Aluminum bulk-plasmon dispersion and its anisotropy

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We present a high-resolution electron-energy-loss spectroscopy (EELS) study on the bulk-plasmon dispersion with respect to absolute value and orientation of the transferred momentum vector $\hbar q$ in an aluminum single crystal. The magnitude q ranges from $q=0$ Å^{-1} to $q=1.5$ Å^{-1}. Orientations are parallel to [100] and [110] crystal directions. The dispersion has been observed to be biquadratic in q, with unique parameters over the entire q range up to the cutoff wave number, in contrast to earlier studies. Substantial deviations from predictions within the random-phase approximation have been found. Furthermore, the anisotropy of the plasmon dispersion, now measured with high resolution, is considerably smaller than previously reported. The results are discussed in terms of exchange and correlation and band-structure effects.

In the last two decades a wealth of experimental and theoretical work has been performed on the momentum-dependent plasmon dispersion of Al (Refs. 1–9) as well as in other metals.[10] The general interest has been the investigation of the electronic structure of nearly-free-electron metals.

Experimental plasmon energies showed quite substantial deviations from theoretical predictions which were made on the basis of the Bohm-Pines random-phase approximation (RPA) applied to a free-electron gas. RPA yields

$$E_p(q)=\hbar\omega_p+\frac{\hbar^2}{m}\alpha_{\mathrm{RPA}}q^2+O(q^4)\ ,$$

where $\omega_p^2=ne^2/\epsilon_0 m$ and $\alpha_{\mathrm{RPA}}=\frac{3}{5}(E_F/\hbar\omega_p)$, E_F being the Fermi energy. It has turned out that the dispersion curves, plotted versus q^2, cannot be fitted by a straight line in the entire q range from $q=0$ Å^{-1} up to the critical wave vector q_c. This circumstance has led many authors to a description of their curves using two dispersion coefficients,[1,11,12] α_{low} (for low q values) and α_{high} (for higher q values up to q_c). Since α_{high} appeared in most cases to be close to α_{RPA}, it has been concluded[3] that it is the high-momentum region which ought to be regarded when studying the plasmon dispersion, and that the low-q region is affected by band-structure effects. This point of view was contested[13] and a controversial debate followed, but no final conclusion has so far been drawn.[11,14,15]

In our opinion, the interpretation in terms of two dispersion coefficients has to be doubted. Several investigations carried out by different authors resulted in quite different values for the crossover on the q scale. In Refs. 1, 8, 11, and 3, the crossover was situated at about 0.37, 0.50, 0.57, and 0.75 Å^{-1}, respectively. Hence, it seems likely that any discontinuity in the slope of the measured dispersion is an artifact.

We have investigated the plasmon dispersion of Al using a high-resolution electron-energy-loss spectroscopy (EELS) spectrometer.[16] We can now clearly state that the Al plasmon dispersion develops smoothly from $q=0$ Å^{-1} up to $q=q_c$. There is no indication of a crossover, but within the given error bars the data fit perfectly to the following function:

$$E_p(q)=E_p(0)+Aq^2+Bq^4\ . \tag{1}$$

Hence, it appears that the use of two dispersion coefficients is unphysical and misleading. It has been puzzling that α_{high} compares better with α_{RPA} than predictions from refined theories. The theory by Singwi[17] *et al.* (STLS) and Vashishta[18] *et al.* (VS) uses local-field correction functions to improve on the treatment of exchange and correlation in RPA. The general trend is a reduction in plasmon dispersion when exchange and correlation effects are taken into account. The puzzle may be resolved by assuming that the near equality of $\alpha_{\mathrm{high}}\approx\alpha_{\mathrm{RPA}}$ is pure coincidence without physical significance. We found a considerable reduction of the plasmon dispersion coefficient $\alpha=\alpha_{\mathrm{low}}$ compared to α_{RPA}, which is presumably caused by exchange and correlation effects.

