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LETTER TO THE EDITOR

Thermal hysteresis phenomena in Eu/Yb superlattices

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Abstract. Thermal hystereses resulting from different cooling conditions were observed in the magnetic properties of epitaxial BCC Eu/Yb superlattices. The magnetic-frozen states, characterized by the cusp in the temperature dependence of the magnetization, were observed for zero-field-cooled superlattices. On the other hand, the magnetization of field-cooled superlattices monotonically increased with decreasing temperature. Such phenomena became more distinct as the Eu interlayer distance in the superlattices decreased. Strong magnetic-field dependence, in which the frozen state weakened and the frozen temperature fell with increasing magnetic field, was also present in the zero-field-cooled Eu/Yb superlattices.

Unique magnetic properties associated with an artificial superstructure are expected in metallic superlattices utilizing rare-earth elements. This is because the magnetic coupling between the 4f spins, which is mediated indirectly through the 6s and/or 5d conduction electrons, is effective over a relatively long range compared with that of d-spin systems. Recently, it has been reported that amorphous Eu/Mn superlattices with atomic-scale periodicity exhibited magnetic properties with some modulation effects due to superperiodicity [1]. We have suggested that the Eu–Mn interfacial interaction and the Eu interlayer interaction constitute the origin of this magnetic behaviour [2]. However, developing a definitive model for the magnetic mechanism occurring in Eu/Mn superlattices is very difficult because two kinds of spins, such as the 4f of Eu and the 3d of Mn, are present and the individual metal layers are in amorphous states. This idea prompted us to study the more simplified case of epitaxial BCC Eu/Yb superlattices [3] containing the non-magnetic metal Yb.

In the present work, Eu/Yb superlattices with various periods were fabricated to provide further insight into the magnetism of superlattices containing rare-earth metals. We report here the magnetic properties of Eu/Yb superlattices as a function of the Eu interlayer distance, Eu thickness, and magnetic-field intensity.

The epitaxial BCC Eu/Yb superlattices were grown at 290 K by a molecular-beam method. The period of the superlattices was repeated 25 times. The details of the preparation have been described previously [3]. The DC magnetization was measured using a superconducting quantum interference device (Hoxan HSM-2000) between 4.2 and 150 K. Samples were placed parallel to the external applied magnetic field to measure the parallel magnetization ($M_{\parallel}$). An applied static magnetic field of up to

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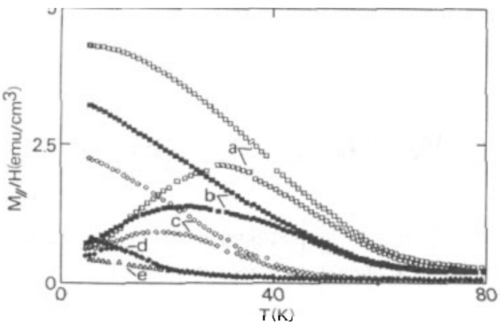

Figure 1. Temperature dependence of parallel magnetizations for epitaxial Eu (20 Å)/Yb superlattices with various Eu interlayer distances measured under 50 G for field-cooled and zero-field-cooled samples. Eu interlayer distances (Å) are 10 (a: □), 20 (b: ■), 40 (c: ◇), 50 (d: ◆), and 70 (e: △).

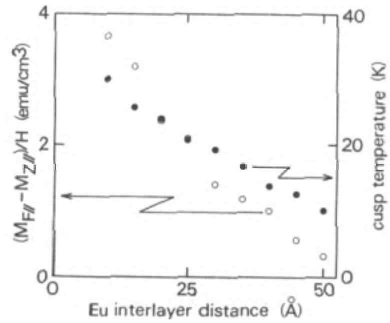

Figure 2. Thermal hysteresis (○) and cusp temperature (●) versus Eu interlayer distance for epitaxial Eu (20 Å)/Yb superlattices.

±5000 G was used. All magnetic data were corrected for magnetism arising from the Au protective layer, Yb component, NaCl substrate, and gelatine sample holder.

