

Interface-mediated structural evolution of immiscible Co-Cu multilayers upon solid-state reactionZ. F. Li,¹ Q. Zhang,¹ D. P. Yu,² C. Lin,¹ and B. X. Liu^{1,*}¹*Advanced Materials Laboratory, Department of Material Science and Engineering, Tsinghua University, Beijing 100084, China*²*Department of Physics, Peking University, Beijing 100871, China*

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Anomalous structural evolution was observed in the Co-Cu multilayers upon solid-state reaction, i.e., the polycrystalline Co and Cu first transformed into a fcc-structured Co-Cu superlattice, which later decomposed into a dodecagonal quasicrystalline phase. The interfacial free energy is thought to be the major driving force for intermixing and alloying between the nanosized Co and Cu layers in solid-state reactions. A detailed thermodynamic calculation is carried out to gain a better understanding of the observed alloying behavior. The possibility of improving the thermal stability of the Co-Cu multilayers for magnetic device application is also discussed.

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I. INTRODUCTION

The giant magnetoresistance (GMR) or spin valve (SV) effect in nanosized bilayers and multilayers has attracted a great deal of interest since the demonstration of their successful use as GMR reading heads¹ and as small-sized device components in magnetoresistive random access memories.² In particular, the Co-Cu multilayers exhibit the largest GMR value and a well-defined oscillatory exchange coupling at room temperature. One of the important properties of GMR materials used for reading heads is good thermal stability, so the reading heads do not change their characteristics considerably within their service time period. In this respect, there have been few papers reporting the effect of thermal annealing on the GMR of the Co-Cu multilayers.^{3–5} In general, thermal annealing of the SV materials leads to some microstructural changes in the SV and, consequently, a degradation of the magnetic as well as transport properties. In these reports, however, the annealing temperature was typically around 350 °C, which has not resulted in the formation of any Co-Cu alloy in the films.

The Co-Cu system is equilibrium immiscible and characterized by a positive heat of formation of +10 kJ/mol calculated by Miedema's model at an equiatomic stoichiometry.⁶ Because of the immiscibility, the solubility between Co and Cu is very small. Nonetheless, there have also been some reports showing some unusual phenomena at the surface/interface associated with the Co/Cu multilayers. For instance, Jesser and Matthews⁷ found that Co deposited on a Cu (001) substrate at room temperature had the same fcc structure of Cu until the deposited Co layer reached a thickness of about 2 nm and deposition of Co on a hot (350 °C) Cu substrate led to the formation of three-dimensional fcc Co crystallites. In the 1990s, Li and Tonner observed some similar growth behaviors when 20 Co monolayers were epitaxially grown on Cu (001) at room temperature.⁸ Furthermore, upon annealing at 670 K, a capping layer of Cu was formed by diffusing the Cu atoms through "pinholes" and step bands onto the free surface of the Co overlayer.^{8,9} Another recent study also showed that if deposition was conducted at 600 K, the Co nanoparticles (about 10 nm diameter) would

burrow into, instead of remaining on, the surface of the Cu (001) substrate.¹⁰

Meanwhile, another approach in materials physics research has been to study the possibility of forming metastable alloys, corresponding to the respective metastable states existing in the Co-Cu system. For instance, a metastable Co-Cu alloy was found to grow on the surface when depositing Co onto Cu at a temperature range of 700–900 K,¹¹ and mostly fcc structured Co-Cu alloys were obtained by mechanical milling¹² in epitaxial monolayers¹³ and directly after sputter deposition,¹⁴ etc. In the present authors' view, however, a thorough study of the Co-Cu system with a scheme of multilayered films covering a wide composition range upon solid-state reaction is still lacking. In other words, more metastable states may exist that may be revealed in the Co-Cu system. Besides, a clear physical picture or understanding concerning the detailed kinetics involved in the interfacial reaction taking place in the systems with positive heat of formation has not yet been reached and therefore requires much more experimental study to clarify the issue. In the present study, we investigate the possibility of the formation of metastable phases by solid-state reaction (SSR) of Co-Cu multilayered films together with comparative ion irradiation experiments. In this paper, the results as well as the respective theoretical modeling are reported.

