Dependence on Fermi Surface Dimensions of Oscillatory Exchange Coupling in $Co/Cu_{1-x}Ni_x(110)$ Multilayers

S. N. Okuno and K. Inomata

Research and Development Center, Toshiba Corporation, Kawasaki 210, Japan

(Received 23 November 1992)

A significant change in the oscillatory exchange coupling corresponding to the contraction of the Fermi surface in the spacer layers is found in the $Co/Cu_{1-x}Ni_x$ (110) system. The oscillation period becomes longer with increasing Ni concentration: from 12 Å (0% Ni) to 23 Å (35% Ni). This change manifests that the long-period oscillation across Cu-Ni (110) arises from the singularity at the neck orbit of the spacer Fermi surface. The neck diameters estimated from the measured periods are in good agreement with those of bulk Cu-Ni Fermi surfaces.

PACS numbers: 75.50.Rr, 73.20.Dx, 75.30.Et

The exchange coupling between ferromagnetic layers across nonmagnetic interlayers has attracted great attention since the discoveries of antiferromagnetic (AF) coupling [1] and "giant" negative magnetoresistance (MR) in AF-coupled films [2]. One of the most striking features is the oscillatory behavior between AF coupling and ferromagnetic (F) coupling as a function of the spacer layer thickness. Since the discovery of this phenomenon by Parkin, More, and Roche in Fe/Cr, Co/Cr, and Co/Ru multilayer systems [3], it has become known to be characteristic of a whole new class of materials [4–9]. Most of the systems show oscillatory behavior with a periodicity of 10–18 Å, which is much longer than would be expected on the basis of a conventional RKKY model. Furthermore, multiperiodic oscillatory coupling has been observed in MBE-grown wedge-shaped sandwiches with sharp interfaces [10–13]. Various theoretical approaches have been proposed to explain this phenomenon [14–16]. A simple approach takes into account the discreteness of the spacer layer thickness in the RKKY model [15]. From the periodicity of the lattice planes, the wave vector is reduced to the first Brillouin zone of the one-dimensional lattice, which leads to the period of more than two monolayers. Further, Bruno and Chappert [16] have argued that the oscillation is determined by the singularity in the Fermi surface of the spacer layer which connects extremal points by the vector **q** along the growth direction of the layered system. They have predicted the number and values of the oscillation periods for noble metal spacers. While this is a most likely approach, because both long and short periods predicted for (001) have been found in Cu and Au [12,13] and an orientation dependence of the coupling strength in accordance with the theory has been reported for Cu [6], it is still not quite clear to what extent the period of the oscillation reflects the Fermi surface of bulk spacer materials. The differences in the spacer materials and in the orientations have not been clearly reflected in the experimental periods. The experimental long periods of the oscillations for fcc Cu(111), (001), and (110) and Au(001) are all within 10–14.5 Å [5–9,12,13]. Furthermore, systematic study for transition metal spacers has shown that all the spacer layers, except for Cr, give the same oscillation period of 9–11 Å [4].

In this Letter, we report a direct relation between the oscillation period and the extremal wave vector at the Fermi surface of the spacer. We have investigated a series of $Co/Cu_{1-x}Ni_x(110)$ multilayers ($x=0$, 0.14, 0.23, and 0.35) by means of field-dependent magnetization and MR measurements. Cu-Ni alloys are a suitable candidate to examine the effect of the spacer layer on the oscillatory coupling because they form solid solutions over the entire concentration range, they are nonmagnetic up to ~45 at.% Ni in bulk Cu-Ni alloys, and the Fermi surface can be altered continuously according to the Ni concentration [17]. We have found that the long-period oscillation becomes longer as the Ni concentration increases. The observed oscillation periods were discussed quantitatively using Fermi surfaces of bulk Cu-Ni alloys.