A thin Al film (thickness about 0.1 μm) was grown epitaxially on the (100) surface of freshly cleaved rocksalt. The film was single crystalline with (100) orientation, and had a grain size >1 μm. The EELS spectra were taken on a 170-keV electron transmission spectrometer described in Ref. 16, with energy and momentum resolution set to 0.18 eV and 0.04 Å^{-1}, respectively.

The anisotropy measurements were carried through by rotating the sample keeping all other spectrometer settings fixed. The rotation was monitored by the diffraction pattern. Spectra were taken at various q values between $q=0$ and 1.5 Å^{-1}. The plasmon lines were almost Lorentzian, and the peak positions were determined from a least-squares fit to the measured intensities. The plasmon peak positions measured along the [100] direction are plotted as circles in Fig. 1(a) versus the squared momentum transfer. The lines in Fig. 1(a) represent calculations of the maximum of the energy-loss

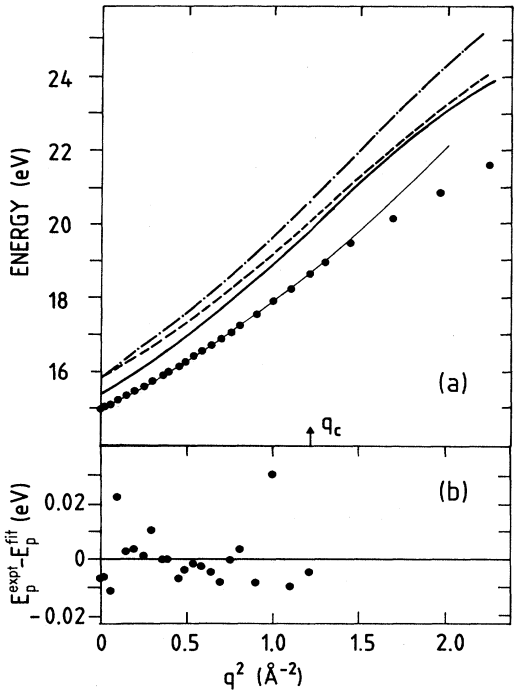

FIG. 1. (a) Measured plasmon dispersion parallel to [100] direction, compared to least-squares-fit curve and calculated predictions. Circles from measurements, thin line from fit, dashed-dotted line calculated from Lindhard-Mermin function, dashed line with inclusion of local-field correction, bold solid line with additional inclusion of core polarization. (b) Deviation of measured points from least-squares-fit curve.

function $\mathrm{Im}[-1/\epsilon(\mathbf{q},\omega)]$ within different models. The dielectric functions $\epsilon(\mathbf{q},\omega+i\Gamma)$ are the Lindhard-Mermin function[19] (dashed-dotted line), the same with inclusion of the local-field correction by VS (dashed line) and the latter after additional inclusion of a core polarization term (bold solid line), respectively. The damping parameter Γ was taken from the plasmon linewidth and the core polarization term from Ref. 5. A least-squares fit (thin solid line) using Eq. (1) to the experimental points yields $E_p(0)=15.01\pm0.01$ eV, $A=2.27\pm0.07$ eV Å^2, and $B=0.65\pm0.02$ eV Å^4. For this fit, only the data points below q_c were used. All these data points are within ±0.04 eV of the fit whereas 90% of the points are within ±0.01 eV [see Fig. 1(b)]. Plotted in an E_p-versus-q^2 diagram, our fit does not indicate any crossover point. Above $q_c=1.1$ Å^{-1}, the dispersion curve enters the band of single-particle excitations and rapidly flattens off as is well known from previous investigations.[12]

From our data we may still define an $\alpha=\alpha_{\mathrm{low}}$ from the initial slope $\alpha=0.30\pm0.01$. However, any choice of an α_{high} would be arbitrary. The value of 0.30 is much smaller than the RPA prediction ($\alpha_{\mathrm{RPA}}=0.44$), a discrepancy which is due to the neglection of short-range correlations and exchange in RPA. Taking account of these latter effects by use of static local field corrections[17,18] we obtain $\alpha_{\mathrm{STLS}}=0.28$ and $\alpha_{\mathrm{VS}}=0.35$, respectively, in better agreement with our experimental value.

