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Dynamics of Domain Walls in the Antiferromagnetic Phase of Dysprosium: a Brief Review

Iu. Liubimova[1] and S. Kustov[2]

[1]School of Photonics, ITMO University, Kronverkskiy pr., 49, lit. A, St. Petersburg, 197101, Russia
[2]Department of Physics, University of Balearic Islands, Cra. Valldemossa km 7.5, Palma de Mallorca E07122, Spain

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Abstract. A brief analysis of several effects related to dynamics of domain walls in antiferromagnetic dysprosium is given. We review experimental studies and interpretations of thermal hysteresis in a number of physical properties of dysprosium and other rare earths that emerges during thermal cycling of samples, scaling laws of magnetic hysteresis, nonequilibrium state of domain walls and relaxation processes in antiferromagnetic phase of dysprosium.

Almost all applied properties of ferroics and multiferroics are due to domain walls (DWs). Detailed studies of new features of DWs and the search for their new unusual properties are subjects of “domain wall engineering” [1]: the science of the principles and ways of creating regular stable domain structures for various applications, like information storage. Domain wall engineering deals also with the unusual intrinsic properties of DWs, like superconductivity of twin boundaries in non-stoichiometric tungsten trioxide (WO_3) [2], magnetoelectric coupling in holmium manganite ($HoMnO_3$) [3], electric conductivity of DWs in lithium niobate ($LiNbO_3$) [4]. In most cases, the unusual properties of DWs stem from their interaction with other lattice defects, e.g. pinning by point-like defects. Examples of such interactions are reviewed by Salje and Dahmen [5].

In recent years, the properties of antiferromagnets have attracted much attention due to their unusual DW dynamics [5]. Devices based on the antiferromagnets can be used for different applications such as: reading, writing, storing and transmitting data [7-11]. The energy of the antiferromagnetic DWs is often lower than domain wall energy of other ferroics [12], enabling creation of a higher DW density. According to theoretical estimates, the domain wall switching rate is an order of magnitude higher in antiferromagnets than in ferromagnets [8,9]. The study of the dynamics of domain walls in antiferromagnetic materials is a difficult experimental task [13]. It requires highly sensitive methods since the motion of an antiferromagnetic domain wall does not change the net magnetic flux across the sample.

Rare-earth elements and their alloys are a particular family of magnetically ordered materials with localized magnetic moments and the highest values of magnetic moment per atom. Dysprosium, as a representative of rare-earth elements, has a complicated temperature-magnetic field phase diagram, shown in Fig. 1, with several ferromagnetic (collinear [14,15], angular [16]) and antiferromagnetic (vortex [17,18], fan [19,20]) spin structures. Rare earths and their alloys are used in innovative research and practical technologies in metallurgy, nuclear energy, optics, medicine, chemical and glass industry, telecommunications

Corresponding author: Iu. Liubimova, e-mail: liubimovaiuliia@itmo.ru

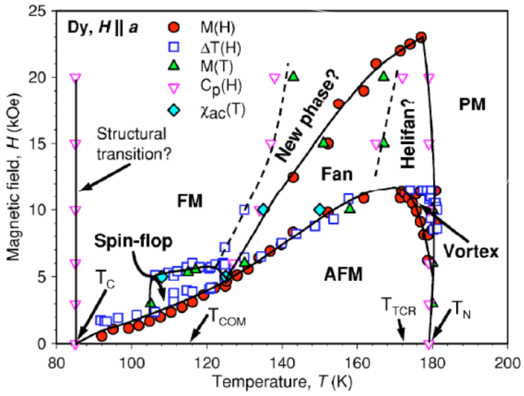

Fig. 1. The magnetic phase diagram of single crystal of dysprosium. At least the following phases are present in the temperature range 200 - 80: ferromagnetic, antiferromagnetic, fan, vortex, paramagnetic. Reproduced from A. S. Chernyshov et al. [17], with permission. Copyright (2005), American Physical Society, DOI: 10.1103/PhysRevB.71.184410.

equipment manufacturing, electronics, laser technology and other applications [6-10,21]. Thus, on one hand, Dy represents a challenging model system for studying magnetically ordered states. On the other hand, despite the complexity of magnetic states, Dy is an important material for various applications.

