ANOMALIES IN THE TEMPERATURE DEPENDENCE OF THE MODULUS OF ELASTICITY DURING THE SPONTANEOUS REORIENTATION OF SPINS IN RARE-EARTH ORTHOFERRITES

K. P. BELOV, A. M. KADOMTSEVA, S. A. MEDVEDEV, V. V. USKOV, and A. Ya. CHERVONENKIS

Moscow State University

Submitted April 30, 1969

Zh. Eksp. Teor. Fiz. 57, 1124–1127 (October, 1969)

The temperature dependence of Young's modulus of a thulium orthoferrite single crystal along the a, b, and c axes of the orthorhombic crystal was measured at temperatures between 300 and 4.2°K. An anomaly in the temperature dependence of Young's modulus was observed at temperatures at which reorientation of the antiferromagnetic vector occurs (80°–90°K). A thermodynamic explanation of the anomaly is proposed and the values of the magneto-elastic constants along the a, b, and c axes of the orthorhombic crystal are determined.

THE elastic and magneto-elastic properties of the rare-earth orthoferrites have so far practically not been investigated. We have investigated the temperature dependence of the elastic moduli of a single crystal of thulium orthoferrite along the a, b, and c axes of the orthorhombic crystal. The single crystals of thulium orthoferrite were grown by the method of no-crucible zone melting with optical heating.[1] A controlled partial pressure of oxygen was kept up in the crystallization chamber. X-ray analysis showed that the obtained crystals had a distorted perovskite structure with the space group Pbnm and lattice parameters

$$a = 5.249 \pm 0.001 \text{ Å}, \quad b = 5.571 \pm \pm 0.001 \text{ Å}, \quad c = 7.582 \pm 0.001 \text{ Å},$$

which is in good agreement with the data of[2]. No extraneous phases were observed by means of metallographic and x-ray analyses. Chemical analysis showed that the deviation from stoichiometry did not exceed one percent and the content of Fe^{2+} ions did not exceed 0.4 percent. The single crystal nature was checked by the Laue method. Young's modulus was measured by a compound vibrator at a frequency of ~150 kHz in the temperature range from room temperature down to 4.2°K.

It is known[3] that on lowering the temperature from room down to liquid nitrogen temperature one observes in thulium orthoferrite a reorientation of the spins accompanied by a transition of the weak ferromagnetic moment from the c to the a axis of the orthorhombic crystal.

The process of reorientation of the spins in thulium orthoferrite does not occur instantaneously but extends over a temperature interval of ~12° centered on ~90°K.[4,5] It should be noted that the values of the temperature at the center of the reorientation region differ in the literature by several degrees for single crystals grown by various methods; this is obviously connected with different content of Fe^{2+} ions. It was shown in[6,7] that in the region of the reorientation temperature of the orthoferrites the first anisotropy constant passes through zero and the effect of the second anisotropy constant which has a positive sign becomes appreciable. This leads to the situation that with the reorientation of the spins in the rare-earth orthoferrites there occur two second-order phase transitions at the temperatures T_1 and T_2 corresponding to the beginning and end of the reorientation process.[7,8]

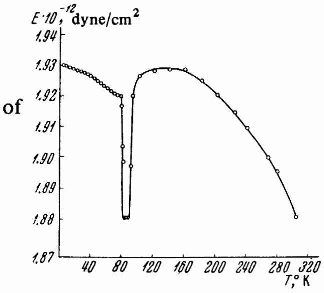

FIG. 1. Temperature dependence of of Young's modulus measured along the c axis of an orthorhombic crystal of thulium orthoferrite.

As our measurements have shown, in the range of temperatures $T_1 < T < T_2$ an anomaly is observed in the temperature dependence of Young's modulus measured along different crystal axes. In Fig. 1 we present the temperature dependence of Young's modulus measured with longitudinal vibrations along the c axis of a crystal of thulium orthoferrite. It is seen that an appreciable decrease of Young's modulus, which can be explained from thermodynamic considerations, is observed in the 80–92°K temperature range.

