MAGNETOSTRICTION CONSTANTS OF SINGLE-CRYSTAL LITHIUM–GALLIUM FERRITES

G. A. PETRAKOVSKIĬ and É. M. SMOKOTIN

Physics Institute, Siberian Division, USSR Academy of Sciences

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We measured the temperature dependences of the magnetostriction constants of single-crystal lithium–gallium ferrites. The measurements were made by the ferromagnetic-resonance method at a frequency of 9000 MHz. The temperature compensation points were determined for the constant λ_{111} of the ferrites $Li_{0.5}Fe_{2.5-x}Ga_xO_4$ with x = 0.06, 0.16, and 0.43. The results are interpreted on the basis of the single-ion magnetostriction theory.

A distinguishing feature of lithium–gallium ferrites is their relatively high magnetization at a considerable Curie temperature T_C, and the small width of the ferromagnetic-resonance line. From this point of view, these ferrites are of practical interest as material for special-purpose microwave ferrite devices. On the other hand, they are convenient for the investigation of the nature of magnetism of ferrites: they contain only one sort of magnetic ions and are characterized by an ordered arrangement of the cations of the octahedral sublattice. Of no little importance is also the fact that their crystal-magnetic and crystal-chemical structures have been investigated in relatively great detail.

In the present paper we present the results of an experimental investigation of the magnetostriction of lithium–gallium ferrites and present their analysis on the basis of the single-ion theory.

SAMPLES AND EXPERIMENTAL PROCEDURE

The magnetostriction constants λ_{100} and λ_{111} were measured in the ordered single crystals

$$Li_{0.5}Fe_{2.5-x}Ga_xO_4 \qquad (1)$$

with x = 0, 0.06, 0.16, 0.43, 0.65, and 1.18 by the ferromagnetic resonance method[1] at 9000 MHz in the temperature range 77–600°K. Spheres with approximate diameter 1.2 mm were used.

It can be shown, using the method described in [2], that the change of the field of the ferromagnetic resonance for a spherical anisotropic ferromagnetic sample subjected to a homogeneous uniaxial compression σ along the [110] axis and magnetized by a field **H** in the (110) plane is given by the relation

$$\delta H(\theta, \sigma, k_1) = \frac{3\lambda_{100}\sigma}{2M} - \frac{9\sigma}{4M}(\lambda_{100} - \lambda_{111})\sin^2(\theta + \delta) + \frac{3\sigma}{2M}\left\{(\lambda_{100} - \lambda_{111})[1 + \cos^2(\theta + \delta)] - 2\lambda_{100}\right\} \times \left\{\frac{4H_0}{H_a[{}^9/_8\sin^2 2(\theta + \delta) - {}^3/_2\sin^2(\theta + \delta)]} + \frac{H_a[{}^9/_8\sin^2 2(\theta + \delta) - {}^3/_2\sin^2(\theta + \delta)]}{2H_0}\right\}^{-1} \qquad (2)$$

where $H_a = 2k_1/M$, $H_0 = \omega/|\gamma|$, θ—angle between the magnetizing field **H** and the [100] direction, and

$$\delta = -\frac{H_a \sin 2\theta[1 - {}^3/_2\sin^2\theta]}{2H_0 + H_a[2 - \sin^2\theta - 3\sin^2 2\theta]}.$$

It follows from this relation that the shift of the field when the stress σ is applied should have an angular dependence as shown in Fig. 1, and should depend linearly

