

The Optical Properties of Metallic Aluminum*

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GENERAL FEATURES I

The optical properties of metallic aluminum are among the most widely measured and analyzed of any material [1–7]. They are dominated by three practically nonoverlapping groups of electronic transitions corresponding to absorptions by conduction band, L-shell, and K-shell electrons. The absorption spectrum for the crystalline solid is shown in Fig. 11.

The 3 e/at. (electrons/atom) donated to the conduction band by the atomic $3s^23p$ valence levels give rise to a typical metallic absorption from zero to ~ 15 eV, the bulk plasmon frequency ω_p . In crystalline samples this portion of the spectrum is dominated by intraband transitions in the far infrared and by two strong interband absorptions at ~ 0.5 eV ($\lambda \approx 2.5 \mu\text{m}$) and ~ 1.5 eV ($\lambda \approx 0.8 \mu\text{m}$). In the noncrystalline or partially crystalline solid state and in the liquid state, the interband spectrum is strongly modified or absent; this effect will be discussed further in Subsection II.C.

Intraband Spectrum A

The intraband contribution to the optical properties can be described phenomenologically to within experimental error with a Drude-model dielectric

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function [8, 9] for the electron gas:

$$\varepsilon_{\text{Drude}} = \varepsilon_0 - [\Omega_p^2 / \omega(\omega + i/\tau)], \quad (1)$$

where the contribution to the dielectric function from core interband transitions has been included in ε_0 . Here Ω_p is the (phenomenological) plasma frequency for intraband transitions and τ the intraband relaxation time. The quantity ε_0 is effectively constant throughout the region of conduction-electron absorption (that is, to well above the plasma frequency) with a value [7] of $1.03\varepsilon_s$.

In addition to using the quantity Ω_p , the strength of the intraband transitions is commonly expressed in terms of the effective number density of electrons participating in intraband transitions

$$n_{\text{eff, intraband}} = m_e \Omega_p^2 / 4\pi e^2. \quad (2)$$

Here m_e and e are the free-electron mass and charge, respectively. A second alternative is the optical mass defined by [8, 9]

$$m_{\text{opt}} = m_e (n_c / n_{\text{eff, intraband}}), \quad (3)$$

where n_c is the density of conduction electrons.

Similarly, the intraband relaxation time is often given in terms of the damping coefficient

$$\gamma = \tau^{-1}. \quad (4)$$

A quantity often cited for comparison with electrical experiments is the dc conductivity in the Drude model

$$\sigma(0) = \Omega_p^2 / 4\pi\gamma. \quad (5)$$

A variety of fits of Eq. (1) have been made to infrared data for polycrystalline samples of metallic aluminum [1, 3, 5, 10–15], but early work was incomplete because the interband transitions at ~ 0.5 and ~ 1.5 eV were not always reckoned with. Some of the more recent determinations of the Drude parameters and the experimental bulk dc conductivity are given in Table XI. The “0.5-eV” interband transition was not accounted for by Ehrenreich *et al.* [1] or by Powell [3], so that some interband absorption strength, as well as damping, is included in their Drude terms. This leads to a conductivity significantly smaller than the measured bulk value. Bennett and Bennett [12] assumed the measured value of $\sigma(0)$ but also neglected the 0.5 eV transition leading to an overestimate of Ω_p . Dresselhaus *et al.* [13], Mathewson and Myers [5], and Benbow and Lynch [14] accounted for both interband transitions, the last two studies with the Ashcroft–Sturm [18] model of the interband spectrum. The values reported by Smith and Segall [15] were derived by fitting the dielectric function of Shiles *et al.* [7] over the range of 0.04 to 3.0 eV, assuming that it is a linear superposition of a Drude term and the two principal interband transitions. No detailed assumptions as to the form of the interband spectrum were made. The resulting parameters thus yield

the least model-dependent description of the room-temperature interband absorption presently available.

