

Baric anomaly in the bulk modulus of the Invar-like alloy $\text{Fe}_{72}\text{Pt}_{28}$

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An analysis based on Landau's theory of phase transitions shows that the anomaly in the bulk modulus of the Invar-like alloy $\text{Fe}_{72}\text{Pt}_{28}$ is magnetic in nature. This theory yields a quantitative interpretation of the anomaly. A new method is proposed for determining a magnetoelastic parameter.

Detailed experimental studies of the pressure dependence of the elastic moduli of the Invar-like alloy $\text{Fe}_{72}\text{Pt}_{28}$ have recently revealed a remarkable property of this alloy: It becomes easier to compress as the applied pressure is raised.^{1,2} Magnetoelasticity arguments had been invoked previously to explain the Invar-like properties of the elasticity of this alloy.³ In the present letter we report some results derived by an approach which starts from the Landau free energy F_M , which is familiar in the theory of phase transitions. These results show how the abrupt change³ in the bulk modulus K_B and the abrupt change^{1,2} in the baric derivative $(\partial K_B/\partial P)_{T,B}$ at a constant temperature T and a constant magnetic induction B are governed by magnetic properties: The Curie temperature, its derivative with respect to volume, the Curie constant, the coefficient of the M^4 term in the expansion of the free energy in the magnetization M , and a magnetoelastic parameter. There is a quantitative agreement between the elastic characteristics predicted theoretically and those found experimentally.

To describe the vicinity of the ferromagnetic transition, we use the Ginzburg-Landau free energy

$$F_M(V, T, M) = V \left(\frac{1}{2} a_1(V, T) M^2 - \frac{1}{4} a_3(V) M^4 \right), \quad (1)$$

where $a_1(V, T) = \alpha(V)[T - T_c(V)]$. We wish to reach an understanding of the unusual changes in the elastic properties at the ferromagnetic transition on the basis of such quantities (found from experiments on magnetic properties) as the Curie temperature $T_c(V)$, its baric derivative, the Curie constant $C(V) = \alpha^{-1}(V)$, the expansion coefficient $a_3(V)$ in expression (1), and the magnetoelastic parameter K' , which is a measure of the magnetization dependence of the bulk modulus at a constant magnetization:

$$K_M \equiv V \left(\frac{\partial^2 F_M}{\partial V^2} \right)_{T, M} = K_0(V, T) + K' M^2. \quad (2)$$

Using (1), we find

$$K' = \frac{1}{2} \left(\frac{\partial a_1}{\partial \ln V} \right)_T + \frac{1}{2} \left(\frac{\partial^2 a_1}{\partial (\ln V)^2} \right)_T = \frac{T_c}{2C} \left[\frac{d \ln T_c}{d \ln V} \left(1 - 2 \frac{d \ln C}{d \ln V} \right) + \frac{1}{T_c} \frac{d^2 T_c}{d (\ln V)^2} \right]. \quad (3)$$

The experimental results correspond to the bulk modulus K_B , at a constant magnetic induction, which is related to K_M by the thermodynamic relation^{4,5}

$$K_B = K_M - \chi_V^{-1} (\partial M / \partial \ln V)_{T,B}^2, \quad (4)$$

where $\chi_V = (\partial M / \partial B)_{T,V}$ is the isothermal magnetic susceptibility at constant volume. The free energy in (1) can be used to rewrite relation (4) as

$$K_B - K_M = - \frac{1}{2a_3} \left(\frac{\partial a_1}{\partial \ln V} \right)_T^2 \frac{1}{1 + \xi} \equiv \frac{\Delta K}{1 + \xi}, \quad (5)$$

where $\xi = (B/2a_3M^3)$ describes the effect of the magnetic induction.

Since we have $\xi \gg 1$ in the paramagnetic state and $\xi \ll 1$ in the ferromagnetic state, the quantity ΔK in fact is the abrupt change in the bulk modulus K_B at the phase transition. Here

$$\Delta K = - \frac{T_c^2}{2a_3 C^2} \left(\frac{d \ln T_c}{d \ln V} \right)^2 \equiv - \frac{K_B^2}{2a_3 C^2} \left(\frac{dT_c}{\partial P} \right)^2. \quad (6)$$

