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Optical and transport properties of ultrathin In quantized films

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Abstract
The differential reflectivity (DR) has been measured during In deposition onto a Si(111)-(6×6)Au surface at 105 K. The experiments were done with s-polarized light from the infrared spectral range (0.26–0.35 eV). The imaginary part of the In dielectric function and mean free path (scattering time) of the electrons were determined from the experimental data. The presented results show two series of maxima which repeat with a period of 1 and 3 monolayers of In(111). They can be explained by periodical changes in surface roughness during layer-by-layer In growth and the influence of the quantum size effect (QSE), respectively. It is shown that the DR technique is a suitable probe for characterization of the size effect phenomena and transport properties in ultrathin metallic films or quantum wells.

1. Introduction

The In/Si system has been the subject of active structural investigation. Adsorption of In on Si(111) substrates gives rise to a variety of surface reconstructions [1, 2] in dependence on the coverage and substrate temperature with a tendency to be semiconducting below 1 monolayer (ML) or to be metallic at larger coverages. The attractiveness of a metal on semiconductor system at film coverages of a few monolayers is connected with the fact that it can form metallic quantum wells.

Previously Stróżak *et al* [3] presented results for growth of In on Si(111) surfaces at 110 K investigated by means of reflection high energy electron diffraction (RHEED). The occurrence of the RHEED intensity oscillations measured during the deposition of In on Si(111)-(7 × 7) and Si(111)-(6 × 6)–Au surfaces indicates a monolayer-by-monolayer growth mode. In turn, the well ordered growth may be revealed in appropriate changes of the optical and transport

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properties of ultrathin films. Structure-dependent electrical conduction was characterized in In films on the Si(111)-(7 × 7) clean surface and the Si(111)-($\sqrt{3} \times \sqrt{3}$)–In reconstructed surface deposited at various substrate temperatures [2]. It was assumed that electrical conductance at lower temperatures differs from the results obtained at room temperature because of the difference in surface structural transformations. The appearance of size effects was observed in the low temperature electrical resistivity measurements of ultrathin Pb and Pb–In films on a Si(111)-(6 × 6)–Au surface [4]. Recently quantum and classical size effects were seen in reflection and transmission spectroscopy studies of the same metal on semiconductor systems under similar growth conditions [5, 6].

In this paper we focus on the optical and transport properties of In quantized films on a Si(111)-(6 × 6)–Au substrate at 105 K. We present data from differential reflectivity (DR) measurements from the infrared spectral range as a function of the deposited In film thickness. Results are supplemented with simultaneously measured RHEED intensity oscillations. Quantitative analysis of changes in optical reflectivity in the frame of the classical McIntyre and Aspnes three-phase model [7] allowed us to determined the behavior of the imaginary part of the dielectric function of ultrathin In films. The analysis of transport properties was done according to classical Drude and quantum Keller–Liu local field theories [8, 9].

2. Experimental details

All experiments were performed in an ultra-high vacuum (UHV) chamber with a base pressure of 5×10^{-11} mbar. The pressure during the evaporation was lower than 2×10^{-10} mbar. The UHV setup was equipped with a RHEED system for monitoring substrate quality and the In deposition process. A substrate with dimensions of $18 \times 4 \times 0.6$ mm^3 was cut from a p-type boron Si(111) wafer with 20 Ω cm specific resistivity at room temperature. Before each measurement the substrate was cleaned using direct resistive heating. A few flashes through the sample for 2–3 s with direct current of more than 13.7 A resulted in the appearance of Si(111)-(7 × 7) superstructure in the RHEED pattern. In order to prepare the Si(111)-(6 × 6)–Au surface reconstruction, about 1.2 ML of Au were deposited onto Si(111)-(7 × 7), annealed for 1 min at about 950 K and then gradually lowered to the initial temperature state. The substrate was cooled using a LN_2 cryostat thermally connected to the sample holder. The appearance of a sharp Si(111)-(6 × 6)–Au superstructure was checked in the RHEED pattern. In films were deposited on the Si substrate at 105 K with an average rate 0.6 ML min^{-1}, controlled by a quartz crystal thickness monitor. RHEED intensity oscillations were used to calibrate the quartz monitor.

