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Oscillatory electron phonon coupling in Pb/Si(111) deduced by temperature-dependent quantum well states*

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Photoemission study of atomically flat Pb films with a thickness from 15 to 24 monolayers (ML) have been performed within a temperature range 75–270K. Well-defined quantum well states (QWSs) are observed, which exhibit interesting temperature-dependent behaviours. The peak position of the QWSs shifts towards higher binding energy with increasing substrate temperature, whereas the peak width broadens linearly due to enhanced electron–phonon coupling strength (λ). An oscillatory λ with a period of 2ML is deduced. Preliminary analysis shows that the oscillation can be explained in terms of the interface induced phase variations, and is thus a manifestation of the quantum size effects.

Keywords: Pb thin films, photoemission, quantum well states, electron–phonon interactions
PACC: 7360D, 7960, 7320D, 6320K

1. Introduction

Many properties of solids, such as resistivity, structural instabilities, specific heat, and particularly superconductivity, depend directly on the electron–phonon coupling strength (λ).[1] Recently, with the advance in thin film growth and electronic structure characterization tools, many unique physical properties of thin films with thickness comparable to the Fermi electron wavelength have been observed and expounded in terms of the quantum size effects (QSEs). Oscillatory superconductivity in atomically flat Pb films has been observed in our previous study, and has been proved to be modulated by the QSEs,[2] where the oscillation of the electron density of states near the Fermi level proves to be the key factor. As we know, another important factor is the electron–phonon coupling, influencing the superconductivity in thin films.

Angle-resolved photoemission spectroscopy (ARPES) has been used successfully to investigate the electronic structure of thin films formed on metal or semiconductor or other surfaces.[3] However, as many other spectroscopy, it does not simply probe the ground state, rather, the resulting decayed excitations (the photohole), and thus many-body effects such as electron–phonon coupling could be involved. In the last few years, prominent progress has been achieved in the study of λ for the surface states in bulk materials[4−6] or for the quantum well states (QWSs) in thin films by ARPES.[7−10] For crystalline metal surfaces, the observed photoemission peak width is proportional to $\hbar/\tau$, where τ is the lifetime of the hole state excitations. Three factors, the Auger decay (via electron–electron interactions), the phonon scattering (via electron–phonon interactions) and impurity scattering contribute to the valence hole decay at zero temperature. Recent investigations show that at

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high temperatures (greater than one-third of the Debye temperature) and small hole energies (lower than the bandwidth), the temperature-dependence of the peak width is actually a good measure of the phonon contribution to the hole lifetime. Since the influence of the electron–electron interactions and the defect scattering are negligible in this particular situation,[1] the inverse hole lifetime can be given by $\hbar/\tau = 2\pi\lambda k_{\rm B}T$, where λ equals $2\pi k$ times the slope of the peak width vs temperature.

In this paper, variable-temperature photoemission spectra from 75 to 270K of Pb films have been collected, and are applied to measure λ at different film thicknesses. What we found is that, while there is a general trend that the λ develops towards the bulk value with increasing film thickness, there is also a quantum oscillation with a period of 2ML, the same as the oscillatory superconducting transition temperature ($T_{\rm c}$) by *ex situ* measurement. The phase variations of π across the confinement gap at the interface prove to be the origin of this oscillation caused by QSEs.

2. Experiments

Pb films were prepared by a special low-temperature growth method, followed by annealing up to room temperature. In the molecular beam epitaxy (MBE) chamber, Pb was evaporated using a Pyrolytic Boron Nitride (PBN) crucible with a flux rate of 0.2ML/min, while the Si substrate was maintained at about 145K by flowing liquid N_2. RHEED patterns were used for real time monitoring of the film growth. After growth, the sample was allowed to warm to room temperature naturally in one hour, and transferred to an analysis chamber with a base pressure of 3×10^{-9}Pa, where an OMICRON scanning tunnelling microscopy (STM) and a GAMMADATA SCIENTA SES-2002 electron energy analyser were installed for *in situ* surface morphology and electronic structure analysis. A sample manipulator in the ARPES analysis chamber, with a button heater attached to the head of an open-cycle He cryostat, was used to control the sample temperature from 20 to 300K with a precision of $\pm$0.1K. The energy resolution (2meV) of the analyser, which is much smaller than the measured QWSs widths, enables us to capture the fine variation of the photoemission spectra with the substrate temperature.