Under zero magnetic field Dy has a helical antiferromagnetic structure below the Neel temperature $T_N \approx 178$ K [22,23]. In the helical structure, which results from a competition between positive and negative exchange interactions [24], magnetic moments are confined in the closed packed basal planes of the hexagonal structure, rotating with respect to the *c*-axis. The first-order magnetostructural antiferromagnetic – ferromagnetic transition occurs in Dy at the Curie temperature, $T_C = 86$ K on cooling, and at 92 K on heating [22,23]. On cooling from T_N to T_C, the rotation angle between spins in the neigh-boring basal planes decreases from 43.2° to 26.5° [23,25].

Thermal and magnetic hysteresis are DW-related phenomena in the antiferromagnetic Dy, other rare earths and their alloys. The former is the difference between magnetic, elastic, anelastic and other properties in the antiferromagnetic phase, measured during cooling and subsequent heating. The thermal hysteresis is especially intense after cooling down to the ferromagnetic state [26–29]. The origin of thermal hysteresis was the subject of intense studies based on a variety of experimental methods. Two types of DWs, represented in Fig. 2, are considered in spiral antiferromagnetic structure [28]. The Type I DWs are perpendicular while the Type II DWs are parallel to the *c*-axis, as is shown schematically in Figs. 2a and 2b, respectively. Two domains separated by Type I DW have different directions

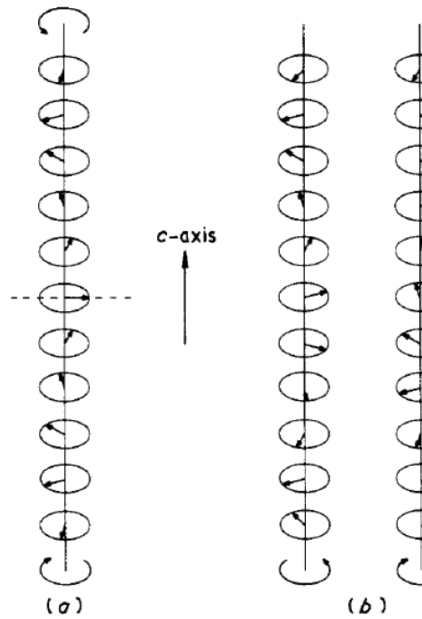

Fig. 2. Spiral domain walls (a) I type, (b) II type. Reproduced from S. B. Palmer [26], with permission. Copyright (1975), IOP Publishing, DOI: https://doi.org/10.1088/0305-4608/5/12/022.

of spin rotation. Type I DWs possess thus a net magnetic moment, responsible for their interactions. The Type II DW runs parallel to the *c*-axis, and the spin rotation also changes from one domain to another. The structure of Type II DW is rather complex.

Initial experimental studies detected thermal hysteresis only after cooling spiral spin antiferromagnets below T_C. Del Moral and Lee [26] assumed that the difference between the temperature spectra of magnetic susceptibility during heating and cooling could be the result of the presence, during heating, of "ferromagnetic" DWs separating antiferromagnetic domains. Similar ideas were employed by Palmer and Lee [27] and Palmer [28]. Palmer [28] suggested that the observed anomalous thermal hysteresis of the elastic and anelastic properties of Dy and other rare-earth alloys was due to spiral spin domains. It was assumed that when the sample is heated from temperature below T_C, the domains of the antiferromagnetic phase "nucleate" from the domains that existed in the ferromagnetic phase. At the Curie temperature the ferromagnetic DWs expand, forming the antiferromagnetic phase. Multiplicity of ferromagnetic domains results in the formation of the antiferromagnetic domains and DWs upon further heating above T_C. This assumption was apparently consistent with experiment: elastic anomalies were detected only when dysprosium was heated from temperatures below T_C. Thus, initial works [26-28] attributed thermal hysteresis in rare earths and their alloys to Type I DWs.