The optical reflectance changes and RHEED intensity oscillations were measured simultaneously with In deposition. The DR spectroscopy consists of measurements of the relative change of a substrate reflectance upon thin film deposition: $\Delta R/R = (R^{\mathrm{Si+In}} - R^{\mathrm{Si}})/R^{\mathrm{Si}}$, where R^{Si} and $R^{\mathrm{Si+In}}$ are the reflectance of a clear substrate and a substrate covered by an In deposit, respectively. The optical system consisted of a globar, prism monochromator, a Brewster's angle polarizer, a system for focusing the mirrors and a PbSe detector. The probe light beam was s-polarized with a chopped frequency 320 Hz. The angle of incidence of the light beam on the sample surface was equal to $49 \pm 0.5°$. The specularly reflected beam was focused on the detector stabilized at $-10\,°$C. A lock-in technique was used to recover the detected optical signal. The stability of the reflected signal during each measurement was better than 10^{-4}. The DR data were recorded as a function of In film thickness at fixed photon energies within the range 0.26–0.35 eV.

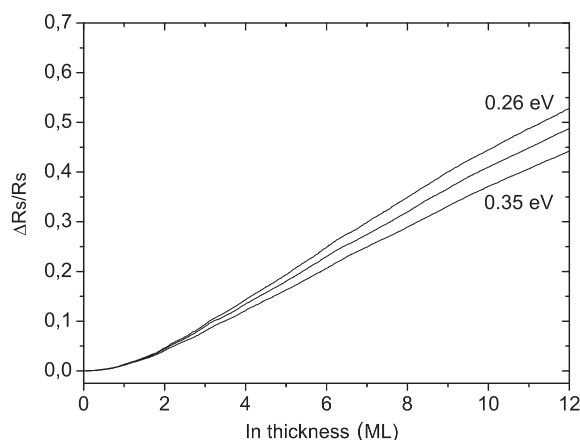

Figure 1. The differential reflectivity $\Delta R/R$ as a function of film thickness measured during In deposition onto a Si(111)-(6 × 6)–Au substrate at 105 K. The presented curves correspond to the DR data obtained for s-polarized light with energies of 0.26, 0.30 and 0.35 eV of the separately prepared thin film sample.

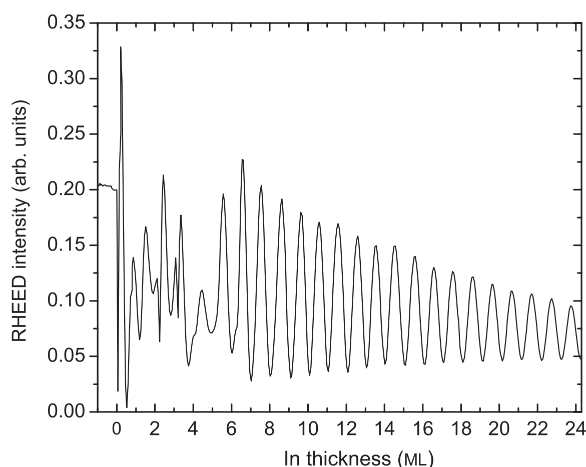

Figure 2. RHEED specular beam intensity variation during In growth on Si(111)-(6 × 6)–Au at 105 K. The electron beam energy is 20 keV, azimuth $[11\bar{2}]$, glacing angle 0.4°.