$Co/Cu_{1-x}Ni_x$ multilayers were grown epitaxially on single crystal MgO(110) substrates by ion beam sputtering at a base pressure of 5×10^{-7} Torr. For Cu-Ni spacer layers, 0, 14, 23, and 35 at.% Ni concentrations were chosen. The individual Co layer thickness was held constant at ~10 Å, and the number of bilayers was 16 for all films studied. The Cu-Ni layer thickness was designed to be between 3 and 50 Å for each Cu-Ni composition. The thicknesses for the Co and Cu-Ni layers were confirmed by a chemical analysis using inductively coupled plasma-optical emission spectroscopy and by the bilayer thickness estimated from x-ray satellite peaks around main diffraction peaks. For x-ray diffraction measurements, each sample exhibits first to third order satellites around the main peak and superlattice peaks at low angle depending on the Cu-Ni thickness, indicating that the films are well layered. A conventional θ-2θ scan and tilted sample measurements reveal that the films are grown with an orientation of Co/Cu-Ni(110)‖MgO(110) and Co/Cu-Ni⟨100⟩‖MgO⟨100⟩. For reference, the lattice spacings of the single-layered $Cu_{1-x}Ni_x$ films (500 Å thickness) deposited on the MgO(110) substrates were examined and were 1.274, 1.270, and 1.267 Å for

$x=0.14$, 0.23, and 0.35, respectively, which are in good agreement with the predictions from Vegard's law.

Magnetization measurements were performed by using a vibrating sample magnetometer with the magnetic field applied in the film plane. The saturation magnetization of the multilayers is almost constant, independent of the spacer thickness, and is nearly the same as that of bulk Co. However, the average magnetization over each Cu-Ni composition becomes somewhat larger as the Ni concentration increases. Assuming that Ni atoms in Cu-Ni alloys near the interface have the same magnetic moment as in the bulk Ni metal, it is supposed that Ni atoms in some 2 Å of Cu-Ni at each interface are polarized by the direct interaction with neighboring Co layers. Torque measurement reveals that a uniaxial in-plane magnetic anisotropy of the order of 10^6 erg/cm^3 is introduced in all the samples deposited on MgO(110).

The magnetic coupling between adjacent Co layers across the Cu-Ni spacer was investigated by means of the field-dependent magnetization and MR measurements. Figures 1(a),1(b) and 1(c),1(d) show typical *M-H* loops and MR curves of Co/$Cu_{0.65}Ni_{0.35}$ multilayers for F- and AF-coupled cases. The corresponding $Cu_{0.65}Ni_{0.35}$ thicknesses are 10.3 and 14.3 Å, respectively. The external magnetic field was applied parallel to the [100] (easy) axis in the film plane. The *M-H* loop characteristic of the F coupling [Fig. 1(a)] is square, and shows low saturation field and high remanent magnetization. The corresponding MR effect [Fig. 1(b)] is too small to be detected. In the case of AF coupling [Fig. 1(c)], on the other hand, the coupling aligns the adjacent Co layers antiferromagnetically, to give a nearly zero remanence, and the external magnetic field which overcomes the AF coupling aligns Co layers parallel to the applied field, yielding a magnetization characteristic of ferromagnetic saturation. Large spin-dependent scattering of the conduction electrons, due to which the resistivity is large in AF alignment and small in F alignment, gives rise to large magnetoresistance [Fig. 1(d)]. A detailed interpretation of the magnetization process can be found in Ref. [18]. The AF coupling strength between two adjacent Co layers with in-plane uniaxial magnetic anisotropy (K_u) follows $J=-dM_sH_s/2$ ($K_u>J/d$), where d and M_s are the magnetic layer thickness and saturation magnetization, respectively, and the saturation field H_s is defined in Fig. 1.

The dependence of the MR ratio on the spacer layer thickness for Co/$Cu_{0.86}Ni_{0.14}$, Co/$Cu_{0.77}Ni_{0.23}$, and Co/$Cu_{0.65}Ni_{0.35}$ multilayers (77 K) is shown in Fig. 2. Three or two oscillations appear in the MR ratio with longer periods with increasing Ni concentration. The saturation field and the remanent magnetization also oscillate correspondingly. These oscillatory behaviors indicate that the exchange interaction oscillates between AF and F coupling depending on the Cu-Ni spacer layer thickness; in Fig. 2, the AF coupling arises at the thickness which shows peaks and the F coupling corresponds to the flat regions. The maximum MR ratios decrease with increasing Ni concentration. The absolute values of ρ_s for Co/Cu-Ni samples are within 25–40 $\mu\Omega$ cm depending on the Ni concentration in the films. It is noticeable that $\Delta\rho$ is rather larger for the samples with a little Ni addition than for the no-addition samples. A detailed description

