NONLINEAR DEPENDENCE OF THE FERROMAGNETIC TRANSITION TEMPERATURE OF TWO INVAR ALLOYS ON THE HYDROSTATIC PRESSURE

L. D. LIVSHITZ and Yu. S. GENSHAFT

Institute of Physics of the Earth, Academy of Sciences, U.S.S.R.

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The dependence of the Curie temperature on pressure was investigated experimentally for Elinvar and Coelinvar alloys at pressures up to 20 kbar. It is shown that the pressure dependence of the Curie temperature shift $\Delta\Theta$ can be represented by an equation of the form of $\Delta\Theta = -ap - bp^2$.

IN investigating second-order phase transitions under pressure, it is obviously interesting to determine the influence of pressure on the magnetic transition temperature.

In the initial stage of the work, it is natural to turn to materials for which—according to the data obtained by other methods,—considerable pressure effects can be expected.[1] This was one of the reasons for investigating Invar alloys. The material for the preparation of the samples was kindly supplied by G. I. Kataev, to whom the authors are greatly indebted.[1)]

The method for establishing and measuring the pressures and temperatures was described in detail earlier.[3] We note only that, although in that method a solid plastic substance (and not a liquid) was used to transmit pressure, we may assume that under test conditions the sample is practically under hydrostatic pressure. The tests consisted of measuring, at fixed values of pressure or temperature ("isobars" or "isotherms"), the dependence of the permeability μ on the appropriate variable —temperature or pressure— $\mu(T)$ or $\mu(p)$. The measurements were carried out in weak (up to 3–5 Oe) alternating fields of 500 cps frequency.

A series of such "isobars," representing the variation of the permeability of one of the samples of alloy No. 2 on increase of temperature, is contained in Fig. 1. Figure 2 gives the $\mu(p)$ curves for alloy No. 1, recorded with the pressure increasing. In these tests, the temperature was usually measured and kept constant to within ±1.5 deg C.

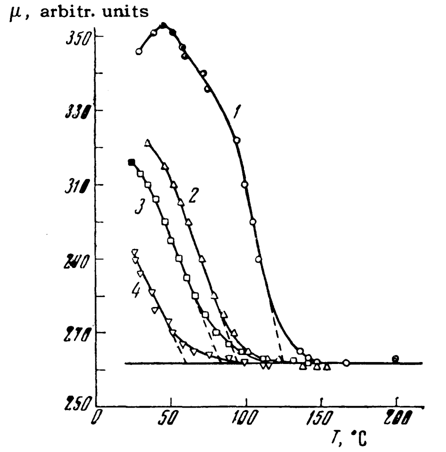

FIG. 1. Dependence of the permeability of alloy No. 2 on temperature at various pressures: 1) p = 0.3 kbar, Θ = 125°C; 2) p = 3.7 kbar, Θ = 95°C; 3) p = 5.9 kbar, Θ = 83°C; p = 8.1 kbar, Θ = 60°C.

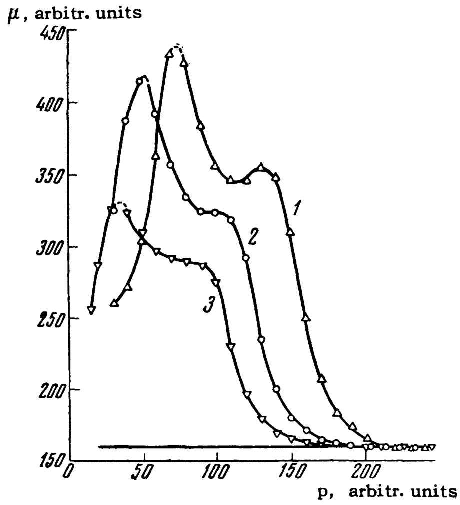

FIG. 2. Dependence of the permeability of alloy No. 1 on pressure at various temperatures: 1) at 0°C; 2) +13.5°C. 3) +19.5°C.

[1)]According to Kataev's data[2], the alloys were in the annealed state (1030–1040°C, 5 hours) and had the following composition: alloy No. 1, Elinvar type with 36% Ni, 12% Cr, 52% Fe; alloy No. 2, Coelinvar type with 53.5% Co, 8.7% Cr, 37.8% Fe.

In the curves of Fig. 2 it is worth noting the first maximum, which appears clearly in samples of alloy No. 1 at temperatures below 20°C. So far such maxima have apparently been observed only in the curves of the temperature dependence of the initial permeability. In the case of nickel, such a maximum of the $\mu(T)$ curve (near 200°C) is ascribed to a change of the crystallographic direction of the easy magnetization.[4] In ternary alloys of iron with 79% Ni and 3—5% Mo, and of iron with 4—14% Si and 0.5—7.5% Al, this maximum is ascribed to ordering processes.[4,5] It is possible that the maximum noted in the $\mu(p)$ curves of alloy No. 1 is due to the $\alpha \rightleftarrows \gamma$ transition, which, as is known,[4] occurs in alloys of iron with 36% Ni and 10—15% Cr at atmospheric pressure in the region of room temperature.

The Curie point was taken to be the intersection of the tangents to the steep parts of the $\mu(p)$ or $\mu(T)$ curves with a line to which these curves tend in the paramagnetic region (as shown by dashed lines in Fig. 1). The ordinate of this line (which in our experiments was parallel to the abscissa axis) is governed by the constants of the measuring device and does not alter, as shown in Figs. 1 and 2, during a series of tests on one sample. The difference between the values of Θ at atmospheric pressure, obtained in the present experiments and in [2], and which is unimportant in the problem discussed here, is due to the difference in the methods of determining the Curie point.

