Magnetoelastic Effects and Magnetization in $LiDyF_4$ and $LiHoF_4$ Single Crystals

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Temperature and magnetic field dependences of the magnetization of $LiHoF_4$ and $LiDyF_4$ single crystals were measured with a dc-SQUID magnetometer MPSM-2 (Quantum Design) with the magnetic field applied along and perpendicular to the *c* axis. Experimental data are well reproduced by simulations based on the microscopic model of the crystal field and magnetoelastic interactions.

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1. Introduction

Double lithium-rare earth fluorides which crystallize in the tetragonal scheelite C_{4h}^{6} structure attract much interest as model objects in physics of dipolar magnets and quantum phase transitions [1]. The unit cell of LiRF$_4$ contains two magnetically equivalent lanthanide R^{3+} ions at sites with the S$_4$ point symmetry. LiDyF$_4$ is a dipolar antiferromagnet with Dy^{3+} magnetic moments normal to the crystal symmetry axis (T_N=0.62 K), LiHoF$_4$ is a dipolar Ising-like ferromagnet with T_c=1.53 K [1]. The main goal of the present study was to elucidate the role of magnetoelastic interactions in formation of the magnetization and the energy level pattern of LiRF$_4$ crystals in the external magnetic fields.

2. Experimental results and discussion

Single crystals of LiDyF$_4$ and LiHoF$_4$ were grown by Bridgeman-Stockbarger method. After X-Rays orientation they were shaped as spheres to acquire a definite demagnetizing factor. To prevent the samples from rotation in the strong magnetic field, they were fixed in Stycast 1266 epoxy resin. The temperature dependences of the magnetization of all single crystals in the temperature range of 2÷300 K and the dependences of the magnetization on the magnetic field in the interval 0÷5 T applied along and perpendicular to the *c* axis were measured with a dc-SQUID.

In the presence of an applied magnetic field **B** (below a direction of **B** relative to the [001] and [100] axes is specified by spherical coordinates θ and φ), we write the Hamiltonian of a single R^{3+} ion in the following form (the nuclear Zeeman energy is neglected):

$$H = H_{cf} - g_J \mu_B \mathbf{BJ} + A\mathbf{JI} + \sum_{\alpha\beta} V'_{\alpha\beta} e_{\alpha\beta} + \sum_{\alpha,s} V''_{\alpha}(s) w_{\alpha}(s)\,. \tag{1}$$

Here the first term is the crystal field Hamiltonian:

$$H_{cf} = \alpha B_2^0 O_2^0 + \beta (B_4^0 O_4^0 + B_4^4 O_4^4 + B_4^{-4} \Omega_4^4) + \gamma (B_6^0 O_6^0 + B_6^4 O_6^4 + B_6^{-4} \Omega_6^4) \tag{2}$$

determined in the crystallographic system of coordinates by the set of seven crystal field parameters B_p^k (O_p^k and Ω_p^k are the Stevens operators, α, β, γ are the reduced matrix elements). The second term in Eq. (1) is the electronic Zeeman energy (μ_B is the Bohr magneton, g_J is the Lande factor, **J** is the total angular momentum). The third term represents the magnetic hyperfine interaction, and the last two terms define linear interactions of rare earth ions with the homogeneous macro- and microdeformations, respectively, where **e** is the deformation tensor, and **w(s)** is the vector of the *s*-sublattice displacement. The electronic operators $V'_{\alpha\beta}$ and V''_{α}(s) can be presented, similar to the crystal field energy, through the linear combinations of Stevens operators with the parameters which have been calculated earlier in the framework of the exchange charge model [2,3].

Taking into account a linear coupling between the lattice macro- and micro-deformations and the equilibrium conditions for the coupled paramagnetic ions and the elastic lattice, we obtain the lattice macrodeformation induced by the magnetic field

$$\mathbf{e}(\mathbf{B}) = -\frac{n}{v_0}\mathbf{S}[\langle \mathbf{V} \rangle_B - \langle \mathbf{V} \rangle_0] \tag{3}$$

and the sublattice displacements, which define the internal magnetostriction,

$$\mathbf{w}(\mathbf{B}) = -\frac{n}{v_0}\mathbf{a}^{-1}[\langle \mathbf{V}'' \rangle_B - \langle \mathbf{V}'' \rangle_0]\,. \tag{4}$$

Here n = 2 is the number of rare earth ions in the unit cell, **S** is the compliance tensor of the lattice, v_0 is the volume of the unit cell, **a** is the dynamic matrix of the lattice at the Brillouin zone centre, and angular brackets $<\ldots>_B$, $<\ldots>_0$ indicate thermal averages over the eigenstates of the rare earth ion Hamiltonian (1) for **B**≠0 and **B**=0, respectively. Operators **V** in Eq.(3) equal to operators **V'** renormalized due to linear coupling of macro- and microdeformations.

