

Field Angle and Thickness Dependence of Coercivity in Electrodeposited CoNi–Cu Multilayer Nanowires

Xue-Ti Tang¹, Gwo-Ching Wang¹, and Mitsuhiro Shima²

¹Department of Physics, Applied Physics and Astronomy, Rensselaer Polytechnic Institute, Troy, NY 12180-3590 USA

²Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180-3590 USA

Variation in the coercivity of CoNi–Cu multilayer nanowires grown by electrodeposition using a hole pattern of alumina templates has been systematically studied as a function of the CoNi layer thickness and the angle between an applied magnetic field and the nanowire axis. We have found that the magnetization reversal for the nanowires with thin disk-shaped CoNi layers is in the coherent rotation mode, while that for the nanowires with long rod-shaped CoNi layers can be interpreted by a combination of the coherent rotation and curling modes. With decreasing CoNi layer thickness $t(\text{CoNi})$ from 6.8 ± 0.8 to 1.7 ± 0.2 nm, the coercivity rapidly decreases from 305 to 10 Oe, indicating that for $t(\text{CoNi}) < 1.7 \pm 0.2$ nm, the CoNi layers of the nanowires become superparamagnetic at room temperature. The room temperature superparamagnetism is further confirmed by magnetization measurements at low temperatures using a superconducting quantum interference device. The estimated blocking temperatures for $t(\text{CoNi}) = 1.7 \pm 0.2$ and 1.0 ± 0.1 nm are 181 and 136 K, respectively.

Index Terms—Magnetic hysteresis, magnetic wire, magnetization reversal, multilayer.

I. INTRODUCTION

THERE has been considerable interest in investigating magnetic/nonmagnetic multilayer nanowires particularly because of their large giant magnetoresistance (GMR) effects observed in the current perpendicular-to-plane (CPP) geometry [1]. The magnetic and magnetotransport properties of the multilayer nanowires can be controlled by changing the magnetic layer thickness. For practical applications of the GMR nanowires, the coercivity is one of the most important parameters that needs to be optimized for device operations. The coercivity of multilayer nanowires is also closely related to various fundamental magnetic phenomena, such as the magnetization reversal process. The magnetization reversal mechanism in multilayer nanowires can sensitively change with the sample dimensions, particularly the magnetic layer thickness, which gives rise to a variation of the coercivity. It has been shown that by measuring the angular dependence of coercivity, one can obtain very useful information about the magnetization reversal mode [2], [3]. Moreover, the coercivity of nanoscale magnetic materials can dramatically decrease upon a transition from the ferromagnetic to the superparamagnetic state as the coercivity in the superparamagnetic state is virtually zero due to the thermal instability of the magnetic moments.

In this work, we report an angular dependence of room temperature coercivity for CoNi–Cu multilayer nanowires grown in a hole pattern of alumina templates by pulsed electrodeposition. The coercivity measurements were carried out as a function of the angle between an applied magnetic field and the nanowire axis. The obtained data were then used to analyze the magnetization reversal mode of the nanowires. When the CoNi layer thickness $t(\text{CoNi})$ in the multilayer nanowires is very small, the coercivity significantly decreases due to an onset of superpara-

magnetism. The temperature dependence of coercivity for multilayer nanowires is also presented.

II. EXPERIMENTAL PROCEDURE

The CoNi–Cu multilayer nanowires with a diameter of about 250 nm were electrochemically grown into a hole pattern of anodized alumina templates, using a method described in [4]. The magnetic properties of the nanowires were systematically studied as a function of the CoNi layer thickness varied from $t(\text{CoNi}) = 1.0$ nm to $7.5 \mu\text{m}$ with the Cu layer thickness kept constant at $t(\text{Cu}) = 4.2$ nm. The total length of nanowires was maintained at about $20 \mu\text{m}$ for all of the samples.

The coercivity values H_C for the multilayer nanowires were determined from magnetic hysteresis loops measured at room temperature using a vibrating sample magnetometer (VSM). The magnetic measurements were carried out as a function of the angle θ between the nanowire axis and the applied magnetic field, varied from 0° to 180° using incremental steps of 15° . The H_C values of the nanowires were also measured at low temperatures using a superconducting quantum interference device (SQUID) in magnetic fields applied perpendicular to the nanowire axis.