II. EXPERIMENTAL PROCEDURE

It has been proven both by experiment¹⁵ and molecular-dynamics simulation¹⁶ that a solid-state reaction is difficult to take place at the interfaces of a metallic multilayer with close-packed atomic planes of both constituent metals parallel to the interfaces. In our experiment, to avoid the close-packed atomic plane stacking at the interfaces of the Co-Cu multilayers, single-crystal NaCl (001) substrates were therefore selected to have both polycrystalline Co and Cu in the multilayered films feature a texture of (001) orientation. Figure 1 shows the multilayer structure schematically. Pure Cu was first deposited onto the NaCl substrate, followed by depositing pure Co in an ultrahigh vacuum e-gun evaporation system with a background vacuum level on the order of 10⁻¹¹ Torr. During deposition, the vacuum level was better

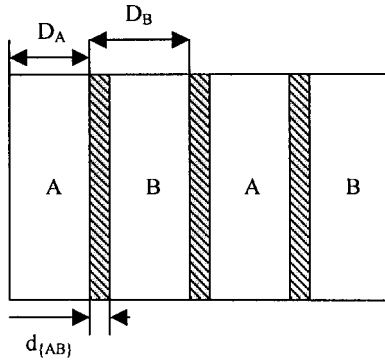


FIG. 1. A schematic configuration of the initial multilayered films with disordered interfaces.

than 1.6×10^{-8} Torr. The deposition rate was controlled at 0.5 \AA/s and no special cooling was provided to the substrates during deposition. The total thickness of the films was about 40 nm, which was required to match the projected range plus the projected-range straggling of 200-keV xenon ions employed in a comparative ion irradiation experiment. The Co-Cu multilayers were designed to consist of five Co and five Cu layers, and the desired overall compositions of the multilayered films were obtained by adjusting the relative thicknesses of the individual Co and Cu layers. Taking the $\text{Co}_{50}\text{Cu}_{50}$ multilayers as an example, the thicknesses of individual Co and Cu layers were calculated to be 39 and 41 \AA , respectively. In the present work, six multilayered films with different compositions of $\text{Co}_{15}\text{Cu}_{85}$, $\text{Co}_{20}\text{Cu}_{80}$, $\text{Co}_{35}\text{Cu}_{65}$, $\text{Co}_{50}\text{Cu}_{50}$, $\text{Co}_{65}\text{Cu}_{35}$, and $\text{Co}_{80}\text{Cu}_{20}$ were designed and prepared for investigation. After deposition, the real compositions of the films were confirmed to be $\text{Co}_{15}\text{Cu}_{85}$, $\text{Co}_{20}\text{Cu}_{80}$, $\text{Co}_{34}\text{Cu}_{66}$, $\text{Co}_{50}\text{Cu}_{50}$, $\text{Co}_{67}\text{Cu}_{33}$, and $\text{Co}_{79}\text{Cu}_{21}$, respectively, by energy-dispersive spectrum analysis, of which the measuring error was around 3–4%. These results proved that thicknesses of the individual layers in the deposited multilayers were fairly close to the designed values. It should be noted that the thicknesses of the individual layers of the above designed multilayered samples were about 4 nm and therefore the length of atomic migration through interfaces for alloying upon SSR and/or ion irradiation was in the range of a couple of nanometers. Some as-deposited films were removed from the NaCl substrates by de-ionized water and placed onto the Mo grids for thermal annealing, which was conducted in a hot stage attached to transmission electron microscopy (TEM) with a base vacuum level on the order of 10^{-7} Torr. The annealing procedure began at $200 \text{ }^\circ\text{C}$ with an increasing step of $50 \text{ }^\circ\text{C}$ and the time at each annealing temperature was at least 30 min. Meanwhile, some as-deposited films were subjected to 200-keV xenon ion irradiation in an implanter with a base vacuum level on the order of 10^{-6} Torr. The xenon ion current density was controlled to be about $0.5 \mu\text{A cm}^{-2}$ and the sample holder was always cooled by liquid nitrogen (77 K) during xenon ion irradiation. All the studied samples were analyzed by TEM bright field examination and selection area electron diffraction (SAD).

