Determination of spin-polarized quantum well states and spin-split energy dispersions of Co ultrathin films grown on Mo(110)

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Abstract

Co thin films were epitaxially grown on Mo(110) and investigated by spin-polarized low-energy electron microscopy (SPLEEM). We find that the spin asymmetry of the electron reflectivity from the Co film alternates its sign as a function of both the electron energy and the Co film thickness as a result of spin polarized quantum well states in the Co film. By measuring spin dependent quantum well states, we are able to resolve the spin-split energy dispersions of the Co film precisely. The determined spin resolved energy bands are further tested by fitting the quantum well states using the phase accumulation model (PAM), and the result shows an excellent agreement between the fitting and the experimental data.

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Electron confinement at the nanometer scale leads to the formation of quantum well states (QWS) that split the electron energy into discrete levels. Research on this subject was initially focused on the effect of QWS on the charge properties of electrons in nanostructures. For example, it was found that the formation of QWS in a thin film could generate phenomena such as "electronic growth" [1,2,3], superconductivity oscillation [4,5,6], Schottky barrier modulation [7], and oscillations of the surface adatom diffusion barrier [8], surface reactivity [9,10], the work function [11,12,13,14], etc. Later it was recognized that QWS also have an important effect on the spin properties of nanostructures. For example, it was shown that the oscillatory magnetic interlayer coupling in a magnetic sandwich can be attributed to the QWS in the spacer layer [15,16]. It was also shown that the Kondo effect of magnetic adatoms could be modulated by QWS in a thin film under the adatoms [17,18]. Even the Rashba spin-split surface state was shown recently to interact with the QWS of a thin Ag film [19]. Noticing that an electron consists of charge and spin freedoms, it is generally believed that the integration of spin and charge freedoms in a nanostructure is a promising future direction for next generation applications in spintronics technologies [20]. Then the interesting question is, how do spin and charge freedoms interplay at the nanometer scale? Although manifested as spin alignment in magnetic materials, the magnetic exchange interaction that drives spin-charge correlation actually originates from the electron-electron Coulomb interaction. It was Heisenberg and Dirac who pointed out that it is quantum mechanical symmetry requirements for identical particles that lead to the exchange interaction by lifting the spin degeneracy in electron energy dispersions [21,22]. Therefore to develop the spintronics research, one of the most important experimental tasks is to determine the spin-split electronic energy dispersions in nanostructures. In fact, it has been a long-standing goal to determine the spin resolved band dispersions since the early stage of research on magnetic ultrathin films [23]. One popular and effective experimental technique for energy band determination is angle resolve photoemission (ARPES) [24]. However, spin-resolved photoemission encounters the difficulty of limited efficiency in detecting the spin-polarization of photoemitted electrons. Typically, efficiency is at least three orders-of-magnitude lower than that of spin-integrated measurements, thus limiting the performance of ARPES for spin-resolved energy dispersion measurements [25]. In practice, spin-resolved ARPES is not applied as widely as spin-integrated ARPES [26,27] and measurements on spin polarized QWS in nanostructures [28] remain rare. An alternative approach was developed recently by performing low energy electron reflectivity measurements on thin films. Adding spatial resolution, this technique is known as Low Energy Electron Microscopy (LEEM) [29]. When the incident electron

beam is spin-polarized, Spin Polarized Low Energy Electron Microscopy (SPLEEM) can probe the spin-resolved electronic states in real time at film growth speed. While the possibility to observe QWS in magnetic thin films by SPLEEM was illustrated in early work [30], the suggestion to explore the dispersion of unoccupied electronic bands from SPLEEM measurements was made more recently [31]. Meanwhile the technique has also been demonstrated to be able to reveal spin polarized QWS in nonmagnetic ultrathin films grown on ferromagnetic substrates [32,33]. In this Letter, we report our study using SPLEEM on ferromagnetic Co thin films grown on Mo(110) substrate. We show that the high quality growth of the Co films enables a real-time layer resolved measurement of spin polarized QWS in the Co layer during film growth, from which the spin-split energy dispersions of the Co film can be determined unambiguously.

