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Observation of a two-dimensional plasmon in a metallic monolayer on silicon surface

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Abstract By use of electron-energy-loss spectrometer with high momentum resolution, we have measured a collective electronic excitation localized at the topmost surface layer on the Si(111)-√3 × √3-Ag surface. The obtained energy dispersion was found to be isotropic with respect to the azimuthal orientation and agreed very well with the plasmon dispersion calculated from 2D nearly-free-electron theory using random phase (RPA) approximation.

1. Introduction

Two-dimensional (2D) plasmon (or sheet plasmon) constitutes an interesting research subject since the charge density distribution of which is restricted in the 2D space and thus shows very different electrodynamical properties compared with those of the 3D-type plasmons (Fig. 1). For example, bulk and surface contributions are always inseparable in the properties of the ***surface plasmons***. On the contrary, properties of the ***2D plasmons*** are mainly determined by those of the 2D electron systems (2DESs) themselves, especially in the cases of 2DESs supported in (semi-)insulating bulk media. We can take advantage of this characteristic for utilizing the 2D plasmon as a sensitive noncontact probe of electronic properties of the 2D electron system (2DES) in the atomically thin region, such as metallic monolayer supported on less conductive dielectric media.

By using electron-energy-loss spectroscopy (EELS), we can determine the plasmon energy as a function of wave number $\boldsymbol{q}_{\parallel}$ with excellent accessible energy range covering from the far-infrared to ultraviolet regimes [1]. The measurement of the energy dispersion curve combined with the theoretical analysis provides us the atomistic information of static and dynamical properties of the 2DES in a great detail.

In the present paper, we report on the direct measurement of the energy dispersion of a 2D plasmon found at the topmost surface layer on the Si(111)-√3 × √3-Ag surface. We have clarified that the quantum statistics and nonlocal effects manifest themselves in the large wave numver region $\boldsymbol{q}_{\parallel} \approx \boldsymbol{k}_F$, where $\boldsymbol{k}_F$ is the Fermi wave number of the 2DES. We have also found good agreement with the calculated dispersion from 2D nearly-free-electron theory [2], and clarified the quantitative agreement with the static electronic properties previously measured by photoelectron spectroscopy.

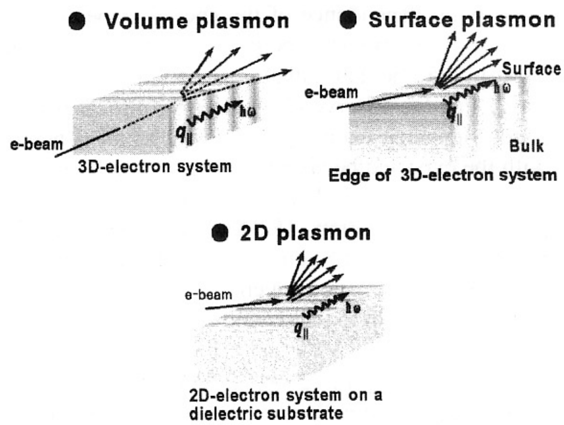

Fig. 1. Charge density wave distribution for the 3D and 2D plasmons.

2. Experimental

The experiment was performed in ultrahigh vacuum with 6×10^{-11} Torr base pressure. The √3×√3-Ag surface was prepared by depositing precisely one monolayer (1 ML) of Ag onto a clean Si(111)-7×7 surface held at around 800 K while keeping the pressure below 1×10^{-9} Torr. Angle resolved electron-energy-loss spectra was recorded by ELS-LEED (energy-loss-spectroscopy with low-energy electron diffraction) which has an energy resolution in meV regime and momentum resolution in 10^{-3} Å^{-1} regime [3].

3. Results and discussion

The Si(111)-√3 × √3-Ag surface is one of the most promising prototype system for investigating 2D plasmons in atomically thin system [4,5]. There is an electron pocket centered at the Γ point of the surface Brillouin zone of a nearly parabolic band, and constitutes a nearly-free 2DES at the topmost surface layer. The carrier density in this electron pocket is $N_{2D}=(1.6\pm0.3)\times10^{13}$ /cm^2 and the effective mass $m^*=(0.29\pm0.05)m_e$ (m_e is the free- electron mass), estimated from photoelectron spectroscopy (PES) measurements [5].