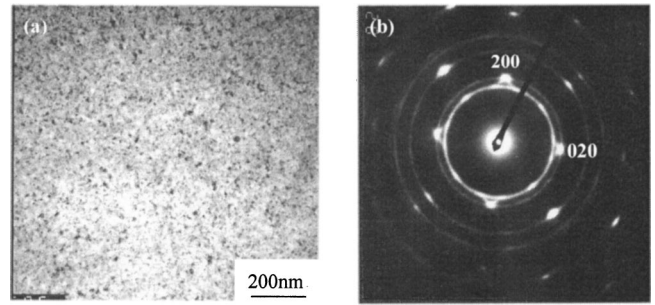


FIG. 2. The morphology (a) and the corresponding SAD pattern (b) of the as-deposited $\text{Co}_{34}\text{Cu}_{66}$ multilayered films.

III. RESULTS

We first checked the structure of the as-deposited Co-Cu multilayered films. Taking the $\text{Co}_{34}\text{Cu}_{66}$ sample as an example, Figs. 2(a) and (b) show a morphology and corresponding SAD pattern, respectively, of the $\text{Co}_{34}\text{Cu}_{66}$ multilayered films in the deposited state. From the SAD pattern, one can see not only the diffraction lines but also strong diffraction spots, confirming that a desired fcc (001) texture did emerge in the polycrystalline films. The Co-Cu multilayered films were then annealed at $300 \text{ }^\circ\text{C}$ for about 40 min and an anomalous structural transition was observed. Figure 3(a) shows a bright field image of the resultant structure after annealing and Figs. 3(b) and (c) exhibit two interesting SAD patterns taken from the films along two different axes, respectively. From the SAD patterns in Figs. 3(b) and (c), it can be deduced that the polycrystalline Co and Cu have reacted to form a unique superlattice. The superlattice was identified to be of fcc structure, in which the Co and Cu (111) close-packed atomic planes stacked in a sequence of Co/Cu/Cu, Co/Cu/Cu, and so on. Figure 3(d) displays a schematic atomic configuration of the fcc-structured superlattice.

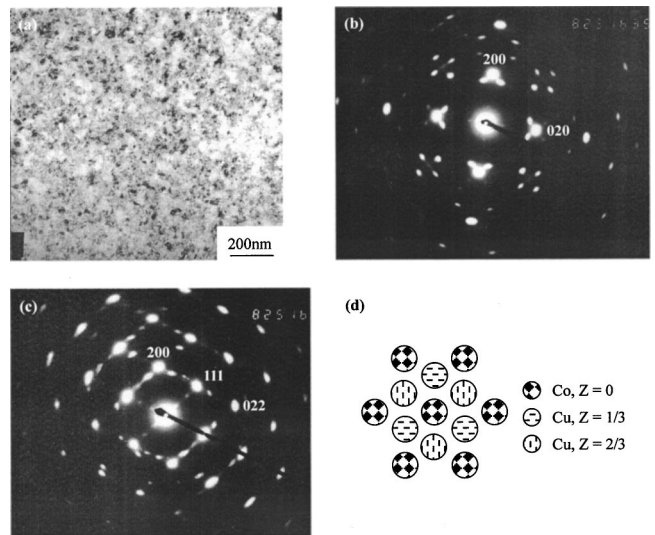
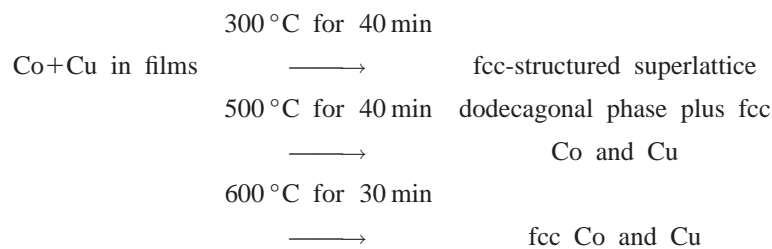


FIG. 3. (a) is the bright field image of the multilayered films after annealing at $300 \text{ }^\circ\text{C}$ for 40 min. (b) and (c) are the SAD patterns of the superlattice along the $[001]$ and $[011]$ axes, respectively. (d) is a schematic $[111]$ projection view of the atomic arrangements in the superlattice.

