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SEMICONDUCTOR BAND STRUCTURE INFORMATION FROM ANGULAR DEPENDENT ELECTRON ENERGY LOSS MEASUREMENTS

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The use of electron energy loss spectroscopy as a probe of band structure is discussed and in particular the ability of this technique to determine energy band shapes which are unobtainable with the more commonly used optical methods is examined. Experimental results for CdI_2 are compared with calculated joint density of states functions.

I. Introduction

In transmission electron energy loss spectroscopy of semiconductors, momentum $\hbar\underline{q}$ as well as energy $\hbar\omega$ can be transferred by the probing electrons. Non-vertical interband transitions can occur and are related to the energy loss probability $P(\underline{q},\omega)$ via the dielectric function $\varepsilon(\underline{q},\omega)$ [1]:

$$P(\underline{q},\omega) = A \frac{1}{q^2} \mathrm{Im}\{\frac{-1}{\varepsilon(\underline{q},\omega)}\} \quad ,$$

where A is independent of $\underline{q}$ and ω but is determined by the particular experimental conditions. $\varepsilon(\underline{q},\omega)$ can thus in principle be determined from the angular dependent electron energy loss probability by Kramers-Kronig analysing the data. Because the energy loss experiment samples the entire Brillouin zone, it is difficult to make exact assignments of structure in $\varepsilon(\underline{q},\omega)$ to energy band shapes, and a discussion of this problem and a method for interpreting $\underline{q}$-dependent results is presented and illustrated for CdI_2.

II. Joint Density of States

The information available about band shapes is illustrated for one dimension in Fig.(1). Peaks will occur at $(\underline{q},\omega)$ in $\varepsilon_2 \equiv \mathrm{Im}\{\varepsilon\}$ if there are large areas of k-space separated by $\underline{q}$ and ω. Such areas are indicated by arrows in Fig.(1) for a fixed value of $\underline{q}$, with the conduction band shifted by $-\underline{q}$ in Figure (1b) for clarity. Several authors have interpreted shifts with $\underline{q}$ of peak positions in ε_2 by referring to the calculated band structures along symmetry directions. Such an interpretation can be seen to be a coarse approximation even if the band structure is taken to be two dimensional. Possible transitions for a particular value of $\underline{q}$ are shown in Fig.(2) for the two dimensional hexagonal Brillouin zone, where the arrows link the positions in the repeated zone scheme of the initial and

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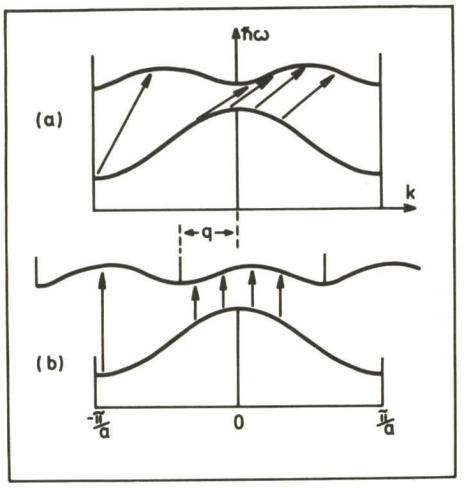
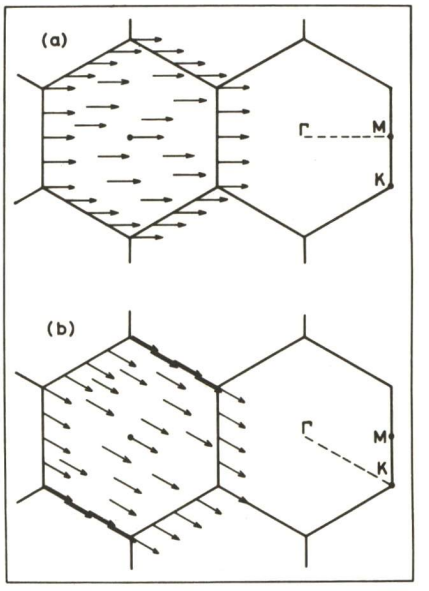


Fig. 1
Hypothetical band structure showing the most probable transition energies for a fixed value of $\underline{q}$

Fig. 2
Non-vertical transitions in the two dimensional Brillouin zone

final states. Reference to Fig.(2) shows that it is only for a few lines in the Brillouin zone that the one dimensional argument illustrated in Fig.(1) is justified.

