sive Brillouin shifts for $\vec{E} \| \vec{c}$ although these shifts are quite different from those for $\vec{E} \perp \vec{c}$. Only the dielectric parameters (background dielectric constant $\epsilon_{b\|}$ and oscillator strength $4\pi\beta_{\|}$) needed to be slightly adjusted.

Recently the k-linear coefficient of the conduction band in CdS has been measured by spin-flip Raman scattering[9] ($C^e = 1.6 \times 10^{-10}$ eV cm). C^e, however, has only a small effect on φ because it enters Eq. (1) in a product with the mass ratio $m_{e\perp}/m_{\perp}$. Using Eq. (1), the corresponding coefficient of the second valence band is found to be $|C^h| = (6.7 \pm 0.7) \times 10^{-10}$ eV cm. The k-linear terms of the energy bands in CdS have been assumed to arise from spin-orbit coupling.[9,16] In this interpretation the ratio $|C^h/C^e|$ was estimated to be $\sim 10^2$.[16] The present experimental values, however, result in the much smaller ratio of 4. Thus the physical origin of the observed k-linear terms requires further theoretical study.

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Bulk Plasmon Dispersion in Si for $0 < q < 1.5 q_F$

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This paper reports measurement of bulk plasmon dispersion in single-crystal silicon along the [100] and [110] directions using electron energy-loss spectroscopy. An electron beam energy of 250 keV allows one to observe the plasmon peak up to wave vectors of $q = 2.55$ Å^{-1} along [100]. It is shown that the plasmon is still dispersive at large q, contrary to what has been reported.

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Despite many experimental efforts[1-6] addressing the problem of plasmon behavior in an electron gas at large wave vectors ($q > q_F$ = Fermi wave vector), results still disagree. Early measurements of plasmon dispersion in Al by electron energy-loss spectroscopy[2,3] indicated that the plasmon energy became constant as a function of q for $q > q_c$ (cut-off wave vector of plasmons) and suggested a possible breakdown of the random-phase approximation (RPA) for the dynamical

structure factor $S(q,\omega)$. This plasmon leveling-off effect was later pointed out by Batson, Chen, and Silcox[5] to be due to a triple scattering process which involved the scattering of a bulk plasmon and a surface plasmon and quasielastic (thermal diffuse scattering or TDS) excitations. After compensating for this effect they find that the plasmon dispersion curve does not level off at large q, but is still dispersive and follows closely the predictions of RPA. However, in that work rather complicated data analysis was used to remove the effects of multiple scattering, and this has produced some resistance to the acceptance of the result.

Very recently, plasmon dispersion at large wave vectors ($q = 2.5$ Å^{-1}) in Si has been reported by Stiebling and Raether.[6] Their measurements indicated that the plasmon dispersion curve became flat at $\hbar\omega = 23.5$ eV for large q, and that the triple-scattering process of Batson, Chen, and Silcox cannot be justified in Si. Examination of their experimental results shown in Fig. 3 of Ref. 6 reveals details which are difficult to understand in terms of plasmon excitation. We first note that the width of the 23.5-eV peak at large q tends to stay relatively constant as a function of q. This is hardly reconcilable with plasmon behavior in which the width should increase rapidly with q. Secondly, we note that in the loss spectra for $q = 0.89$ and 1.11 Å^{-1}, their plasmon peaks seem to be narrower than the $q = 0$ plasmon-TDS double scattering peak at about 16.6 eV. Since the plasmon width increases with q, it is inconceivable that plasmons at $q = 1$ Å^{-1} should be narrower than the plasmon peak at $q = 0$. Finally, we note that the intensity of the dispersed plasmon is less than that of the double scattering peak for q as small as 0.89 Å^{-1}, clearly indicating that multiple scattering involving TDS is rather strong.

Here we would like to report a measurement of the plasmon dispersion at large q in Si, using electron energy-loss spectroscopy with 250-kV electrons,[7] five times higher than that used by Stiebling and Raether, thus reducing multiple scattering considerably. This is the first measurement of plasmon dispersion at large q with such high-energy electrons. Our as-measured plasmon dispersion curve (prior to any data processing) shows that the plasmon peak is still dispersive at large q, in contrast to all the measurements that have been previously reported. At $q \sim 2.5$ Å^{-1} our measured plasmon energy is ≈ 26 eV which is more than 2 eV above what has been reported. Furthermore, our data can be easily reconciled within the framework of RPA.

