Dispersion and Damping of a Two-Dimensional Plasmon in a Metallic Surface-State Band

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(Received 18 September 2000)

We have studied, for the first time, the energy and the linewidth dispersion of a plasmon in a dense two-dimensional electron system in a metallic surface-state band on a silicon surface. As expected from the considerably high effective density and long Fermi wavelength of the system, the plasmon energy dispersion exhibited an excellent agreement with the nearly free-electron theory. However, in a small wave number region below the Landau edge, we have observed an anomalous linewidth dispersion which nearly free-electron theories do not predict.

DOI: 10.1103/PhysRevLett.86.5747 PACS numbers: 73.20.Mf, 71.45.Gm

A considerable number of studies exist on *volume* and *surface plasmons* in three-dimensional electron systems, discussing correlations between plasmons and optical properties [1–3], as well as elucidating nonlocal effects in the electrodynamics at the solid-vacuum interfaces [4–6]. The *two-dimensional (2D) plasmon* (or *sheet plasmon*), whose oscillating charge density distribution is strictly confined to a 2D space, is expected to constitute another attractive topic through which we can look into the dynamical aspect of a 2D conductive phase in solids. Experimentally, the 2D plasmon was first observed for a dilute classical 2D electron system (2DES) on a liquid He surface [7], and later in dilute but already degenerate quasi-2DESs in Si–metal-oxide semiconductor field-effect transistors or in GaAlAs quantum well structures [8,9]. In these artificially tailored systems, large electron spacing, or long Fermi wavelengths ($\sim$1000 Å) were (unconsciously) utilized to avoid the complexity associated with the background short-period ion lattice. Their energy dispersions were studied in a tiny wave number region ($\mathbf{q}_\parallel \ll 0.01$ Å^{-1}) where they are describable by classical local response theory and are kept free from lifetime broadening by Landau damping.

In spite of its broad interest, study on the dispersion and damping of the 2D plasmon in a "dense" and "atomically thin" system has been lacking so far. The reason is mainly ascribed to the lack of an appropriate spectroscopic technique with a wide accessible $\mathbf{q}_\parallel$ range (0–2 Å^{-1}) as well as a wide energy range from the far-infrared to the ultraviolet regime, necessary for observing the peculiar dispersion of 2D plasmon. High-resolution electron-energy-loss spectroscopy (HREELS) is the strongest candidate which nearly meets the above requirements, but its typical momentum resolution (0.05–0.1 Å^{-1}) is not high enough for resolving the steeply dispersing loss by 2D plasmon.

In the present Letter, we report on the first experimental result on both the energy and the linewidth dispersion of a 2D plasmon found in a dense 2DES in a surface-state band on Si(111)-($\sqrt{3} \times \sqrt{3}$)-Ag. The atomic and electronic structures of this surface have been well studied, and the presence of a surface-state band with an electron pocket centered at the Γ point of the surface Brillouin zone was clarified [10,11]. This free-electron-like band (called $S1$ band) is energetically well separated from other surface and bulk bands and, thus, keeps the 2D plasmon free from coupling to any interband transitions. This simple and clean situation renders this surface one of the most suitable and promising arenas of opportunity for exploring a plasmon in a dense 2DES confined in an atomically thin region.

Our experiment was performed in ultrahigh vacuum (6×10^{-11} Torr base pressure) using an ELS-LEED (energy-loss spectroscopy with low-energy electron diffraction, Henzler-type EELS) which can be operated as an HREELS with an improved momentum resolution in the 10^{-3} Å^{-1} regime [12]. The energy resolution was 20–30 meV, limited by the presence of multiplasmon excitations in the bulk and the metallic $S1$ band in the specular tuning geometry. The ($\sqrt{3} \times \sqrt{3}$)-Ag surface was prepared by depositing precisely one monolayer (1 ML) of Ag onto a clean Si(111)-(7 × 7) surface heated at around 800 K. The domain size of the $\sqrt{3} \times \sqrt{3}$ structure was larger than 600 Å (corresponds to 0.01 Å^{-1} in reciprocal space), as confirmed by the ELS-LEED operated in the high-resolution LEED mode, to minimize the scattering of plasmon at domain boundaries.