3. Results and discussion

By our special growth method, we have successfully fabricated uniform Pb films on the Si(111) surface with atomic level precision, and established its particular growth behaviour over a wide thickness range. A critical thickness of 10ML for the formation of flat Pb film stable at room temperature is established under the low-temperature growth approach. A QSEs-induced bi-layer growth mode dominates up to 21ML with an alternation of the relative stability variation from even layers to odd layers at 13ML, and then a quasi layer-by-layer mode follows. The uniformity of the Pb films prepared for the present photoemission study can be seen both from the STM morphologies and the corresponding photoemission spectra. The insets in Fig.1 illustrate the typical morphologies of the films of 22 and 23ML thick. Although stepped surfaces can be observed from the STM image, the number of steps on the Pb surface are equal to the number of steps on the Si(111) substrate before Pb growth, therefore, the films are essentially perfect as there are exactly same number of atomic layers on each terrace. The photoemission spectra shown in Figs.1(a) and 1(b) further confirm this, where only one set of QWSs appears in the spectrum of a particular thickness.

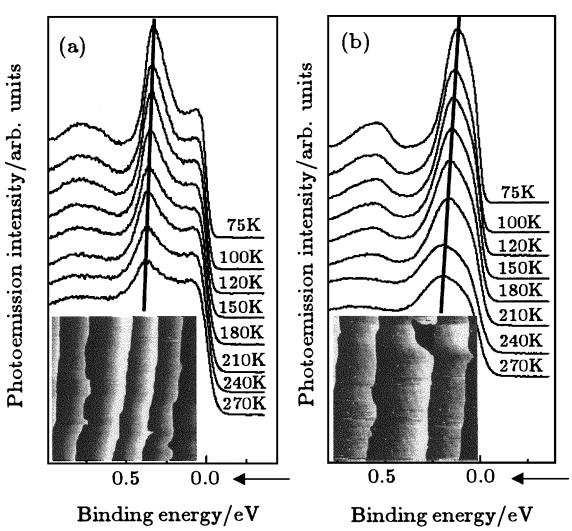


Fig.1. Normal emission spectra of the Pb layers taken from 75 to 270K. The insets display the RT STM images of the atomically flat Pb films with a thickness of 22ML (a) and 23ML (b), with an image size of 2000nm×2000nm.

We can see that with increasing substrate temperature, the peak position of the QWSs shifts towards higher binding energies (Figs.1(a) and 1(b)). A linear temperature dependence of the binding energy of the QWSs is evident, as seen in Fig.2(a). To find out the exact relation between the linewidth (ΔE)

and the temperature, a curve fitting by Voigt profile with Lorentzian line shape was made. It is clear from the quality of the fitting that the peaks are good Lorentzian, indicating that the interpretation of the width as inverse lifetime is reasonable. Typical fit results are plotted in Fig.2(b) for the films of 22 and 23ML, where ΔE increases linearly with increasing temperature and exhibits prominent different slopes for the adjacent layers. According to the previous discussion, this slope is related to the λ value via

$$\lambda = \frac{1}{2\pi k_{\mathrm{B}}} \frac{\mathrm{d}\Delta E}{\mathrm{d}T}. \tag{1}$$

The λ values derived from the QWSs peaks for different film thicknesses are shown in Fig.3 (triangles). Because of the special growth mode of Pb films on Si(111), only the values for stable layers (15, 17, 19, 21, 22, 23, 24ML) are acquired in this study. Besides an overall gradual increase of the λ values towards the bulk value (1.55),[11] an oscillation of λ with a period of 2ML is clearly seen from 21 to 24ML. As a comparison, the measured T_{c} of the Pb films is also added in Fig.3 (dots). A clear one-to-one correspondence in the oscillatory λ and the experimental T_{c} for different layers is obvious, but the variation trend is inversed in amplitude vs thickness above 21ML. That is to say, a larger λ value corresponds to a lower T_{c} in the Pb films. Note that the experimental T_{c} was acquired with the Pb films covered by 4ML Au, while the photoemission data for measurement of λ is taken from the clean Pb film surfaces.