In this experiment we have used two thin single-crystal silicon samples ($t \sim 1000$ Å) with [001] orientation prepared by two different methods. One was prepared by mechanical polishing followed by ion milling as in Ref. 6, and the other, which was boron doped, was prepared by chemical etching. Our measurements with these two samples show identical results. This rules out speculation that the dispersionless 23.5-eV peak observed in Ref. 6 might be an artifact of ion milling. A sample with [001] orientation was chosen in preference to the [011] orientation used in Ref. 6 so that the multiple scattering due to Bragg peaks can be minimized. This point will be discussed in detail later. We have measured plasmon dispersion for momentum transfer $\vec{q}$ along both the [100] and [110] directions. The samples were oriented by x-ray diffraction before being mounted in the electron-scattering spectrometer, whose energy and momentum-transfer resolution are 0.4 eV and 0.2 Å^{-1}, respectively. In Fig. 1, we show the electron energy-loss spectra for $0 \leq q \leq 2.55$ Å^{-1} in the [100] direction. At $q = 0$, a plasmon peak at 16.6 eV with width 3.3 eV is observed, and as q increases, the plasmon peak shifts to higher energies. At $q \sim 1.3$ Å$^{-1} \gtrsim q_c$, the double scattering (bulk plasmon plus TDS) peak begins to emerge from the real plasmon peak. The double-scattering peak (at ~ 16.7 eV) stays constant in energy as q increases as expected. The true plasmon peak disperses upward in energy and becomes broader as q increases. We also note that the intensity of the plasmon peak falls below the intensity of the plasmon-TDS peak for $q = 1.8$ Å^{-1}, clearly indicating that multiple scattering is much less serious in our work than in Ref. 6. Shown above the raw data for 2.45 and 2.55 Å^{-1} is an expanded view of the same data, after removal of a simple straight line background chosen to make the dispersed plasmon peak appear roughly symmetrical. While this presentation tends to lower the observed plasmon peak position somewhat, it shows that the dispersed plasmon peak is still well resolved. The shoulders located at lower energies as shown in Fig. 1 are most likely to be due to the excitation of direct nonvertical interband transitions which has been reported previously.[8]

Stiebling and Raether have argued that the triple processes (bulk plasmon–surface plasmon–thermal diffuse) and losses due to a thin SiO_2 film on the surface can be ruled out as cause for the ~ 23.5-eV peak observed at large q. Here we

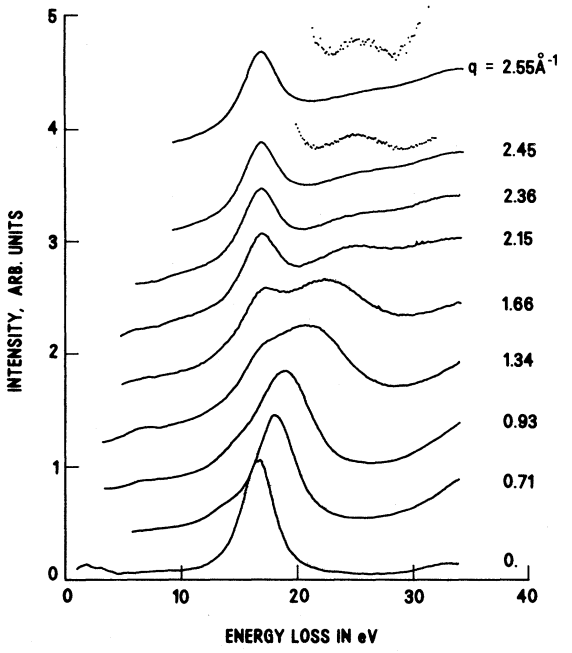

FIG. 1. Intensity vs energy loss for Si[100] (in arbitrary units). Shown above the raw data for 2.45 and 2.55 $Å^{-1}$ is an expanded view of the same data after removing a simple straight-line background.

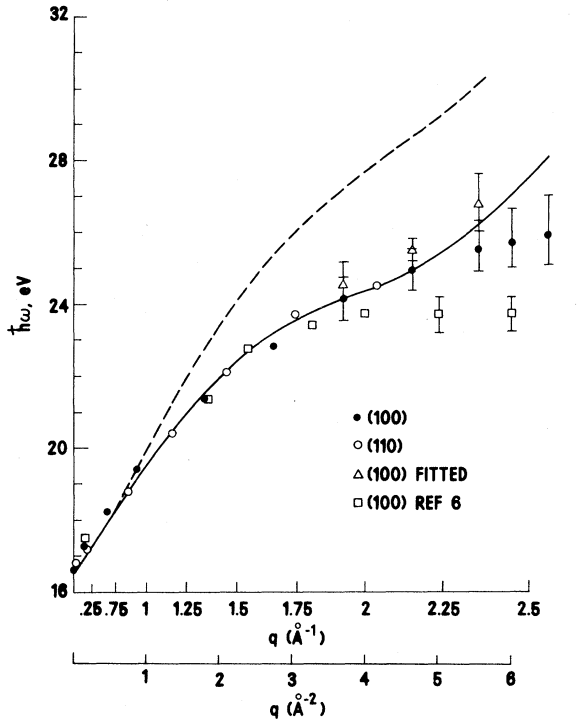

FIG. 2. Plasmon dispersion [$\hbar\omega_p(q)$ vs q^2] in silicon. Both [100] and [110] data points are included at small q. Dashed curve is RPA with Hubbard's exchange correction and solid curve is RPA with exchange correction 1.8 times larger than Hubbard's.

