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Magnetically Remanent Single Layer Neutron Polarizers

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A study was carried out to determine the optimal sputtering parameters for the production of single layer neutron polarizers consisting of a $Co_{60}Fe_{40}$ layer with a high remanence on an anti-reflecting layer. In a triode sputtering machine the working gas pressure, the substrate bias, and the angle of incidence of the atoms onto the substrate were varied. Annealing procedures were applied to some of the samples. Characterization was done by neutron scattering, SQUID magnetometry, x-ray diffraction, transmission electron microscopy, and Rutherford backscattering. Conditions were determined under which the polarization values of the samples lay between 89 and 99% above $0.3\,\theta_c$ in a parallel field of 3mT, and between 80 and 95% above $0.4\,\theta_c$ for opposite fields of 3mT, sometimes showing flipping fields as high as 15mT.

KEYWORDS: polarizers for neutrons, neutron optics, magnetic layers

Introduction

Using remanent neutron polarizers and analysers is a novel possibility in polarized neutron work[1], which has not yet been realized in full-scale experiments. One potential application is to use prepolarizing guides before a transmission cavity polarizer[2,3] in order to push the threshold wavelength for polarization to lower values. In this case the prepolarizer has to be magnetized opposite to the guide field to avoid the necessity of using a broad band, large cross section spin flipper between polarizing guide and the transmission polarizer. Another important application is for building large solid angle analyser systems. Here the goal is to avoid the use of large gap magnetizing coils, which would produce large stray fields, particularly disturbing in spin echo spectroscopy. This can be achieved with coatings in which saturation can be maintained by a small guide field in the mT range. We report on the development of sputter deposition techniques and performances achieved with single layer coatings for these two kinds of uses.

Experimental

The layers were prepared in a triode sputtering machine[4] with two fixed targets in between, of which a rotatable substrate holder was placed. The distance between target and substrate is 150 mm.

The above mentioned alloy was chosen because it has for the spin down component a slightly negative scattering length density. Thus the critical angle remains an imaginary quantity until the magnetization falls to 90% of the saturation value.

The guide was made of polished borkron glass, the standard material for neutron guides, which contains about 13 weight% of B_2O. For the analyser 1mm thick float glass was used because it is much cheaper and sufficient for this application. However, nearly all the results, which are to be presented in Ref.5 refer to layers which were sputtered onto 3 mm thick float glasses with the dimensions of 500 mm x 200 mm. Inside the machine the substrates were placed with their longer side being perpendicular to the ground.

The thickness of the sputtered layers was controlled *in situ* by quartz balances, the sputter current on the target, *ex situ* by Rutherford backscattering (RBS), and x-ray reflection. The magnetic properties were studied by SQUID magnetometry. Hysteresis curves were taken for two perpendicular directions in the plane of the film which correspond to the vertical and horizontal directions inside the machine. The easy axis of magnetization lies always in the plane of the film. For the direction out of plane the saturation field B_s amounts to 2.5T.

The structural properties were characterized by transmission electron microscopy (TEM) and x-ray diffraction. Finally, the most promising samples and later all coated glasses for the production of the guide were tested with neutrons at the reflectometer V6 at BENSC.

Since structural and magnetic properties strongly depend on the production parameters, we varied working gas pressure, substrate bias, magnetic field, and angle of incidence of the atoms onto the substrate. It was also checked what effect the addition of small amounts of tantalum could have. Afterwards annealing procedures were applied to some of the samples.

Gas pressures

The pressure of the argon gas was varied in the range from 0.05Pa to 1Pa where ten different values were used. In this pressure range the number of collisions occurring between argon atoms and sputtered atoms on the way from target to substrate varies from about one to twenty.

While the layer thickness did not depend strongly on the gas pressure, the magnetic properties showed remarkable effects. For pressures up to 0.1Pa the saturation fields were slightly above 0.1T, the remanences at 50% and the coercive fields (H_{cf}) around 20mT. At higher pressures H_{cf} fell to around 5mT with remanences sometimes above 90%. Unexpectedly, at particular pressures magnetic anisotropies were found. In the pressure region ranging from 0.3 - 0.5Pa the easy axis was in the vertical, and ranging from 0.7 - 0.8Pa it was in the horizontal direction.

Substrate Bias

The sputtering machine allows us to apply either dc or rf bias to the substrate. This accelerates argon ions to the surface of the growing film whereby loosely bound atoms, e.g., from gases, are preferentially removed and the growth is influenced by the increased amount of energy available at the surface.

As expected, the growth rate of the layers was reduced when a bias was applied and decreased to 50% at either 250V dc or 250W rf.

