MAGNETOSTRICTION OF RARE-EARTH METALS IN THE PARAMAGNETIC, ANTI-FERROMAGNETIC, AND FERROMAGNETIC RANGES

K. P. BELOV, R. Z. LEVITIN, and B. K. PONOMAREV

Moscow State University

Submitted to JETP editor July 15, 1965

J. Exptl. Theoret. Phys. (U.S.S.R. 49, 1733-1740 (December, 1965)

The magnetostriction of the rare-earth metals Tb, Dy, Ho, and Er (polycrystalline specimens) has been measured in pulsed magnetic fields up to 150 kOe, in the temperature interval 90 to 300°K. In all the metals, a magnetostriction large in absolute value (of order 100×10^{-6}) was observed in the paramagnetic range. In Ho, a magnetostriction due to the transition from the antiferromagnetic to the ferromagnetic state was observed. In Tb and Dy in the ferromagnetic state, the magnetostriction attains values of 3300×10^{-6} and 2200×10^{-6} respectively.

THE magnetostriction of weakly magnetic substances in pulsed magnetic fields was first observed by Kapitza.[1] He established that the magnetostriction of bismuth, antimony, and graphite varies quadratically with the field and reaches values of 10×10^{-6} to 30×10^{-6} in fields of order 280 kOe. It has been shown recently that antiferromagnetic substances characteristically possess a magnetostriction of considerable magnitude.[2,3] The magnetostriction of weakly magnetic substances is of great interest, since it is an immediate result of a manifestation of basic types of interaction connected with the magnetic state of the substances in question. In the present work it is shown that the magnetostriction in the paramagnetic range is especially large in the rare-earth metals Tb, Dy, Ho, and Er.

In rare-earth ferromagnets below the point of transition Θ_2 to the magnetically ordered state, the magnetostriction reaches values of order 10^{-3}.[4-8] Because of large anisotropy, however, it was not possible, in the fields used in [4-8], to attain the saturation magnetostriction in polycrystals; and in single crystals, the saturation magnetostriction was measured only along individual "easy" directions. In the present work, therefore, the magnetostriction of Tb, Dy, and Ho in fields up to 150 kOe was also measured below the magnetic-ordering temperature.

1. The measurement of the magnetostriction in pulsed magnetic fields in the temperature interval 90 to 300°K was carried out by means of a remote piezoelectric sensor (a description of the method of measurement will be given elsewhere). Temperatures of 90 to 300°K were produced by blowing nitrogen vapor through the bore of the solenoid; the stability of the temperature during the time of an experiment was ± 0.3°K, and the temperature gradient along the specimen did not exceed 2°K. The error in the determination of the relative value of the strain (in its dependence on the field or the temperature) was 3 to 5%; the absolute value of the strain was measured with an accuracy of 10 to 12%.

2. In the measurement of magnetic, magnetostrictive, and other properties in pulsed magnetic fields, it must be conceded that the magnetization processes occur adiabatically. From thermodynamic relations, the relation between the adiabatic and isothermal magnetostrictions, λ_a and λ_T, is[9]

$$\Delta\lambda_a = \lambda_a - \lambda_T = -\int_0^H \alpha_H \frac{T}{c_H}\left(\frac{\partial I}{\partial T}\right)_H dH. \qquad (1)$$

Here I is the magnetization, and α_H and c_H are, respectively, the coefficient of thermal expansion and the specific heat at constant field. The values of α_H and c_H depend on the field:

$$\alpha_H = \alpha_I - \left(\frac{\partial \lambda}{\partial H}\right)_T \left(\frac{\partial H}{\partial T}\right)_I, \qquad (2)$$

$$c_H = c_I - \left(\frac{\partial I}{\partial T}\right)_H \left(\frac{\partial H}{\partial T}\right)_I. \qquad (3)$$

Thus in order to find the value of λ_T, it is necessary to subtract the adiabatic increment $\Delta\lambda_a$. We mention in advance that in the metals we studied, taking account of the adiabatic increment $\Delta\lambda_a$ is important in the paramagnetic range and close to the transition point Θ_2; below this temperature,

