Strong clamping of the ferromagnetic transition in epitaxial Er

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We report the magnetization behavior of single-crystal epitaxial films of Er lattice matched to Y_xLu_{1-x} alloy substrates. As the Er is largely strain-free, the main epitaxial constraint on its ferromagnetic transition arises solely from the interaction of the magnetostriction with the clamping effect of epitaxy. We observe large modifications of the magnetic behavior which establish that the clamping effect is large. The very different epitaxial behaviors of Dy and Er are explained in terms of accessible domain structure.

The way that strictions derived from phase change are accommodated in epitaxial systems remains a topic at the forefront of research.[1,2] It frequently happens that the phase transitions of bulk systems are accompanied by dimensional changes, most frequently just a volume change. Ferromagnetic and ferroelectric transitions also can produce length changes up to ~0.3% that are, moreover, anisotropic.[3–5] In epitaxial material, however, the crystal is constrained by its attachment to the substrate. This generally alters the resulting dimensional changes, and modifies the energetics such that the phase diagram may undergo substantial change.[1,2,6] These effects are of special interest for magnetic thin-film systems and thin-film ferroelectrics where the epitaxial constraint may be employed as a control parameter. It can be used, for example, to select a particular phase or to augment its range of stability. In this work we report research on epitaxial Er in which the effect of one specific mechanism, lattice clamping, is clearly identified.

In an earlier study of the phase relationships in epitaxial Dy,[1] Tsui and Flynn (TF) distinguished between the effects of epitaxial strain and of epitaxial clamping. The physical constraint that maintains the epilayer at a constant in-plane dimension is called clamping; in addition, the epitaxial process often changes the in-plane atomic spacing, for example, to maintain atomic registry at the interface, and this is the epitaxial strain ε of interest here. By use of substrates that cover a range of atomic spacings, TF explore the epitaxial phase diagram of Dy as a function of temperature T and strain ε. They find large changes except, surprisingly, that epitaxial Dy grown with zero strain magnetizes much like bulk Dy, even in a crystal only 100-Å thick. These authors reject, on energetic grounds, the straightforward interpretation that the fact the crystal is clamped causes negligible effects. They show instead that Dy evades the epitaxial constraint by breaking into a domain structure in which neighboring domains have different magnetization directions, such that the mean strain vanishes (see also Ref. 6). Thus the small change of the phase diagram observed near $\varepsilon=0$ for Dy did not necessarily show that the clamping itself is a minor perturbation. However, the possible size of the clamping effect remained undetermined.

In what follows we report results for epitaxial Er. Earlier research on Er films[7] has shown that epitaxy has the strong effect of suppressing spontaneous ferromagnetism entirely in films of up to ~1-μm thick. The origins of this remarkable effect in terms of strain and clamping have not previously been analyzed. Here we report magnetic measurements on Er grown epitaxially on nonmagnetic Y_xLu_{1-x} alloy substrates with x adjusted to give a perfect lattice match. With strain effects thus removed we find that the Er magnetic phase diagram remains strongly perturbed. The effect of clamping alone is therefore very large. It is obviously of great interest that Dy evades this strong constraint, while Er does not. In what follows, we first present the experimental results and then explain the reasons why Dy and Er behave differently.

The rare-earth film growth took place at about 400 °C in the University of Illinois EpiCenter by well-practiced procedures[8] with (0001) Y-Lu substrates from e-beam sources grown on fresh buffer layers of (011) Mo on sapphire (1120). Er was then deposited from a pyrolytic boron nitride effusion cell and the assembly was capped with 50 Å of Au. An Y_xLu_{1-x} alloy of composition of $x=39$ at. % was chosen to best reproduce the Er lattice spacing. Between Y and Lu the $\hat{a}$ axis lattice spacing varies from +2.5 to −1.5% of the Er spacing. With reasonable composition uncertainties we estimate that any residual misfit is less than the bulk magnetostrictive strains of ~0.3%. This places the crystals in the regime where clamping is the principal perturbation on the epitaxial system.