Temperature modulation experiments [30,31] showed that during heating from the ferromagnetic phase thermal hysteresis exists in polycrystalline Dy up to ca. 170 K. This temperature was consistent with the observations of a small effect in the real part of the AC susceptibility at a frequency of 1 kHz, cooling-heating rate about 0.3 K/min and a magnetic field H = 600 A/m. The authors supposed that an intermediate domain structure existed in Dy during heating from the ferromagnetic state. Thus, the existence of different antiferromagnetic domain structure during heating from below T_C and cooling from above T_N was assumed [31]. This assumption was consistent with the conclusions of Palmer [28].

Corró et al. [32] studied both thermal and magnetic hysteresis of the reversible inverse magnetostriction in polycrystalline Dy using so called mechanomagnetic spectroscopy (MMS) [33]. MMS employs ultrasonic resonant oscillations of a sample and detects periodic stress-induced flux variations across the sample. MMS yields reversible inverse magnetostriction (reversible Villari effect) in magnetically ordered materials as a function of oscillatory strain amplitude, magnetic field, and temperature. Under cyclic magnetic field, following [34], MMS yields magnetization *M(H)* hysteresis and is much more sensitive as compared with conventional measurements of *B(H)* hysteresis in antiferromagnetic materials (with low susceptibility values). Corró et al. reported a disappearance of magnetoelastic coupling in antiferromagnetic polycrystalline Dy under low magnetic fields at 166 K. This temperature thus represented the so-called Villari point [32]. Quite remarkable was the observation that thermal hysteresis during heating from the ferromagnetic state disappeared in the Villari point. The authors suggested that the thermal hysteresis was due to the presence of a residual ferromagnetic phase, presumably stabilized by lattice defects, like dislocations, creating high local stresses/strains. The residual ferromagnetic phase existed only up to the temperature of the Villari point, where the magnetoelastic coupling changed the sign. The crucial role of the Villari point was independently confirmed by measurements of real and imaginary components of magnetic susceptibility, Fig. 3 [34].

Dynamics of ferromagnetic DWs is a classical subject of experimental studies [35]. The magnetization of ferromagnets under a slowly growing magnetic field H is often the result of a rearrangement of domain structure through the avalanche-like motion of DWs. Scaling of ferromagnetic hysteresis is well documented [36-38]. On the other hand, very little is known on the DW dynamics in antiferromagnets, mostly due to experimental difficulties. Kobayashi [29] studied *B(H)* magnetic hysteresis and its scaling

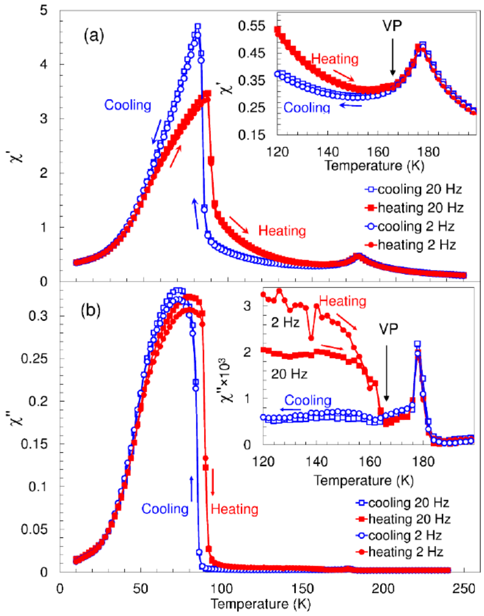

Fig. 3. AC magnetic susceptibility on cooling and heating in the temperature range 9 - 240 K: (a) real part χ' and (b) imaginary part χ''. Excitation field 3.5 Oe and frequencies 20 Hz and 2 Hz. The insets show components χ' and χ''on an expanded scale in the range 120 - 200 K. Reproduced from Iu. Liubimova et al. [31], permission provided by Creative Commons Attribution License. Copyright (2017), Multidisciplinary Digital Publishing Institute, DOI: https://doi.org/10.3390/met7060215.