At a gas pressure of 0.1Pa, a rf bias ranging from 100 W and 500 W decreases H_{cf} from 22mT to 15mT, while it increases H_{cf} for pressures of 0.2Pa from 10mT to 20mT, and for 0.75Pa from 5 to 18mT. Thus the final value is the same for these pressures, i.e., the structure of the growing film is determined by the bias and not by the energy of the arriving atoms.

A dc bias in the range of 100 Vto 200V showed similar but not so pronounced effects.

However, neither an anisotropy was induced nor a high remanence reached in any of these bias experiments.

Oblique incidence

It is well known that magnetic anisotropies occur in films grown from atoms with oblique incidence on the substrate. Our machine allows us to rotate the substrate holder. Thus it can be turned at different angles relative to the targets. However, since the targets are fixed, the distance between the target and the substrate differs, depending on the angle, by a factor of two to four over the horizontal of the sample. Different solid angles of the target as seen from the substrate lead to different layer thicknesses. If the required properties depend on the thickness of the layers (as for supermirrors) this method can not be used. Since for single layer polarizers the thickness of the layers is less critical we produced layers with the substrate holder rotated to angles of 30° and 70° between the direction of the incident atoms and the normal to the surface of the substrate.

For a pressure of 0.06 (0.75)Pa we found a thickness ratio of the thick to the thin side amounting to 1.15 (1.4) at 30° and 3 at 70°.

At a pressure of 0.06Pa the anisotropy increased only slightly, while it was much stronger at 0.75Pa where values of B_S = 20 (80)mT, H_{cf}= 9 (4)mT, and remanence R = 96 (18)% were measured for the thin (66nm) and the thick (200nm) side, respectively . It is interesting to note that in this case at 0.75Pa the easy axis was in horizontal direction, i.e., perpendicular to the normally occurring direction. This means that the effect of oblique incidence is stronger than the - still unknown - effect which leads to the anisotropy occurring in our machine "naturally" at 0.75Pa.

Already in 1970 Okamoto et al.[6] explained the effect of oblique incidence by a tilt of the columns in which the material grows. Due to shadowing effects a structure develops with an easy axis perpendicular to both the direction of the incoming atoms and the surface normal. TEM pictures of our samples showed columns tilted by 10° from the surface normal at a sample rotation angle of 70°.

This also explains why the effect is stronger on the side of the substrate where the layer was thinner: due to the larger distance to the target the solid angle and hence the angular spread of the incoming atoms are smaller. This and also another experimental finding led us to further experiments where the angles of the atoms were better defined by placing a collimator between the target and the substrate.

Collimators

A collimator with a length of 50 mm and a slit width of 15 mm was constructed.

If installed in front of the substrate, strong anisotropies can be produced at all three tested pressures: 0.06, 0.2, and 0.75Pa. If the collimator is turned by 90°, the anisotropy turns as well. The easy axis was found to be always perpendicular to the direction of the slits.

In all cases remanences above 97% were measured with H_{cf} ranging from 1 to 10mT.

This is once again explained by the shadowing effect of growing islands or columns. If the angle of the incident atoms is restricted in one but not in the other direction, the structure grows anisotropically. By our collimator this angle interval is reduced from 70° in the vertical or 45° in the horizontal direction to 9° perpendicular to the slit direction while it does not change in the other direction. After removing every other segment of the collimator the effect vanishes.

However, this whole setup reduces the growth rate by a factor of nearly five and leads to an inhomogeneous layer thickness due to the shadows of the segments. If the collimator is moved a bit into the direction of the target to reduce this inhomogeneity, the anisotropy is quickly destroyed. This phenomenon even occurs at a pressure of 0.06Pa and at a distance of 25 mm from the substrate, although the transmitted angle interval is not altered. Nearly no additional collisions occur; otherwise the flight direction might be altered since the mean free path of the atoms is about 100 mm.

Thus, further development is necessary before this effect can be utilized in our machine. It should be noted here that, due to the line source character of the sputtered trench, this effect occurs "naturally" in magnetron sputtering machines with a moving substrate.

Addition of tantalum

The addition of small amounts of tantalum increases the magnetic hardness of iron.[7] We added tantalum during the sputtering process at four different pressure values. The amount of tantalum added was determined after the sputtering by RBS to be 1.0 ± 0.1 at%.

By adding tantalum, the saturation magnetization was reduced by nearly 10% and the saturation field significantly increased to 0.2T. Most strikingly, the normally occurring anisotropies vanished. The other values were: H_{cf} = 100mT and R = 50%.

Since the saturation magnetization was already significantly lowered by adding no more than 1 at% Ta and the annealing of these samples (see below) did not lead to better results either, this method was not pursued further.

Annealing

Annealing processes introduce additional energy into the material which can improve the crystal structure, remove obstacles for the movement of Bloch walls, and thus lower the coercivity.

