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LETTER TO THE EDITOR

Magnetic-frozen states in Eu/Mn superlattices

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Abstract. Eu/Yb and Mn/Yb superlattices, in which the Yb is a non-magnetic element, were fabricated by a molecular beam method in order to provide further insight into the magnetic properties of Eu/Mn superlattices. The magnetic susceptibility and magnetisation measurements made on the experimentally prepared superlattices indicated that the unique magnetic behaviour of Eu/Mn superlattices, as typified by the magnetic-frozen state and unidirectional anisotropy, was associated with the amorphous Eu interlayer interaction and interfacial interaction between the amorphous Eu and amorphous Mn layers.

The magnetic properties of metallic superlattices have attracted much attention in both fundamental and practical fields [1]. Most studies reported to date have focused on interface magnetism [2, 3] and magnetic anisotropy [4–7] of d-spin superlattices, including ferromagnetic elements such as Fe, Co and Ni. However, no magnetic modulation effect due to artificial periodicity has been observed in these systems. In contrast, new magnetic properties associated with a superstructure are expected in superlattices utilising rare-earth elements. This is expected because the magnetic coupling between the 4f spins, which is mediated indirectly through the conduction electrons, is effective over a relatively long range compared to that of d-spin systems. In fact, magnetic modulation effects introduced by superperiodicity have been observed in rare-earth epitaxial superlattices, such as Gd/Y [8–10], Dy/Y [11], and Er/Y [12]. Recently, we have found that amorphous Eu/Mn superlattices with atomic-scale periodicity possessed spin-glass-like behaviour, in which the magnetic-frozen state appeared in zero-field-cooled samples [13]. However, this magnetic behaviour was not observed in the Eu/Mn superlattices with long periods, in which amorphous Mn (a-Mn) and [110]-oriented crystal Eu (c-Eu) (this grew over the amorphous Eu (a-Eu) layer when the Eu layer thickness was larger than about 25 Å [13]) layers became thick. Therefore, a model in which the Ruderman–Kittel–Kasuya–Yosida interaction exists between amorphous Eu layers was proposed to explain the observed magnetic properties.

In the present work, amorphous Eu/Yb and Mn/Yb superlattices, in which the Yb is a non-magnetic element, were fabricated in order to provide further insight into the magnetic properties of amorphous Eu/Mn superlattices. We report here the results of detailed magnetic studies on the prepared superlattices.

The component metal layers were deposited alternately at the rate of 0.2 Å s^{-1} under ultra-high vacuum at 9×10^{-10} Torr using Knudsen cells in the molecular beam epitaxy apparatus (base pressure 7×10^{-11} Torr). Metallic Eu (99.99%), Mn (99.999%), and

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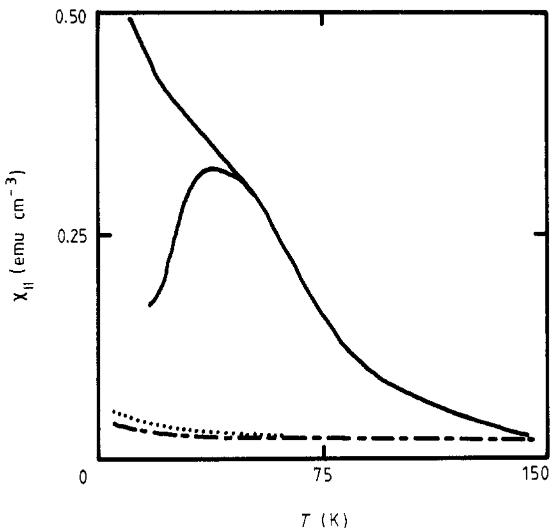

Figure 1. Temperature dependence of parallel magnetic susceptibilities for [Eu20/Mn20]25 (———), [Eu20/Yb20]25 (· · · ·), and [Mn20/Yb20]25 (·—·) as measured in a magnetic field of 500 G.

Yb (99.99%) were used as the source materials. The growth temperature was kept at about 260 K, except when otherwise indicated, in order to suppress interlayer diffusion. The period of the superlattice was repeated 25 times for each sample. The details of the preparation have been described previously [13]. X-ray diffraction (XRD) using a Rigaku CN2013 diffractometer and *in situ* reflection high-energy electron diffraction (RHEED) with a 30 keV electron beam were used to analyse the structures of the superlattices. Energy dispersive x-ray spectroscopy (EDX, using a Tracor Northern TN5500) and Auger electron spectroscopy (AES, Jeol JAMP-10S) were used to check impurity levels. The DC magnetic susceptibility and magnetisation curves were 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 in order to measure the parallel magnetic susceptibilities. The applied static magnetic field was up to ±5000 G. All magnetic data described below were corrected for diamagnetism arising from the Si substrate and the gelatine sample holder.