Interband Spectrum **B**

The two strong interband transitions of the conduction-electron spectrum in the crystalline material are a consequence of the “parallel-band” effect [1, 18–20] that occurs in almost free-electron polyvalent metals. In these materials the occupied and unoccupied conduction bands are effectively parallel over substantial regions of k space in the vicinity of high-symmetry planes parallel to the zone faces. Allowed transitions between these parallel bands lead to prominent interband absorptions at energies approximately twice the Fourier component of the effective crystal potential $V_{\mathbf{k}}$ for the reciprocal lattice vector $\mathbf{K}$ corresponding to the face in question. In aluminum, absorption between almost parallel bands occurs in the neighborhood of surfaces parallel to the hexagonal (111) and the square (200) zone faces. The corresponding energy gaps predicted by a two-band model [18] using parameters derived by Ashcroft [21] from de Haas–van Alphen measurements below 4 K are $2V_{111} = 0.487$ eV and $2V_{200} = 1.53$ eV. The parallel-band absorption shows sharp rises near these energies followed by long tails toward higher energies that joins smoothly onto the band-to-band absorption arising from the remainder of the Brillouin zone [18, 20]. In addition, theory predicts [22, 23] an accidental degeneracy of the energy bands leading to a weak but finite interband absorption down to zero energy. However, this has not been verified experimentally because the interband component is overwhelmed by the intraband absorption below a few tenths of an electron volt.

The 1.5-eV (0.8- μm) interband absorption is readily apparent as a pronounced drop in the reflectance for crystalline samples (Fig. 12) and as a small peak in the extinction coefficient (Fig. 11). The former appears to have been first reported by Strong [24] and the latter by Schulz [25]. The absorption responsible for these features was identified as a transition between pairs of parallel bands by Ehrenreich *et al.* [1]. Shklyarevskii and Yarovaya [26] independently identified the absorption as an interband transition but did not recognize the crucial importance of energy-band parallelism near the Fermi energy.

The second major interband absorption at ~ 0.5 eV is almost completely hidden by the Drude absorption. There is a suggestion of it in the early infrared measurements of Beattie [27] and in the ultrahigh vacuum (uhv) reflectance data of Bennett *et al.* that show a small drop in reflectance near 25 μm . (see Bennett *et al.* [28] Fig. 3). This transition was first definitively observed in low-temperature absorptivity experiments by Bos and Lynch [29]. Figure 13 shows subsequent results of Benbow and Lynch [14] for single-crystal aluminum at 4.2 K together with Smith and Segall's [15] separation of room-temperature data for uhv evaporated films. The results are

presented in terms of the real part of the interband conductivity

$$\sigma_{\text{interband}}(\omega) = (-i\omega/4\pi)[\varepsilon(\omega) - \varepsilon_{\text{Drude}}(\omega)]. \quad (6)$$

Below approximately 0.3 eV the uncertainty in the experimental data and in the Drude parameters is so large that the interband contribution cannot be determined reliably from the available measurements.

Note the shift of the interband absorption toward higher energies as the temperature is lowered. Mathewson and Myers [5] studied this temperature dependence in detail for the “1.5-eV” transition (see also Fig. 17).

At low temperatures (~ 20 K) a weak fine structure is superimposed on the tail of the 1.5-eV absorption in the range 2–2.5 eV [30]. Some of these structures appear to arise [31] from transitions near the corners of the Brillouin zone in the vicinity of the intersections of the two groups of planes responsible for the two strong parallel-band absorptions. Along these lines of intersection three bands are almost parallel with band gaps determined by both V_{200} and V_{111} . However, the transitions arising from this “trio” of parallel bands along the intersections are not as prominent as those associated with the “duos” of parallel bands in the planes because of the smaller momentum space involved in the neighborhood of the lines of intersection. The result is two weak absorptions, one near the strong 0.5-eV interband transition and the second just above the strong 1.5-eV transition. These have been predicted in detail by band-structure calculations [23, 31]. (See also Hunderi and Nilsson [30].)

Further interband fine structure has been resolved in recent piezo-modulation experiments by Jiles and Staines [31a]. This includes strain-induced changes around 2–2.5 eV and 4.6 eV, in addition to structure near the 1.5 eV parallel-band transition.

