Dynamics of antiferromagnetic domains in spiral antiferromagnets

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We have studied the dynamics of spontaneous relaxation and reorientation induced by an external magnetic field of domain structures in a spiral antiferromagnet. We find that the rate of induced reorientation exhibits a threshold in the magnitude of the reorienting field. The results of our experiments indicate that the induced reorientation of the domain structure is not a result of smooth motion of the domain walls. It is likely that the reorientation occurs via fluctuation-induced formation of seeds with directions of the spiral vector which are energetically more favorable. The dynamics of spontaneous relaxation and induced reorientation are described by logarithmic slowing-down, which is a characteristic of the relaxation of magnetization in spin glasses, and can be explained using the Palmer-Anderson hierarchical model of the dynamics.

1. INTRODUCTION

As a rule, antiferromagnets (AFM) are not single-domain, but rather are broken up into regions in which the orientation of the antiferromagnetism vector, which can be directed along any of several equivalent crystal axes according to the crystal lattice symmetry, is different. Antiferromagnetic domain structures (ADS) depend on the distribution of dislocations in the sample. Up until now the domain structure of antiferromagnets has received little study. This is due to experimental difficulties which arise when investigating the domain structure in antiferromagnets, for which the net magnetic moment of each domain equals zero. Therefore, traditional methods applied to the investigation of domain structures in ferromagnets are not suitable for antiferromagnets.[1] Still less studied are ADS dynamics. It turns out that a convenient method of investigating ADS dynamics is a method based on measurement of magnetostriction, since the reorientation of ADS in an antiferromagnet is usually accompanied by strain of its critical lattice.

In this paper we discuss the magnetostriction connected with ADS reorientation in the cubic antiferromagnet $ZnCr_2Se_4$, which is a spiral AFM. In Ref. 2 it was shown by neutron diffraction that this AFM breaks up into domains in which the propagation vector **q** of the spiral in each domain is parallel to one of three equivalent directions of type [100]. X-ray diffraction experiments[3] have shown that the cubic crystal lattice of single crystals of $ZnCr_2Se_4$ undergoes tetragonal distortions of magnitude $\Delta l/l \sim 5\cdot 10^{-4}$ in the AFM phase, and that the lattice is compressed along **q**. Because the magnetic susceptibility $\chi_\parallel$ along the vector **q** in $ZnCr_2Se_4$ is an order of magnitude larger than $\chi_\perp$, i.e., the susceptibility in the direction perpendicular to **q**, reorientation of the ADS is observed when a magnetic field is applied. As a result of this reorientation the vectors **q** of the domains are aligned along that one of the axes of type [100] which makes the smallest angle with the direction of **H**. This reorientation will be accompanied by a compression of the lattice in the direction of the external field **H**.

The single-crystal system $Zn_xCd_{1-x}Cr_2Se_4$ is a cubic spiral AFM for $x \geqslant 0.5$. This system is convenient for studying the dynamics of domain structures in spiral AFMs, since we can smoothly vary the parameters of the spiral AFM structure by changing the value of x. For example, as x varies from 1 to 0.5 the value of the anisotropy $(\chi_\parallel - \chi_\perp)$ of the susceptibility varies from $4.8\cdot 10^{-3}$ to $2.4\cdot 10^{-2}$ in cgs units, the angle of rotation of the spins in the spiral AFM structure changes from 42° to 28°, the Néel temperature changes from 21 to 13 K, etc.[4] In order to measure magnetostriction in single-crystals of $Zn_xCd_{1-x}Cr_2Se_4$, we developed the method described in Ref. 5. From single crystals of size ~ 1 mm we cut sheets with thickness ~ 0.5 mm parallel to the crystal plane (100). On the polished surface of the sheet we deposited a film of chromium of width ~ 100 μm and thickness ~ 0.1 μm, using the method of vacuum evaporation. At the end of this film we deposited contact pads of copper with thicknesses of 0.2 μm, onto which we welded copper microbond wires with diameters of 30 μm.

The resistance of the chromium films came to $\sim 200\ \Omega$. We recorded the change in resistance as the film was deformed. The coefficient of strain sensitivity $k_{ij} = (\Delta R/R)/e_{ij}$ for the film was measured beforehand in the following way: a film was deposited on a thin (0.1 μm) glass sheet, and one end of the sheet was clamped while the other was displaced by a screw whose pitch was known. We then measured the change in resistance of this film and calculated its strain. It was found that the coefficient of strain sensitivity was practically independent of temperature in the range 4 to 300 K, and equalled $k_\parallel = 1.8 \pm 0.1$ in the case where the film deformed along the current direction and $k_\perp = 0.5 \pm 0.1$ when the film deformed perpendicular to the current direction. This strain gauge was inertialess and had practically no mechanical effect on the samples under study.

