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H. Pascard, A. Globus. Volume anomaly in ferrimagnetism. Journal de Physique Lettres, 1981, 42 (13), pp.319-322. 10.1051/jphyslet:019810042013031900 . jpa-00231936

HAL Id: jpa-00231936

<https://hal.science/jpa-00231936>

Submitted on 1 Jan 1981

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Classification

Physics Abstracts

65.70 — 75.50G — 75.80

Volume anomaly in ferrimagnetism (*)

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(Reçu le 13 février 1981, révisé le 30 avril, accepté le 6 mai 1981)

Résumé. — L'anomalie de volume $\Delta V/V$ due à l'énergie magnétique associée aux interactions d'échange est déterminée expérimentalement pour $Y_3Fe_5O_{12}$. Les valeurs mesurées de 77 K à T_c coïncident avec les valeurs obtenues à partir de l'expression basée sur l'approximation du champ moléculaire et sur les théories de Néel de l'anomalie de dilatation et du ferrimagnétisme. Ces résultats sont comparés à ceux obtenus par d'autres auteurs pour des matériaux ferromagnétique et antiferromagnétique à moments localisés : une courbe réduite est obtenue.

Abstract. — The volume anomaly $\Delta V/V$ due to the magnetic energy corresponding to the exchange interactions is experimentally determined for YIG. The experimental values (from 77 K to T_c) agree with the values deduced from the theoretical expression based on the Néel's theories of volume anomaly and of ferrimagnetism. These results are compared with those obtained by other authors on ferromagnetic and antiferromagnetic materials with localized magnetic moments : a reduced curve is obtained.

1. Introduction. — This letter reports some results of a systematic investigation in order to check experimentally a hypothesis proposed previously by one of the authors about the relation between the magnetoelastic anisotropy found in ferrimagnetic polycrystals, and the volume anomaly [1]. So, the results about the volume anomaly in ferrimagnetism are presented.

The volume anomaly of cubic magnetic materials corresponds to the isotropic spontaneous deformation accompanying the occurrence of the magnetic-order state associated with exchange interactions at and below the critical temperature T_c .

For ferromagnetic materials Néel showed that this volume anomaly was due to the variation of the interaction energy (coupling forces) between magnetic atoms as a function of the distance of the magnetic layers [2].

For insulating cubic magnetic materials Bloch stated precisely the law for the volume dependence of the superexchange interactions [3] and related quantitatively the volume anomaly at 0 K to the variation of T_c with pressure [4].

2. Theoretical expression of the volume anomaly. — Let us consider a magnetic material with localized

moments and one kind of magnetic ion and assume that the exchange coupling is of the Heisenberg type $-2 J_{ij} S_i S_j$. In the approximation of the molecular field theory the magnetic energy per unit volume is written as [5] :

$$E_m = - N \frac{3}{2} k_B T_c \frac{S}{S+1} \sigma^2, \quad (1)$$

where

N is the number of magnetic ions per unit volume, T_c the critical temperature (directly proportional to the exchange interactions),

S the spin of the magnetic ion and

σ the reduced magnetization $m(T)/m(0)$.

Assuming a volume dependence of the exchange interaction of the form V^β , for a small volume variation $\Delta V/V$, T_c can be written as [6] :

$$T_c = T_0 \left(1 + \beta \frac{\Delta V}{V} \right) \quad (2)$$

T_0 would be the critical temperature if the lattice were not compressible, β may be positive or negative.

The sum of the magnetic and elastic energies per unit volume is :

$$E = - N \frac{3}{2} k_B T_0 \left(1 + \beta \frac{\Delta V}{V} \right) \times \frac{S}{S+1} \sigma^2 + \frac{1}{2\chi} \left(\frac{\Delta V}{V} \right)^2 \quad (3)$$

where χ is the compressibility.

(*) La version française de cet article a été acceptée aux Comptes Rendus de l'Académie des Sciences et elle est insérée dans le n° du 2 février 1981, série II, p. 439.