3. Results and discussion

3.1. Differential reflectivity and growth mode of In on Si(111)-(6 × 6)–Au

Figure 1 presents experimental results of DR measurements for s-polarized light during deposition of In thin film onto a Si(111)-(6 × 6)–Au substrate at 105 K. The data show that the optical response in the infrared spectral region (0.26–0.35 eV) increases with increasing film thickness. Up to a thickness of 1.5 ML In the dependence of $\Delta R/R$ is the same for all curves. For thicknesses greater than 2 ML In divergence of the curves begins. The changes are more significant for smaller energies of incident light. Some unusual features in the form of small deviations from the linear growth of the $\Delta R/R$ are observed.

Simultaneously measured RHEED intensity oscillations are presented in figure 2. Transition between two types of oscillation behavior can be observed. Up to 5 ML In

oscillations with additional peaks were seen from the very beginning. It has been shown that the existence of the extra maxima (additional peaks) indicate a perfect layer growth [10]. In our case it indicates that growth of In on Si(111)-(6 × 6)–Au was in layer-by-layer mode with a 2 ML growth front. After 5 ML In a new set of regular oscillations emerges which is a sign of a rigorous monolayer-by-monolayer film growth (1 ML growth front). Such a behavior of the RHEED intensity corresponds to the crystal structure of the growing film. Another type of RHEED oscillation transition was observed during Pb deposition onto a Si(111)-(7 × 7) substrate. However, the occurrence of transition was connected with a structural transformation from an amorphous-like to a well-ordered crystalline thin film [11].

3.2. Dielectric function of the In films

Several classical models have been proposed to describe changes in reflectance of the sample due to formation of a thin surface layer [7, 12]. In the long-wave approximation, theories that describe differential reflectivity of a thin isotropic and anisotropic layer on a substrate give the same results. It was shown that for p-polarized oblique incident light the main contribution to the optical response is due to the component parallel to the surface. We notice that dielectric functions calculated from s-polarization and p-polarization DR data were the same within an accuracy ±3% for all thicknesses and certain photon energies [5]. In this work we analyzed the results for s-polarized light.

In the classical description of isotropic layer McIntyre and Aspnes showed that when the thickness of a film d is substantially smaller with respect to the wavelength λ of the incident light, DR for s-polarization can be expressed as follows:

$$\frac{\Delta R_s}{R_s} = \frac{R_s^{\mathrm{In+Si}} - R_s^{\mathrm{Si}}}{R_s^{\mathrm{Si}}} = \frac{8\pi d}{\lambda} \cos\varphi \, \mathrm{Im} \left\{ \frac{\varepsilon^{\mathrm{In}} - \varepsilon^{\mathrm{Si}}}{\varepsilon^{\mathrm{Si}} - 1} \right\}, \tag{1}$$

where $\varepsilon^{\mathrm{In}} = \mathrm{Re}\,\varepsilon^{\mathrm{In}} + \mathrm{Im}\,\varepsilon^{\mathrm{In}}$ and $\varepsilon^{\mathrm{Si}} = \mathrm{Re}\,\varepsilon^{\mathrm{Si}} + \mathrm{Im}\,\varepsilon^{\mathrm{Si}}$ are the complex dielectric function of In film and Si substrate, respectively.

Using equation (1) and experimental data from figure 1 we calculate the imaginary part of the dielectric function $\mathrm{Im}\,\varepsilon^{\mathrm{In}}$ which is the main parameter that describe electromagnetic interaction of the light with the sample. For Si the bulk dielectric function after extrapolation to 0.25 eV is equal to $\varepsilon^{\mathrm{Si}} = 13 + \mathrm{i}0.001$ [13]. In the calculations we assumed that the real part of the dielectric function $\mathrm{Re}\,\varepsilon^{\mathrm{In}}$ is equal to the In bulk value [14]. However, equation (1) is almost insensitive to the real part of the dielectric function of In because the imaginary part of the substrate is close to zero in the studied spectral range.