Figure 1(A) shows typical angle-resolved EELS spectra taken at 90 K along the $\Gamma M'$ direction from the 1 ML ($\sqrt{3} \times \sqrt{3}$)-Ag surface (hereafter referred to as the "pristine" $\sqrt{3} \times \sqrt{3}$ surface). A single loss peak is clearly observed to disperse as a function of $\mathbf{q}_\parallel$ accompanied with monotonic linewidth broadening. Figure 1(A′) shows another series of spectra from the pristine $\sqrt{3} \times \sqrt{3}$ surface taken along the $\Gamma K'$ direction. Figure 1(B) shows the

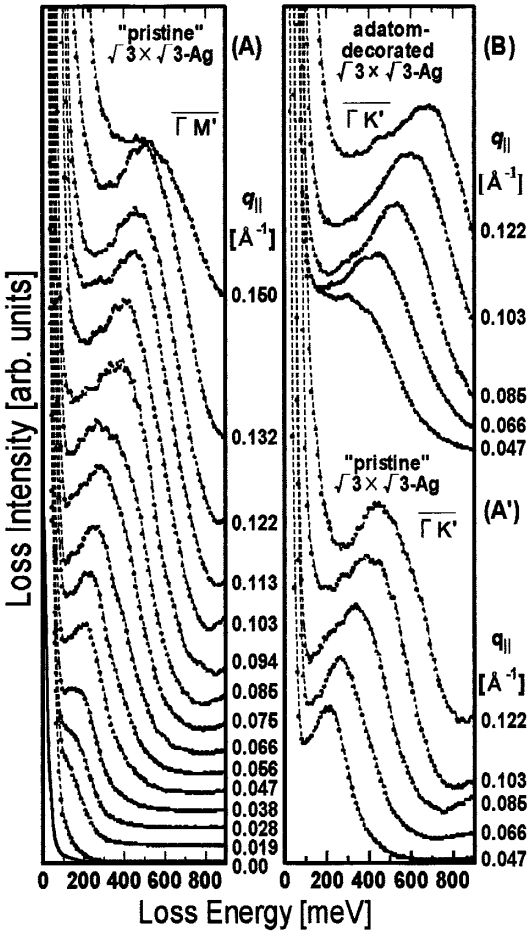

FIG. 1. Angle-resolved EELS spectra taken with electron primary energy $E_p = 12.4$ eV. (A) An example of $\mathbf{q}_\parallel$ scanning along the $\Gamma M'$ direction is shown. (A′) and (B): The blueshift in energy dispersion due to the additional adatom decoration is demonstrated. Each spectrum was magnified to compensate for the intensity drop as a function of $\mathbf{q}_\parallel$.

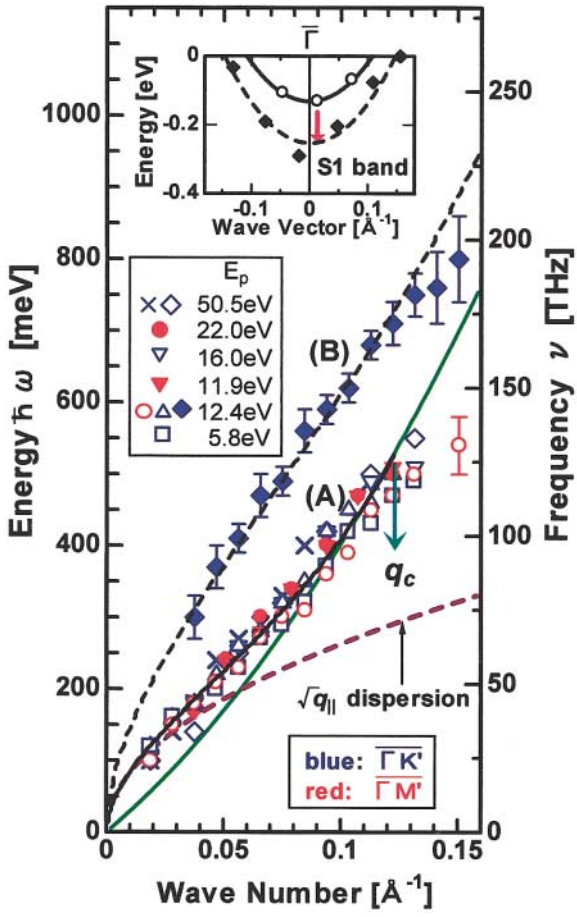

FIG. 2 (color). Plasmon dispersions measured at 90 K from (*A*) the "pristine" $\sqrt{3} \times \sqrt{3}$ surface (1 ML) and (*B*) the $\sqrt{3} \times \sqrt{3}$ surface decorated with 0.15 ML of additional Ag (1.15 ML in total). Blue and red plots represent the data obtained by scanning $\mathbf{q}_\parallel$ along the $\Gamma K'$ and $\Gamma M'$ directions, respectively. The bold solid and dashed curves are the fits by full RPA dispersion. The green curve is the upper edge of the single-particle excitation continuum and the green arrow indicates the Landau edge, $\mathbf{q}_c$, calculated for the pristine surface. The inset is a 2D band dispersion diagram showing the "sinkage" of the $S1$ band (the solid curve corresponds to the pristine surface, and the dashed curve corresponds to the adatom-decorated surface) [11].

