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The evolution of sheet-plasmon behavior in silver monolayers on Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag surface

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ABSTRACT

The dispersion of sheet plasmons in two-dimensional electron gas (2DEG) systems prepared on Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag surface was investigated by angle-resolved high-resolution electron-energy-loss spectroscopy (HREELS) in an in-situ ultrahigh vacuum environment. The evolution of dispersion at the low coverage of additional silver adatoms shows a behavior associating with two-dimensional adatom gas phase and the critical coverage to sustain this phase is also determined. Dispersion at higher coverage of additional silver adatoms features a gap opening in the long wavelength limit (momentum $q \rightarrow 0$), suggesting a new mode of plasmon. A calculation based on the theory of surface plasmons implies this plasmon is 3D-type surface plasmon and reveals a smooth transition from a 2D sheet plasmon to a 3D surface plasmon.

1. Introduction

Quasi two-dimensional plasmons were first observed on the liquid helium surface [1], and in inversion layers of Si–SiO_2 structures [2]. The characterization of dispersion for both systems were limited by the structures and the experimental techniques employed, where only electron-density-dependent dispersion was characterized, with a plasmon energy in the order of 10^{-3} meV for liquid helium surface and 1 meV for the inversion layer of Si–SiO_2 due to low electron density. Until recently, a more strictly confined two-dimensional gas system was reported in a silver-induced surface superstructure Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag [3] and a wavevector-dependent dispersion was reported on this surface by using electron energy loss spectroscopy [4]. As those electrons involved in collective excitation are well confined within the top layer of surface, the so-called "sheet plasmons" distinguish themselves primarily from surface plasmons by having a vanishing energy in the long wavelength limit ($q = 0$) rather than a finite value. Angle-revolved HREELS is unique tool to characterize wavevector-dependent dispersion of plasmons on the surface, with rich information in loss structures [5] and scattering mechanism [6]. In this work, HREELS measurements were performed on Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag surface of various thicknesses of Ag-adatom overlayer, disclosing the evolution of sheet-plasmon dispersion from a 2D-type plasmon to a 3D type plasmon. Since surface plasmons of silver single-crystal surface and silver thin-film on Si(1 1 1)-(7×7), including recently discovered acoustic surface plasmon, are well-studied 3D-type plasmon both theoretically and experimentally [7–11], the novel two-dimensional electron system of Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag provides us a proving ground to those established systems and models to further explore electronic properties of a strictly two-dimensional electron gas (2DEG).

2. Experimental

The silicon substrates were cut from commercial-manufactured Si(1 1 1) wafers including both p-type and *n*-type bulk doping. Before introducing into ultra-high vacuum, samples were chemically cleaned to remove organical and metallic particles adsorbed on wafer surfaces. The base pressure of ultrahigh vacuum chamber is 2×10^{-10} Torr, samples were degassed at around 200 °C for about 12 h. After degassing, samples were slowly heated to 900 °C and flashed to 1200 °C for 10 s to remove silicon oxides, followed by gradually cooling down to room temperature. Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag was prepared by depositing 1 ML silver onto a Si(1 1 1)-(7×7) substrate at 500 °C. The deposition rate was monitored by quartz crystal oscillator which can be moved directly in front of sample. The thickness of silver, expressed here in the unit of ML (monolayer, corresponding to 7.8×10^{14} atoms cm^{-2}, areal density of silicon atom on Si(1 1 1) surface), was controlled by tuning the time of exposure to the source. A low-energy electron diffraction (LEED) was used to check the surface superstructure in situ during the sample preparing process. The prepared sample with Si(1 1 1)-($\sqrt{3} \times \sqrt{3}$)-Ag surface was transported into HREELS chamber and measurements were performed in situ. A medium energy resolution

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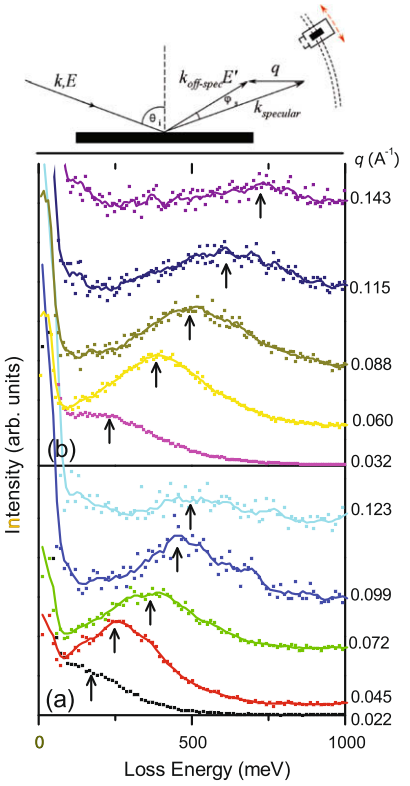

Fig. 1. HREELS spectra of (a) pristine Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface and (b) surface deposited with additional 0.02 ML silver adatoms. The lines in magnified spectra are smoothed by averaging five nearest neighbor points. Electrons impact energy E_o = 20.29 eV and incident angle $\theta_i = 60°$. Inset shows the scattering geometry.

of ~ 10 meV was set to achieve strong signal, and the momentum resolution was better than 0.01 Å^{-1}[1], which is small enough to resolve the concentrated peak profile of dipole scattering [6].

