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The effect of hydrogen on the magnetostriction of rare-earth compounds $Tb_xDy_{1-x}Fe_2$

S. A. Nikitin, I. S. Tereshina, A. P. Touliakov, and E. A. Tereshina

Department of Physics, Moscow State University, Vorobyevy Gory, 119899, Moscow, Russia
E-mail: nikitin@rem.phys.msu.su

V. N. Verbetsky and A. A. Salamova

Department of Chemistry, Moscow State University, Vorobyevy Gory, 119899, Moscow, Russia

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The crystal structure and the magnetic and magnetoelastic properties of $Tb_xDy_{1-x}Fe_2H_y$ ($x = 0.27$; 0.41; $y \le 3$) are investigated. The hydrides are obtained by the interaction of $Tb_xDy_{1-x}Fe_2$ samples with hydrogen gas at room temperature. It is established that the hydrides have the same cubic type of structure as the parent compounds but the lattice parameter in the hydrides is larger than in the hydrogen-free samples. Both the Curie temperature and the magnetic moment decrease drastically as the hydrogen concentration increases. Measurements of the longitudinal $\lambda_{||}$ and transverse magnetostriction $\lambda_{\perp}$ are carried out in magnetic fields up to 12 kOe in the temperature range 78–300 K using strain gauges.The thermal expansion coefficients as a function of temperature are obtained for hydrides with high hydrogen concentration. The magnetostriction of hydrides is strongly different from that of hydride-free samples. The small and negative value of the magnetostriction at low temperatures proves that hydrogen atoms make a strong contribution to the crystal field acting on the rare-earth ions.

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Introduction

Intermetallic compounds of rare earths (R) and 3*d*-transition metals having $MgCu_2$-type crystal structure are attracting much attention in recent years as giant magnetostrictive materials [1]. These compounds interact actively with hydrogen gas at room temperature and form stable hydrides. Upon hydrogenation these compounds exhibit substantial changes in their magnetic properties [2]. The magnetoelastic properties of $Tb_xDy_{1-x}Fe_2$ compounds have been investigated insufficiently up to now, because the samples are degraded in the process of hydrogen absorption. We have prepared powder-pressed samples for all of the compounds, including the host alloys, and have measured the magnetostriction of these compounds. This method was successfully applied by Clark et al. [3]. The present paper reports on the magnetic and magnetoelastic properties of $Tb_xDy_{1-x}Fe_2H_y$ ($x = 0.27$; 0.41) with different hydrogen concentrations ($y \le 3$).

Experimental methods

Samples of $Tb_{0.27}Dy_{0.73}Fe_2$ and $Tb_{0.41}Dy_{0.59}Fe_2$ were prepared by melting constituent metals of rare earth and Fe metals with purities of 99.95% and 99.98%, respectively, in an induction furnace, followed by annealing for 40 h at 1000°C. X-ray diffraction analysis carried out with a powder diffractometer using Cr K_α-radiation showed that all of the compounds studied were almost single-phase and had crystalline structures of the $MgCu_2$-type. The amount of extraneous phases in the samples was no more than 2%.

The hydrides were obtained by the interaction of the $Tb_xDy_{1-x}Fe_2$ samples with hydrogen gas at room temperature. The concentration of absorbed hydrogen in the samples was calculated using the van der Waals equation and additionally was measured by the full-burning method.

Thermomagnetic analysis (TMA) was used to measure the Curie temperature in a field of 1 kOe. The magnetic measurements were made for aligned and free powder samples in the temperature range

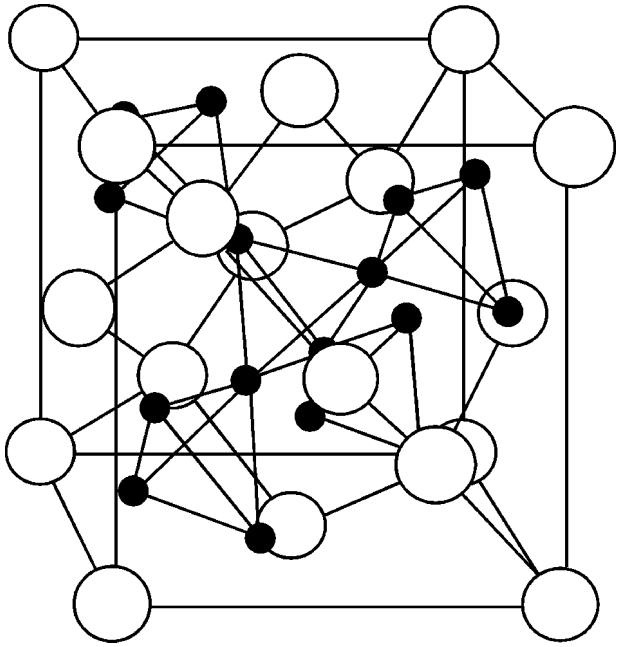

Fig. 1. Cubic Laves phase ($C15$). The large circles represent the rare-earth ions and the small circles represent 3d ions.