To make an exact comparison between experiment and theory, it is thus necessary to calculate for each $\underline{q}$ a joint density of states which is proportional to



the number of valence and conduction states separated by $\underline{q}$ and ω. For a semiconductor or insulator, this function can be defined by

$$J(\underline{q},\omega) \propto \sum_{\ell\ell'} \int d\underline{k}\ \delta\{E'_{\ell'}(\underline{k}+\underline{q}) - E_{\ell}(\underline{k}) - \hbar\omega\},$$

where the sum is over all occupied bands ℓ and all empty bands ℓ'. Structure in the experimentally determined curves of $\varepsilon_2(\underline{q},\omega)$ simply reflects that in $J(\underline{q},\omega)$.

As an illustration of the effects expected, $J(\underline{q},\omega)$ was determined numerically for the 2-D Brillouin zone. The quantity $[E'(\underline{k}+\underline{q})-E(\underline{k})]$ was calculated at 2.6×10^4 points in the Brillouin zone for $\underline{q}$ in the two directions shown in Fig.(2) and using two energy bands $\hbar\omega \propto \cos(\pi k/\Gamma K)$ where $k = |\underline{k}|$. In each case one of the bands was 1 eV wide and the other 2 eV wide but similar results were obtained with a band-width ratio of 1:0.75. The results shown in Figs.(3a-d) reveal that the relative curvatures of the bands and the magnitude of $\underline{q}$ rather than the direction of $\underline{q}$ have the greatest effect on J.

These results can be used to find average band shapes from experimentally determined curves of $\varepsilon(\underline{q},\omega)$. Comparison of experiment with these results may, in many cases, yield more information about real band shapes than would comparison with $J(\underline{q},\omega)$ calculated in detail from theoretical band structures, especially for more complex materials with closely spaced energy bands.

