Ballistic magnetoresistance in a nanocontact between a Ni cluster and a magnetic thin film

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We present measurements of ballistic magnetoresistance in nanocontacts grown by electrodeposition of Ni microclusters on magnetic thin films covered by aluminum oxide layers, using a technique proposed by Schad *et al.* [D. Allen, R. Schad, G. Zangari, I. Zana, D. Yang, M. C. Tondra, and D. Wang, *J. Vac. Sci. Technol. A*, **18**, 1830 (2000); *Appl. Phys. Lett.* **76**, 407 (2000); D. Allen, R. Schad, G. Zangari, I. Zana, D. Yang, M. C. Tondra, D. Wang, and D. Reed, *J. Appl. Phys.* **89**, 6662 (2001)]. The measurements are made on single Ni clusters in contact with a Ni and Co thin film. We measure the magnetoresistance and observe the relaxation of the magnetization and electrical resistance as a function of time. The clusters are electrodeposited under several different experimental conditions. Some are deposited randomly on an unpatterned film and some through various patterned photoresists that control the location at which the cluster is grown. The typical contact size is estimated from the electrical resistance to be 10–30 nm. Ballistic magnetoresistance values up to 14% are obtained in these first experiments. © 2001 American Institute of Physics. [DOI: 10.1063/1.1413734]

Magnetoresistive devices and sensors are the basis for new applications in an area often called magnetoelectronics.¹ Recently it has been shown in atomic nanocontacts,² as well as in nanoscopic (scale ≈ 10 nm) electrodeposited contacts,³ of Co and especially of Ni that values of up to 700% ballistic magnetoresistance (BMR) can be achieved at room temperature in moderate applied fields. These contacts were grown between macroscopic wires. The ideal case for nanomagnetoelectronics capable of high levels of device integration will be contacts between nanoscale current leads. In this regard, there have been recent developments by Schad *et al.*⁴ showing that it is possible to grow Cu clusters by electrodeposition on top of aluminum oxide layers deposited on a magnetic thin film (MTF). Their approach is to apply a potential in the electrolyte solution between the electrode and the MTF. The potential causes an electrical arc or breakdown in the oxide. The breakdown appears to create a pinhole in the oxide, probably in preexisting thin spots or partial pinholes, and a Cu cluster electrodeposits on the underlying exposed MTF. In the present work, we have used this effect to make pinhole nanocontacts,^{5–7} between magnetic metals, with the aim of making nanocontacts that exhibit the BMR effect.^{2,3}

The best substrates we have found for the electrodeposition of Ni BMR nanocontacts consist of metallic multilayers such as: Si(100)/thermal oxide/1 nm Ta/100 nm Au/10 nm Ni or Co/1 nm Al, with the metallic films deposited in a UHV magnetron sputtering system. The purpose of the Au layer is to reduce the lead resistance to a negligible value compared to the nanocontact resistance. The best results are obtained

when the Al is oxidized by exposure to air. The electrodeposition is carried out in a solution of 1.5 M $\text{Ni}(\text{SO}_3\text{NH}_2)_2$ and 0.5 M H_3BO_3 at a pH of 3.5. The potential, referenced to a saturated calomel electrode, was -1.5 V for times varying from 5 to 60 s.

Ideally, it would be desirable to make BMR measurements on individual Ni clusters. However, the Ni clusters are randomly distributed on the substrate surface with a mean separation on the order of tens of microns. This close spacing of Ni clusters makes it difficult to make electrical contact to a single cluster. To assist in making electrical contact to a single cluster we have patterned a photoresist on some samples to open a regular array of holes 5–20 μm in diameter spaced several mm apart. In this way, only one Ni cluster is likely to be deposited in the exposed area of the sample at the bottom of each hole in the photoresist. No deposition occurs on the insulating photoresist. Electrical contact to the Ni cluster electrodeposited in the hole is made by attaching a Cu wire to it with silver paint. This method avoids any mechanical stress, which could damage the nanocontact.

The resistance observed for contact to a single cluster ranges from 0.2 to 80 Ω , essentially all of which is attributable to the nanocontact since the lead and contact resistances are negligible. We estimate the cross section of the contact by assuming that an atom occupies 0.1 nm² and that this has a quantum resistance of 12 900 Ω .⁶ We obtain values of the contact size between 3.5 and 75 nm, although the majority of them are around 10–30 nm. In some cases we have contacts much smaller than 1 nm.

The clusters are imaged with a field-emission scanning electron microscope (SEM) and an atomic force microscope (AFM). SEM analysis showed that the clusters have different structures: some resemble a cauliflower, as described in Ref.

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FIG. 1. (a) The sample geometry and the contacts made to measure the sample resistance through the pinhole nanocontact, (b) the SEM micrographs of the two kinds of growth (cauliflower and ring clusters), and (c) an AFM image and a topographic line of a ring of clusters.

4, and some have the appearance of ring clusters. Figure 1(b) presents SEM images of these two kinds of growth. AFM studies confirm the case of the ring structure, as in Fig. 1(c), in which the profiles as well as the topography are presented. Energy dispersive x-ray chemical analysis by the SEM indicated that the clusters were indeed Ni.

