Direct nonvertical interband transitions at large wave vectors in aluminum*

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We report electron-energy-loss measurements of the direct nonvertical interband transition in aluminum at an incident electron energy of 75 keV identifying a loss at 1.5 eV at a scattering vector $q = 0$ and an initial linear dispersion. The present experiments demonstrate that the loss continues to exist for $q > 1.0$ Å^{-1} and that the dispersion levels off at a value of ~ 8.8 eV at $q = 1.56$ Å^{-1}. A comparison with recent calculations is given.

Recently, Petri and Otto[1] reported electron-energy-loss spectra of single-crystal aluminum, in which a sharp dispersive peak lying below the bulk plasmon peak was identified as due to the excitation of direct nonvertical interband transitions. Specifically, for momentum transfer $\vec{q}$ in the [100] direction, a linearly dispersive peak which starts at 1.5 eV for $q \simeq 0$ and goes up to ~7.2 eV for $q \sim 1$ Å^{-1} was observed. The measured data, however, terminated at $q \simeq 1$ Å^{-1}. Beyond that, no well-defined peak was observed. Since this peak is sitting on a fast-rising diffuse background, it is not clear from the previous measurement whether the peak at large q is lost in the broad diffuse background or is simply nonexistent. Detailed measurements in the region of large momentum transfer ($q > 1$ Å^{-1}) should be valuable in understanding the nature of this peak. At small q, the predominant transitions come from states at the Fermi level. However, at large q, it is quite likely that the transitions could start from states well below the Fermi level and become a significant contribution to the scattering cross section. Furthermore, measurement at large q might also pose a crucial test of the existing theory. Here we report the electron-energy-loss spectra extended to larger q values of single-crystal aluminum with [001] orientation. The sharp dispersive peak was found to exist in the region of $q > 1$ Å^{-1}, with approximately the same peak width and intensity as in the smaller q region.

Single-crystal aluminum films of [001] orientation were obtained from a bulk crystal by chemical polishing. Incident electrons of 75-keV kinetic energy were used with experimental procedures described in detail elsewhere.[2] Energy-loss spectra for $\vec{q}$ in the [100] and [110] direction were obtained and recorded on photographic plates. Data were obtained subsequently by microdensitometry.

Figure 1 shows the microdensitometer traces of the loss spectra for $q = 0.5$ and 1.04 Å^{-1} in the [100] direction. The sharp peaks at 4.4 eV ($q = 0.5$ Å^{-1}) and 7.3 eV ($q = 1.04$ Å^{-1}) are attributed to the direct nonvertical interband transitions, and the broad diffuse peaks at higher energies are due to intraband transitions as suggested by Petri and Otto.[1] It is most interesting that the width (~2 eV) and the intensity of the interband peak stay relatively constant over the entire q region of interest, in contrast with bulk plasmons at large q, in which the width increases and the intensity decreases rapidly according to $1/q^2$ approximately.[3] These are the most unusual features of the peak. Our measured width is significantly narrower than that reported in the previous measurement (~3 eV).[1] This might indicate that the single-crystal films (thinned chemically from bulk crystals) we use seem to have higher quality than the epitaxial films used in the previous experiment. Experimental data for q in the [110] direction, on the other hand, showed no sharp peaks and only broad diffuse peaks were observed. This is consistent with the

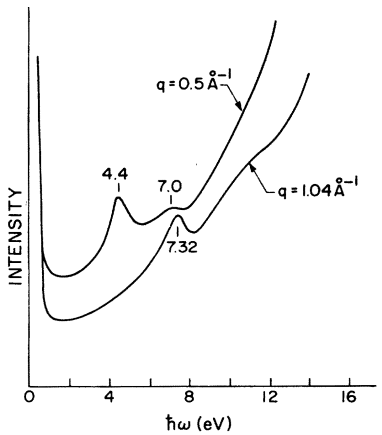

FIG. 1. Microdensitometer traces of the energy-loss spectra in single-crystal Al of [001] orientation with $q = 0.5$ and 1.04 Å^{-1} in the [100] direction.

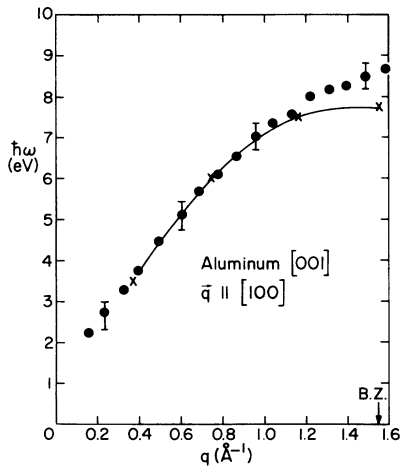

FIG. 2. Dispersion of direct nonvertical interband transitions in the [100] direction. Experimental data are represented by dots, and the theoretical results by crosses.

observations of Petri and Otto.[1]

