The magnetostriction of the $HoMnO_3$ hexagonal single crystals.

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The magnetostriction of $HoMnO_3$ hexagonal single crystals was investigated for a wide range of applied magnetic fields with strengths up to H = 14 T for all possible combinations of magnetic field orientation H and magnetostriction $\Delta L/L$. The anomalies found in the magnetostriction measurements of $HoMnO_3$ correlate well with the phase diagram of these compounds. For the first time the measurements of magnetostriction of $HoMnO_3$ single crystal were made in all five possible configurations.

Introduction.

Manganites $R MnO_3$ (R=Gd to Lu, Y, Sc) are multiferroics [1,2]. These materials can crystallize in two structural types: orthorhombic or hexagonal. Magnetic phase diagrams of hexagonal $RMnO_3$ are very complicated, as shown perfectly in [1-3], where it was concluded that these systems have at least two different magnetic orderings associated with 4f- and 3d- subsystems. Also it is well established by now that hexagonal manganites are antiferromagnetic as well as ferroelectric, with high ferroelectric Curie temperatures between 590 and 1000 K. The antiferromagnetic order emerges in low temperatures (below 100 K) and coexists with ferroelectricity.

Hexagonal $HoMnO_3$ is ferroelectric below 830 K and antiferromagnetic below $T_N \sim 76$ K. But the magnetic phase diagram of this material is more complicated [1]. As we can see in this diagram, the magnetic moments of manganese can have two possible positions: in the P63cm magnetic structure ($33\ K < T < T_N$ at zero fields, Mn-spins perpendicular to the hexagonal axis) and the P63cm configuration ($5.2\ K < T < 33$ K, Mn-spins parallel to the hexagonal axis). The magnetic structure of the intermediate (INT) phase is described by the $P6_3$ symmetry group; it has lower magnetic anisotropy [1,2]. Also it is possible that in the $P63cm$ symmetry only 1/3 of the Mn spins are aligned with the crystallographic hexagonal axis, whereas in the $P63cm$ phase the Mn spins are rotated in-plane by 90° [4-6].

But the role that Ho^{3+} ions play in the ordering of the $HoMnO_3$ single crystals remains unclear. In [7] it was proposed that the Ho^{3+} moments order noncollinearly in the hexagonal plane. Furthermore, neutron scattering experiments suggested an antiferromagnetic ordering along the hexagonal axis of a fraction of the Ho^{3+} moments below or close to the temperature of Mn spin reorentation [8,9].

The symmetry of the $HoMnO_3$ hexagonal single crystals forbids direct magnetoelectric coupling, so the magnetoelastic effect is the logical explanation for this coupling [10]. To obtain more data points, we measured

the magnetostriction effect with a large number of configurations of the direction of the strictional effect, for values of applied magnetic fields ranging up to 14 T, and for a wide range of temperatures.

Experiment.

High-quality hexagonal single crystals of $HoMnO_3$ were grown using the optical floating zone method with a four-mirror furnace FZ-4000 (Crystal System Corp). Magnetic measurements were made with a PPMS 6000 Quantum Design facility and a vibrating sample magnetometer [11]. The magnetostriction measurements were made in the Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw, Poland, using an experimental facility for magnetostriction measurements based on the Oxford 15 T superconducting solenoid [12].

Results and discussion.

Figure 1 shows the dependency of magnetic moment on temperature in applied magnetic field H = 1 kOe, magnetic field is parallel to c-axis. The first such point, around T = 4.7 K, corresponds to the first manganese moment reorientation, or Ho^{3+} ion ordering [2]. The second point at T = 37 K corresponds to the manganese ion reorientation from the P63cm to the P63cm configuration. The Neel temperature corresponding to manganese ions ordering at T = 76 K is not seen clearly due to paramagnetism of Ho^{3+} ions. Inset plots magnetic moment M versus applied magnetic field H in the $HoMnO_3$ single crystal for different temperatures and for the field applied along hexagonal c-axis. We can observe the "spin-flop-like" behavior. This behavior stops above 30 K, which is the temperature at which magnetic ordering of manganese ions transitions from P63cm to P63cm.

Fig. 1 Temperature dependence of magnetization in magnetic field H = 0.1 T along crystallographic c-axis. Inset: Magnetization versus the applied magnetic field. Temperatures depicted on the figure. Magnetic field was applied along the *c*-axis.