III. RESULTS AND DISCUSSION

Fig. 1 shows magnetic hysteresis loops measured at room temperature for CoNi–Cu nanowires with $t(\text{CoNi}) = 7.5 \pm 1.0 \mu\text{m}$ (long rod-shaped), 6.8 ± 0.8 nm (thin disk-shaped), and 1.0 ± 0.1 nm at the angles $\theta = 90^\circ$ and 0° . The nanowires with $t(\text{CoNi}) = 7.5 \pm 1.0 \mu\text{m}$ measured at $\theta = 90^\circ$ exhibit a sheared loop in the low field range with a saturation field of ~ 5 kOe, while at $\theta = 0^\circ$, the loop is in a more squared shape with the saturation field of ~ 2 kOe [Fig. 1(a)], indicating that the magnetic easy axis lies along the nanowire axis. In contrast, the nanowires with $t(\text{CoNi}) = 6.8 \pm 0.8$ nm show a largely sheared hysteresis loop with the saturation field of more than 8 kOe at $\theta = 0^\circ$ and ~ 5 kOe at $\theta = 90^\circ$ [Fig. 1(b)],

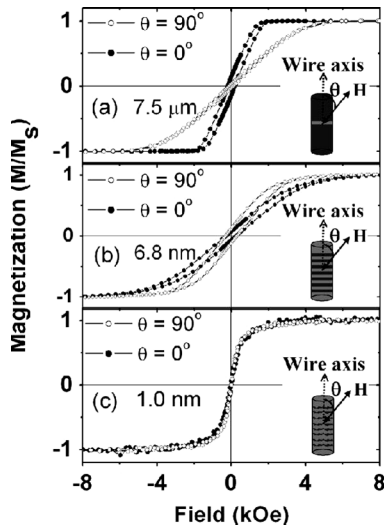


Fig. 1. Magnetic hysteresis loops measured by VSM at room temperature for CoNi–Cu nanowires with (a) $t(\text{CoNi}) = 7.5 \mu\text{m}$, (b) $t(\text{CoNi}) = 6.8 \text{ nm}$, and (c) $t(\text{CoNi}) = 1 \text{ nm}$. Schematic representations shown on the right define the sample geometry with respect to the applied magnetic-field direction.

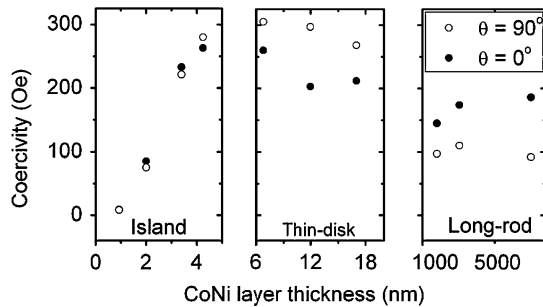


Fig. 2. Coercivity measured at room temperature by VSM in magnetic-field directions perpendicular and parallel to the nanowire axis of CoNi–Cu samples, plotted as a function of the CoNi layer thickness.

indicating that the easy axis is perpendicular to the nanowire axis. At $t(\text{CoNi}) \sim 51 \text{ nm}$, the magnetic easy axis rotates from a direction parallel to the nanowire axis to that lying in a plane perpendicular to the nanowires. When $t(\text{CoNi}) < 1.7 \text{ nm}$ (Fig. 1(c) shows a result for $t(\text{CoNi}) = 1.0 \pm 0.1 \text{ nm}$), the hysteresis loops yield a very small coercivity H_C ($\sim 10 \text{ Oe}$) with a negligibly small angular dependence, indicating that the magnetic layers are superparamagnetic at room temperature.

The CoNi layer thickness dependence of the coercivity H_C for CoNi–Cu multilayer nanowires measured at $\theta = 90^\circ$ and 0° is summarized in Fig. 2. When $t(\text{CoNi}) > 1000 \text{ nm}$ (“long rod-shaped” region), the values of H_C for $\theta = 0^\circ$ are consistently larger than those for $\theta = 90^\circ$. On the other hand, when $6 \text{ nm} \leq t(\text{CoNi}) \leq 19 \text{ nm}$ (“thin disk-shaped” region), the values of H_C for $\theta = 90^\circ$ are consistently larger than those for $\theta = 0^\circ$. The observed tendency for the magnetic layer thickness dependence of H_C can be explained by the magnetization reversal mode, which will be discussed later. When $t(\text{CoNi}) \leq 5 \text{ nm}$ (“island shaped” region), the values of H_C measured at $\theta = 0^\circ$ are fairly close to those at $\theta = 90^\circ$ with no obvious angular dependence, indicating that the anisotropy energy KV (K : anisotropy constant for CoNi, V : volume of each CoNi layer) of the CoNi

