

Off-gap interface reflectivity of electron waves in Fabry-Pérot resonators

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(Received 19 September 2008; revised manuscript received 12 December 2008; published 23 January 2009)

The interface reflectivity in the Mg/W(110) metallic quantum well is determined from line-shape analysis of high-resolution photoemission spectra. Moreover, a quick reflectivity drop is found away from projected band gaps of the appropriate symmetry near E_F , such that the interface reflectivity overcomes the bulklike quasi-particle lifetime as the dominant line broadening mechanism at the Fermi energy. A nearly-free-electron model calculation for the W(110) substrate band structure demonstrates that coherent wave-function scattering is the relevant mechanism that determines the interface reflectivity in the resonator.

DOI: [10.1103/PhysRevB.79.033410](https://doi.org/10.1103/PhysRevB.79.033410)

PACS number(s): 73.21.-b, 79.60.-i, 73.50.Gr

Ultrathin metal films are simple realizations of quantum wells.¹ Valence electrons are free to move in the (x,y) plane of the film, while bouncing back and forth between the surface and the interface in the z direction. The technical challenge is achieving plane-parallel surface and/or interfaces, namely, thin films with atomically sharp interfaces and minimum surface roughness. When these conditions are met the characteristic discretization of the band structure is generally observed, either for quantum well (QW) states that are completely confined inside the film or for QW resonances coupled to bulk states in the substrate.²

Quantum confinement in a metallic film leads to a strong modulation of the density of states close to the Fermi energy that depends on the film thickness. Occasionally this gives rise to thickness-dependent physical properties, such as the surface roughness or the mode of growth.^{3,4} Although the thickness dependence allows one to tune thin-film properties, the appearance of new exotic phenomena are rather related to the scattering properties of the interface.⁵⁻⁹ As a matter of fact, only substrates with Fermi energy gaps of the appropriate symmetry lead to the thickness-dependent modulation in the density of E_F states required for quantum-growth phenomena.^{3,4} Using magnetic substrates with spin-dependent band structure one can modulate the electron spin in the film,⁷ i.e., strongly confined states build up for one spin, whereas weaker resonances remain for the other. Thereby a nonmagnetic material, such as Cu, can transmit magnetic information in trilayers and multilayers.⁵ Beyond magnetic polarization, the parallel effective mass,⁶ the spin-orbit splitting,¹⁰ and in general, the crystal symmetry of the substrate lattice is picked up by the thin film via interface scattering.^{8,9}

Interface engineering offers enormous possibilities for QW tuning.^{11,12} In general, changes at the interface define new phase boundary conditions, and hence an overall shift of QW levels.¹¹ Of particular interest for applications is the fact that QW spectra can be smoothly modulated via substrate doping,¹² in contrast to the limited thickness-dependent variation in discrete atomic-size steps. Conversely, important substrate properties can be determined from the QW spectrum analysis,⁶ for example, the actual size of the substrate band gap, which is otherwise underestimated in regular photoemission measurements due to photohole screening effects.¹³

Despite the importance of the interface in defining the electronic properties of a quantum well, fundamental questions still remain. The reflection of electron waves at projected spin and symmetry band gaps is well established.² The essential physics are captured in phase accumulation models,¹⁴ which account rather well for the data. However, many interesting systems exhibit QW resonances where projected band gaps are not present.⁵ The basic question that arises is how the interfacial reflectivity is reduced away from the gap, and whether one can determine an effective interface roughness based on general substrate electronic properties, such as the density of states of the appropriate symmetry to couple with QW states.

In order to address this issue, we have carried out high-resolution angle-resolved photoemission experiments for the model Mg/W(110) quantum wells, and performed a line-fit analysis of the spectra. This system allows us to examine a wide energy range that, extending away from a projected substrate band gap, reaches the Fermi level. In contrast to the slow energy-dependent variation found in Ag QW states,¹ the Mg/W(110) system exhibits a fast interface reflectivity change that makes interface scattering the dominant broadening mechanism near E_F . In fact, the reflectivity broadening inverses the tendency of the QW state linewidth to scale as $(E-E_F)^2$ (Ref. 1), i.e., sharp QW peaks become broader as we move away from the gap boundary and closer to the Fermi level. The interface reflectivity function can be retrieved from a scattering analysis of free-electron waves at the W(110) crystal, which can be sufficiently modeled for the energy of interest in a two-band model. This approach shows that the reflectivity for out-of-gap QW resonances mainly depends on the strength of the crystal potential that opens the gap.