It is worth mentioning that after the superlattice was formed and cooled down to room temperature, it remained unchanged for more than one month. The superlattice induced in the immiscible metallic multilayer upon thermal annealing deserves much more investigation, e.g., by cross-sectional TEM examination to observe the real stacking of the atomic planes in the superlattice, etc. For some samples, thermal annealing was continued with increasing temperature up to 500 °C for 40 min; another interesting structural transition was observed as shown by a SAD pattern in Fig. 4(a). In contrast to that seen in Figs. 3(b) and (c), Fig. 4(a) shows twelvefold rotational-symmetry diffraction spots and their spatial distributions are quite analogous to the cases of the well-identified quasicrystals featuring a twelvefold rotational symmetry.¹⁷ This result indicated that the previously formed

superlattice decomposed into a metastable Co-Cu phase. From the above characterization results, a Co-Cu dodecagonal quasicrystal was formed by medium-temperature annealing of nanosized Co-Cu multilayered films, and it corresponds to a metastable state of the Co-Cu system. Noting the intensity distributions between the twelvefold rotational-symmetry diffraction spots and the diffraction spots from the fcc structure, it is concluded that the transition from the fcc-structured superlattice to the dodecagonal phase was not yet complete. Further raising the annealing temperature to 600 °C for 30 min, the corresponding SAD pattern displayed in Fig. 4(b) shows that only those diffraction signals reflected from the fcc structure can be observed and identified to be the equilibrium fcc Co and/or Cu. The above structural evolution can be summarized as follows:



According to the above structural evolution, the polycrystalline Co and Cu in the deposited multilayers first changed into a fcc-structured superlattice upon 300 °C annealing for 40 min, which was evidenced by the SAD patterns exhibited

in Figs. 3(b) and (c). From the figures, one sees that the satellite peaks in the SAD patterns are fairly bright, indicating that a more or less uniform mixing of the Co-Cu multilayer was achieved through interfacial reaction between the nanosized Co and Cu layers, resulting in the formation of a Co-Cu alloy. In the next section, the related thermodynamic and kinetic analyses will be discussed in detail to show that such uniform mixing is, in fact, driven by the interfacial free energy stored in the multilayer. Second, the fcc-structured superlattice partially decomposed and some Co-Cu grains of the dodecagonal phase were thus formed. These results suggest that the dodecagonal phase is likely to appear in a relatively large portion, rather than in a limited region close to the Co-Cu interfaces of the multilayers.

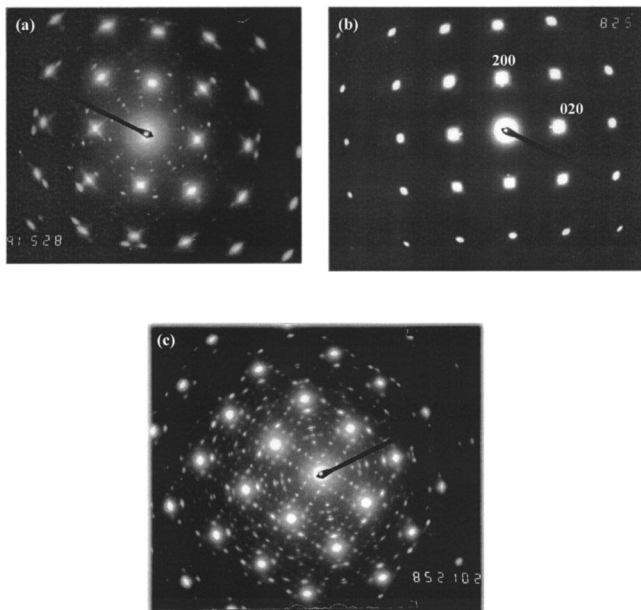


FIG. 4. (a) is a SAD pattern of the multilayered films after annealing at 500 °C for 40 min. (b) is a SAD pattern of the films after annealing at 600 °C for 30 min. (c) The SAD pattern of Co₃₄Cu₆₆ multilayered films upon irradiation at a dose of 7×10^{15} Xe⁺/cm².

The above experimental results are well reproducible, as the same fcc-structured superlattice was also observed in two other Co-Cu multilayers with overall compositions of Co₅₀Cu₅₀ and Co₂₀Cu₈₀, respectively, upon similar annealing conditions. Moreover, the same dodecagonal phase was also obtained in the Co₅₀Cu₅₀ multilayers upon annealing at 550 °C for 30 min.