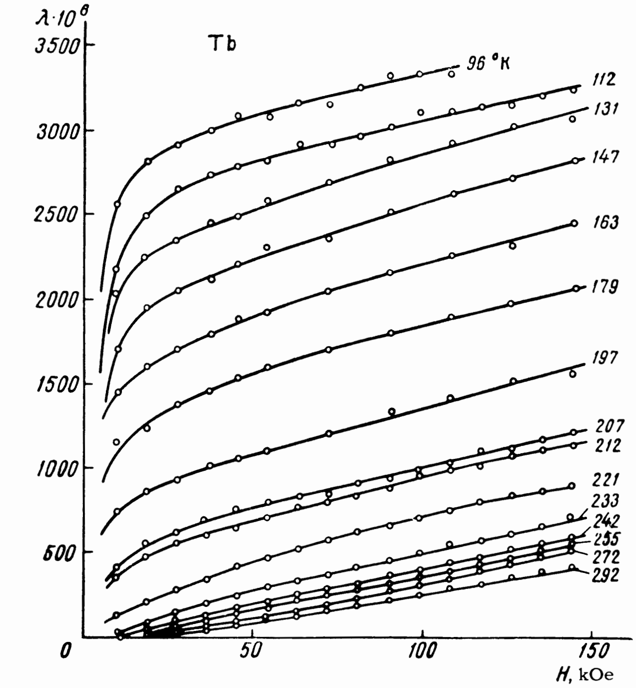

FIG. 1. Dependence of magnetostriction on field at constant temperature in Tb.

the adiabatic increment may be neglected, since in this case $\lambda_T \gg \Delta\lambda_a$.

3. Figures 1 to 4 present isotherms of the magnetostriction of Tb, Dy, Ho, and Er; Figs. 5 to 8 give the dependence of the magnetostriction of these substances on temperature at various fields.

The magnetostriction below Θ_2 was studied in three metals, Tb, Dy, and Ho. In the first two metals, the measurement of the magnetostriction was carried out only in fields above the critical field for transition from the helicoidal to the ferromagnetic state (H_{cr} = 0.2 kOe for Tb and 10 kOe for Dy). For these metals, the magnetostriction

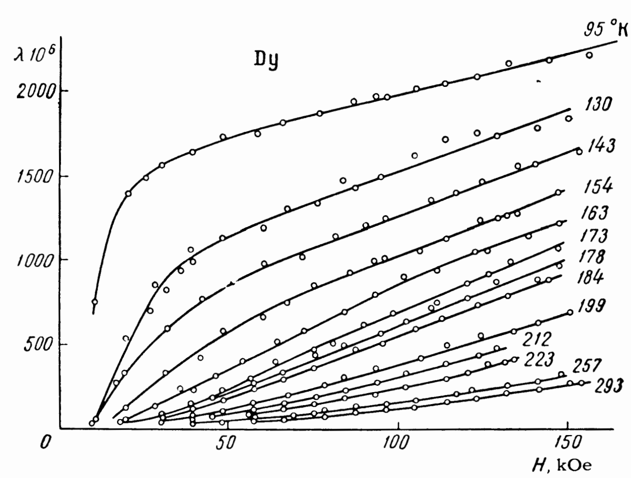

FIG. 2. Dependence of magnetostriction on field at constant temperature in Dy.