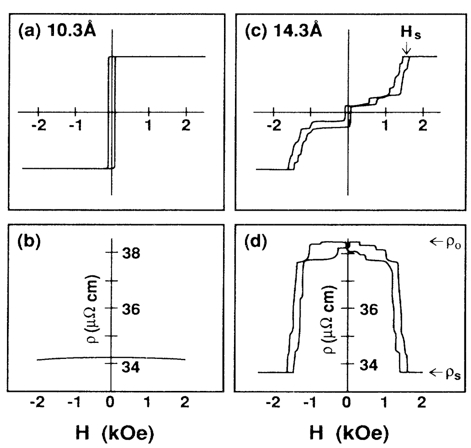

FIG. 1. *M-H* loops and MR curves for Co(9.5 Å)/$Cu_{0.65}$-$Ni_{0.35}$(110) multilayers which are (a),(b) F coupled across 10.3 Å $Cu_{0.65}Ni_{0.35}$, and (c),(d) AF coupled across 14.3 Å $Cu_{0.65}Ni_{0.35}$.

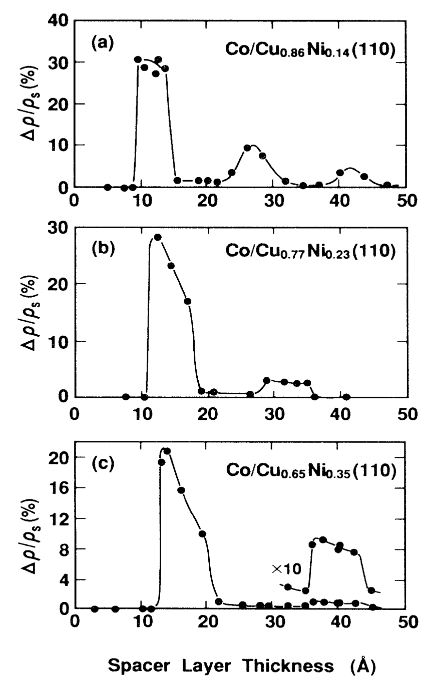

FIG. 2. MR ratio as a function of spacer layer thickness for (a) 16×[Co(9.3 Å)/$Cu_{0.86}Ni_{0.14}$(t Å)](110), (b) 16×[Co(10.0 Å)/$Cu_{0.77}Ni_{0.23}$(t Å)](110), and (c) 16×[Co(9.5 Å)/$Cu_{0.65}$-$Ni_{0.35}$(t Å)] multilayers at 77 K. The solid lines serve as guides to the eye. $\Delta\rho$ is defined as $\rho_0-\rho_s$, which are shown in Fig. 1.

of the magnetoresistance will be presented in a forthcoming publication. For $Co/Cu_{0.86}Ni_{0.14}$ multilayers [Fig. 2(a)], the position of the first AF peak is 11 Å and the oscillation period is 12 Å. The maximum saturation field yields an AF-coupling strength $J=-0.26$ mJ/m^2. For $Co/Cu_{0.77}Ni_{0.23}$ and $Co/Cu_{0.65}Ni_{0.35}$ multilayers [Figs. 2(b),2(c)], two wide peaks appear with a right-shoulder-dropped shape with the same width. Since the oscillation periods become noticeably longer, the width of the AF peaks becomes wider. Consequently, a decrease of the MR ratio through the increase of the spacer thickness appears in one AF region, giving the distorted shape. The positions of the first AF peak and oscillation periods for Figs. 2(b) and 2(c) are 14.5 Å, 17 Å and 17 Å, 23 Å, respectively. The maximum AF coupling strengths are -0.13 and -0.08 mJ/m^2, respectively.

For the Co/Cu (Ni 0%) multilayers the MR shows three oscillations in the Cu thickness range up to 40 Å; the first peak is at 10 Å and the intervals are 12 Å, which is the same as the observations in $Co_{0.9}Fe_{0.1}/Cu(110)$ multilayers on the same substrates [19]. Johnson *et al.* [6] have observed a similar oscillation period (12.5 Å) but different first peak position (8.5 Å) in a Co/Cu/Co(110) MBE-grown wedge-shaped sandwich.

