75**, 764–779 (August 1978)

Three-point dynamic spin correlations in ferromagnets in the critical region above the Curie point are studied and physical effects due to them are discussed. It is shown that, because of the three-point correlations, there exists a strong dependence of the dynamic spin form factor, measured in neutron-scattering experiments, on the magnitude of the atomic spin. Evidently, this explains the difference in the character of the energy dependences of the form factors of Fe [J. W. Lynn, *Phys. Rev.* **B11**, 2624 (1975)] and EuO [O.W. Dietrich, J. Als-Nielsen, and L. Passell, *Phys. Rev.* **B14**, 4923 (1976)]. The three-point correlations in uniaxial ferromagnetics can lead to a strong dependence of the critical absorption on the magnetic field, and it is clear that this has been observed in  $\text{GdCl}_3$  [G. Kamleiter and J. Kötler, *Sol. State. Commun.* **14**, 787 (1974)]. Finally, three-point correlations lead to the appearance of polarization of neutrons when they are scattered in magnets in the paramagnetic phase, and to dependence of the scattering cross section on the polarization of the incident neutrons. These effects are due to interference of the first and second terms of the perturbation-theory series for the magnetic-scattering amplitude and are very small in the far paramagnetic region. However, near the Curie point, because of the growth of critical fluctuations, they increase strongly. Estimates of the expected effect are made, and agree with experimental results for the dependence of the cross section on the polarization for iron near  $T_c$  [A.V. Lazuta, S.V. Maleev, B.P. Toperverg, A.I. Okorokov, A.G. Gukasov, Ya. M. Otchik, and V. V. Runov, *LIYaN (Leningrad Nuclear Physics Institute) Preprint no. 366 (1977)*], and A. I. Okorokov, A. G. Gukasov, Ya. M. Otchik, and V.V. Runov, *Phys. Lett.* **65A**, 60 (1978)]. In the last section, the polarization that arises is calculated in the framework of spin-wave theory for a multi-domain unmagnetized ferromagnet.

PACS numbers: 75.25.+z, 75.30.Gw, 61.12.-q, 75.30.Ds

## 1. INTRODUCTION

As is well known, terms containing odd powers of the magnetic-moment density  $\mathbf{M}(\mathbf{r})$  are absent in the Landau expansion for the free energy of a ferromagnet near the Curie point. At the same time, above  $T_c$  in zero external magnetic field there are no static correlation functions of an odd number of quantities  $\mathbf{M}(\mathbf{r})$ . The static correlation functions are related in a well known way to vertex parts in the diagrammatic perturbation-theory series (see, e.g., the book by Patashinskii and Pokrovskii<sup>[1]</sup>), in which, as is well known, there are also no odd vertices above  $T_c$ . Essentially, the absence of odd static correlations is a consequence of the symmetry of the system with respect to time reversal (we consider this question in more detail below). Therefore, in the dynamical theory odd correlations of the magnetization should certainly be present.

In the present paper we discuss the properties of three-point dynamic correlations of the magnetization, and certain physical effects that arise from their existence. The principal results obtained are the appearance of polarization of the neutrons in critical neutron scattering in ferromagnets above the Curie point and the dependence of the neutron-scattering cross section in this region on the polarization of the incident beam. These effects arise because of the interference of the first and second terms of the perturbation-theory series for the magnetic-scattering amplitude, the interference term being found to be proportional to the integral of

the three-point dynamic correlation function of the magnetizations. This result has been published by us in a short communication.<sup>[2,3]</sup> Despite its small size, the effect under consideration has been observed experimentally by Okorokov, Gukasov, Otchik, and Runov,<sup>[3,4]</sup> and their results agree satisfactorily with the theoretical estimates made on the basis of the dynamic-scaling hypothesis for three-point correlations. It should be noted that, so far as we are aware, only pair correlations of critical fluctuations have been studied experimentally up to now, at least in magnets. Thus, Ref. 4 must be regarded as the first attempt to study more-complicated correlations. Nonzero "bare" odd correlators of the magnetization arise as a result of the non-commutativity of the spin projections of the individual atoms. Therefore, for large values of the atomic spin  $S$  odd correlations should be suppressed. This can lead to a large difference in the form of the formfactors describing the dynamics of pair correlations for ferromagnets with small and large spins. This form factor was measured in Fe ( $S=1$ ) by Lynn<sup>[5]</sup> and in EuO ( $S=7/2$ ) by Dietrich *et al.*<sup>[6]</sup> In the former case, at a fixed momentum and at nonzero energies, Lynn observed broadened peaks, which he interpreted as spin waves above the Curie point, while in the latter case such peaks were not observed. It is not excluded that this difference is, in fact, associated with the suppression of odd correlations.

Finally, we note one more phenomenon that is most probably associated with three-point dynamic correla-

tions. This is the anomalous temperature dependence, observed by Kamleiter and Kötztler,<sup>[7]</sup> of the critical absorption of electromagnetic radiation in the uniaxial ferromagnet GdCl<sub>3</sub>. One of the authors<sup>[8]</sup> once explained this effect by starting from the assumption that an external magnetic field, when it interacts with a three-point dynamic magnetization correlation, violates the symmetry selection rule that leads to the so-called normal behavior in which the temperature dependence of the lifetime of a critical fluctuation is the same as that of the static susceptibility. The correctness of this explanation could be checked by studying the dependence of the critical absorption in GdCl<sub>3</sub> on the magnetic field. Unfortunately, up to now this has not been done.

Concluding the Introduction, we shall discuss in somewhat more detail the content of the subsequent sections of the paper. In Sec. 2 we analyze the general properties of three-point correlations in magnets in the absence of long-range order (the paramagnetic phase) and discuss, in more detail than in the Introduction, the question of the effect of the spin magnitude on the dynamics of the paramagnetic phase of ferromagnets. In Sec. 3 of the paper we discuss the polarization effects that arise from three-point correlations and occur in the scattering of neutrons in magnets in the paramagnetic phase, and, on the basis of the dynamic-scaling hypothesis, make estimates of the magnitude of the expected effects for the case of scattering in ferromagnets above the Curie point.

Finally, in the last section we calculate the polarization arising as a result of three-point correlations in an unmagnetized multi-domain ferromagnet at low temperatures, when the spin-wave theory is valid. The size of the polarization obtained is small, and its experimental observation should be further complicated by the depolarizing influence of the magnetic fields of the domains. At the same time this calculation appears to us to be of fundamental interest, since an unmagnetized multi-domain ferromagnet possesses, in effect, the same magnetic symmetry as the ferromagnet above the Curie point. However, unlike in the paramagnetic phase, where only more or less reliable estimates of the expected effect are possible, in the case of a multi-domain ferromagnet all the calculations can be carried through to the end. As a result, using this example it is possible to verify by means of direct calculations the correctness of the basic ideas, used in the earlier sections of the paper, concerning the properties of three-point correlations.

## 2. PROPERTIES OF THREE-POINT CORRELATIONS

Below, in place of the magnetic-moment density  $M(\mathbf{r})$  it will be convenient for us to use the atomic spins  $S_{\mathbf{R}}$  and their Fourier components

$$S_{\mathbf{r}} = N^{-1/2} \sum_{\mathbf{R}} e^{i\mathbf{r}\cdot\mathbf{R}} S_{\mathbf{R}}.$$

Following Vaks, Larkin, and Pikin,<sup>[9]</sup> we define the three-spin Matsubara Green function by the equality

$$F_{\alpha,\alpha,\alpha}(\mathbf{R}_1, i\omega_1, \mathbf{R}_2, i\omega_2, \mathbf{R}_3, i\omega_3) = \frac{1}{T} \delta_{\omega_1+\omega_2+\omega_3} \times F_{\alpha,\alpha,\alpha}(\mathbf{R}_1, \omega_1, \mathbf{R}_2, \omega_2, \mathbf{R}_3, \omega_3) = \int_0^{1/T} \prod_{j=1}^3 (d\tau_j \exp(i\omega_j \tau_j)) \langle T_{\tau} S_{\mathbf{R}_1}^{\alpha}(\tau_1) S_{\mathbf{R}_2}^{\alpha}(\tau_2) S_{\mathbf{R}_3}^{\alpha}(\tau_3) \rangle. \quad (1)$$