Magnetic measurements were undertaken in a Quantum Design superconducting quantum interference device system capable of measurements at fields up to 5.5 T and at temperatures down to 1.5 K. Er in the bulk undergoes an antiferromagnetic-to-ferromagnetic transition at $T_c=20$ K,[9]

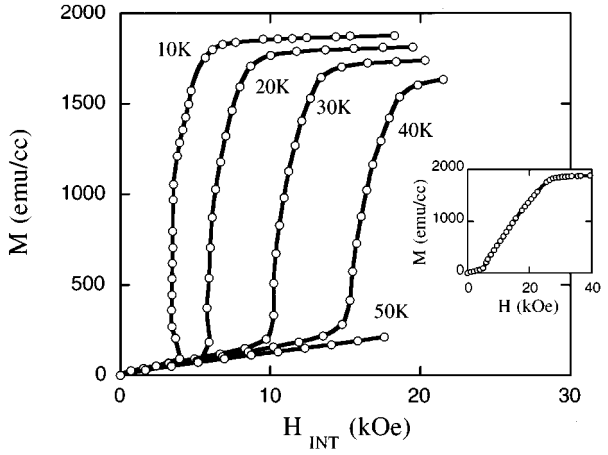

FIG. 1. Magnetization vs internal field at temperatures from 10 to 50 K. These measurements were taken in an ascending magnetic field and have been corrected for the demagnetizing factor. Inset: Magnetization vs applied field at 10 K without demagnetizing correction.

and the evidence of the magnetic transition is found in the magnetization curves below this temperature. Samples were cut to a size suitable for magnetic measurements using a diamond saw. The mass of each sample along with the known thickness and density of the sapphire substrate was used to estimate the area of the sample. Taking the known thickness of Er, the volume of Er was calculated and used to determine its absolute magnetic moment in emu/cm^3 . The internal field is given by

$$\mathbf{H}_{\text{int}}=\mathbf{H}_{\text{app}}+\mathbf{H}_d\,, \tag{1}$$

where the demagnetizing field is defined by

$$\mathbf{H}_d=-4\pi\alpha\mathbf{M}. \tag{2}$$

Here $\mathbf{M}$ is the sample magnetization and α depends on the sample geometry, with $\alpha=1$ for an infinite thin film. The numerical value of α was determined to be 0.9 by requiring that the 10 K magnetization curve be essentially single valued and have a nearly first-order transition from the helimagnetic state to a fully magnetized state in the applied c-axis magnetic field. The reason α departs from unity has never been clearly identified, but may well relate to the domain structure. Using this value of α and Eqs. (1) and (2), we determined the internal field H_c required to initiate substantial magnetic alignment.

Magnetization measurements for a 2000-Å single-crystal Er film, which was grown strain-free on an alloy base layer, are summarized in Fig. 1. The main part shows the magnetization vs internal field at various temperatures, while the inset shows the 10 K magnetization curve without correction for demagnetization effects. Figure 2 shows the resulting critical field as a function of temperature for three samples: the 2000 Å Er film grown on $Y_{39}Lu_{61}$ prepared in this study, a 1750 Å Er film grown on Y from a previous study,[7] and a bulk Er sample, also from a previous study.[10] Note that the two thin-film samples have nearly the same thickness, but their critical fields are markedly different. We emphasize that the film grown on an alloy should have no epitaxial strain but

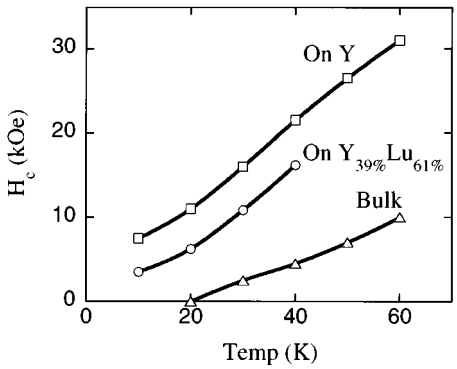

FIG. 2. Critical field (H_c) vs temperature for three Er samples: 1750-Å film on Y, 2000-Å film on strain-free alloy (this study), and a bulk Er sample.

nevertheless remains clamped to the substrate. In contrast the film grown on Y has both clamping and epitaxial strain. In the case of bulk Er the critical field is 0 at a temperature of about 20 K, at which point an external field is no longer required to maintain the c-axis ferromagnetic alignment, which thus occurs spontaneously. In the film samples, however, the critical field curves do not extrapolate to zero field at any temperature, thus indicating complete suppression of the ferromagnetic transition even for the present unstrained sample. We have confirmed this result for a number of samples, including some superlattices.