It is known that ion irradiation is capable of inducing intermixing between the metal layers through dynamic atomic collision and driving the resultant mixture into a highly energetic and disordered state. In the following relaxation period, the highly energetic state would somehow relax towards equilibrium and reside in one of the possible metastable states in the system, thus obtaining the corresponding nonequilibrium solid phase.¹⁸ In the present study, 200-keV xenon ion irradiation was performed with some Co-Cu mul-

tilayered films to provide further evidence for the existence of the metastable state observed in solid-state reactions and some interesting results were obtained. In the $\text{Co}_{50}\text{Cu}_{50}$ multilayered films, the same dodecagonal phase was indeed formed after 77-K xenon ion irradiation to a dose of $5 \times 10^{15} \text{Xe}^+/\text{cm}^2$, as evidenced by the observed SAD pattern (not shown) similar to that shown in Fig. 4(a), confirming the corresponding metastable state in the Co-Cu system. Incidentally, a more interesting result was observed in another sample, i.e., in the $\text{Co}_{34}\text{Cu}_{66}$ multilayered films, a two-dimensional pattern was observed after 77-K xenon ion irradiation to a dose of $7 \times 10^{15} \text{Xe}^+/\text{cm}^2$. The pattern was assembled by the ion-induced dodecagons and featured a fourfold rotational symmetry, as shown in Fig. 4(c).

IV. DISCUSSION

We now discuss the formation mechanism of the metastable phases in the Co-Cu system upon solid-state reaction. The key issue is how to understand the first structural transition from the nanosized polycrystalline Co and Cu in the as-deposited films into a fcc-structured superlattice, as the second transition is believed to be a minor atomic adjustment realized under a medium-temperature annealing.¹⁹ Very recently, Lin, Yang, and Liu²⁰ proposed a criterion for predicting the possibility of solid-state amorphization in binary metal systems with either positive or negative heat of formation. Applying a similar principle, we perform some thermodynamic calculations to predict the formation of the metastable superlattice upon solid-state interfacial reaction in the equilibrium immiscible Co-Cu system.

Accordingly, a thermodynamic factor ΔF was defined as a Gibbs free-energy difference between a metastable superlattice phase and the initial energetic state of the multilayers. If $\Delta F < 0$, this free-energy difference will act as a thermodynamic driving force for interfacial reaction. Figure 1 has shown a schematic configuration of the initial multilayered films with disordered interfaces. The Gibbs free energy of the initial multilayers can be calculated as follows,

$$G_{\text{init}} = 2 \left(\gamma_{[A]-\{AB\}} + \gamma_{[B]-\{AB\}} + d_{\{AB\}} \frac{\Delta G_{\{AB\}}}{V_{\{AB\}}} \right), \quad (1)$$

in which $\gamma_{[A]-\{AB\}}$ and $\gamma_{[B]-\{AB\}}$ are the interfacial free energy between metal $[A]$ or $[B]$ and amorphous phase $\{AB\}$, respectively. $V_{\{AB\}}$ is the mean atomic volume of the amorphous phase. $d_{\{AB\}}$ is the thickness of the interfacial layer in a disordered state in the as-deposited films.²¹ Accordingly, $d_{\{AB\}}$ in Eq. (1) is assumed to be 0.6 nm. $\Delta G_{\{AB\}}$ is the Gibbs free energy of the amorphous layer.

The interfacial free energy $\gamma_{[A]-\{AB\}}$ between crystal A and the amorphous phase consists of an enthalpy term from the solid phase at the interface, an entropy term from the amorphous phase at the interface, and a chemical term reflecting the interaction between metal $[A]$ and amorphous $\{AB\}$,

$$\gamma_{[A]-\{AB\}} = \gamma_{[A]-\{AB\}}^{\text{enth}} + \gamma_{[A]-\{AB\}}^{\text{entr}} + \gamma_{[A]-\{AB\}}^{\text{chem}}, \quad (2)$$

in which

$$\begin{aligned} \gamma_{[A]-\{AB\}}^{\text{chem}} &= \frac{f_B^A \Delta H_{\text{int}}^{A \text{ in } B}}{C_0 V_A^{2/3}}, \\ \gamma_{[A]-\{AB\}}^{\text{enth}} &= \frac{\Delta H_A^{\text{fuse}}}{C_0 V_A^{2/3}}, \\ \gamma_{[A]-\{AB\}}^{\text{entr}} &= \frac{2RT}{C_0 V_{AB}^{2/3}}, \end{aligned} \quad (3)$$

where C_0 is a constant equal to 4.5×10^8 . ΔH_A^{fuse} is the heat of fusion of metal A .