3. Results and discussion

3.1. Sub-monolayer deposition of silver adatoms

Fig. 1 shows the energy-loss peaks in the HREELS spectra from Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface, increasing and dispersing with increasing momentum transfer parallel to the surface q given in Å^{-1}. The black arrows in Fig. 1 indicate the center of loss peaks corresponding to the plasmon energy at the certain momentum transfer q.

In Fig. 2, plasmon energies of various thicknesses of silver adatoms are plotted against momentum transfer to show how sheet-plasmon dispersion behavior evolves with deposition of additional silver adatoms on Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface. All measurements were performed consecutively on the same sample prepared in situ. For each deposition, HREELS measurements started from scattering angle $\phi_s = 0°$ (specular direction) to $\phi_s = 5°$ with the step of 1° (inset of Fig. 1).

A model describing two-dimensional electron gas was derived by Stern within the framework of the nonlocal response theory by random phase approximation (RPA) [12]. The following is an approximation of plasmon frequency ω_{2D} up to the second-order term in q:

$$\omega_{2D} = \left[\frac{4\pi n_{2D}e^2}{m^*(1+\epsilon_0)}|q| + \frac{3}{4}v_F^2q^2 + \ldots\right]^{1/2}, \quad (1)$$

where n_{2D} is areal density of electrons, m^* is effective mass, ϵ_0 is static dielectric constant of substrate and v_F is the Fermi velocity of electrons gas. In Fig. 2a, by setting fitting parameter as n_{2D} and m^* in best fit of the Stern model, Eq. (1) [4,6], the areal density of electrons of various thickness were obtained $n_{2D} = 2.04, 3.09, 1.77 \times 10^{13}$ cm^{-2} for surfaces of 0-ML, 0.02-ML, and 0.05-ML additional silver adatoms, respectively. The dashed line in Fig. 2a is the classic $\sqrt{q}$ dispersion behavior with the same electron density as pristine Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface (0 ML additional silver adatoms).

The blue shift and increased steepness in the dispersion of surface with additional 0.02-ML silver adatoms are due to the charge transfer from silver adatoms to surface state of Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag. This observation is consistent with a photoemission study on Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface, in which the filling of surface state band and corresponding enlargement of Fermi surface were observed by depositing silver adatoms, indicative of an increase of electron density [13].

The other feature from the evolving dispersions is that a critical coverage $\Theta_c \sim 0.02$ ML of additional silver adatoms exists on Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface, which is borne out in Fig. 2a.[2] This critical coverage was also reported in measurements of electrical conduction [14] and ultraviolet photoelectron spectroscopy [15]. Below Θ_c, a so-called two-dimensional adatom gas phase is formed, and remarkably increases the electron density of the surface, contributing to higher plasmon energy and surface conductance. A frozen two-dimensional adatom gas was also imaged by low-temperature STM study [16]. Continued deposition exceeding Θ_c, the two-dimensional adatom gas phase is destroyed by formations of silver micro-crystals where nucleation centers work like a sinker to trap all the highly mobile silver adatoms.

Fig. 2b shows plasmon dispersion behaviors measured up to 1.08-ML additional silver adatoms. There is not much difference between 0.05 ML (red curve, Fig. 2b) and 0.08 ML (cyan curve, Fig. 2b), however, a striking change was observed when 1.08 ML additional silver was deposited (purple curve, Fig. 2b). The smooth extrapolation into the long wavelength limit suggested a finite gap and the linear dispersion changed to a more complicated curvature where Stern's model failed to fit this dispersion. Quantitatively speaking, the best fitting of the 1.08-ML surface gave $n_{2D} = 3.60 \pm 3.48 \times 10^{13}$ cm^{-2}, in which the uncertainty was in the same magnitude as electron density itself. In order to characterize this behavior, a series of more detailed measurements starting with 0.07 ML to 3.75 ML of additional silver adatoms were performed in situ, followed by a discussion of gap energy within the framework of an evolving surface plasmon in the next section.

3.2. Few-layer deposition of silver adatoms

As shown in Fig. 3, the tendency to the opening of a gap at long wavelengths becomes more distinct with the increasing thickness of silver adatoms. Due to the specular beam intensity in HREELS, we cannot experimentally investigate this behavior at $q = 0$). However, this energy gap can be extrapolated from explicit behavior of

[1] The instrument was a commercial LK2000-14-R spectrometer (LK Technologies) with slits of 0.13 mm of two primary monochromators. The angular acceptance α is 0.15°, the estimated momentum resolution Δq is given by $\sqrt{2mE_i/\hbar}(\cos\theta_i + \sqrt{1 - E_{loss}/E_i}\cos\theta_s)\alpha$ (Ref. [7]). Typical setting for our spectrometer: the impact energy E_i = 20 eV, θ_i = 60°, and loss energy $E_{loss} \sim$ 200 meV around specular direction, the Δq=0.0059 Å^{-1}; 5° awary from specular direction where θ_s = 55°, the Δq=0.0063 Å^{-1}.

[2] We noticed there is a discrepancy in the coverage of additional silver adatoms deposited on Si(1 1 1)-($\sqrt{3}\times\sqrt{3}$)-Ag surface, between our result 0.02 ML (1.02 in total) and their single measurement of 0.15 ML reported by Nagao et al. [4]. However our 0.02 ML coverage is in agreement with the critical coverage reported by this same group in later publications [14–16] for the formation of the two-dimensional electron gas phase.