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A theoretical study of interfacial structure of Co/Cu and Co/Pd multilayers

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Abstract. The electronic structures and energies of Co/Cu and Co/Pd multilayers with abrupt or mixed interfaces have been calculated using the linear muffin-tin orbital method in the atomic sphere approximation. The mixed interface is modelled by exchanging atoms between Co layers and Cu (Pd) layers. The calculated total energies of Co/Cu multilayers with mixed interfaces are higher than those of Co/Cu multilayers with abrupt interfaces. The calculated total energies of Co/Pd multilayers with mixed interfaces are lower than those of Co/Pd multilayers with abrupt interfaces. Stable interface structure appears to be abrupt in Co/Cu multilayers and mixed in Co/Pd multilayers, as revealed by experimental observations. Total-energy differences between the multilayer with abrupt interfaces and the multilayer with mixed interfaces are caused by Co 3d-band narrowing for Co/Cu multilayers and Pd spin polarizations for Co/Pd multilayers.

1. Introduction

Since the discovery of perpendicular magnetic anisotropy (PMA) in Co/Pd multilayers [1] and giant magnetoresistance (GMR) in Fe/Cr and Co/Cu multilayers [2, 3], several studies have investigated the magnetism of multilayers. These studies include attempts to clarify the magnetism of multilayers by analysing the electronic structure using first-principles band calculation [4, 5]. However, application of this method is usually limited to multilayers with abrupt interfaces, and the electronic structure of multilayers with mixed interfaces is not well understood.

Some experiments showed that interface structure plays an important role in the magnetism of multilayers. Fullerton *et al* [6] found that in Fe/Cr multilayers, the GMR was enhanced by interfacial roughness, which was controlled by the growth conditions of the multilayers. Moreover, Ono and Shinjo [7] prepared a Co/Cu/NiFe multilayer with rough interfaces on a substrate having a rough surface and found enhancement of the GMR. Thus, the influence of interface structure on the electronic structure of magnetic multilayers should be examined.

Magnetic multilayers have either abrupt or mixed interfaces. Germar *et al* [8] reported that Co film grew layer by layer on Cu(100) substrate and found no mixing between the Co film and the Cu substrate. Co/Cu multilayers are believed to have abrupt interfaces. However, Kim *et al* [9] found a considerable amount of Co–Pd alloy-like phase near the interfaces of Co/Pd multilayers. However, the electronic structure of the Co/Pd multilayer with interfacial mixing is not well understood.

In recent years, some authors have studied the interfacial mixing using band calculations. One test is that of treating the averaged solution for the abrupt interfaces. Schep and Bauer modelled the interface with randomly distributed scatterers and discussed the conductor for a

dirty interface [10]. Herman *et al* averaged the energies for the abrupt interfaces and took the structural and chemical imperfections into account [11]. Another test is that of calculating the local environment modelled by exchanged structure. Fullerton *et al* suggested the interfacial ordered compounds (IOC) model and applied it to Fe/Pd, Co/Ru, Co/Rh, with semi-empirical and/or *ab initio* calculation [12]. Since the local electronic structure in the interface is thought to play an important role as regards the characteristics of the multilayers, this model seems to be adequate for studying the effect of the local environment, like the crystal field and/or the coordination number for the Co/Pd multilayer.

In the present paper, calculations for the multilayers with and without mixed interfaces are made using the linear muffin-tin orbital method in the atomic sphere approximation (LMTO-ASA) [13]. The mixing interfaces are modelled using the IOC model suggested by Fullerton *et al*. The structural stability and the electronic structures of abrupt and mixed interfaces in Co/Cu and Co/Pd multilayers are discussed.

2. Calculation method

The electronic structures of Co_nX_n ($n = 2, 3$) with fcc(001) stacking are calculated, where X = Cu, Pd, and the subscript, n , represents the number of atomic layers. Figure 1 shows the calculation models for Co_3X_3 and Co_2X_2 multilayers. Figures 1 show cross sections along