Complete data for the dispersion of the nonvertical interband peak is shown in Fig. 2. Our data for $q<1$ Å^{-1} agrees very well with Petri and Otto's data. In this region, the dispersion is approximately linear. For $q>1$ Å^{-1}, our new data show that the dispersion deviates from the linear relationship, and tends to level off at ~8.8 eV for $q=1.56$ Å^{-1} (zone boundary in the [100] direction). This suggests that the transitions start from states below the Fermi level. In Fig. 3, we plot the peak intensity of the interband peak as a function of q. Relatively uniform intensity of the peak was observed for $q \gtrsim 0.4$ Å^{-1}. This variation is clearly slower than $1/q^2$. The wave-vector and frequency-dependent energy-loss function $\mathrm{Im}[-1/\epsilon(q,\omega)]$ for aluminum has recently been calculated with the inclusion of energy band structure and exchange-correlation effects.[4] Sharp peaks were reproduced in the calculated energy-loss function only for q in the [100] direction. The width of the peak is ~1.8 eV and is relatively constant with respect to q, which is in good agreement with our observation. The calculated dispersion also agrees remarkably well with our experimental data for $q \lesssim 1.2$ Å^{-1} (Fig. 2). For $q \gtrsim 1.2$ Å^{-1}, the calculated points fall below the experimental curve, and at the zone boundary ($q=1.56$ Å^{-1}), the difference between the theory and the present measurement is ~1 eV. As we mentioned earlier, one of the most intriguing features of the peak is its intensity variation with q as shown in Fig. 3. The uniformity of the intensity for $q \gtrsim 0.4$ Å^{-1} indicates that the peak height of the energy-loss function should grow quadratically in q since the energy loss cross section is proportional to $(1/q^2)$ Im $[-1/\epsilon(q,\omega)]$. The calculated curve for Im $[-1/\epsilon(q,\omega)]$, indeed, does show that the peak height of the energy-loss function grows with q, but, however, at a rate more or less linearly rather than quadratically. Thus the theory is still inadequate to explain this very interesting intensity variation behavior. Calculations done on a more finely divided mesh in the Brillouin zone, perhaps, could improve the agreement between theory and the experiment. Although the peak positions were found to be not significantly dependent on the form of exchange-correlation employed,[4] the peak intensity at large q is still quite sensitive to the forms of exchange-correlation corrections. Therefore, a better treatment of exchange-correlation corrections might also improve the agreement between theory and experiment. Since the transition matrix elements were found to be weakly dependent on q our data suggests that the joint density of states of the transition should increase quadratically in q.

Although sharp peaks were obtained in the theoretical calculation it, however, gave no further detailed discussion as to the relationship of this peak to the energy band structures. The physical origin of this direct nonvertical interband peak was first proposed by Petri and Otto.[1] It was suggested that the nonvertical interband transition arises from energy bands in the X-W-X' direction[1] on the square face of the Brillouin zone. On the weak pseudopotential approximation, energy bands of Al within the square face are nearly parallel,[5] and separated by an amount $2U_{200}=1.5$ eV. The linear portion of the dispersion curve ($q \lesssim 1$ Å^{-1}), can be interpreted as transitions in-

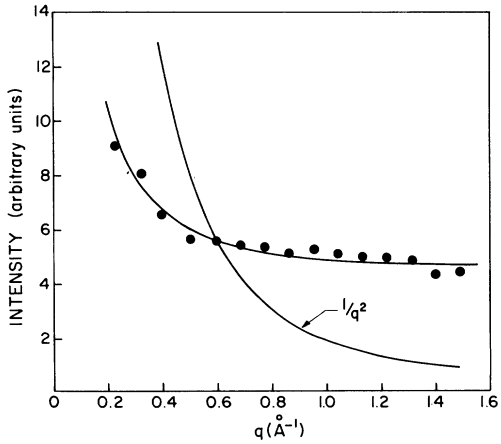

FIG. 3. Wave-vector dependence of the peak intensity of the nonvertical transition peak. The $1/q^2$ curve normalized to the intensity of experimental data at $q=0.6$ Å^{-1} is also shown.

volving electrons at the Fermi level as suggested previously.[1]

At large q (>1 Å^{-1}), transitions would originate from states well below the Fermi level and it has been pointed out to us by Ashcroft that the transition energy from X to X' ($q = 1.56$ Å^{-1}) is ~9 eV which agrees very nicely with our data. In addition, the parallel band model will involve a high density of states for nonvertical transitions with q lying in the square face of the Brillouin zone.[6] The reason for the discrepancy between our experimental value of 8.8 eV and the value of 7.8 eV found by the detailed calculation[4] is not clear.

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[1]E. Petri and A. Otto, Phys. Rev. Lett. 34, 1283 (1975).

[2]G. H. Curtis and J. Silcox, Rev. Sci. Instrum. 42, 630 (1971); C. H. Chen *et al.*, Phys. Rev. Lett. 36, 525 (1976).

[3]P. E. Batson, Ph.D. thesis (Cornell University, 1976) (unpublished).

[4]S. P. Singhal, Phys. Rev. B 14, 2352 (1976).

[5]N. W. Ashcroft and K. Sturm, Phys. Rev. B 3, 1898 (1971).

[6]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart, and Winston, New York, 1976), p. 303.