Neutron depolarization measurements on a Dy-single crystal[1]

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Abstract

The depolarization of polarized thermal neutrons by transmission through a Dy-single crystal near the Curie point was measured in order to obtain information about the micromagnetic structure of this substance. The magnetic phase transition observed is sharper than for polycrystals but a marked broadening and hysteresis of the transition is measured for the single crystal too. The results demonstrate a strong relaxation effect for the micromagnetic structure near the transition line with time constants in the order of 30 min. It is assumed that the hysteresis and magnetic after-effect are connected with the lattice clamping effect and the strong magnetostriction of this substance.

Zusammenfassung

Neutronen-Depolarisationsmessungen an einem Dy-Einkristall

Um Aussagen über die mikromagnetische Struktur zu erhalten, wurde die Depolarisation polarisierter thermischer Neutronen beim Durchgang durch einen Dy-Einkristall gemessen. Der magnetische Phasenübergang ist schärfer als bei polykristallinen Proben, obwohl auch hier eine markante Verbreiterung und Hysterese des Überganges gemessen werden konnte. Nahe der ferro-antiferromagnetischen Phasengrenze zeigte sich ein ausgeprägter Relaxationseffekt der mikromagnetischen Struktur mit Zeitkonstanten bis zu 30 min. Es wird vermutet, daß die beobachtete Hysterese und magnetische Nachwirkung mit dem Gitter-»clamping«-Effekt und der Magnetostriktion in engem Zusammenhang steht.

EURATOM/INIS DESCRIPTORS

NEUTRON BEAMS
DEPOLARIZATION
MEASURED VALUES
DYSPROSIUM
MONOCRYSTALS
THERMAL NEUTRONS
CURIE POINT
DOMAIN STRUCTURE
MAGNETISM
PHASE TRANSFORMATION
HYSTERESIS
RELAXATION
TIME MEASUREMENT
CRYSTAL LATTICES
MAGNETOSTRICTION

1. Introduction

The measurement of the neutron depolarization action of a ferromagnetic material is a favourable method for the investigation of the micromagnetic structure. For this method the change of polarization of polarized thermal neutrons transmitting the sample without deflection is measured. The degree of polarization behind the sample is related to the mean domain size and mean magnetization within the domains of the bulk material. Therefore this method is not restricted to the observation of the domain structure at the surface of the samples or to very thin samples. Earlier measurements on Dy-polycrystals [1; 2] gave an indication for rather small magnetic domains, a strong hysteresis of this structure, a large broadening of the magnetic phase transition and a kind of an internal coercive field for the wall mobility. First measurements on a Dy-single crystal [3] yield new information about the micromagnetic structure and indicate a sort of strong magnetic after-effect near the transition temperature.

Dy has a ferromagnetic structure below 85 K, and between 85 K and 178 K an antiferromagnetic spiral structure with magnetic moments within the basal plane of the hexagonal lattice [4; 5]. The antiferromagnetic structure is changed to the ferromagnetic structure by applying a sufficiently large magnetic field $H \geqq H_c = h(T - 85)$, ($h = 125$ Oe/K). The antiferro-ferromagnetic transition is a first order transition with a change from the hexagonal to the orthohombic lattice symmetry resulting in an expansion of the a-(11$\bar{2}$0) and c-(0001) axis [6]. The driving force of this transition is the gigantic magnetostriction ($> 10^{-3}$) whithin the basal plane [7; 8; 9]. The magnetostriction energy (E_{MS}) is the elastic energy (E_E) and the magnetoelastic energy (E_M). For equilibrium the following condition is fulfilled:

$$E_{MS} = E_M + E_E = \\ = -1/8\, C^{\gamma}(\lambda^{\gamma})^2 \simeq -0{,}48\, J/cm^3 \triangleq 1{,}1\ \text{K/atom} \qquad (1)$$

$$C^{\gamma} = 2(C_{11} - C_{12})$$

$$\lambda^{\gamma} = \lambda(a,a) - \lambda(b,a)$$

C_{ij} elastic constants ([10])
$\lambda(a, b)$ saturation magnetostriction measured in b-direction and magnetization parallel to the a-direction

2. Neutron depolarization

The depolarization factor (D_M) is defined as the ratio of the beam polarization (in z-direction) behind the sample to the incident beam polarization (P_0).

