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LETTER TO THE EDITOR

Periodic change of the magnetic and structural properties with thickness of Er thin films

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Abstract. Er thin films with thicknesses ranging from 88 to 196 Å and with their *c*-axis perpendicular to the film plane were prepared on the Nb buffer layer deposited on $Al_2O_3(11\bar{2}0)$ substrate. The magnetic phase transition temperatures showed periodic change as a function of Er thickness with a period of 20 Å, which corresponds to about seven atomic layers of Er. The periodic change in the magnetic property was found to be accompanied by that in the *c*-axis lattice constant. A possible origin of the periodic changes is discussed.

In the recent studies on Er/Y [1–3] and Er/Lu [4, 5] films and superlattices, there exists considerable interest particularly in the coherent propagation of the longitudinally modulated spin structure through nonmagnetic layers and in the effect of epitaxial strain on their magnetic properties. In the recent studies, the thickness dependence of the magnetic properties has been discussed only in terms of a change in epitaxial strain which is relaxed as the film thickness increases. This strain relaxation gives a gradual change in magnetic and structural properties over hundreds of Å in thickness [4]. We expect, however, to find some influence of the intrinsic *c*-axis-modulated spin structure on the thickness dependence of the magnetic properties when we fabricate a series of Er thin films by finely tuning the thickness. In this letter, we present a comprehensive investigation on the thickness dependence of the magnetic properties in Er thin films. We found that the magnetic phase transition temperatures and the *c*-axis lattice constant change periodically with the thickness of Er film.

By the molecular beam epitaxy technique using electron-beam evaporation, Er thin films with thicknesses ranging from 88 to 196 Å were prepared on the Nb buffer layer with a thickness of about 250 Å deposited on $Al_2O_3(11\bar{2}0)$ substrate. The substrate was 1×1 cm^2 in area and 0.3 mm in thickness. The Er thin film and the Nb buffer layer were deposited at the rate of 0.2 and 0.3 Å s^{-1} respectively at the substrate temperature of 400 and 800 °C. The base pressure was less than 5×10^{-11} Torr. For *in situ* rate and thickness control, a quartz oscillator was used. The thickness monitor using the quartz oscillator had been calibrated in advance using the satellite peaks in x-ray diffraction measurements for the Er/Y multilayers with different thicknesses of bilayer. From this calibration, the error in thickness was evaluated to be within 2.3% of the thickness for the thicknesses between 50 and 200 Å. During the sample preparations, fine RHEED patterns were observed showing a good epitaxial growth. The samples were always handled in dry N_2 gas or He gas to avoid oxidization in Er thin films because we did not use any capping layers.

X-ray diffraction patterns obtained by θ–2θ scans at room temperature show that the *c*-axis of the Er film is oriented perpendicular to the sample plane. A typical diffraction

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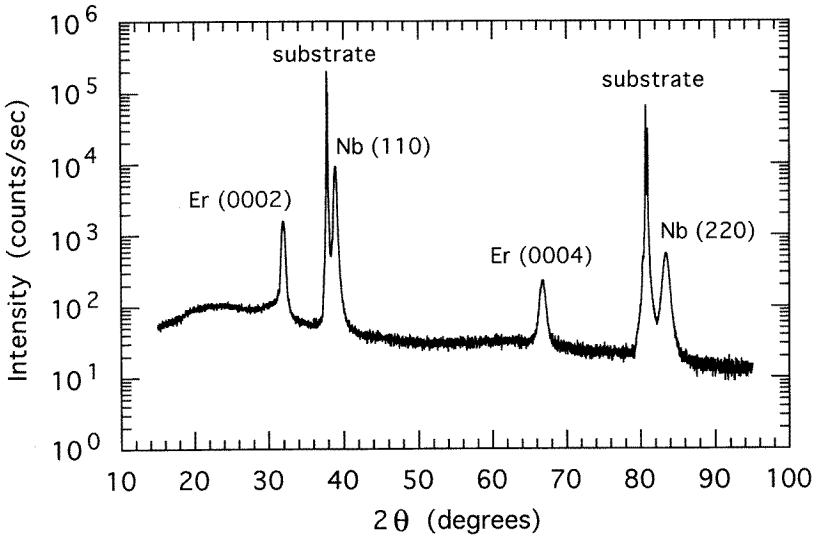

