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Optical response of Pb quantum wells on Si(111)-(6 × 6)-Au

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Abstract

Ultrathin Pb films deposited on Si(111)-(6 × 6)-Au at 105 K in ultrahigh vacuum (UHV) conditions were studied by means of differential reflectance (DR) spectroscopy. The optical signal from spectral range 0.25–0.60 eV has been measured *in situ* during Pb film growth. Base on DR data the imaginary part of the Pb dielectric function, the optical conductivity and the plasma frequency were determined. The scattering time of the electrons involved in optical transitions was obtained according to the Keller and Liu local field theory [O. Keller, A. Liu, Phys. Rev. B 49 (1994) 2072].

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1. Introduction

Differential reflectance spectroscopy has been successfully applied for investigation of metallic ultrathin films and nanostructures formed on semiconductor substrates. Achievement in observing variety of intriguing features such as phase transition during Pb deposition onto Si(111)-(7 × 7) [1] and optical anisotropy during 1D Pb nucleation on vicinal Si(335) [2] demonstrate high potential and wide opportunities of using optical methods in mesoscopic regime.

Low-dimensional Pb/Si systems are a subject of great scientific interest. They morphology and structural properties strongly depend on certain growth conditions. This in turn revealed in appropriate changes of the optical properties. Self-organized atomic structures in form of highly uniform-height Pb islands with flat tops and steep edges have been observed in temperature range 120 K $< T <$ 250 K [3]. Depending on the substrate surface reconstruction the preferred island height was five steps in the case Si(111)-($\sqrt{3} \times \sqrt{3}$)-Pb [4] and seven steps on the Si(111)-(7 × 7) [5]. The driving force of the preferred island growth is believed to be related to quantum size effects (QSE) [6].

Quantum size effect appears when the one or more dimensions of a crystal become comparable to the Fermi wavelength of the electrons. The spatial confinement of the electron motion leads to a quantization of the component of the electronic wave vector and consequently to a discrete set of electron energy levels. Clear experimental evidence of the QSE in Pb/Si system was initially observed in electrical resistivity measurements [7]. Simultaneously measured reflection high energy diffraction (RHEED) intensity oscillations have been shown that at low temperatures Pb on Si(111)-(6×6)-Au substrate growth in *quasi*- monolayer-by-monolayer mode. QSE related periodical changes have been found in Hall effect studies [8]. In addition the existence of the quantized levels in Pb ultrathin films was proved by photoemission and electron tunneling experiments [9,10], which demonstrate that Pb/Si system at low temperature

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can be very attractive as a simple prototype of metallic quantum well. Previously the optical properties of Pb films on Si(111)-(6 × 6)-Au were studied in the spectral range from 0.5 eV to 2.0 eV [11], where the main contribution to optical properties are due to the interband transitions.

In this Letter we study infrared optical properties of ultrathin Pb films deposited on Si(111)-(6×6)-Au substrate at 105 K. The optical measurements were held in the spectral range where only intraband transitions (i.e., free electron conductivity) take place. The DR changes were measured during Pb growth using linear polarized light within energy range $h\nu = 0.25$–0.40 eV. We also presented data obtained for light energies 0.50–0.60 eV, where both intraband and interband transition mechanisms are present. The analysis of the optical reflection changes was held according to classical McIntyre–Aspnes three phase model and quantum Keller–Liu local field theory. Presented results are compared with the data obtained from electric contact four-probe and structural RHEED spot profile measurements. Finally, the influence of the electron density oscillations during ultrathin film growth on observed changes in optical properties of Pb is discussed.

2. Experimental

All experiments were performed in UHV chamber with the base pressure 5×10^{-11} Torr. The substrate quality, Pb film morphology and deposition process have been monitored with RHEED diffractometer. A substrate with dimensions of $18 \times 4 \times 0.6\ \text{mm}^3$ was cutted from a p-type boron Si(111) wafer with 20 Ω cm specific resistivity at room temperature. A gas-flow liquid nitrogen cryostat was used for sample cooling up to 105 K. The stable low substrate temperature during deposition was essential for forming smooth films. The surface cleaning procedure consisted in a flashing for a few seconds. For that purpose direct resistive heating with the dc current about 13.7 A through the sample was used. This procedure results in a sharp Si(111)-(7 × 7) superstructure RHEED pattern. In order to prepare the Si(111)-(6 × 6)-Au surface reconstruction, 1.2 ML of Au were deposited on Si(111)-(7 × 7) superstructure. Annealing for 1 min at about 950 K and slow cooling to initial temperature stage resulted in appearance of a clear Si(111)-(6 × 6)-Au RHEED pattern. The studied Pb films were grown in the (111) orientation. The average deposition rate controlled by a quartz crystal thickness monitor was equal 0.25 ML min^{-1}. RHEED specular beam intensity oscillations were used to calibrate the quartz thickness monitor.