The plasmon energy dispersion is plotted as a function of wave vector $q_\parallel$ in Fig. 2. As clearly seen, there is no dependence of the energy dispersion on the electron primary energy E_p (electron probing depth) and the azimuthal orientation. These fact imply that the observed loss has a strong 2D nature. Also, vanishing excitation energy at small $q_\parallel$ agrees with the expected behavior of 2D plasmon [2,6].

Using the "nonlocal" response theory with random phase approximation (RPA), we analyzed the plasmon in a 2DES on a semi-infinite dielectric substrate. For convenience, we show an approximation up to the second-order term in $q_\parallel$,

$$\omega_{2D}(q_\parallel) = [\,4\pi N_{2D} e^2 m^{*-1} (1+\varepsilon_{Si})^{-1}\, q_\parallel + 6N_{2D}\hbar^2\pi\,(2m^*)^{-2}\, q_\parallel^2 + O(q_\parallel^3)\,]^{1/2}.$$

Here, $\omega_{2D}(q_\parallel)$ is the 2D plasma frequency, N_{2D} is the areal density of electrons in the 2DES, $\varepsilon_{Si}=11.5$ is the dielectric constant of the Si, m^* is the electron's effective mass, and e is the elementary electric charge.

The first term is identical to the $\sqrt{q_\parallel}$ term from the classical local response theory [6]. Second- and higher-order terms originate from nonlocal effects and reflect the quantum statistics of the 2DES; for example, the second term is rewritten as $(3/4)v_F^{\,2}q_\parallel^{\,2}$ with the Fermi velocity v_F of the 2DES and exhibits the degeneracy of the electron system.

The solid curve in Fig.2 is the best fit to the nonanalitic full RPA dispersion. The electron density and the electron effective mass are determined to be $N_{2D}=1.9\times10^{13}$ /cm^2 and $m^*=0.30\ m_e$, respectively which agreed very well with the values form the PES measurements given above. The overall fit is excellent, and compared with the "local" $\sqrt{q_\parallel}$ dispersion calculated using the same N_{2D} and m^* values shown by the dashed curve, improvement in the fit at larger $q_\parallel$ value is clearly seen. This indicates the importance of including nonlocal effects and quantum statistics in analyzing the energy dispersion of the 2D plasmons. Moreover, fitting over a wide $q_\parallel$ range with the "nonlocal" full RPA dispersion yields the values of N_{2D} and m^* simultaneously, while in the "local" $\sqrt{q_\parallel}$ case, only their ratio is obtained

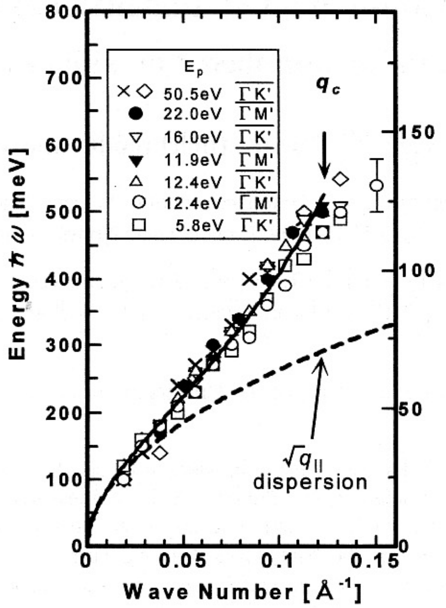

Fig. 2. Plasmon dispersion plot measured with $q_\parallel$ scanning along ΓK′ and ΓM′ directions at 90 K.

4. Summary

We have measured the dispersion of a 2D plasmon localized at the topmost atomic layer of the Si(111)-√3 × √3-Ag surface. We have found that the energy dispersion is isotropic with respect to the azimuthal orientation and shows no dependence on the electron probing depth. We have clarified that the observed energy dispersion does not agree with $\sqrt{q_\parallel}$ dispersion by the classical local response theory, but agrees very well with the full RPA dispersion, especially at large $q_\parallel$ values.

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