would like to suggest another possible channel of multiple scattering, involving Bragg reflection, that might cause the appearance of an extraneous peak at large q for the [011] orientation. If we measure the plasmon dispersion along [111], when q exceeds the zone boundary in that direction, plasmons excited from the (1$\bar{1}$1) Bragg peak dominate and obscure plasmons excited from the undiffracted beam. So, in general, the plasmon cannot be measured to large q in the [111] direction by electron energy-loss spectroscopy. Along the [100] direction, the (200) reflection is forbidden, so one can, in principle, go out quite far in q until one reaches the (400) reflection at $q = 4.64$ $Å^{-1}$. However, when [011] sample orientation is used, there are four Bragg peaks [two (111) and two (113)] which are close to the [100] symmetry line. As soon as q approaches the forbidden (200) reflection at ~ 2.3 $Å^{-1}$, these four Bragg spots are equidistant (~ 2 $Å^{-1}$) from it and the plasmons excited from those diffracted electrons become a major contribution of the intensity observed in the region $q \gtrsim 2$ $Å^{-1}$, producing a peak at ~ 23.5 eV, the energy at which Stiebling and Raether reported leveling off of the dispersion curve. We feel that this might be what has been observed in their work. In our experiment, using samples with [001] orientation, since the distance from (220) spots to the forbidden (200) spots is the same as that from (000) to the (200) spots, the contribution from the two (220) spots would become important for $q \cong 2.4$ $Å^{-1}$ in the [100] direction. However, even under this extreme condition, the plasmon energy loss will be the same because the momentum transfer is the same as that for plasmons excited to the (200) diffracted beam from the incident beam. Of course, when q becomes greater than ≈ 2.4 $Å^{-1}$, the effect of the (400) beams becomes more important, and any peak positions observed above this q value will appear lower than expected. Therefore, we conclude that [001] orientation is far superior to [011] orientation if we are concerned with measurement of the plasmon dispersion along [100] at large q. In addition, we have minimized the effects of scattering from the (220) and (400) Bragg reflections by tilting the crystal so that they are weakly excited.

In Fig. 2, we show the plasmon dispersion for q in the [100] and [110] directions. Peak posi-

tions were simply read from the data by hand. Within our experimental errors, we observe no difference in the plasmon dispersion along these two directions in the small-q region. The plasmon dispersion coefficient, α, obtained is 0.4 $\pm$ 0.02, in good agreement with the [100] results in Ref. 6. For comparison, data points for $q \parallel [100]$ from Ref. 6 are also shown in Fig. 2. It is clear that at large q the plasmon is still dispersive and our data points are at least 2 eV higher than those reported in Ref. 6.

We have also made attempts to fit our data with a simple RPA calculation including a constant lifetime[9] and an exchange correction. The lifetime was determined from our measured plasmon width at $q = 0$, ≈ 3.3 eV (full width at half maximum). At present, there exists no well-established theory to correct the dielectric function for exchange and correlation effects. Here we used the general expression

$$\epsilon(q,\omega) = 1 + (\epsilon_{\mathrm{RPA}} - 1)/[1 - G(q)(\epsilon_{\mathrm{RPA}} - 1)]$$

with $G(q) = q^2/2(q^2 + q_{\mathrm{F}}^2)$ as the exchange correction. The static form of $G(q)$, proposed by Hubbard,[10] was chosen for reasons of simplicity, although there exist in the literature many suggestions as to which is the best form for $G(q,\omega)$.[11-14] The calculated plasmon dispersion under these assumptions is also shown in Fig. 2. At small q the simple RPA results agree reasonably well with the measured results; however, at large q, the measured dispersion curve falls below the RPA predicted curve. At $q \approx 2.4$ Å^{-1}, RPA puts a point ~ 3.5 eV higher than our observed data. Of course, one can always choose $G(q)$ as a fitting parameter to obtain RPA results which would agree with the observed data. In case of Al, it was found[5] that a $G(q)$ twice as large as one proposed by Hubbard[9] had to be used in order to fit the experimental dispersion curve. In the present case, we also find that much better fit can be obtained if we used a $G(q)$ which is ~ 1.8 times larger than the one proposed by Hubbard,[8] as shown also in Fig. 2.

It is evident that multiple scattering still poses as a serious problem at large q even in our experiment. If multiple-scattering effects could be removed completely, it is expected that the data points at large q would move to higher energies. Since there is no simple way to deal with multiple scattering in this case, we have attempted to determine the peak positions by nonlinear least-squares fitting of Lorentzians. At most two Lorentzians were used to fit any one data set, and an eigenvalue-eigenvector analysis of the fitting results was done to determine fitting error estimates and the extent of parameter correlation. In all cases reported here the effects of correlation were essentially negligible. The fit residuals were essentially randomly distributed. The error bars shown in Fig. 2 include our estimate of systematic error and a perhaps overly conservative measure of the fitting error based on a doubling of the sum of squares of the fit residuals. As a result of this analysis, peak positions for the large-q data do not move to higher energies, as shown in Fig. 2. However, we are unable to fit the last two curves for $q = 2.45$ and 2.55 Å^{-1} with confidence because the plasmon peak is too broad and too weak compared to the background.

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