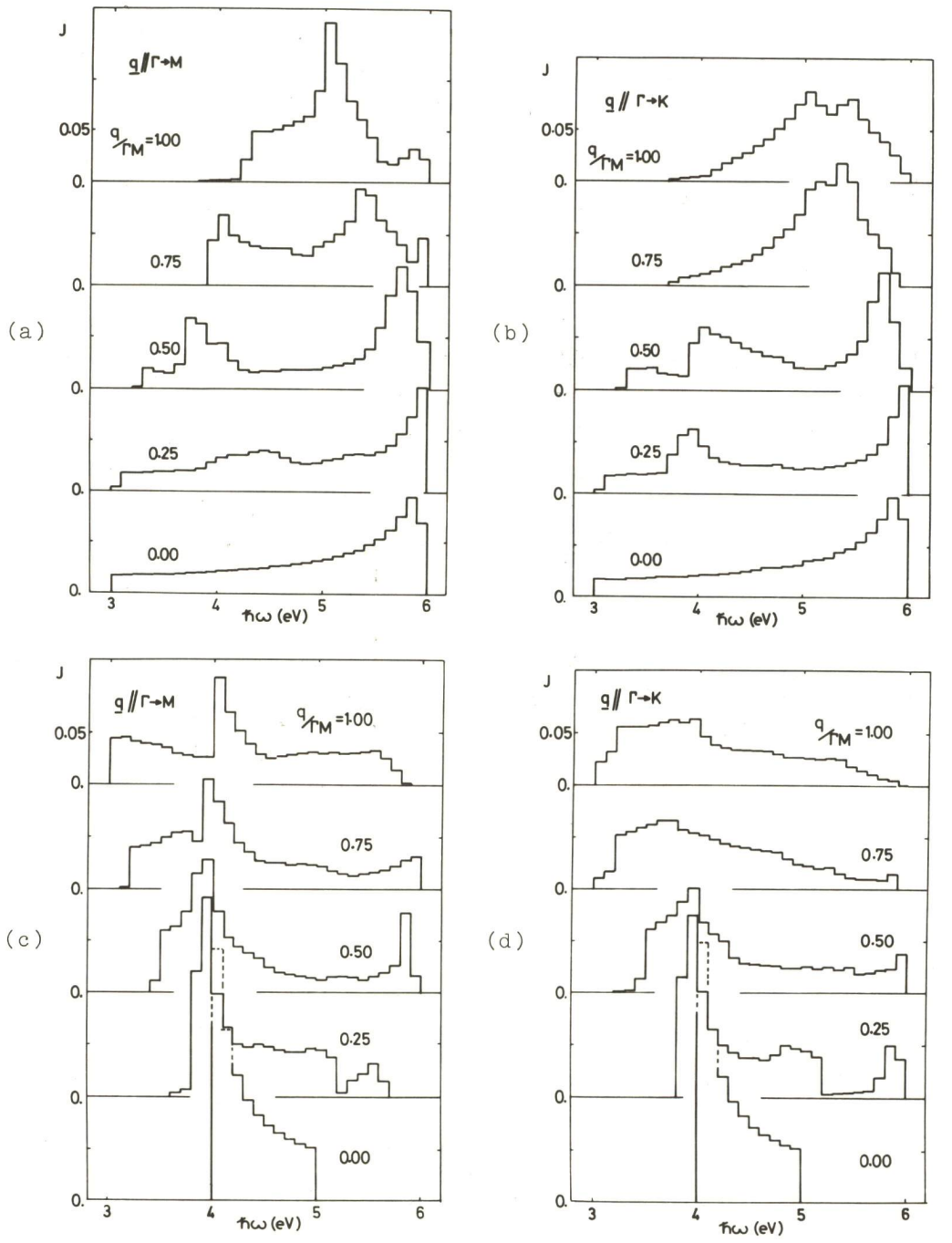


Fig. 3

J(q,ω) for the 2-D hexagonal Brillouin zone: two bands with opposite curvature and minimum separation at Γ for (a) $\underline{q}||\Gamma\vec{M}$ and (b) $\underline{q}||\Gamma\vec{K}$; two bands with parallel curvature and maximum separation at $\bar{\Gamma}$ for (c) $\underline{q}||\Gamma\vec{M}$ and (d) $\underline{q}||\Gamma\vec{K}$

III. Results for CdI_2

Curves of $\varepsilon_2(q,\omega)$ derived from electron energy loss measurements of CdI_2 are shown in Fig.(4). Thin polycrystalline films of this layered material were used and these were found to be highly oriented with the basal planes of the crystallites parallel to the substrate. The energy loss experiment could thus be carried out in such a way that $\underline{q}$ was in the plane of the layers. The calculated energy bands for CdI_2 (see, for example, [2]) are much more complicated than those from which $J(\underline{q},\omega)$ in Fig.(3) were derived, however the same arguments can be applied to estimate the coarse shape of the energy bands. The peaks in $\varepsilon_2(q,\omega)$ have been assigned elsewhere [3] to particular interband transitions using energy loss results and photoemission data [4]. The assignments are shown in the schematic

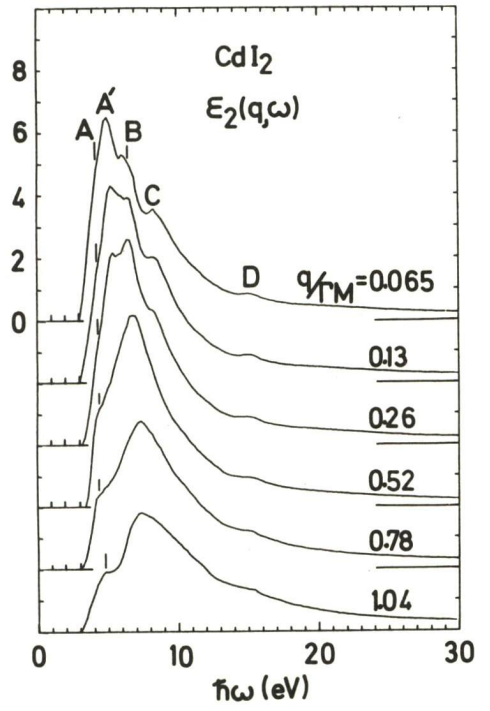


Fig. 4

$\varepsilon_2(q,\omega)$ determined for CdI_2 from electron energy loss measurements

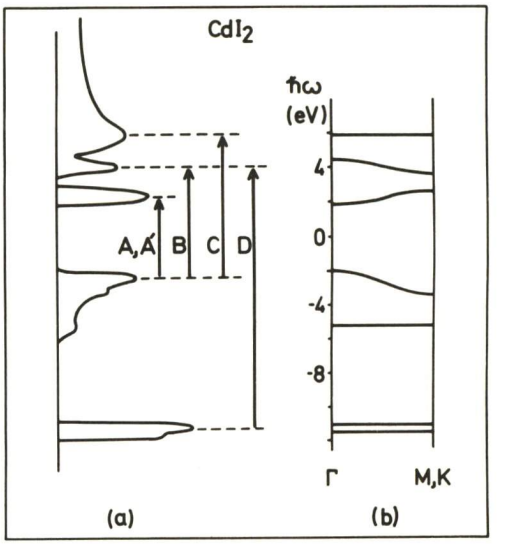


Fig. 5

Schematic density of states (a) and band curvatures (b) for CdI_2

density of states diagram Fig.(5a). The behaviour with q of peaks A and A', namely: A' disappearing, A becoming stronger and moving to higher energies, is very similar to that seen in Fig.(3a) and (3b) and suggests that the first conduction band has an opposite curvature to the top of the valence band and comes closest to it at Γ. The feature B moves to higher energies and broadens as q increases, suggesting that the bands involved have parallel curvature with minimum separation at Γ. Features C and D show no dispersion indicating that both the third conduction band density of states maximum and the Cd d level are dispersionless. These band shapes are shown schematically in Fig.(5b).

References

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