Figure 3 shows the results of the imaginary part of the dielectric function $\mathrm{Im}\,\varepsilon^{\mathrm{In}}$ as a function of In film thickness. The thickness-dependent $\mathrm{Im}\,\varepsilon^{\mathrm{In}}$ shows several prominent features. The dielectric function increases linearly with the film thickness at the initial stage of the deposition process. It is clearly seen that divergence of the curves really begins after deposition of the first monolayer. After that, two series of peaks are observed where the positions of the peaks are independent of the energy of incident light. The small peaks with a period equal to 1 ML In are visible in the thickness range up to 5 ML. They reflect strong changes in the surface roughness over the 2 ML growth front of the indium films. The surface scattering of the free electrons caused by periodic changes in the surface roughness is only important during the initial stage of In growth. This scattering mechanism does not have a considerable influence on the optical properties of the In films at the higher thickness. The absence of the 1 ML periodic variations after deposition of 5 ML In support this idea; however, the surface roughness changes with a period of 1 ML In still exist as one can see from RHEED intensity oscillations (figure 2).

More developed peaks are observed after deposition of 3 ML In and 6 ML In. Similar maxima were observed in the dielectric function dependence of ultrathin Pb films and were

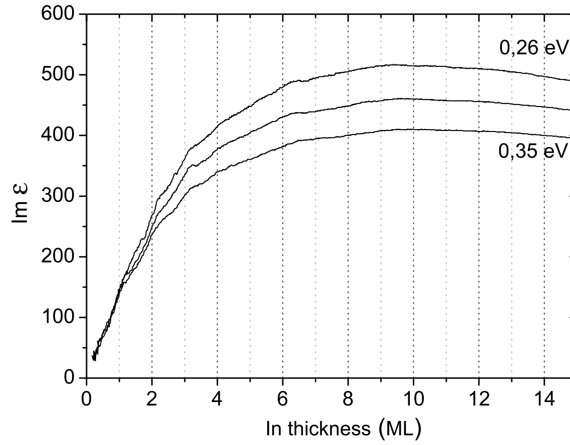

Figure 3. Imaginary part of the dielectric function $\mathrm{Im}\,\varepsilon^{\mathrm{In}}$ of the indium films calculated from experimental data figure 1.

interpreted as the manifestation of the quantum size effect (QSE) [5]. In the free electron model the QSE peaks are expected when the following condition for film thickness is fulfilled: $d = n\lambda_F/2 = n\pi/k_F$. As d can change in multiples of the monolayer thickness d_0 the QSE condition is satisfied only over limited thickness regions. For In the Fermi velocity $v_F = 1.7 \times 10^8$ cm s^{-1} [2] and the Fermi wavevector $k_F = 1.47$ Å^{-1}, so that $\lambda_F/2 = 2.14$ Å, where λ_F is the de Broglie wavelength at the Fermi level. The In(111) ML thickness is $d_0 = a_{\mathrm{In}}/\sqrt{3} = 2.85$ Å. Thus we have $\lambda_F{:}d_0 = 2.14{:}2.85 \approx 3{:}4$ and in consequence the QSE condition $md_0 = n\lambda_F/2$ is approximately fulfilled for $(m, n) = (3, 4)$ and multiples of these pairs. In other metals it can be a different number, for example in Pb films this relation is valid for $(m, n) = (2, 3)$ and QSE oscillations with 2 ML Pb periodicity were experimentally observed [4, 5].

3.3. Transport properties of ultrathin In films

Observed changes in optical properties of ultrathin In films could be interpreted as an optical diamagnetic response of a metallic quantum well in the frame of self-consistent local-field theory [9]. The diamagnetic response plays the main role in the quantum well electrodynamics when the frequency of the light is so low that only intraband transitions take place. For a metallic quantum well it corresponds to frequencies from the infrared spectral range.