2. EXPERIMENTAL RESULTS

In Fig. 1 we show the results of the following experiment: a single-crystal sample of $ZnCr_2Se_4$ was magnetized into a single-domain state along the [100] direction. The magnetic field decreased to zero, after which a spontaneous relaxation of the ADS to the equilibrium state began. The strain was measured along the [010] direction. As the temperature decreases the relaxation rate also decreased: at a temperature of 4.2 K the sample remained single-domain over the course of an hour after the field was switched on, while for $T \geqslant 12$ K the ADS relaxed to its equilibrium state in a time less than 1 sec. We found that the dynamics of the spontaneous relaxation of the ADS was described not by

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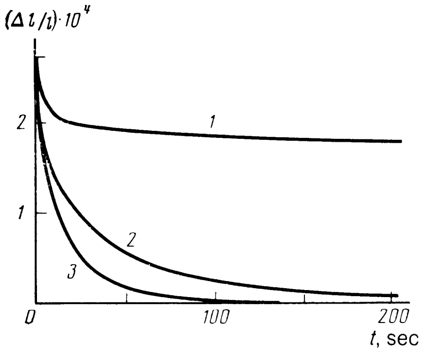

FIG. 1. Dependence of the strain $\Delta l/l$ on time during spontaneous relaxation of ADS from a single-domain state to an equilibrium multidomain state at temperatures of 4.2 K (curve 1), 7 K (curve 2), and 9 K (curve 3).

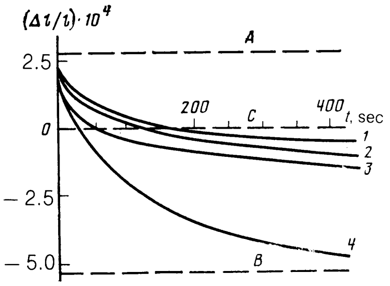

FIG. 3. Time dependence of the magnetostriction $\Delta l/l$ for induced reorientation of ADS in $ZnCr_2Se_4$. **H**|| [010], $T = 4.2$ K. Curve 1 is for $H = 2$, curve 2 for 4.0, curve 3 for 5.2, and curve 4 for 6.5 kOe. Line A corresponds to a single-domain state with **q**|| [100], line B to a single-domain state with **q**|| [010], line C to the equilibrium multidomain state at $H = 0$. $\Delta l/l$ was measured || [010].

exponential relaxation but rather by logarithmic slowing-down, i.e., the system possesses a broad spectrum of relaxation times. The spontaneous relaxation curves are well approximated by the so-called "stretched-exponential," i.e., $\Delta l/l \sim \exp(-(t/\tau)^{\beta})$, where $0 < \beta < 1$. The temperature dependences of β and τ for the compound $ZnCr_2Se_4$ are shown in Fig. 2. It turns out that the curves for spontaneous relaxation to the equilibrium state from states with **q** parallel to the directions [100] and [010] can differ considerably from one another in the same sample. The values of the parameters β and τ corresponding to these curves can differ as much as 15 to 20%. This fact can be explained by the inequivalent distributions of dislocations in the [100]-type directions. As the temperature decreases from T_N to 4.2 K, this inequivalency of the [100] directions becomes more marked.

In Fig. 3 we show the results of the following experiment: a magnetic field of $H = 9$ kOe was switched on parallel to the [100] direction in a single-crystal $ZnCr_2Se_4$ sample at a temperature of 4.2 K. In this case the sample was magnetized into a single-domain state with **q** parallel to [100]. The field was then reduced to zero and the spontaneous relaxation of the ADS began. During the next 300–600 seconds the rate of spontaneous relaxation decreased significantly, after which the sample stayed in a metastable, almost single-domain state with **q** parallel to [100] for several hours. After this a remagnetizing field was applied along [010] and a reorientation of the ADS began, as a result of which the vectors **q** of the domains aligned along [010]. The strain was measured along [010]. We will refer to this reorientation of the ADS in an external field as an "induced" reorientation. It turns out that there is a threshold of induced reorientation of the ADS, i.e., it is necessary to apply a field H_0 of rather large value in order for a substantial reorientation of the ADS to occur in the course of an hour. In order for the reorientation to an almost single-domain state with **q** parallel to **H** to occur in the course of several minutes, it is necessary to apply a field H_1 of a somewhat larger value, i.e., $H_1 \geqslant H_0$. These facts indicate that the reorientation of the ADS is not a result of a smooth shifting of the domain walls. The quantities H_0 and H_1 depend significantly on temperature: for $ZnCr_2Se_4$ at 4.2 K they come to 2 and 5 Oe, respectively. The dynamics of induced reorientation for $t > 3$ to 4 seconds is described by the relation

$$\Delta l/l = U(H)\exp(-(t/\tau)^{\beta}), \qquad (1)$$

where $0 < \beta < 1$.