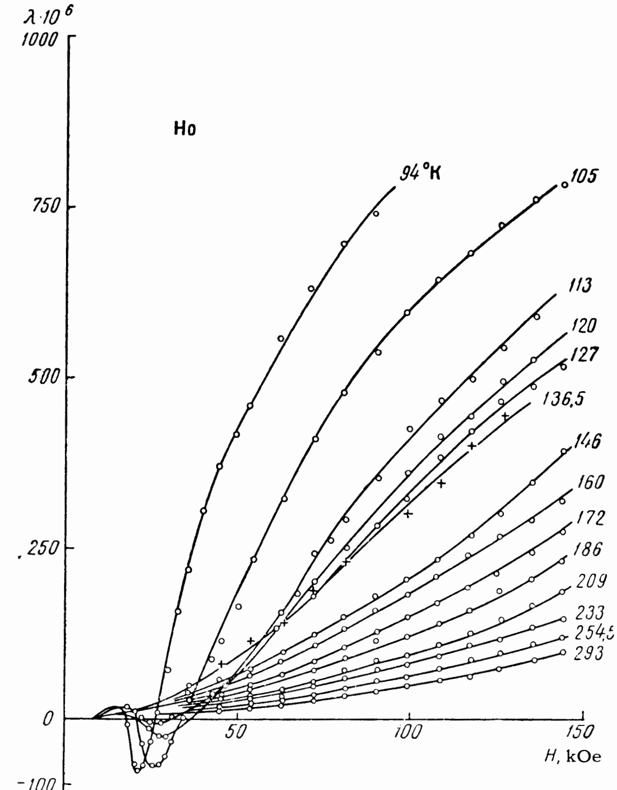

FIG. 3. Dependence of magnetostriction on field at constant temperature in Ho.

below the point Θ_2 in fields above 40 to 50 kOe can be described by the relation (cf. Figs. 1 and 2)

$$\lambda = \lambda_s + \chi_\lambda H, \qquad (4)$$

where $\chi_\lambda = d\lambda/dH$ is the slope of the straight line $\lambda(H)$ in strong fields. Although the measurements were carried out on polycrystals, it can be stated with assurance that the value of λ_s is related to

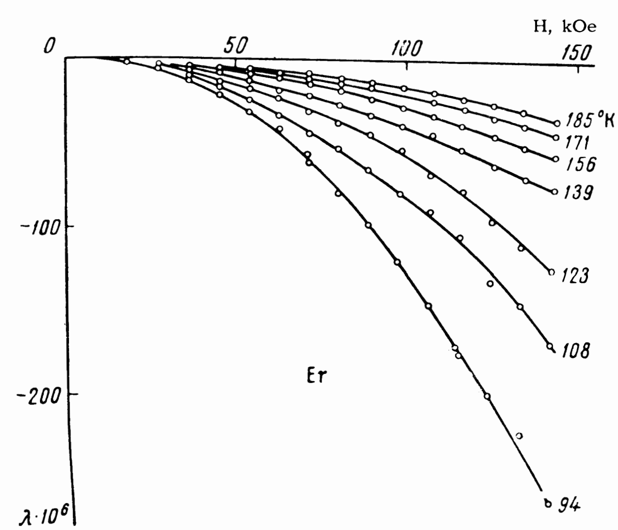

FIG. 4. Dependence of magnetostriction on field at constant temperature in Er.

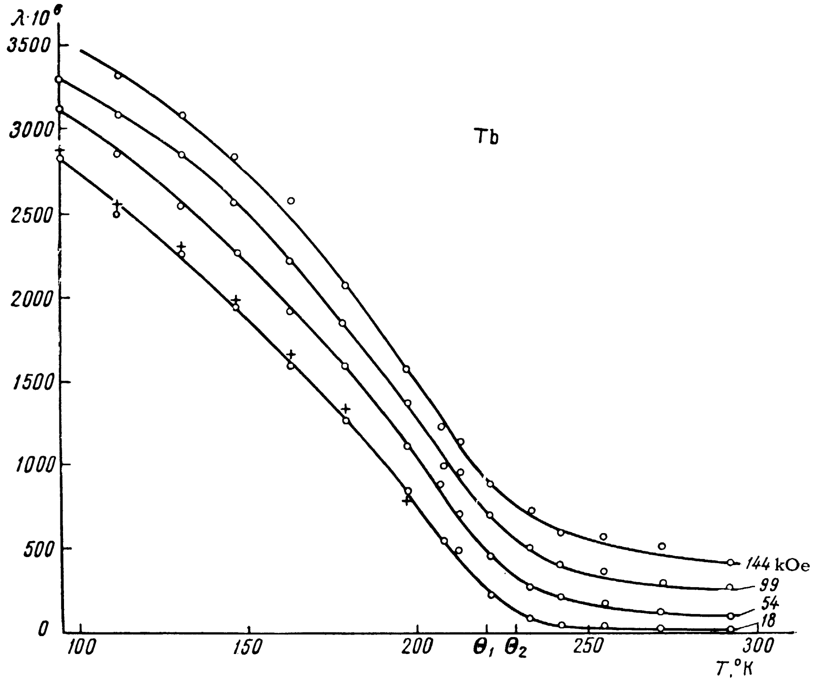

FIG. 5. Dependence of magnetostriction on temperature at constant field in Tb; +, dependence of $\lambda_s(T)$.