$$D_M = \frac{P}{P_0} \qquad (2)$$

The depolarization is 1-D_M. The calculation of the depolarization action of a ferromagnetic sample starts from the Bloch-equation for the expectation value of the neutron spin $\langle \mathbf{s} \rangle$

$$\frac{d\langle \boldsymbol{s} \rangle}{dt} = g\langle \boldsymbol{s} \rangle \times \boldsymbol{B} \qquad (3)$$

g gyromagnetic ratio of the neutron
B magnetic induction

This equation can be used because the mean domain size $\bar{\delta}$ is much larger than the neutron wave length. *Halpern* and *Holstein* [11] solved this equation for a ferromagnetic domain system with the assumption of a nonadiabatic spin transition between the domains. This condition is very well met by the thin domain walls predicted for Dy [12]. The expectation value of the spin at the end of the domain i is related to its value at the end of the domain i-1 by the depolarization dyadic A_i

$$\langle \boldsymbol{s}_i \rangle = A_i \langle \boldsymbol{s}_{i-1} \rangle \qquad (4)$$

The average over many neutron paths has to be carried out ($\langle\ \rangle_{av}$) and for an uncorrelated domain structure the result is:

$$D_M = \frac{P}{P_0} = \left\langle \prod_{i=1}^{N} \left(1 - 2\sin^2\vartheta_i \sin^2 \frac{g\, B_i\, \delta_i}{2\, v}\right) \right\rangle_{av} \qquad (5)$$

ϑ_i is the angle between the magnetization within domain i and the sample magnetization, v is the neutron velocity,

[1] Herrn Prof. Dr. E. Bagge zum 60. Geburtstag gewidmet.

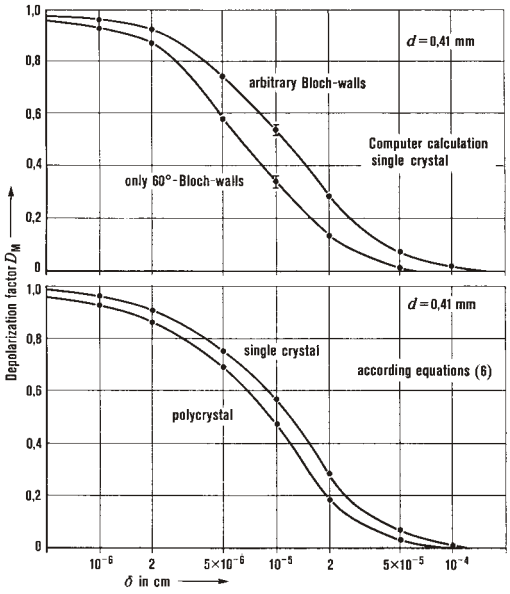

Fig. 1: Depolarization factor for a 0,41 mm thick Dy-crystal as a function of the mean magnetic domain size and various assumptions about the correlations between the domain magnetization

Fig. 1: Abhängigkeit des berechneten Depolarisationsfaktors von der mittleren magnetischen Bereichsgröße für 0,41 mm dicke Dy-Proben und verschiedene Annahmen über die Korrelation der Bereichsmagnetisierung

δ_i the neutron path in domain i and N is the mean number of domains traversed by the neutrons $N = D/\delta$ (D ... sample thickness).

The calculation can be done numerically rather easily. For the limiting cases of small domains ($\delta_i \ll \delta_0$) and large domains ($\delta_i > \delta_0$) one obtains simple relations for poly- and single-crystals for the cases of unmagnetized samples and samples near magnetic saturation. δ_0 is defined as the distance which needs the neutron for a whole Larmor precession $\left(\delta_0 = \frac{2\pi v}{g B}\right)$. The difference between poly- and single-crystals lies in the fact that for a Dy-single crystal the domain magnetization is in the basal plane and therefore in a defined direction to the neutron beam (in our case 90°). The formulas for the limiting cases are [13]

$$\delta_i \ll \delta_0 \qquad\qquad \delta_i > \delta_0$$

Unmagnetized sample:

$$\text{poly crystal} \quad D_M = e^{-\frac{g^2}{3v^2} B_s^2 \bar{\delta} D} \qquad D_M = e^{-\ln 3 \frac{D}{\bar{\delta}}}$$

$$\text{single crystal} \quad D_M = e^{-\frac{g^2}{4v^2} B_s^2 \bar{\delta} D} \qquad D_M = e^{-\ln 2 \frac{D}{\bar{\delta}}}$$

Sample near magnetic saturation ($B_z \sim B_s$) (6)

$$\text{poly and single crystal} \quad D_M = e^{-\frac{g^2}{2v^2}(B_s^2 - B_z^2) \bar{\delta} D} \qquad D_M = e^{-\frac{2\Delta B}{B_z} \cdot \frac{D}{\bar{\delta}}}$$

The various δ-dependences at various temperatures and magnetic fields can be used to decide between the limiting cases. Fig. 1 shows the results according to formula (5) with various assumptions about the correlation between the domains and the dependence as given by the equations (6).