In Fig. 4 we show the dependence of the quantities $U(H)/U_0$, τ, β on the value of the remagnetization field H for $ZnCr_2Se_4$ at 4.2 K; U_0 denotes the value of the strain along [010] corresponding to a transition from a single-domain state with **q** parallel to [100] to a single-domain state with **q** parallel to [010]. For compounds with concentrations x equal to 0.5 and 0.8, the character of the dependences $U(H)$, τ, β on field and temperature are the same as for the compound $ZnCr_2Se_4$. The quantity H_1 decreases with decreasing x. It is found that the threshold energy density

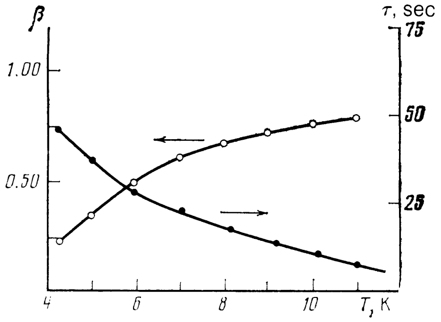

FIG. 2. Temperature dependence of the parameters β and τ in the "stretched exponential" which approximates the dynamics of spontaneous relaxation of ADS in $ZnCr_2Se_4$.

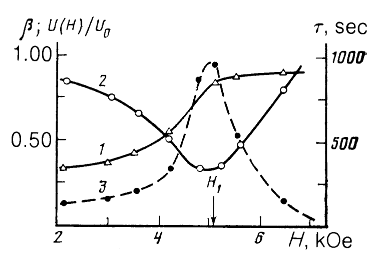

FIG. 4. Dependence on the magnitude of the field of the parameters β and τ in the "stretched exponential" which approximates the dynamics of induced reorientation in $ZnCr_2Se_4$; $U(H)/U_0$ is curve 1, β is curve 2, τ is curve 3.

$\Delta E_M = H_1^2(\chi_\parallel - \chi_\perp)/2$ for reorientation of the ADS comes to roughly $6\cdot 10^4$ erg/cm^3. This value varies by a factor of 1.1 to 1.2 from sample and does not depend significantly on the Zn concentration, or consequently the spiral AFM structure parameter. The scatter in the values of ΔE_M is caused by different degrees of sample imperfection. For certain samples the dependence of the induced reorientation of the ADS on time for values of the external field $H \approx H_1$ was nonmonotonic. We were even able to observe oscillations $\Delta l(t)$ relative to some level which depended on the magnitude of H_1. These features can be explained by the motion of dislocations during the ADS reorientation.

We found that reorientation of ADS does not take place via uniform rotation of the vector **q** in the domains, but apparently is due to fluctuation-induced formation and growth of seeds of domains whose vectors **q** are directed in the energetically more favorable direction. The results of the following experiment bear this out: the sample was first magnetized to a single-domain state with **q**|| [100], after which a field **H**|| [110] was applied; we then recorded the strain associated with the ADS reorientation. It can be shown that the torque acting on a domain in this case is proportional to $H^2(\chi_\parallel - \chi_\perp)\sin 2\phi$, where ϕ is the angle between the directions of **q** and **H**. Therefore this torque is maximal for $\phi = \pi/4$, i.e., for **H**|| [110]. However, in this case the reorientation of the ADS takes place much more slowly than for **H**|| [010]. When the field is oriented along [010], the torque acting on a domain with **q**|| [100] equals zero; on the other hand, the difference in domain energies between **q**|| [100] and **q**|| [010] is maximal. Apparently, as a result of thermal fluctuations seeds of domains arise with **q**|| [010] in a sample with **q**|| [100]; these seeds then grow and reorient the entire domain.

3. DISCUSSION OF RESULTS

Relaxation $\sim\exp(-(t/\tau)^\beta)$, where $\beta < 1$, is usually observed in disordered systems, e.g., the magnetization of a spin glass relaxes in this way. This type of relaxation law can be explained within the hierarchical dynamics model.[6] According to this model, a system can be treated as a collection of interacting subsystems, each of which can be placed in correspondence with a certain hierarchical level with index n. For this model, a subsystem with index $n+1$ can change its original state only when a certain quantity of a subsystem on the preceding nth order level changes its state. This means that the model builds in correlations between subsystems with respect to the various hierarchical levels. We will show that the correlations between neighboring portions of the ADS are caused by the presence of tetragonal magnetostriction and the strain field of the lattice caused by dislocations.