The possibility that excitons may be associated with the interband absorption edges has been investigated theoretically [32]. While excitonic states are predicted, their binding energies are found to be only 5 (singlet) and 20 (triplet) meV as a result of the highly effective screening of the coulomb interaction by the conduction electrons. Such structure is unobservable in conventional optical experiments but may contribute to the broadening of the interband peaks.

II OPTICAL MEASUREMENTS AND SAMPLE CONDITIONS

A Polished Polycrystalline Samples

Serious studies of the optical properties of aluminum were begun in 1874 by Quincke [33]. Early studies employed polished bulk or rolled samples [33–40]. Even though bulk aluminum takes a “high polish,” it always retains

a “hazy white surface” [36] and has a reflectance well below that of an uncontaminated evaporated film. As will become apparent, this is presumably the result of both scattering and absorption by the microscopically rough, oxide-coated surface. A composite of reflectance data for polished surfaces is given in Fig. 12. The curve generally follows the data of Coblenz [36] (IR), Luckiesh [38] (visible + UV), Quincke [33] (visible), and Wulff [40] (visible + UV). The polished bulk-metal data are strongly affected by surface contamination and light scattering, particularly at shorter wavelengths. In the visible, Nutting [35] reports reflectance values $\sim 10\%$ higher than those shown, while values measured by Coblenz and Stair [39] are 10% lower. An even more dramatic range is given by Taylor [41], who quotes reflectances from 33 (mill finish) to 84% (electropolished) at 296.7 nm .

Thin-Film Polycrystalline Samples **B**

The key factor in obtaining accurate values of the optical properties was the discovery that aluminum could be sputtered [42] or evaporated [43] to form highly reflecting metal films. Early sputtered films were described as “brilliant” [44, 45]. However, evaporation by the tungsten-coil method [46, 47] has become the preferred method of sample preparation because of its high speed and potential for low contamination. The major impetus for these developments was the production of interferometer and telescope mirrors [24]. (The superior reflectance of evaporated aluminum films over silver at short wavelengths extended stellar spectroscopy some 250 Å into the violet.)

Evaporated films show reflectances much higher than polished surfaces particularly in the ultraviolet. Generally, the higher the vacuum and the faster the evaporation, the higher is the reflectance [48–51]. Two curves for evaporated surfaces are shown in Fig. 12. The upper is for unoxidized, smooth films produced in ultrahigh vacuum (10^{-9} – 10^{-10} torr), and the other is for films evaporated in conventional high vacuum ($\sim 10^{-5}$ torr). The first is believed to correspond to a smooth, uncontaminated aluminum surface. The second is for more-or-less contaminated surfaces having oxide coating with no special attempts made to eliminate surface roughness. The uhv curve generally follows the data of Bennett *et al.* [28], Hass and Waylonis [51] (though not uhv films, a high evaporation rate was employed that has been shown to yield the same results as an uhv preparation [52]), and Endriz and Spicer [53]. Beyond 11.8 eV the curve follows the analysis of Shiles *et al.* [7], which relies heavily on electron-energy-loss measurements [54] near ω_p and on transmission and reflectance data of Ditchburn and Freeman [55] and x-ray absorption measurements[†] beyond ω_p .

The high-vacuum curve generally follows the data of Beattie [11], Strong [24], Hass [48, 56], Sabine [57], Banning [58], and Walker *et al.* [59]. The

[†] For a list of sources used for x-ray cross sections, see Shiles *et al.* [7].

curve shown is in close agreement from 250 to 2500 nm with the U.S. National Bureau of Standards' reference standard first-surface aluminum mirror # 2003a [60], which has an aged oxide coating. The curve also generally has the same reflectance from 200 to 2000 nm as commercially available evaporated mirrors.[†] Films evaporated under higher vacuum or at high deposition rates generally have reflectances between the high-vacuum and uhv curves shown. (For references to additional measurements see Shiles *et al.* [7] and the sources cited in Schopper [61].)

The differences between the various evaporated film data arises from surface oxide layers [48, 62–66], residual gas incorporated in the film [49, 50], surface roughness [53, 67, 68], and film morphology [52, 69–71].