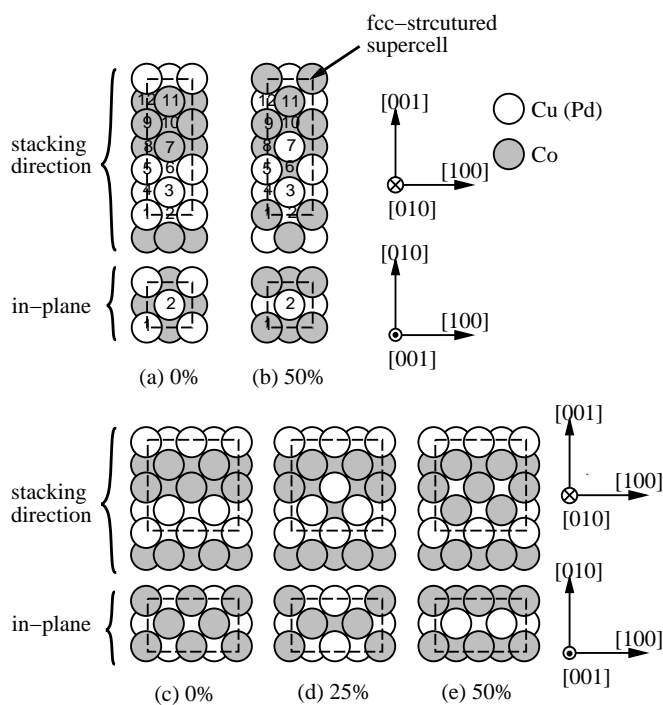


Figure 1. Calculation models for multilayers. The upper and lower schematic diagrams in (a)–(e) indicate the cross sections along the stacking direction and in-plane film. (a) and (b) shows the model for Co_3X_3 (X = Cu, Pd). (a) shows the cross sections of a multilayer without interfacial mixing. (b) shows the cross sections of a multilayer with 50% mixing. (c), (d) and (e) show the model for Co_2X_2 (X = Cu, Pd): (c) no mixing; (d) 25% mixing; (e) 50% mixing. The Co and Cu (Pd) atoms correspond to the solid grey circles and open circles, respectively.

the stacking direction [001] and cross sections along the film plane $\langle 001 \rangle$. The atomic radii of all atoms are assumed to be the averages for the bulk materials. Broken lines represent fcc-structured supercells. For Co_3X_3 multilayers, the cross sections of multilayers without interfacial mixing and with 50% mixing are shown in figures 1(a) and 1(b), respectively. Supercells for Co_3X_3 contain six atomic layers and two in-plane atoms. Each site in the supercell is numbered from 1 to 12. We employ the interfacial ordered compounds (IOC) model suggested by Fullerton *et al* [12]. The multilayer with 50% mixing is represented by exchanging the atoms between sites 1 and 12 and between sites 6 and 7.

For Co_2X_2 multilayers, the cross sections of multilayers without interfacial mixing are shown in figure 1(c) and the cross sections of multilayers with 25% and 50% mixing are shown in figures 1(d) and 1(e), respectively. The supercells for Co_2X_2 contain four atomic layers and four in-plane atoms. The mixing for these cells is modelled by exchanging the atoms on only one side of interface; i.e., the supercells for the 25% and 50% mixing models are $\text{Co}/\text{Co}_{0.75}\text{X}_{0.25}/\text{Co}_{0.25}\text{X}_{0.75}/\text{X}$ and $\text{Co}/\text{Co}_{0.5}\text{X}_{0.5}/\text{Co}_{0.5}\text{X}_{0.5}/\text{X}$, respectively.

The electronic structures are calculated within the framework of the local spin-density approximation using the linear muffin-tin orbital method in the atomic sphere approximation (LMTO-ASA) [13], which has the advantages of treating large numbers of atoms and being a reliable method for the close-packed systems. We use the Vosko–Wilk–Nusair form for the exchange–correlation potential [14]. The core electrons for each of the atoms are treated by the frozen-core approximation. The wavefunctions for the valence electrons of Co, Cu and Pd are expanded in the basis consisting of s, p and d partial waves. The number of k -points is 135 or 243 in the irreducible Brillouin zone, which is equivalent to an eighth of the first Brillouin zone. The atomic radii, the lattice parameters and the numbers of k -points are listed in table 1. The convergence criteria for the energies and magnetic moments at each of the sites are that they are less than 10^{-2} eV and $10^{-3} \mu_B$ after one self-consistent iteration. Even if the number of k -points is increased, the absolute energy differences among the various k -points are less than 10^{-2} eV. The absolute energies for the number of k -points used in this calculation may be unreliable but the relative energies can be discussed.