A general phenomenon that we observe (with few exceptions) during current flow is a relaxation or decay in the resistance of the nanocontact to a stable value on a typical time scale of 500 s. In a few cases, the resistance remained stable or increased. In several samples, the resistance returned to its initial value after the sense current was turned off overnight. When the sense current was turned on again, the relaxation appeared again. However, in all cases the relaxation to first approximation is independent of the applied magnetic field. Typical results are presented in Fig. 2(a). The initial resistance of 9.80Ω relaxes to 9.60Ω , while exhibiting a small BMR, as the magnetic field is cycled. The time dependence of the resistance is presented in Fig. 2(b). The relaxation at other applied fields is very similar. The relaxation is not understood, but it is found in nearly all samples we have studied, including the earlier work on electrodeposited contacts using macroscopic wires.³ However, in the earlier samples the relaxation time characteristically was longer, in some cases days.³ We believe the relaxation phenomenon is an important challenge for this field, and that an understanding of it will help greatly in the development of more stable contacts.

FIG. 2. (a) Resistance vs applied field for a sample with Ni as an underlayer and (b) the fitted relaxation of the resistance with time for the same data as above. In this case the BMR is about 1%.

We have investigated the BMR in the present samples for several reasons, one of which is for comparison with previous measurements of BMR in nanocontacts electrodeposited between two macroscopic wires. In that case, one might have argued that magnetostriction has some interplay with the observed magnetoresistance. However, in the present samples the geometry is completely different. In the present samples the Ni cluster diameter is a few microns, and the MTF is a few nanometers thick, having nanometric magnetic volumes. Since we observe very similar behavior in the magnetoresistance, magnetostrictive effects can probably be ruled out.^{2,3}

In Fig. 3 we present BMR measurements on the cauliflower-type clusters that have been deposited without a patterned photoresist. Here the number of clusters in the electrical contact is not limited to one. The sample structure is Si(100)\thermal oxide\2 nm Ta\5 nm Co\1.5 nm Al (oxidized by air exposure)\electrodeposited Ni. This case is of interest because we observed no time-dependent relaxation in the resistance and only a small field-dependent relaxation. The lower panel in Fig. 3 is the initial BMR-versus-field loop, with the arrows indicating the length of time that current has been flowing through the sample. The first loop is

FIG. 3. Resistance vs applied field for a sample with Co as an underlayer without a patterned photoresist. In this case no relaxation is observed and the BMR is $\approx 4\%$.

FIG. 4. Resistance vs applied field for a sample with Ni as an underlayer (upper graph). The data exhibit relaxation. The inset represents a linear fit to the time-dependent relaxation. The lower graph presents a plot of the resistance vs applied field after normalizing the data in the upper panel to the fit in the inset to present the plot that would be expected if there were no relaxation.

finished after 883 s. The upper curve is the loop recorded between 1816 and 2690 s. The loops are similar in resistance and BMR but the order of the resistance variation with field is inverted from the lower to the upper panel. The BMR is on the order of 4%. This small value probably occurs because this particular sample did not contain the 100 nm Au needed to make the lead resistance negligible and also because the contact is made with several clusters at once, reducing the total nanocontact resistance.

In Fig. 4 we present a more typical case in which there is both a resistance relaxation and a BMR effect.

The sample structure is: Si(100)\thermal oxide\1 nm Ta\100 nm Au\10 nm Ta\10 nm Ni\1.5 nm Al (oxidized by air exposure) electrodeposited Ni. The upper plot is the resistance versus magnetic field for a Ni MTF. The inset presents a linear fit to the time dependence of the resistance at the three zero-field crossings in the data. The nonlinearity of Fig. 2 is not observed in Fig. 4 because the relaxation is incomplete and still linear. Note that the slope is not zero at zero field. The linear fit is used to normalize the data to the value it would have had if there were no time dependence to

the resistance but only a field dependence. The result is presented in the lower graph. The lower graph shows a BMR of 14%. This value obtained for the BMR is of the same order as those published previously,² suggesting the same basic effect is present.

The major conclusions of this work may be summarized as follows.

- (1) We have observed BMR effects in a thin-film geometry in which Ni clusters are electrodeposited on pinholes in an aluminum oxide layer covering a MTF substrate.
- (2) The resistivity suggests that the pinhole nanocontacts are typically 10–30 nm in diameter, although occasionally some are much smaller than 1 nm.
- (3) The BMR effects, including the time-dependent relaxation in the resistance, are similar to the results obtained earlier for nanocontacts electrodeposited between macroscopic Ni wires.
- (4) The similarity of results between these two very different sample geometries appears to rule out magnetostriction as an explanation for the data.
- (5) The very small volume of magnetic metal in the present samples is an additional reason for ruling out magnetostriction.
- (6) The similarity between the present results and the earlier ones suggests that it is only a matter of time before BMR values in the samples of the present geometry reach the level achieved earlier with macroscopic wires (700%)(2).
- (7) The pinholes seem to be concomitant with the oxide thin layers and play a significant role in magnetoresistive effects.

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