1 Surface Oxide Layers

A layer of Al_2O_3 forms rapidly on an aluminum surface even in high vacuum [72, 73]. The layer is both adherent and relatively impervious so that once formed it protects the underlying metal from further oxidation [74]. The thickness of the surface layer and its composition depend strongly on sample structure and history, particularly on chemical treatment [24, 52, 69, 71, 74, 75]. Polarimetric studies of evaporated films simply exposed to the atmosphere indicate that the surface oxide layer is from 20 to 55 Å thick [50, 62–66] but that thicker layers readily form in moist environments [74]. The surface layers are probably hydrated to varying degrees depending on formation conditions. Preparation conditions [48, 52, 76], purity [76], structure [69, 71], partial oxidation in vacuum before exposure to the atmosphere [65], etc.; all influence the reported thickness. In addition, exposure of aluminum surfaces in air to ultraviolet light has been found to decrease their reflectance at short wavelengths; this is attributable to an increase in the oxidation rate of the surface [48, 69, 76, 77].

The oxide of aluminum is highly transparent from approximately 6 μm (0.2 eV) [78] to 180 nm (6.8 eV) [6, 79], so that a surface oxide film does not decrease the reflectance significantly over this range. Moreover, in the infrared a light wave does not “see” the film [48]. (The incident and reflected electric waves must be out of phase by approximately π to create the required node in the electric field at the metal surface. Thus, provided that the oxide thickness is much less than the wavelength of light, the electric fields of the incident and reflected waves almost cancel in the oxide layer and coupling to the layer is negligible.) However, at the shorter wavelengths of the vacuum ultraviolet, the oxide layer has a strong effect. For example, at 121.6 nm

[†] The authors are indebted to Professor Wayne Major of the University of Richmond for providing measurements for a commercially supplied, aluminum-film ($\sim 1000\text{-}\text{\AA}$) reflector on optical glass prepared by D. and E. Technology, Santa Clara, California. Evaporation was performed at $\sim 10^{-6}$ torr.

(~ 10 eV) an oxide layer $17 \text{ \AA}$ thick decreases the reflectance by a factor of two [63, 76].

Considerable research has been devoted to developing thin-film dielectric coatings to preserve or enhance the optical qualities of aluminum front-surface mirrors. For many applications, the natural oxide film is either too thin to offer the required protection from chemical change and mechanical damage—particularly when frequent cleaning is needed—or too absorbing for use in the ultraviolet. Coatings of the silicon oxides and Al_2O_3 that offer a high degree of mechanical and chemical protection have been developed [48, 80, 81] for use in the IR, visible, and near UV. At high angles of incidence, silicon oxide coatings show a reduced reflectance in the infrared. For such applications, protective coatings of Y_2O_3 and HfO_2 have been found to be highly successful [82]. At shorter wavelengths, overcoatings of MgF_2 and LiF preserve the high reflectance of aluminum down to $\sim 1000 \text{ \AA}$ by preventing surface oxidation [48, 83]. Multiple dielectric layer coatings to enhance reflectance over a limited wavelength range have also been developed; enhancement of a 90% reflector to 99% reflectance have been reported [84].

Bulk Inclusion of Residual Gas 2

Studies of films evaporated at different rates and pressures indicate that the optical constants are very sensitive to residual gas incorporated into the bulk of the films [49, 50], specifically to the film composition within the penetration depth [50]. This is generally attributed [48–50, 85] to oxide formation arising from the gettering of molecular oxygen or to the reduction of water vapor to form Al_2O_3 and hydrogen. The relevant parameter appears to be the ratio of the residual gas pressure to the deposition rate, which is a measure of the arrival rate of residual gas molecules to that of aluminum atoms [49, 50]. Films deposited from high-purity starting material at terminal pressure-to-rate ratios of the order 10^{-8} – 10^{-9} torr min $\text{\AA}^{-1}$ or less [50] have similar reflectances and appear to be uncontaminated for the purpose of optical property measurements, provided that subsequent oxide formation is avoided.