Table 1. The atomic radii (r_A), the lattice parameters (a , b and c) and the numbers of k -points (N_k).

	r_A (Å)	a (Å)	b (Å)	c (Å)	N_k
Co_3Cu_3	1.278	3.615	3.615	10.845	243
Co_2Cu_2	1.278	7.230	3.615	7.230	135
Co_3Pd_3	1.315	3.718	3.718	11.153	243
Co_2Cu_2	1.315	7.435	3.718	7.435	135

3. Results and discussion

Table 2 shows the calculated total energies of Co/Cu and Co/Pd multilayers. The total energies of Co/Cu multilayers without mixing are lower than those of Co/Cu multilayers with 25% mixing and 50% mixing. In contrast, the total energies of Co/Pd multilayers without mixing are higher than those of Co/Pd multilayers with 25% mixing and 50% mixing. Thus, abrupt interfaces are stable in Co/Cu multilayers and mixed interfaces are stable in Co/Pd multilayers, as revealed by experimental observations [8, 9].

The energies at each of the sites in the Co_3Cu_3 multilayers are shown in table 3. Co sites 1 and 6 in the multilayer with 50% mixing show 0.4 eV higher energy than the corresponding

Table 2. Total energies (in keV) of Co/Cu and Co/Pd multilayers without mixing, with 25% mixing and with 50% mixing.

	No mixing	25% mixing	50% mixing
Co ₃ Cu ₃	-13.3111		-13.3100
Co ₂ Cu ₂	-17.7476	-17.7472	-17.7466
Co ₃ Pd ₃	-9.8260		-9.8274
Co ₂ Pd ₂	-13.1021	-13.1026	-13.1033

Table 3. Energies (in keV) at each of the sites in the Co₃Cu₃ multilayers without mixing and with 50% mixing.

Site	No mixing		50% mixing	
	Type	Energy	Type	Energy
1	Cu	-1.3988	Co	-0.8193
2	Cu	-1.3988	Cu	-1.3989
3	Cu	-1.3988	Cu	-1.3988
4	Cu	-1.3988	Cu	-1.3988
5	Cu	-1.3988	Cu	-1.3989
6	Cu	-1.3988	Co	-0.8193
7	Co	-0.8197	Cu	-1.3989
8	Co	-0.8197	Co	-0.8197
9	Co	-0.8198	Co	-0.8197
10	Co	-0.8198	Co	-0.8197
11	Co	-0.8197	Co	-0.8197
12	Co	-0.8197	Cu	-1.3989

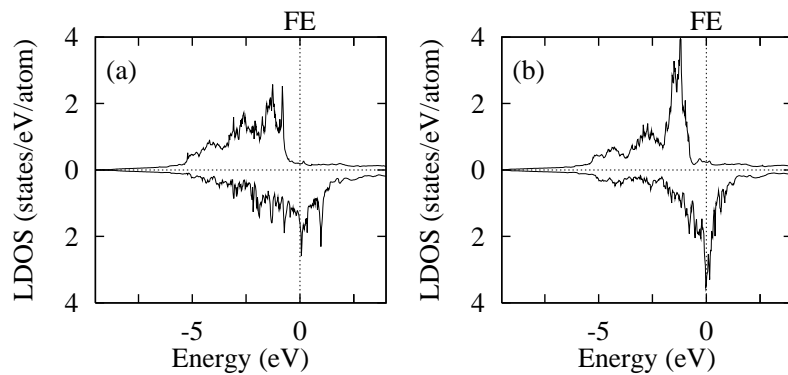
Co sites, sites 12 and 7, in the multilayer without mixing. The other Co sites in the multilayer with 50% mixing show nearly the same energy as the corresponding Co sites in the multilayer without mixing. Furthermore, every Cu site in the multilayer with 50% mixing shows nearly the same energy as the corresponding Cu sites in the multilayer without mixing. The high energy at Co sites 1 and 6 in the multilayer with 50% mixing contributes to the higher total energy of the multilayer with 50% mixing compared to the multilayer without mixing.