Figure 1. A typical x-ray diffraction pattern obtained at room temperature for the sample with the Er film thickness of 160 Å.

pattern is shown in figure 1. We see that Er(0002) and Nb(110) planes are preferentially oriented in the film plane. The *c*-axis lattice constant at room temperature calculated from the Er(0002) and (0004) peaks is 5.6146 Å on average, which is about 0.5% longer than the lattice constant of 5.5874 Å for bulk Er at room temperature. Figure 2 shows the structural coherence lengths parallel to the *c*-axis. The coherence length was evaluated from the expression $2\pi/\Delta Q$, where ΔQ is the full width at half maximum of the intensity profile. The coherence lengths coincide with the thicknesses of the Er thin films. This means that the Er thin films are in the form of single crystals in all of the thicknesses prepared.

Using a SQUID magnetometer (Quantum Design MPMS2), magnetization M in the field-cooling procedure was measured down to 10 K with an external field of 100 Oe applied parallel to the *c*-axis. Typical examples of the results are shown in figure 3. For all the samples, an anomaly (marked by a circle) which corresponds to a local minimum in $\mathrm{d}M/\mathrm{d}T$ was observed at around 80 K. As we see for the sample of 144 Å Er, a monotonic increase of the magnetization with lowering temperature was found in some samples. Others have a maximum (marked by a triangle) and a minimum (marked by an inverted triangle) like the sample of 152 Å Er. We define T_{max} and T_{min} as the temperatures at which the magnetization has a maximum and a minimum, respectively. The temperature variation accompanied by a maximum and a minimum is similar to that observed in an [Er(38 Å)/Y(72.5 Å)]$_{100}$ superlattice [3]. For this superlattice, a series of magnetic phase transitions has been observed by neutron scattering measurements. Assuming that our samples show the same sequence of magnetic ordering, the temperature dependence of the magnetization for the sample of 152 Å Er is explained as follows. The anomaly around 80 K corresponds to the Néel temperature T_N. The onset of the 2/7 spin phase is marked by a maximum at T_{max} near 45 K. The decrease in M below 45 K is related to the formation of the 3/11 spin phase which has no net moment. The upturn below T_{min} near 17 K comes from the formation of the 4/15 spin phase. Like the Er/Y thin films and superlattices [3], our Er thin films are not expected to experience the full sequence of the bulk commensurate states [6], especially the low-temperature phases including the ferromagnetic phase. This

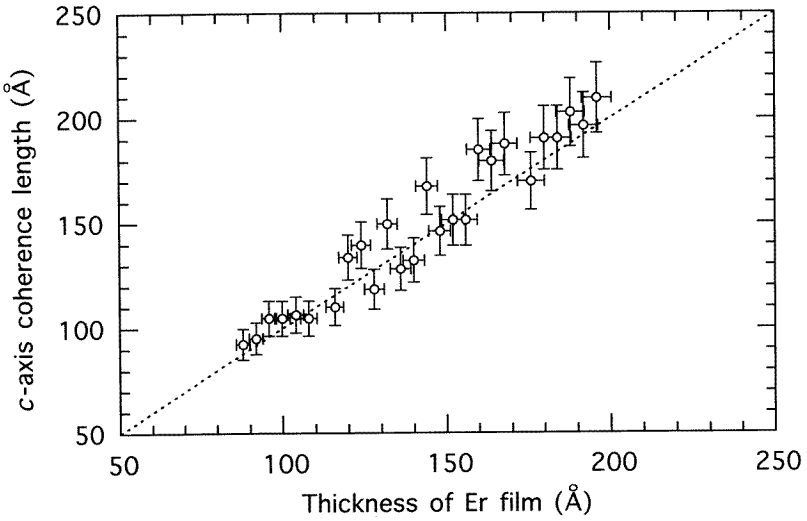

Figure 2. The *c*-axis coherence length against thickness of Er thin films determined from the room-temperature x-ray scans.