80–700 K and in magnetic fields up to 12 kOe, using a pendulum magnetometer.

The magnetostriction of $Tb_xDy_{1-x}Fe_2H_y$ was measured in fields up to 12 kOe in the temperature range 80–300 K by using a conventional strain-gauge bridge. Since the hydrides were available only in the powder form, the measurements were made with powder-pressed samples. The fine powder of $Tb_xDy_{1-x}Fe_2H_y$ compounds was compacted under pressure near 3 GPa in disks with dimensions of 0.8 mm×1.5 mm×8 mm. The densities of the samples produced in this way were nearly 70% of the powder x-ray densities. We took as the magnetostriction value for the investigated compounds the values measured on the pressed sample multiplied by the ratio of the powder x-ray density to the pressed sample density [3]. This method was checked on solidified precursor samples. Magnetostrictive measurements were made as a function of magnetic field strength and temperature.

Results and discussion

$Tb_xDy_{1-x}Fe_2$ ($x = 0.27;\ 0.41$) compounds were found to be crystallized in the cubic $MgCu_2$-type structure (Fig. 1). The lattice constant a and unit cell volume V are listed in Table. It was observed that hydrogenation leads to a lattice expansion of the compounds without change of the cubic structure from the $MgCu_2$ type. The relative cell volume

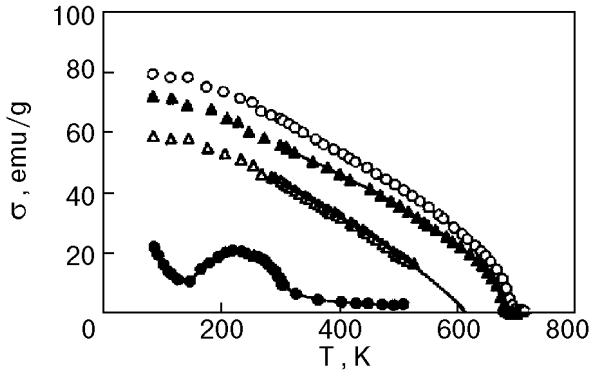

Fig. 2. The temperature dependence of magnetization σ of the $Tb_xDy_{1-x}Fe_2$ compounds measured at an external magnetic field of 1 kOe: $Tb_{0.41}Dy_{0.59}Fe_2$ (○); $Tb_{0.27}Dy_{0.73}Fe_2$ (▲); $Tb_{0.27}Dy_{0.73}Fe_2H_1$ (Δ); $Tb_{0.41}Dy_{0.59}Fe_2H_3$ (●).

increases $\Delta V/V$ show a consistent ~ 9% (at $x = 1$) and ~ 20% (at $x = 3$) volume expansion over the hydrogen-free unit cell for all the compounds.

Table

Crystallographic and magnetic data for the $Tb_xDy_{1-x}Fe_2$ compounds and their hydrides.

Compound	a, Å	V, Å^3	$\Delta V/V$, %	T_C, K
$Tb_{0.27}Dy_{0.73}Fe_2$	7.325	393.03	–	686
$Tb_{0.27}Dy_{0.73}Fe_2H_1$	7.537	428.15	8.9	596
$Tb_{0.41}Dy_{0.59}Fe_2$	7.324	392.87	–	697
$Tb_{0.41}Dy_{0.59}Fe_2H_3$	7.789	472.547	20.2	275

The Curie temperatures T_C of $Tb_xDy_{1-x}Fe_2$ compounds and their hydrides were determined using TMA, as was mentioned above (Fig. 2). In cases when T_C was close to room temperatures, it was obtained from a Belov–Arrott plot (for example, see Fig. 3). The Curie temperatures of $Tb_xDy_{1-x}Fe_2H_y$ are summarized in Table in comparison with their original counterparts.

Both the Curie temperature and the magnetization decrease with increasing hydrogen concentration. At $y = 3$ a compensation temperature appears in the compound. Such behavior has been observed earlier [7] and is typical for all RFe_2 compounds (where R is a heavy rare earth).

Figure 4,*a* shows the temperature dependence of the magnetization $\sigma(T)$ measured in an external magnetic field of 12 kOe. It can be seen from Fig. 4 that the σ-curve measured on heating of the sample is characterized by a single minimum at $T \approx 130$ K (compensation temperature) and a broad maximum at $T \approx 220$ K.