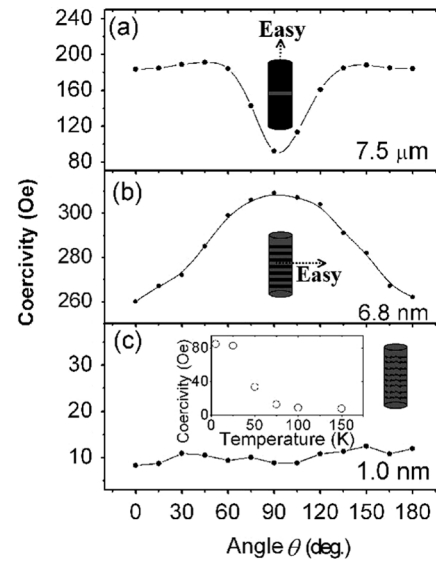


Fig. 3. Coercivity measured as a function of the angle θ between an applied magnetic field and the nanowire axis for CoNi–Cu nanowires with magnetic layer thicknesses (a) $t(\text{CoNi}) = 7.5 \mu\text{m}$, (b) $t(\text{CoNi}) = 6.8 \text{ nm}$, and (c) $t(\text{CoNi}) = 1 \text{ nm}$. The easy axis direction in (c) is not defined since the sample is superparamagnetic. The solid curves in (a), (b), and (c) are guides to the eyes. The inset in (c) shows a temperature dependence of the coercivity measured by SQUID in magnetic fields applied perpendicular to the nanowire axis as a function of the temperature for $t(\text{CoNi}) = 1.0 \text{ nm}$.

layers significantly decreases with decreasing V . The decrease in CoNi layer thickness may introduce a discontinuity in the layers, which can further reduce the volume of each segment in the magnetic layers that eventually becomes superparamagnetic at room temperature [5], [6]. Indeed as $t(\text{CoNi})$ decreases from 6.8 to 1 nm, H_C decreases rapidly from 305 to 10 Oe.

In order to identify the magnetization reversal mode for the CoNi–Cu nanowires, we extracted the coercivity data from the hysteresis loops measured at various angles θ . Fig. 3 shows the angular dependence of H_C for samples with various values of $t(\text{CoNi})$. Nanowires with $t(\text{CoNi}) = 7.5 \pm 1.0 \mu\text{m}$ [Fig. 3(a)], the magnetic easy axis lies in the direction parallel to the nanowire axis. The dependence of H_C on the angle between the easy axis and the field direction shows the same trend as the dependence of H_C on θ . A slight increase in H_C is observed when θ increases from 0° to 45° . The observed increase in H_C infers that the magnetization reversal proceeds in a curling mode. According to the Aharoni’s theory of the curling mode magnetization reversal [7], the coercivity H_C should increase with increasing the angle between the easy axis and the field direction. When θ further increases from 45° to 90° , H_C dramatically decreases due to a transition of the magnetization reversal mode from the curling to the coherent rotation. The angular dependence of H_C in the coherent rotation mode can be calculated from the classical Stoner–Wohlfarth (SW) model [8]. According to the SW model, H_C should decrease with increasing the angle between the easy axis and the field direction.

The transition from the curling mode at low angles θ to the coherent rotation mode at high angles θ can be explained by a mechanism described in the theoretical study by Sun *et al.* [9]. In this case, the coercivity H_C in the curling mode is smaller than that in the coherent rotation mode when θ is small, while

H_C in the curling mode becomes larger than that for the coherent rotation when θ is larger. For the curling mode, the spins in the rod-shaped CoNi layers are in a configuration with no net magnetization along the hard axis as reported in [9]. The results we obtained for CoNi–Cu nanowires with long rod-shaped CoNi layers show a similar trend to that reported for single-element magnetic nanowires [2], [3]. In the case of multilayer nanowires with long rod-shaped magnetic layers, a head-to-tail alignment of magnetization between adjacent magnetic layers is energetically more favorable so as to minimize the magnetostatic energy of the system. Hence, the magnetization reversal in the CoNi–Cu nanowires with long rod-shaped CoNi layers is believed to proceed in the same way as that for single-element magnetic nanowires.

In the case of CoNi–Cu multilayer nanowires with thin disk-shaped magnetic layers, the easy axis lies in the direction perpendicular to the nanowire axis (i.e., $\theta = 90^\circ$) [Fig. 3(b)]. The trend in the variation of H_C with the angle between the easy axis and the field direction is opposite that of the angle θ in the case of the disk-shaped magnetic layers. The coercivity H_C decreases with decreasing θ and with increasing the angle between the easy axis and the field direction, indicating that a coherent rotation of spin occurs in the CoNi layers. It has been theoretically shown that the magnetization reversal mode is governed by minimization of the total energy of the system that is given by the sum of the exchange energy E_{ex} and the magnetostatic energy E_{ms} . In the coherent rotation mode, E_{ex} can be minimized since all of the spins rotate in unison during the magnetization reversal process. On the other hand, E_{ms} can be minimized in the curling mode since the spin directions in the magnetic entity are distributed in a curling pattern so that there is no net magnetization along the hard axis. The coherent rotation mode is therefore more favorable when E_{ex} needs to be minimized, while the curling mode is more favorable when E_{ms} should be minimized. For CoNi–Cu nanowires with small $t(\text{CoNi})$, the angles between neighboring spins would be significantly large, which increase the exchange energy in the CoNi layers if the spins were in a curling pattern. Hence, the coherent rotation mode is more favorable for small $t(\text{CoNi})$ than the curling mode in order to minimize the exchange energy.