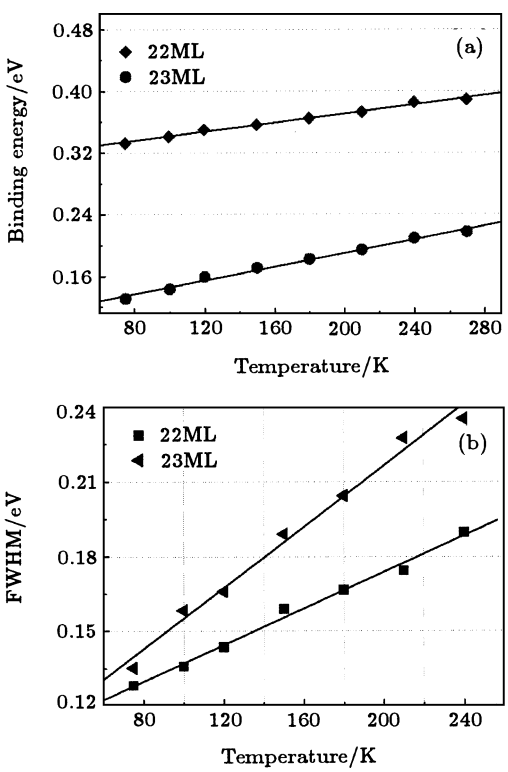


Fig.2. Variations of binding energy with temperature (a) and that of full width at half maximum (FWHM) (b) with temperature for different thicknesses.

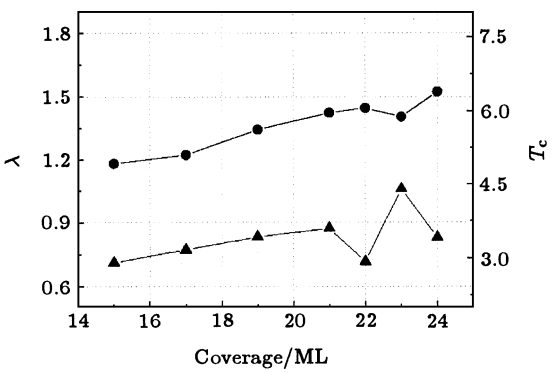


Fig.3. Experimental λ values for QWSs of the Pb films as a function of film thickness (triangles), with the T_{c} of the films by *ex-situ* measurement also shown (dots).

According to the phase accumulation model,[12,13] three factors associated with the binding energy variations of the QWSs can be expected: 1) thermal expansion of the epitaxial film results in an enhancement of the quantum well width, and thus an increase of the binding energy, 2) temperature dependence of the band structure and the lowering of Fermi level of the substrate originated from the decreased electron density of states, can lead to a reduction of the binding energy, 3) a phase change in the film and substrate boundary contributes minor effect to the binding energy variation. While, experimentally, the net shift of the binding energy depends on the relative importance of the first two conflicting factors, further quantitative results rest with the calculations based on the different thermal expansion coefficient of the substrate and epitaxial materials.[9] In our Pb/Si(111) system, the thermal expansion coefficients of Si and Pb are 2.8×10^{-6} and $2.89\times10^{-5}\mathrm{eV\cdot K^{-1}}$, respectively. Therefore, a dominant expansion of the epitaxial Pb film over that of the Si substrate would induce the shift of the binding energy towards a higher value, and this is actually observed in our experiment.

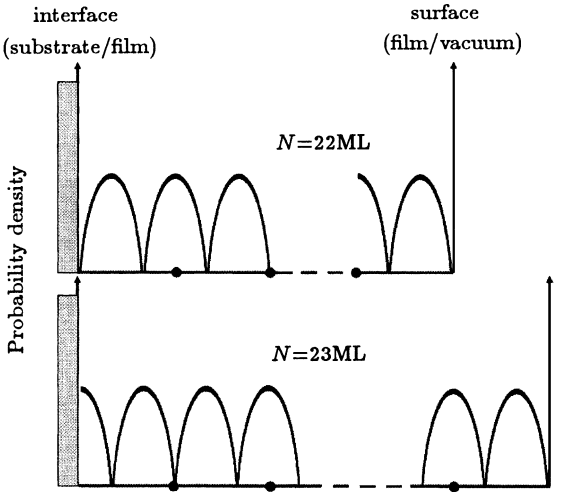


Fig.4. Probability densities of the QWSs for typical even (22ML) and odd (23ML) Pb layers plotted in a sketch map. The dots indicate the position of each atomic plane.

