

Quantum Oscillations in Coupled Two-Dimensional Electron Systems

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Quantum oscillations of the electron-phonon coupling of a Shockley surface state induced by the coupling to a metallic quantum well are observed experimentally as the thickness of the quantum well is increased. Microscopic calculations allow us to assign these oscillations to changes in the Eliashberg function, caused by a modulation in the electronic density overlap between the 2D surface state band and the 2D electron system of the quantum well and by the gradient of the one-electron potential in combination with the localized character of the Shockley-type surface state wave function. The findings are important in the context of the control of surface processes such as film growth and surface chemical reactions.

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A variety of fundamentally important condensed matter phenomena have been discovered and investigated in electronic systems with reduced dimensionalities. Two prominent examples of such kinds of electronic systems, which are both addressed in this letter, are Shockley-type surface (SS) states of the (111) oriented noble metal surfaces [1] and quantum-well (QW) states in ultrathin metallic films [2,3]. The former are states which are restricted to the outermost atomic surface layers and therefore represent an almost ideal example for two-dimensional electron systems. Their localization at the surface furthermore makes them strong sensors to any surface perturbation [4]. They are therefore particularly interesting for processes in nanotechnology and catalysis.

QW states evolve by the confinement of electrons in ultrathin metal films. When the thickness of a film is reduced to values comparable to or smaller than the electron coherence length, the electronic band structure evolves into a quantized electron spectrum in the direction perpendicular to the surface. The quantization arises from the standing electron wave pattern supported by the film and depends critically on the film thickness. This has been shown to influence a variety of other physical properties, which also show quantum size effects. Examples include superconducting properties [5,6], magnetic coupling in GMR-type ferromagnetic and non-magnetic multilayer stacks [7–10], the electron-phonon (e -ph) coupling parameter [11–13], and variations in the chemical activity of the QW surface [14–16]. In quite a number of these systems these properties show a clear oscillatory dependence on the film thickness (quantum oscillations), which can be directly related to periodic modulations in the electronic QW state spectrum.

In this Letter we show that such a quantum-oscillatory behavior can efficiently be transferred between two coupled two-dimensional electron systems. Experimentally, and from microscopic calculations, we have studied the e -ph coupling parameter λ_{SS} of the Ag(111) SS band as modified by the thickness of a supporting silver QW. By means of photoelectron spectroscopy we have identified clear oscillations in λ_{SS} as a function of QW thickness. Our microscopic model calculations quantitatively reproduce these experimental findings. It enables us to assign the origin of the SS state quantum oscillations to charge density variations at the surface caused by the underlying QW and to peculiarities of the Eliashberg function of the QW. Coupled metallic QWs have been studied already in the past, but an oscillation transfer as reported here was not observed [17–19]. The impact of the QW electronic structure on specific surface properties may represent an efficient means to control processes of relevance in fields such as nanofabrication and surface catalysis.

The strength of the e -ph coupling in a one-electron state ($\Psi_{\mathbf{k}_i}, \epsilon_{\mathbf{k}_i}$) described by $\lambda_{\mathbf{k}_i}$ is defined as the first reciprocal moment of the Eliashberg spectral function $\alpha^2 F_{\mathbf{k}_i}(\omega)$ [20]

$$\lambda_{\mathbf{k}_i} = 2 \int_0^{\omega_{\max}} \frac{\alpha^2 F_{\mathbf{k}_i}(\omega)}{\omega} d\omega. \quad (1)$$

The spectral function in quasielastic scattering approximation $\delta(\epsilon_{\mathbf{k}_i} - \epsilon_{\mathbf{k}_f} \pm \omega_{\mathbf{q},\nu}) \approx \delta(\epsilon_{\mathbf{k}_i} - \epsilon_{\mathbf{k}_f})$ is given by

$$\alpha^2 F_{\mathbf{k}_i}(\omega) = \frac{1}{(2\pi)^2} \int d^2\mathbf{q} \sum_{\nu,j} \delta(\omega - \omega_{\mathbf{q},\nu}) \times |g(\mathbf{k}_i, \mathbf{k}_f, \mathbf{q}, \nu)|^2 \delta(\epsilon_{\mathbf{k}_i} - \epsilon_{\mathbf{k}_f}), \quad (2)$$