To take into account magnetic dipole-dipole interactions between the rare earth ions, we use the mean field approximation. The local field $\mathbf{B}_{\mathrm{loc}} = \mathbf{B} + (\mathbf{Q} - N\mathbf{I})\,\mathbf{M}$ in the spherical sample (**Q** is the tensor of dipole lattice sums, N=4π/3, **M** is the magnetization, **I** is the unity matrix) is substituted for **B** in the Hamiltonian (1), and the expressions (3) and (4) are substituted for **e** and **w**, respectively. Thus we obtain the effective self-consistent single-ion Hamiltonian parametrically dependent on the magnetization. When studying magnetic properties of the system, it is enough to consider the matrix of this Hamiltonian in the subspace of states of the ground multiplet of a rare earth ion. The magnetic moment of an ion satisfies to the self-consistent equations

$$m_{\alpha} = \frac{\sum_i g_J \mu_B < i \,|\, J_{\alpha} \,|\, i > e^{-E_i(M)/kT}}{\sum_i e^{-E_i(M)/kT}}\,, \tag{5}$$

where $E_i(M)$ are the energy levels of a rare earth ion (eigenvalues of the Hamiltonian (1)), T – temperature, k is the

Boltzman constant. To obtain energies of sublevels of the ground multiplet and the magnetization $\mathbf{M} = n\mathbf{m}/v_0$, the following actions are performed: the matrix of the effective Hamiltonian with $\mathbf{M} = \mathbf{e} = \mathbf{w} = 0$ is diagonalized, and the macro- ($\mathbf{e}(\mathbf{B})$) and microdeformations ($\mathbf{w}(\mathbf{B})$), and the magnetic moment (5) are calculated. At the next step the obtained values of $\mathbf{M}$, $\mathbf{e}$, $\mathbf{w}$ are substituted into the Hamiltonian, and the procedure is repeated (up to five times) to get a steady solution.

The calculations are essentially simplified when making use of symmetry properties of a system. Really we worked with linear combinations of the deformation tensor and the sublattice displacements corresponding to irreducible representations A_g, B_g, E_g of the lattice factor group C_{4h}^6. In particular, the magnetic field directed along the crystal symmetry axis c brings about only totally symmetric A_g deformations, and the field in the basal plane induces only A_g and rhombic B_g deformations. The corresponding internal A_g and B_g deformations are described by three and five independent linear combinations of sublattice displacements, respectively. We used in calculations the low-temperature compliance constants of LiErF$_4$: $S(A_g11)=7.79\cdot10^{-12}$ m^2/H; $S(A_g12)=-2.5\cdot10^{-12}$ m^2/H; $S(A_g22)=3.26\cdot10^{-12}$ m^2/H; $S(B_g11)=63.2\cdot10^{-12}$ m^2/H; $S(B_g12)=24.2\cdot10^{-12}$ m^2/H; $S(B_g22)=29.4\cdot10^{-12}$ m^2/H; and the parameters of a rigid ion model of the lattice dynamics [1–3].

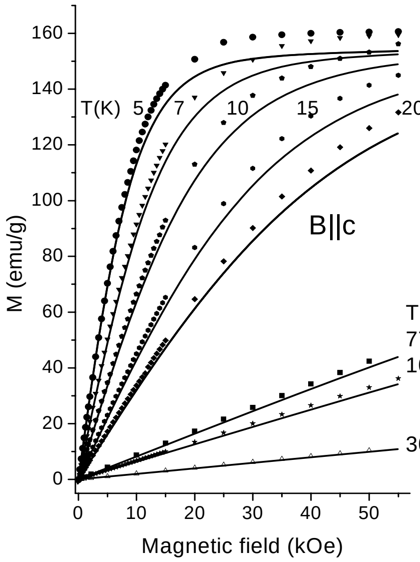

Fig. 1. Magnetic field dependences of LiHoF$_4$ magnetization. Solid lines are theoretical results, points are experimental data.

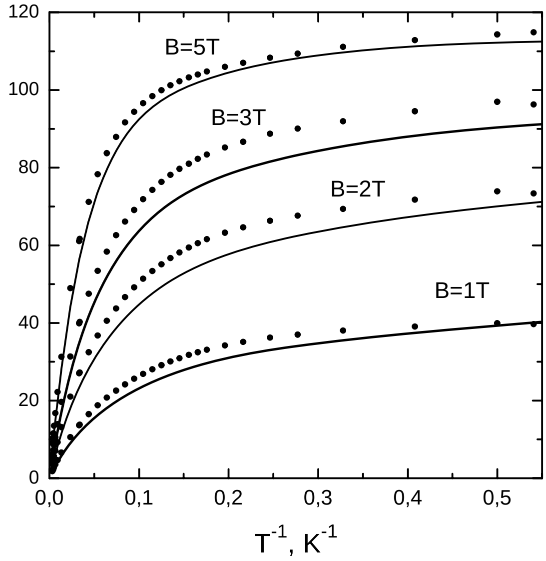

Fig. 2. Temperature dependences of LiHoF$_4$ magnetization. Solid lines are theoretical results (θ=83^0, φ= –15^0), points are experimental data.