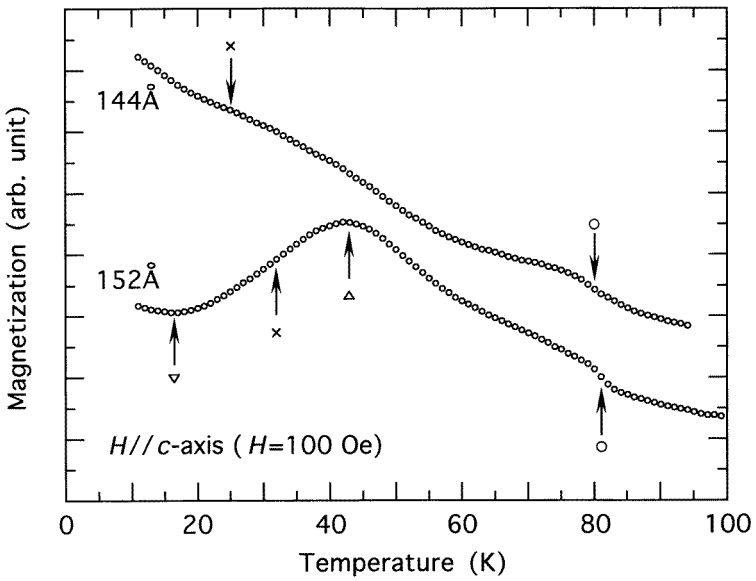

Figure 3. Typical temperature dependence of the magnetization for the samples with the Er film thicknesses of 144 and 152 Å.

is probably because of the epitaxial strain which suppresses the magnetic phase transitions accompanied by structural phase transitions.

In figure 4, the magnetic phase transition temperatures T_N, T_{max} and T_{min} determined from the magnetization measurements are shown as a function of the thickness of Er thin films. The inflection point (marked by crosses) between T_{max} and T_{min} is also shown for reference. For the samples without extrema, only the lowest inflection point is shown. The

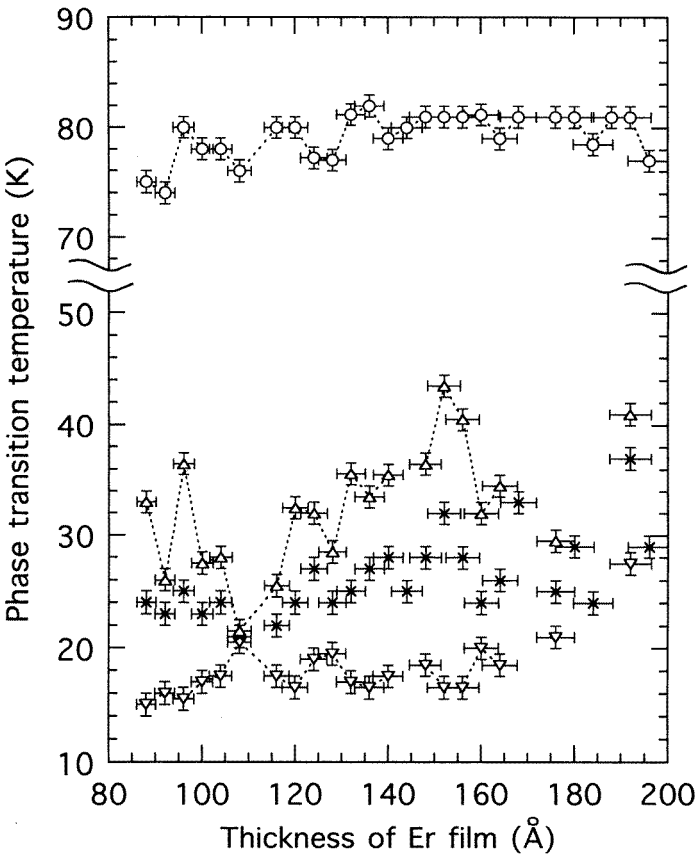

Figure 4. Magnetic phase transition temperatures as a function of the thickness of Er film. The symbols correspond to those in figure 3.

Néel temperature T_N shows a periodic change with a period of about 20 Å superposed on a monotonic increase approaching about 80 K as the thickness increases. Both T_{max} and T_{min} also have the same period of about 20 Å but with opposite phase in the case of T_{min}. We see that there is a correspondence between the changes of T_N, T_{max} and T_{min} below 164 Å. At around the temperatures where the T_N versus thickness curve shows a dip, the temperature range between T_{max} and T_{min}, where the 2/7 spin phase is expected to change to the 3/11 spin phase, becomes narrower. For the samples without T_{max} and T_{min}, like the sample of 144 Å Er, this phase transition may have disappeared or taken place in a very narrow temperature range. The suppression of this phase transition seems to happen with a period of about 20 Å.