The experiment was performed at the SPLEEM user-instrument at Lawrence Berkeley National Laboratory. A Mo(110) single crystal disk was cleaned by cycles of flashing the substrate in oxygen (10^{-8} Torr) at 2200 K to remove carbon contamination from the Mo substrate. Co films were grown on the Mo substrate at room temperature by e-beam evaporation, and the pressure remained below $3x10^{-10}$ Torr during the film growth. Co grows epitaxially on Mo(110) in the Nishiyama-Wassermann orientation, i.e., with the hcp Co hexagonal base plane Co(0001) parallel to Mo(110) [34]. Our Low Energy Electron Diffraction (LEED) result confirms the epitaxial single crystalline growth of Co on Mo(110) at least up to 15ML (Fig. 1), in agreement with literature results [35]. For the SPLEEM measurement, a spin-polarized low energy electron beam is produced by illuminating a GaAs (100) surface, activated with Cs and O_2 to negative electron affinity, with circular polarized light from a diode laser. The instrument is provided with electron optics to conveniently adjust azimuthal and polar orientation of the polarization of the incident electron beam [36]. The spin polarized electron beam is directed at the sample surface at normal incidence, and the specular beam is magnified in an electron-optical column to form a real-space image of the sample. Energy spectra were taken continuously during the Co film growth, sweeping the energy range at a rate of typically 1 sweep/minute. The typical growth rate used in our experiment was ~0.1ML/min, so that the whole spin dependent measurement can be finished in about 2 hours for a 12ML thick Co film.

Under illumination with varying azimuthal and polar orientation of the spin polarization of the incident electron beam we find that magnetic domain contrast is maximized as the electron spin is in-plane and parallel to the Mo $[1\,\bar{1}\,0]$ axis, showing that the Co magnetization easy axis in the Co/Mo(110) system is parallel to the Mo $[1\,\bar{1}\,0]$ axis, in agreement with the Magneto-Optic Kerr Effect measurement result [35]. Spin

resolved energy spectra were taken by alternating the incident electron spin direction parallel and antiparallel to the Co magnetization direction during the Co film growth. Fig. 2(a) and (b) show representative spin resolved electron reflectivity spectra. Both spin-up and spin-down electron reflectivities oscillate with the Co film thickness at fixed electron energy, and oscillate with the electron energy at fixed Co thickness. The observed oscillations clearly show the existence of QWS in the Co film.

To better illustrate the spin dependence of the QWS, we define the spin asymmetry as the difference between spin-up and spin-down electron reflectivity, $A \equiv (R_{up} - R_{down})/(R_{up} + R_{down})$, where R_{up} and R_{down} are the reflectivity for spin-up and spin-down electrons, respectively. Fig. 2(c) shows the spin asymmetry as a function of the electron energy and the Co film thickness. We found a clear non-vanishing spin asymmetry throughout the energy and thickness ranges studied. Moreover, the asymmetry oscillates with both the electron energy and with Co film thickness. This is a clear evidence of the spin polarization of the QWS in the Co film, i.e. electron interference conditions in the Co film depend on the electron spin direction relative to the Co magnetization. In fact, the spin asymmetry not only oscillates in magnitude, but also alternates between positive and negative signs. To ensure that the sign change in the spin asymmetry is not an artifact of different incident intensities for spin-up and spin-down electrons, we located an area containing a 180 degree magnetic domain wall and performed the experiment again by fixing the incident electron spin direction. The domain imaging result (Fig. 3) shows that the domain contrast indeed alters its sign by changing the electron energy or by changing the Co film thickness. This result proves rigorously that the spin asymmetry of the reflectivity results from polarization of the QWS in the Co film. The alternation of the domain contrast sign in a Co thin film can be explained by the fact that constructive interference from the two surfaces of the Co thin film is satisfied at different conditions for spin-up and spin-down electrons, so that spin-up and spin-down electron reflectivities are enhanced or suppressed at different conditions.

The QWS in a thin film are generally described by the so-called phase accumulation model (PAM) which gives the quantization condition of [16]

$$2kd_{Co} + \phi = 2n\pi \qquad \text{n=integer} \qquad (1)$$

Here d_{Co} is the Co film thickness, k is the electron wave vector, and ϕ is the phase accumulation of the electron upon reflection at the two boundary surfaces of the Co film. Since the phase ϕ depends on the electron energy only [16], the electron wave vector at a

given energy should then depend only on the thickness periodicity (Λ) of the QWS oscillations at that energy: $k(E) = \pi / \Lambda(E)$. Therefore, a precise determination of the oscillation periodicity at different energies will give the *k-E* relationship, i.e., the energy band dispersion of the film, without the need of any model fitting. Then for a ferromagnetic thin film, a precise determination of the spin-dependent QWS will give the spin-split energy bands. Fig. 4(a) and (b) show the Co QWS for spin-up and spin-down electrons, respectively. QWS peaks appear at integer multiples of the Co atomic layer thickness, this is seen particularly clearly in the case of spin-up electrons. This result not only shows the atomically flat quality of the film [2], but also ensures a precise determination of the Co film thickness, which is crucial to the determination of the energy band dispersion $k(E) = \pi / \Lambda(E)$. This high quality QWS measurement for both spin-up and spin-down electrons permits clear resolution of the energy bands for spin-up and spin-down electrons [Fig. 4(c)]. *ab-initio* calculation of the spin-split Co energy bands show inconsistent results in the literature. Among the published calculation results, we find one result [solid lines in Fig. 4(c)] that agrees mostly with our experimental data [37]. Even there is an overall agreement after shifting the theoretical result by 2.5 eV, there is a notable difference between the theoretical and the experimental results at higher electron energy. This discrepancy could either come from insufficient accuracy of the band structure calculation or from the fact that the energy bands of a Co thin film could be different from those of bulk Co. To estimate the accuracy of our experimentally determined energy bands, we compare the QWS maxima seen in Fig. 4(a) and (b) with constructive interference resonances predicted from eqn. (1), using the spin resolved energy bands in Fig. 4(c) and assuming a linear dependence of the phase shift on the energy [16]. The predicted resonances [dots in Fig. 4(a) and (b)] are in excellent agreement with the experimentally observed reflectivity maxima.