Clearly, the function  $F$  is symmetric under permutations of "particles," i.e., of pairs  $(\alpha_n, \mathbf{R}_n, i\omega_n)$  and  $(\alpha_m, \mathbf{R}_m, i\omega_m)$ . Using the usual expansion over intermediate states (see the book by Abrikosov, Gor'kov, and Dzyaloshinskii,<sup>[10]</sup> and the paper by one of the authors<sup>[11]</sup>) it is not difficult to obtain the following spectral representation for  $F$ :

$$F_{\alpha,\alpha,\alpha}(\mathbf{R}_1, \omega_1, \mathbf{R}_2, \omega_2, \mathbf{R}_3, \omega_3) = \sum_{abc} \frac{\exp(-E_a/T)}{Z} \left\{ \frac{S_{ab}^{(1)} S_{bc}^{(2)} S_{ca}^{(3)}}{(\omega_{ba}-\omega_1)(\omega_{ca}+\omega_3)} + \frac{S_{ab}^{(3)} S_{bc}^{(2)} S_{ca}^{(1)}}{(\omega_{ba}-\omega_2)(\omega_{ca}+\omega_1)} + \frac{S_{ab}^{(2)} S_{bc}^{(3)} S_{ca}^{(1)}}{(\omega_{ba}-\omega_2)(\omega_{ca}+\omega_1)} + \frac{S_{ab}^{(1)} S_{bc}^{(3)} S_{ca}^{(2)}}{(\omega_{ba}-\omega_1)(\omega_{ca}+\omega_2)} + \frac{S_{ab}^{(2)} S_{bc}^{(1)} S_{ca}^{(3)}}{(\omega_{ba}-\omega_2)(\omega_{ca}+\omega_3)} + \frac{S_{ab}^{(3)} S_{bc}^{(1)} S_{ca}^{(2)}}{(\omega_{ba}-\omega_2)(\omega_{ca}+\omega_2)} \right\}. \quad (2)$$

Here  $S^{(m)} = S^{\alpha m}(\mathbf{R})_m$ ,  $\omega_{ba} = E_b - E_a$ , and all the imaginary frequencies  $i\omega_m$  are replaced by  $\omega_m$ . As can be seen from this expansion, the function  $F$  is an analytic function of the variables  $\omega_m$ , in each of which it has a cut along the real axis. All the observable physical effects associated with dynamic three-spin correlations can be described by means of appropriately chosen combinations of the different branches of the function  $F$  at real frequencies. Here, however, it must be remembered that, by virtue of the invariance of the system under translations in time, the real frequencies should be related by a conservation law ( $\omega_1 + \omega_2 + \omega_3 = 0$ ), so that the function  $F$  is, in fact, not a sum of three analytic functions depending on the pairs of arguments  $(\omega_1, \omega_2)$ ,  $(\omega_2, \omega_3)$ , and  $(\omega_1, \omega_3)$ , but a sum of two functions of two independent pairs of arguments, e.g.,  $(\omega_1, \omega_2)$  and  $(\omega_1, \omega_3)$ . The decomposition of the function  $F$  into two such functions  $F^{(1)}(\omega_1, \omega_3)$  and  $F^{(2)}(\omega_1, \omega_2)$  is easily performed by expanding the last two terms in formula (2) in partial fractions and taking the frequency conservation law into account; as a result, for  $F^{(1)}$  we have

$$F_{\alpha,\alpha,\alpha}^{(1)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \omega_1, \omega_3) = \sum_{abc} \frac{\exp(-E_a/T)}{Z} \times \left[ \frac{S_{ab}^{(1)} S_{bc}^{(2)} S_{ca}^{(3)} (1 - \exp(-\omega_{ca}/T))}{(\omega_{ba}-\omega_1)(\omega_{ca}+\omega_3)} + \frac{S_{ab}^{(3)} S_{bc}^{(2)} S_{ca}^{(1)} (1 - \exp(-\omega_{ba}/T))}{(\omega_{ba}-\omega_3)(\omega_{ca}+\omega_1)} \right], \quad (3)$$

while  $F^{(2)}$  is obtained from this expression by replacing  $\omega_3$  by  $\omega_2$  and interchanging  $S^{(2)}$  and  $S^{(3)}$ .

In the next section of this paper it will be shown that the polarization effects of interest to us can be expressed in terms of the Fourier transform of the ternary correlator of the spins:

$$\int dt_1 dt_2 dt_3 \exp(i\omega_1 t_1 + i\omega_2 t_2 + i\omega_3 t_3) \langle S^{(1)}(t_1) S^{(2)}(t_2) S^{(3)}(t_3) \rangle = 2\pi \delta(\omega_1 + \omega_2 + \omega_3) F_{\alpha,\alpha,\alpha}^{(0)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3; \omega_1, \omega_2, \omega_3), \quad (4)$$

where

$$F_{\alpha,\alpha,\alpha}^{(0)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3; \omega_1, \omega_2, \omega_3) = 4\pi^2 \sum_{abc} \frac{\exp(-E_a/T)}{Z} S_{ab}^{(1)} S_{bc}^{(2)} S_{ca}^{(3)} \delta(\omega_{ba}-\omega_1) \delta(\omega_{ca}+\omega_3).$$

It is clear that the function  $F^{(0)}$  differs by only a factor from the discontinuity<sup>1)</sup> of the first term in the expression (3) for  $F^{(1)}$  in the variables  $\omega_1$  and  $\omega_3$ . It is not difficult to express the function  $F^{(0)}$  in terms of the quantities  $\Delta_1 \Delta_3 F^{(1)}$  and  $\Delta_1 \Delta_2 F^{(2)}$ . In fact, it follows from (3) and the analogous expression for  $F^{(2)}$  that  $\Delta_1 \Delta_3 F^{(1)}$  and  $\Delta_1 \Delta_2 F^{(2)}$  are sums of two terms, each of which contains a sum, over intermediate states, of three  $S^{(m)}$  and two  $\delta$ -functions. Taking into account the energy conservation law ( $\omega_1 + \omega_2 + \omega_3 = 0$ ), it is easy to transform the sums over intermediate states appearing in  $\Delta_1 \Delta_2 F^{(2)}$  in such a way that they will be proportional to the sums appearing in  $\Delta_1 \Delta_3 F^{(1)}$ . As a result, a system of two linear equations is obtained; solving this, we find

$$F^{(0)} = 4n(-\omega_1) [n(\omega_1) \Delta_1 \Delta_3 F^{(1)} - n(-\omega_1) \Delta_1 \Delta_2 F^{(2)}] - 4T^2 \left[ \frac{\Delta_1 \Delta_3 F^{(1)}}{\omega_1 \omega_3} + \frac{\Delta_1 \Delta_2 F^{(2)}}{\omega_1 \omega_2} \right] = -4T^2 \left[ \frac{\Delta_1 \Delta_3}{\omega_1 \omega_3} + \frac{\Delta_1 \Delta_2}{\omega_1 \omega_2} \right] F. \quad (5)$$

Here  $n(\omega) = [\exp(\omega/T) - 1]^{-1}$ , and the approximate equality on the right is valid for  $|\omega| \ll T$ . This formula is analogous to the well known expression for the Fourier transform of the correlator  $\langle S_{R_1}^a(t) S_{R_2}^a(0) \rangle$  in terms of the imaginary part of the susceptibility (see e.g., Refs. 5 and 6).

We now consider the question of the properties of the function  $F$  that follow from the symmetry of the system with respect to time reversal. As is well known, when  $t$  is replaced by  $-t$  the spectrum of the system is not altered but the matrix elements of the spins before and after such a transformation are connected by the relation  $S_{ab}^a = -S_{\bar{a}\bar{b}}^a$ , where  $\bar{a}$  and  $\bar{b}$  are the states  $a$  and  $b$  reversed in time. Using these properties and also the hermiticity of the spin operators, it is not difficult to convince oneself that the function  $F^{(0)}$  is purely imaginary and to obtain from (2) the equality

$$F_{\alpha\beta\gamma\delta}^{(0)}(R_1, R_2, R_3; \omega_1, \omega_2, \omega_3) = -F_{\alpha\beta\gamma\delta}^{(0)}(R_1, R_2, R_3; -\omega_1, -\omega_2, -\omega_3). \quad (6)$$

It follows immediately from this formula that at zero frequencies, i.e., in the static limit, the function  $F$  is identically equal to zero.

To conclude this section we shall discuss the question of the effect of the spin magnitude on the critical dynamics of ferromagnets above  $T_c$ . We shall assume that the atomic spin  $S$  is a large number, and elucidate the parametric dependence of the quantities of interest on  $S$ . First we shall consider the static theory. The many-particle Green functions of this theory are defined by the equality

$$\frac{1}{T} G^{(2n)} = \int_0^{1/T} d\tau_1 \dots d\tau_{2n} \left\langle T_\tau S_1 \dots S_{2n} \exp \left[ - \int_0^{1/T} d\tau \mathcal{V}(\tau) \right] \right\rangle, \quad (7)$$

$$\mathcal{V} = - \frac{1}{2} \sum_{RR'} V_{RR'}^{\alpha\beta} S_{R\alpha}^{\alpha} S_{R'\beta}^{\beta},$$

where  $S_m = S_{\alpha m}(R_m, \tau_m)$ , and  $V_{RR'}^{\alpha\beta}$  is the interaction energy of the atomic spins, which includes not only the exchange energy but also other forms of interaction, e.g., dipole-dipole interaction. From this formula it follows immediately that the spin dependence of the static  $2n$ -particle Green function has the form

$$G^{(2n)} = \frac{S^{2n}}{T^{2n-1}} g_{2n} \left( \frac{S^2 V}{T} \right), \quad V = \sum_{\mathbf{R}} V_{\mathbf{R}}^{\alpha\alpha} \quad (8)$$

and, in particular, for the ordinary Green function we obtain the formula

$$G = G^{(2)} = \frac{S^2}{T} g_2 \left( \frac{S^2 V}{T} \right),$$

while for the Curie temperature we have the estimate  $T_c \sim S^2 V / 9$ .