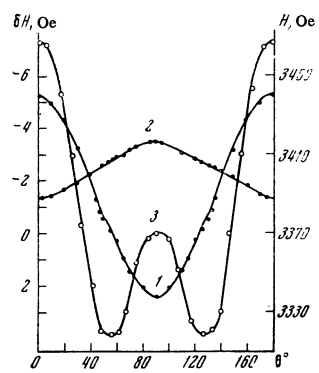

FIG. 1. Angular dependencies: 1 – of the shift of the resonance field $\delta H(\theta, k_1)$ for single-crystal ferrites $Li_{0.5}Fe_{2.07}Ga_{0.43}O_4$ (at $\sigma = 60 \times 10^6$ dyne/cm²); 2 – of the shift of the resonance field $\delta H(\theta, k_1)$ for yttrium ferrites (at $\sigma = 87 \times 10^6$ dyne/cm²); 3 – of the resonant field $H(\theta)$ for yttrium ferrites.

on the magnitude of σ within the limits of the elastic deformation. The points on the same figure represent the dependence $\delta H(\theta)$, obtained experimentally on single-crystal ferrites (1) with x = 0.43, for which H_a = −250 Oe. For comparison, we present also an analogous dependence, measured with single-crystal yttrium ferrite together with the $H(\theta)$ dependence. The agreement between the calculated and experimental curves is good. A check shows that δH is linear in σ. All this gives grounds for assuming that the apparatus and procedure used by us are sufficiently reliable.

EXPERIMENTAL RESULTS AND THEIR ANALYSIS

The points of Fig. 2 show the measured temperature dependence of the magnetostriction constants. At 77°K the constant $|\lambda_{100}|$ increases first with increasing gallium content, reaches a maximum at $x \approx 0.2$, and then decreases with further increase of x. The constant λ_{111} decreases at 77°K monotonically with increasing x. It

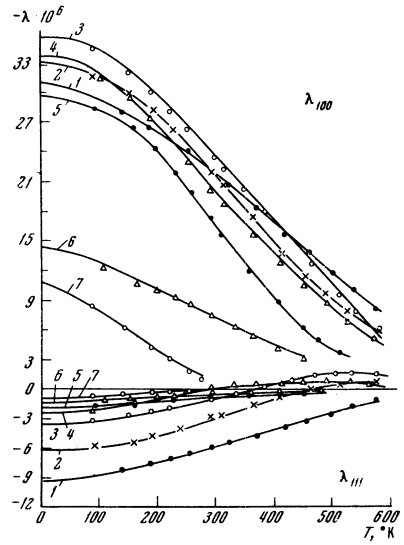

FIG. 2. Temperature dependence of the magnetostriction constants λ_{100} and λ_{111} of single-crystal ferrites $Li_{0.5}Fe_{2.5-x}Ga_xO_4$: 1 – x = 0; 2 – x = 0.06; 3 – x = 0.16; 4 – x = 0.43; 5 – x = 0.65: 6 – x = 0.9; 7 – x = 1.18

is interesting that λ_{111} has a temperature compensation point already at x = 0.06. The compensation temperature decreases with increasing gallium contents and reaches room temperature for a composition near x = 0.4.

We have interpreted the temperature dependences of the magnetostriction constants on the basis of Callen's single-ion theory.[3] It was assumed in the calculations that the elastic constants of lithium–gallium ferrites do not depend on the temperature and on the gallium constant. It was also assumed that the constants were equal to those of lithium ferrites $Li_{0.5}Fe_{2.5}O_4$.[4] The theoretical relations were fitted to the experimental data by least squares using an electronic computer. The necessary temperature dependences of the sublattice magnetization were calculated by the Prince method[5] from the known temperature dependences of the total magnetization. The latter were measured by a static method in a field of 12 000 Oe. The results of the matching are shown in Fig. 2 by solid lines. The agreement between experiment and theory is good. This reduction of the experimental data shows that the constant λ_{100} is determined by the predominant influence of the octahedral sublattice. Therefore, when the Fe^{3+} are replaced by Ga^{3+} ions, the constant λ_{100} at first increases. This growth, however, is hindered by the partial distribution of the gallium ions in the octahedral site and by the rapid decrease of the number of active bonds of the Fe^{3+} ions of the B-sublattice. The latter causes an appreciable portion of the Fe^{3+} ions of the B positions to become inactive with increasing x, i.e., they do not take part in the ferromagnetism.[6] Therefore at $x \approx 0.2$ $|\lambda_{100}|$ reaches a maximum and then decreases. The sound of the constant λ_{111} is also determined by the predominant influence of the B-sublattice.