Measured coupling data for Co/Cu-Ni(110) systems are summarized in Table I. The oscillation behavior in the Co/Cu-Ni(110) system strongly depends on the Ni concentration in the spacer layer; with increasing Ni concentration, the oscillation period becomes longer, the position at the first AF peak shifts toward greater thickness, and the maximum AF coupling strength becomes weaker. The change in J should be also affected by a difference in the peak position. Thus, we compared J at the same spacer layer thickness using the decay of J versus spacer thickness derived from the first and second AF peaks for each CuNi composition. It still showed a small decrease with increasing Ni concentration; J at 10 Å for 14, 23, and 35 at.% Ni samples was -0.32, -0.29, and -0.26 mJ/m^2, respectively.

A plausible cause of the change in oscillatory period is a change in electronic state in the spacer layer. Although Ni addition to the Cu spacer contributes to the change in lattice spacing, this change is at most 0.86% for $Cu_{0.65}Ni_{0.35}$ compared with Cu, which is too small to explain the approximate doubling of the oscillation period. Ni addition should also change the interface state. Polarized Ni atoms at the interface might contribute to the increase of magnetic randomness at the interface. In order to confirm the effect of roughness on oscillation periods experimentally, we prepared Co/Cu multilayers with rougher interface by increasing the argon acceleration voltage in ion beam sputter deposition. They did not show the oscillation between AF and F coupling, but showed an oscillation in the weak AF coupling strength with the same period as mentioned previously.

In a Cu-Ni alloy, the Fermi surface gradually contracts with increasing Ni content, maintaining the topology of the Cu Fermi surface [20]. In the light of the theory [16], we discuss the relation between the measured oscillation periods and the extremal points at the spacer Fermi surface. In the Cu-Ni Fermi surface, there are several extremal wave vectors along the [110] direction: three wave vectors linking extremal points on neighboring Fermi surfaces which include wave vectors relating to the belly orbit, and one wave vector passing through the neck diameter in the [110] direction, linking a pair of saddle points of the opposite faces with velocities parallel and antiparallel to [110]. We can select an extremal vector in Cu-Ni(110) which determines the observed oscillations. The former wave vectors become longer with increasing Ni concentration. Thus, their oscillation periods $\Lambda = 2\pi/|\mathbf{q}|$ become smaller as the Ni concentration increases, opposing the observed change in the period. The size of the latter wave vector, which is equal to the neck diameter, becomes smaller with Ni concentration in-

TABLE I. Summary of measured coupling data for the Co/CuNi (110) multilayers, including the oscillation period (Λ), the position of the first AF coupling (t), and the maximum coupling strength at first AF coupling. The short-period oscillations are not found.

Ni concentration (at.%)	Λ (Å)	t (Å)	$-J$ (mJ/m^2)
0	12	10	0.40
14	15	11	0.26
23	17	14.5	0.13
35	23	17	0.08

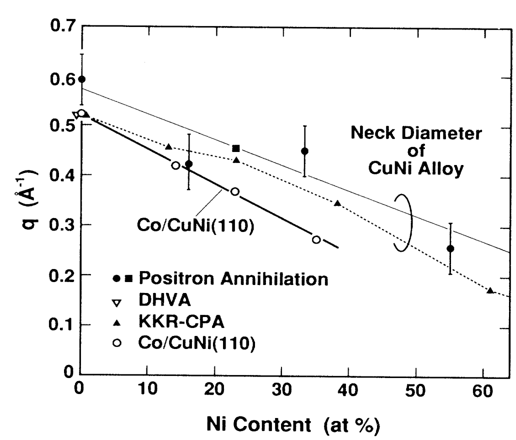

FIG. 3. Comparison of the composition dependences of the wave vectors obtained from measured oscillation periods of exchange coupling and neck diameters in Cu-Ni alloys. The open circles give the wave vectors from periods of oscillations; closed circles with error bars and squares give the positron annihilation experimental results of Hasagawa and McGervey, respectively [21]; and closed triangles give the calculated results from KKR-CPA [20]. An open inverted triangle is the neck diameter of pure Cu from the de Haas–van Alphen effect [22].

crease. Thus, the oscillation period becomes longer with Ni addition, which agrees with the experimental result. In Fig. 3, the wave vectors obtained from measured oscillation periods in Co/Cu-Ni(110) multilayers are compared with the neck diameters of the bulk Cu-Ni alloys. They are in good quantitative agreement. This manifests that the long-period oscillatory coupling across Cu-Ni(110) arises from the extremal points of the neck orbit, strongly supporting the theoretical approach of Bruno and Chappert [16]. Moreover, this suggests that the bulk Fermi surface is maintained even in ultrathin films of about 10 Å thickness. The oscillations arising from the other extremal points are expected to have short periods of less than 4.2 Å. These short-period oscillations were not found in our multilayers probably because of the somewhat rough interface.