Intense and sharp diffraction peaks, accompanied by higher-order reflections, due to the artificial periodic structure were observed in small-angle XRD patterns of all samples. This indicates that the superlattices have been successfully grown with the formation of smooth and clear interfaces. The amorphous structure of each layer was confirmed by the halo pattern observed by *in situ* RHEED and by the broad nature of the middle-angle XRD peaks.

The temperature dependence of the parallel magnetic susceptibilities of Eu/Mn, Eu/Yb and Mn/Yb superlattices are shown in figure 1. The thickness of the individual layers was held constant at 20 Å. A superlattice of amorphous Eu/Yb was prepared at a growth temperature of 90 K, because this system crystallised in the synthesis at 260 K. Unique magnetic behaviour reflecting the difference of the cooling condition, as seen in the Eu/Mn superlattice, was not observed in Eu/Yb or Mn/Yb superlattices. That is, a remarkable increase in the magnetic susceptibility was not observed in the field-cooled samples. The zero-field-cooled samples also did not show any magnetic-frozen states. This finding leads to the conclusion that the unique magnetism is associated with the inherent nature of the amorphous Eu/Mn superlattices.

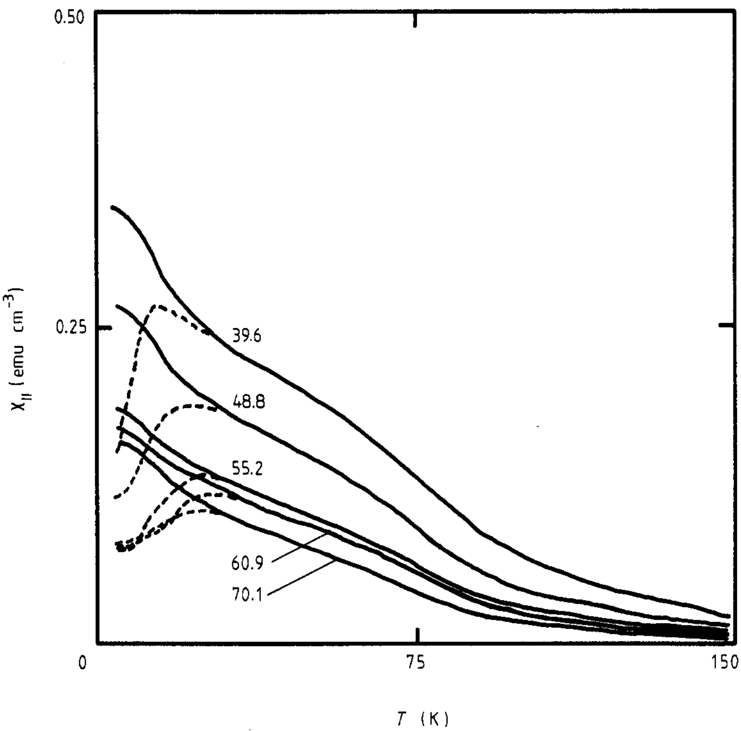

Figure 2. Temperature dependence of the parallel magnetic susceptibilities for Eu/Mn superlattices with various Eu interlayer distances (Å) as measured in a magnetic field of 1000 G for field-cooled (full curve) and zero-field-cooled (broken curve) samples. The periodic thicknesses are described.

Figure 2 shows the temperature dependence of the parallel magnetic susceptibilities measured for the Eu/Mn superlattices, in which the Eu layer thickness is held constant at 20 Å and the thickness of the intervening Mn layers are varied from 20 to 50 Å. The magnetic susceptibility obtained under 1000 G for field-cooled and zero-field-cooled samples are described by the solid and broken lines, respectively. The cusp is observed in the magnetic susceptibilities of all zero-field-cooled superlattices. This indicates that these materials are in a magnetic-frozen state at low temperature. Note that the cusps observed become more well defined as the Eu interlayer distance (equivalent to the Mn layer thickness) decreases. On the other hand, the absolute value of the magnetic susceptibility obtained for field-cooled samples decreased exponentially with increasing Eu interlayer spacing, as shown in figure 3. The Mn layers themselves are not likely to play a role in the observed magnetism because the magnetic susceptibilities decreased with increasing a-Mn layer thickness. Since the superlattices prepared possess the same Eu layer thickness and same periodicity number, all samples are equivalent as far as the contribution from each a-Eu layer. Therefore, we can conclude that a magnetic interaction between a-Eu layers, which is mediated by the 6s conduction electrons of Eu, is also responsible for the observed magnetism, in addition to the interfacial magnetic interaction between a-Eu and a-Mn layers. This can be supported by the experimental result that such unique magnetic behaviour was not observed in the Eu/Mn superlattices

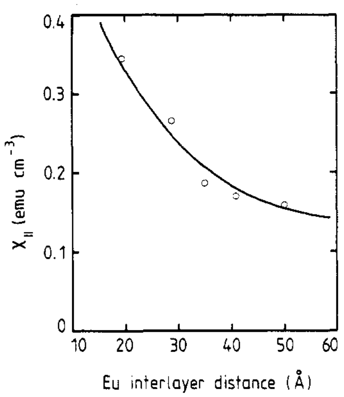

Figure 3. Parallel magnetic susceptibilities at 4.2 K of field-cooled samples versus Eu interlayer distance.