The transverse magnetostriction of $HoMnO_3$ single crystal is shown in figure 2. It is easy to observe the non-monotonic behavior of the magnetostriction graph for all three possible configurations of the direction of magnetic field/striction (see fig. 2). Striction reaches a sharp minimum for the temperature range T = 4.2 K ÷ 6 K, that is, close to the first "critical" point of the M(T) dependence (fig. 1), which, as mentioned above, corresponds to the first manganese moment reorientation, or Ho^{3+} ion ordering. The magnitude of magnetostriction in this configuration is about ~ 10^{-5}, which is not surprising for 4f- and 3d- elements. So neither manganese nor holmium can be responsible for this effect.

Fig. 2 Transversal magnetostriction of $HoMnO_3$ at different temperatures in various configurations. The conditions of measurements are depicted on the graph.

Figure 3 depicts the longitudinal effect of magnetostriction in the $HoMnO_3$ single crystal. The non-linear behavior of these graphs in strong applied magnetic fields was surprising. The first part of these graphs was perfectly described in [1, 2], but the behavior of magnetostriction in the area with the strongest fields is unexplained. It is easy to see that after a smooth peak the graphs fall at low temperatures in both directions along the axes of the sixth and the second order. At temperatures higher than T = 30 K the graph have no further anomalies. Most importantly, these graphs show that the non-monotonic behavior of magnetostriction remains confined to temperatures of about T = 30 K, about the same as the temperature of the manganese spin reorientation [1, 2].

Fig. 3 Longitudinal magnetostriction of $HoMnO_3$ at different temperatures in various configurations. The conditions of measurements are depicted on the graph.

Conclusions.

In conclusion, we can argue that Ho^{3+} ions and Mn^{3+} can be a major influence on the magnetostriction effects in $HoMnO_3$: these ions affect the coupling mechanism in hexagonal crystals $HoMnO_3$. Notably, the effects were observed in the area of the phase diagram where the Mn^{3+} ions are antiferromagnetically ordered. Further investigations must be carried out to develop a role of different ions on the magnetostriction effect [13].

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References.

[1] F. Yen, C. D. Cruz, B. Lorenz, E. Galstyan, Y. Y. Sun, J. Mater. Res. **22**, 2163 (2007).

[2] B. Lorenz, ISRN Condens. Matter Phys. 2013, 43 (2013).

[3] O. P. Vajk et all., Appl. Phys. **99**, 08E301 (2006).

[4] J. Vermette, S. Jandl, M. Orlita, M. M. Gospodinov, Phys. Rev. B **85,** 134445 (2012).

[5] M. Fiebig et all., J. Appl. Phys. **91**, 8867 (2002).

[6] B. Lorenz, A. P. Litvinchuk, M. M. Gospodinov, and C.W. Chu, PRL **92**, 087204 (2004).

[7] H. Sugie et all., J. Phys. Soc. Jpn. **71**, 1558 (2002).

[8] S. Nandi, et all., PRL **100**, 217201 (2008).

[9] A. Munoz et all., Chem. Mater. **13**, 1497 (2001).

[10] Seongsu Lee et all., Nature **451**, 805 (2008).

[11] A.D. Balaev, Yu.V. Boyarshinov, M.M. Karpenko, B.P. Khrustalev, Prib. Tekh. Eksp. **3**, 167 (1985).

[12] V.I. Nizhankovskii, Eur. Phys. J. B **71**, 55 (2009).

[13] We had therefore to measure the magnetostriction in single crystal samples which include another non-magnetic 4-f element to clarify the role of Ho-ion in magnetic properties of the hexagonal $HoMnO_3$.