The photoemission experiments were performed at 150 K and room temperature using a Scienta 200 high-resolution angle-resolved hemispherical analyzer. The sample was illuminated with monochromatized photons of $h\nu=40.8$ eV emitted by a Gammadata VUV 5000 high-intensity He discharge lamp. Energy and angular resolutions were set to 50 meV and 0.3° , respectively. Figures 1 and 2 show the normal-emission QW spectra for Mg(0001)/W(110) layers of varying thickness. These are prepared and characterized as described in previous works.^{9,15} Despite their different crys-

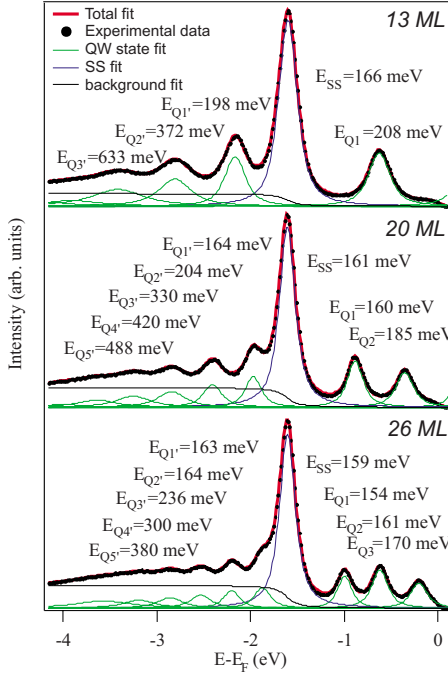


FIG. 1. (Color online) High-resolution photoemission spectra from Mg/W(110) quantum wells of relatively small thickness taken at $h\nu=40.8$ eV. Solid lines represent peak fit to the data done by Lorentzian functions. The resulting linewidth δE is indicated on top of each peak. The error of the linewidth from the peak fit amounts to 5 meV.

tal lattices, Mg films on W(110) relax to the bulk hexagonal structure immediately after the second layer. The strong d band emission of the W(110) substrate is visible up to 5 ML, and hence the spectra in Fig. 1 contain only Mg emissions. The intense peak at approximately -1.6 eV is the Mg surface state. The other emissions correspond to QW states and resonances due to the electron confinement in the film.

The positions of the QW peaks in Figs. 1 and 2 are well described by the Bohr-Sommerfeld quantization rule,¹⁵

$$kNt + \frac{\Phi}{2} = \pi n, \quad (1)$$

where the electron wave vector k and the total boundary phase shift Φ depend on the binding energy E , N is the number of atomic layers, t is the Mg interlayer distance, and n is the quantum number.² The different spectra in Figs. 1 and 2 have been fit following Eq. (1) and using Lorentzian lines for the surface state and each QW peak and an additional convolution with a Gaussian to account for the experimental resolution. The interest is focused on the resulting linewidth δE [full width at half maximum (FWHM)], also indicated in the figure, which shows, for a given thickness, a characteristic binding-energy dependence. At the left side of the surface state, QW peaks become narrower when their binding energy is reduced. At the right side of the surface state we observe the same trend for relatively large thickness (Fig. 2), but for smaller thickness we find the opposite behavior (Fig. 1), namely, peaks broaden as we get closer to E_F .

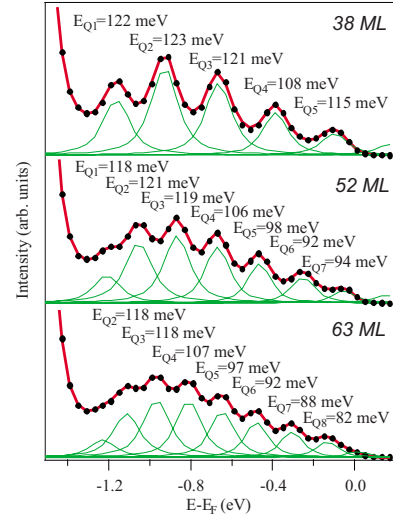


FIG. 2. (Color online) High-resolution photoemission spectra from Mg/W(110) quantum wells for higher thickness and close to E_F . The fit procedure is the same as for Fig. 1.