Going over to the dynamical theory in accordance with the dynamic-scaling hypothesis of Halperin and Hohenberg,<sup>[12]</sup> as generalized by Polyakov<sup>[13]</sup> to many-particle Green functions, we have

$$G^{(2n)} = \frac{S^{2n}}{T^{2n-1}} g_{2n} \left( \frac{S^2 V}{T}, \frac{\omega_i}{\Omega(S, \tau)} \right), \quad (9)$$

where  $\Omega(S, \tau)$  is the characteristic energy in the dynamic scaling, whose spin dependence we shall not specify, and  $\tau = (T - T_c) T_c^{-1}$ .

We turn now to the odd dynamic Green functions. Such functions are nonzero only because of the noncommutativity of the atomic spins. For the single-cell bare three-particle Green function (single-cell block) of discrete frequencies an expression has been obtained<sup>[9]</sup> which, in zero magnetic field, is proportional not to  $S^3$  but to  $S^2$ . In an analogous way one can convince oneself that the bare Green function of order  $2n+1$  is proportional to  $S^{2n}$ .<sup>2)</sup> Since the exact Green function is obtained by complicating the single-cell function by attaching other single-cell blocks to it by means of interaction lines  $V_{RR'}$  (see Ref. 9), the expression for the odd Green functions in the leading approximation in  $S$  should have the form

$$F^{(2n+1)} = \frac{S^{2n}}{T^{2n}} f_{2n+1} \left( \frac{S^2 V}{T}, \frac{\omega_i}{\Omega(S, \tau)} \right), \quad (10)$$

where, by virtue of the dynamic-scaling hypothesis,  $\Omega(S, \tau)$  is the same quantity as in (9).

We shall examine now how the existence of the odd correlations affects the properties of the Green function  $G(\mathbf{q}, \omega)$ . In Ref. 9  $G(\mathbf{q}, \omega)$  was expressed in terms of the irreducible part  $\Sigma(G^{-1} = \Sigma^{-1} - V_{\mathbf{q}})$ , which can be represented in the form of the diagrammatic series

$$\Sigma = \text{---} \bullet \text{---} + \text{---} \bullet \text{---} \text{---} \bullet \text{---} + \text{---} \bullet \text{---} \text{---} \bullet \text{---} \text{---} \bullet \text{---} + \dots \quad (11)$$

Here the third term arises from the existence of odd vertices. In the static theory these vertices are equal to zero, and therefore, this term, represented in the form of a sum over discrete frequencies,<sup>[9,10]</sup> is not singular near the Curie point, i.e., does not contain terms in which all Green functions appear with zero discrete frequencies. The same is true for all the other diagrams with odd vertices. In the static limit  $\omega_n = 0$  all such diagrams lead to a certain additional renormalization of the Curie temperatures, not taken into account in the usual static theory.<sup>[1]</sup> After this renormalization and analytic continuation in the frequency, the contribution to  $\Sigma$  and  $G$  from diagrams with odd vertices should vanish for  $\omega = 0$ . Furthermore, as is well

known, for ImG there exists the representation

$$\text{Im } G_{ab}(\omega) = \pi(1 - e^{-\omega/T}) \sum_{ab} Z^{-1} \exp(-E_a/T) S_{ab}^x S_{ba}^y \delta(\omega_{ba} - \omega). \quad (12)$$

Because of the odd correlations, in the sum over  $a$  and  $b$  there are terms in which the  $t$ -parities of the states  $a$  and  $b$  are different. This means that the product of the phases of the wavefunctions of the states  $a$  and  $b$  differs in sign from that for the states  $\bar{a}$  and  $\bar{b}$ . Then, in order that the equality  $S_{ab}^x = -S_{\bar{b}\bar{a}}^x$  hold, a factor canceling this difference in sign is needed. From what has been said above it is clear that this factor should be the difference  $\omega_{ba}$ . We must suppose, therefore, that the contribution to ImG due to odd correlations is proportional to  $\omega^3$  for small  $\omega$ . As a result, we arrive at the following parametric representation for the Green function  $G$  in the critical region above  $T_c$ :

$$G(\mathbf{q}, \omega) = \frac{1}{\pi} \int \frac{d\omega' g_0(\mathbf{q}, \omega')}{\omega' - \omega} + \frac{\omega}{\pi} \int \frac{d\omega' g_1(\mathbf{q}, \omega')}{\omega'(\omega' - \omega)}, \quad (13)$$

where

$$g_0(\mathbf{q}, \omega) = \frac{S^2}{T} \frac{\omega}{\Omega} \rho_0 \left( \frac{S^2 V}{T}, \mathbf{q}, \frac{\omega}{\Omega} \right),$$

$$g_1(\mathbf{q}, \omega) = \frac{S^2}{T} \left( \frac{\omega}{\Omega} \right)^2 \frac{1}{S^2} \rho_1 \left( \frac{S^2 V}{T}, \mathbf{q}, \frac{\omega}{\Omega} \right).$$

Here,  $\rho_0$  and  $\rho_1$  are even functions of  $\omega$ , finite at  $\omega = 0$ . In analyzing the critical dynamics above  $T_c$ , for  $G$  it has been found to be convenient to use the representation<sup>[8, 14]</sup>

$$G(\mathbf{q}, \omega) = G(\mathbf{q}, 0) \Gamma(\mathbf{q}, \omega) [-i\omega + \Gamma(\mathbf{q}, \omega)]^{-1},$$

$$\Gamma(\mathbf{q}, \omega) = [i\omega G(\mathbf{q}, 0)]^{-1} [\Phi(\mathbf{q}, \omega) - \Phi(\mathbf{q}, 0)], \quad (14)$$

where  $\Phi$  is the retarded Green function of the operators  $dS_a/dt$ . Using reasoning analogous to that given above for  $\Gamma$ , we obtain the representation

$$\Gamma(\mathbf{q}, \omega) = \frac{1}{\pi i} \int \frac{d\omega' [\gamma_0(\mathbf{q}, \omega') + \gamma_1(\mathbf{q}, \omega')]}{\omega' - \omega} \quad (15)$$

where

$$\gamma_0(\mathbf{q}, \omega) = \Omega \varphi_0 \left( \frac{S^2 V}{T}, \mathbf{q}, \frac{\omega}{\Omega} \right), \quad \gamma_1(\mathbf{q}, \omega) = \Omega \left( \frac{\omega}{\Omega} \right)^2 \frac{1}{S^2} \varphi_1 \left( \frac{S^2 V}{T}, \mathbf{q}, \frac{\omega}{\Omega} \right)$$

are even functions of  $\omega$ , finite at  $\omega = 0$ . In neutron-scattering experiments one measures the dynamic form factor

$$N(\mathbf{q}, \omega) = \frac{T}{\omega} \text{Im } G(\mathbf{q}, \omega). \quad (16)$$

It is clear that  $N = N_0 + N_1$ , where  $N_{0,1} \propto g_{0,1}$ . As functions of  $\omega$  the quantities  $N_0$  and  $N_1$  behave differently.  $N_0(\mathbf{q}, 0) \neq 0$ , and  $N_0$  decreases with increase of  $\omega$  for  $\omega \gg \Omega$  (see Ref. 14), whereas  $N_1(\mathbf{q}, 0) = 0$ , and  $N_1$  has a maximum somewhere near  $\omega \sim \Omega$  and only then begins to decrease. It is obvious, then, that the character of the behavior of the form factor  $N$  depends on the relative contributions of  $N_0$  and  $N_1$ . If the contribution of  $N_1$  is large the function  $N$  should have a maximum at  $\omega \sim \Omega$ , whereas for small  $N_1$  there will be no such maximum. It is obvious that the larger is  $S$ , the smaller is the contribution of  $N_1$  to  $N$ . This is in qualitative agreement with the results of Refs. 5 and 6: in Fe, where  $S = 1$ , a maximum

of  $N$  was observed at  $\omega \neq 0$ , while in EuO, where  $S = 7/2$ , such a maximum was not observed.