What does the theory predict regarding the value of ΔK for $\text{Fe}_{72}\text{Pt}_{28}$? According to Refs. 1 and 2, we have T_c 367 K and K_B (360 K) = 87 GPa. According to Fig. 4 of Ref. 3, on the other hand, the value is $K_B(T_c) = 115$ GPa. This difference will correspond to our two estimates. We take the value of the baric derivative of the Curie temperature from Ref. 6 for an alloy with $T_c = 367$ K: $dT_c/dP = -36$ K/GPa. From Ref. 4 we have $C = 0.575$ K. Finally, we take the value $a_3 C = 4.7 \times 10^{-5}$ K/G² from Fig. 2 of Ref. 7 for the temperature dependence of the magnetization at $B = 0$, since near T_c we have $a_3 C M^2 = T_c - T$ according to Eq. (1). From (6) we then find $(\Delta K)_{\text{theor}} = -(19-32)$ GPa. To compare this theoretical value with the experimental value of the jump in the modulus K_B , which was studied in Ref. 3, we use the temperature dependence of the change in K_B from its value at the Curie point to its minimum value at $T \approx 333$ K from Table I and Fig. 4 of that paper. We then have $(\Delta K)_{\text{exp}} = -28$ GPa. Above the Curie point, there is a substantial change in K_B over a temperature range ≈ 200 K wide. According to Eq. (5), however, this change in K_B cannot be associated with a phase transition within the framework of Landau's theory.

The approximate agreement between the jump ΔK given by (6) and that found experimentally supports the idea that magnetism has a governing effect on the softening of the bulk modulus of the alloy $\text{Fe}_{72}\text{Pt}_{28}$. Working from this correspondence, we can attempt to explain the anomalous behavior of the baric derivative $(\partial K_B / \partial P)_{T,B}$ on the basis of Landau's theory of phase transitions.

Differentiating (4) with respect to the pressure P , and using (1), we find

$$\left(\frac{\partial(K_B - K_0)}{\partial P}\right)_{T,B} = \frac{3K'}{K_B a_3 (1 + \xi)} \left(\frac{\partial a_1}{\partial \ln V}\right)_T + \frac{2\Delta K}{K_B (1 + \xi)} \times \left(1 + \frac{6 + 15\xi}{4(1 + \xi)^2} \frac{d \ln a_3}{d \ln V} + \frac{3\xi}{4a_3 (1 + \xi)^2} \left(\frac{\partial a_1}{\partial \ln V}\right)_T \frac{1}{M^2}\right). \quad (7)$$

Here we have used the relation $(\partial/\partial P)_{T,B} = -K_B^{-1}(\partial/\partial \ln V)_{T,B}$, and K' and ΔK are given by (3) and (6). The jump caused in the baric derivative in (7) by the magnetism at the ferromagnetic transition ($T = T_c$) is given by

$$\Delta \left(\frac{\partial(K_B - K_0)}{\partial P}\right)_{T,B} = \frac{3K'}{a_3 C} \frac{dT_c}{dP} + \frac{2\Delta K}{K_B} \left(1 + \frac{3}{2} \frac{d \ln a_3}{d \ln V}\right). \quad (8)$$

Using the experimental value given above, along with the experimental result $K' = 1.0 \times 10^5$ from Ref. 3, we find an estimate of -24 for the first term on the right side of (8). Theoretical estimates of $d \ln a_3 / d \ln V$ lead to values on the order of a few units, while $2\Delta K / K_B$ is less than unity. In making the comparison with experiment, we will accordingly ignore the second term in (8). Since we have $\Delta(\partial K_B / \partial P)_{\text{exp}} \simeq -30$ according to Refs. 1 and 2, we draw the (satisfying) conclusion that the magnetic (or magnetoelastic) explanation of the anomalous properties of the bulk modulus of the Invar-like alloy $\text{Fe}_{72}\text{Pt}_{28}$ is confirmed. Since the absolute value of the jump in (8) is so large, we conclude that the magnetic component which we have been discussing here which is responsible for the large negative value of the baric derivative observed in Refs. 1 and 2.

Having drawn this conclusion, we can immediately suggest a new method for determining the magnetoelastic parameter K' in (3). This new method, which differs from the model-based approach of Ref. 3, is based on the use of Eq. (8) and experimental data on the jump in the baric derivative of K_B . As an example we consider the Invar-like alloy $\text{Fe}_{72}\text{Pt}_{28}$. If we ignore the second term on the right side of (8) and use the experimental values of $\Delta(\partial K_B / \partial P)$, $a_3 C$, and dT_c / dP given above, we find the following value for the magnetoelastic parameter: $K' = 1.3 \times 10^5$. This value is not greatly different from the estimate of K' in Ref. 3.

Our analysis shows how one can work from known magnetic properties, to determine the elastic properties of ferromagnets, in particular, to identify the reason for the remarkable property of this Invar-like iron-platinum alloy of becoming more compressible as the pressure is raised.

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