In two-dimensional states, such as surface and QW states, the single contribution to the photoemission peak width δE is the inverse quasiparticle lifetime Γ , i.e., the decay rate of the excited photohole. The latter is usually expressed as¹⁶

$$\Gamma = \Gamma_{e-e} + \Gamma_{e-ph} + \Gamma_{e-def}. \quad (2)$$

Assuming constant electron-phonon (Γ_{e-ph}) and electron-defect (Γ_{e-def}) contributions, the energy-dependent lifetime results from the inelastic electron-electron scattering as $\Gamma_{e-e} \propto (E - E_F)^{-2}$ (Ref. 16). This results on a steady decrease of δE by approaching the Fermi energy, as indeed found in Ag/Fe(100) QWs and in Fig. 2 for Mg films of large thickness. The same behavior is found in Fig. 1 for QW states with binding energies higher than the surface state, but not for QW states above the surface state. As we shall see, this is due to the fact that out of the substrate band gap, the bulklike lifetime contribution in the thinnest films is overcome by the interface reflectivity broadening.

We may address the out-of-gap interface reflectivity in the simplest way, i.e., by studying the scattering of free-electron waves at the W crystal, modeled in a one-dimensional nearly-free-electron approach. Let us assume a one-dimensional chain of W atoms with lattice constant a , reciprocal-lattice wave vector $G=2\pi/a$, and crystal potential $V(z)=V_G e^{iGz} + V_G^* e^{-iGz}$. The two level approximation results in an electron wave function in atomic units of the form ($\hbar=m=e^2=1$),

$$\Psi_W = \alpha e^{i(q+\pi/a)z} + \beta e^{i(q-\pi/a)z}, \quad (3)$$

where $q = \sqrt{2(E+E_G) - \sqrt{4EE_G + |V_G|^2}}$ is the two-band model momentum, $E_G = \frac{1}{2}(\frac{G}{2})^2$, and the coefficients α and β satisfy the Schrödinger equation. At the Mg side ($z > 0$) we consider the scattering of one-dimensional free-electron waves,

$$\Psi_{Mg} = A e^{ikz} + B e^{-ikz} = A(e^{ikz} + R e^{-ikz}). \quad (4)$$

Here $k = \sqrt{2(E - V_0)}$ is the free propagating Mg wave vector, $V_0 = 10.55$ eV is the crystal potential, and $R = B/A$ is the total

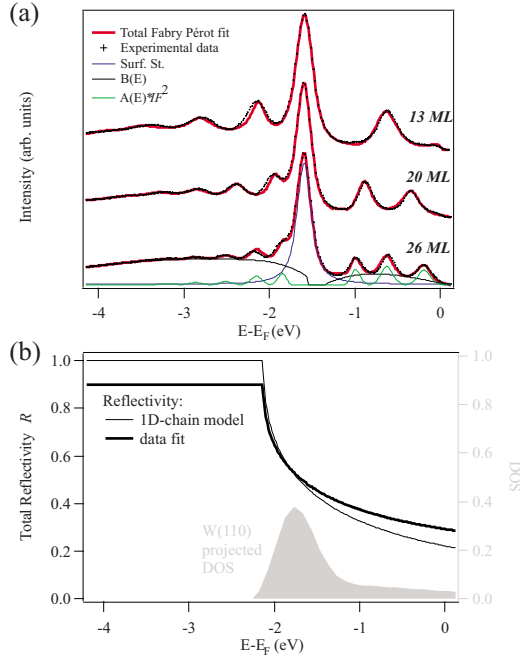


FIG. 3. (Color online) (a) Peak fit based on the Fabry-Pérot resonator model described in Eq. (6). The surface-state contribution is fitted separately with a Lorentzian function. An example for the fit and background functions is shown on the bottom of the 26 ML spectrum. (b) Reflectivity functions as determined from the one-dimensional chain model and the fit to the reflectivity to the experimental data. Also shown is the projected density of states of bulk W onto the (110) plane, which leaves a gap below -2 eV. Only W states allowed to interact with Mg QW states are included (see the text).