the magnetostriction in the basal plane; this is due to the fact that the uniaxial anisotropy is very large in Tb and Dy, so that in fields of order 10^5 Oe, saturation of the magnetostriction along the hexagonal axis of the crystal is not attained.[10] As for the value of χ_λ, it can be due to two causes: either the paraprocess magnetostriction, or the magnetostriction connected with rotation of the magnetic moments out of the basal plane toward the hexagonal axis of the crystal. To clear up this question it is necessary to carry out measurements of magnetostriction on single-crystal rare-earth ferromagnets in strong fields. The enormous value of λ_s attracts attention: at nitrogen temperatures it is 1500×10^{-6} for Dy and 2850×10^{-6} for Tb. This indicates that the magnetoelastic energy in the basal plane must exert an appreciable influence on the magnetic structure of Tb and Dy.

4. In holmium, the critical field for destruction of the helicoidal structure (in the neighborhood of $T = \Theta_2$) is equal to about 20 kOe. In this metal, we succeeded in observing the magnetostriction connected with this breakdown (cf. Figs. 3 and 7). In fields up to about 20 kOe the magnetostriction of Ho is positive, at the critical field it changes sign, and then it again becomes positive. This type of dependence of the magnetostriction on the field near the point Θ_2 was observed in Dy.[6] An explanation of this phenomenon was given in [11].

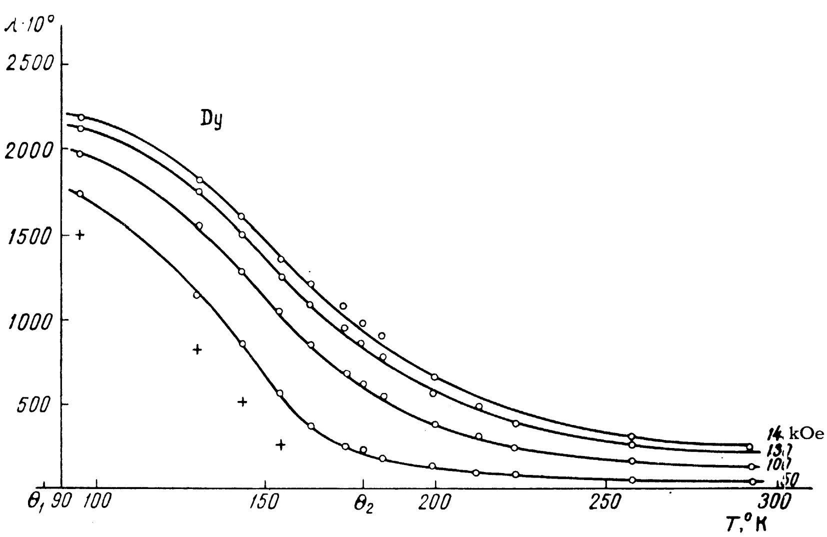

FIG. 6. Dependence of magnetostriction on temperature at constant field in Dy; +, dependence of $\lambda_s(T)$.