where $g(\mathbf{k}_i, \mathbf{k}_f, \mathbf{q}, \nu)$ —the e -ph matrix element which re-

flects the probability of electron scattering from the initial state $\Psi_{\mathbf{k}_i}$ with energy $\epsilon_{\mathbf{k}_i}$ to the final state $\Psi_{\mathbf{k}_f}$ with energy $\epsilon_{\mathbf{k}_f}$ by the phonon $\omega_{\mathbf{q},\nu}$. The sum in Eq. (2) is carried out over all final electron states $\Psi_{\mathbf{k}_f}$ and all possible phonon modes (\mathbf{q}, ν) . The e -ph matrix element is

$$g(\mathbf{k}_i, \mathbf{k}_f, \mathbf{q}, \nu) = \left(\frac{1}{2M\omega_{\mathbf{q},\nu}} \right)^{1/2} \times \langle \Psi_{\mathbf{k}_i} | \hat{\epsilon}_{\mathbf{q},\nu} \cdot \nabla_{\mathbf{R}} V_{\text{sc}} | \Psi_{\mathbf{k}_f} \rangle, \quad (3)$$

where M is the atomic mass, $\hat{\epsilon}_{\mathbf{q},\nu}$ are the phonon polarization vectors, and $\nabla_{\mathbf{R}} V_{\text{sc}}$ is the gradient of the screened one-electron potential with respect to atom displacements from their equilibrium positions \mathbf{R} . In the calculations, we simulate the Ag(111) surface by using a slab model with 15–55 atomic layers of Ag(111). With these slab thicknesses, *ab initio* computations of e -ph coupling are currently not feasible. Therefore, in the present calculations we use a model proposed for studying e -ph interaction in surface and QW states [21,22]. This model combines three independent approximations to evaluate the e -ph coupling matrix elements [Eq. (3)]: (i) one-electron wave functions and energies are calculated with one-dimensional potential [23–25]; (ii) phonon frequencies and polarizations are obtained from a force constant method or from an embedded atom method; and (iii) a gradient of one-electron potential is represented by the Ashcroft pseudopotential [26] screened within Thomas-Fermi approximation. Details of the model used are given in Ref. [27]. The embedded atom interatomic potentials produce the equi-

librium Ag(111) surface crystal structure in close agreement with experimental data. These potentials also describe phonon spectra of the Ag(111) in good agreement with available experimental data and *ab initio* computational results [28].

Experimentally, the e -ph coupling parameter λ_{SS} has been obtained from temperature dependent measurements of the photoemission linewidth Γ of the SS state. Details of the used experimental setup are summarized in Refs. [11,29].

Figure 1(a) shows a 2D photoemission map ($h\nu = 21.22$ eV) recorded at 200 K for a 40 ML thick QW that was prepared by evaporation of Ag on a Cu(111) substrate and subsequent annealing to 600 K [29]. Spectral signatures of the two neighboring 2D electron systems—silver QW and Ag(111) SS state—are clearly resolved. For normal emission ($\bar{\Gamma}$ -point) the dispersive SS state appears at a binding energy of about 40 meV as the most intense feature in the spectrum. At higher binding energies between 300 and 850 meV (above the Cu gap edge) a series of three dispersing QW states ($\nu = 1, 2$ and 3) is visible. Figure 1(b) shows energy distribution curves (EDC) of the SS state at the $\bar{\Gamma}$ -point for different temperatures (QW thickness: 36 ML). The full width at half maximum linewidth (note log scale) of the SS state increases with increasing temperature. The broadening is induced by the increased phonon occupation at higher temperatures and the consequent increase in electron-phonon scattering processes, which reduce the SS state lifetime. In the high temperature asymptotic behavior the e -ph coupling parameter λ_{SS} can be evaluated from the linear slope of this temperature dependence of the SS state linewidth $\Gamma_{e\text{-ph}}$ according to [11,21,30–34]

