ions are, indeed, heated during the period of low-frequency oscillation, leading us to think that the heating is caused by the ion acoustic wave, as mentioned by many authors.[6-9]

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COLLECTIVE EFFECTS IN THE OPTICAL PROPERTIES OF Cs ABOVE THE PLASMA FREQUENCY[*]

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Optical and dielectric constants of Cs have been obtained above the plasma frequency by measuring reflectance as a function of angle at a Cs-substrate interface. We find a strong, broad peak centered at 5 eV in the conductivity σ. The peak is attributed primarily to plasmon-assisted transitions, though direct transitions may contribute to the overall absorption in this energy region.

There has been much recent interest in the optical properties of the alkali metals. Much of the work, both theoretical[1-3] and experimental,[4-6] has been devoted to the lighter metals, Na and K, and to the energy region below the plasma frequency. Several recent theoretical studies suggest that excitation of collective modes makes an important contribution to the optical absorption above the plasma frequency.[1-3] Sutherland, Hamm, and Arakawa[7] reported structure in the optical constants of Na and K that is consistent with these predictions but does not distinguish between the absorption process suggested by Hopfield[1] as opposed to those of Lundqvist and Lydén[2] and Janow and Tzoar.[3]

We report here measurements of the optical and dielectric constants of Cs at photon energies above the plasma energy.

Experimental methods.—Reflectance measurements were made at the interface of Cs films evaporated onto quartz and CaF$_2$ substrates with light incident through the substrate. The techniques and equipment are described elsewhere.[7,8] Cesium (99.7% purity) was evaporated onto cooled substrates (10°C) in a 10^{-6} Torr vacuum, and thereafter maintained at 3×10^{-7} Torr. Films about 1 μm thick were evaporated in typical times of 10 sec. Since oxidation proceeds from the vacuum side of the cesium film, measuring times of many hours were available before oxidation of the film penetrated to the interface.

The reflectance was measured as a function of angle of incidence, and the optical constants derived from the behavior of the reflectance near