spectra obtained from the $\sqrt{3} \times \sqrt{3}$ surface decorated with a small additional amount of Ag adatoms loosely bound to the substrate $\sqrt{3} \times \sqrt{3}$ surface held at 90 K [11,13]. As clearly seen by comparing Fig. 1(A′) with Fig. 1(B), the energy position of the loss shifts, sensitively reflecting the change in the surface electronic structure induced by the adatom decoration.

Figure 2 shows the energy dispersion plot as determined from the peak positions of the losses in the EELS spectra. Peak positions of the losses were determined by deconvolutions, using two Lorentzians and a constant background. The linewidth values were determined by further deconvolution of the loss peaks with respect to the energy and $\mathbf{q}$-space resolution of the spectrometer.

It is clear that the change in electron primary energy E_p (or, change in the probing depth) does not affect the energy position of the losses. Also, the difference in bulk doping of Si has negligible influence on the energy dispersion. For example, in curve 2(*A*), solid red circles and triangles represent the data obtained from an *n*-type silicon wafer with 15 Ω cm resistivity, and the remainder were obtained from a *p*-type 0.05 Ω cm wafer. These findings indicate that the excitation has its origin neither in the surface space-charge layer nor in the bulk, and thus identify it as the excitation in the topmost surface layer. The energy of a 2D plasmon is expected to approach 0 as $\mathbf{q}_\parallel$ vanishes since the restoring force for a charge density wave in 2D approaches zero in the long-wavelength limit. This is clearly seen in our data. As hallmarked from the above observations, the observed loss is thus assigned to the 2D plasmon in the $S1$ band.

Within the framework of the nonlocal response theory using random phase approximation (RPA), Stern has calculated the plasmon dispersion of a 2D nearly free-electron (NFE) system [14]. Using his theory, we analyze plasmons in the 2DES on a semi-infinite dielectric substrate. The following is an approximation up to the second-order term in $|\mathbf{q}_\parallel|$:

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

Here, $\omega_{2D}(\mathbf{q}_\parallel)$ is the 2D plasma frequency, N_{2D} is the areal density of electrons, m^* is the effective electron mass, and e is the elementary electric charge. ε_{Si} is the dielectric constant of the Si substrate which is nearly dispersionless ($\varepsilon_{Si} = 10.5-11.5$) within the frequency range of interest [15]. Since the difference in the plasmon frequency is only less than 3.5% over the entire dispersion curve for the constant ε_{Si} case and for the dispersing ε_{Si} case, we used a constant value $\varepsilon_{Si} = 11.5$ and adjusted N_{2D} and m_e values for the fitting. The first term in the right-hand side is identical to $\sqrt{|\mathbf{q}_\parallel|}$ dispersion from the classical local response theory for a very thin film [16]. The second- and higher-order terms originate from nonlocal effects and reflect the degeneracy, or quantum statistics of the 2DES; for example, the second term is rewritten as $\frac{3}{4}v_F^2\mathbf{q}_\parallel^2$ with the Fermi velocity v_F of the 2DES.