In conclusion, we have observed the oscillatory exchange coupling in a series of $Co/Cu_{1-x}Ni_x(110)$ multilayers and shown that the variation of the oscillation period is well correlated with the variation of the neck diameter of the Fermi surface of the bulk $Cu_{1-x}Ni_x$ alloy. The present result has provided a strong experimental proof for the recent theory [16].

The authors wish to thank H. Endo for measurements of the inductively coupled plasma-optical emission spectroscopy.

[1] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).

[2] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988); G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).

[3] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2301 (1990).

[4] S. S. P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).

[5] D. H. Mosca, F. Petroff, A. Fert, P. A. Schroeder, W. P. Pratt, Jr., and R. Loloee, J. Magn. Magn. Mater. **94**, L1 (1991); S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).

[6] M. T. Johnson, R. Coehoorn, J. J. de Vries, N. W. E. McGee, J. aan de Stegge, and P. J. H. Bloemen, Phys. Rev. Lett. **69**, 969 (1992).

[7] F. Petroff, A. Barthelemy, D. H. Mosca, D. K. Lottis, A. Fert, P. A. Schroeder, W. P. Pratt, Jr., R. LaLoe, and S. Lequien, Phys. Rev. B **44**, 5355 (1991).

[8] J. J. de Miguel, A. Cbollada, J. M. Gallego, R. Miranda, C. M. Schneider, P. Schuster, and J. Kirshner, J. Magn. Magn. Mater. **93**, 1 (1991); Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. B **46**, 8659 (1992).

[9] W. R. Bennett, W. Schwarzacher, and W. F. Egelfoff, Jr., Phys. Rev. Lett. **65**, 3169 (1990).

[10] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **67**, 140 (1991).

[11] S. T. Purcell, M. T. Johnson, N. W. E. McGee, R. Coehoorn, and W. Hoving, Phys. Rev. B **45**, 13064 (1992).

[12] M. T. Johnson, S. T. Purcell, N. W. E. McGee, R. Coehoorn, J. aan de Stegge, and W. Hoving, Phys. Rev. Lett. **68**, 2688 (1992).

[13] A. Fuss, S. Demokritov, P. Grunberg, and W. Zinn, J. Magn. Magn. Mater. **103**, L221 (1992).

[14] Y. Wang, P. M. Levy, and J. L. Frey, Phys. Rev. Lett. **65**, 2732 (1990); H. Hasegawa, Phys. Rev. B **42**, 2368 (1990); D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, Phys. Rev. Lett. **67**, 493 (1991).

[15] R. Coehoorn, Phys. Rev. B **44**, 9331 (1991); C. Chappert and J. P. Renard, Europhys. Lett. **15**, 553 (1991); D. M. Deaven, D. S. Rokhsar, and M. Johnson, Phys. Rev. B **44**, 5977 (1991).

[16] P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991); Phys. Rev. B **46**, 261 (1992).

[17] In disordered alloys the imprint of the Fermi surface disappears when the impurity broadening exceeds the effective temperature of 5–10 K. However, as shown in Ref. [20], this broadening is quite small and the Fermi surface is well defined.

[18] K. Inomata and Y. Saito, Appl. Phys. Lett. **61**, 726 (1992).

[19] Y. Saito, S. Hashimoto, and K. Inomata, Appl. Phys. Lett. **60**, 2438 (1992).

[20] B. Gordon, W. M. Temmerman, B. L. Gyorffy, and G. M. Stocks, *Transition Metals—1977,* IOP Conf. Proc. No. 39 (Institute of Physics, London, 1978), p. 402.

[21] B. W. Murray and J. D. McGervey, Phys. Rev. Lett. **24**, 9 (1970); M. Hasegawa, T. Suzuki, and M. Hirabayashi, J. Phys. Soc. Jpn. **37**, 85 (1974).

[22] W. J. O'Sullivan and J. E. Schirber, Phys. Rev. **181**, 1367 (1969).