Previously, the oscillatory behaviour of λ as a function of the film thickness or the binding energy of the QWSs was reported for the Ag/V(100) and Ag/Fe (100) systems, and was attributed to the interface or the surface effect.[9,10] When the characteristic scale of a system is comparable to the Fermi electron wavelength, the surface and interface effect becomes much more important. To understand the relation of quantized energy levels with the quantum well width (film thickness) and the boundary conditions, a phase accumulation model[12,13] was proposed within a finite potential well

$$2k(E)Nd + \phi_{\rm s}(E) + \phi_{\rm i}(E) = 2\pi n, \qquad (2)$$

where $k(E)$ is the electron wavevector in Pb along the ΓL direction (perpendicular to the substrate surface), $\phi_{\rm s}(E)$ and $\phi_{\rm i}(E)$ are the phase shifts upon electron reflection at the surface and the interface respectively, n (an integer) is the index of the QWSs, and N is the number of atomic layers. The phase shift at the interface can be described as

$$\phi_{\rm i}(E) = {\rm Re}\left[-\cos^{-1}(2\frac{E+{\rm i}\gamma-E_{\rm l}}{E_{\rm u}-E_{\rm l}}-1)\right]+\phi_{\rm I}^{0}, \quad (3)$$

where $E_{\rm l}$ and $E_{\rm u}$ are the energies of the lower and upper edges of the confinement gap, γ is a broadening parameter. $\phi_{\rm I}^{0}$ is a constant. The arccosine function yields a phase shift of $-\pi$ at the lower edge and 0 at the upper edge of the substrate gap. The Fermi electron wavelength ($\lambda_{\rm F}$) in Pb is nearly four times the atomic plane spacing (a_0) along the [111] crystallographic direction, namely $\lambda_{\rm F} \approx 4a_0$, and the periodicity for the QWSs crossing the Fermi level is about 2ML.[14] The oscillatory binding energy of the QWSs in Pb films should lead to a similar oscillatory behaviour of the phase shift at the interface. A direct explanation can be seen in the schematic graph in Fig.4, where the probability densities of typical even and odd layered films of 22 and 23ML are displayed.

As suggested previously for the Ag/Fe(100) system, the matrix element for electron–phonon interaction, within the rigid-ion model, is proportional to $\langle\varphi'|\nabla V|\varphi\rangle$, where φ and φ' are respectively the initial and final states of the electrons in the crystal potential.[15] The scattering probability at the surface or the interface is proportional to $\langle\varphi'|\nabla V|\varphi\rangle^2$, where ∇V is the boundary potential step. If the interface is define at $Z = 0$, the origin of the quantum oscillation depends on $|\varphi(0)|^4$, the scattering rate of phonons. For a Pb film with a large binding energy, an interface phase shift is close to $-\pi$ (just as the film of 22ML thick), rather small amplitude introduces a small value of $|\varphi(0)|^4$, and thus a suppressed interface contribution follows with a small λ value. On the contrary, a film with a lower binding energy allows a small phase shift close to 0 at the interface and a large λ result, as observed for the film with a thickness of 23ML in Fig.4. Based on the above analysis, the variations of the phase shift at the interface can be expected to account for the oscillatory λ in Pb thin films.

Similar analysis can be employed to analyze the contribution from the film surface. Since the vacuum barrier is close to an infinitely high barrier, the energy levels of interest are far below the vacuum level. Therefore, the wave function at the surface plane should be a node, and the phase shift at the surface potential step should be always close to $-\pi$, thus represents small variations with film thickness alternation. A suppressed contribution allows for a small electron phonon-coupling from the surface. Another question is about the global reduction of the λ values for all the film thicknesses, as suggested by De Gennes,[16] no enhancement is expected if a film with reduced dimension and a large λ value is grown on a nonsuperconducting material. So in our experiment, an overall reduction in λ compared with a bulk value should be reasonable.

In summary, the same oscillatory behaviour in λ and $T_{\rm c}$ with a period of 2ML indicates clearly that they are both modulated by the QSEs. The antiphase may originate from the different boundary conditions between the clear Pb films for λ measurement and the Pb films with Au overcap for determination of $T_{\rm c}$.

4. Summary

Many properties can be modulated by the QSEs, and the superconductivity is one of the examples demonstrated recently. The electron–phonon coupling, one of the two important factors that influence superconductivity, is investigated in this study, which shows a similar oscillatory behaviour as that of the T_c. The similarity of the 2 ML oscillatory period in λ and T_c indicates clearly that they are both modulated by the QSEs. This work may provide helpful but indirect information for the understanding of the oscillatory superconductivity in Pb films with reduced dimension.

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