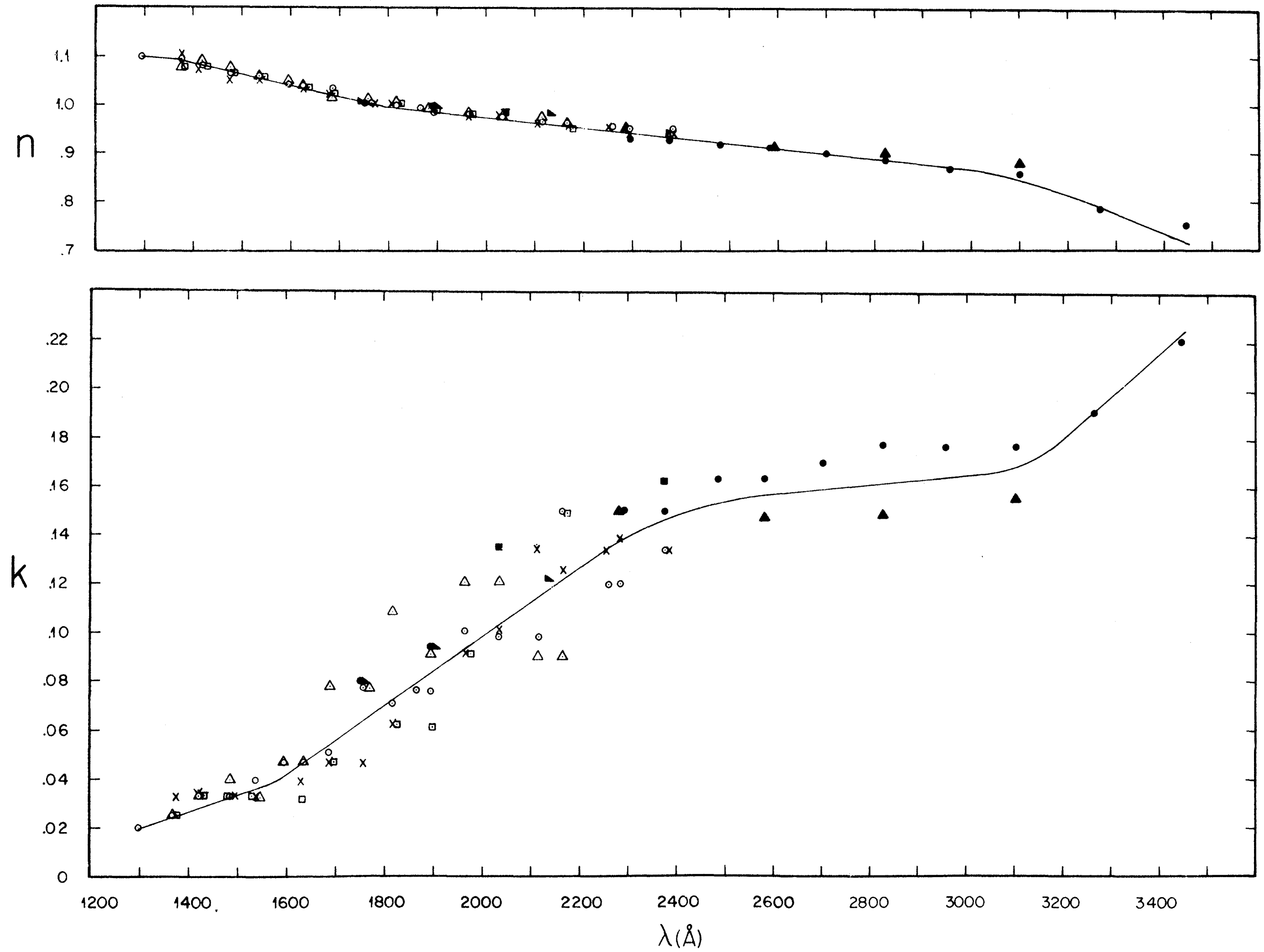

FIG. 1. Experimentally determined n and k values for Cs plotted against photon wavelength. Film A, open circles; film B, open triangles; film C, open squares; film D, crosses; film E, closed circles; film F, closed triangles; film G, closed squares; and film H, closed right triangles. Solid lines give the smoothed average values.

the angle for total internal reflection. When $n \simeq 1$ and $k \lesssim 0.05$, the real part of the complex refractive index n is determined by finding the angular position of the critical angle for total internal reflection and making a small correction for finite k, the imaginary part of the refractive index. The value of k is calculated from the slope of the reflectance curve at the critical angle, which has only a weak dependence on n.[9]

Experimental results.—Measurements with a CaF_2 substrate extended from 5 to 9.6 eV and averaged results from four films were used in the analysis. Maximum deviations from n_{av} ranged from <2% at 9.6 to 4% at 5 eV. Values of k showed scatter of about ±12% about the average value. The scatter in k is large with the CaF_2 substrates because surface contamination distorted the shape of the reflectivity curves above the critical angle and made accurate determination of the slope difficult. Measurements were made with rigorously cleaned quartz substrates from 3.6 to 6 eV. These measurements gave reflectance curves as a function of incident angle which were a nearly perfect fit to Fresnel's equations. Somewhat less scatter was obtained in the values of k measured on these films. Data points and smoothed curves of n and k used in the subsequent analysis are shown in Fig. 1.

The real part of the dielectric constant ϵ_1 and the optical conductivity σ may be obtained from n and k values by the relations $\epsilon_1 = n^2 - k^2$ and $\sigma = \omega\epsilon_2/4\pi = \omega nk/2\pi$. Since $k \ll n$, the value of ϵ_1 is obtained with the accuracy of n^2. Percentage uncertainties in σ are approximately the same as in k.

Discussion.—Values of ϵ_1 show the qualitative behavior expected for a nearly free-electron model with relatively small (15%) corrections due to other absorption mechanisms. A future paper will discuss these data in some detail.