### 3. POLARIZATION EFFECTS IN CRITICAL SCATTERING ABOVE $T_c$

First of all we shall explain why an initially unpolarized beam of neutrons becomes polarized on scattering in a ferromagnet above  $T_c$ . It is well known that the Born scattering amplitude has the form

$$f_{\mathbf{q}} = -r_0 \gamma \int F(\mathbf{q}) \exp(i\mathbf{q}\mathbf{R}_j) (\delta_{\alpha\beta} - \epsilon_{\alpha\beta\gamma}) S_j^\alpha \sigma^\beta, \quad (17)$$

where  $\mathbf{q} = \mathbf{p} - \mathbf{p}'$ ,  $\mathbf{p}$  and  $\mathbf{p}'$  are the initial and final momenta of the neutron,  $\mathbf{e} = \mathbf{q}\mathbf{q}^{-1}$ ,  $r_0$  is the classical electron radius,  $\gamma$  is the gyromagnetic ratio of the neutron,  $1/2\sigma$  is its spin,  $\mathbf{S}_j$  is the spin of the atom at the point  $\mathbf{R}_j$ , and  $F(\mathbf{q})$  is the magnetic form factor of the atom. The polarization is a pseudovector. Since the Born amplitude (17) depends on the momentum transfer  $\mathbf{q}$ , in the Born approximation there is no pseudo-vector along which the polarization vector of the scattered neutrons can point. As shown below, the expression for the polarization  $\mathbf{P}$ , arising from the interference of the first and second orders of perturbation theory, contains the pseudo-tensor  $\langle S_1^\alpha S_2^\beta S_3^\gamma \rangle$ . If we confine ourselves to taking only exchange forces into account, this is proportional to  $\epsilon_{\alpha\beta\gamma}$ , and, since in second order the amplitude depends on  $\mathbf{p}$  and  $\mathbf{p}'$  separately, the pseudo-vector  $\mathbf{p} \times \mathbf{p}'$  is formed, along which the polarization points. Since  $\mathbf{p} \times \mathbf{p}'$  is the only pseudo-vector in the system, the polarization will also point along it when dipolar forces are taken into account, although in this case  $\langle S_1^\alpha S_2^\beta S_3^\gamma \rangle$  has a more complicated tensor structure. In the scattering of slow neutrons with momentum of the order of the inverse lattice constant, far from the critical region the polarization is small. It is proportional to the ratio of the energy of the magnetic interaction of the neutron and the atomic spin at a distance of the order of the inverse lattice constant to the interaction energy in the spin system, which can be written in the form  $r_0 E_0 / aV$ , where  $a$  is a quantity of the order of the lattice constant and  $E_0 = \hbar^2 / 2m a^2$  ( $E_0 \approx 24$  K for  $a = 1$  Å). In the critical region the correlations between the spins grow and the polarization is enhanced (up to  $3 \times 10^{-4}$  in the experiments of Refs. 3 and 4).

Since the existence of three-spin correlations is a dynamical phenomenon, the polarization depends strongly on the relative magnitudes of the lifetime of a fluctuation and the time of flight of the neutron across the fluctuation, and also on the actual dynamical behavior of the fluctuations. As has been shown,<sup>[15]</sup> the dynamical properties of a ferromagnet in the range of temperatures in which  $4\pi\chi \ll 1$  (the exchange region) differ from the properties in the region  $4\pi\chi \gg 1$  (the dipolar region). Therefore, the dependences of the polarization on  $\tau = (T - T_c)T_c^{-1}$  and the scattering angle  $\vartheta$  in the exchange region differ from the dependences in the dipolar region. This makes it possible to make an additional check on the predictions of the theory concerning the influence of the dipolar forces on the critical dy-

namics of ferromagnets. In Refs. 3 and 4 it was not the polarization that was measured, but the difference of the cross sections for scattering of neutrons with initial polarizations parallel and antiparallel to  $\mathbf{p} \times \mathbf{p}'$ . The existence of this difference and the appearance of polarization are due to the same causes, and the estimates of the dependence of the polarization on  $\vartheta$  and  $\tau$  are also valid for this difference. Below we shall obtain the formula for the polarization of the neutrons and indicate the changes that lead to expressions for the difference of the cross sections.

The polarization vector of the scattered neutrons is determined by the formula

$$\mathbf{P} \frac{d^2\sigma}{d\Omega dE'} = \frac{p'}{p} \left(\frac{m}{2\pi}\right)^2 \frac{1}{T_0 N} \langle (S^+ - 1)_{p',\sigma} (S^-)_{p,\sigma'} \rangle. \quad (18)$$

Here  $S_{p,\sigma}$  is a matrix element of the S-matrix, the averaging is performed over the states of the sample and over the spin states of the incident neutron beam,  $T_0$  is an infinitely long time interval, to which the expression in angular brackets is proportional, and  $N$  is the total number of magnetic atoms (we normalize the cross section to that for one magnetic atom, and take  $\hbar = 1$ ).

The amplitude  $(S^-)_{p,\sigma}$  is expressed in the usual way in the form of a series in powers of the magnetic interaction  $U_q$ :

$$(S^-)_{p,\sigma} = -i \int_{-\infty}^{\infty} dt U_q(t) e^{-i\omega t} + (-i)^2 \int \frac{d\mathbf{p}_1}{(2\pi)^3} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 U_{-q_1}(t_1) U_{-q_2}(t_2) \exp(i\omega_1 t_1 + i\omega_2 t_2) + \dots \quad (19)$$

$$U_q = -\frac{2\pi}{m} r_0 \gamma N^h F(\mathbf{q}) (S_q^+ \sigma), \quad (20)$$

where

$$\begin{aligned} \mathbf{q}_1 &= \mathbf{p}' - \mathbf{p}, & \mathbf{q}_2 &= \mathbf{p}_1 - \mathbf{p}, & \mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q} &= 0, \\ \omega_1 &= E_{p_1} - E_p, & \omega_2 &= E_{p_2} - E_p, & \omega_1 + \omega_2 + \omega &= 0, \\ E_{p_1} &= p_1^2/2m, & S_q^{\pm\alpha} &= (\delta_{\alpha\beta} - e_\alpha e_\beta) S_q^\beta, & e_i &= q_i q_i^{-1}. \end{aligned}$$

Since polarization does not arise in first order in  $U_q$ , we take into account both the terms written out in (19), and, in the expression (18) for the polarization  $\mathbf{P}$ , consider the interference term of order  $U_q^3$ . Making use of (20), for a crystal with a center of inversion we obtain from (18)

$$\begin{aligned} P_\mu \frac{d^2\sigma}{d\Omega dE'} &= \frac{p'}{p} (r_0 \gamma)^2 \frac{2\pi i N^h}{m T_0} \int \frac{d\mathbf{p}_1}{(2\pi)^3} F(\mathbf{q}) F(\mathbf{q}_1) F(\mathbf{q}_2) \\ &\times \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \{ \langle S_q^{\pm\alpha}(t) S_{q_1}^{\pm\beta}(t_1) S_{q_2}^{\pm\gamma}(t_2) \rangle \exp(i\omega t + i\omega_1 t_1 + i\omega_2 t_2) \\ &- \langle S_{q_1}^{\pm\alpha}(t_1) S_{q_2}^{\pm\beta}(t_2) S_q^{\pm\gamma}(t) \rangle \exp(-i\omega t - i\omega_1 t_1 - i\omega_2 t_2) \} \text{Sp}(\sigma_\alpha \sigma_\beta \sigma_\gamma). \end{aligned} \quad (21)$$

Using the symmetry under time reversal, it is not difficult to transform the expression (21) to the following final form:

$$\begin{aligned} P_\mu \frac{d^2\sigma}{d\Omega dE'} &= \frac{p'}{p} (r_0 \gamma)^2 \frac{2\pi i N^h}{m T_0} (\delta_{\alpha\mu} \delta_{\beta 1} + \delta_{\alpha 1} \delta_{\mu\beta} - \delta_{\alpha\beta} \delta_{\mu 1}) \\ &\times \int \frac{d\mathbf{p}_1}{(2\pi)^3} F(\mathbf{q}) F(\mathbf{q}_1) F(\mathbf{q}_2) \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \\ &\times \exp(i\omega t + i\omega_1 t_1 + i\omega_2 t_2) \langle S_q^{\pm\alpha}(t) S_{q_1}^{\pm\beta}(t_1) S_{q_2}^{\pm\gamma}(t_2) \rangle. \end{aligned} \quad (22)$$

We also quote the formula for the cross section for

scattering of polarized neutrons:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{d^2\sigma_0}{d\Omega dE'} (1 + \mathbf{P}_0 \bar{\mathbf{P}}), \quad (23)$$

where  $\mathbf{P}_0$  is the initial polarization and the expression for  $\bar{\mathbf{P}} d^2\sigma_0/d\Omega dE'$  differs from (22) in the signs of the last two pairs of  $\delta$ -symbols.