The solid curve in Fig. 2 is the best fit to the experimental data (A) according to Stern's full RPA dispersion. The electron density and the electron effective mass are determined to be $N_{2D} = 1.9 \times 10^{13}$ cm^{-2} and $m^* = 0.30m_e$, respectively, where m_e is the free-electron mass. The overall fit is excellent, and, compared with the "classical" $\sqrt{|\mathbf{q}_\parallel|}$ dispersion calculated using the same N_{2D} and m^* values shown by the brown dashed curve, better agreement in the larger $\mathbf{q}_\parallel$ region is evident. This demonstrates the necessity of going up to large $\mathbf{q}_\parallel$ in examining quantum effects in 2D plasmons. Moreover, fitting over a wide $\mathbf{q}_\parallel$ range with the full RPA dispersion yields the values of N_{2D} and m^* simultaneously, while, with the classical $\sqrt{|\mathbf{q}_\parallel|}$ dispersion, only their ratio is obtained. The above values of N_{2D} and m^* agree with the values obtained from photoelectron spectroscopy (PES) measurements for the $S1$ band, $N_{2D} = (1.3-1.9) \times 10^{13}$ cm^{-2} and $m^* = (0.24-0.34)m_e$ [11]. Consequently, the estimated Fermi wave number $\mathbf{k}_F = (2\pi N_{2D})^{1/2} = 0.109$ Å^{-1} and the Fermi energy $E_F = 2\pi N_{2D}\hbar^2(2m^*)^{-1} = 0.15$ eV also agree very well with the reported ones ($\mathbf{k}_F = 0.10-0.12$ Å^{-1}, $E_F = (0.14-0.16)$ eV [11]).

Upon depositing 0.15 ML of randomly adsorbed Ag adatoms onto the pristine $(\sqrt{3} \times \sqrt{3})$-Ag surface, a dramatic blueshift in the plasmon dispersion is observed, as shown in Fig. 1(B) and curve 2(B). The fit by Stern's full RPA dispersion (bold dashed curve) again agrees well with the data and yields $N_{2D} = (6-8) \times 10^{13}$ cm^{-2} and $m^* = (0.34-0.38)m_e$. The m^* value for 2(B) is close to that for 2(A), which indicates that the dispersion of the surface-state band is not much changed. Most of the increase in N_{2D} is therefore ascribed to the sinkage of the $S1$ band, as displayed in the inset of Fig. 2 [11]. If we tentatively adopt here the simplest Langmuir-Gurney model (electron transfer model), in order to have a quantitative measure of the phenomenon, the amount of nominal charge donated to the $S1$ band is $\Delta Q = (0.35-0.51)e$/Ag, which is half of the value estimated from a PES result obtained at a lower adatom density (0.022 ML) [11]. The smaller electron transfer in our case will be ascribed to electron backdonation to the $S1$ state due to the increase in adatom-adatom interactions [17].

Since Stern's theory describes a longitudinal plasma in a 2D NFE system, the nearly perfect agreement found between the experiment and the theory suggests that electron coupling in our 2DES is not very strong. The effective density parameter $r_s^* = (\pi N_{2D})^{-1/2} a_B^{*-1}$, which expresses the extent of electron-electron coupling in the 2DES, is 1.2 for the pristine $\sqrt{3} \times \sqrt{3}$ surface and 0.63–0.83 for the "adatom-decorated" $\sqrt{3} \times \sqrt{3}$ surface where the effective Bohr radius a_B^* is defined by $a_B^* = [(\varepsilon_{Si} + 1)/2](m_e/m^*)a_B$ with the Bohr radius $a_B = 0.529$ Å. That the r_s^* values are located outside the conventional metallic density region, even smaller than the density parameters of aluminum ($r_s = 2.1$), implies that electron-electron coupling in the present 2DES is moderate, as we have inferred above.

It is surprising that we can develop the above discussions without encountering any difficulty due to the presence of the background ion lattice which is regarded as the main cause of the disagreements between the NFE theories and the experiments for volume plasmons, even for the simplest alkali metals [2,3]. The reasons why our 2DES is free from such complexity are considered as follows. (i) The plasmon energy is small enough compared with any possible excitation energy for the core and the valence band electrons. (ii) Ion potential is partly screened by the 2DES with high effective density. (iii) Because of the long Fermi wavelength ($\lambda_F = 42-58$ Å) of our 2DES, the wavelengths of the constituent electronic wave functions in the ground state and of the plasmon modes are substantially longer than the ion spacing, thus rendering the excited state of the 2DES insensitive to the short-period lattice. Because of these advantages, all that must be considered is the mean static polarizability contribution at the substrate-vacuum interface, which is well represented by $(\varepsilon_{Si} + 1)/2$. This "clean and ideal" situation makes our 2DES a nearly perfect experimental realization of an ideal 2D Fermi fluid with a metallic density.