Minimizing the energy with respect to the volume variation we obtain directly the volume anomaly corresponding to the equilibrium state at $T < T_c$:

$$\frac{\Delta V}{V} = + N \frac{3}{2} k_B T_0 \beta \chi \frac{S}{S+1} \sigma^2 \quad (4)$$

ΔV is the difference between the volume of the magnetic material and the volume that a non magnetic material (with a similar crystallographic structure) would have (at the same temperature $T < T_c$).

We have

$$\frac{\Delta V}{V} = - N \frac{3}{2} k_B \frac{dT_c}{dP} \frac{S}{S+1} \sigma^2$$

$$\left(\text{since } \frac{dT_c}{dP} = \beta T_0 \frac{d(\Delta V/V)}{dP} = - T_0 \beta \chi \right). \quad (5)$$

For a ferrimagnetic material with 2 sublattices, a and d , and with only one kind of magnetic ions, and assuming antiferromagnetic interaction between the two sublattices to be dominant, the magnetic energy is [5] :

$$- N \frac{3}{2} k_B T_c \frac{S}{S+1} 2 \lambda \sigma_a \sigma_d \quad (6)$$

where σ_a and σ_d are the reduced magnetizations of the two sublattices a and d . λ is the fraction of magnetic ions in site a ($\lambda = 0.4$ for YIG) hence :

$$\frac{\Delta V}{V} = - N \frac{3}{2} k_B \frac{dT_c}{dP} \frac{S}{S+1} 2 \lambda \sigma_a \sigma_d. \quad (7)$$

3. Experimental determination of the volume anomaly in YIG. — The thermal expansion was measured between 77 K and 1 000 K by using a single-rod type silica dilatometer. The length variation of the samples was detected with a linear variable differential transformer. A precision of 0.025×10^{-3} was obtained. Figure 1 shows the thermal expansion measured on YIG.

Fig. 1. — Thermal expansion for $Y_3Fe_5O_{12}$ and $Y_3Ga_5O_{12}$ versus temperature.

In order to separate the magnetic thermal expansion contribution from that due to the lattice vibrations, we compare the $Y_3Fe_5O_{12}$ thermal expansion curve with that of the non magnetic garnet $Y_3Ga_5O_{12}$, whose thermal expansion coefficient, when far from T_c (in the ranges 0-100 K [7] and 650-1 000 K [8]), is the same as the one of $Y_3Fe_5O_{12}$ in these temperature ranges.

Then, one can separate at any temperature $T \lesssim T_c$ the magnetic contribution from that due to the lattice vibrations :

$$\left(\frac{\Delta l}{l} \right)_{\text{magn.}} = \left(\frac{\Delta l}{l} \right)_{\text{YIG}} - \left(\frac{\Delta l}{l} \right)_{\text{non magn. garnet}}. \quad (8)$$

Figure 2 shows the magnetic thermal expansion contribution $(\Delta l/l)_{\text{magn.}}$ of YIG versus temperature. One can see that at T_c a deformation is measured, which is due to the short-range order effects (or spin correlation effects).

Fig. 2. — Magnetic thermal expansion for $Y_3Fe_5O_{12}$ versus temperature.

We assume here that, for the analysis of the magnetic thermal expansion contribution $(\Delta l/l)_{\text{magn.}}$, one can separate the ferrimagnetic contribution $(\Delta l/l)$ due to the ferrimagnetic order energy ($(\Delta l/l) \neq 0$ for $T < T_c$ and $(\Delta l/l) = 0$ for $T \geq T_c$) and the short-range order contribution $(\Delta l/l)_{\text{spin corr.}}$, taking as a first approximation

$$\left(\frac{\Delta l}{l} \right) = \left(\frac{\Delta l}{l} \right)_{\text{magn.}} - \left(\frac{\Delta l}{l} \right)_{\text{spin corr.}} \quad (9)$$

with $(\Delta l/l)_{\text{spin corr.}} = (\Delta l/l)_{T_c}$.

This hypothesis seems justified, since we analyse only the length anomaly corresponding to the ferrimagnetic order energy, below T_c . This method is comparable with that used to obtain the value of the spontaneous magnetization deduced from a difference between the total saturation magnetization and the magnetization part due to the paramagnetism (this value of spontaneous magnetization is also zero at T_c).