It is of interest to predict the possible compositions with zero values of $\lambda_S = (2\lambda_{100} + 3\lambda_{111})/5$, λ_{100} or $2B_2 + B_1$ (or $\sim 2\lambda_{111} + \lambda_{100} = 0$). All these three conditions, in principle, can be satisfied when the Fe^{3+} ions of the B positions are replaced by nonmagnetic ions. Such may be Al^{3+}, Ti^{4+}, and In^{3+}. To the contrary, the maximum value of the indicated quantities are reached when diamagnetic ions are introduced in the tetrahedral sites. From this point of view, the lithium–gallium and lithium–zinc ferrite systems are promising for applications in devices using magnetoelastic interactions.

Thus, we have found good agreement between the experimental temperature dependences of λ_{100} and λ_{111} and Callen's single-ion theory when a suitable choice is made of the magnetoelastic-coupling coefficients (see the table). We can attempt to calculate these coefficients

Coefficients of magnetoelastic coupling of tetrahedral (A) and octahedral (B) sublattices for the ferrites $Li_{0.5}Fe_{2.5-x}Ga_xO_4$ [erg-cm^{-3}]

Gallium content, x	$B^{\gamma}_{0,2}(A)\cdot10^{-6}$	$B^{\gamma}_{0,2}(B)\cdot10^{-6}$	$B^{\varepsilon}_{0,2}(A)\cdot10^{-6}$	$B^{\varepsilon}_{0,2}(B)\cdot10^{-6}$
0	59,0	—88,7	—53,0	70,5
0,06	61,0	—95,0	—20,0	25
0,16	42,0	—80	—11,8	14,7
0,43	14,2	—47,0	—6,1	8,2
0,9	2,16	—17,0	—0,90	2,3
1,18	3,6	—14	—0,91	1,38

on the basis of the microscopic theory of magnetostriction.[7,8] This raises difficulties as a result of the incomplete understanding of the role of different mechanisms in the magnetostriction of S-ferrites. Thus, Tsuya[7] regards the predominant mechanism to be the magnetic dipole–dipole interaction, whereas an analysis by Kanamori,[8] and particularly experimental investigations, offer evidence more readily favoring the single-ion origin of the magnetostriction of the S-ferrite. A convincing proof of this premise is contained in the papers of Phillips and White.[9,10]

Our results agree also qualitatively with the single-ion model and diverge strongly from the dipole–dipole interaction model. Indeed, for example from the point of view of the magnetostriction theory based on the dipole–dipole model,[7] both constants should vanish simultaneously. Moreover, the very presence of a compensation point of λ_{111} of the ferrites (1) cannot be explained from the point of view of the dipole–dipole model of magnetostriction. There are also many other investigations in which it is shown that the dipole–dipole model encounters unsurmountable difficulties. All this justifies to some extent the attempt to ascribe an appreciable part of the magnetostriction of S-ferrites to the single-ion magnetoelastic coupling.

The single-ion magnetostriction results from an individual interaction of the spin of the paramagnetic ion with the field of the deformations of the crystal. This interaction leads to a change of the energy spectrum of the ion as a result of the distortion of the electric crystal field upon deformation of the lattice. The spin Hamiltonian for the S-ion at $S > 1/2$, with allowance for the magnetoelastic interaction, can be written in the form

$$\mathcal{H} = g\beta \mathbf{H}\mathbf{S} + \sum_{i,j=1,2,3} S_i D_{ij} S_j + \mathcal{H}'', \qquad (3)$$

where i, j = 1, 2, 3 are the coordinate axes (x, y, z) connected with the local symmetry of the position of the paramagnetic ion, H is the effective molecular field connected with the exchange interaction, $\mathcal{H}''$ is the part of the Hamiltonian which determines the splitting of the levels of the S-ion in a zero magnetic field also in the

absence of deformations, and D_{ij} is the tensor of the spin-phonon interaction, the form of which is determined by the point symmetry of the position of the paramagnetic ion.[11, 12]