In summary, we studied spin dependent reflectivity from Co thin films grown on Mo(110) using SPLEEM. From our spin resolved observations of QWS and the atomic layer precise film thickness control spin resolved energy bands in the Co films were derived without the need of any model fitting. The spin-up and spin-down energy bands agree reasonably well with an *ab-initio* calculation result, while we also observe a noticeable discrepancy at higher energy.

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Figures:

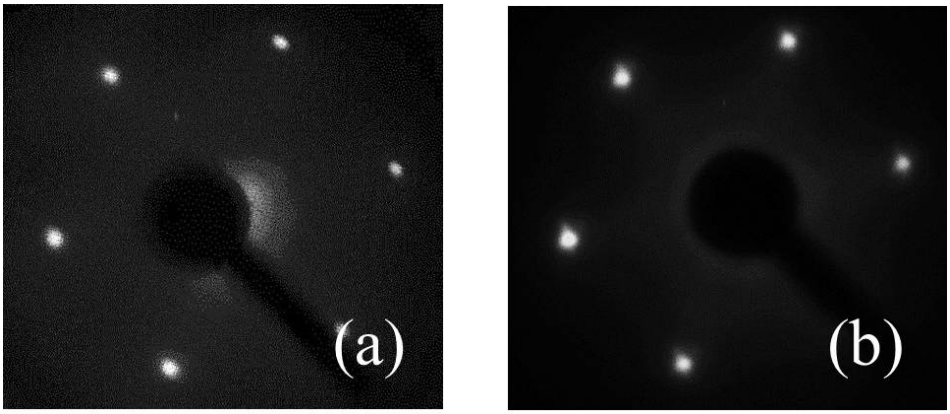

Fig.1: LEED patterns of (a) 9ML and (b) 15ML Co films grown on to Mo(110) substrate taken at 140 eV electron energy.

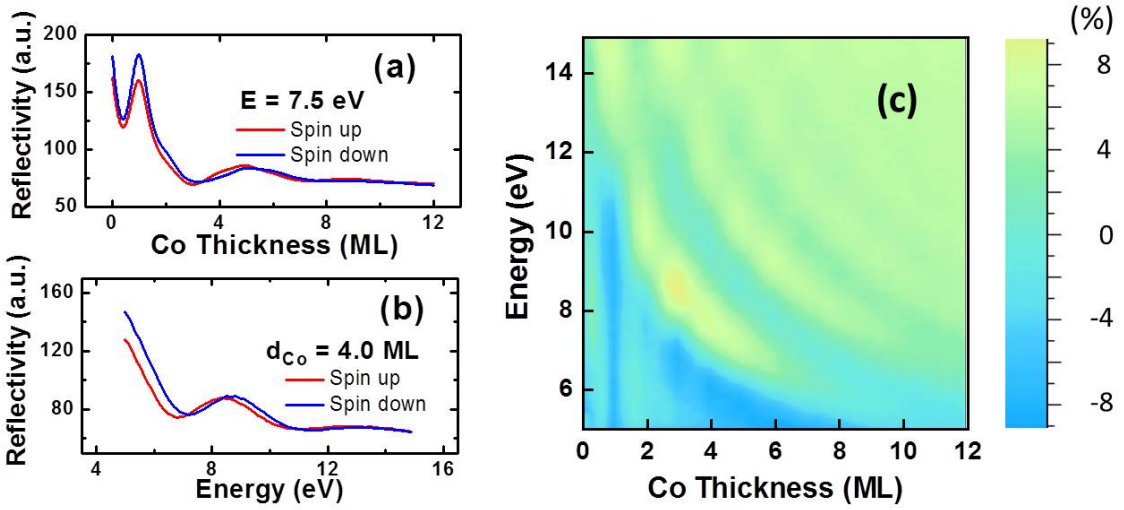

Fig.2: (color online) Spin-dependent electron reflectivity (a) as a function of Co thickness at 7.5 eV electron energy, and (b) as a function of electron energy at 4.0 ML Co thickness. (c) The spin asymmetry of the reflectivity oscillates with the electron energy and the Co thickness.