both in the ferromagnetic and in the helicoidal antiferromagnetic phase of polycrystalline Dy at low fields up to 20 kA/m. Due to resolution limitations, magnetic hysteresis was detected only during heating from the ferromagnetic state and only up to ca. 130 K. Experimental hysteresis loops registered by Kobayashi [29] are shown in Fig. 4. The hysteresis above ca. 130 K was not resolved and the temperature range with discovered Villari point [32] has not been explored. The temperature range of coexistence of the ferromagnetic and antiferromagnetic phases in polycrystalline Dy was assumed to be $T_C \pm 10$ K. The magnetic hysteresis in the antiferromagnetic state was explained by the existence of Type I domain walls [29]. Kobayashi [29] attempted to study scaling of antiferromagnetic hysteresis. The dependence of hysteresis losses ΔW_{loss} versus remanent magnetic flux density B_R is shown in Fig. 5 [29]. Experimental data show power-law scaling $\Delta W_{loss} \sim (B_R)^n$ with the same exponent n = 1.5 for antiferromagnetic and ferromagnetic phases. Kobayashi attributed universality of the ΔW_{loss} (B_R) scaling to the similarity of the dissipative properties of the motion of DWs in ferro- and antiferromagnetic states.

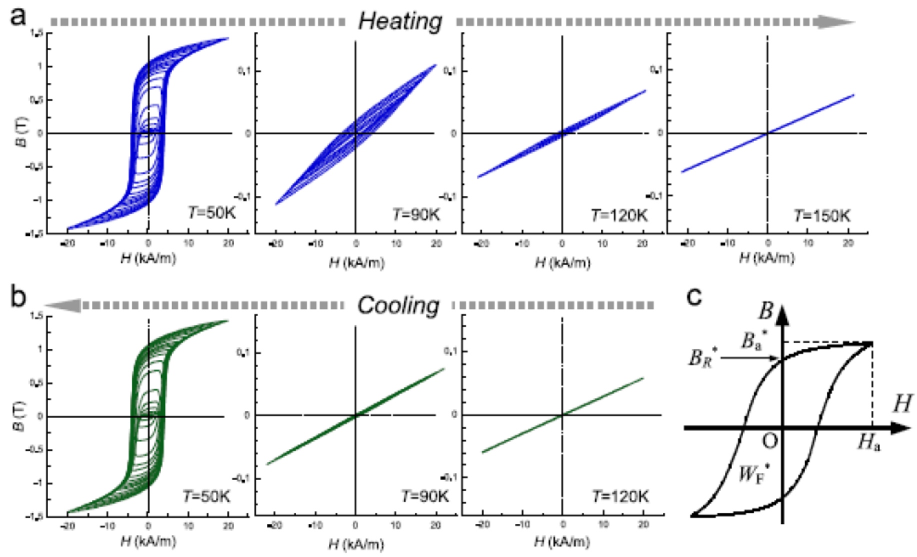

Fig. 4. *B-H* hysteresis loops with different amplitudes of the magnetic field *H*, shown at different temperatures during (a) heating, (b) cooling; (c) the hysteresis loop parameters B_a^*, B_R^* and W_F^* are the maximum flux density, remanent flux density and hysteresis losses, respectively. Reproduced from S. Kobayashi [29], with permission. Copyright (2011), American Physical Society, DOI: 10.1103/PhysRevLett.106.057207.

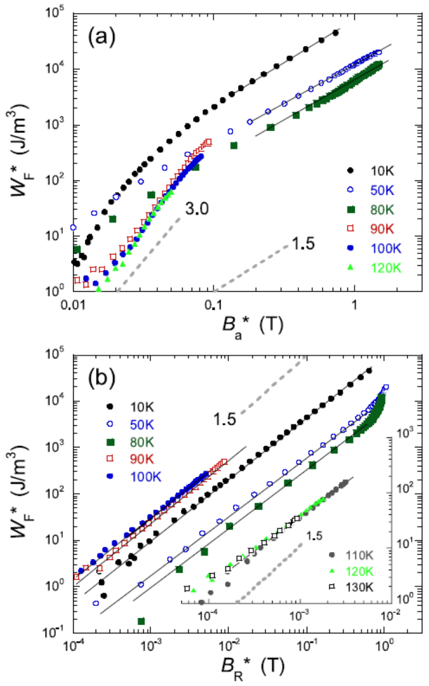

Fig. 5. Relationship between (a) hysteresis losses W_F^* and maximum magnetic flux density B_a^*; (b) between the hysteretic losses W_F^* and the remanent magnetic flux density B_R^* at different temperatures during heating, plotted on a double logarithmic scale for a polycrystalline dysprosium sample. Reproduced from S. Kobayashi [29], with permission. Copyright (2011), American Physical Society, DOI: 10.1103/PhysRevLett.106.057207.