For spinel ferrites

$$Fe_{\mu}^{3+}M_{1-\mu}[Fe_{\lambda}^{3+}M_{2-\lambda}]O_4 \quad (4)$$

it can be shown, starting from the Hamiltonian (3), that the magnetostriction constants have at 0°K the values

$$\lambda_{100} = -\frac{4S^2}{a^3}\left[3\mu C_{11}^{A} + \frac{2}{3}\lambda(C_{11}^{B} - C_{12}^{B} - \sqrt{2}C_{16}^{B} - 2\sqrt{2}C_{54}^{B} + 2C_{55}^{B})\right],$$
$$\lambda_{111} = -\frac{8S^2}{a^3}\left[\mu C_{44}^{A} + \frac{1}{9}\lambda(7C_{11}^{B} - C_{12}^{B} + 3C_{33}^{B} + 2\sqrt{2}C_{16}^{B} + 4\sqrt{2}C_{54}^{B} + 2C_{55}^{B})\right]. \quad (5)$$

Here M is the diamagnetic ion, C_{ij} the coefficients of the tensor of the Hamiltonian of the spin-phonon interaction,[11, 13] and a the crystal-lattice constant. The superior index of the coefficients C_{ij} designates the corresponding position of the spinel. The coefficients C_{ij} can be determined from the deformation dependences of the EPR spectra of the Fe^{3+} ions in diamagnetic crystals that are isomorphic to those investigated, and the coefficients of the magnetoelastic coupling can then be estimated. In view of the absence of data on the deformation dependence of the EPR spectra of the iron-group ions in spinels, we used the results obtained by Feher[13] for the crystal MgO. For the Fe^{3+} ion the coefficients C_{ij} are equal to: $C_{11} = 26 \times 10^{-13}$ and $C_{44} = -5.5 \times 10^{-18}$ (cm/dyne). Recognizing that the octahedral cell of MgO is close in its dimensions to the octahedral cell of $Li_{0.5}Fe_{2.5}O_4$ we can employ these for a rough estimate of magnetoelastic energy of the octahedral sublattice of the lithium ferrite. We ignore the trigonal component of the crystal field of the B-position of spinel. After simple calculations, we obtain

$$B_1(B) = -\frac{15}{2\sqrt{4\pi}}B_{0,2}^{\gamma}(B) = +200 \cdot 10^6 \text{ erg-cm}^{-3}$$
$$B_2(B) = -\sqrt{\frac{15}{4\pi}}B_{0,2}^{\varepsilon}(B) = -20 \cdot 10^6 \text{ erg-cm}^{-3}$$

Our experimental data yield

$$B_1(B) = +190 \cdot 10^6 \text{ erg-cm}^{-3} \quad B_2(B) = -75 \cdot 10^{-6} \text{ erg-cm}^{-3}$$

Consequently, the single-ion magnetoelastic energy gives the correct order of magnitude and the correct signs of the magnetoelastic coefficients.

Thus, the introduction of diamagnetic ions of gallium in the lithium ferrite leads to an increase of the constant λ_{100} and to the appearance of the compensation point of λ_{111}. The obtained temperature dependences are in agreement with Callen's single-ion theory.[3] Moreover, a calculation of the coefficients of the magnetoelastic coupling for the octahedral sublattice of the lithium ferrite, made by us on the basis of the single-pion theory and the spin-phonon coupling coefficients C_{ij} measured by Feher,[13] gives values that agree reasonably with those obtained experimentally. All this gives grounds for hoping that the single-ion theory is capable of explaining the greater part of the magnetostriction of S-ferrites.

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