The Gibbs free energy of the amorphous layer $\Delta G_{\{AB\}}$ can be calculated by Miedema's model,⁶

$$\Delta G_{\{AB\}} = \Delta H^{\text{amor}} - T \Delta S^{\text{amor}}, \quad (4)$$

where ΔH^{amor} and ΔS^{amor} are the formation enthalpy and entropy of the amorphous phase, respectively. T is the annealing temperature assumed to be 600 K. ΔH^{amor} is given by

$$\Delta H^{\text{amor}} = \Delta H_{\text{chem}}^{\text{amor}} + \alpha_0 (C_A T_A + C_B T_B - T). \quad (5)$$

C_i ($i=A, B$) is the atomic concentration of metal A or B . α_0 is an empirical parameter being 3.5 J/mol K, and T_i ($i=A, B$) is the melting point of component i . The $\Delta H_{\text{chem}}^{\text{amor}}$, resulting from the electron redistribution generated at the contacted surface between the dissimilar atomic cells, is expressed as

$$\Delta H_{\text{chem}}^{\text{amor}} = C_A f_B^A \Delta H_{A \text{ in } B}^{\text{int}} \quad (6)$$

where f_B^A is a function that accounts for the degree to which an atom A is surrounded by an atom B . $\Delta H_{A \text{ in } B}^{\text{int}}$ is the microscopic interfacial energy at 0 K between the cells of dissimilar components A and B . The entropy of the amorphous phase is assumed to be that of an ideal solid solution, namely,

$$\Delta S^{\text{amor}} = -R(C_A \ln C_A + C_B \ln C_B), \quad (7)$$

where R is the gas constant.

The calculation of the Gibbs free energy of the superlattice is discussed in detail. First, the superlattice has an ordered structure, and the entropy term can be ignored because it has only a minor effect on the free-energy change, such as that of an ordered equilibrium compound. The Gibbs free energy of the metastable superlattice can therefore be expressed as⁶

$$G_{\text{ms}} = \Delta H_{\text{ms}}^e + \Delta H_{\text{ms}}^s + \Delta H_{\text{ms}}^c \quad (8)$$

where the three terms on the right side are chemical, structural, and elastic contributions, respectively. The elastic term of a metastable compound (MC) phase is related to the lattice distortion caused by the size mismatch between the two constituent metals.⁶ It is well known, however, that in an equilibrium compound the elastic energy can be neglected, since the lattice distortion caused by the atomic size mismatch of the two constituent metals can be relaxed down to a minimum by an ordering arrangement of the atoms. Similarly, if an MC phase is formed near a eutectic point, where

no corresponding equilibrium compound exists, the elastic energy can also be neglected. For the Co-Cu system, there is no equilibrium compound in the equilibrium phase diagram except terminal solid solutions. The elastic term of the metastable superlattice is therefore negligible and the Gibbs free energy of the superlattice can be expressed as

$$G_{\text{ms}} = \Delta H_{\text{ms}}^c + \Delta H_{\text{ms}}^s. \quad (9)$$

The chemical term ΔH_{MC}^c is accordingly calculated by

$$\Delta H_{\text{ms}}^c = \Delta H_{\text{amp}} X_A V_A^{2/3} f_{AB}, \quad (10)$$

where ΔH_{amp} is a constant amplitude for a specific system. X_A and V_A are the atomic concentration and volume of atom A , respectively. f_{AB} is a function, namely, the extent of atom A surrounded by atom B . In the calculation of f_{AB} , a constant γ is used to describe the chemical short-range-order (CSRO) difference of the solid solution, the amorphous state, and the ordered compound.²² As an MC phase is commonly considered to be compoundlike, it is reasonable to assume that the CSRO in the MC phase is similar to that in an ordered compound and therefore γ is taken as 8.