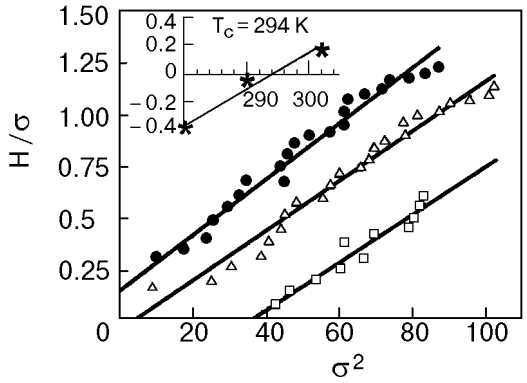

Fig. 3. The dependence of H/σ versus σ^2 for $Tb_{0.41}Dy_{0.53}Fe_2H_{2.8}$ at different T, K: 280 (❑); 290 (Δ), and 302 (●).

Figure 4,*b* shows the temperature dependence of longitudinal magnetostriction $\lambda_{\|}$ measured at different external magnetic fields. The maximum of the longitudinal magnetostriction $\lambda_{\|}$ for all the curves is observed near the $T_C \approx 270$ K, obtained by the TMA. Near the compensation temperature the $\lambda_{\|}$ longitudinal magnetostriction (Fig. 4,*b*) changes its sign and becomes negative at low temperatures. Anomalies at these temperatures can be also seen on the thermal expansion curve $\Delta l/l$ (see Fig. 4,*c*).

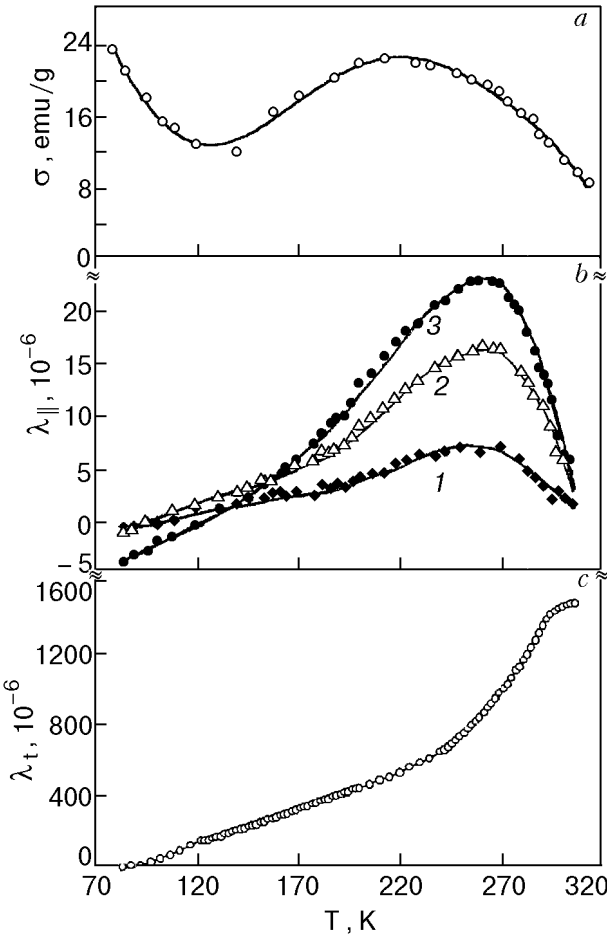

Fig. 4. The temperature dependence of magnetization σ of crystalline $Tb_{0.41}Dy_{0.59}Fe_2H_3$ samples measured at an external magnetic field of 12 kOe (*a*). The temperature dependence of the longitudinal magnetostriction $\lambda_{\|}$ measured at different values of external magnetic fields H, kOe: 2 (*1*), 10 (*2*), 12 (*3*) (*b*); Temperature dependence of the thermal expansion $\lambda_t = \Delta l/l$ (*c*).

From these results the following comments can be made: (i) hydrogenation leads to a simple volume expansion of the cubic lattice up to 20%; (ii) the Curie temperature, magnetic moment, and magnetostriction decrease drastically with increasing hydrogen concentration. As follows from Ref. 8, hydrogen in RFe_2 occupies different positions depending on concentration. Our measurements are evidence that hydrogen incorporation at high concentration ($y = 3$) strongly influences the crystal field acting on the rare-earth atoms.

Conclusions

Thus a comprehensive study of the structural, magnetic, and magnetoelastic properties of the $Tb_xDy_{1-x}Fe_2$ compounds ($x = 0.27$; 0.41) and their hydrides has been made. The interstitial atoms occupy sites adjacent to the rare-earth atoms, creating a change of crystal field that reflects the local symmetry and induces a significant change in the magnetic properties (Curie temperature, saturation magnetization, and magnetostriction).

Acknowledgments

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