Regarding the theoretical papers available up to now, it is not clear to what extent band structure influences plasmon dispersion. Figure 1(a) indicates that there is a nearly-q-independent downward shift of plasmon energy compared to the theoretical curves which can be explained by band-structure effects according to Sturm.[13] In other words, the coefficient α (low q dispersion coefficient) is not strongly depending on band structure. As a test for these calculations,[13,20] measurements on the anisotropy of plasmon dispersion are presented in Fig. 2. The data scatter considerably as is indicated by the error bars. The reason for this is the steep plasmon dispersion together with minuscule shifts in apparatus settings. For instance, a 1% momentum misfit at 1 Å^{-1} will provoke a 0.07-eV plasmon energy error, obscuring seriously the measurement. The statistical uncertainties for the energy dispersion measurement of Fig. 1 are far below 0.07 eV. This is due to the rapid data collection as well as the high apparatus stability.

One would have expected anisotropy values larger by a factor of 2 or 3 from earlier measurements[6,7] which, on the other hand, suffer from extremely large error limits. While a theoretical calculation by Bross[21,22] was in closer accordance with those data,[6,7] a calculation by Sturm,[20] based on pseudopotential theory, compares quite favorably with the very small anisotropy we found (see solid line in Fig. 2). The agreement of Sturm's calculations with our experimental data indicates also that Sturm's prediction of a negligible influence of band-structure effects on the long-wavelength dispersion coefficient α is correct. The change in sign of the anisotropy near 1.2 Å^{-1} is questionable. Sturm[23] points out that a lower plasmon dispersion, as resulting from exchange and correlation corrections, would shift the zero crossing to higher q values. The measured anisotropy tends to zero for q above q_c: On entering the above-q_c regime the spec-

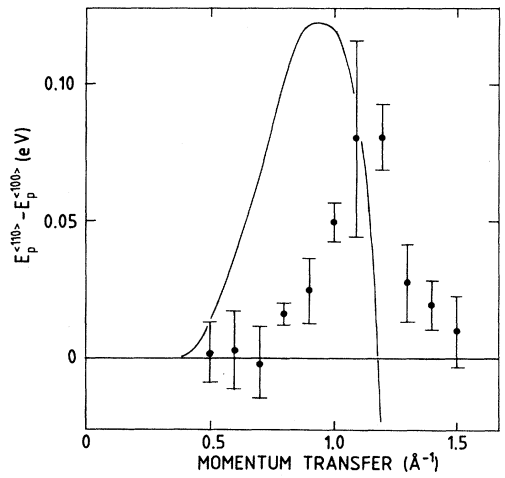

FIG. 2. Anisotropy of Al bulk-plasmon dispersion. Difference between plasmon peak energies measured with momentum transfer parallel to [110] and [100] directions, respectively. The solid line is a theoretical calculation from Ref. 20.

tral line called "bulk plasmon" loses more and more its collective character hybridizing with the single particle excitations. What is measured is the structure factor of the particle-hole continuum, showing a plasmon dispersion which is not only weak but isotropic.

In summary we have presented measurements on the aluminum bulk plasmon performed with high energy and momentum resolution. Unlike earlier authors, we have found the following: (i) There is no discontinuity in the dispersion curve slope. (ii) A function $E_p(0)+Aq^2+Bq^4$ perfectly fits the dispersion curve within narrow error limits. (iii) The anisotropy of plasmon dispersion is very small, barely exceeding 100 meV. (iv) Exchange and correlation effects reduce the dispersion coefficient compared to the RPA value.

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