Keller and Liu calculated a local field in the quantum well induced by a plane monochromatic electromagnetic wave of cyclic frequency ω incident at an angle θ [9]. This allowed the s-polarized amplitude-reflection coefficient to be obtained:

$$\tilde{r}_s = \frac{r_s + K_{yy}}{1 - K_{yy}}, \tag{2}$$

where r_s is the amplitude-reflection coefficient at the vacuum–substrate interface and K_{yy} takes into account the global charge neutrality condition [15], and can be written in the form

$$K_{yy} = \frac{\mu_0 e^2 \omega\tau(1 + r_s)Nd}{2\pi\hbar^2 q_\perp^0(\omega\tau - 1)Z} \tag{3}$$

where μ_0 is the magnetic permeability of free space, $q_\perp^0$ is the vacuum wavevector component of the incident field perpendicular to the sample surface and Z is the valence of the ions. For

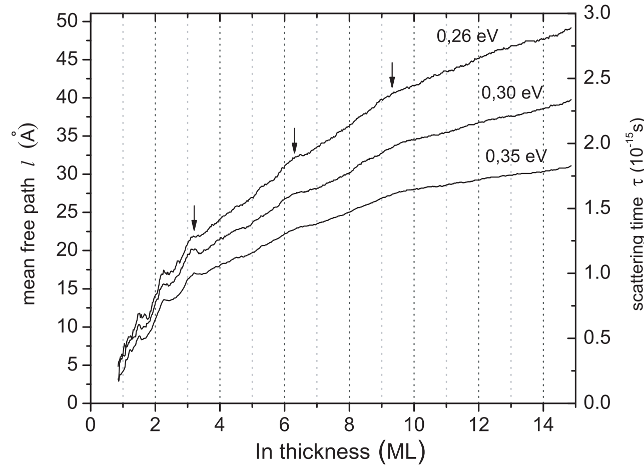

Figure 4. The mean free path l (left) and scattering time τ (right) as a function of thickness for ultrathin indium films calculated using equations (2)–(4). The arrows denote the peaks which are concerned with the quantum size effect.

an infinite potential quantum well in the low-temperature approximation, the electron density N fluctuates with the thickness of the film to maintain a constant chemical potential [16]. It assumed that all electrons are scattered uniformly with a momentum relaxation time denoted as τ. Once the amplitude-reflection coefficient has been determined the energy-reflection coefficient for s-polarized light can be found and compared with the experimental values presented in figure 1 in the form:

$$\frac{\Delta R_s}{R_s} = \frac{|\tilde{r}_s|^2}{|r_s|^2} - 1. \quad (4)$$

We use equations (2)–(4) to derive the mean free path of electrons $l = \upsilon_F\tau$, where υ_F is the electron velocity at the Fermi level. Figure 4 presents the scattering time τ and the mean free path l, respectively, calculated from experimental data figure 1.

The thickness-dependent mean free path (scattering time) shows periodic peaks which are more pronounced than on the dielectric function curves. Besides, for thicknesses above 9 ML In we can observe a peak that confirms the existence of the QSE oscillations with a period 3 ML during In film growth. Modulation of the transport properties with a period of 1 ML at low thicknesses is due to surface scattering, and this supports suggestions of a strong change in surface roughness during the initial stage of In film growth, whereas at higher thicknesses periodic changes in the scattering time are due to the QSE. We suppose that at a thickness of 3 ML In both peaks with 1 and 3 ML oscillation periods are mutually superimposed. Such behavior of the transport properties also shows the crossover from a surface-dominated regime to a regime where electrons scatter on a thickness-dependent number of QSE states. Earlier, Motulevich and Shubin studied the optical constants of In films with a thickness of 0.6 μm at room temperature. They showed that for incident light with $\lambda = 4\ \mu$m ($\hbar\omega = 0.31$ eV) the mean free path of In films is equal to 50 Å [17].

