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ATOMIC STRUCTURE AND MAGNETIC PROPERTIES OF RARE-EARTH-IRON MULTILAYERS

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Abstract. – Mössbauer spectroscopy and magnetization measurements were carried out on Fe-RE multilayers where RE=Nd, Tm. Perpendicular and in-plane anisotropy direction for Nd-Fe and Tm-Fe multilayers was respectively observed. The atomic structure was studied by Mössbauer spectroscopy, small angle X ray scattering and transmission electron microscopy. The structure of the layers is built from pure iron and pure neodynium sandwiching a NdFe paracrystalline interface layer. We demonstrate that multilayer structure can be used to give a choosen magnetic anisotropy.

One of the principal aims in growing metallic superlattices is to create new materials with properties different from those of their constituent and alloys. But they can also be used to understand some fundamental properties of matter.

Recently Nd$_2$Fe$_{14}$B has been shown to exhibit important magnetic properties [1]. These magnetic properties are related to the magnetocrystalline anisotropy, but the details of the mechanisms are not clear. In this paper we show that rare-earth iron multilayers which modelize the crystalline structure of Nd$_2$Fe$_{14}$B permanent magnet which has a layer of Nd and B between three layers of iron [2], can be used to understand the origin of these anisotropy. We have choosen two different and opposite rare-earth:

a) neodynium which is a light rare-earth (ferromagnetic coupling with iron) and has a negative second order stevens factor $\alpha_2$;

b) thullium which is a heavy rare-earth (antiferromagnetic coupling with iron) and has a positive $\alpha_2$.

We report here the results of Mössbauer spectroscopy and magnetization measurements of the Fe-Nd and Fe-Tm multilayers.

The samples were prepared by the alternate evaporation of RE and Fe layers in an ultra-high vacuum chamber (10$^{-8}$ Torr during evaporation) onto a substrate held at 410 K which was the optimal temperature for these multilayers quality. Characterization of the samples regarding periodicity, interfaces quality and atomic structure were made by means of X-ray diffraction, electron microscopy and resistivity measurements. All films were continuous and microcrystalline. The RE layer thickness was kept constant: 38 Å for Nd and 36 Å for Tm while Fe layer thickness was varied between 13 Å and 31 Å. The total thickness of each sample was about 1 µm. Mössbauer spectroscopy and magnetization measurements were carried out on RE-Fe multilayers. A Foner type magnetometer was used for magnetization measurements. Figure 1 shows the Mössbauer spectra for two Nd-Fe and Tm-Fe samples with the same Fe layer thickness of 31 Å. Note the different velocity scales.

Fig. 1. – Mössbauer spectra of Nd-Fe and Tm-Fe multilayers at $T = 4.2$ K and $H = 0$ for the samples with Fe layer thickness of 31 Å. Note the different velocity scales.
are parallel to the film plane in Tm-Fe. The spectra are very different in the two cases. In the 31 Å Nd-Fe sample, pure Bcc iron can clearly be detected and represent 60% of the total iron, the rest (40% or 12 Å) are in a more distributed neighbourhood. In the 31 Å Tm-Fe samples 60% of iron are in a magnetic site but the neighbourhood seems to be more distributed. X rays diffraction at small angle [3] gives three peaks and can be simulated [4] with an interface width of about 10 Å, consistent with Mössbauer result if one attribute the non Bcc iron in NdFe to interface iron atoms. Transmission electron microscopy shows well defined rare-earth and iron crystals about 100 Å width. Therefore one can conclude that the structure of the multilayer is made of pure iron, pure rare-earth and a paracrystalline interface [5] in between. Figure 2 shows the hysteresis loop at 4.2 K of Nd-Fe and Tm-Fe, one can see again that Nd-Fe sample exhibit strong perpendicular anisotropy and Tm-Fe sample inplane anisotropy.

Fig. 2. – Hysteresis loops of Nd-Fe and Tm-Fe multilayers at \(T = 4.2\) K for the samples with Fe layer thickness of 31 Å and different applied field direction.

As in Fe-RE compounds a ferromagnetic coupling of 3d-4f moments in Nd-Fe and antiferromagnetic coupling in Tm-Fe multilayers were found. On the basis of Fe hyperfine field distribution a mean magnetic Fe moment was obtained using the usual conversion factor of 145 kOe/\(\mu_B\) [6]. For the Nd-Fe system the iron moment was found to be \(\mu_{Fe} = 1.74\) and 2.1 \(\mu_B/\text{at.}\) for Fe layer thickness of 13 Å and 31 Å, respectively. For Tm-Fe, \(\mu_{Fe} = 0.6\) and 1.2 \(\mu_B/\text{at.}\) for iron thickness of 13 Å and 31 Å respectively. One can see that the antiferromagnetic coupling in Tm-Fe leads to some frustrated situated at the Tm-Fe interfaces and then to an iron moments reduction. The opposite behavior of the anisotropy in the two systems is also interesting. Two different phenomena have been observed in the past concerning anisotropy in thin films:

a) in soft ferromagnetic materials, the demagnetizing field produces in plane anisotropy [7];
b) in this films of rare-earth and transition metals alloys, perpendicular anisotropy is generally observed; the explanation of these results is related to pairing effect [8].

We believe that in our case, a different phenomenon plays a major role: the interface crystal field. This crystal field induce a big positive second order term on the rare-earth (\(A_2^0\) in the Hutchings notation [9]). This second order term being dominant over the higher order term existing in the hexagonal crystals of neodymium and thulium. Therefore the moment direction in rare-earth is determined by \(\alpha_s\) and the moment direction in iron (determined by the exchange coupling between iron and rare-earth), is parallel or perpendicular to the film plane.

In conclusion, we have demonstrated in this study that a natural explanation of strong anisotropy in layered intermetallics can be the existence of an interface crystal field.