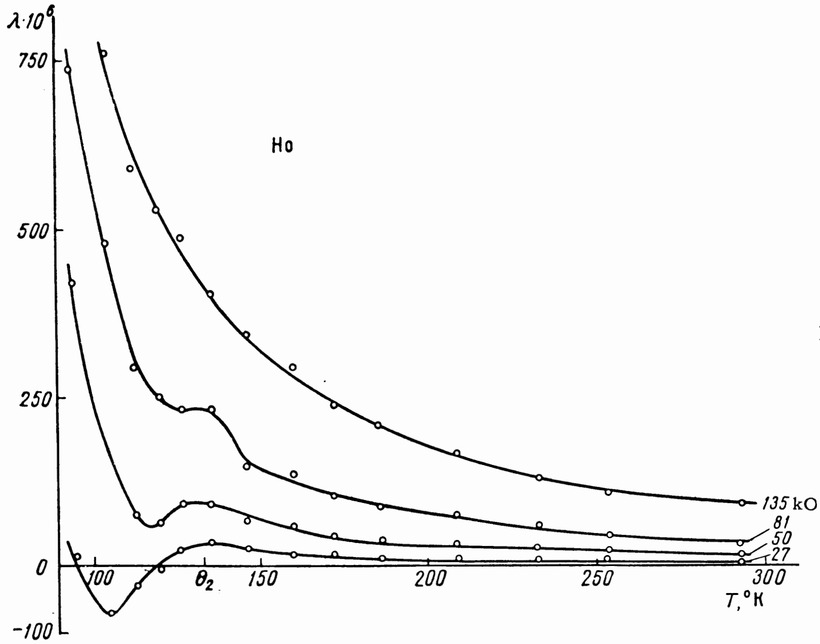

FIG. 7. Dependence of magnetostriction on temperature at constant field in Ho.

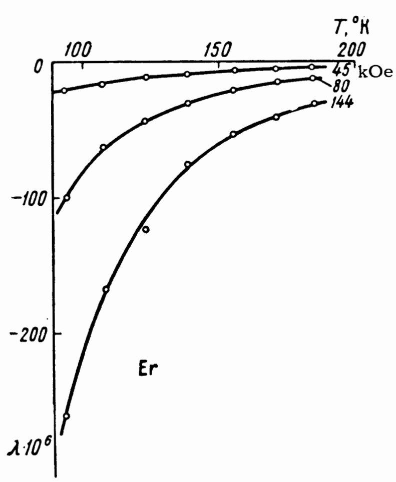

FIG. 8. Dependence of magnetostriction on temperature at constant field in Er.

5. It was discovered by us that the rare-earth metals studied possess a very large magnetostriction in the paramagnetic range (100 to 150 degrees above the Θ_2 point). Here the magnetostriction in a field of order 150 kOe reaches a value of (100 to 400) $\times\ 10^{-6}$. In Kapitza's work[1] it was shown that in the paramagnetic range, the dependence of magnetostriction on field can be represented in the form of a series,

$$\lambda_a = aH^2 + bH^4 + \ldots, \tag{5}$$

where a and b are constant coefficients for adiabatic magnetostriction. Our measurements showed that for Dy, Ho, and Er in the paramagnetic range, in the part where the Curie-Weiss law is obeyed, the adiabatic magnetostriction λ_a satisfies the relation (5), and that in fields up to 150 kOe it is sufficient to take into account only the first two terms of the series. For Tb in the paramagnetic range, the magnetostriction depends on the field in a more complicated manner; apparently it is necessary in the case of this metal to take account of terms of higher order in formula (5).

As has already been pointed out above, in measurements in pulsed fields it is necessary to take into account the effect of the adiabaticity of the process. A calculation carried out according to formulas (1) to (3), with use of experimental data for the paramagnetic susceptibility, the specific heat, and the thermal expansion,[12, 13] showed that within experimental error, the coefficient b is entirely connected with the adiabaticity of the magnetization process. As regards the coefficient a, for it the adiabatic increment amounts to 20 to 60% of the value measured in pulsed fields. Thus our measurements indicate that in the paramagnetic range, in fields up to 150 kOe, the isothermal magnetostriction of the metals investigated satisfies the relation

$$\lambda_T = a_T H^2, \tag{6}$$

where a_T is a constant for isothermal magnetostriction. This result agrees with the deductions of Akulov,[14] who showed that the magnetostriction should be proportional to the square of the magnetization:

$$\lambda_T = a_T' I_T^2. \tag{7}$$

Since in fields up to 150 kOe paramagnetic saturation can be neglected,

$$I_T = \chi_T H \tag{8}$$

and from (7) it follows that

$$\lambda_T = a_T' \chi_T^2 H^2, \tag{9}$$

in agreement with the relation (6).