Chen et al. [39] used resonant magnetic X-ray photon correlation spectroscopy to detect DW fluctuations in 500 nm epitaxial dysprosium layers (yttrium/dysprosium/yttrium, 20/500/20 nm). Based on autocorrelation of speckle images, exemplified by Fig. 6, the authors argued that i) DWs underwent prolonged fluctuations in the narrow temperature range below the Neel temperature T_N = 180 K; ii) the fluctuations of the domain structure froze at temperatures approximately 10 K below T_N. It has to be emphasized that the supposed "freezing" temperature is close to the temperature where the thermal hysteresis disappears in Dy. Experimental studies of the ultrafast magneto-optical Kerr effect for the same 500 nm epitaxial dysprosium layer in the Y/Dy/Y system were reported by Langner et al. [40]. The temperature dependence of the magneto-optical Kerr angle showed a peculiarity close to 166 K, not commented upon.

Elastic and anelastic properties of Dy were studied mostly at low frequencies below ca. 10^3 Hz [41-43] and by means of a pulse-echo technique in the MHz frequency range [15,25,27,28,44,45]. Palmer and Lee [27] measured the elastic constants of Dy between 4.2 and 300 K. Their data also pointed to a peculiarity of the C_{13} elastic constant close to the Villari point at 166 K, which remained unexplained. The most informative frequency range for acoustic studies of the DW

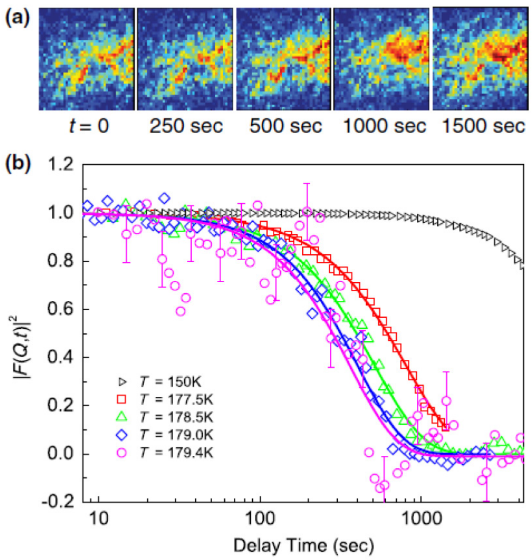

Fig. 6. (a) Snapshots of speckles versus time obtained using X-ray photon correlation spectroscopy at 178.5 K for a thin film Y/Dy/Y; (b) normalized autocorrelation function at various temperatures, the rate of change of function as it approaches Neel's temperature becomes higher. Reproduced from S. W. Chen et al. [39], with permission. Copyright (2013), American Physical Society, DOI: 10.1103/PhysRevLett.110.217201.