In order to clarify the influence of the structural change on the magnetic property, the *c*-axis lattice constants at room temperature and the Néel temperatures are plotted in figure 5 as a function of the thickness of Er thin films. We see that the *c*-axis lattice constant at room temperature shows the same periodic change as T_N with a period of about 20 Å and there is a correlation in the change of the lattice constant and T_N. This correlation is shown more clearly in figure 6, where the data point for bulk Er is also plotted. The

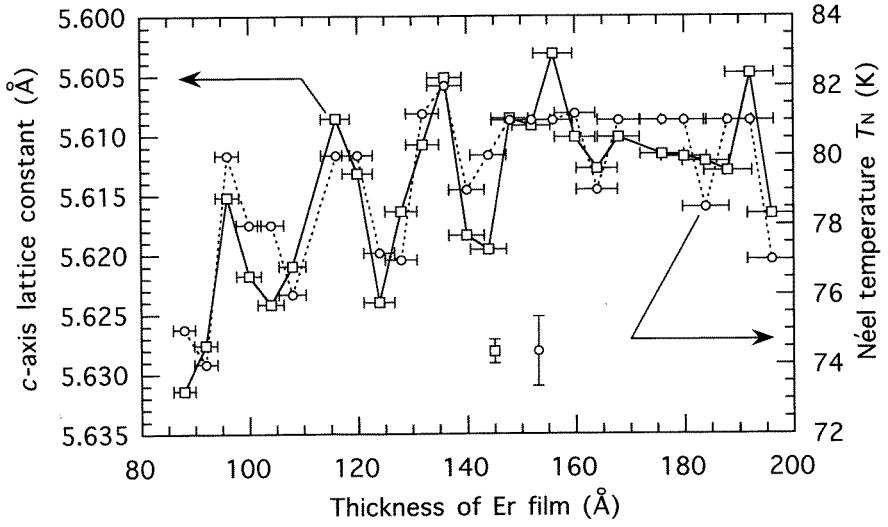

Figure 5. The variations of the *c*-axis lattice constant and the Néel temperature T_N as a function of the thickness of Er film. The lattice constant was determined from the room-temperature x-ray scans. Typical error bars for the vertical direction are separately shown for clarity. Note that on the left vertical axis numbers are given in descending order.

data for our Er thin films seem to extrapolate to that for bulk Er. The Néel temperature is found to become lower as the *c*-axis lattice constant increases. It is clear that the periodic change observed in the magnetic phase transition temperatures is accompanied by that in the structure. It is expected that, as the lattice constant increases, the strength of the indirect exchange coupling between the Er magnetic moments decreases. This causes the lowering of T_N. The suppression of some spin phases and the change in the temperature dependence of spin phases may be explained in terms of a change in the exchange coupling as has been theoretically shown for the Ising model with competing interactions [7].

The overall monotonic decrease of the lattice constant as the thickness increases shown in figure 5 can be understood in terms of the relaxation of the epitaxial strain which comes from the lattice mismatch at the interface between Er and Nb. This relaxation effect on the lattice constant has been already reported in Er/Lu thin films [4]. The periodic change with a period of about 20 Å, however, is hard to understand in terms of strain relaxation only. If this periodicity comes from the periodic variations of the film morphology with alternating filled and incompletely filled atomic layers, we would have an oscillation period of one atomic layer as observed in Co films [8] and Fe/Au superlattices [9]. In our Er thin films, one atomic layer corresponds to about 2.8 Å. The period of 20 Å corresponds to about seven atomic layers, which is almost equal to the period of the intrinsic *c*-axis-modulated spin structure in the high-temperature phase. For the 2/7 and 3/11 spin phases, the periods are evaluated to be 7 (phase angle is 51.4°/layer) and 7.29 (49.4°/layer) atomic layers, respectively [6].