$$\Gamma_{e\text{-ph}}(T) = 2\pi\lambda_{\text{SS}}k_B T. \quad (6)$$

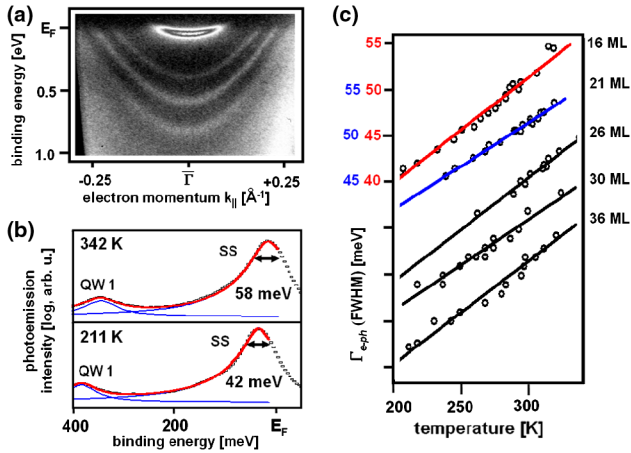


FIG. 1 (color online). (a) 2D $E(k_{\parallel})$ photoemission map for 40 ML Ag/Cu(111), $T = 200$ K, $h\nu = 21.22$ eV. (b) Temperature-dependent energy distribution curves at the $\bar{\Gamma}$ -point of 36 ML Ag for 211 and 342 K recorded with $h\nu = 6$ eV. The full width at half maximum linewidth (note log scale) of the SS state increases with increasing temperature. (c) Linewidths (dots) and linear fits (solid lines) of the SS state as a function of temperature for five different films in the thickness regime between 16 and 36 ML. The $\Gamma_{e\text{-ph}}$ scale is plotted for 16 ML Ag film thickness in red, for 21 ML in blue, and changes subsequent by 5 meV from 26 to 36 ML.

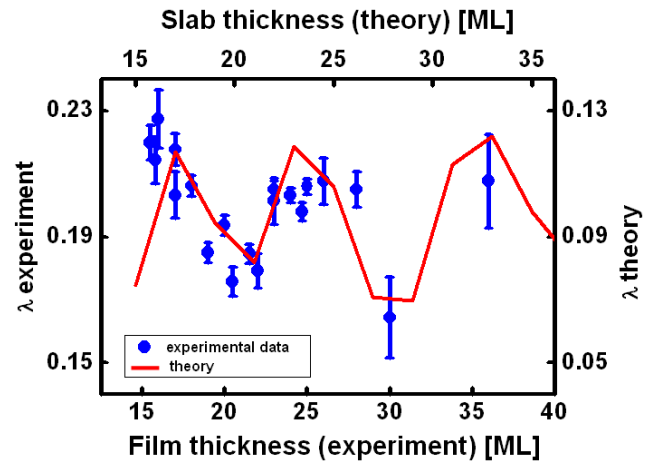


FIG. 2 (color online). Electron-phonon coupling parameters for the SS state as a function of Ag-film thickness. Blue dots show the experimental data, the red line the theoretical data. The meaning of the different scales in the experimental and the theoretical data set is addressed in the text.

Figure 1(c) compares the temperature dependence of the SS linewidth for five different films in the thickness regime between 16 ML to 36 ML thick QW as a function of temperature T . It is evident that the slope of $\Gamma_{e-ph}(T)$ and, in consequence, the e -ph coupling parameter λ_{SS} is modified by the thickness of the supporting QW. This observation provides first evidence of an efficient coupling between the two 2D electronic systems.

Figure 2 shows experimental and theoretical λ_{SS} values of the SS state as a function of the thickness of the silver QW between 15 and 40 ML. The variations in λ_{SS} of both data sets follow a clear oscillatory behavior. It exhibits an amplitude of ≈ 0.02 and an oscillation period of about 10–12 ML. The slightly varying thickness scale between experiment and theory is due to uncertainties in the determination of the film thickness in the experiment [29]. The overall lower values of the theoretical data (≈ 0.1) were expected, since the calculations do not include the constant background of phonon-mediated scattering processes from the Cu substrate as well as from defects scattering contribution to the total linewidth [35]. As mentioned above, oscillations in distinct properties of a QW are a rather common behavior arising from the quantization of the

electron spectrum in the QW. Our results provide for the first time evidence that such a quantum-oscillatory behavior can efficiently be transferred to another low-dimensional electronic system that is coupled to the QW. Below we identify the relevant peculiarities which are responsible for this transfer process.

