Double quantum well states in Cu/Co/Cu grown on Co(001)

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A double quantum well (QW) system of Cu/Co(1 ML)/Cu grown on Co/Cu(001) has been investigated by photoemission spectroscopy. Energy spectra of the valence band from one of the Cu QW's have been measured as a function of the other Cu QW thickness. The results show strong resonance between the two Cu QW states across a 1 ML Co barrier. In particular, we observe that quantum well coupling removes the degeneracy of the two Cu QW states, resulting in a state crossing effect. A phase accumulation model is developed to explain these observations.

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The ability to grow atomically flat thin films opens a great opportunity for nanostructures research, especially for electron confinement or quantum well (QW) states in metallic layered structures.[1] Because of the role of electrons near the Fermi level in the electronic properties of materials, spin-polarized valence electron QW states in magnetic nanostructures[2,3] have attracted wide interest recently in the study of magnetoelectronic properties in nanostructures. A specific example is the intrinsic connection between QW states and oscillatory interlayer coupling[4] in magnetic multilayers. The photoemission technique plays a key role in such studies because QW states in momentum space can be resolved by performing angle resolved photoemission spectroscopy (ARPES).[5,6] By measuring the momentum-resolved QW states, the physical origin of the long- and short-period oscillations of the interlayer coupling as well as the relationship between the two oscillations have been unambiguously identified.[7,8]

In spite of this progress, most of the QW studies have been focused on a single QW and its relation to the interlayer magnetic coupling. As coupling of electrons from different layers generates new properties not realizable in individual layers, it is important to investigate the electronic interaction between QW's in nanostructures. Recent results on magnetic tunnel junctions show that the insertion of a QW film between the ferromagnetic and insulating layers can significantly alter the magnetoresistance of the junction,[9,10] suggesting that interaction of electrons between different layers may play an important role in the overall electronic states of a nanostructure. Photoemission results also indicate that the electron wave function in one QW can tunnel across a thin barrier layer to interact with the wave function in another QW.[11,12] While these discoveries are promising, it is unclear how new electronic states are generated by the interaction between QW's. For instance, it is yet unclear if the phase accumulation model (PAM),[13] which successfully describes single QW systems, can be extended to multi-QW systems.

In this Brief Report, we report photoemission results on a Cu/Co(1 ML)/Cu double QW system grown on Co(001). We choose the Cu/Co system because of the following reasons. First, Cu/Co(001) has become a representative system for studying QW's in magnetic nanostructures.[2,3,12,14] Second, Cu has a simple Fermi surface whose sp band can be easily separated from the other energy bands. Finally, Cu and Co grow epitaxially on each other in the (001) orientation, giving rise to an atomically flat interface in this system.[15,16] Photoemission results show that as QW states from the two individual Cu layers reach the same energy level, the coupling between the two QW's splits the degeneracy to result in state crossings. We extend the PAM by matching the boundary conditions at all interfaces of the double QW system and successfully reproduce this state crossing effect.

A Cu(001) single crystal was prepared by mechanical polishing down to 0.25 μm diamond paste followed by electrochemical polishing. The Cu crystal was then cleaned *in situ* with cycles of Ar ion sputtering at 1.5 keV and annealing at $\sim 600\,^{\circ}$C. Co and Cu layers were subsequently deposited at room temperature by molecular beam epitaxy at 0.7 ML/min and 1.7 ML/min, respectively. Thickness of the film was monitored by a quartz crystal oscillator, the thickness reading of which is consistent with the reflection high energy electron diffraction oscillations[17] and the periodicity of quantum well oscillations.[18] 15 ML Co was grown on Cu(001) as the ferromagnetic base layer, on top of which a Cu/Co(1 ML)/Cu sandwich was grown to form a double QW. The two Cu layers were grown as perpendicular wedges for independent thickness variation of the two Cu QW's. Photoemission measurements were performed at beam line 7.0.1.2 of the Advanced Light Source at Lawrence Berkeley National Laboratory. The beam spot size of 50–100 μm gave a thickness resolution of better than 0.4 ML on the wedged sample. Normal emission with 83 eV photon energy was used to select the electronic states near the belly of the Cu Fermi surface; quantum well states derived from the sp band with Δ_1 symmetry were thus probed with this geometry.[5]

We first present the photoemission results of the double QW with the outer Cu well thickness (d_{out}) fixed. Figure 1 shows a series of photoemission spectra at $d_{\mathrm{out}}=17$ ML with different inner Cu thicknesses (d_{in}). The dominant features of these spectra are the three peaks located at 0.1, 0.8, and 1.4 eV below the Fermi level (dotted vertical lines) that coincide with the three QW states in a 17 ML Cu film grown on Co(001) ($d_{\mathrm{in}}=0$ ML). Additional features are observed in the spectra. First, the intensities of the three dominant QW peaks vary with the inner Cu film thickness. Second, additional peaks are present in the spectra, the positions of which