The energies at each of the sites in the Co₃Pd₃ multilayers are shown in table 4. Every Co site in the multilayer with 50% mixing shows nearly the same energy as the corresponding Co sites in the multilayer without mixing. Pd sites 7 and 12 in the multilayer with 50% mixing show 0.4 eV lower energy than the corresponding Pd sites, sites 6 and 1, in the multilayer without mixing. The other Pd sites in the multilayer with 50% mixing also show lower energies than the corresponding Pd sites in the multilayer without mixing. However, the energy differences are smaller than 0.4 eV. The low energy at Pd sites 7 and 12 in the multilayer with 50% mixing contributes to the lower total energy of the multilayer with 50% mixing compared to the multilayer without mixing.

A large energy difference is calculated between site 7 in the Co₃Cu₃ multilayer without mixing and site 6 in the Co₃Cu₃ multilayer with 50% mixing. The local densities of states (LDOS) at these Co sites are shown in figure 2. Figure 2(a) shows the LDOS at site 7 in the Co₃Cu₃ multilayer without mixing and figure 2(b) shows the LDOS at site 6 in the Co₃Cu₃ multilayer with 50% mixing. Each spin LDOS at site 6 in the multilayer with 50% mixing has more states at the high-energy end of the 3d band than that at site 7 in the multilayer without mixing.

Table 4. Energies (in keV) at each of the sites in the Co_3Pd_3 multilayers without mixing and with 50% mixing.

Site	No mixing		50% mixing	
	Type	Energy	Type	Energy
1	Pd	-0.8179	Co	-0.8198
2	Pd	-0.8179	Pd	-0.8180
3	Pd	-0.8176	Pd	-0.8179
4	Pd	-0.8176	Pd	-0.8179
5	Pd	-0.8179	Pd	-0.8180
6	Pd	-0.8179	Co	-0.8198
7	Co	-0.8198	Pd	-0.8183
8	Co	-0.8198	Co	-0.8198
9	Co	-0.8199	Co	-0.8199
10	Co	-0.8199	Co	-0.8199
11	Co	-0.8198	Co	-0.8198
12	Co	-0.8198	Pd	-0.8183

**Figure 2.** Calculated LDOSs at (a) site 7 in the Co_3Cu_3 multilayer without mixing and (b) site 6 in the Co_3Cu_3 multilayer with 50% mixing. Upper and lower panels show the components of the majority- and minority-spin, respectively. FE is the Fermi energy.

However, the numbers of electrons at the two sites that occupy each spin band are approximately equal. Figure 3 shows the local numbers of states (LNOSs) at the same sites as are shown in figure 2. Within the energy range from -3 to 0 eV, each spin LNOS at site 6 in the multilayer with 50% mixing is smaller than that at site 7 in the multilayer without mixing. At the Fermi energy, however, the LNOSs at the two sites are approximately equal, showing that the bands at the two sites have the same area and differ only in shape.

The difference in shape can be explained in the following way. A Co atom at site 6 in the Co_3Cu_3 multilayer with 50% mixing has two Co and ten Cu nearest-neighbour atoms. A Co atom at site 7 in the Co_3Cu_3 multilayer without mixing has eight Co and four Cu nearest-neighbour atoms. Thus, the Co atom at site 6 in the Co_3Cu_3 multilayer with 50% mixing is surrounded by more Cu atoms than the Co atom at site 7 in the Co_3Cu_3 multilayer without mixing. Co 3d bands are not filled, whereas Cu 3d bands exist far below the Fermi energy. The Co 3d band at site 6 in the Co_3Cu_3 multilayer with 50% mixing does not hybridize with the neighbouring Cu 3d bands. The isolated Co 3d band at site 6 in the Co_3Cu_3 multilayer with 50% mixing becomes narrow.

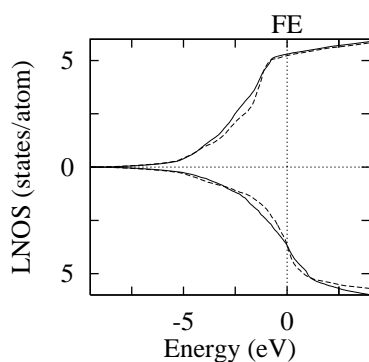


Figure 3. Calculated LNOSs at site 7 in the Co_3Cu_3 multilayer without mixing (solid line) and site 6 in the Co_3Cu_3 multilayer with 50% mixing (broken line). Upper and lower panels show the components of the majority- and minority-spin, respectively.