reflectivity. Matching conditions at $z=0$ enable us to obtain the analytical form of R as

$$R = \frac{\left(k - q - \frac{\pi}{a}\right) V_G + \left[E - \frac{1}{2} \left(q + \frac{\pi}{a}\right)^2\right] \left(k - q + \frac{\pi}{a}\right)}{\left(k + q + \frac{\pi}{a}\right) V_G + \left[E - \frac{1}{2} \left(q + \frac{\pi}{a}\right)^2\right] \left(k + q - \frac{\pi}{a}\right)}. \quad (5)$$

In Fig. 3(b) we represent the theoretical reflectivity function $R(E)$ (thin line), calculated using Eq. (5) with $2V_G=3.8$ eV, i.e., the width of the projected W gap considering only bands with symmetries (s , p_z , $d_{3z^2-y^2}$, and $d_{x^2-y^2}$) that can interact with the Mg s - p bands. $R(E)$ is thus defined as a constant plateau [$R(E)=1$] inside the W band gap, followed by a rapid decay toward the Fermi energy. As we see in Fig. 3, the functional form of $R(E)$, in particular the rapid drop in reflectivity away of the gap, explains the energy-dependent linewidth of QW peaks in Fig. 1.

The separate contribution of lifetime and interface reflectivity to the energy-dependent δE can be determined from a fit to the classical Fabry-Pérot (FP) interferometer model, where QW states are treated as Fabry-Pérot modes.¹ This is done for the data of Fig. 1 and is presented in Fig. 3(a). The

solid lines fit the QW, without the surface state, using the following function:

$$I = \frac{1}{1 + \frac{4f^2}{\pi^2} \sin^2\left(kNt + \frac{\Phi}{2}\right)} A(E) + B(E), \quad (6)$$

where $A(E)$ and $B(E)$ are smooth functions of E that account for the photon-energy-dependent photoemission cross section and the inelastic background, respectively. Both A and B are polynomial functions of the order of the number of QW peaks in the spectra. Note that $I(E)$ is a single oscillating sinus-square-like function, not a sum of Lorentzians or Gaussians, such as those used in Figs. 1 and 2. The prefactor of $A(E)$ produces oscillating values between 0 and 1 that have to be offset by $B(E)$ to account for the nonzero intensity between the QW peaks and enveloped by $A(E)$ to account for their intensity. Therefore, $B(E)$ is not the usual photoemission background, but a smooth polynomial that passes just under the QW peaks. On the other hand the attenuation (broadening) of Fabry-Pérot modes is accounted for within the interferometer finesse f , which is defined as

$$f = \frac{\pi \sqrt{R_e} e^{-Nt/2\lambda}}{1 - R_e e^{-Nt/\lambda}}, \quad (7)$$

where λ denotes the quasiparticle mean-free path that relates to the quasiparticle inverse lifetime Γ and the group velocity v by $\Gamma = v/\lambda$ and R describes the total reflectivity at the interface and the vacuum barrier.¹⁷ Both Γ and R enter the fitting procedure in Fig. 3 as energy-dependent parameterized functions that capture the essential physics. For $\Gamma(E)$ we use a parabolic function that matches the extrapolated surface-state linewidth of the infinitely thick Mg film, i.e., $\Gamma(-1.6 \text{ eV})=145$ meV that agrees very well with the Mg single-crystal linewidth of the surface state at the same measuring temperature of 150 K.¹⁸ For the experimental reflectivity $R_e(E)$, we assume the theoretical reflectivity function $R(E)$ derived in Eq. (5) but allow the simple linear scaling $R_e = \eta \cdot R + \chi$. The simultaneous fit to the spectra in Fig. 1 gives $\Gamma(E)=0.07 \text{ eV} + 0.028 \text{ eV} \cdot (E-E_F)^2$, with $E-E_F$ in eV, and $R_e=0.78R+0.12$. Inside the gap, the reflectivity is lower than $R=1$, as expected for incoherent leaking of the QW. Out of the gap, R_e is slightly higher than the expected $R(E)$. This shows the validity of the theoretical approach, although it also reflects its limitations. Indeed, the nearly-free-electron model cannot account for hybridization effects or W/Mg lattice mismatch. These would require a more sophisticated theoretical approach, probably including W and Mg electronic band structures as input parameters.