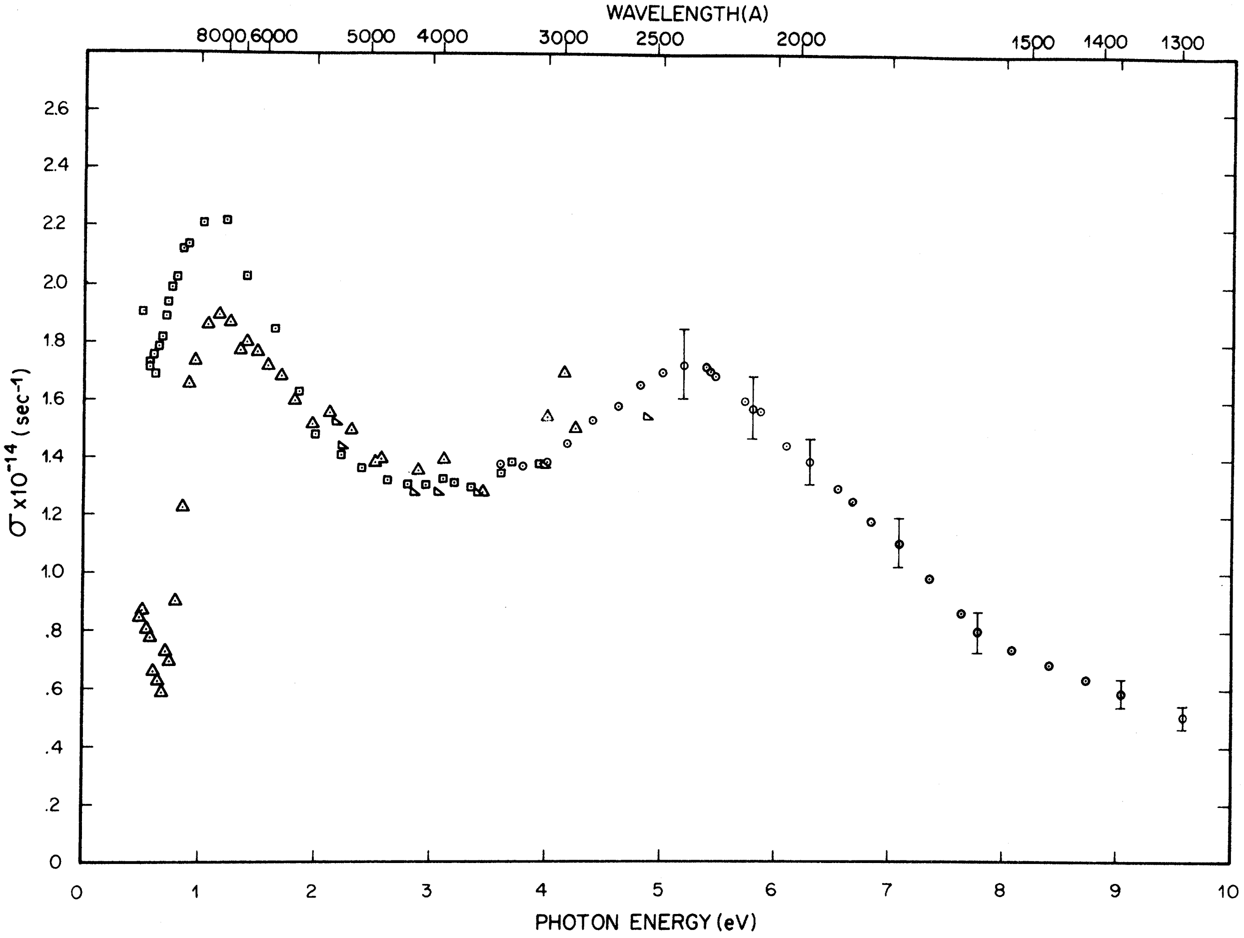

FIG. 2. Optical conductivity σ versus photon energy. Present data, open circles; Smith, open squares; Mayer and Hietel, open triangles; and Ives and Briggs, open right triangles.

The absorption effects responsible for the deviation in ϵ_1 are most easily discussed in terms of σ, which is a direct measure of the optical energy absorption of the material. Our values of σ are plotted in Fig. 2 along with lower-energy data of Ives and Briggs,[4] Smith,[10] and Mayer and Hietel.[5] The low-energy peak (<1 to 3 eV) is generally attributed to an interband transition and is discussed by Butcher[11] and Meessen.[12] The broad peak with threshold at 3.5 to 4 eV and maximum at 5 eV is the new feature we report here, though its existence may be surmised from earlier lower-energy data. It is of particular interest because its strong features are not predicted by calculations of absorption by direct transitions in simple alkali metals. Its explanation must be given in terms of either peculiarities in the band structure of Cs or absorption processes involving collective excitations.

Band structures of Cs have been calculated by Ham[13] and Kenney,[14] the latter extending 8 eV above the conduction-band minimum. No optical-absorption calculations based on "these" or similarly detailed calculations have been reported.

Mixed bands of s, p, and d character occupy the region 2 to 4 eV above the conduction-band states and are too low in energy to account for the observed absorption though they may account for part of the absorption in the region between the two peaks. A prominent set of "f bands" lies 5 to 8 eV above the conduction-band states and lies too high in energy to account for the observed peak. In addition, transitions to such states are thought to be weak. It appears that in the absence of serious errors in the band-structure calculation the absorption cannot be explained as an interband transition.

Several authors[1-3] have described absorption mechanisms involving collective effects that may provide absorption processes comparable in strength with interband transitions in the alkali metals where interband absorption is weak. Hopfield[1] shows that there should be an increase in free-carrier absorption as one passes through the plasma frequency due to a reduction in the effectiveness of screening of the lattice ions. Such a mechanism should have a threshold at $\hbar\omega_p$ and reach a peak at slightly higher energy. For Cs the threshold would be at $\hbar\omega_p \simeq 2.9$ eV and the peak no higher than 4 eV. For this reason and because free-carrier absorption is very weak at 5 eV compared with other absorption mechanisms, this explanation may be ruled out. Lundqvist and Lydén[2] and Janow and Tzoar[3] calculate absorption with a threshold at $\hbar\omega_p + \hbar\omega_G$, where ω_G is the threshold for interband transitions. The final state of this process is a plasmon and an electron-hole pair. Consequently, we would expect the related absorption to have a maximum near $\hbar\omega_p$ plus the energy of the maximum of the interband absorption peak. The observed absorption seems to fit this description very well with a threshold between 3.5 and 4 eV as compared with $\hbar\omega_p + \hbar\omega_G = 3.5$ to 3.8 eV, depending on the value taken for ω_G. The peak at 5 eV is very close to the plasmon energy plus the energy of the interband absorption peak. Lundqvist and Lydén predict a magnitude for the absorption as measured by the value of σ of 30 to 50% of the interband peak for Na. For Cs we observe relative values closer to 75%. If the interpretation in terms of plasmon-assisted transitions proves to be correct, this magnitude of the absorption and the fact that this absorption is larger in Cs than in Na will have to be explained. It is interesting to speculate that the absorption becomes stronger for alkali metals of higher atomic number. We are currently measuring Rb to see if such a trend does in fact exist.

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