Figures 4(a) and 4(b) show the LDOSs at the Co sites in the Co_3Pd_3 multilayer that correspond to the sites described in figures 2(a) and 2(b), respectively. The LDOS difference between site 6 in the multilayer with 50% mixing and site 7 in the multilayer without mixing is smaller than that in the Co_3Cu_3 multilayer. The Co atom at site 6 in the Co_3Pd_3 multilayer with 50% mixing is surrounded by more Pd atoms than the Co atom at site 7 in the Co_3Pd_3 multilayer without mixing. However, since neither Co 3d bands nor Pd 4d bands are filled, the Co 3d band at site 6 in the Co_3Pd_3 multilayer with 50% mixing hybridizes with the neighbouring Pd 4d bands. The Co 3d band at site 6 in the Co_3Pd_3 multilayer with 50% mixing is not as isolated as that in the Co_3Cu_3 multilayer with 50% mixing.

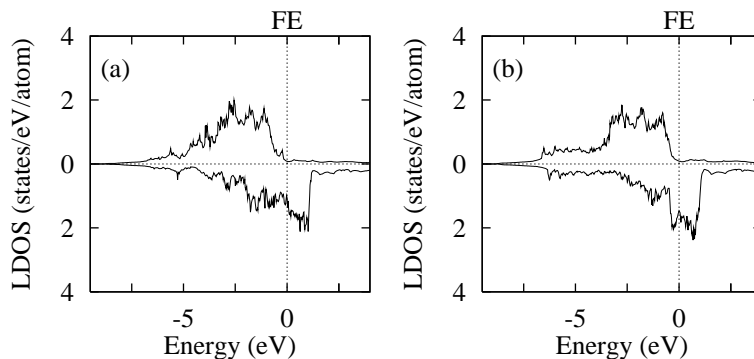


Figure 4. Calculated LDOSs at (a) site 7 in the Co_3Pd_3 multilayer without mixing and (b) site 6 in the Co_3Pd_3 multilayer with 50% mixing.

A large energy difference is calculated between site 6 in the Co_3Pd_3 multilayer without mixing and site 7 in the Co_3Pd_3 multilayer with 50% mixing. The LDOSs at these Pd sites are shown in figure 5. Figure 5(a) shows the LDOS at site 6 in the Co_3Pd_3 multilayer without mixing and figure 5(b) shows the LDOS at site 7 in the Co_3Pd_3 multilayer with 50% mixing. The spin polarization in the LDOS at site 7 in the multilayer with 50% mixing is larger than that at site 6 in the multilayer without mixing. The calculated local magnetic moments at each of the sites in the Co_3Pd_3 multilayers are shown in table 5. The Pd local magnetic moment at site 7 in the multilayer with 50% mixing is larger than that at site 6 in the multilayer without mixing.

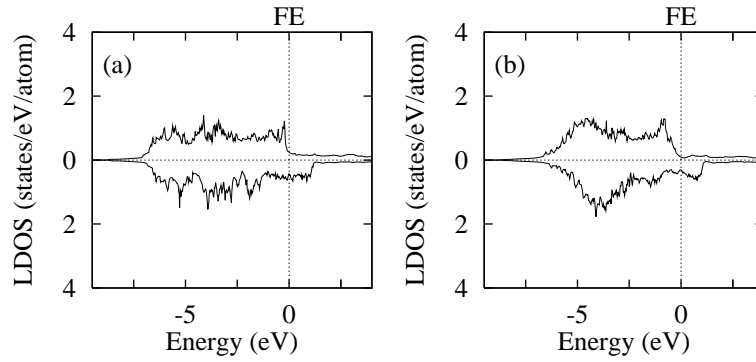


Figure 5. Calculated LDOSs at (a) site 6 in the Co_3Pd_3 multilayer without mixing and (b) site 7 in the Co_3Pd_3 multilayer with 50% mixing.

Table 5. Local magnetic moments (in μ_B) at each of the sites in the Co_3Pd_3 multilayers without mixing and with 50% mixing.