Let us investigate the process of subdivision of a sample into ADS domains. Suppose that a spiral AFM is magnetized to a one-domain state with **q**|| [100], after which the external field is reduced to zero. Let us discuss an edge dislocation for which the dislocation line is parallel to the [010] direction. In this case there exists a region of elastic stress along the dislocation line in which the lattice is stretched in the (010) plane,[7] so that a state with **q**|| [010] is energetically more favorable in this region. As a result of fluctuations, the seed of a **q**|| [010] domain can then arise in this region. We can calculate the critical size r_c of such a seed by minimizing the sum of the elastic and domain-wall energies of the seed. Using a value of $T_N = 21$ K for $ZnCr_2Se_4$ and an anisotropy constant $K_A \sim 10^6$ erg/cm^3 (Ref. 4) for confinement of the spiral axis along the crystallographic directions of type [100], and an energy of spontaneous tetragonal magnetostriction $E_{str} = E_0^2/2 \approx 10^5$ erg/cm^3, we obtain $r_c \approx 10^{-6}$ cm. Here, E is the Young's modulus, and e_0 is the value of the spontaneous magnetostriction $e_0 \approx 5\cdot 10^{-4}$. Domains of smaller size than this will collapse.

Let us estimate the maximum value r_d of an already-formed domain. The increase in size of a domain ceases when the gain in elastic strain energy corresponding to the reorientation of the vector **q** is less than the increase in energy due to increasing the area of the domain wall. It follows that r_d will be on the order of the length over which the value $\Delta l/l$ of the strain caused by a single dislocation decreases to the value $\approx 5\cdot 10^{-4}$ caused by tetragonal magnetostriction. Using the results of Ref. 7, we can show that $r_d \approx (2\text{–}8)\cdot 10^{-6}$ cm. Consequently, the increase in size of a domain ceases almost immediately after it nucleates.

Let us now investigate two dislocations such that the lines of these dislocations are parallel to the [010] axis. Suppose that the dislocations are situated so that between them there exists a region in which the lattice is stretched along the (010) plane. It can be shown that the value of the strain in this region is considerably increased if at one of the dislocations a seed of a domain with q parallel to [010] arises. Therefore, the appearance of a seed at one of the dislocations stimulates the appearance of a seed at its neighbor, which leads to the hierarchical dynamics of the ADS reorientation.

One of the results of the hierarchical model is that the stronger the correlations between subsystems of different levels, the smaller the value of β. Our experiments show that β reaches its minimum value (Fig. 4) for reorientation in a field of that value H_1 for which the gain in magnetostatic energy $H^2(\chi_\parallel - \chi_\perp)/2 \approx 10^5$ erg/cm^3 equals the loss in elastic energy of the tetragonal strain. This experimental result is found to be in agreement with the hierarchical dynamics model. Let us show that the correlation between different portions of the ADS is most marked in the field H_1. Suppose the sample is magnetized to a single-domain state with **q**|| [010], after which a field is applied along [010]. The ADS reorientation at the field value H_1 has already gone to the single-domain state (Fig. 3, curve 4). In actuality, for $H \gtrsim H_1$ there is also a reorientation of those portions of the ADS located near dislocations in whose neighborhood a state with **q**|| [010] is energetically unfavorable in the absence of the field. During reorientation of **q** in a field of value H_1, the decrease in the density of magnetostatic energy in these portions of the ADS is roughly equal to the increase in elastic energy. Therefore, in these portions of the ADS no seed of the domain with **q**||**H** appears. However, if in the neighboring portions of the ADS the vectors **q** are already parallel to **H**, then as a result of the change in the strain field in the portions under discussion the seeds with **q**||**H** can form. Thus, the correlation between different portions of the ADS is most marked for reorientation of the ADS in a field of value H_1. In Fig. 4 we show that for $H \approx H_1$ the value of β is a minimum, while the value of τ is a maximum. This result coincides with the conclusions obtained from the hierarchical dynamics model.[6]

Therefore, the reorientation of domains in spiral AFM belonging to the system $Zn_xCd_{1-x}Cr_2Se_4$ takes place in a threshold fashion. The threshold energy density $\Delta E_M = H^2(\chi_{\parallel} - \chi_{\perp})/2$ for reorientation of the ADS comes to roughly $6\cdot 10^4$ erg/cm^3. This value is independent of the parameters T_N, $|q|$, K_A, etc. of the spiral AFM structure, and coincides with the value of the tetragonal magnetostriction which in our experiments amounted to $\sim 10^5$ erg/cm^3 for various x and parameters of the spiral AFM structure.

The reorientation of ADS takes place via fluctuation-induced formation of seeds of domains with energetically more favorable directions of the vectors **q**. The reorientation dynamics of the domain structure in these AFM can be described by using the hierarchical relaxation model, developed to describe the relaxation of magnetization in spin glasses.

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