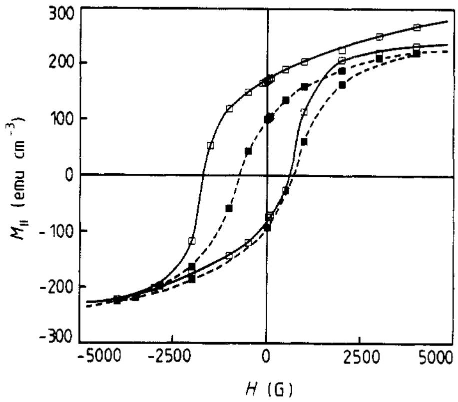

Figure 4. Parallel magnetisation curves at 4.2 K for field-cooled (□) and zero-field-cooled (■) [Eu20/Mn50]25.

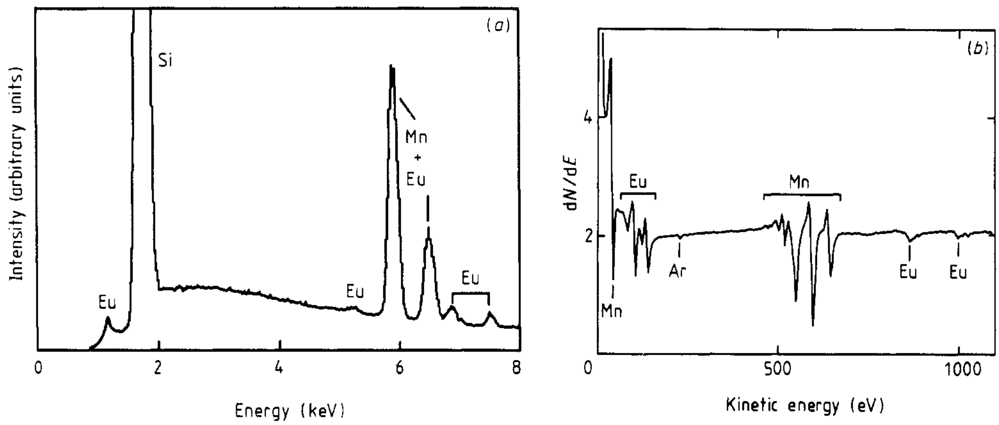

Figure 5. Typical (*a*) EDX and (*b*) AES spectra of Eu/Mn superlattices.

with long periods, in spite of the presence of an a-Eu/a-Mn interface [13]. That is, a long-range magnetic interaction [14, 15] between the a-Eu layers becomes stronger as the thickness of the 'spacer', such as the a-Mn and/or c-Eu layers, is decreased.

Typical magnetisation curves obtained at 4.2 K for amorphous Eu/Mn superlattices are shown in figure 4. The field-cooled samples were initially cooled under 5000 G. The observation of saturated and residual magnetisation suggests the presence of ferromagnetic components. However, no elements that can act as ferromagnetic impurities were detected by EDX or AES measurements, as shown in figure 5. Therefore, the appearance of ferromagnetism here is due to the superlattice structure, since Eu and Mn are antiferromagnetic in their bulk states. The hysteresis loops obtained for zero-field-cooled superlattices were symmetrical. In contrast, anti-symmetrical hysteresis loops [16] were observed for field-cooled Eu/Mn superlattices. This indicates that unidirectional anisotropy exists in this system. Since unidirectional anisotropy generally arises from the interlayer exchange interaction between ferromagnetic and antiferromagnetic components, the magnetic feature of Eu/Mn superlattices is that of mictomagnetism. The presence of magnetic-frozen states in zero-field-cooled samples can be also explained by this magnetism.

In conclusion, amorphous Eu/Mn superlattices with short periodicity cooled under zero-field possessed magnetic-frozen states at low temperature. Unidirectional anisotropy was observed in the hysteresis loop of the field-cooled samples. These magnetic behaviours were not observed in the amorphous Eu/Yb and Mn/Yb superlattices, nor in Eu/Mn superlattices with thick a-Mn and c-Eu layers. Therefore, it is believed that this magnetic behaviour is due to a mictomagnetic state associated with the a-Eu interlayer interaction and interfacial interaction between the a-Eu and a-Mn layers.

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