As a matter of further interest, Fig. 3 shows how the magnetization of the strain-free film sample varies with temperature in a small (200 G) applied magnetic field. Below its Néel temperature, which for this sample is marked by the cusp in the magnetization, Er has a helimagnetic c-axis modulated magnetic structure. In this state each Er atom has a magnetic moment aligned along the c axis, and the sign of the moment oscillates with a period of about eight atomic layers. The two peaks on the left are quite sharp and can be associated with known spin-slip states in which the oscillations lock to the lattice before completing a harmonic cycle, thus leaving a small net magnetic moment. Since the signatures of the helimagnetic state are present in Fig. 3 and the magnetic moment is small, the helimagnetic state in the strain-free film sample evidently persists to zero temperature.

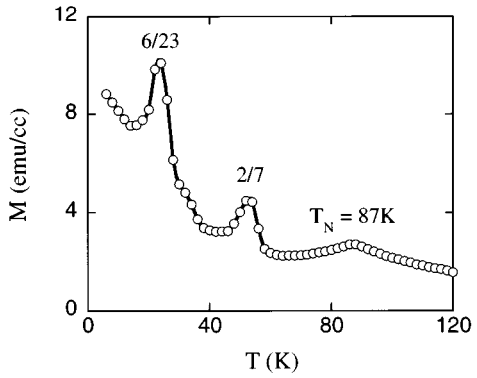

FIG. 3. Magnetization vs temperature at an applied field of 200 G. Evidence of the Néel transition from the paramagnetic to helimagnetic regimes can be seen at 87 K.

From the fact that large fields are required to induce magnetization we deduce that the epitaxial constraint has modified the magnetic phase diagram and suppressed the ferromagnetic transition. In the absence of substantial strain this major change is due almost entirely to epitaxial clamping alone, which is thus revealed as a powerful agent for the control of magnetism in strongly magnetostrictive materials. In point of fact, the critical field of 6.25 kOe required at 20 K in the present material (2000 Å) is slightly more than half the critical field of 11 kOe for a strained Er film of about the same thickness (1750 Å), but grown on Y. While the latter film is expected to exhibit effects from both strain and clamping, the former should be affected by clamping alone. Thus the results lend support to the argument by TF that the clamping effect in Dy is not in fact weak, but rather is comparable to strain effects.

It remains still to be explained why the Dy lattice less than 100-Å thick can evade the clamping constraint by distorting into a domain structure, while Er does not, even for films one to two orders of magnitude thicker. We show that this is a direct consequence of a critical difference between the Dy and Er magnetic structures. The difference is that Dy magnetizes in the basal plane, whereas Er magnetizes perpendicular to the plane even in thin films, because the crystal-field anisotropy is large enough to overcome the shape anisotropy. The in-plane magnetization of Dy with its tetragonal in-plane distortion permits a domain structure in which neighboring domains magnetized in orthogonal directions produce in-plane magnetostrictive strains which average nearly to zero. This is illustrated in Fig. 4(a). In this way clamping and magnetostriction can coexist. For Er, however, with its perpendicular magnetization, the alternative domains polarized in and out of the surface *produce identical magnetostrictions.* This different geometry, as illustrated in Fig. 4(b), therefore, offers no Dy-like mechanism whereby a domain structure can evade the powerful effect of clamping on the magnetostrictive film.

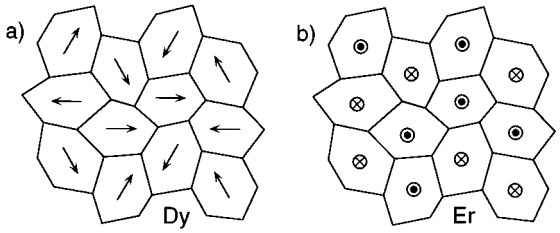

FIG. 4. (a) Proposed magnetic domain structure of Dy which permits magnetostriction despite clamping, since the distortions of the various domains cancel each other when averaged together. (b) Analogous perpendicular magnetic domain structure of Er. The in-plane magnetostriction for each domain does not depend on the sign of the c-axis magnetization, so the resulting in-plane lattice distortions do not cancel each other and are subject to clamping.

Interaction effects of this type between epitaxy and strictions of phase transitions are as yet little studied and remain poorly understood. They evidently can induce complex changes of phase relationships, with a rich dependence on the symmetry of the magnetization process. We expect that much the same phenomena prevail in the ferroelectric process of epitaxial films also. These materials are finding increasing use in switching and manipulating (e.g., piezo) applications. From the present perspective it appears likely that the interaction between epitaxy and the strictions offers a means not only to control the direction of polarization, but also to manipulate the domain structure which, in turn, determines the mean strain in the polar material. For these reasons alone, the phenomena revealed in the present research are likely to be of interest for years to come.

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