The temperature dependence of the parallel magnetizations for epitaxial Eu/Yb superlattices with various Eu interlayer distances is shown in figure 1. It is important to realize that the Eu interlayer distance is defined by the Yb spacer thickness. The Eu thickness of these samples was held constant at 20 Å. The magnetic data were obtained in a magnetic field of 50 G for field-cooled (under 50 G) and zero-field-cooled samples. Thermal hystereses resulting from different cooling conditions were observed for these samples. The field-cooled superlattices showed a monotonic increase in the magnetization at low temperature. On the other hand, the magnetic-frozen states, characterized by the cusp in the temperature dependence of the magnetization, were observed for the zero-field-cooled superlattices. It is worth noting that this unique magnetic behaviour systematicaly changed with increasing thickness of the non-magnetic Yb layers. That is, the absolute values of magnetization gradually became smaller as the Yb thickness increased, even though the total volume of 4f magnetic spins was the same in all of the superlattices. The thermal hysteresis, which is estimated by the difference between the magnetizations of field-cooled ($M_{F\|}$) and zero-field-cooled ($M_{Z\|}$) samples, also became smaller as the Eu interlayer distance increased, as shown in figure 2. Moreover, the frozen temperature, as defined by the cusp position, decreased with increasing Eu interlayer distance as shown in figure 2. As a result, the thermal hysteresis and cusp were not clearly detected in epitaxial Eu/Yb superlattices with Eu interlayer distances greater than 70 Å. From the results from the studies on the magnetic properties of the Eu/Mn [1, 2] and Eu/Yb superlattices, we can conclude that the individual Eu layers in superlattices are magnetically coupled to each other and that the Eu interlayer interaction contributes to the thermal hysteresis phenomena including the magnetic-frozen states. Here such a long-range interaction must be a RKKY-type interaction which is mediated by the 6s conduction electrons of Eu metal because the direct interaction between 4f spins which are localized in the inner core is negligibly small and is only effective over a short range.

Next we studied epitaxial Eu/Yb superlattices with various Eu thickness. Here the Yb thickness was fixed at 20 Å. As shown in figure 3, the magnetic properties of this

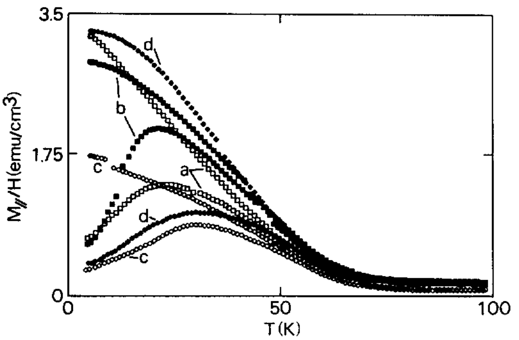

Figure 3. Temperature dependence of parallel magnetizations for epitaxial Eu/Yb (20 Å) superlattices with various Eu layer thicknesses measured under 50 G for field-cooled and zero-field-cooled samples. Eu layer thicknesses (Å) are 20 (a: □), 40 (b: ■), 60 (c: ◇), and 80 (d: ◆).

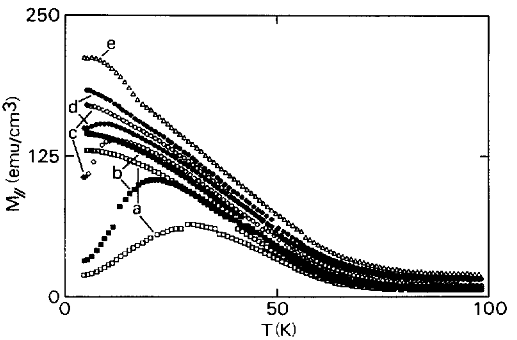

Figure 4. Temperature dependence of parallel magnetizations for the epitaxial Eu (40 Å)/Yb (20 Å) superlattice measured under various magnetic fields for field-cooled and zero-field-cooled samples. Applied magnetic fields (G) are 30 (a: □), 50 (b: ■), 100 (c: ◇), 150 (d: ◆), and 300 (e: △).

series did not systematically change with increasing Eu thickness. This is probably due to a complex combination of factors which affect the interaction between the Eu layers in different ways. Such factors might include the pseudo-two-dimensionality of the layers which become more bulk-like with increasing thickness.