3. Experimental set-up

The polarized neutron set-up at the tangential beam hole of the TRIGA-Mark II reactor was used for the measurements [1]. Co-Fe single crystals in reflection position are functioning as polarizer and analyzer. The beam polarization was determined by the combined shim-spin-flip method [14]. An accuracy of about 0,4 % was achieved within a total counting time of 15 min. The measurements were carried out at a neutron wave length of 1,4 Å and a reactor power of 250 kW.

The single crystal was grown by a horizontal boat cold-crucible method from a 99,9 % pure material. The dimensions of the crystal after growing were 28 cm long and 7 mm in diameter. A disc with a thickness of 0,41 mm was cut off this crystal with the basal plane of the hexagonal lattice parallel to the surface of the disc. The disc has a nearly ellipsoidal shape with axis of 7 and 22 mm. An area of only 5×15 mm² was exposed to the neutron beam using a suitable Cd-cover. The strong magnetic anisotropy between the basal plane and the c-axis causes the magnetic induction of the various domains being always parallel to the surface of the disc and therefore perpendicular to the neutron beam.

The sample was mounted together with an Al sheet and the Cd-cover to an evaporation cryostat where the temperature can be varied between 4,2 K and room temperature. The stability of temperature and the relative accuracy was ± 0,1 K, any other temperature gradient within the sample being smaller than 0,1 K too. Various thermocouples, carbon resistors and thermistors are used for temperature measurement. It was possible to establish a magnetic field varying between 10 Oe and 2,2 kOe by means of on oil-cooled coil. Measurements are carried out with the easy a-(11$\bar{2}$0)-axis and the harder b-(1$\bar{1}$00)-axis parallel to the magnetic field.

4. Experimental results

Results of depolarization measurements for a single crystal below the Curie-point are shown in Fig. 2 together with the results for a polycrystal. The D_M-factor for the single crystal decreases very rapidly with increasing field, corresponding

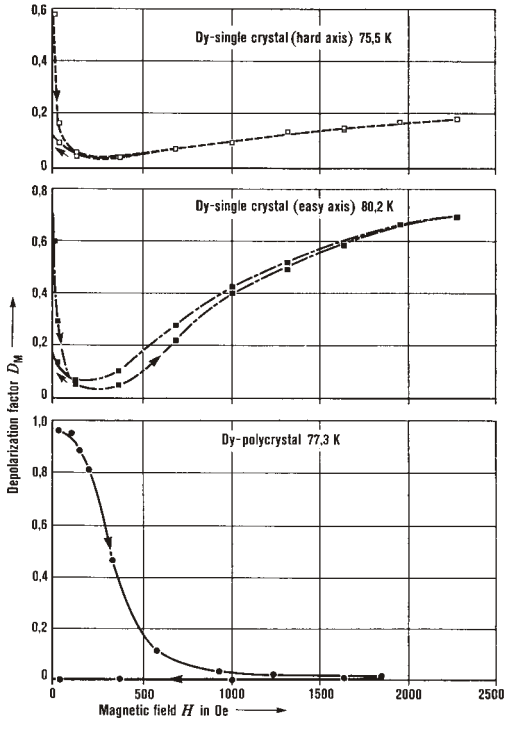

Fig. 2: Measured depolarization factor as a function of the magnetic field for Dy single and polycrystals below the Curie point

Fig. 2: Abhängigkeit des gemessenen Depolarisationsfaktors vom angelegten Magnetfeld für ein- und vielkristalline Dy-Proben unter der Curie-Temperatur

to a fast growth of the domains (equ. (6); from $10^{-5} \rightarrow 10^{-4}$ cm). For cooling the sample at 10 Oe a mean domain size of $\bar{\delta} = 1{,}7 \times 10^{-5}$ cm was measured at 30 Oe, whereas for cooling at 30 Oe the mean domain size at 30 Oe was $2{,}5 \times 10^{-5}$ cm indicating a larger wall mobility at higher cooling fields. In higher fields D_M increases as a result of the sample magnetization and orientation of the domains parallel to the magnetic field. This increase is more pronounced for magnetization parallel to the easy a-axis. The polycrystal shows higher D_M-values or smaller domains and a strong hindrance for the growth of larger domains. This points to a forceful influence of internal stresses on the wall mobility.