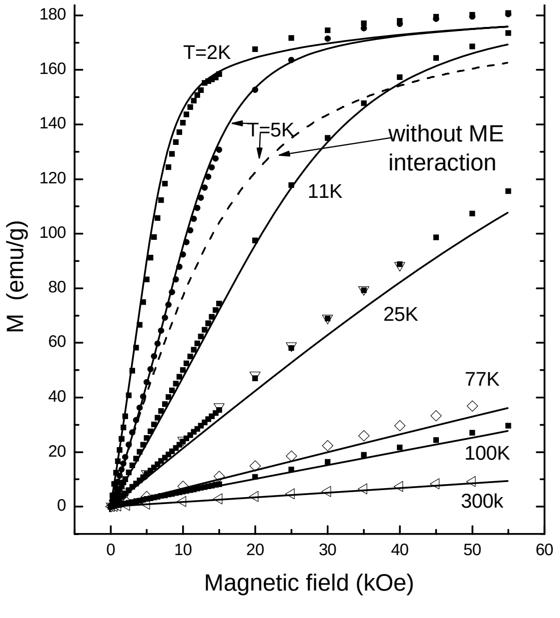

Fig. 3. Field dependences of LiDyF$_4$ magnetization. Solid lines are theoretical results (θ=90^0, φ=-5^0), points are experimental data.

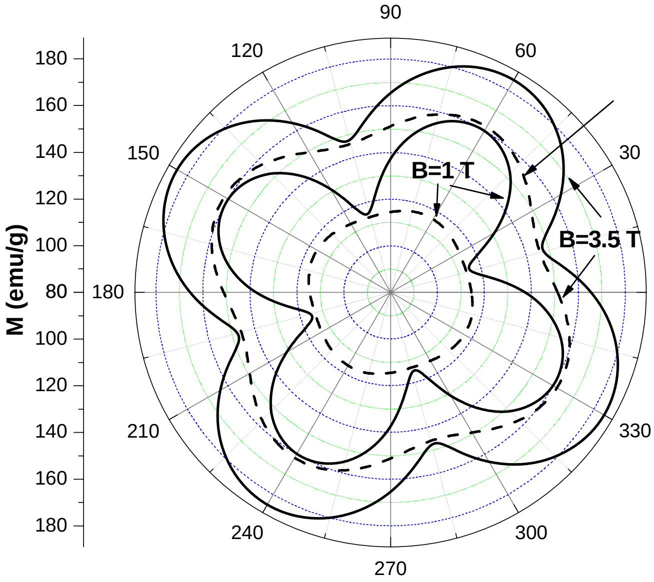

Fig. 4. Angular dependences of magnetization in the basal plane of LiDyF$_4$ (T=2 K). The dashed curves correspond to zero magneto-elastic coupling.

The measured temperature and field dependences of the magnetization in $LiHoF_4$ and $LiDyF_4$ single-crystals are compared with results of simulations in Figs.1-3. The calculated angular dependences of the magnetization in the basal plane of $LiDyF_4$ are shown in Fig. 4.

Mixing of the ground multiplet with the excited multiplets of a rare earth ion due to spin-orbit coupling was taken into account by making use of slightly renormalized matrix elements of Stevens operators and values of Lande factors g_J (1.22 and 1.3133 instead of 5/4 and 4/3 for pure $^5I_8$ and $^6H_{15/2}$ multiplets of Ho^{3+} and Dy^{3+}, respectively). From fitting the calculated dependences to the experimental data, small corrections for the published earlier crystal field parameters in diluted isomorphic crystals $LiYF_4$:Dy and $LiYF_4$:Ho were determined (Table 1).

Table 1. *Crystal field parameters B_p^k (cm^{-1}) for $LiHoF_4$ and $LiDyF_4$ single-crystals*

p k	Dy^{3+} ($4f^9$) $^6H_{15/2}$		Ho^{3+} ($4f^{10}$) $^5I_8$	
	This work	Ref. [4]	This work	Ref. [5]
2 0	170	165	219.7	190.35
4 0	-85	-88	-87.3	-78.25
6 0	-4.2	-4.4	-3.55	-3.25
4 4	-721	-980	-710	-657.2
4 -4	-661	0	-612	-568.6
6 4	-390	-427	-387	-364
6 -4	-248	-65	-253.7	-222.3

3. Conclusion

As it is seen in Figs.1-3, simulated temperature and magnetic field dependences of the magnetization are in good agreement with experimental results. It follows from calculations that magnetoelastic interactions in double lithium-rare earth fluorides bring about essential contributions to the magnetization in external magnetic fields at liquid helium temperatures. In particular, theory predicts a large anisotropy of the magnetization in the basal plane of $LiDyF_4$ (see Fig.4) at temperatures lower than 5 K in magnetic fields larger than 0.7 T.

A possible reason for some discrepancies between the calculated and experimental data is the neglect of dependences of compliance constants on temperature and magnetic field and of the interaction between the paramagnetic ions induced by the phonon exchange.

Results of this work are very important for the correct interpretation of the magnetization measurements in very large pulsed magnetic fields [6].

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