The correlator of three  $S^{\pm}$  appearing in (22) is expressed, in complete analogy with (5), in terms of the three-spin Green function  $F^{\pm}$ . In order that the dynamic-scaling theory can be used in estimates of the magnitude of the polarization, it is necessary to express  $F^{\pm}$  in terms of the spin Green functions  $G$  and the vertex part  $\Gamma$ . Here, unlike in the usual relationship<sup>[10]</sup> between  $F$  and the functions  $G$  and  $\Gamma$ , in the expression for  $F^{\pm}$  only the transverse (to the external momenta) parts of the Green functions appear:

$$\begin{aligned} F_{\alpha\beta\gamma}^{\pm}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3; \omega_1, \omega_2, \omega_3) &= G_{\mathbf{q}_1}^{\pm\alpha\alpha}(\omega_1) G_{\mathbf{q}_2}^{\pm\beta\beta}(\omega_2) G_{\mathbf{q}_3}^{\pm\gamma\gamma}(\omega_3) \Gamma_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3}^{\pm\alpha\beta\gamma}(\omega_1, \omega_2, \omega_3), \\ G_{\alpha\beta}(\mathbf{q}) &= G_{\mathbf{q}}^{\pm\alpha\beta} + G_{\mathbf{q}}^{\pm\beta\alpha} = (\delta^{\alpha\beta} - e^\alpha e^\beta) G_{\mathbf{q}}^{\pm} + e^\alpha e^\beta G_{\mathbf{q}}^{\pm}. \end{aligned} \quad (24)$$

Before proceeding to the transformation of the expression (22), we shall discuss estimates for  $G^{\pm}$  and  $\Gamma^{\pm}$  in the exchange and dipolar regions, and also the tensor properties of  $\Gamma^{\pm}$ . In the so-called exchange region  $4\pi\chi \ll 1$  (see Ref. 15), where the influence of the dipole forces can be neglected,  $G_{\mathbf{q}}^{\pm} \approx G_{\mathbf{q}}^{\pm}$ . In the dipolar region  $4\pi\chi \gg 1$  the isotropy is violated, and at small  $\mathbf{q}$  we have  $G_{\mathbf{q}}^{\pm} \gg G_{\mathbf{q}}^{\pm}$ . But if the momentum  $\mathbf{q}$  is greater than the characteristic dipolar momentum  $q_0 = a^{-1} \omega_0^{1/2} T_c^{-1/2}$  ( $\omega_0 = 4\pi(g\mu_0)^2 v_0^{-1}$ , where  $g$  is the atomic  $g$ -factor,  $\mu_0$  is the Bohr magneton, and  $v_0$  is the volume of the unit cell), then  $G_{\mathbf{q}}^{\pm} \approx G_{\mathbf{q}}^{\pm}$ .

It is obvious that in the exchange region  $\Gamma^{\pm\alpha\beta\gamma}$  is an isotropic antisymmetric pseudo-tensor, and, therefore,  $\Gamma^{\pm\alpha\beta\gamma} \propto \epsilon^{\alpha\beta\gamma}$ . In the dipolar region, besides  $\epsilon^{\alpha\beta\gamma}$  the expression for the vertex also contains terms proportional to pseudo-tensors constructed from contractions of  $\epsilon^{\alpha\beta\gamma}$  with components of the vectors  $\mathbf{q}_1$ ,  $\mathbf{q}_2$ , and  $\mathbf{q}_3$ .

We shall give the expression for the polarization that follows from (22), (24), and (5), confining ourselves in the expression for  $\Gamma^{\pm\alpha\beta\gamma}$  to the term proportional to  $\epsilon^{\alpha\beta\gamma}$ , i.e., putting  $\Gamma^{\pm\alpha\beta\gamma} = 1/6\epsilon^{\alpha\beta\gamma}\Gamma$ :

$$\begin{aligned} P_\mu \frac{d^2\sigma}{d\Omega dE'} &= -i \frac{4\pi}{3m} N^h \frac{p'}{p} \int \frac{d\mathbf{q}_1}{(2\pi)^3} F(\mathbf{q}) F(\mathbf{q}_1) F(\mathbf{q}_2) \frac{T^2 \Delta}{\omega_1} \left( \frac{\Delta_1}{\omega_1} + \frac{\Delta_2}{\omega_2} \right) \\ &\times \langle G_{\mathbf{q}_1}^{\pm}(\omega_1) G_{\mathbf{q}_2}^{\pm}(\omega_2) \Gamma_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}}^{\pm\alpha\beta\gamma}(\omega_1, \omega_2, \omega) \Phi(\mathbf{e}, \mathbf{e}_1, \mathbf{e}_2) \rangle, \end{aligned} \quad (25)$$

where

$$\begin{aligned} \Phi(\mathbf{e}, \mathbf{e}_1, \mathbf{e}_2) &= [\mathbf{e}_1 \times \mathbf{e}_2] (\mathbf{e}, \mathbf{e}_2) - [\mathbf{e} \times \mathbf{e}_1] (\mathbf{e}, \mathbf{e}_2) - [\mathbf{e}, \mathbf{e}_2] (\mathbf{e} \times \mathbf{e}_2) \\ &= \left( \frac{q^2 - q_2^2}{(q + q_2)^2} - 1 \right) [\mathbf{e} \times \mathbf{e}_2] (\mathbf{e}, \mathbf{e}_2). \end{aligned}$$

This expression for  $\mathbf{P}$  is exact in the exchange region and suitable for estimates in the dipolar region.

Above the Curie point the exact functional forms of  $G$  and  $\Gamma$  are not known; therefore, we can obtain only rather crude estimates for  $\mathbf{P}$ . Here we give the expressions that follow from the dynamic-scaling theory for the functions  $G$  and  $\Gamma$  appearing in (25), and the final results for the polarization. The procedure for evalu-

ating the integral in (25) has been transferred to the Appendix.

We shall consider the exchange region ( $4\pi\chi < 1$ , or  $\kappa > q_0$ ). For  $G$  we have the expression<sup>[12]</sup>

$$G_q(\omega) = G_q(0)g(q/\chi, \omega/\Omega_q), \quad g(q/\chi, 0) = 1, \quad (26)$$

where  $G_q(0)$  is the static Green function, for which we shall use the Ornstein-Zernike formula  $G_q(0) = Z[(q^2 + \chi^2)a^2]^{-1}$ , where  $Z \sim T^{-1}$  and  $\chi = \tau^\nu a^{-1}$  ( $\nu = 2/3$ );  $\chi$  is the inverse correlation length, and the characteristic energy  $\Omega_q = T_c(qa)^{5/2}$  for  $q > \chi$  and  $\Omega_q = \Omega_\chi = T_c(\chi a)^{5/2}$  for  $q < \chi$ . In the dynamic-scaling theory the vertex part  $\Gamma$  has the following functional form, which is easily obtained from the unitarity conditions<sup>[13,15]</sup>:

$$\Gamma_{q_1, q_2}(\omega) = T_c(\chi a)^{3/2} \gamma(q/\chi, \omega/\Omega_\chi); \quad (27)$$

for  $q \leq \chi$  and  $\omega \sim \Omega_\chi$ , the function  $\gamma \sim 1$ . If all momenta are large compared with  $\chi$ , the dependence on  $\chi$  in  $\Gamma$  drops out. In addition, in the case when one of the momenta, e.g.,  $q$ , is small compared with  $q_1$  and  $q_2$ , in accordance with the principle of coalescence of correlations<sup>[16]</sup> we have

$$\Gamma_{q_1, q_2}(\omega) \sim \begin{cases} q_1, q_2 \gg q \gg \chi, \\ q_1, q_2 \gg \chi \gg q. \end{cases} \quad (28)$$

Before writing out the final formulas for the polarization in the case of small-angle scattering, we shall discuss the character of the behavior of  $P(q, \chi)$  as a function of  $q$  for fixed  $\chi$ . Using the expressions given for  $G$  and  $\Gamma$ , it is not difficult to convince oneself that as  $q$  decreases ( $q \gtrsim \chi$ ) the polarization increases so long as  $\omega \leq \Omega_q$ . For  $\omega \gg \Omega_q$  the functions appearing in (25) begin to fall off rapidly<sup>[14]</sup>; therefore, the maximum value of the polarization can be achieved in the region  $\omega \leq \Omega_q$ . The energy transfer in scattering through a fixed angle  $\vartheta$  is a function of the momentum transfer, and, if  $\omega(p^2/2m)^{-1} \ll 1$ , it can be represented in the form  $\omega = pm^{-1}(q^2 - (p\vartheta)^2)^{1/2}$ . Here the inequality  $\omega \leq \Omega_q$  means that  $q \gtrsim q_c = (2E_0 pa T_c^{-1})^{2/3} a^{-1}$ ,  $E_0 = (2ma^2)^{-1}$ . This condition is the opposite of the condition for quasi-elasticity in the scattering and corresponds to the statement that the time of flight of the neutron across a fluctuation of size  $\sim q^{-1}$  is of the same order as or greater than the characteristic lifetime of the fluctuation. Thus, the largest value of the polarization arises when the energy exchange between the neutron and the spin fluctuations is maximal.