In the case of nanowires with $t(\text{CoNi}) = 1.0 \pm 0.1$ nm [Fig. 3(c)], the coercivity H_C is fairly small (~ 10 Oe), independent of the measurement angle. In the temperature dependence of H_C shown as the inset of Fig. 3(c), H_C for $t(\text{CoNi}) = 1.0 \pm 0.1$ nm decreases with increasing temperature, due to the onset of superparamagnetism. The observed temperature dependence of coercivity is consistent with the trend predicted from the SW model for the coherent rotation mode. According to the SW model, the coercivity can be expressed as $H_c = H_{c0}[1 - (T/T_B)^{1/2}]$ where H_{c0} and T_B are the coercivity at 0 K and the blocking temperature of the magnet, respectively [10]. The values for T_B at $t(\text{CoNi}) = 1.0 \pm 0.1$ nm and 1.7 ± 0.2 nm (the data are not shown here) are about 136 and 181 K, respectively. The superparamagnetism

observed in the CoNi–Cu nanowires with $t(\text{CoNi}) < 1.7$ nm can be attributed to thermal instability of the magnetic moments in the CoNi layers due to the reduction in the volume of each layer with decreasing the layer thickness and eventual discontinuity in the magnetic layers [5], [6]. The discontinuity of very thin CoNi layers is indeed inferred in the SEM images obtained from CoNi–Cu multilayer nanowires, which reveal some waviness in the interfaces of the multilayer nanowires. Such wavy interfaces are likely to induce a thickness variation in each layer. When each magnetic layer becomes very thin and even discontinuous, the multilayered nanowires readily become superparamagnetic at ambient temperature.

IV. CONCLUSION

The coercivity of CoNi–Cu multilayer nanowires has been studied as a function of the field angle, magnetic layer thickness, and temperature. The angular dependence of the coercivity measured with respect to the nanowire axis shows that the magnetization reversal for nanowires with thin disk-shaped CoNi layers is in the coherent rotation mode, while that for long rod-shaped CoNi layers is explained by a combination of the coherent rotation and curling modes. For $t(\text{CoNi}) \leq 6.8$ nm, the coercivity dramatically decreases with decreasing $t(\text{CoNi})$, indicating that the magnetic layers become superparamagnetic. Our experimental findings will hopefully stimulate the current development of theoretical modeling for magnetic/nonmagnetic multilayer nanostructures and will help the design of novel magnetic nanosensors and ultra-high density data-storage devices.

ACKNOWLEDGMENT

This work was supported in part by the Rensselaer Polytechnic Institute seed grant and in part by the National Science Foundation Award 05 06 738.

REFERENCES

- [1] P. R. Evans, G. Yi, and W. Schwarzacher, *Appl. Phys. Lett.*, vol. 76, pp. 481–483, 2000.
- [2] S. Goolaup, N. Singh, A. O. Adeyeye, V. Ng, and M. B. A. Jalil, *Eur. Phys. J. B*, vol. 44, pp. 259–264, 2005.
- [3] G. C. Han, B. Y. Zong, P. Luo, and Y. H. Wu, *J. Appl. Phys.*, vol. 93, pp. 9202–9207, 2003.
- [4] X.-T. Tang, G.-C. Wang, and M. Shima, *J. Appl. Phys.*, vol. 99, pp. 033 906–033 906-7, 2006.
- [5] F. Spizzo, E. Angeli, D. Bisero, P. Vavassori, and F. Ronconi, *Appl. Phys. Lett.*, vol. 79, pp. 3293–3295, 2001.
- [6] J. Xu, M. A. Howson, B. J. Hickey, and D. Greig, *Phys. Rev. B*, vol. 55, pp. 416–422, 1997.
- [7] A. Aharoni, *J. Appl. Phys.*, vol. 82, pp. 1281–1287, 1997.
- [8] E. C. Stoner and E. P. Wohlfarth, *Phil. Tr. R. Soc. S-A*, vol. 240, pp. 599–642, 1948.
- [9] L. Sun, Y. Hao, C.-L. Chien, and P. C. Searson, *IBM J. Res. Dev.*, vol. 49, pp. 79–102, 2005.
- [10] B. D. Cullity, *Introduction to Magnetic Materials*. Reading, MA: Addison-Wesley, 1972, pp. 410–421.