The principle mechanism which couples the SS to the electron spectrum of the QWs is illustrated by the black arrows in Fig. 3(a). The SS state hole, which is created within the photoemission process, couples to the QW states by the refilling due to phonon-mediated electron scattering processes. It is this kind of scattering process which is described by the e -ph matrix element given by Eq. (3). The initial state Ψ_{k_i} corresponds here to the SS state wave function Ψ_{SS} , the final state Ψ_{k_f} is a QW state Ψ_{QW} . Figure 3(b) separately shows the calculated individual contributions of the first and the second QW state to the total value of λ_{SS} as a function of film thickness. Both partial contributions show similar oscillatory behavior at a reduced amplitude. It is the sum of these partial contributions of all QW states resulting in the total λ_{SS} that has been probed experimentally and is shown in Fig. 2. The QW state with higher quantum number exhibits a larger partial contribution to total λ_{SS} for a given QW thickness, due to the larger electronic density overlap with the SS [Fig. 3(c)]. Furthermore, as the film thickness increases one observes an overall decrease of the individual contributions, due to decreasing electronic density overlap of the SS and individual QW states [Fig. 3(d)].

The overall decrease of the partial λ contributions is compensated by the charge density of additional QW states which evolve in the film with increasing film thickness. Figure 4 displays results from a calculation of a partial λ of the SS state ($\lambda_{\text{gap-QWS}}$) which considers exclusively QW

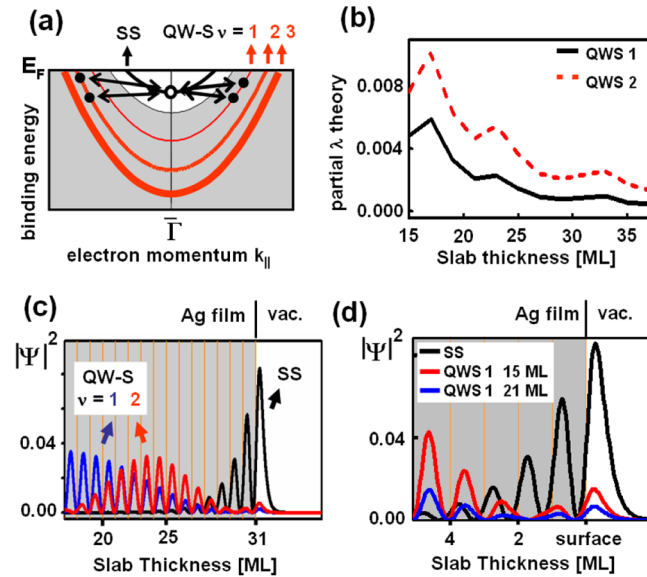


FIG. 3 (color online). (a) Schematic illustration of the electronic band structure. The white area represents the Ag(111) band-gap supporting the SS state, the gray area represents the projected electronic bands of full bulk Ag(111) to the surface, which is quantized due to the confinement of electrons in the ultrathin Ag films (red parabola). The arrows indicate the phonon-mediated relaxation paths of the photoemitted SS hole (energies are highly exaggerated). (b) Partial λ to the electron-phonon coupling parameter of the SS state from the $\nu = 1$ and $\nu = 2$ QW states as a function of film thickness. (c) Calculated quadratic wave function $|\Psi|^2$ of the $\nu = 1$ and $\nu = 2$ QW states and the SS state for a 31 ML thick Ag film. (d) Calculated quadratic wave function $|\Psi|^2$ of the $\nu = 1$ QW state for 15 and 21 ML slabs.