Moreover, we find that the YIG thermal expansion

curves for polycrystals and for single crystals (in the [111] direction) are the same (within the limit of experimental precision $\pm 0.025 \times 10^{-3}$). Thus, the volume anomaly is defined as

$$\frac{\Delta V}{V} = 3 \frac{\Delta l}{l} \tag{10}$$

Then, we find the experimental volume anomaly at 0 K for YIG :

$$\left(\frac{\Delta V}{V}\right)_{0\text{ exp.}} \simeq -3.00 \times 10^{-3}.$$

We can compare this value with that obtained from the theoretical expression (7) using the variation of T_c with the pressure dT_c/dP [15] :

$$\left(\frac{\Delta V}{V}\right)_{0\text{ calc.}} \simeq -2.95 \times 10^{-3} \text{ (table I).}$$

Both values agree in magnitude and sign.

Table I. — Comparison of the calculated volume anomaly at 0 K $(\Delta V/V)_{0\text{ calc.}}$ obtained from the theoretical expression (deduced from the molecular field theory) using the variation of T_c with the pressure dT_c/dP with the experimental volume anomaly at 0 K $(\Delta V/V)_{0\text{ exp.}}$ obtained after subtraction of the deformation at T_c .

	N 10^{22} cm^{-3}	S	$\left(\frac{dT_c}{dP}\right)$ 10^{-9} $\text{K. dynes}^{-1}.\text{cm}^2$	$\left(\frac{\Delta V}{V}\right)_{0\text{ calc.}}$ 10^{-3}	$\left(\frac{\Delta V}{V}\right)_{0\text{ exp.}}$ 10^{-3}
YIG	2.10	5/2	+ 1.20 [15]	- 2.95	- 3.00
EuO	2.94	7/2	+ 0.37 [13]	- 1.75	- 1.65 [11]
MnO	4.60	5/2	+ 0.30 [14]	- 2.05	- 2.25 [12]

Fig. 3. — Comparison of the volume anomaly temperature dependence with that of the product of the sublattice reduced magnetizations, for $\text{Y}_3\text{Fe}_5\text{O}_{12}$.

Figure 3 shows the experimental variation of $\frac{\Delta V}{V}(T)$ as a function of temperature for YIG. This temperature dependence is compared with the product $\sigma_a(T) \sigma_d(T)$ of the temperature dependences of the reduced magnetizations of the 2 sublattices a and d found by Anderson [9] on the basis of Néel's theory of ferrimagnetism : the temperature dependences are in good agreement.

4. Comparison with other materials : insulating ferromagnetic, antiferromagnetic. — Using the same method to obtain the volume anomaly corresponding to the exchange energy associated with the magnetic order state below T_c , we deduced the quantity $\Delta V/V$ from the experimental curves published for europium oxide EuO [11] (insulating ferromagnetic) and for manganese oxide MnO [12] (antiferromagnetic). When comparing these experimental curves with those corresponding to the thermal expansions of the lattice vibrations deduced from a Debye-type law [11, 12], and subtracting the deformation at T_c we find :

$$\begin{aligned} \left(\frac{\Delta V}{V}\right)_{0\text{ exp.}} &\simeq -1.65 \times 10^{-3} \text{ for EuO} \\ \left(\frac{\Delta V}{V}\right)_{0\text{ exp.}} &\simeq -2.25 \times 10^{-3} \text{ for MnO.} \end{aligned}$$

These experimental results were also compared with those obtained from the theoretical expression (5) which gives the volume anomaly as a function of the variation of T_c with the pressure dT_c/dP (table I). The results are in good agreement.

Moreover, when plotting $(\Delta V/V)_{\text{exp.}}/(\Delta V/V)_{0\text{ calc.}}$ versus T/T_c (Fig. 4) we obtain a reduced temperature-dependence curve for all the three magnetic structures (ferri-, antiferro- and ferromagnetic) with localized moments.

Fig. 4. — Reduced curves of volume anomaly versus temperature for ferrimagnetic, antiferromagnetic and ferromagnetic materials with localized magnetic moments.

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