Figures 3(A) and 3(B) show the $\mathbf{q}_\parallel$ dependence of the relative loss intensity [normalized to the (00) spot intensity] and the plasmon linewidth $\hbar\Delta\omega$ for the pristine $\sqrt{3} \times \sqrt{3}$ surface, respectively. The relative intensity rapidly drops by 3 orders of magnitude within a small $\mathbf{q}_\parallel$ range, reflecting the sharp angular distribution of the dipole lobe [18]. At around $\mathbf{q}_\parallel = 0.13$ Å^{-1}, the intensity drops even more rapidly and swiftly disappears. This additional drop suggests the onset of Landau damping where the plasmon branch enters single-particle excitation continua and decays into electron-hole pairs via strong wave-particle interaction [1,2]. Our RPA calculation predicts that this damping sets in at $\mathbf{q}_c = 0.125$ Å^{-1} (where the calculated dispersion ends; see Fig. 2) which nearly corresponds to the experimental value given above.

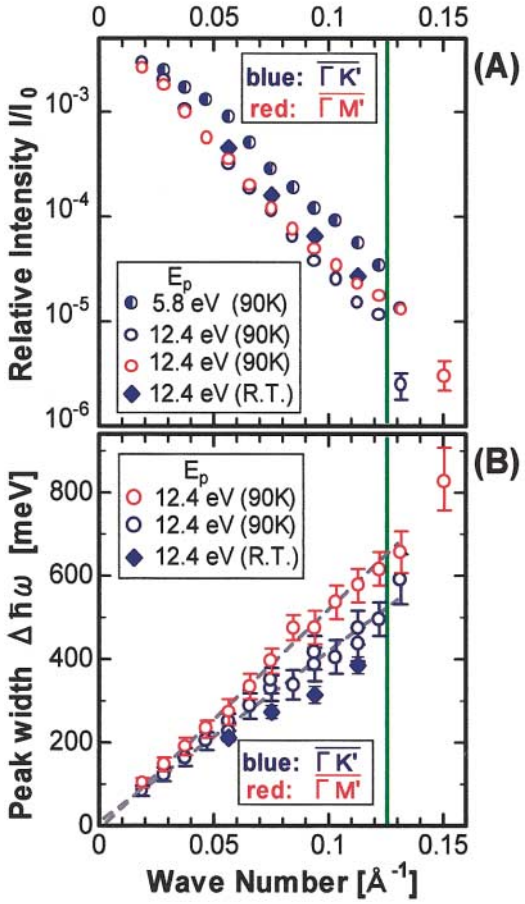

FIG. 3 (color). (A) Relative loss intensity plotted as a function of wave number $\mathbf{q}_{\|}$. (B) Plasmon linewidth plotted as a function of $\mathbf{q}_{\|}$. The Landau edge, $\mathbf{q}_c$, is indicated by the green lines.

At smaller wave numbers below $\mathbf{q}_c$, where no Landau damping is operative, a clear, strongly dispersing damping is observed, as shown in Fig. 3(B). It should be noted that the interband transitions, which are regarded as the main decay channel for volume plasmons [2,3], are evidently excluded here since there is no transition energy close to the plasmon energy. We have clarified that the observed damping has no relevance to phonon scattering or phonon-assisted intraband transitions, since the linewidth does not increase upon elevating the sample temperature [compare the blue open circles and solid diamonds in Fig. 3(B)]. Neither can it be attributed to impurity scattering or impurity-assisted processes since introducing scattering centers by the additional adatom-decoration causes a parallel upward shift of the linewidth-dispersion curve, rather than affecting its slope [19].

The most possible mechanisms left, which also meet energy and momentum conservation, are the decay via two (and higher multiple) electron-hole pairs, induced by Coulomb collision or electron-electron scattering [1,3,20]. Presence of this type of damping was strongly suggested for volume plasmon in alkali metals, where the r_s values are of the same order as the present system [3,20]. Thus, the observed linewidth dispersion strongly suggests that the electronic correlation effect plays a more important role and manifests itself more clearly in the aspect of plasmon damping than in the energy dispersion of our 2D plasmon.

In conclusion, we have measured the dispersion and the damping of a 2D plasmon in a dense 2DES in a silicon surface-state band. The energy dispersion of the plasmon behaves similar to that of a nearly ideal 2D Fermi gas because of its high effective density and the small influence of the lattice and band structures. On the contrary, the plasmon linewidth revealed an unexpected dispersion which NFE theories do not predict. We consider it as the plasmon damping via multiple electron-hole pair excitations induced by electron-electron scattering.

T. N. acknowledges Professor T. Inaoka, Professor C. Oshima, Professor V. U. Nazarov, Professor A. Liebsch, and Professor H. Ishida for helpful discussions.

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