Fig. 3 shows the D_M-factor at 96,6 K. In fields below $H_c = 1450$ Oe the sample is antiferromagnetic with theoretically no depolarization action ($D_M \sim 1$). The depolarization starts before this value indicating the existence of some regions with a net ferromagnetic moment even below H_c and shows a sharper transition to the ferromagnetic state. In the ferromagnetic region the increase of D_M is observed according to the sample magnetization. A marked hysteresis of the ferromagnetic domain structure in the pure ferromagnetic region and in the transition region can be noticed. The transition between the anti-ferromagnetic and ferromagnetic state shows, even for the single crystal, a strong broadening, a shift for increasing or decreasing magnetic field and a marked time dependence of D_M or of the magnetic domain structure near the transition field, which will be discussed later on.

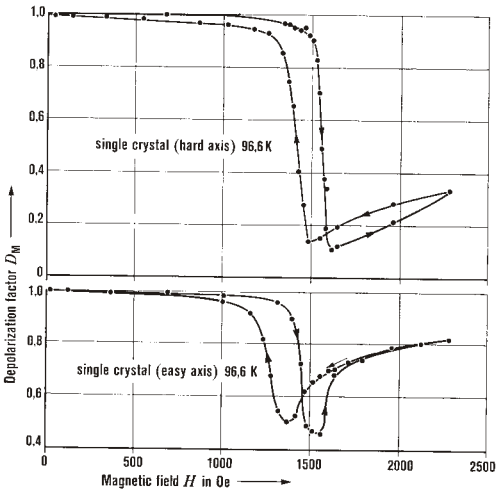

Fig. 3: Measured depolarization factor as a function of the magnetic field for 96,6 K where the antiferro-ferromagnetic transition line is crossed at $H_c = 1450$ Oe

Fig. 3: Abhängigkeit des gemessenen Depolarisationsfaktors vom angelegten Magnetfeld bei 96,6 K wo bei $H_c = 1450$ Oe die antiferro-ferromagnetische Umwandlungsgerade überschritten wird

The mean domain size for the ferromagnetic region, where an increase of D_M is observed, could be calculated by equation (6) and the B_s values measured by *Behrendt* [4]. The results are demonstrated in Fig. 4, where the last diagram shows the temperature dependence of the mean domain size at constant field. A larger wall mobility at higher temperatures can be deduced from these results as well.

The shift of the magnetic transition field observed while measuring at constant temperature (Fig. 3) is converted to a shift of the transition temperature for measurements at constant magnetic fields (Fig. 5). A shift of about 2 K was noticed for an increase or decrease of temperature respec-

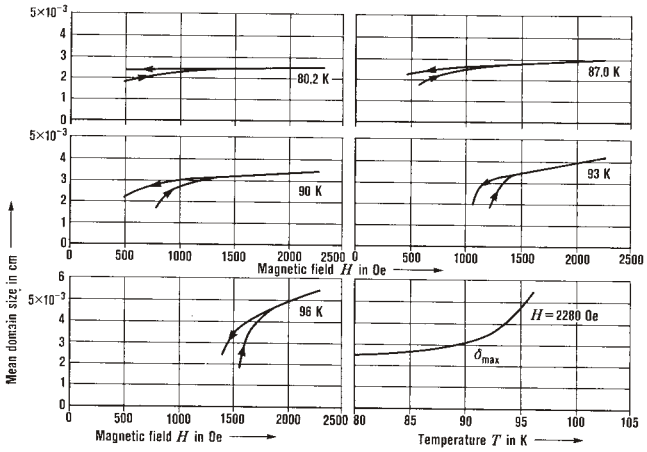

Fig. 4: Mean magnetic domain size as a function of the magnetic field and temperature (last diagram) as obtained from the measurements in the ferromagnetic region

Fig. 4: Mittlere magnetische Bereichsgröße als Funktion des Magnetfeldes und der Temperatur im ferromagnetischen Bereich

tively. This value is of the same order of magnitude as the magnetostriction energy mentioned before (equ. (1)). The smaller slope of the D_M-curve at 370 Oe is explained by a predominant parallel orientation of the domains at larger fields. The measured thermal hysteresis of the transition at 10 Oe was 2,1 K, being somewhat smaller than the value of 5 K reported by *Wilkinson* et al. [5] and the value of 3 K stated by *Bartholin* et al. [15], evidently depending on the quality of the crystal.