Differential reflectance spectroscopy based on measurements of a relative change of the sample reflectivity $\Delta R/R = (R^{\text{Si+Pb}} - R^{\text{Si}})/R^{\text{Si}}$ upon thin film deposition, where R^{Si} and $R^{\text{Si+Pb}}$ are reflectance of a bare substrate and the substrate covered with Pb film, respectively. The optical system consisted of a globar, stabilized with water cooling system, prism monochromator, Brewster's angle polarizer and temperature stabilized PbSe detector. The linear p-polarized light entered the UHV chamber through a MgF_2 window and was focused on the sample with the system of the mirrors. The angle of incidence on the sample was equal to 49.2°. The reflected light after passing second window was focused on the detector. The light was chopped with frequency 320 Hz. A lock-in technique was used to recover measured signal. The whole optical setup was optimized in order to achieve a high stability signal and high accuracy of the measurements. The stability of the reflected signal during each measurement was better than 10^{-4}.

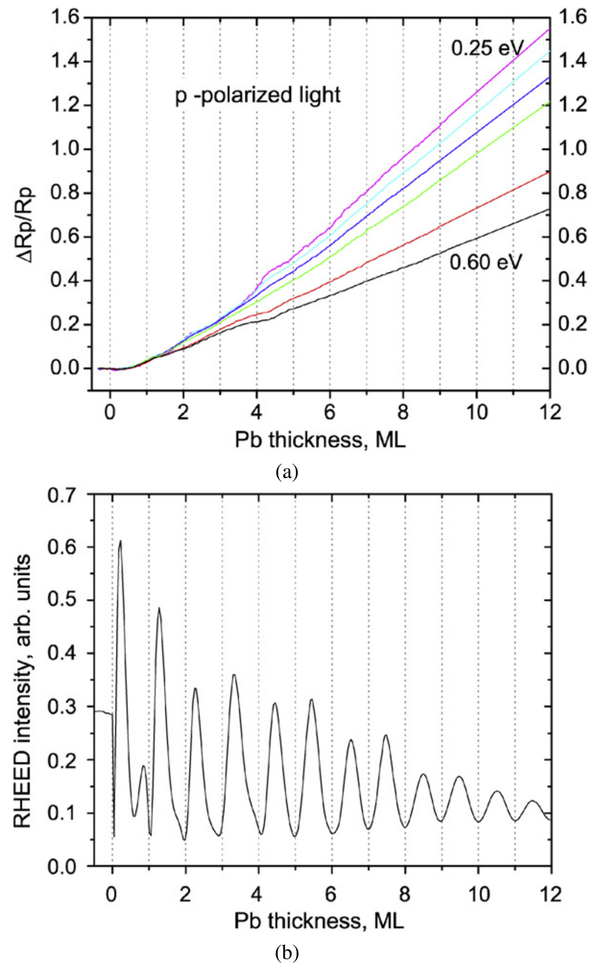

Fig. 1. (a) The differential reflectance $\Delta R/R$ measured *in situ* during Pb deposition on Si(111)-(6 × 6)-Au at 105 K for p-polarized light with fixed energies of incident light 0.25, 0.30, 0.35, 0.40, 0.50, 0.60 eV. (b) RHEED beam intensity oscillations recorded simultaneously with reflectance measurements. The electron energy is 20 keV, glancing angle 0.35°, azimuth Si[11$\bar{2}$].

3. Results and discussion

3.1. Differential reflectance and RHEED intensity measurements

The DR data obtained during Pb growth on Si(111)-(6 × 6)-Au are shown on Fig. 1(a). Each curve was measured a few times for separately prepared thin film samples.

For the thickness greater than 1.5 ML Pb a divergence of the DR curves begins. Changes in reflectance are more essential in the case of the lowest energies. Some unusual features in the form of peaks and dips are observed. Only the curve measured with incident light energy $h\nu = 0.40$ eV increases mostly linearly with the film thickness. The specular RHEED beam