The annealing was done by putting samples, cut for the SQUID measurements to an area with dimensions 4.5 mm x 4.5 mm, onto a thick steel plate in a temperature controlled oven. The coated side of the glass touched the plate to guarantee that the temperature would quickly reach the desired value and to reduce oxidation.

Annealing at 150° C for 100 h already showed the expected behaviour but only for samples produced with bias: H_{cf} was reduced by about 20%, R increased by 5 to 10%. No oxidation was detected.

However, due to oxidation, annealing at 250°C for 68 h led to a saturation magnetization reduced by 5%. The layers produced with rf bias oxidized most quickly, followed by those with dc bias and those without bias. For all samples the coercivity decreased from 15 to 20mT to around 1mT. Within the first 2.5 h the remanence increased by 30 to 50% and after 68 h roughly decreased to the initial value.

Annealing in vacuum of one of these samples led to the same saturation magnetization as before annealing, but to a reduction of H_{cf} from 24 to 1.5mT.

Annealing of ten different samples at 250° C for 68 h in a magnetic field of 66mT showed no other effects compared to annealing without a magnetic field. Sputtering in magnetic fields between 25 and 95mT also showed no additional effects at two different sputtering pressures compared to sputtering without field.

The samples with tantalum added were annealed for 4 and 18 h at 200°C, again with and without a magnetic field. After 18 h the saturation magnetization did not decrease. However, in these totally isotropic samples a strong anisotropy developed in vertical direction at production pressures of 0.5 and 0.75Pa. Again, whether a magnetic field was applied or not during annealing did not make a difference.

Production

After more than 500 SQUID measurements described above we checked the most promising layers with neutrons. It turned out that only the layer sputtered at 0.4Pa fulfilled the requirements for the prepolarizing guide. The first sample showing the proper qualities was remeasured half a year later and the reflectivity was still found to be the same, which leads to the assumption that the remanence does not decay within this period of time.

Finally we started to coat the glasses which will be used for the prepolarizing guide at the position IN 15 at the ILL, Grenoble with an antireflecting layer of 300 nm $Gd_{16}Ti_{84}$ and the polarizing layer of 150 nm $Co_{60}Fe_{40}$. Each piece of coated glass was tested on a neutron reflectometer. First each of the samples was magnetized in a field of -52mT. After reversing the direction of the magnetic field the reflectivity was measured in a field of 3mT. Figure 1 shows a typical example with the two spin components and the resulting polarization. In Fig.2 the reflectivity at an angle of 0.4° is given for different magnetic fields, revealing a flip of the magnetization at 13mT. Some samples did not change their direction of magnetization even at 15mT.

The neutron tests revealed that only about two thirds of the glasses showed the required properties while for the remaining third an insufficient remanence lead to an intolerably high spin down component. Until now we have

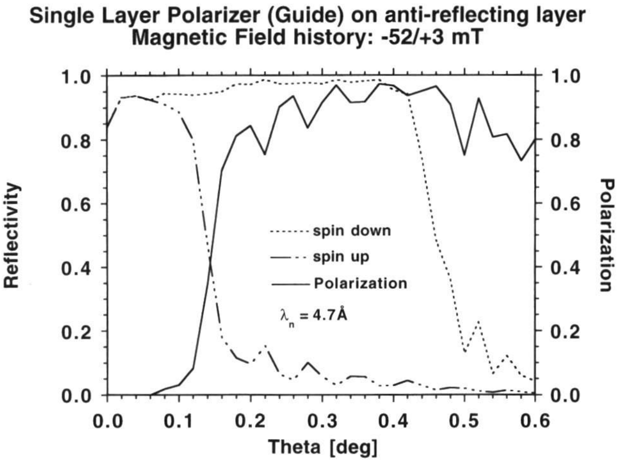

Fig. 1 Experimental reflectivities of a single layer polarizer on an anti-reflecting layer used for the prepolarizing guide for spin-up and spin-down neutrons and the corresponding polarization. After having magnetized the sample to -52mT a field of 3mT was applied during measurement.

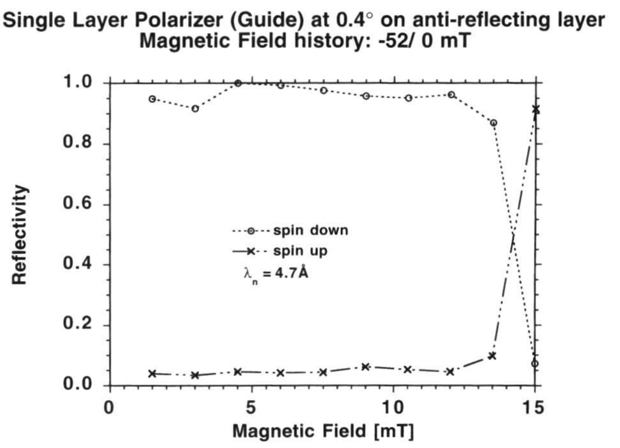

Fig. 2. Experimental reflectivities of a single layer polarizer on an anti-reflecting layer used for the prepolarizing guide for spin-up and spin-down neutrons at a grazing angle of 0.4°. The field was set to -52mT field before this measurement.

not found the reason of this scatter in properties. We tried twice to coat these glasses with insufficient magnetization again but gained no improvement. Finally we etched the layers away without damaging the surface of the glass and coated the pieces again. Luckily this series converged rapidly to zero.