Since  $qa \ll 1$ , the restriction  $q \gtrsim q_c$  also means that  $T_c(2paE_0)^{-1} \gg 1$ , i.e., critical enhancement of the polarization is possible in the scattering of sufficiently cold neutrons in high-temperature ferromagnets. Generally speaking, then, the behavior of  $P(q)$  depends on the relative magnitudes of  $q$ ,  $q_c$ , and  $\chi$ . On the other hand, the exchange region is defined by the inequality  $\chi > q_0$ , and for most ferromagnets is not large. Taking into account also that we can only make asymptotic estimates for the polarization, there is no particular sense in considering the two cases  $\chi > q_c > q_0$  and  $q_c > \chi > q_0$ . Therefore, we confine ourselves to the case  $q_c \lesssim q_0$ . In this case we obtain a parametrically fairly well defined maximum for the polarization in the exchange region, and we can carry out matching with the

dipolar region.

The corresponding expression for the polarization for scattering through a given angle  $\vartheta$  has the form

$$P(\vartheta) \sim \frac{r_0 E_0 \mathbf{n}}{a T_c a^2} \begin{cases} \left(\frac{q_c}{p\vartheta}\right)^{3/2} \frac{1}{\chi p\vartheta}, & p\vartheta \gg \chi > q_0, \\ \left(\frac{q_c}{\chi}\right)^{3/2} \frac{p\vartheta}{\chi^3}, & \chi \gg p\vartheta, \chi > q_0, \end{cases} \quad (29)$$

where  $\mathbf{n} = \mathbf{p} \times \mathbf{p}' / |\mathbf{p} \times \mathbf{p}'|^{-1}$ . As  $\vartheta \rightarrow 0$  the polarization vanishes. This is a consequence of the additional symmetry of the three-point vertex (the special  $\mathbf{pp}'$  plane disappears) and of the regularity of  $G$  and  $\Gamma$  as  $\vartheta \rightarrow 0$ . It can be seen from (29) that  $P$  falls off with decrease of  $q_c$ ; therefore, the optimal value is  $q_c \approx q_0$ . In this case the maximal value of the polarization is attained on the boundary of the exchange and dipolar regions ( $\chi \approx q_0 \approx p\vartheta$ ) and is of the order of  $r_0 a^{-1} (q_0 a)^{-2} E_0 T_c^{-1}$ .

Finally, we shall discuss the estimation of the polarization in the dipolar region. As was noted above, in this region the three-spin correlator acquires anisotropy, due both to the anisotropy of  $G$  and to the anisotropy of the vertex  $\Gamma$ . At the same time, in the expression (25) for the polarization, from the vertex  $\Gamma^{\alpha\beta\gamma}$  only the part proportional to  $\varepsilon^{\alpha\beta\gamma}$  was taken into account. However, it can be shown that, although taking other parts of  $\Gamma^{\alpha\beta\gamma}$  into account does lead to a different angular dependence of the integral, it does not change the estimate obtained on the basis of formula (25). The only difference from the calculations in the exchange region is that one takes into account the different dependence of the characteristic energy on the momentum:

$$\Omega_q = \begin{cases} T_c(q_0 a)^{5/2} q a, & q_0 > q > \chi, \\ T_c(q_0 a)^{5/2} \chi a, & q_0 > \chi > q. \end{cases} \quad (30)$$

It is necessary to note that, in this dependence of the characteristic energy on the momentum, restrictions on  $q$  (like those in the exchange region) do not arise (for  $q_c \lesssim q_0$ ).

Evaluating the integral in (25) in accordance with the procedure described in the Appendix for  $q_c \approx q_0$ , we obtain

$$P(\vartheta) \sim \frac{r_0 E_0 \mathbf{n}}{a T_c a^2} \begin{cases} \left(\frac{q_0}{p\vartheta}\right)^{5/2} \frac{1}{p\vartheta}, & p\vartheta \gg q_0 \gg \chi, \\ \frac{1}{q_0^{5/2} (p\vartheta)^{5/2}}, & q_0 \gg p\vartheta \gg \chi, \\ \frac{p\vartheta}{q_0^{5/2} \chi^{5/2}}, & q_0 \gg \chi \gg p\vartheta. \end{cases} \quad (31)$$

As has been shown above, the polarization is most easily observed in ferromagnets with high  $T_c$  (e.g., in iron or nickel). It is known<sup>[17]</sup> that for Fe the quantity  $a \approx 1 \text{ \AA}$ , and the optimal conditions  $q_c \approx q_0$  for comparison with the theory are achieved with neutron wavelengths  $\lambda \approx 20 \text{ \AA}$ . In this case the polarization in the dipolar region for  $\tau \sim 10^{-4}$  reaches the value  $10^{-3}$ . In the exchange region the polarization does not exceed the value  $10^{-4}$ , which is reached on the boundary of the dipolar and exchange regions. The estimates for nickel give approximately the same results.

#### 4. POLARIZATION ON SCATTERING BY SPIN WAVES IN A MULTI-DOMAIN SAMPLE

In this section we shall calculate the polarization that arises in the scattering of neutrons in a ferromagnet at a sufficiently low temperature, when spin-wave theory is valid. We shall assume that the sample is divided into a large number of randomly oriented domains, so that there is no macroscopic magnetic moment. The neutron polarization arising in scattering in one domain consists of two parts. One part depends on the direction of the magnetization  $M$  of the domain and disappears on averaging over the orientations of the domains. The other part of the polarization, which, as above  $T_c$ , is due to three-spin correlations, has a component along  $n$  that does not depend on  $M$  and does not vanish on averaging. We shall calculate this part of the polarization. We shall neglect the effect of the dipolar forces and anisotropy on the spin waves.

It can be seen from the formula (23) for  $P$  that to determine the polarization we need an expression for  $\langle S_1^\alpha S_2^\beta S_3^\gamma \rangle$  averaged over the orientations of the domains. We shall express the spin projections  $S^\alpha$  in the fixed laboratory coordinate frame in terms of the spin projections in a coordinate frame with its  $z$  axis pointing along the domain magnetization:

$$S^\alpha = S^m m_x^\alpha + S^m m_y^\alpha + S^m m_z^\alpha, \quad (32)$$

where  $m_i$  are the unit vectors of the coordinate axes associated with the domain. Having written the spin components in terms of magnon creation and annihilation operators:

$$S^z = -S + a^\dagger a, \quad S^x = (S/2)^{1/2} (a^\dagger + a), \quad S^y = -i(S/2)^{1/2} (a^\dagger - a),$$

we average  $\langle S_1^\alpha S_2^\beta S_3^\gamma \rangle$  over the domain orientations. This averaging is easily performed if we take into account that the spatial and thermodynamic averaging are not coupled, and  $\langle m_x^\alpha m_x^\beta m_x^\gamma \rangle = 1/6\epsilon^{\alpha\beta\gamma}$ . As a result, from formula (29) we obtain

$$P \frac{d^2\sigma}{d\Omega dE'} = -\frac{N(r_0\gamma)^3}{3m} \frac{p'}{p} S \int \frac{d\mathbf{p}_1}{(2\pi)^3} F(\mathbf{q}) F(\mathbf{q}_1) F(\mathbf{q}_2) \times n(E_p, -E_p) n(E_{p'}, -E_{p'}) \times \{\delta(E_p - E_{p'} - \epsilon_{q_1}) \delta(E_p - E_{p'} - \epsilon_{q_2}) - \delta(E_{q_1} - E_{p'} + \epsilon_{q_2}) \delta(E_p - E_{p'} + \epsilon_{q_1})\} \times \{[\mathbf{e}_1 \times \mathbf{e}_2](\mathbf{e}_1 \mathbf{e}_2) - [\mathbf{e} \times \mathbf{e}_1](\mathbf{e}_1) - [\mathbf{e} \times \mathbf{e}_2](\mathbf{e}_2)\}, \quad (33)$$