Several series of tests on both alloys gave plots of the pressure dependence of the Curie temperature, shown in the (p, T) diagram of Fig. 3. Each

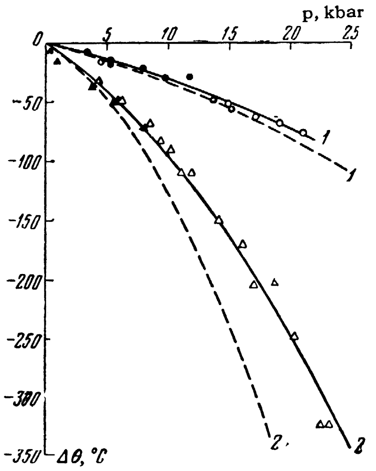

FIG. 3. Shift of the Curie temperature with pressure: 1) alloy No. 1; 2) alloy No. 2. Black symbols denote experimental values obtained by isobars and open symbols those determined from isotherms; dashed curves represent the results of calculations using Eq. (3).

point of Fig. 3 represents an arithmetic average value of Θ obtained with the pressure (or temperature) rising and falling. The observed hysteresis of the magnetic transition (which is influenced mainly by the experimental method) does not usually exceed the indeterminacy in the Curie temperature, found by the method just described. The diagram shows that the points obtained from the isotherms and isobars satisfactorily fit continuous curves.

$\Delta\Theta(p)$ curves of both alloys may be represented in the (p, T) plane by a quadratic equation with two constants:

$$\Delta\Theta = \Theta_p - \Theta = -ap - bp^2, \quad (1)$$

where Θ_p is the magnetic transition temperature at a pressure p. The coefficients a and b were determined by the least squares method for a linear equation in terms of coordinates $\Delta\Theta/p$ and p:

$$\Delta\Theta/p = -a - bp. \quad (2)$$

The average relative deviation of individual experimental points from the curves which were plotted using Eq. (1) does not exceed 6.5% in pressure, and 8% in temperature.

It is interesting to compare the results of direct measurements of the dependences $\Theta(p)$ with those of calculations using thermodynamic formulas. Thus, from a series expansion of the thermodynamic potential in the region of the Curie point we can obtain a quadratic expression for the pressure dependence of the Curie temperature:[2]

$$\Delta\Theta = -\frac{3\gamma}{\alpha'_\Theta} p - \frac{3(\varepsilon_1 - 2\varepsilon_2)}{\alpha'_\Theta} p^2, \quad (3)$$

where $\alpha'_\Theta = \alpha/(T - \Theta)$, and α, γ, ϵ_1 and ϵ_2 are thermodynamic coefficients in the notation used by Kataev and determined by him from his own experimental data.[2)] A comparison of the coefficients of p and p^2 in the expressions (3) and (1) is given in the table [p is expressed in kilobars (10^9 dyn/cm^2), and $\Delta\Theta$ —in deg C].

Here, P_a and P_b are the probable deviations of the coefficients in Eq. (2). The table and Fig. 3

Alloy	$a \pm P_a$	$3\gamma/\alpha'_\Theta$	$b \pm P_b$	$3(\varepsilon_1-2\varepsilon_2)/\alpha'_\Theta$
№ 1	2.6±0.3	2.6	0.05±0.01	0.07
№ 2	6.7±0.6	6.2	0.27±0.01	0.63

2)Equation (3) is similar to Eq. (8) in[2], which was obtained for uniaxial loads.

show that the initial parts of the calculated $\Theta(p)$ curves are in good agreement with the experimental data.

There is some interest in an estimate of the dependence of Θ on the variation of the interatomic spacing R with pressure:

$$\frac{d\Theta}{dR} = \frac{d\Theta}{dp}\frac{dp}{dR} \approx -\frac{3K}{R}\frac{d\Theta}{dp}, \tag{4}$$

where K is the bulk modulus. The coefficients in the right-hand part of Eq. (4) are functions of temperature and pressure. A rough estimate of the relative slope of the $\Theta(R)$ curves of the investigated alloys may be obtained by substituting in Eq. (4) the initial values of K and $d\Theta/dp$. It is then found that $Kd\Theta/dp$ of both alloys differs by only 15%. If it is arbitrarily assumed that $R = 2.5$ Å, then $d\Theta/dR$ of these alloys is found to be of the order of $3-3.5 \times 10^{11}$ deg/cm. Obviously, more reliable estimates need an exact knowledge of the bulk modulus and of the actual phase composition of the samples under pressure.[3)]

We have thus demonstrated experimentally the existence of nonlinear pressure dependences of the ferromagnetic transition temperature. Such dependences of temperatures of phase transitions of the second kind follow, for example, from the analyses of Kholodenko [6] and Ginzburg. [7] However, in the experiments on the ferromagnetic transition (cf. reviews [1] and [4]) only linear $\Theta(p)$ dependences were observed, which was probably due to the small range of pressures used in such investigations and in some cases due to the smallness of the effects themselves. Thus, for example, according to our preliminary measurements up to 26 kbar, the experimental results for gadolinium are equally well described by a curve of the type of Eq. (1) with coefficients $a = 1.44$ and $b = -0.004$ and by a straight line with a slope $d\Theta/dp = -1.37$ deg/kbar.

In conclusion, we regard it as our pleasant duty to thank Yu. N. Ryabinin and R. Z. Levitin for their interest in the present work.

3)The value of K was calculated from the formula $K = E/(9 - 3E/G)$, where E and G are values of the elasticity moduli given in[2] for the ferromagnetic phase at $T = \Theta$.

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