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Hydrogen-induced change of the magnetic coupling in Nb/Fe multilayers

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Abstract

Nb/Fe multilayers, consisting of typically 10–20 doublelayers with individual layer thicknesses in the nanometer range, were investigated by neutron reflectometry, SQUID magnetization and magnetoresistivity measurements. The main purpose of this investigation was to study the influence of hydrogen on the magnetic properties of these systems. We find that the magnetic coupling of the Fe layers over the nonmagnetic Nb layers can be switched between antiferro- and ferro-magnetic by hydrogen charging. The change of the coupling upon hydrogen charging is continuous and reversible.

Keywords: Magnetic coupling; Multilayers; Hydrogen; Neutron reflectometry; Magnetoresistance

1. Introduction

The magnetic coupling of Fe layers in Nb/Fe multilayers oscillates between ferro- and antiferro-magnetic coupling as a function of the Nb spacer layer thickness. This had been reported in the literature [1,2] and was verified in our own neutron reflectivity measurements [3,4]. We find antiferro-magnetic superstructure peaks (as the one shown in Fig. 1) for Nb layer thicknesses in the range around 1.5 nm and 2.3 nm but not for thicknesses around 1.9 nm. This clearly demonstrates the oscillating behavior of the coupling. In a simple version the coupling energy is described by [5]

$$J \sim \frac{1}{(2k_F r)^2} \cdot \sin(2k_F r) \tag{1}$$

where k_F is the Fermi vector and r the spacer layer thickness.

The aim of the present experiment was to change the magnetic coupling not via the spacer layer thickness but via hydrogen charging assuming that hydrogen changes the electronic properties, in particular the Fermi vector, in the spacer layers. We will show here that this is indeed possible.

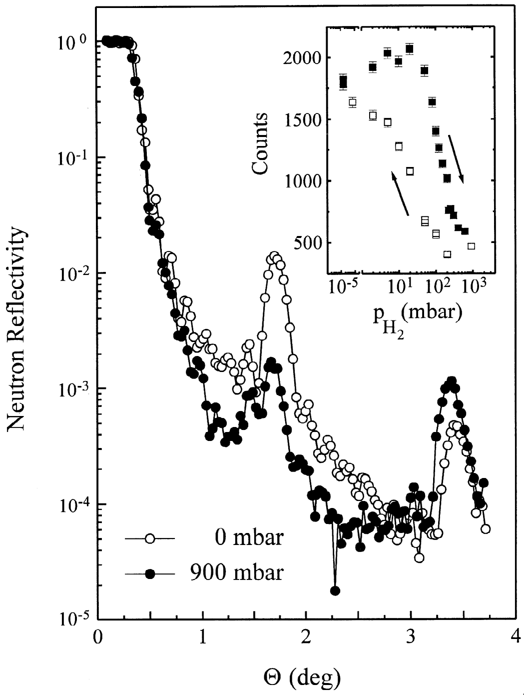

Fig. 1. Neutron reflectivity measurements on a Nb/Fe multilayer with the composition [1.5 nm Nb/2.6 nm Fe]×18. Open circles are for the uncharged sample, closed circles for the same sample after hydrogen charging at 900 mbar at 200 °C. The insert shows the height of the antiferromagnetic peak for increasing and decreasing external hydrogen pressure in the second charging/decharging cycle. The measurements were performed with the hydrogen atmosphere around the sample.

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2. Experimental details and results

The samples were prepared by e-beam evaporation of the materials on Si wavers. We used a 5 nm thick Cr buffer layer between Si and the Nb/Fe layers to avoid interdiffusion and terminated the Nb/Fe stack by a 4–5 nm thick Pd cap to facilitate hydrogen uptake. The Fe layers were always 2.6 nm thick but the Nb layer thicknesses were varied. The sample for which the change of the coupling is reported here had a Nb layer thickness of 1.5 nm. Hydrogen charging was performed from the gas phase at 200 °C. The pressure of the external H_2 atmosphere was varied between 0 and 900 mbar.

Fig. 1 shows neutron reflectivity data for a [1.5 nm Nb/2.6 nm Fe]×18 sample before and after hydrogen charging. The peak at 3.4 degree is due to the structural periodicity of the multilayer. The peak shift and the intensity change upon hydrogen charging are a clear indication of hydrogen uptake. Assuming that hydrogen is only in Nb (see Ref. [6]), the fit yields an expansion of the Nb layers by 0.05 nm (3.3%) and a hydrogen concentration of $[H]/[Nb] \approx 25\%$ in the charged state.