A marked relaxation effect was observed in the transition region. Fig. 6 shows some typical results. For these measurements the magnetic field was changed suddenly from 130 Oe, where the sample was in the anti-ferromagnetic state, to the specified value. The final field values, 530 and 660 Oe, are in the transition region, whereas 850 Oe belong to the region where the D_M values are increasing, caused by a slow but predominant growing of the domains orientated parallel

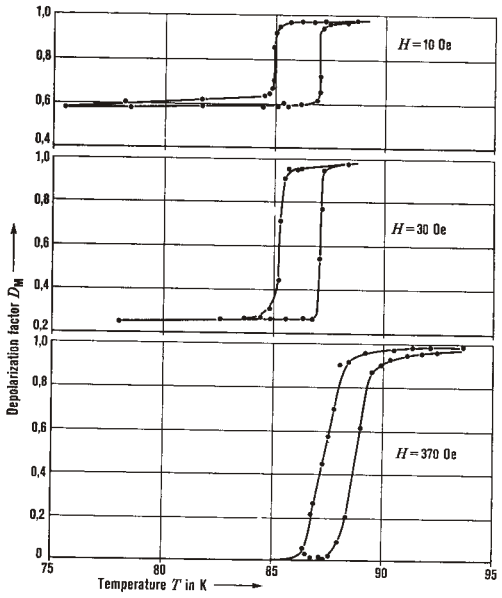

Fig. 5: Temperature dependence of the depolarization factor at various magnetic fields

Fig. 5: Temperaturabhängigkeit des Depolarisationsfaktors bei verschiedenen Magnetfeldern

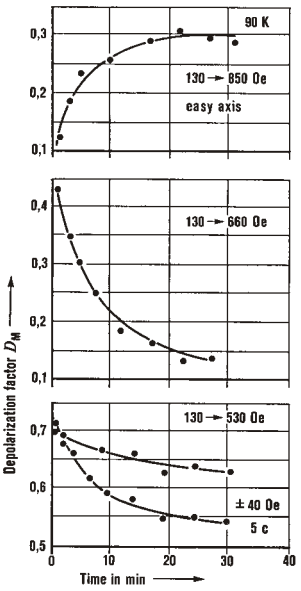

Fig. 6: Time dependence of the depolarization factor observed near the magnetic transition line (lowest curve: magnetic field is altered between 530 ± 40 Oe with a period of 5 c)

Fig. 6: Beobachtete Zeitabhängigkeit des Depolarisationsfaktors bzw. der mikromagnetischen Struktur nahe der magnetischen Umwandlungsgeraden (unterste Kurve: Magnetfeld zwischen 530 ± 40 Oe mit einer Periode von 5 Hz variiert)

to the field. The activation energy ΔE for this relaxation process can be calculated according to equation

$$P = C\,e^{-\frac{\Delta E}{kT}} \qquad (7)$$

where P stands for the probability that the activation is produced by thermal fluctuations. C is a factor of the order of the Debye-frequency [16; 17]. Our measurements yield values of $\Delta E \sim 0{,}25$ eV. *Belov* et al. [18] observed for magnetization measurements on Dy in fields of 50 Oe near 85 K a strong magnetic viscosity too. He concluded that the magnetostriction plays a dominant role in this process.

5. Discussion

In contrast to the measurements on polycrystals a sharper transition at the Curie point could be observed, even the broadening is remarkable for a first order transition.

The size of the ferromagnetic domains could be calculated from the measurements in low fields and from these near magnetic saturation. The field and temperature dependence of the domain sizes is quite common. The domains of the single crystal are much larger than for polycrystals where the grain boundaries represent an upper limit for the domain size in this material with a strong anisotropy. The existence of an inner coercive force for the wall mobility was noticed in single crystals too. This coercive force is small near the transition temperature and increases with decreasing temperature. The existence of an inner coercive field agrees with the calculations of *Egami* et al. [12].

Thermal hysteresis was observed for the measurement at constant magnetic fields. The difference of the transition temperature can be connected with the difference of the magnetoelastic energies between the antiferro- and ferromagnetic state, which was calculated to −2,2 K/atom [9], the so-called lattice clamping effect [8]. It is assumed that the thermal hysteresis effect is a consequence of this lattice clamping effect.

The relaxation of the ferromagnetic structure near the critical transition field H_c demonstrates a strong magnetic aftereffect in this substance. Internal stresses (τ_i) caused by the lattice deformation at the transition point can give an additional term ($\tau_i \lambda$) for the driving force. The strong pressure dependence of the transition is known [15] and therefore a reduction of the stresses by thermal activation can shift the transition of the sub-domain [19] to the value determined by the saturation magnetostriction.

Further measurements with crystals prepared by various thermal treatments and the additional measurements of the magnetic Bragg-reflection are planned.

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