The presence of surface effects has also been demonstrated in x-ray absorption measurements in the neighborhood of the L edge [86, 87]. This has not been investigated in detail, but it has been associated with an apparent systematic overestimate of the L-shell absorption [7].

Surface Roughness and Morphology 3

An extremely important factor in determining effective optical properties, particularly in the ultraviolet, is the roughness of the sample surfaces [53, 67, 68]. Surface roughness causes both scattering [88] and coupling to surface

plasmons [89] (the latter lie at an energy of $\hbar\omega_p/\sqrt{2} \approx 10.6$ eV for an aluminum-vacuum interface). Reflectance curves for two low-contamination films [53, 90, 91] with slight surface roughness are shown in Fig. 12 for comparison with the smooth, uhv surface reflectance. The main reflectance drop lies near the surface-plasmon frequency, but the drop extends well into the visible, largely as a result of scattering. Even a slight surface roughness produces dramatic effects. For example, a film with an rms surface roughness of only 27 Å is reported [53] to exhibit a reflectance drop of 20% at 3 eV (400 nm). These effects account for the wide variation of reflectance found in the UV and often previously attributed to contamination.

Film morphology, particularly grain size and orientation, has a substantial effect on optical properties. This is particularly apparent in aluminum films deposited by evaporation at oblique angles of incidence [92–94]. The optical properties of these films are anisotropic and the films develop diffusely reflecting surfaces with increasing thickness as a result of an increasingly coarse microstructure [80]. During the initial stages of deposition, films prepared in good vacuum systems appear to be compact and to have relatively small grain size, regardless of deposition angle; but beyond a critical film thickness diffuse scattering becomes substantial. For example, aluminum films 300 to 400 Å thick show surface roughness (as measured by scattering in the visible) for deposition angles of 60° or more. Holland [92] showed that the critical film thickness for diffuse reflection decreases sharply as the angle of incidence increases. These properties result from the preferential growth of crystallites in the form of columns parallel to the vapor beam [95] that are “self-shadowing.” The amount and nature of the residual gas in the evaporating system have a major effect on the microstructure and, hence, the optical properties both because of gettering [96] and scattering of the metal vapor that alters the angle of incidence on the substrate [94].

C Noncrystalline and Liquid Samples

Vapor-quenched solid and liquid aluminum exhibit typical metallic properties but with a wide range of modifications in the interband spectrum. In the liquid state, aluminum exhibits no interband structure [97]. Rather, the conduction-electron absorption appears to follow a free-electron Drude model to within experimental error. This is in agreement with the expectation that in the liquid state there is no long-range order and, hence, no well-defined band structure nor Brillouin-zone boundaries [98].

Similar properties might be anticipated for samples with a low degree of crystallinity. However, the optical spectrum of vapor-quenched aluminum films deposited at low temperature exhibits a complex behavior [5, 99]. Films deposited at 25, 140, and 198 K show a progressive reduction in the intensity

of the 1.5-eV interband absorption with decreasing substrate temperature. Essentially no 1.5-eV absorption remains in films deposited at 25 K [99]. The reduced 1.5-eV absorption does not show a shift in position or broadening, only a decrease in magnitude. Further, annealing of the films at room temperature restores the 1.5-eV absorption. These observations are taken [5] to indicate that deposition from the vapor on a cooled substrate yields a two-phase system that is partly crystalline and partly amorphous but that crystallizes on annealing.

This interpretation cannot be complete since it is not consistent with analysis [99] of the 25-K films. These films show no 1.5-eV interband component, and consequently they would be considered to be noncrystalline in this interpretation. However, their optical properties are not described by the Drude model as those of the liquid can be. Rather, the conduction-electron absorption appears to consist of a Drude term plus a residual absorption near 0.5 eV that is attributed to interband transitions characteristic of a $2V_{111}$ energy gap. A possible explanation [99] is that the film consists of small-diameter, close-packed planar clusters with characteristic (111) translational symmetry but lacking the (200) translational symmetry of bulk aluminum.