Assuming $R_e(E)$ and $\Gamma(E)$ and the FP model of Eq. (6), we may retrieve the thickness and energy-dependent linewidth function for Mg QWs near E_F . This function is shown in Fig. 4(a). It can be compared with the measured linewidth of Mg QWs in Figs. 1 and 2 that is summarized in Fig. 4(b). In both theory and experiment we observe that the increasing trend of the QW linewidth (Fig. 1) extends to QWs with thickness up to ~ 50 ML, and smoothly evolves into the bulklike decreasing function for thicker films (Fig. 2). The interface reflectivity indeed remains a relevant broadening

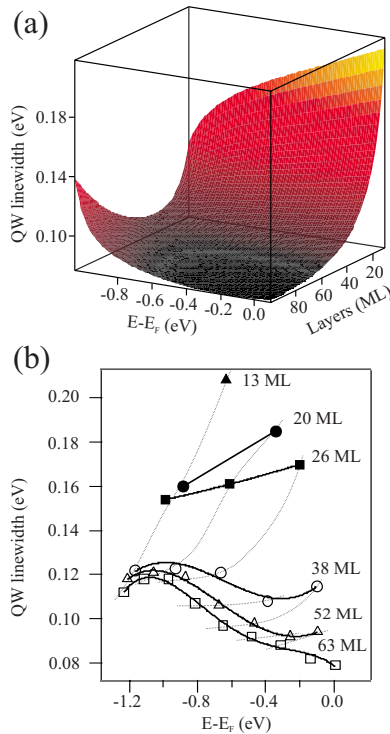


FIG. 4. (Color online) (a) Theoretical and (b) experimental QW linewidth as a function of energy and layer thickness in the 30 ML–100 ML range, respectively. Thick solid lines are polynomial fits that connect the experimental linewidth (FWHM) results for QW states at the indicated thickness. The reflectivity-related increasing trend toward E_F transforms into the bulklike decreasing function only for very thick films.

mechanism also for relatively thick films, far beyond the bulk mean-free path.

The validity of the functional form of $R(E)$ indicates that coherent scattering, i.e., wave-function matching, is the key feature determining the reflectivity of electron waves in me-

tallic QWs. The important parameter in Eq. (5) is, in fact, the strength of the electron potential V_G , namely, the size of the bulk band gap $2V_G$ relative to the total bandwidth. Thus, even for quantum well resonances that lie out of a projected band gap, the latter is determinant: for a large gap we will have a smooth decrease in reflectivity, but for a small gap the reflectivity will drop much faster. Note the difference with the interface reflectivity drop caused by inelastic interface scattering or leakage, which has been proposed for the Ag/Fe(100) QW.¹ Leakage may be viewed as a tunneling process, such that one can expect maximum transmission and minimum interface reflection for maxima in the density of substrate states (DOS) projected along the perpendicular direction. In fact, in the Ag/Fe(100) QW the small reflectivity change found (from $R=0.8$ at $E=E_F$ to $R=0.6$ at $E=E_F - 2$ eV) nicely correlates with the increasing (from E_F) DOS of Fe(100) perpendicular to the surface. In the Mg case the contrary behavior is observed, proving that QW leaking cannot explain the quick reflectivity drop away from the gap edge. In fact, Fig. 3(b) shows the projected DOS of bulk W directed along the $[110]$ direction, i.e., the sum of s , p_z , and d_z states ($d_{3x^2-z^2}$ and $d_{x^2-y^2}$). The DOS is maximum close to the edge of the projected band gap, decreasing smoothly toward E_F . Thus, the maximum overlap between thin film and bulk states takes place at the gap edge, where we actually find the largest reflectivity.

In summary, following a line-shape analysis of photoemission data we have proved the rapid interface reflectivity drop in out-of-gap quantum well resonances in the Mg/W(110) QW system. Such a reflectivity drop is well explained by our theoretical analysis, which indicates that quantum well resonances can build up with significant intensity depending only on the proximity and the size of the totally reflecting bulk band gap.

The work was supported through projects of the University of the Basque Country (Grant No. GIU06/27) and the Spanish Ministerio de Educacion y Ciencia (Grant No. MAT2007-63083).

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