When an external stress is applied along the c axis of an orthorhombic crystal, the thermodynamic potential of the orthoferrite in the range of temperatures in which a reorientation of the spins in the (ac) plane is observed can be written as follows:

$$\Phi = \Phi_0 + k_1 \sin^2\theta + k_2 \sin^4\theta + L_z\xi_z \sin^2\theta + {}^1/_2 M_z\xi_z^2 \sin^2\theta + {}^1/_2 E_{0z}\xi_z^2 + \xi_z p_z. \tag{1}$$

Here Φ_0 is the part of the potential which does not depend on the orientation of the antiferromagnetic vector and on the strain ξ_Z, θ is the angle between the direction of the magnetic moment and the c axis of the crystal, k_1 and k_2 are the first and second magnetic anisotropy constants, L_Z and M_Z are the magneto-elastic energy constants, E_{0Z} is Young's modulus, p_Z is the external compressive stress, and ξ_Z is the relative strain along the c axis of the orthorhombic crystal.

The general expression for the thermodynamic potential[9] contains a whole series of terms linear and quadratic in the strains u_{ij}. However, for fixed $u_{zz} = \xi_z$ all u_{ij} are expressed in terms of ξ_z by means of the equilibrium condition so that the contribution from these terms can be considered to be included in the terms of formula (1). The equilibrium values of ξ_z and θ can then be found from the minimum condition of the thermodynamic potential:

$$\frac{\partial\Phi}{\partial\xi}=0, \quad \frac{\partial\Phi}{\partial\sin^2\theta}=0; \tag{2}$$

$$L_z\sin^2\theta + p_z + [E_{0z} + M_z\sin^2\theta]\xi_z = 0, \tag{3}$$

$$k_1 + 2k_2\sin^2\theta + L_z\xi_z + {}^1/_2 M_z\xi_z^2 = 0. \tag{4}$$

Substituting the value

$$\sin^2\theta = -\frac{k_1}{2k_2} - \frac{L_z\xi_z}{2k_2}, \tag{5}$$

found from (4) in (3) and ignoring terms quadratic in ξ_z, we obtain

$$\xi_z = \frac{L_z k_1}{E_z 2k_2} - p_z\left(E_{0z} - \frac{k_1 M_z}{2k_2} - \frac{L_z^2}{2k_2}\right)^{-1}. \tag{6}$$

At temperatures $T < T_1$ when $\sin\theta = 1$

$$E_z = E_{0z} + M_z; \tag{7}$$

for $T > T_2$ when $\sin\theta = 0$

$$E_z = E_{0z}. \tag{8}$$

In the temperature range $T_1 < T < T_2$ in which the spontaneous reorientation of the spins is observed Young's modulus decreases in accordance with (6) and becomes

$$E_z = E_{0z} - \frac{k_1 M_z}{2k_2} - \frac{L_z^2}{2k_2}. \tag{9}$$

Consequently one should observe at the temperatures T_1 and T_2 two jumps of Young's modulus corresponding to two second-order phase transitions.

According to (9) the jump of Young's modulus on the right at the temperature $T_2 = 92°K$ should be

$$\Delta E_z = L_z^2 / 2k_2, \tag{10}$$

since the first anisotropy constant k_1 vanishes at T_2.[7] The second anisotropy constant k_2 depends weakly on the temperature and in accordance with[5,6] its value can be assumed to be 30×10^3 erg/cm³. One can thus estimate from the magnitude of the jump of Young's modulus the value of the magneto-elastic constant L_z which turns out to be $(5.3 \pm 0.5) \times 10^7$ erg/cm³. On the other hand, according to (6), one can determine the magnitude of the magneto-elastic constant from the formula

$$L_z = \Delta\xi_z E_z. \tag{11}$$

The strain appearing with the reorientation of the spins $\Delta\xi_z$ determined from a measurement of the magnetostriction along the c axis[10] is 2×10^{-5}. The value of L_z obtained from (11) is $(4.0 \pm 0.5) \times 10^7$ erg/cm³, i.e., it is close to the value of L_z obtained from the jump of Young's modulus (10).