TABULATED DATA III

Room-Temperature Optical Constants A

The most reliable optical data presently available are for uncontaminated polycrystalline aluminum films prepared in uhv. Measurements generally are made in reflectance at room temperature (18–25°C) and are presently available over the range 0.04 eV (32 μm) to 11.8 eV (105 nm) [28, 53]. In addition, a number of ellipsometric studies [5, 65, 71, 100, 101] of thin films in the near IR and visible have recently become available, particularly through the studies of Mathewson and Myers [5, 100] and of Liljenvall *et al.* [101]. (The former also includes high- and low-temperature measurements.) Studies [14, 29] of electropolished polycrystalline and single-crystal samples have also been reported, but absolute optical properties were not measured. The results are, however, consistent with absolute measurements on thin films.

Ellipsometric measurements yield the optical constants directly, but reflectance measurements at normal incidence must be analyzed by using dispersion theory. This has recently been done over a wide spectral range for the room-temperature uhv reflectance measurements by Shiles *et al.* [7],

who used a self-consistent Kramers–Kronig analysis. This involved combining the reflectance measurements below 11.8 eV with (1) electron-energy-loss measurements in the vicinity of the plasma frequency and (2) transmission and angular-variation-of-reflectance measurements in the extreme UV and x-ray regions. The energy-loss measurements were mainly those of Gibbons *et al.* [54]. The higher-energy optical data were taken primarily from Ditchburn and Freeman [55], Haensel *et al.* [102], Gähwiler and Brown [103], Fomichev *et al.* [104–106], Singer [107], Ershov *et al.* [108, 109], Cooke and Stewardson [110], Bearden [111], Henke *et al.* [112, 113], Singman [114], Lublin *et al.* [115], Hubbel *et al.* [116], and Davisson [117]. These additional data were for room temperature but were not for uhv samples and may suffer from surface effects to some degree, particularly in the transmission measurements on thin films near the $L_{II,III}$ edge [86, 87] and to a lesser degree between the plasmon energy and the $L_{II,III}$ edge [104] (see later in this section).

Since the Kramers–Kronig relations require knowledge of one optical function over the entire spectrum, the analysis proceeded by successive approximations. A trial reflectance function was constructed by employing estimates from transmission, electron-energy-loss, and similar data above 11.8 eV. This was then analyzed and the resulting optical functions compared with the input. The trial function was successively modified—primarily by the substitution of experimental data where available—until the calculated and measured optical functions agreed.

Throughout, the results were checked against a number of optical sum rules to ensure agreement with independent theoretical and experimental constraints (see Chapter 3 of this volume). This disclosed that the reported oscillator strength in the L-absorption region was consistently too high by $\sim 14\%$. The absorption coefficient in this region has generally been measured in transmission by using thin films that exhibit a surface component to their absorption [86, 87]. To compensate for this, an *ad hoc* reduction was made in the data based on the thin-film measurements of Lukirskii *et al.* [118], Fomichev and Lukirskii [104, 105], Haensel *et al.* [102], and Gähwiler and Brown [103]. This brought the calculated absorption coefficient into agreement with both the f sum rule and later measurements by Balzarotti *et al.* [87], who separated surface and bulk absorption. While the data of Balzarotti *et al.* [87] extend only 10 eV above the L edge, the $k(\omega)$ values are consistently some 15% below those used by Shiles *et al.* [7] in preparing their original input in this range. This is in agreement with the conclusion that previous measurements of the absorption cross section in the neighborhood of the $L_{II,III}$ edge were systematically too large.

While this correction is very important in the soft x-ray region of the spectrum, it was found to have negligible effect on the optical constants below the plasma frequency as determined by the Kramers–Kronig analysis.

The results of the analysis for the refractive index and the reflectance between the infrared and the $L_{II,III}$ edge are listed in Table XII. Given for

comparison are the *in situ* uhv ellipsometric measurements of Mathewson and Myers [100], the uhv reflectance data of Bennett *et al.* [28], and the vacuum ultraviolet transmission and reflectance-angular-variation measurements of Ditchburn and Freeman [55]. The index results are graphed in Fig. 14. It will be seen that the Kramers-Kronig analysis is in good agreement with the ellipsometric measurements of Mathewson and Myers [100] in the visible and the IR. The differences—less than 11% in $n(\omega)$ and 8% or less in $k(\omega)$ —give an indication of the uncertainty of the data.