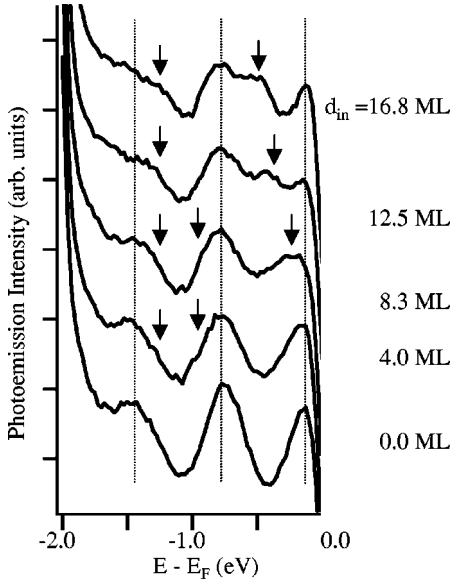

FIG. 1. Photoemission spectra at fixed outer Cu layer thickness of 17 ML with various inner Cu layer thickness. Dotted lines show the original states of the 17 ML Cu/Co single quantum well in the absence of an inner Cu layer. Arrows point at additional features arising from the inner Cu layer at various thickness.

depend on the inner Cu film thickness (indicated by arrows in Fig. 1). Since photoemission measures the outer Cu layer with a probing depth of only ~5 ML, the dependence of the energy spectra on the inner Cu film implies that the electronic states of the two Cu QW's are coupled. This is consistent with our recent results on the symmetric double QW Cu/Ni(1 ML)/Cu, which shows that electronic coupling of two Cu wells lifts the degeneracy of QW states.[12]

To gain a detail understanding of the double QW interaction, we measured the energy spectra as a function of the inner Cu thickness at a fixed outer Cu thickness [Fig. 2(b)] as well as energy spectra as a function of the outer Cu thickness at a fixed inner Cu thickness [Fig. 2(c)]. In this way, we tune the energy levels of one Cu QW by changing the well width while fixing the energy levels of the other Cu QW. Spectra of single Cu QW on Co(001) are shown in Fig. 2(a) as a reference. The dashed lines in Fig. 2(a) are calculated results from PAM.[13] To see the effect of the double QW interaction, we plot in Figs. 2(b) and 2(c) (dashed lines) the locations of QW states that correspond to isolated inner and outer Cu QW's. Recalling that photoemission probes only the outer Cu electronic states, we would expect Figs. 2(b) and 2(c) to give results of the isolated outer Cu QW only (dashed line labeled by ν_{out}) if the two Cu QW's were totally decoupled. The fact that the results shown in Figs. 2(b) and 2(c) are distinguishable from the isolated QW states prove the existence of electronic coupling between the two Cu QW's.

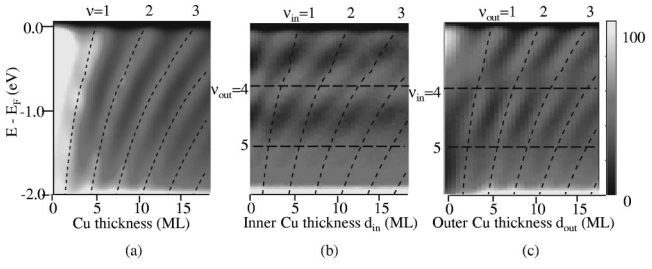

FIG. 2. (a) Photoemission spectra in Cu/Co(001) structure. Dashed lines are calculated results from the phase accumulation model. Photoemission intensity as a function of (b) the inner Cu thickness at fixed outer Cu layer thickness of 17 ML, and (c) the outer Cu thickness at fixed inner Cu layer thickness of 17 ML. Dashed lines show the isolated outer (labeled by quantum number ν_{out}) and inner (ν_{in}) single quantum well states, respectively.