The structural contribution is expressed by

$$\Delta H_{\text{ms}}^s = E(Z) - X_A E(Z_B) - X_B E(Z_A), \quad (11)$$

where $E(Z)$, $E(Z_A)$, and $E(Z_B)$ are the lattice stability of the MC phase and pure metals A and B , respectively, and Z , Z_A , and Z_B are the mean numbers of valence electrons in the MC phase and the numbers of valence electrons in pure metals A and B , respectively. The dependence of the lattice stability on Z for paramagnetic and ferromagnetic transition metals has been calculated by Nissen *et al.*²²

From the above deduction, the thermodynamic factor ΔF for interfacial reaction in the multilayered films can be expressed as

$$\Delta F = \frac{(D_A + D_B)(\Delta H_{\text{ms}}^c + \Delta H_{\text{ms}}^s)}{V_{\text{ms}}} - 2 \left(\gamma_{[A]-\{AB\}} + \gamma_{[B]-\{AB\}} + d_{\{AB\}} \frac{\Delta G_{\{AB\}}}{V_{\{AB\}}} \right). \quad (12)$$

The calculation results showed that the ΔF was indeed negative and was about -5 kJ/mol, which can in turn provide a thermodynamic driving force for the interfacial reaction. Kinetically, since the Co and Cu metals in the multilayered films are both of polycrystalline structures, the atomic diffusion pathways may be of those grain boundaries, other possible defects in the films, and/or even ‘‘pinholes’’ suggested in the literature.¹¹ It is worth mentioning that a kinetic factor κ was defined very recently by Lin, Yang, and Liu to correlate with a diffusivity difference between the constituent metals of the system, and the readiness of solid-state amorphization was proportional to the κ value.²⁰ Accordingly, κ of the Co-Cu system was calculated to be about 0.45. Such a κ value seems to be too small for driving a significant dif-

ferential atomic diffusion, which is necessary to induce disordering in the multilayered films. In fact, in our experiments, solid-state reactions conducted so far has not resulted in amorphization in all the studied Co-Cu multilayered films. However, such a κ may be the physical origin for crossing interfacial atomic diffusion that resulted in more or less uniform mixing between the nanosized Co and Cu layers and the formation of the metastable superlattice as well as the dodecagonal phase in the Co-Cu multilayered films.

In addition, we performed a constant-pressure molecular-dynamics (MD) simulation with a many-body Co-Cu potential derived by Mazzone, Rosato, and Pintore²³ to investigate the atomic mixing between the Co and Cu metals and the stability of the metastable Co-Cu phase. In simulation, we constructed a uniformly mixed Co-Cu solid solution of fcc structure in a composition range of 10–80 at% of Co. After running the MD simulation for several hundred picoseconds, all the related dynamic variables showed no secular variation, this suggesting that such a structure could remain unchanged, i.e., such a Co-Cu alloy was likely to be stable against spontaneous decay.

According to our experimental results and MD simulations, the metastable phase features relevant thermal stability and is therefore likely to be encountered in the Co-Cu system upon moderate-temperature annealing. It is generally agreed that GMR is associated with the reorientation of the magnetic moments in the neighboring magnetic layers and has been interpreted on the basis of spin-dependent electron scattering at the magnetic/nonmagnetic interface and in the bulk of the magnetic layer.²⁴ If the microstructure of the multilayered films changes, the characteristics of the GMR will also vary accordingly. In this sense, the thermal stability of the GMR materials is an important factor in determining their suitability for device applications. The respective multilayered structures should be able to survive high-temperature materials processing, such as photoresist baking, and they must survive some unexpected change due to the operating environment or high operating current densities. In short, despite the immiscibility between Co and Cu, the interfacial free energy stored in the multilayered films can promote the solid-state interfacial reaction between the Co and Cu metals upon moderate-temperature annealing and should be taken into account when preparing the Co-Cu multilayers for some specific applications.

V. SUMMARY

(1) In the equilibrium immiscible Co-Cu system, the properly prepared Co-Cu multilayered films are able to undergo a solid-state interfacial reaction upon annealing at a moderate temperature for a relatively short time resulting in the formation of a fcc-structured superlattice and/or a dodecagonal quasicrystal phase.

(2) According to the calculation, if the individual layer thickness was on the order of a nanometer, formation of the metastable fcc-structured superlattice as well as the dodecagonal phase in the multilayered films upon solid-state reaction is thermodynamically and kinetically possible.

(3) Technically, our results seem to suggest that to improve the thermal stability of the Co-Cu multilayered films for magnetic device application, one should consider choosing an appropriate preparation method with relevant parameters to decrease the excess interfacial free energy in the films.

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