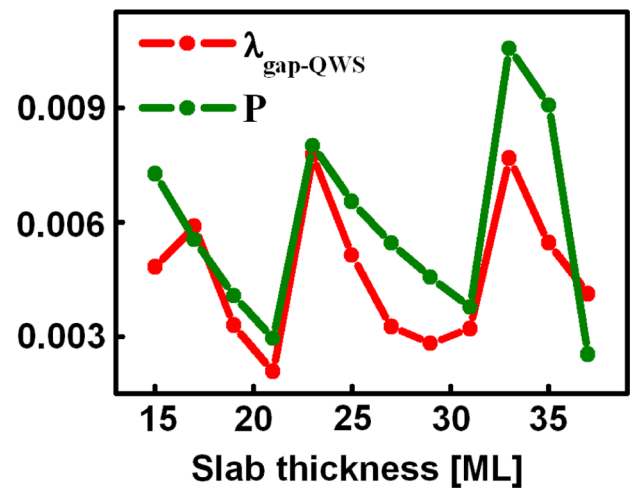


FIG. 4 (color online). Partial λ to the electron-phonon coupling parameter of the SS state from the QW states localized in the Cu(111) $\bar{\Gamma}$ band gap as a function of film thickness and electronic density overlap of corresponding wave functions $P = \int \sum_{\text{QWS}} |\Psi_{SS} \Psi_{\text{QWS}}|^2 dz$.

states in the energy regime of the Cu(111) substrate band gap (below 850 meV binding energy). At distinct film thicknesses an additional QW state moves into resonance with the Cu band gap and, hence, increases the number of QW states, which affect $\lambda_{\text{gap-QWS}}$, by one. The appearance of this additional QW state within the band gap is equivalent to a sudden localization of the QW wave function in the Ag film [11]. Such localization takes place for instance at a slab thickness of 22 ML and obviously contributes considerably to the amplitude of the second oscillation maximum of $\lambda_{\text{gap-QWS}}$ (see black line Fig. 3(b) and red line in Fig. 4). The magnitude of $\lambda_{\text{gap-QWS}}$ is, however, by far too small to explain the total amplitude of λ_{SS} and accounts for only 10% of the observed experimental value. The overlap of the SS wave function with QW states outside of the Cu(111) band gap accounts for the remaining 90% of the amplitude. The latter, the sum over oscillations of all partial contributions, is obviously a crucial factor responsible for the pronounced oscillations in λ_{SS} . However, these oscillations cannot be explained by a simple overlap effect.

For further evaluation, we also analyzed the effect of the two wave function constituents, Ψ_{SS} and Ψ_{QW} , of the e -ph matrix element given by Eq. (3). We checked the scenarios where either Ψ_{SS} or Ψ_{QW} were set constant as a function of the film thickness. The former case corresponds to the delocalization of the SS state and mimics therefore the complete loss of its surface localized character. In both cases the total value of λ_{SS} as well as the partial contributions from the individual QW states increase with increasing film thickness in contrast to the results shown in Fig. 3(b). For $\Psi_{\text{QW}} = \text{const}$ oscillations are still visible; however, for $\Psi_{\text{SS}} = \text{const}$ any oscillation has vanished. The localization of the SS state wave function at the surface of the QW is obviously an essential ingredient for the observed oscillatory behavior. Another significant ingredient is the gradient of the one-electron potential [Eq. (3)] which enhances the oscillation amplitude of λ_{SS} . If we replace the gradient by the potential itself the oscillation amplitude is substantially reduced. Moreover, for some thicknesses the maxima and minima positions of λ_{SS} become opposite to those we obtain with the nonzero gradient of the potential.

We also calculated the partial contribution of the surface Rayleigh wave phonon mode. Recently, it has been shown that, for ultrathin Pb(111) films, oscillations in the e -ph coupling occur due to a corresponding shift in the surface phonon frequencies originating from the oscillatory behavior of the first interlayer spacing with the slab thickness [36]. That is not true in our case. Nevertheless, calculations exclusively considering effects associated with the Rayleigh wave phonon mode yield oscillations with amplitudes around ≈ 0.007 , which is 1/3 of the total amplitude (≈ 0.02) for including the full phonon spectrum.

In conclusion, we have reported on quantum oscillations of a SS state as transferred from an underlying metallic

QW by means of (I) the overlap of SS and QW state wave functions and by (II) the gradient of the one-electron potential in combination with the localized character of the SS wave function. An overall modulation of the SS state e -ph coupling parameter λ_{SS} by about 20% was observed. Because of its surface localization, the surface Rayleigh wave phonon mode is of high relevance to the SS modulations and contributes about 1/3 of the total oscillation amplitude. Our findings suggest that further control of surface properties due to coupling to the electronic QW should be possible, such as the chemical activity of the metal surface.

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