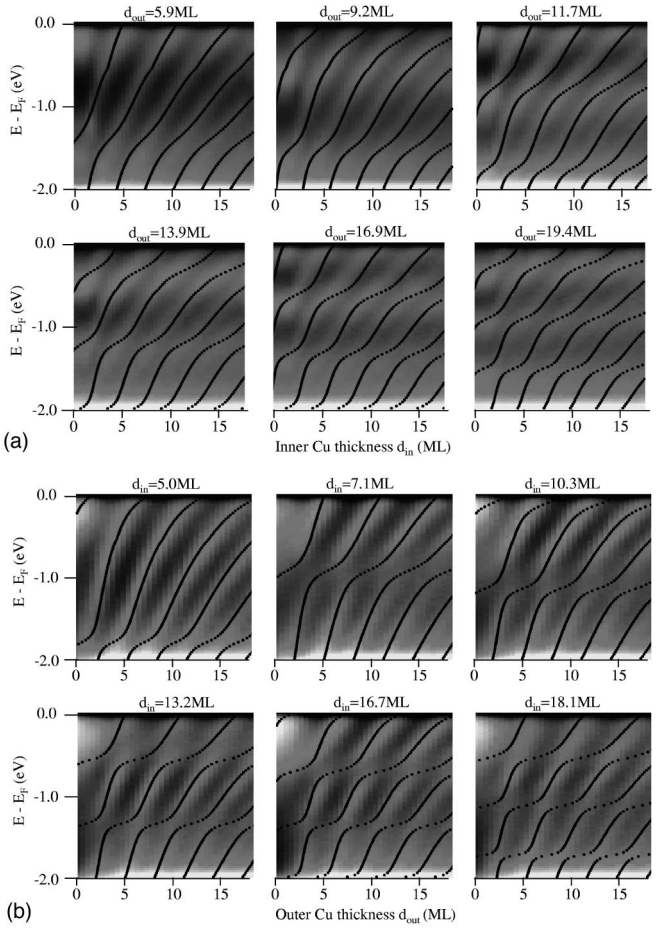

FIG. 3. Photoemission intensity as a function of (a) the inner Cu thickness at various fixed outer Cu layer thickness and (b) the outer Cu thickness at various fixed inner Cu layer thickness. Dotted lines show the calculated results from the phase accumulation model for the double quantum well states.

Let us look at Fig. 2(b). The QW states in this case evolve with the inner Cu thickness in a way as to avoid the crossing points of states from the two isolated QW states. As the crossing points correspond to degeneracy of the two isolated QW's, the results of Fig. 2(b) can be easily understood since coupling of two degenerate states would lead to symmetric and antisymmetric states, which lift the energy degeneracy. This leads to an avoided-crossing behavior such that a ν_{in}-like state evolves continuously into a ($\nu_{\mathrm{in}+1}$)-like state as it passes a ν_{out}-like state, where "ν_{in}- (ν_{out}-)like state" refers to a state derived from the isolated inner (outer) well with

quantum number ν_{in} (ν_{out}). This is exactly what we observed in Fig. 2(b). Similar state-crossing behavior also occurs in double QW's with fixed inner Cu thickness [Fig. 2(c)].

To confirm that the state crossing occurs where the isolated inner and outer wells have degenerate states, we took a series of spectra as a function of the inner Cu thickness at various fixed outer Cu thickness [Fig. 3(a)] and as a function of outer Cu thickness at various fixed inner Cu thickness [Fig. 3(b)]. It is obvious that state crossings always occur near the energy levels of the QW states in the fixed Cu layer, supporting our analysis.

For a quantitative understanding of the state crossing, we extended the PAM to calculate the quantization condition of the double quantum well states. In the PAM, the QW problem is reduced to that of an electron in a square potential well where the continuity of the wave functions at the boundary is embodied in a QW phase factor. For single Cu QW states, mismatching of minority spin energy bands between Cu and Co determines the Cu/Co phase ϕ_{Co}, and the image potential at the vacuum interface determines the Cu/vacuum phase ϕ_{vac}.[13] For double QW's, additional boundary conditions need to be satisfied at each side of the middle Co barrier within which the wave function is a superposition of exponentially decaying wave functions from the two Cu QW's. If the middle Co barrier is infinitely thick, the two wave functions decaying from the two Cu/Co interface will have negligible overlap inside the Co barrier and this automatically brings back the single QW case. For ultrathin Co barriers, however, the significant overlap of the two decaying wave functions inside the Co correlates the boundary conditions at the two Co/Cu interfaces so that QW states in the two Cu layers have to be adjusted to match the correlated boundary conditions. This is the physical origin of the QW interaction. By introducing a phase ϕ to relate the decaying wave vector κ in the Co barrier to the electron wave vector k in the Cu layer such that $\kappa = -k\tan(\phi/2)$, we found the quantization condition of the double QW's, given by

$$\tan\left(k_{eff}d_{out}-\frac{\phi_{vac}+\phi}{2}\right)=\frac{f(L,k,\phi)\sin\left(k_{eff}d_{in}-\frac{\phi_{Co}-\phi}{2}\right)}{\sin\left(k_{eff}d_{in}-\frac{\phi_{Co}+\phi}{2}\right)-f(L,k,\phi)\cos\left(k_{eff}d_{in}-\frac{\phi_{Co}-\phi}{2}\right)}, \tag{1}$$

where

$$f(L,k,\phi)=\frac{\sin\phi}{\exp\left[-2kL\tan\left(\frac{\phi}{2}\right)\right]-1}.$$

