BEHAVIOR OF THE INITIAL SUSCEPTIBILITY OF THE PARA-PROCESS IN FERRO- AND FERRIMAGNETS NEAR THE CURIE TEMPERATURE

K. P. BELOV, A. N. GORYAGA, and C. SRINIVASAN

Moscow State University

Submitted to JETP editor July 4, 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) 51, 1639–1642 (December, 1966)

The initial susceptibility of the para-process χ_0 near the Curie point (approach from the high-temperature side) is investigated in some ferro- and ferrimagnets (ferrites). Some refinements are introduced into the method for determining χ_0. It is shown that for a number of ferrites the relation $\chi_0^{-1} = A(T - \Theta)^{\gamma}$ is valid in the indicated temperature range. The value of γ in this case is approximately the same (1.30–1.37) as for simple ferromagnetics.

RECENTLY, much attention has been given to the study of magnetic transformations in ferro- and antiferromagnetic substances, since such study results in data necessary for the improvement of the theory of phase transitions.

According to molecular-field theory and Landau's thermodynamic theory of second-order phase transitions,[1] in which only long-range magnetic order is taken into account, the initial susceptibility χ_0 of ferromagnets near the Curie point (approaching from the high-temperature side) depends on the temperature as

$$\chi_0^{-1} = A(T - \Theta)^{\gamma}, \qquad (1)$$

where $\gamma = 1$.

This relation is not satisfied for real ferromagnets, however. Right near the Curie temperature the dependence (1) is destroyed as a consequence of the appearance of spin correlations (short-range magnetic order). There are a great many theoretical and experimental papers[2-13] in which an attempt is made to define more accurately the character of the behavior of the susceptibility of ferromagnets near the magnetic transformation temperature and to determine the value of γ. The majority of the theoretical calculations lead to the value $\gamma = 4/3$ for simple ferromagnets.[2-8] Experimental values of γ for iron, nickel, gadolinium, etc., have been found both from purely magnetic measurements[9-13] and from experiments on the critical scattering of neutrons near the magnetic transition,[14] and they confirm these calculations ($\gamma = 1.30-1.37$).

In the present paper we are interested in the question of the validity of Eq. (1) for the description of the magnetic susceptibility in ferrimagnets, in particular in ferrites, near their Curie points ($T > \Theta$) and in their values of γ. Ferrites are uncompensated antiferromagnets, i.e., they are ferrimagnetic. As is known, the paramagnetic susceptibility of ferrites in the region $T > \Theta$ does not follow the Curie-Weiss law but the more complicated Néel law. Our experiments, however, showed that very close to the Curie point (on the high-temperature side), Eq. (1) is valid for the description of the initial susceptibility of the para-process.

Before we consider the results of the measurements of γ in ferrites, we should like to explain which means of determining the initial susceptibility χ_0 in the vicinity of the Curie point is the most accurate and what physical meaning should be attached to this quantity. There is some vagueness about this in the various investigations of $\chi_0(T)$ in region of the Curie point.

In the theoretical papers devoted to a calculation of γ near the Curie point, a single-domain ferromagnet is considered, and consequently χ_0 should be understood as the initial susceptibility of the para-process. Thus, in order to correctly evaluate γ in Eq. (1), it is necessary to apply to a real ferromagnet an external magnetic field of such size as to reduce it to the single-domain state. The fact is, in real ferromagnets near the Curie temperature and even somewhat above it, domains can still exist as a consequence of residual spontaneous magnetization ("tail" of σ_S). As experiments have shown,[15] the "tail" of σ_S is due mainly to inhomogeneity of composition over the sample volume.[16] This leads to fluctuations in the exchange integral and consequently to a diffuseness of the magnetic transformation region. Hence, it is very risky to try to verify Eq. (1) when the values of χ_0 are determined in weak fields, since that part of the susceptibility due to rem-

Table I. Determination of the quantity γ in Eq. (1) for invar* and nickel.

Material	γ			Temperature interval in which Eq. (1) is valid
	By the method of thermodynamic coefficients	In a field H = 1 Oe	In a field H = 5 Oe	
Invar, 10 h at 1000°C	1.16±0.0	0.79	0.98	$\Theta-18°$
Invar, 20 h at 1000°C + 20 h at 1200°C	1.20 0.04	0.53	0.52	$\Theta-7°$
Electropolished nickel	1.30±0.03	1.91	0.64	$\Theta-10°$

*Composition 36 wt% Ni + 64 wt% Fe.

nants of domain structure near the Curie point can also contribute to the quantity χ_0.

We measured, in the vicinity of the Curie temperature, the magnetic susceptibility of an alloy of composition 36 wt% Ni–64 wt% Fe in both weak and strong fields, and, in addition, the sample was subjected to two different homogenizing heat treatments. The samples were in the form of rods 4.5 mm in diameter and 130 mm long (the measurements were made by the ballistic method, and the temperature gradient along the length was negligibly small). Since the magnetization is very small near the Curie temperature, the demagnetizing field in these samples was consequently small and could not significantly affect the results of the measurement.

Table I gives the values of γ for this invar alloy for two homogenizing heat treatments and for electropolished nickel; χ_0 was determined from the magnetization curves in weak fields (1 and 5 Oe) and in strong magnetic fields (up to 2000 Oe). In the latter case χ_0 was found by treating the data by the method of thermodynamic coefficients that we presented earlier.[17]

Table II. Values of γ in ferrites.