where  $d^2\sigma/d\Omega dE'$  is the cross section, averaged over the domain orientations, for scattering by spin waves, and  $\epsilon_q = 2m\alpha q^2$ ;  $\alpha \sim 100$  for Fe and Ni.<sup>[17]</sup> Solving the equations obtained from the requirement that the arguments of the  $\delta$ -functions in (33) be equal to zero, we can conclude that the polarization is nonzero only when the inequality  $\alpha q \leq 2p$  is fulfilled, i.e., in scattering through small angles  $\vartheta \leq 2/\alpha$  and with small energy transfer  $|E_p - E_{p'}| \leq E_p [(2/\alpha)^2 - \vartheta^2]^{1/2}$ . Analogous conditions are also imposed on the virtual momentum and energy transfers; therefore, at high temperatures the statistical weights can be replaced by  $T(E_p - E_{p_1})^{-1}$  and  $T(E_{p'} - E_{p_1})^{-1}$ . As a result, it follows from (33) that

$$P \frac{d^2\sigma}{d\Omega dE'} = {}^2/_{3n} (2mr_0\gamma)^3 SN \frac{p'}{p} \frac{1}{(2\pi)^3} \int \sin \vartheta_2 d\vartheta_2 \int_0^{2\pi} d\varphi \int_0^\infty q_2^2 dq_2 \frac{T^2}{\alpha^2 q_2^2 (q - q_2)^2} \cos \vartheta_2 \sin \vartheta_2 \cos \varphi \left\{ \frac{q^2 - q_2^2}{(q - q_2)^2} - 1 \right\} \times \{\delta[2pq_2(\cos \theta \cos \vartheta_2 + \sin \theta \sin \vartheta_2 \cos \varphi) - \alpha q_2^2] \times \delta[2pq \cos \theta + \alpha q^2 - 2\alpha q q_2 \cos \vartheta_2]\} - \{\delta[2pq_2(\cos \theta \cos \vartheta_2 + \sin \theta \sin \vartheta_2 \cos \varphi) + \alpha q_2^2] \times \delta[2pq \cos \theta - \alpha q^2 + 2\alpha q q_2 \cos \vartheta_2]\}, \quad (34)$$

where  $\cos \vartheta_2 = (\mathbf{e} \cdot \mathbf{e}_2)$  and  $\cos \theta = (\mathbf{p} \cdot \mathbf{q})(pq)^{-1}$ . In this interval the second pair of  $\delta$ -functions differs from the first only in the sign in front of  $\alpha$ , and therefore it is sufficient to calculate the integral with the first term in curly brackets.

We introduce the notation  $u = \cos \theta$ ,  $u_1 = \cos \vartheta_2$ ,  $\alpha q/2p = v$ ,  $\alpha q_2/2p = v_1$ . Integrating over  $\vartheta_1$  and  $\varphi$  in formula (34), we obtain

$$P \frac{d^2\sigma}{d\Omega dE'} = -{}^2/_{3nm} \left(\frac{r_0\gamma}{2\pi}\right)^3 SN \frac{p'}{p} \left(\frac{T}{2E_p}\right)^2 \frac{1}{q} \frac{1}{(1-u^2)^{1/2}} \frac{1}{uv} \times \left[ \frac{u+v}{|u+v|} I^+ - \frac{u-v}{|u-v|} I^- \right], \quad (35)$$

where

$$I^\pm = \int_{u_1}^{u_2} u_1^2 du_1 \left\{ 1 + \frac{\Lambda(1-\Lambda)}{u_1^2 - \Lambda} \right\} \frac{1}{[(\beta_2 - u_1^2)(u_1^2 - \beta_1)]^{1/2}},$$

$$\Lambda = \frac{(u+v)^2}{4uv}, \quad \beta_{1,2} = \frac{uv+1}{2} \mp [(1-u^2)(1-v^2)]^{1/2}.$$

The expression for  $I^-$  differs from the formula for  $I^+$  by the replacement of  $v$  by  $-v$ . The integration limits  $\beta_{1,2}$  are determined from the requirement  $|\cos \varphi| \leq 1$ . Taking the integral in (35) does not present any difficulty; therefore, we give the final formulas for the polarization:

$$P \frac{d^2\sigma}{d\Omega dE'} = {}^1/_{3n} \frac{m\alpha}{p} (r_0\gamma)^3 SN \left(\frac{T}{2E_p}\right)^2 \frac{q^2/p^2}{(p-p')^2/p^2 - (\alpha/2)^2(q/p)^4} \left\{ \begin{matrix} a \\ b \end{matrix} \right\}, \quad (36)$$

where  $a = p\vartheta/2|p-p'|$  for

$$\begin{cases} \frac{1}{\alpha} - \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2} \leq \frac{p-p'}{p} \leq \frac{1}{\alpha} + \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2}, \\ -\frac{1}{\alpha} - \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2} \leq \frac{p-p'}{p} \leq -\frac{1}{\alpha} + \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2}. \end{cases}$$

and  $b = -[1 - (\alpha q/2p)^2]/\alpha\vartheta$  for

$$\begin{cases} \frac{p-p'}{p} \geq \frac{1}{\alpha} + \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2}, \\ \frac{p-p'}{p} \leq -\frac{1}{\alpha} - \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2}, \\ -\frac{1}{\alpha} + \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2} \leq \frac{p-p'}{p} \leq \frac{1}{\alpha} - \left(\frac{1}{\alpha^2} - \vartheta^2\right)^{1/2}, \\ \frac{2}{\alpha} > \vartheta > \frac{1}{\alpha}. \end{cases}$$

From formula (36) it can be seen that in scattering through a fixed angle,  $P \parallel n$  for all energy transfers. This is sufficient for the integral of the polarization over the energy to be nonzero. However, the expressions for  $P$  in the region of angles  $\vartheta < 1/\alpha$  diverge as the energy transfers approach the threshold for crea-

tion of a spin wave, and, therefore, in this region it is necessary to take into account the gap in the spin-wave spectrum.

For  $1/\alpha < \vartheta < 2/\alpha$  the expression (36) does not have singularities in the energy transfer, and for the polarization on scattering through angle  $\vartheta$  we have

$$P(\theta)\sigma(\theta) = \frac{1}{2}nSN \frac{(r_0\gamma)^3}{\theta} p \left( \frac{T}{2E_p} \right)^2 \times \left\{ \frac{\theta^2}{\alpha(\theta^2 - 1/\alpha^2)^{1/2}} \left[ \operatorname{arctg} \frac{1/\alpha + [(2/\alpha)^2 - \theta^2]^{1/2}}{(\theta^2 - 1/\alpha^2)^{1/2}} - \operatorname{arctg} \frac{1/\alpha - [(2/\alpha)^2 - \theta^2]^{1/2}}{(\theta^2 - 1/\alpha^2)^{1/2}} \right] + \frac{\theta^2}{2\alpha} \ln \frac{2/\alpha + [(2/\alpha)^2 - \theta^2]^{1/2}}{2/\alpha - [(2/\alpha)^2 - \theta^2]^{1/2}} - [(2/\alpha)^2 - \theta^2]^{1/2} \right\}. \quad (37)$$

For  $\vartheta > 1/\alpha$  the one-magnon scattering cross section, associated with the correlator  $\langle S_+ S_- \rangle$ , is equal to zero; therefore,  $\sigma(\vartheta)$  in (37) is determined by the longitudinal correlator, i.e., by processes in which two spin waves take part:

$$\sigma(\theta) = \frac{1}{2}nN \left( \frac{r_0\gamma}{2\pi} \frac{T}{E} \right)^2 \left( \frac{ap}{\alpha} \right)^2 \frac{C}{\theta^2}, \quad (38)$$

where  $C$  is a number of order unity.

It follows from the formulas (37) and (38) that the polarization in ferromagnets with  $\alpha \sim 100$  can reach a value of the order of one per cent. However, to observe it is rather complicated, since it has this magnitude in the small region of angles  $1/\alpha < \vartheta < 2/\alpha$ , where the total intensity of the scattered neutrons is small. Moreover, in the case that we have considered it is practically impossible to observe this polarization because of the depolarizing field of the random domains. At the same time, in the case of a band domain structure the depolarizing influence of the domains can be small, and, clearly, the value of the polarization will not differ greatly from that obtained above, so that in this case the effect considered can be observed experimentally.

In conclusion, the authors express their gratitude to A. I. Okorokov, V. V. Runov, A. G. Gukasov, and V. A. Ruban for a large number of interesting discussions.

## APPENDIX

We shall estimate the polarization in the exchange region ( $\kappa > q_0$ ) for  $p\vartheta \gg \kappa$ . The estimates for the other regions are made in an analogous manner.