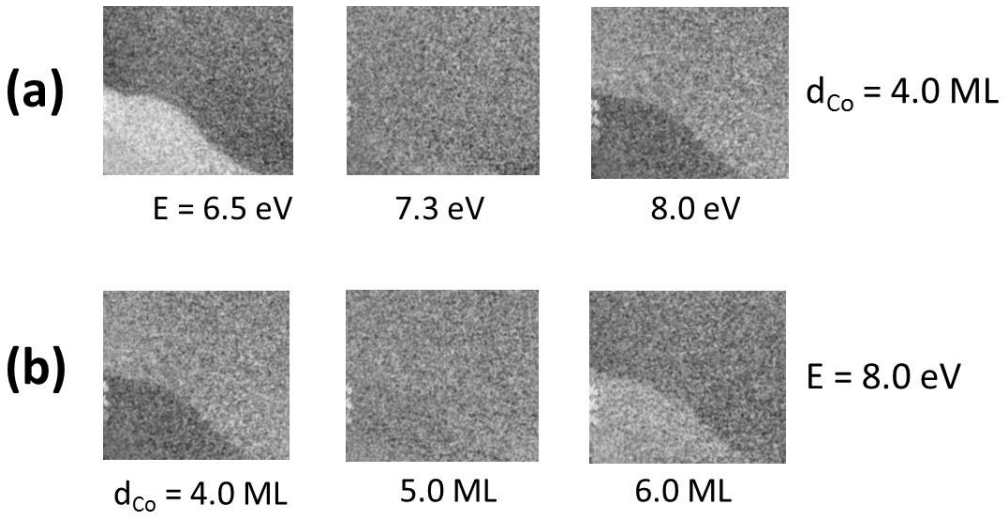

Fig.3: Co magnetic domain image (5μm×5μm) (a) as a function of electron energy at 4ML Co thickness, and (b) as a function of Co thickness at 8.0 eV electron energy. The domain contrast reversal shows clearly the spin polarization of the quantum well states in the Co film.

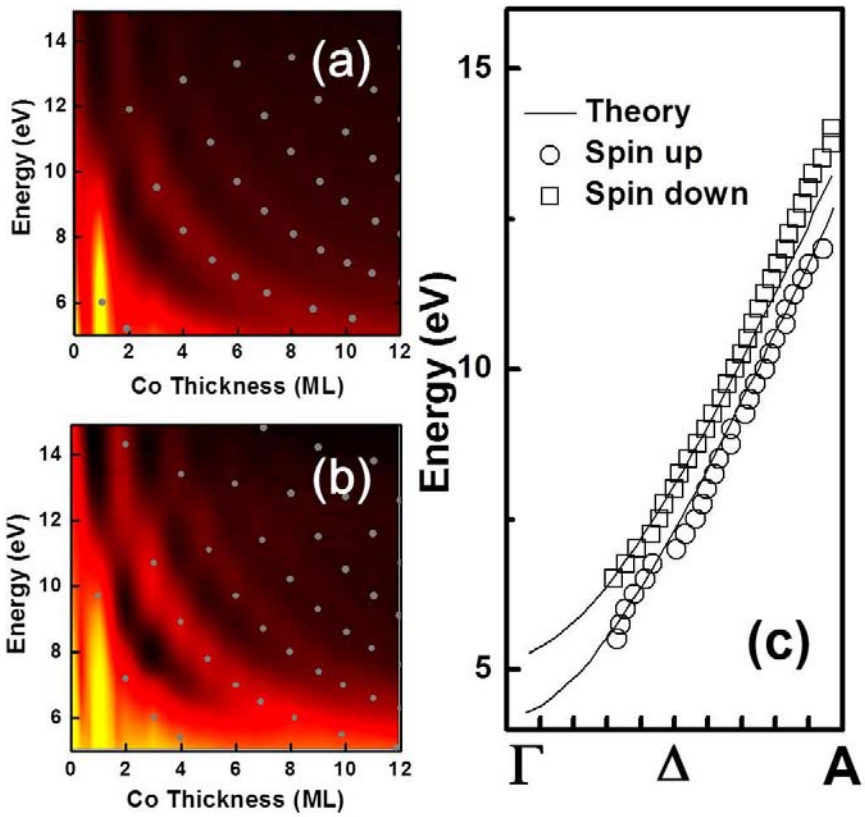

Fig. 4: (color online) Electron reflectivity for (a) spin-down electrons, and (b) spin-up electrons shows layer resolved quantum well states. (c) Spin-split energy bands retrieved from the spin-polarized quantum well states.

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