Ferrite	γ*	Temperature interval in which Eq. (1) is valid
$Ni_{0,25}Zn_{0,75}Fe_2O_4$	1,37±0,02	Θ--26°
$Ni_{0,3}Zn_{0,7}Fe_2O_4$	1,31±0,03	Θ--18°
$Ni_{0,4}Zn_{0,6}Fe_2O_4$	1,30±0,02	Θ—55°
$Ni_{0,5}Zn_{0,5}Fe_2O_4$	1,38±0,03	Θ--58°
$Ni_{0,6}Zn_{0,4}Fe_2O_4$	1,35±0,03	Θ—25°
$Ni_{0,7}Zn_{0,3}Fe_2O_4$	1,31±0,04	Θ—19°
$Ni_{0,8}Zn_{0,2}Fe_2O_4$	1,30±0,04	Θ—11°
$Ni_{0,9}Zn_{0,1}Fe_2O_4$	1,33±0,04	Θ-- 9°
$NiFe_2O_4$	1,32±0,04	Θ-- 8°
$MnFe_2O_4$ (single crystal)	1,35±0,02	Θ—31°

*Determined by the method of thermodynamic coefficients.

The results of the measurements were plotted in H/σ, σ^2 coordinates; the values of χ_0^{-1} were found by extrapolating the straight lines obtained (in the region $T > \Theta$) to the ordinate axis. It is seen from Table I that the magnitudes of γ found by the method of thermodynamic coefficients differ sharply from those determined in weak fields. In addition, the former in invar do not depend on heat treatment, whereas the magnitudes determined in weak fields depend strongly on this; this is evidence that inhomogeneities of composition significantly affect the behavior of the magnetization curves in weak fields near Θ.

In our opinion, the most accurate means of determining the magnitude of the initial susceptibility of the para-process near the Curie point is the method of thermodynamic coefficients. We note that Graham obtained by this method $\gamma = 1.33$ for a crystal of gadolinium,[12] and Kouvel and Fisher[13] found for nickel $\gamma = 1.35 \pm 0.02$.

In our work, we found for nickel $\gamma = 1.30 \pm 0.03$, and for invar this quantity is somewhat smaller. The latter requires special explanation, since the magnetic transformation in invar alloys is anomalous.[1]

We now return to the results of measurements of γ in ferrites. The measurements were carried out on ferrites of the system $Ni_{1-x}Zn_xFe_2O_4$, where x = 0.0 to 0.75. The samples were prepared by the usual ceramic technique. Final sintering took place in air for 6 h at 1250°C. The samples were in the form of parallelepipeds of length 60 mm and cross-section 3 × 4 mm. In addition a single crystal of manganese ferrite, prepared by the Verneuil method, was studied. The values of χ_0 were determined by the method of thermodynamic coefficients.

As is seen from Table II, for all compositions the value of γ varies within the limits 1.30–1.37, i.e., in the first approximation the same as in sim-

ple ferromagnets. It follows from this that the magnetic phase transition in ferrimagnets has the same basic features as in simple ferromagnets.

[1] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Gostekhizdat, 1957 (English Transl., Pergamon, N. Y., 1960); K. P. Belov, Magnitnye prevrashcheniya (Magnetic Transitions), Fizmatgiz, 1959 (English Transl., Consultants Bureau, N. Y., 1961).

[2] C. Domb and M. F. Sykes, Proc. Roy. Soc. (London) **A240**, 214 (1957); Phys. Rev. **128**, 168 (1962).

[3] G. A. Baker, Phys. Rev. **124**, 768 (1961); **129**, 99 (1963); **136**, A1376 (1964).

[4] C. Domb, Advances in Physics **9**, 149, 245 (1960).

[5] N. W. Dalton and D. W. Wood, Phys. Rev. **138**, A779 (1965).

[6] G. Dresselhaus, Phys. Rev. **139**, A855 (1965).

[7] J. Gammel, W. Marshall, and L. Morgan, Proc. Roy. Soc. (London) **A275**, 257 (1963).

[8] A. Z. Patashkinskiĭ and V. L. Pokrovskiĭ, JETP **50**, 439 (1966), Soviet Phys. JETP **23**, 292 (1966).

[9] J. E. Noakes and A. Arrot, J. Appl. Phys. **35**, 931 (1964).

[10] S. Arajs and R. V. Colvin, J. Appl. Phys. **35**, 2424 (1964).

[11] S. Arajs, J. Appl. Phys. **36**, 1136 (1965).

[12] C. D. Graham, J. Appl. Phys. **36**, 1135 (1965).

[13] Y. S. Kouvel and M. E. Fisher, Phys. Rev. **136**, A1626 (1964).

[14] L. Passell, K. Blinowski, T. Brun, and P. Nielsen, Phys. Rev. **139**, A1866 (1965).

[15] K. P. Belov and A. N. Goryaga, Izv. AN SSSR, ser. fiz., **21**, 1038 (1957).

[16] K. P. Belov and A. N. Goryaga, FMM **2**, 3 (1956).

[17] K. P. Belov and A. N. Goryaga, FMM **2**, 441 (1956).

Translated by L. M. Matarrese
198