The glasses for the analyser have not been coated yet but the reproducibility of the coating procedure has been tested several times. Figs.3~5 give the curves corresponding to the two figures above. The sample with the reflectivity curve shown in Fig.3 was in a second measurement measured at 60mT which results in virtually the same curve, i.e., the sample is fully remanent at 3mT. Fig.4 shows the behaviour of the layer after magnetization in an opposite field, indicating that a field of 20mT is needed to saturate the layer. Fig.5 reveals that a field of 3mT is already sufficient to keep the magnetization after application of a field in the same direction.

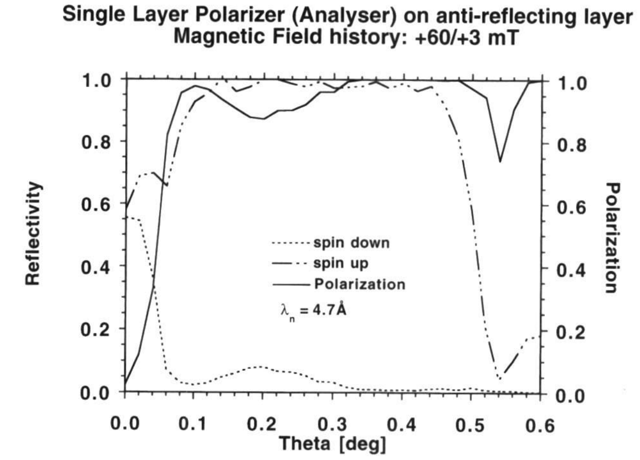

Fig. 3 Experimental reflectivities of a single layer polarizer on an anti-reflecting layer used for the analyser for spin-up and spin-down neutrons and the corresponding polarization. After having magnetized the sample to +60mT a field of 3mT was applied during measurement.

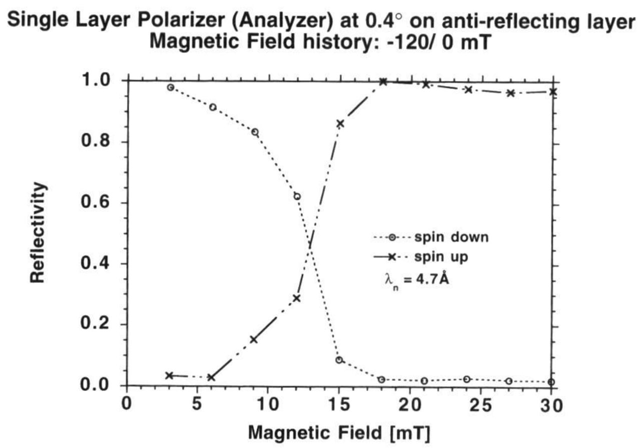

Fig. 4 Experimental reflectivities of a single layer polarizer on an anti-reflecting layer used for the analyser for spin-up and spin-down neutrons at a grazing angle of 0.4°. The field was set to -120mT before the measurement.

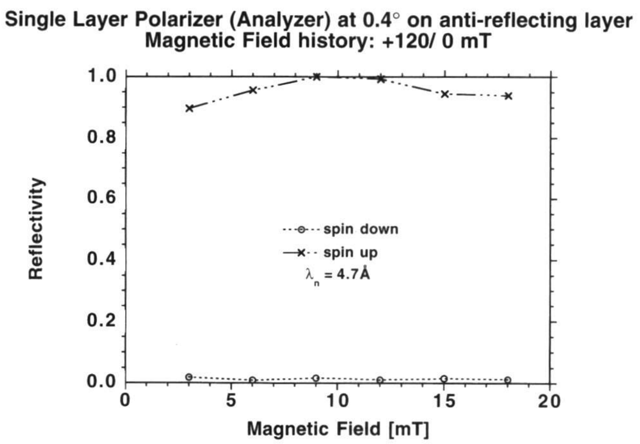

Fig. 5 Experimental reflectivities of a single layer polarizer on an anti-reflecting layer used for the analyser for spin-up and spin-down neutrons at a grazing angle of 0.4°. The field was set to +120mT before the measurement.

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