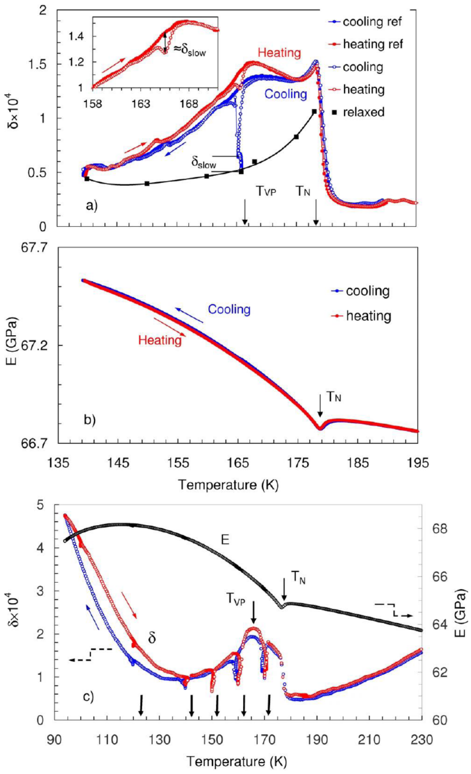

Fig. 7. (a) Temperature spectra of internal friction, δ, in two consecutive thermal cycles 210 K–140 K, cooling-heating rate – 1 K/min. The first cooling process was interrupted at 166 K for 2400 s, resulting in a significant relaxation of the internal friction. The reference cooling-heating curves correspond to the uninterrupted thermal cycle. Black squares represent final values of the internal friction after interrupting cooling scan for 2400 s at fixed temperatures: 140, 150, 160, 166, 168, 175, and 178 K. The inset shows details of the memory effect on heating. (b) Temperature spectra of Young's modulus during thermocycling for as cast sample. (c) Dependencies of internal friction and Young's modulus, E, in a thermocycle 230-95 K (cooling-heating rate 2 K/min) with interruptions of thermal cycling for as received (rolled) sample. Reproduced from S. Kustov et al. [48], permission provided by Creative Commons Attribution 4.0 International License, http:/creativecommons.org/licensesby/4.0/. Copyright (2019), Nature Publishing Group, DOI: https://doi.org/10.1038/s41598-019-41566-7.

dynamics in ferromagnets is usually between 10^3 and 10^5 Hz. These frequencies are high enough to make detectable the microeddy current component, proportional to the frequency, and do not exceed strongly the relaxation frequency of macroeddy current component, see e.g. [46]. Frequencies from this range were used to study elastic and anelastic properties in the antiferromagnetic state of polycrystalline Dy [47,48]. Resonant longitudinal oscillations of bar-shaped samples at a frequency around 90 kHz were employed. Fig. 7 shows an example of temperature spectra of ultrasonic internal friction (IF), expressed as logarithmic decrement δ, and Young´s modulus [48]. The IF is extremely low in the paramagnetic state ($\delta \approx 10^{-5}$) and increases nearly an order of magnitude upon antiferromagnetic ordering. Temperature hysteresis is clearly detected in elastic and anelastic properties even during thermal cycling within antiferromagnetic phase. These two observations pointed to the predominant contribution of DWs to anelasticity of antiferromagnetic Dy [48]. Interrupted cooling and heating scans exemplified in Fig. 7c, revealed intense IF relaxation and T-dot dependences [47,48] with a maximum intensity located at the Villari point, T_{VP} = 166 K, marked also by an IF peak. A conclusion was drawn that antiferromagnetic DWs in polycrystalline Dy are in the non-equilibrium "glassy" state down to ca. 150 K, characterized also by memory and temperature chaos effects [48].

Direct observations of intense anelastic relaxations [47,48] and frequency dependence of imaginary part of susceptibility [34] contradicts observations of DW stability by means of X-ray photon correlation spectroscopy in thin epitaxial films [39]. Kustov et al. [48] concluded that DW fluctuations just below the Néel temperature reported in [39], reflect DW fluctuations during the second order para-antiferromagnetic phase transition. The IF relaxation in the antiferromagnetic polycrystalline dysprosium down to 150 K showed two kinetic processes: “fast” and “slow” [48]: The former scales with the temperature rate, T-dot, and was attributed to the rapid rearrangement of the DW structure similar to the so-called transitory damping term. The latter showed logarithmic kinetics, typical in glassy structures. Only the slow relaxation component was found to be associated with the memory effect [48], a hallmark of spin glasses.

CONCLUSIONS

Experimental results accumulated so far and their interpretations allows one to draw the following conclusions.

The thermal hysteresis that occurs during thermal cycling of Dy, other rare earths and their alloys can be consistently explained by the existence on heating of “extra” Type I DWs, inherited from the ferromagnetic phase or from the ferromagnetic nuclei. The residual ferromagnetic phase can be stabilized by lattice defects up to the Villari point, wherein magnetoelastic coupling in polycrystalline Dy vanishes and changes the sign.

The Villari point around 166 K in polycrystalline Dy is an important characteristic temperature. This temperature controls the behavior of several magnetic properties as detected by a number of different methods.

The observation of relaxation of the reversible Villari effect and internal friction in the antiferromagnetic state from the Néel temperature down to ca. 150 K, as well as the existence of the “memory” effect, point to the non-equilibrium glassy state of antiferromagnetic domain walls over this temperature range.

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