Fig. 1

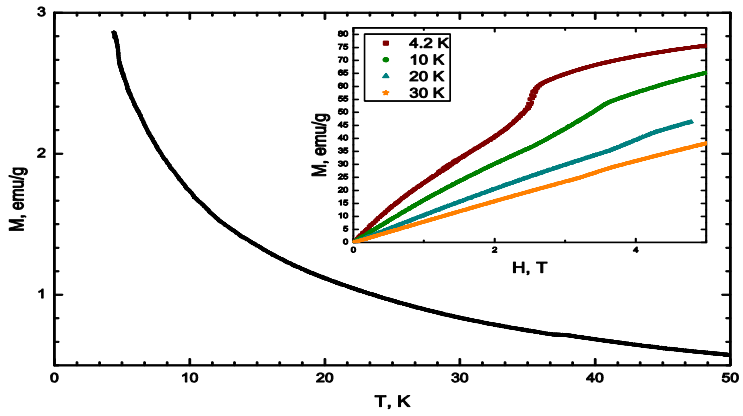
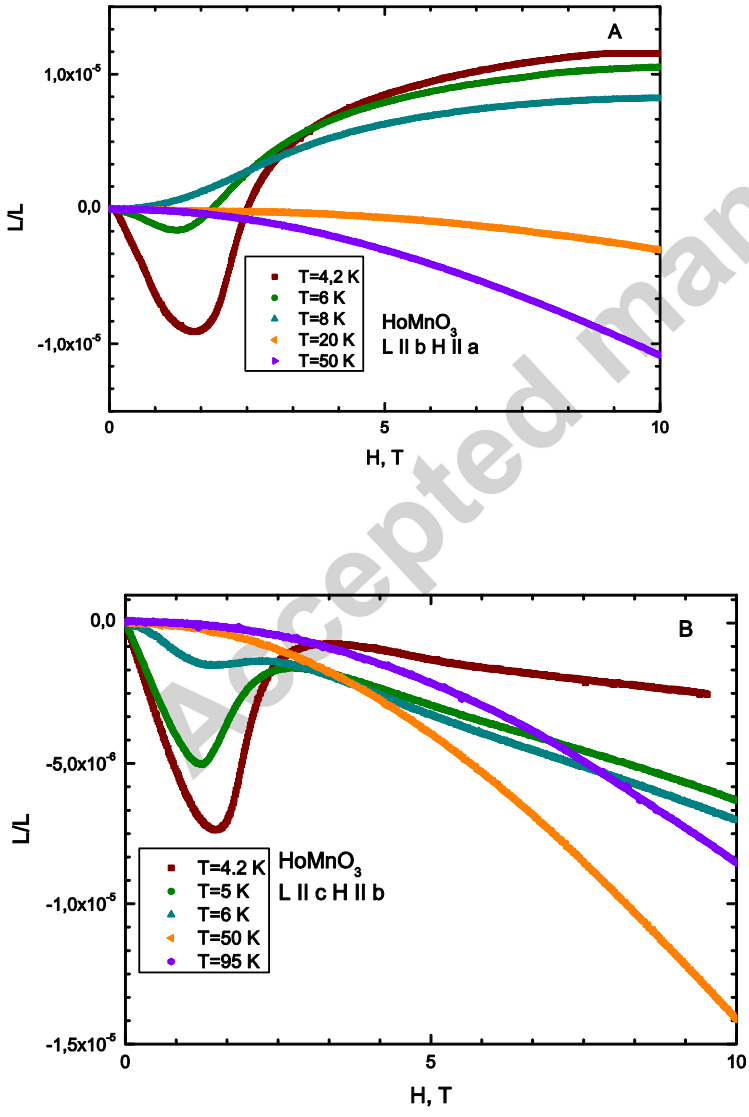


Fig. 2



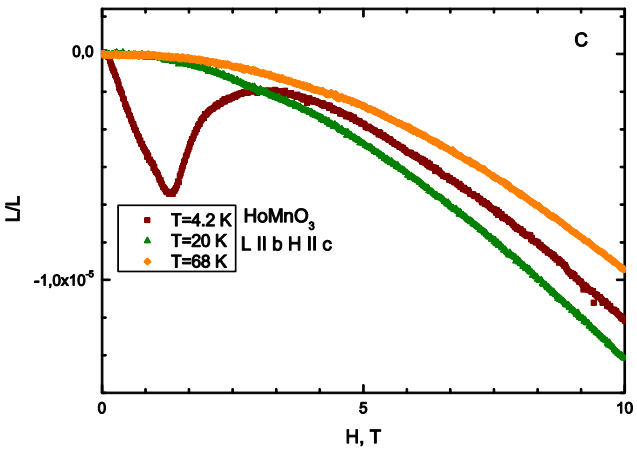


Fig. 3