The expression (25) for  $P$  in the coordinate frame in which the  $z$  axis is parallel to  $q$  and the  $zx$  plane is the scattering plane has the form

$$P \frac{d^2\sigma}{d\Omega dE'} = -i \frac{4\pi}{3m} N^2 \frac{p'}{p} \int \frac{\sin \vartheta_2 d\vartheta_2 d\varphi q_2^2 dq_2}{(2\pi)^2} \frac{T\Delta}{\omega} \left( \frac{T\Delta_1}{\omega_1} + \frac{T\Delta_2}{\omega_2} \right) \times [G_+(\omega) G_{q_+q_1}(\omega_1) G_{q_1}(\omega_2) \Gamma_{q_1-q_1-q_1}(\omega, \omega_1, \omega_2)] \times \cos \vartheta_2 \sin \vartheta_2 \cos \varphi \left( \frac{q^2 - q_2^2}{q^2 + 2qq_2 \cos \vartheta_2 + q_2^2} - 1 \right), \quad (A.1)$$

where

$$\begin{aligned} \omega_1 &= E_{p'} - E_{p+q_1} = E_{p'} - E_p - 2E_0 a^2 p q_2 \cos \psi - E_0 a^2 q_2^2, \\ \omega_2 &= E_{p+q_1} - E_p = 2E_0 a^2 p q_2 \cos \psi + E_0 a^2 q_2^2, \quad \omega = E_p - E_{p'}, \\ \cos \psi &= \cos \vartheta_2 \cos \theta + \sin \vartheta_2 \sin \theta \cos \varphi, \quad \cos \theta = (\mathbf{p}\mathbf{q}) / (pq)^{-1}; \end{aligned}$$

for small-angle scattering,  $\sin \theta \approx p\vartheta/q$ .

It can be seen from (A.1) that the integration over  $\varphi$  gives a nonzero result only when the dependence of the expression in square brackets (the discontinuities of the three-spin Green function) on the energies containing  $\cos \varphi$  is taken into account. As a result of taking the discontinuities, several terms arise. One of the terms giving the principal contribution is the simplest to estimate and has the form

$$T \frac{\Delta G_q(\omega)}{\omega} T \frac{\Delta_2 G_{q_1}(\omega_2)}{\omega_2} \Delta_1 G_{q_+q_1}(\omega_1) \Gamma_{q_1-q_1-q_1}(\omega, \omega_1, \omega_2), \quad (A.2)$$

where

$$\begin{aligned} \Gamma(\omega, \omega_1, \omega_2) &= \Gamma(\omega + i\delta, \omega_1 - i\delta, \omega_2 + i\delta) - \Gamma(\omega + i\delta, \omega_1 + i\delta, \omega_2 - i\delta) \\ &+ \Gamma(\omega - i\delta, \omega_1 - i\delta, \omega_2 + i\delta) - \Gamma(\omega - i\delta, \omega_1 + i\delta, \omega_2 - i\delta). \end{aligned}$$

The expression for  $\tilde{\Gamma}$  in the form of a combination of different branches of the function  $\Gamma$  has been obtained taking into account the fact that  $\Gamma$  is an analytic function of two independent pairs of arguments. Like  $F^{(0)}$  [cf. (6)], the expression (A.2) is purely imaginary and changes sign under the replacement  $\omega_i \rightarrow -\omega_i$ . If we take into account that  $\Delta G(\omega)$  is an odd function, it follows from this that  $\tilde{\Gamma}(\omega_i) = \tilde{\Gamma}(-\omega_i)$ . Therefore, for  $\omega_i \lesssim \Omega_{q_i}$  we shall assume  $\tilde{\Gamma}(\omega_i)$  to be a weakly varying function of  $\omega_i$ , finite at  $\omega_i = 0$ . The dependence of  $\tilde{\Gamma}$  on the momenta is determined by the formulas (27), (28).

We shall evaluate the integral over  $q_2$ . Since for  $\omega_2 \gg \Omega_{q_2}$  the functions  $G$  and  $\Gamma$  fall off rapidly,<sup>[14]</sup> the principal contribution arises from the region  $\omega_2 \lesssim \Omega_{q_2}$ . This, generally speaking, leads for fixed  $q_2$  to a restriction on the range of integration over the angles, and to an extra small factor. However, it is not difficult to convince oneself that for  $q_c \approx q_0$ , in the entire important range of integration  $q_2 \gtrsim q_0$ , this does not happen. Since the region  $p\vartheta \gg \kappa$  has been chosen,  $q \gg \kappa$ . Using the expressions (26) for  $G$  and (27), (28) for  $\Gamma$  it is easy to see that the integral over  $q_2$  converges and the principal contribution arises from the region  $q_2 \sim \kappa$ . For  $q \gg q_2 \sim \kappa$  we have

$$\Delta G_{q_1}(\omega_2) / \omega_2 \sim \Omega_{q_1}^{-1} \kappa^{-2} \sim \kappa^{-3/2}, \quad \Gamma(q_1 \sim q \gg \kappa) \sim q^{1/2} \kappa.$$

A factor proportional to  $\cos \varphi$  arises from  $\Delta G_{q_1}(\omega_1)$  and is of the order of

$$\Delta G_{q_+q_1}(\omega_1) \sim \frac{p\theta}{q} \frac{2E_0 p}{T_c \kappa^{1/2}} \frac{1}{q^2}.$$

Finally, we consider the remaining integral, over  $\vartheta_2$ :

$$\int_0^\pi \sin^2 \vartheta_2 \cos \vartheta_2 \left( \frac{q^2 - q_2^2}{q^2 + 2qq_2 \cos \vartheta_2 + q_2^2} - 1 \right) d\vartheta_2. \quad (A.3)$$

For  $q_2 = 0$  this expression vanishes. Therefore, for  $q_2 \sim \kappa \ll q$  an extra small factor  $\kappa q^{-1}$  appears.

As a result, taking into account all the estimates given above, we have

$$P(q) \sim \frac{r_0 E_0}{a} \frac{1}{T_c} \frac{1}{a} \left( \frac{q_c}{q} \right)^{1/2} \frac{1}{q\kappa} \frac{p\theta}{q}. \quad (A.4)$$

Here we have taken into account that

$$\frac{d^2\sigma}{d\Omega dE'} \sim \frac{T}{\omega} \Delta G_q(\omega),$$

and introduced the dimensional constant  $a$ , which was omitted above.

To obtain the polarization on scattering through angle  $\theta$  it is necessary to integrate  $P d^2\sigma/d\Omega dE'$  over the energy transfer and divide by  $\sigma(\theta)$ . The integration over  $E'$  is equivalent to integration over  $q$  over the region  $q \geq p\theta$ ; this leads simply to the replacement  $q \rightarrow p\theta$  in (A.4) and gives the expression (29) of the main text.

<sup>1</sup>We recall that by the discontinuity of a function across the cut we mean the quantity  $\Delta_{\pm} f(x) = [f(x+i\delta) - f(x-i\delta)]/2i$ .

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Translated by P. J. Shepherd

## An alternative explanation of the anomalies of the physical properties of Invar alloys

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(Submitted 30 March 1978)

*Zh. Eksp. Teor. Fiz.* **75**, 780-784 (August 1978)

The residual magnetic moment and unidirectional exchange anisotropy of a chromium-rich iron-chromium alloy and an iron-nickel Invar alloy were investigated as a function of temperature. In both cases the magnetic moment and the unidirectional anisotropy were retained in the temperature range between the Curie and Néel points. These experimental results were used to draw conclusions on the nature of the physical anomalies of Invar alloys. The proposed model was found to be in agreement with all the currently available experimental data.

PACS numbers: 75.50.Bb, 75.30.Gw, 75.10.-b

Several models have been suggested to explain the anomalies of the physical properties of Invar alloys. They include the latent antiferromagnetism model of Kondorskiĭ,<sup>[1-3]</sup> the model of two  $\gamma$  states of Weiss,<sup>[4]</sup> the models based on the alloy heterogeneity (Schlosser<sup>[5,6]</sup>) and allowing for the ordering in alloys (Kachi and Asano<sup>[7-9]</sup>), the model of weak band ferromagnetism of Wohlfarth,<sup>[10,11]</sup> and others. All these models explain more or less satisfactorily the anomalies of the dependence of the magnetic moment and Curie temperature of the alloys (FeNi, FePt, FePd) on their compositions, anomalous values of the high-field susceptibility, etc. However, none of these models explains all the physical properties of Invar alloys, for example, the temperature dependence of the linear expansion co-

efficient (see Fig. 2b below).<sup>[12]</sup> Some of them are even in conflict with the experimental observations.

In the models postulating antiferromagnetism of Invar alloys<sup>[3]</sup> there still remains the question how to explain the influence of antiferromagnetism on the physical properties of Invar alloys at temperatures above the Néel point  $T_N$  of the antiferromagnetic component (this temperature is  $\sim 50^\circ$  K for the Fe-Ni alloys).

We investigated chromium-rich Fe-Cr alloys from which samples with a low expansion coefficient, known as "nonmagnetic Invars," were prepared. The investigated alloys with 78-93% Cr exhibited unidirectional exchange anisotropy.<sup>[13]</sup> One of the investigated alloys