Comment on the line shape of the plasma resonance in simple metals*

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The line width of the long-wavelength plasma resonance in the alkali metals, when corrected for instrumental broadening, is found to bear a simple relation to the magnitude of the smallest-wave-vector Fourier component of the lattice potential.

A number of studies, both experimental[1,2] and theoretical,[3,4] of the width of the plasma resonance in simple metals have been carried out. All of these, however, have concentrated on the momentum-transfer dependence of the width. This problem still remains a mystery, at least as far as the small momentum increase in width is concerned. We wish to point out that another aspect of this problem, namely the width measured at zero momentum transfer, bears a simple relation to the magnitude of the nearest zone-boundary band gap.

Figure 1 shows the width of the plasma resonance in the forward-scattering characteristic energy-loss spectra of the alkalis,[2,5-8] plotted versus the square of the energy gap $N_1 - N_1'$. The band gaps used in the figure are derived, where possible, primarily from de Haas–van Alphen effect measurements. Theoretical calculations and optical measurements are also used.[9-15] The values used are given in Table I. The widths have been observed to be independent of temperature from 150 to 300 K in sodium, potassium, and also in aluminum, in our laboratory. Sample quality is also unimportant for all crystallite sizes greater than 100 Å. This result has been known for many years,[16] and has been confirmed in our measurements.

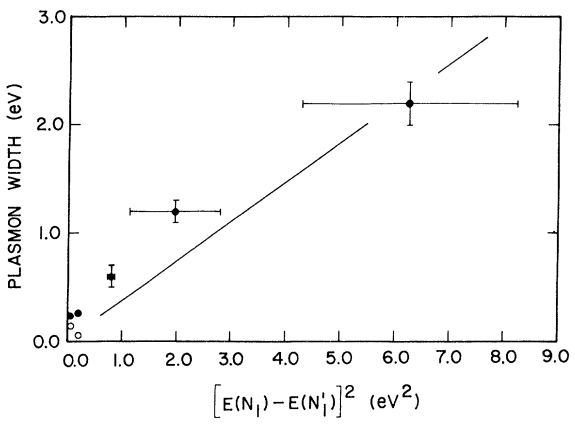

FIG. 1. Full width at half maximum of the plasma resonance plotted vs the square of the energy gap at the nearest zone boundary. From left to right, the materials are K, Na, Rb, Cs, and Li. The data come from unpublished work of the authors and from Refs. 2 and 5–8. The dots are data corrected only for instrumental resolution. The open circles for K and Na are also corrected for grain boundary scattering as discussed in Ref. 17. The band gaps come from Refs. 9–15. The slope of the line sketched in is 0.37 eV^{-1} with an estimated uncertainty of 0.05 eV^{-1}.

As a plasmon can be thought of as a superposition of pair excitations, its damping might depend on an average of electron scattering taken over the entire conduction band. The band gap involves only states near the zone boundary at which the gap occurs. Whence the simple relation shown in Fig. 1?

Qualitatively, the behavior can be understood as follows. The plasmon width is proportional to the magnitude of the imaginary part of the dielectric response function evaluated at ω_p, $\epsilon_2(\omega_p)$. Both intraband (Drude) and interband transitions contribute to $\epsilon_2(\omega_p)$. The relative importance of the two can be determined using optical data. In potassium, for example, the plasmon width is 0.21 eV full width at half maximum (instrumental broadening of 0.060 eV removed). The prediction derived from optical data at the plasma frequency is 0.2 eV,[8] while the prediction derived from infrared data is 0.02 eV.[8,16] The optical measurements were made at room temperature. Thus interband transitions are the dominant broadening mechanism. The magnitude of ϵ_2 due to interband transitions is proportional to the square of an interband

TABLE I. The band gaps at the zone boundary nearest the origin in the alkali metals. The values were determined as discussed in the text, using results from Refs. 9–15.

Material	$E(N_1) - E(N_1')$ (eV)
Li	2.5 ± 0.4
Na	0.45 ± 0.03
K	−0.16 ± 0.01
Rb	−0.90 ± 0.03
Cs	−1.4 ± 0.3

matrix element, while the zone-boundary band gaps are linear in the same matrix element. That only one band gap is important is due to the fact that the others all occur only in those interband transitions that are at energies above ω_p, and so cannot contribute to the decay of the plasmon.[18] This mechanism for plasmon damping is temperature independent and does not assume any specific degree of sample quality, as the data require.

The main conclusion from this observed correlation is that band-structure effects dominate the zero-momentum-transfer plasmon width, and that the variation of observed widths among the alkalis is as expected. We suggest that future work on the momentum transfer dependence of the plasmon width include the effects of band structure, which have been ignored to date.

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