Deviant behaviour of magnetization of microsized powder of Ising dipolar antiferromagnet LiDyF$_4$ at temperatures $T > T_N$

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Introduction

Lithium–rare-earth double fluorides attract interest as model objects in physics of dipolar magnets [1]. Crystal symmetry of concentrated LiDyF$_4$ is $I4_1/a$, unit cell contains two magnetically equivalent Dy^{3+} ions at sites with the S_4 point symmetry [1, 2, 3]. Single crystal is Ising dipolar planar antiferromagnet, $T_N = 0.610(15)$ K, magnetic moments are ordered normally to *c* axis [4].

Methods and Materials

Micropowder of LiDyF$_4$ is prepared by sintering powders of LiF and DyF$_3$ taken in proportions according to phase diagrams from [2]. Initial batches, 0.121 g of LiF and 0.642 g of DyF$_3$ are dried for 2 hours at 150°C and $4 \cdot 10^{-3}$ Pa in vitreous carbon crucible. Sintering is performed in the same crucible for 18 hours at 600^{0}C in Ar atmosphere. X-ray powder diffraction and optical microscopy are used for characterization of the samples. Size of particles is about 1 μm according to images of sample taken via optical microscopy.Magnetization of the sample is measured in the range of applied magnetic fields 0–90 kOe and in the temperature range 2–300 K on vibration magnetometer *VSM* at *Physical Property Measurement System* (*PPMS*®).

Theoretical analysis

The Hamiltonian of rare earth ion in crystal lattice and external magnetic field **H** is diagonalized in the basis of the first 146 levels of the full space of states of $4f^9$ electronic configuration. Considered form of the one-ion Hamiltonian H is

$$H = H^0 + H_{\mathrm{Zee}} + H_{\mathrm{cf}} + H^{(\mathrm{P})}.$$

The first term is the free ion energy (the Hamiltonian H described in [5]), the second is the electron Zeeman energy

$$H_{\mathrm{Zee}} = \mu_{\mathrm{B}} \mathbf{H}_{\mathrm{loc}} (2\mathbf{S} + \mathbf{L}).$$

$\mathbf{H}_{\mathrm{loc}}$ is the local magnetic field that includes the applied field **H**, the molecular field $Q\mathbf{M}$ and the demagnetizing factor of sphere $N_{\mathrm{m}} = 1$ [6]:

$$\mathbf{H}_{\mathrm{loc}} = \mathbf{H} + \sum_{s`} \mathbf{M}(\mathrm{s`}) \left(Q(\mathrm{s},\mathrm{s`}) - \frac{4\pi}{3v} N_{\mathrm{m}} \right).$$

The third term is the crystal field on Dy^{3+} ion in S_4 point symmetry:

$$H_{\mathrm{cf}} = B_0^2 O_0^2 + B_0^4 O_0^4 + B_4^4 O_4^4 + B_{-4}^4 O_{-4}^4 + B_0^6 O_0^6 + B_4^6 O_4^6 + B_{-4}^6 O_{-4}^6,$$

where set of parameters $B^k{}_p$ is taken from [6].

The fourth term of Hamiltonian represents linear interaction of rare-earth ion with homogeneous macrodeformations $e_{\alpha\beta}$ [6]:

$$H^{(\mathrm{P})} = \sum_{\alpha\beta} V_{\alpha\beta} e_{\alpha\beta}, \text{ where } V_{\alpha\beta} = \sum_{pk} B_{p,\alpha\beta}^k O_p^k.$$

Magnetization M for powder of spherical particles, distributed equiprobably respecting the direction of applied field $\mathbf{H}$ is calculated as follows ($1/\beta = k_B T$):

$$M = \frac{1}{4\pi}\int_0^{2\pi} \mathrm{d}\varphi \int_0^{\pi} \mathrm{d}\theta \, \sin\theta \, \frac{\mu_B g_L}{m} \frac{\mathrm{Sp}\left[\mathbf{J}\frac{\mathbf{H}}{|\mathbf{H}|}\exp(-H\beta)\right]}{\mathrm{Sp}\left[\exp(-H\beta)\right]}.$$

Results and discussion

Experimental and calculated field dependencies of magnetization are presented at Fig. 1. Qualitative agreement between them appears at at $T > 10$ K. At $T = 5$ K measurements of magnetization have a shape of loop: measurements in increasing and decreasing field do not converge. Full hysteresis forms two loops connecting at zero (Fig. 2). Temperature dependence of the area of hysteresis is presented at Fig. 3. As it may be seen, effect of magnetization loops appear at $T < 7$ K and area of loops grows with descending temperature.

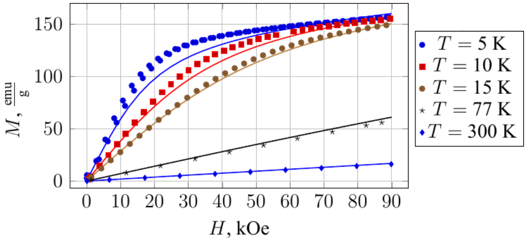

Figure 1. Field dependence of magnetization M at different temperature T
Symbols: experimental data. Lines: calculations

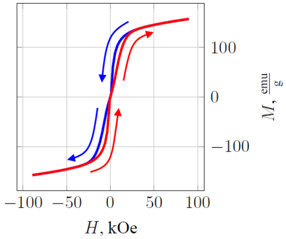

Figure 2. Full hysteresis at 2 K
Arrows represent direction of measurement

Magnetic longitudal relaxation time τ is measured by time-resolved investigation of magnetization. Applied field is changed from 0 kOe to 12 kOe or from 90 kOe to 12 kOe. Start of the measurements of magnetization is defined by the moment of establishment of the desired value of external field (12 kOe). Relaxation time τ is got via aproximation of time dependence by exponent function $M = A + B \cdot \exp(-t/\tau)$. Obtained values are about seconds and tens of seconds in the temperature range 2-7 K.

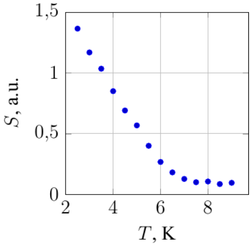

Figure 3. Temperature dependence of area S of hysteresis

Butterfly hysteresis and slow magnetic relaxation were already described in literature: for single-molecule magnets (SMM) [7] and for spin-phonon coupling [8]. Whether described case is related to one of these two or represents another phenomenon is a subject of following studies.

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