Here $k_{eff}=k_{BZ}-k$, where k_{BZ} is the magnitude of the Brillouin zone vector along [001] direction, and L is the width of the Co barrier. A similar equation for the inner QW can be obtained with a simple variation of Eq. (1). The coupling between the two QW's manifests in Eq. (1) most prominently in the factor $f(L,k,\phi)$. In the limit of infinitely thick Co barrier, $f(L,k,\phi)$ approaches zero ($-\pi<\phi<0$ in PAM) so that the solution of Eq. (1) is $2k_{eff}d_{out}-\phi_{vac}-\phi=2\pi\nu_{out}$, which is the expected single QW solution. In the limit of zero thickness Co barrier, $f(L,k,\phi)$ approaches infinity so that Eq. (1) yields the solution $2k_{eff}(d_{out}+d_{in})-\phi_{vac}-\phi_{Co}=2\pi\nu$, with the inner and outer Cu layers joining together to form a single QW. For $L\sim 1/k$, which corresponds to our experimental condition, Eq. (1) describes the state crossing behavior due to QW coupling. For a numerical evaluation of Eq. (1), experimental values d_{in} and d_{out} are substituted and k_{eff}, ϕ_{Co}, ϕ_{vac} are calculated using the same method as that of Kawakami *et al.*[18] We substitute ϕ with the bulk Co/Cu value ϕ_{Co} and use L as a fitting parameter (we will discuss the validity of this operation later). The calculated results are shown as dashed lines with $L=1.0\pm0.3$ Å for all spectra in Figs. 3(a) and 3(b). The general trends of the double QW states, especially the state crossing behavior, are reproduced reasonably well. Therefore, we conclude that the state crossing comes from the overlap of QW wave functions inside the Co barrier.

Despite the overall agreement, the calculated curves deviate quantitatively from the experimental data. We believe that comes from the assumption in modeling the 1 ML Co barrier. Even though a nearly perfect 1 ML Co interface layer should be obtained in this system under our growth condition,[16,19] a square well potential used in the PAM is still an oversimplification. First, we notice that the fitting value of L is smaller than our experimental value of 1 ML (1.8 Å), which is a sign that the step potential may no longer be a good approximation for a monolayer-thick Co film. This shortcoming arises since using a step potential assumes a complete change of the Cu-to-Co electron wave functions at a sharp boundary, which should break down in the monolayer limit. When a step potential is casually used without justification, the potential width L would be reduced from 1.8 Å in modeling the 1 ML Co as the more localized Co d band is expected to draw the Co/Cu electronic boundary much closer to Co than the itinerant s band in Cu. Second, the value of ϕ for 1 ML Co should be different from the bulk value of ϕ_{Co}. As x-ray magnetic linear dichroism measurements show that 1 ML Co is not ferromagnetic, an averaged potential barrier of the minority and majority energy gaps should be used to account for the spin fluctuations. Nevertheless, we find that changing ϕ by varying the energy gap from minority-spin energy gap to majority-spin energy gap do not yield a significantly different Co barrier thickness for

the best fitting in Fig. 3. We thus leave the discussion of ϕ as a topic for future study. Spin-dependent QW measurement will be especially illuminating. Finally, the potential energy barrier of Co comes from the *s*-*d* hybridized energy gap that is located at $k \neq 0$, which should complicate the boundary condition between Co and Cu. Tight-binding calculation shows that the *s* and *d* orbitals have different electronic boundaries and that the orbitals exhibit oscillatory character within the energy gap.[13] A more quantitative understanding of the double QW states requires theoretical calculation of the 1 ML Co electronic structure. Nevertheless, disregarding the detailed structure of the interface, the 1 ML Co has unequivocally formed a barrier coupling two QW's and our simple model has successfully described the overall qualitative behavior of the double QW system.

In summary, we investigated double QW states in Cu/Co(1 ML)/Cu grown on Co(001) with photoemission. Thickness dependent measurements show clear evidence of the coupling between the two Cu QW's across the Co barrier. In addition, the evolution of double QW states shows state crossing behavior near degenerated energy levels of the corresponding two isolated Cu QW's. The phase accumulation model has been developed to explain the double QW state evolution. It has been shown that the QW coupling originates from the overlap of the wave functions from the two Cu QW's in the Co barrier.

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