The superstructure peak at 1.7 degree in Fig. 1 is due to the antiferro-magnetic ordering of the Fe layers (ferromagnetic within the layers but opposite directions in adjacent layers). It can be seen from Fig. 1 that the intensity of the antiferro-magnetic (AF) peak is strongly reduced (note the logarithmic scale) after hydrogen charging. The height of this peak as a function of the external hydrogen pressure around the sample is displayed in the insert of Fig. 1 for increasing and decreasing hydrogen pressure, respectively. The reduction of the peak height is the main evidence for our conclusion that hydrogen changes the magnetic coupling (see Section 3 below).

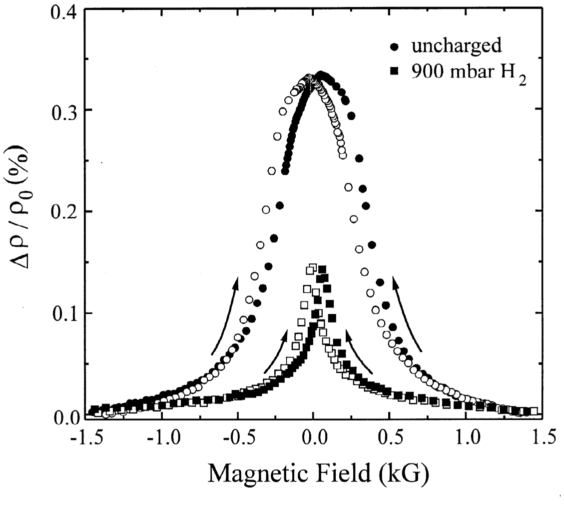

Fig. 2. Magnetoresistance (relative change of the resistivity $\Delta\rho/\rho$ in an applied magnetic field) for the sample [1.5 nm Nb/2.6 nm Fe]×18. Circles are for the measurement before hydrogen charging, squares after hydrogen charging at 900 mbar (in situ), open symbols for increasing, closed symbols for decreasing field.

Further evidence comes from magnetization and magnetoresistance measurements. The magnetization hysteresis curves (not shown here) of the uncharged sample have the typical shape of antiferromagnetic materials, i.e., a gradual increase of M over H, a large saturation field, and a small remanence. The hysteresis curves of charged samples are ferro-magnetic-like, i.e., they have the form of a square loop with a small saturation field and a high remanence. The magnetoresistance (see Fig. 2) of the uncharged sample is considerably larger than that of the hydrogen charged sample. This shows that the resistivity contribution from the antiferro-magnetic ordering (''giant'' magnetoresistance) is reduced or has disappeared in the charged sample.

3. Discussion and conclusion

The reduction of the intensity of the antiferromagnetic peak in Fig. 1 shows that the antiferromagnetic ordering in the sample has been reduced by hydrogen charging but it does not necessarily mean that the ordering has been changed to ferromagnetic. However, we know from other experiments [3,4] that, due to large pinning effects, the ordering remains as it is, except if internal or external forces drive it in an other direction.

For example, Nb/Fe multilayers with $d_{Nb} \geq 1.9$ nm where the exchange coupling is weaker than in the present case, hydrogen charging has no effect on the magnetic ordering, i.e., the ordering of the uncharged sample (ferro- or antiferro-magnetic) persists after charging and is not changed. In addition, if these samples are magnetized in an external field, they remain in the ferro-magnetic state after the field is switched off, even if the virgin sample was antiferro-magnetic. Thus in the present case where experimentally a change of the ordering is observed (Fig. 1) the internal magnetic coupling J must have changed the sign.

The picture which evolves from these data is as follows: in the uncharged sample, the Fe layers are antiferromagnetically ordered in a columnlike domain structure. Hydrogen charging changes the sign of the exchange coupling and reverses the ordering in most of the domains. Some antiferro-magnetism persists due to local inhomogeneities, either in the coupling or in the pinning. The large hysteresis in the charging/decharging cycle (insert of Fig. 1) is due to the magnetic pinning since the ordering is reversed only after the exchange coupling energy surmounts the pinning energy.

We suggest that the change of the exchange coupling upon hydrogen charging is caused by a change of the effective Fermi vector in Nb. (The influence of the layer expansion is minimal since it is only 0.05 nm whereas the oscillation length of the magnetic coupling is 0.9 nm [2,3].) A change of the Fermi vector with hydrogen incorporation is generally conceivable but a quantitative understanding is complicated considering the complex band structure of Nb.

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