Site	No mixing		50% mixing	
	Type	Moment	Type	Moment
1	Pd	0.31	Co	1.80
2	Pd	0.31	Pd	0.36
3	Pd	0.22	Pd	0.35
4	Pd	0.22	Pd	0.35
5	Pd	0.31	Pd	0.36
6	Pd	0.31	Co	1.80
7	Co	1.82	Pd	0.39
8	Co	1.82	Co	1.82
9	Co	1.79	Co	1.80
10	Co	1.79	Co	1.80
11	Co	1.82	Co	1.82
12	Co	1.82	Pd	0.39

The large Pd magnetic moment can be explained by the following. A Pd atom at site 7 in the Co_3Pd_3 multilayer with 50% mixing has ten Co and two Pd nearest-neighbour atoms. A Pd atom at site 6 in the Co_3Pd_3 multilayer without mixing has four Co and eight Pd nearest-neighbour atoms. Thus, the Pd atom at site 7 in the Co_3Pd_3 multilayer with 50% mixing is surrounded by more Co atoms than the Pd atom at site 6 in the Co_3Pd_3 multilayer without mixing. A Pd atom at site 7 in the Co_3Pd_3 multilayer with 50% mixing is spin polarized and has a large magnetic moment due to the neighbouring Co atoms, because Pd is easily spin polarized by ferromagnetic metals.

Figures 6(a) and 6(b) show the LDOSs at the Cu sites in the Co_3Cu_3 multilayer that correspond to the sites shown in figures 5(a) and 5(b), respectively. Spin polarization is not found in either LDOS. The calculated local magnetic moments at each of the sites in the Co_3Cu_3 multilayers are shown in table 6. Cu atoms have no magnetic moments.

The Cu atom at site 7 in the Co_3Cu_3 multilayer with 50% mixing is surrounded by more Co atoms than the Cu atom at site 6 in the Co_3Cu_3 multilayer without mixing. However, the Cu atom at site 7 in the Co_3Cu_3 multilayer with 50% mixing is not spin polarized, because Cu 3d bands exist far below the Fermi energy and Cu is not easily spin polarized by ferromagnetic metals.

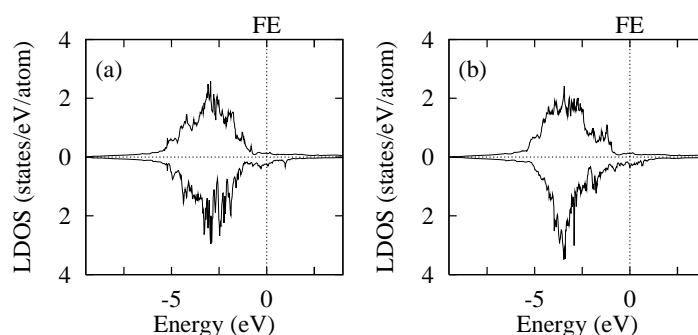


Figure 6. Calculated LDOSs at (a) site 6 in the Co_3Cu_3 multilayer without mixing and (b) site 7 in the Co_3Cu_3 multilayer with 50% mixing.

Table 6. Local magnetic moments (in μ_B) at each of the sites in the Co_3Cu_3 multilayers without mixing and with 50% mixing.

Site	No mixing		50% mixing	
	Type	Moment	Type	Moment
1	Cu	0.00	Co	1.56
2	Cu	0.00	Cu	0.00
3	Cu	-0.02	Cu	0.01
4	Cu	-0.02	Cu	0.01
5	Cu	0.00	Cu	0.00
6	Cu	0.00	Co	1.56
7	Co	1.63	Cu	0.03
8	Co	1.63	Co	1.69
9	Co	1.61	Co	1.63
10	Co	1.61	Co	1.63
11	Co	1.63	Co	1.69
12	Co	1.63	Cu	0.03

4. Summary

The electronic structures and energies of Co/Cu and Co/Pd multilayers and their change with interfacial mixing were studied by the LMTO-ASA method using the IOC mixing model. In Co/Cu multilayers, since Cu 3d bands exist far below the Fermi energy, Co atoms that are exchanged for Cu atoms by mixing have narrow 3d bands and high energies. The total energies of Co/Cu multilayers with mixing are higher than those of Co/Cu multilayers without mixing. In Co/Pd multilayers, since Pd 4d bands are not filled, Pd atoms that are exchanged for Co atoms by mixing show large spin polarization and have low energies. The total energies of Co/Pd multilayers with mixing are lower than those of Co/Pd multilayers without mixing. Abrupt interfaces appear to be stable in Co/Cu multilayers and mixed interfaces appear to be stable in Co/Pd multilayers.

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