We discuss the periodic change of the lattice constant along the *c*-axis with film thickness. It is well established [10] that conduction electrons play an important role in determining the structure of a metal. Especially in heavy rare earths, conduction electrons have an influence not only on their structures but also on their magnetic properties, which are dominated by the localized magnetic moments of 4f electrons coupled via the

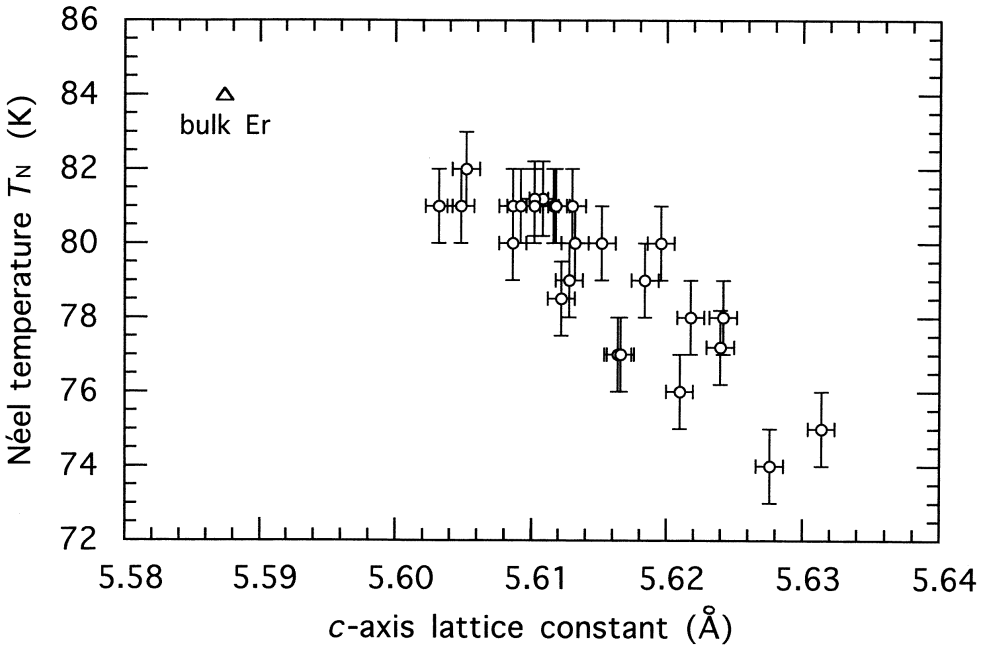

Figure 6. The correlation between the *c*-axis lattice constant at room temperature and the Néel temperature T_N. The triangle shows the data point for bulk Er.

Ruderman–Kittel–Kasuya–Yoshida (RKKY) indirect exchange interaction. In characterizing the conduction electrons, the Fermi surface topology plays a decisive role. The webbing structures of the Fermi surface for heavy rare earths provide some parallel surface sheets for nesting vectors which drive their magnetic ordering. The nesting vectors give the period of the modulated spin structure below T_N. Though Er has no long-range magnetic ordering at room temperature, it is expected that the nesting feature of the conduction electrons remains above T_N. For thin films, one possible origin of the enhancement in the nesting feature is the quantum-interference effect [11]. In the quantum-interference state, the rapidly oscillating Bloch functions of conduction electrons are modulated by a slowly varying envelope function. This envelope function gives the modulation period $2(k_{ZB} - k_F)$ of the distribution of conduction electrons, where k_{ZB} is the wave vector at the zone boundary and k_F is the Fermi wave vector. In the topology of the Fermi surface of Er, it is theoretically concluded that there are flat surface sheets perpendicular to the *c*-axis reciprocal vector [12, 13]. For these flat surface sheets, the period $2(k_{ZB} - k_F)$ in the quantum-interference state is equal to the period for the nesting vector between the flat surface sheets across the middle zone boundary. When the thickness of the Er film is a multiple of this period, the modulation structure in conduction electrons is stabilized via electron–lattice interaction. As the thickness of the Er thin film increases, therefore, the averaged lattice constant evaluated in the x-ray diffraction studies changes periodically with a period of seven atomic layers (about 20 Å) which is equal to the period for the nesting vector.

In summary, the magnetic phase transition temperatures and *c*-axis lattice constant of Er thin films were found to change periodically with a period of about 20 Å as a function of

the thickness. With the same period, some spin phases disappear. Though further detailed studies are necessary, this periodicity probably originates from the nesting feature in the Fermi surface of Er.

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