Figure 4 shows the temperature dependence of the parallel magnetizations measured under various magnetic fields for the epitaxial Eu (40 Å)/Yb (20 Å) superlattice. The frozen temperature exponentially decreased as the magnetic field became stronger, as shown in figure 5. On the other hand, a critical magnetic field was observed for the change in the thermal hysteresis, as shown in figure 5. Namely, the thermal hysteresis increased until the magnetic field reached 50 G, at which point it gradually decreased. Finally, the thermal hysteresis and cusp were not observed above 4.2 K under 300 G. This means that the Eu interlayer interaction which forms the thermal hysteresis possesses a strong magnetic-field dependence and is stopped when the magnetic field reaches 300 G. The shift of frozen temperature observed here is extremely big and has not been observed before for spin-glass materials. In spin-glass materials, the magnetic-field dependence has been evident as a broadening of the cusp, where the frozen temperature changes little during measurement under static magnetic fields [4, 5]. Therefore, it appears that the Eu/Yb superlattices are not spin-glass materials. This idea is supported by the lack of time-dependent change of the magnetic states, which normally occurs as one of the important characteristics of spin-glass materials [6].

A typical magnetization curve obtained at 4.2 K for an epitaxial Eu/Yb superlattice is shown in figure 6. The arrow indicates the starting point for the magnetic-field cycling on the sample which was cooled from ambient temperature to 4.2 K under 0 G. The observation of saturated and residual magnetization implies the presence of ferromagnetic components in the Eu/Yb superlattices. Here no elements that can act as ferromagnetic impurities were detected by energy dispersive x-ray spectroscopy and

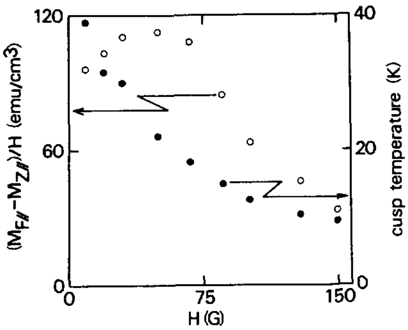

Figure 5. Thermal hysteresis (○) and cusp temperature (●) versus magnetic field for the epitaxial Eu (40 Å)/Yb (20 Å) superlattice.

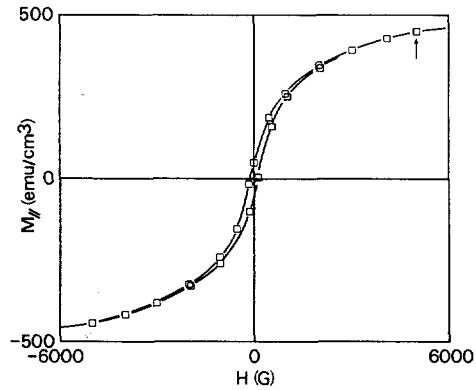

Figure 6. Parallel magnetization curve at 4.2 K for the epitaxial Eu (20 Å)/Yb (40 Å) superlattice.

Auger electron spectroscopy [3]. Therefore, the appearance of ferromagnetic behaviour here is due to the superlattice structure since Eu and Yb are antiferromagnetic and non-magnetic, respectively, in their bulk states. The extrapolated magnetization at 10 000 G was about 475 emu cm^{-3} at 4.2 K. The magnetic moment of the Eu can be estimated to be 2.46 μ_B based on this value. Comparison with the theoretical saturated magnetic moment (7 μ_B) indicates that about 35% of the Eu atoms are in a ferromagnetic state. Since such a big magnetic moment does not occur in the spin-glass state in diluted spin systems, the magnetic-frozen behaviour observed is associated with the inherent nature of the rare-earth superlattices.

In conclusion, the magnetization of field-cooled BCC Eu/Yb superlattices monotonically increased as the temperature decreased. On the other hand, the magnetic-frozen states, characterized by the cusp in the temperature dependence of the magnetization, were observed for the zero-field-cooled samples. Such thermal hysteresis, which reflected the difference in cooling conditions, became more distinct with decreasing thickness of the non-magnetic Yb layer. This result indicates that the unique magnetism observed in Eu/Yb superlattices is associated with the Eu interlayer interaction mediating by 6s conduction electrons. Strong magnetic-field dependence, in which the frozen state weakened and the frozen temperature fell with increasing field, was also observed in the zero-field-cooled Eu/Yb superlattices. Since there was no time dependence observed for the magnetic states of Eu/Yb superlattices, the observed magnetism is associated with the inherent nature of rare-earth superlattices and is not believed to be a spin-glass characteristic.

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