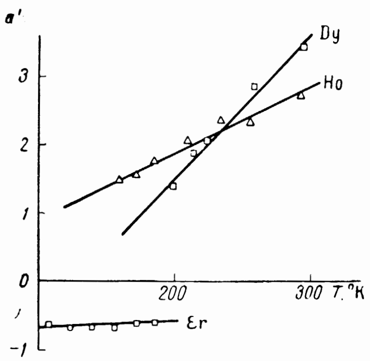

FIG. 9. Dependence of the coefficient a' (in 10^{-8} Oe^{-2}) on temperature.

Figure 9 shows the temperature dependence of the adiabatic coefficients $a' = a_T' + \Delta a'$ for Dy, Ho, and Er. It is clear that within the limits of experimental error, a' can be expressed in the form

$$a' = a_0' + a_0'' T. \tag{10}$$

From the work of Clark et al.[15] it follows that in the single-ion approximation, the coefficient a_T' should not depend on temperature; and simple thermodynamic calculations show that the adiabatic increment $\Delta a'$ is proportional to the temperature. However, the value of $\Delta a'$ calculated from literature data on the specific heat, susceptibility, and thermal expansion[12,13] turns out somewhat smaller than $a_0''T$. To clear up the problem of the dependence of a_T' on temperature, it is necessary to make measurements of these quantities on the specimens investigated, in order to make sure that the temperature dependence of a_T' obtained from our data is not a consequence of a deviation of α_I, χ_I, and c_I for our specimens from the literature data.

6. The following circumstance demands attention: the magnetostriction in the paramagnetic range is negative for Er, whereas the magnetostriction of Tb, Dy, and Ho is positive. As has already been pointed out, the magnetoelastic energy apparently exerts a significant influence on the form and type of antiferromagnetic structure. In Er, in contrast to Tb, Dy, and Ho, there is observed not a helicoidal but a cycloidal magnetic structure.[11] Possibly this is directly connected with the negative sign of the magnetostriction of erbium.

[1] P. L. Kapitza, Proc. Roy. Soc. (London) **A135**, 537 (1932).

[2] K. P. Belov and R. Z. Levitin, JETP **37**, 565 (1959), Soviet Phys. JETP **10**, 400 (1960).

[3] T. Nakamichi and M. Yamamoto, J. Phys. Soc. Japan **17**, Suppl. B-I, 214 (1962)(Proc. Internatl. Conf. on Magnetism and Crystallography, Kyoto, 25-30 Sept., 1961).

[4] K. P. Belov, R. Z. Levitin, S. A. Nikitin, and A. V. Ped'ko, JETP **40**, 1562 (1961), Soviet Phys. JETP **13**, 1096 (1961).

[5] E. W. Lee and L. Alberts, Proc. Phys. Soc. (London) **79**, 977 (1962).

[6] K. P. Belov and S. A. Nikitin, JETP **42**, 403 (1962), Soviet Phys. JETP **15**, 279 (1962).

[7] S. Legvold, J. Alstad, and J. Rhyne, Phys. Rev. Letters **10**, 509 (1963).

[8] A. E. Clark, R. M. Bozorth, and B. DeSavage, Phys. Letters **5**, 100 (1963).

[9] K. P. Belov, Uprugie, teplovye i élektricheskie yavleniya v ferromagnetikakh (Elastic, Thermal, and Electrical Phenomena in Ferromagnets), Fizmatgiz, 1957.

[10] W. Henry, in the collection Novye issledovaniya redkozemel'nykh metallov (New Investigations of Rare-Earth Metals), edited by E. M. Savitskiĭ, Mir, 1964.

[11] K. P. Belov, R. Z. Levitin, and S. A. Nikitin, UFN **82**, 449 (1964), Soviet Phys. Usp. **7**, 179 (1964).

[12] K. A. Gschneider, in the collection Svoistva i primenenie redkozemel'nykh metallov (Properties and Applications of Rare-Earth Metals), (Translations) edited by E. M. Savitskiĭ, IIL, 1960.

[13] F. J. Darnell, Phys. Rev. **130**, 1825 (1963).

[14] N. S. Akulov, Ferromagnetizm (Ferromagnetism), 1938.

[15] A. E. Clark, B. DeSavage, W. Coleman, E. R. Callen, and H. B. Callen, J. Appl. Phys. **34**, 1296 (1963).

Translated by W. F. Brown, Jr.
222