The electromagnetic interaction of the light can be discussed in terms of optical conductivity. In the infrared region the main contribution to optical properties of metals comes from free electrons. For this purpose it worth using the classical Drude model, which is valid even in mesoscopic metallic structures [18]. The Drude formula defines the relationship

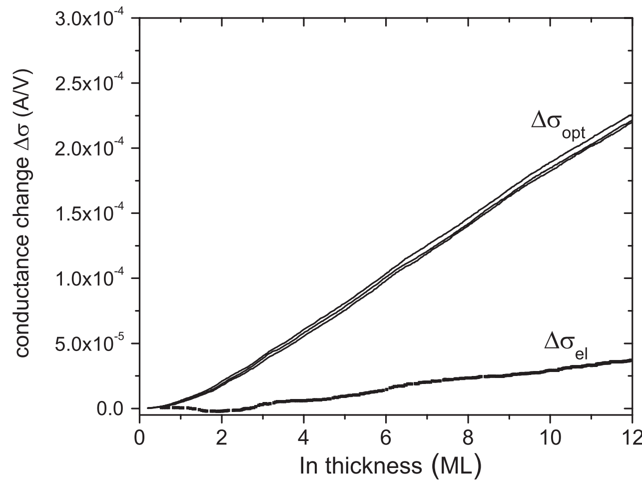

Figure 5. The optical conductance changes $\Delta\sigma_{opt}$ of In films on Si(111)-(6 × 6)–Au at 105 K calculated from the data in figure 3 for energies 0.26–0.35 eV and electrical conductance changes $\Delta\sigma_{el}$ measured during In deposition on Si(111)–($\sqrt{3} \times \sqrt{3}$)–In at 160 K, reproduced from [2].

between the frequency-dependent conductivity $\sigma(\omega)$ and the dielectric function $\varepsilon(\omega)$:

$$\varepsilon(\omega) = 1 + \mathrm{i}\frac{4\pi\sigma(\omega)}{\omega}. \tag{5}$$

Figure 5 shows changes in optical conductance $\Delta\sigma_{opt}$ obtained from the data presented in figure 3 using equation (5) and taking into account continuous changes in film thickness. All curves demonstrate the same thickness-dependent behavior within an accuracy of ±2%. For comparison we present electrical conductance data for the In film deposited onto Si(111)-($\sqrt{3} \times \sqrt{3}$)–In at 160 K. Despite higher values of the conductance in the Si(111)-(6 × 6)–Au substrate [2], changes in both optical and electrical conductance have similar values at the beginning of the In growth. The difference in values of the conductance after further growth can be explained by the different temperature of the substrates during In deposition. For instance, the electrical conductance of 12 ML In film on Si(111)-($\sqrt{3} \times \sqrt{3}$)–In at 88 K is 20 times higher than that observed at 160 K [2]. Strong temperature-dependent conductance can be explained due to the minimization of phonon scattering and more ordered film growth which are characteristic of the low temperature regime. The presented changes in optical and electric conductance show similar behavior, especially in the thickness range 2.5–7 ML In. Peaks with a period of 3 ML In can be observed and, as mentioned above, are recognized as the appearance of the quantum size effect. The oscillations in electrical resistance with a period of 1 nm (3.5 ML In(111)) were previously measured during In deposition on Au film substrate at 80 K [19]. They were explained by the transverse quantization electron states in In films. All of these considerations support the assumption that for a wide temperature range (up to 160 K) In growth in the layer mode and ultrathin films can be regarded as quantum well structures.

4. Conclusion

The optical reflectivity changes during epitaxial growth of In on Si(111)-(6 × 6)–Au have been studied by means of the differential reflectivity technique. The imaginary part of the dielectric function and optical conductivity of ultrathin indium film was determined using the classical McIntyre-Aspnes and Drude theories, respectively. The mean free path (scattering time) was

derived from experimental data using the local-field theory of Keller and Liu. A crossover between two types of periodical maxima has been observed. The maxima with a period of 1 ML In(111) are explained as a consequence of the monolayer-by-monolayer In growth mode. The origin of the peaks with period 3 ML In(111) is due to a QSE effect. The QSE peaks are correlated with those observed in electrical conductance and their occurrence supports the assumption of monolayer-by-monolayer growth of In.

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