From the obtained agreement of the experimental data with the results of the thermodynamic treatment one can conclude that the Young's modulus anomaly in the temperature range in which reorientation occurs is due mainly to the rotation of the antiferromagnetic vector under the action of external elastic stresses. The magneto-elastic constants $L_{x,y,z}$ and the elastic moduli $E_{x,y,z}$ can be expressed in terms of the constants δ_i and λ_i, with the aid of which the magneto-elastic and elastic energy of the rare-earth orthoferrites was written in[11].

As is seen from Fig. 1, the magnitudes of the jump of Young's modulus on the left and on the right in the region of spin reorientation differ somewhat from one another. According to relations (7) and (8) one can determine the second magneto-elastic constant M_z in formula (1) from the difference in the jumps of Young's modulus at the temperatures T_1 and T_2; it is found to be $(6.0 \pm 0.6) \times 10^9$ erg/cm³.

We have also observed an anomaly in the temperature dependence of Young's modulus accompanying the reorientation of spins along the a and b axes of an orthorhombic crystal of thulium orthoferrite (Fig. 2). From the magnitude of the jump of Young's modulus at the temperature T_2 we determined the values of the first magneto-elastic constants along the a and b axes of the crystal: $L_x = (2.2 \pm 0.2) \times 10^7$ erg/cm³ and $L_y = (2.8 \pm 0.3) \times 10^7$ erg/cm³; from the difference in the jumps of Young's modulus at the temperatures T_1 and T_2 we determined the values of the second magneto-elastic constants: $M_x = -(3.0 \pm 0.3) \times 10^9$ erg/cm³ and $M_y = -(3.5 \pm 0.4) \times 10^9$ erg/cm³.

In conclusion we express our gratitude to R. Z. Levitin for participating in a discussion of the results and for a number of valuable remarks.

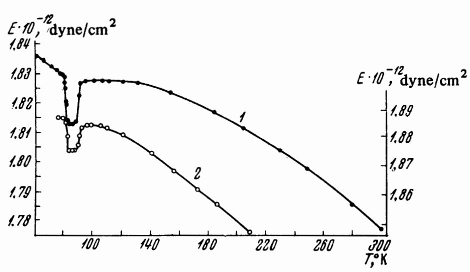

FIG. 2. Temperature dependence of Young's modulus measured along the a axis (curve 1, left-hand scale) and along the b axis (curve 2, right-hand scale) of an orthorhombic crystal of thulium orthoferrite.

[1] S. A. Medvedev, A. M. Balbashev, and A. Ya. Chervonenkis, Monokristally tugoplavkikh redkikh metallov (Single Crystals of Refractory Rare Metals), Nauka, 1969, p. 27.

[2] G. Will and O. Eberspächer, Z. Anorg, und Allgem. Chem. 356, 163 (1968).

[3] C. Kuroda, T. Miyadai, A. Naemura, N. Nuzeki, and H. Tokata, Phys. Rev. 122, 446 (1961).

[4] E. M. Gyorgy and J. P. Remeika, J. Appl. Phys. 39, 1369 (1968).

[5] J. R. Shane, Phys. Rev. Letters 20, 728 (1968).

[6] K. P. Belov, A. M. Kadomtseva, R. Z. Levitin, V. A. Timofeeva, V. V. Uskov, and V. A. Khokhlov, Zh. Eksp. Teor. Fiz. 55, 2151 (1968) [Sov. Phys.-JETP

28, 1171 (1969)].

[7]K. P. Belov, R. A. Volkov, B. P. Goranskiĭ, A. M. Kadomtseva, and V. V. Uskov, Fiz. Tverd. Tela 11, 1148 (1969) [Sov. Phys.-Solid State 11, in press].

[8]H. Horner and C. M. Varma, Phys. Rev. Letters 20, 845 (1968).

[9]L. D. Landau and E. M. Lifshitz, Teoriya uprugosti (Theory of Elasticity), Nauka, 1965, p. 54 [Addison-Wesley].

[10]K. P. Belov, A. M. Kadomtseva, T. L. Ovchinnikova, and V. V. Uskov, ZhETF Pis. Red. 4, 252 (1966) [JETP Lett. 4, 170 (1966)].

[11]A. S. Pakhomov, Fiz. Metal. i Metalloved. 25, 595 (1968).

Translated by Z. Barnea
131