A large number of optical constant measurements [10, 11, 25–27, 51, 55, 56, 65, 66, 73, 75, 91, 119–129] have been reported for films prepared on normal substrates in high vacuum and subsequently exposed to the air. The resulting “effective” optical constants describe good but contaminated aluminum surfaces made with no special attempt to produce ultrasmooth films. A number of these measurements are compared with values derived from uhv films in Fig. 14. Over most of the spectrum the effective optical constants for the oxidized surface lie below those for the uhv films. This is most pronounced in the IR, where effective optical constants 25% or more smaller than the uhv values are common. In the visible the effective constants are generally 10 to 15% lower than the uhv values, while in the UV the effective constants are surprisingly close to the uhv values. The exception is for the effective $n(\omega)$ near the surface plasmon frequency ω_s . Here there is a “bump” in the effective index, presumably arising from dispersion associated with the apparent “absorption” of the surface plasma and light scattering by films with rough surfaces.

Reflectance at Normal Incidence **B**

The reflectance for smooth unoxidized films at normal incidence as calculated from the results of Shiles *et al.* [7] by using the Fresnel relation

$$R(\omega) = \frac{[n(\omega) - 1]^2 + k^2(\omega)}{[n(\omega) + 1]^2 + k^2(\omega)}, \quad (7)$$

is given in Table XII and Fig. 15. Also given in Table XII are the direct reflectance measurements of Bennett *et al.* [28] and the reflectance calculated from the ellipsometric measurements of Mathewson and Myers [100]. These data cover the infrared and visible and apply to films prepared in uhv. Values of the reflectance in the vacuum ultraviolet are also given; these were taken from the work of Ditchburn and Freeman [55] on films evaporated in high vacuum and are likely to suffer from surface-roughness and surface-oxide effects. (See Fomichev and Lukirskii [104].)

The last column of Table XII gives a composite set of reflectance data for the more common high-quality evaporated aluminum mirrors that are prepared in conventional high vacuum (10^{-5} – 10^{-6} torr) and subsequently

exposed to the atmosphere. These same data are plotted as the curve marked high-vacuum films in air in Fig. 12. The properties of such films vary widely depending on formation conditions, atmospheric exposure history, and substrate surface roughness. The composite given is for films formed by rapid evaporation in high vacuum on optically polished (but not supersmooth) substrate and subsequently exposed to clean dry air. Such films generally show reflections $\sim 1\%$ lower than smooth oxide-free films in the infrared and visible. In the ultraviolet, their reflectance drops rapidly from the smooth oxide-free film value as a result of scattering and surface-plasmon excitation. A wide range of reflectance is reported at these wavelengths because surface roughness is generally not controlled.

C Room-Temperature Dielectric Function

The dielectric function covering the complete energy range involved in the Kramers–Kronig analysis is given in Table XIII and Fig. 16. This set of data includes the 14% reduction in the high-energy side of the $L_{II,III}$ edge needed to obtain agreement with the f sum rule and recent transmission measurements [87] taking surface effects into account. The energy intervals used in the table have been chosen to give closely spaced points in regions of pronounced structure such as interband absorption and x-ray edges. Intermediate points can be found by interpolation noting that the dielectric function generally has a power-law dependence on energy in regions far removed from interband edges. Near the edges the energy spacing is too coarse to show all the x-ray fine structure. For details of this the reader is referred to Balzarotti *et al.* [87] and Haensel *et al.* [130] for L-edge structure and to Kiyono *et al.* [131] for K-edge spectra.

The $\epsilon_2(\omega)$ curve in Fig. 16 discloses unexpected discontinuities in slope: a small discontinuity at approximately 7 eV and a larger discontinuity near 15 eV. These are not readily apparent in the other optical functions, specifically in $n(\omega)$ and $k(\omega)$. These features may be extraneous, arising from the joining of data from different sources. This is most probably the case for the discontinuity near 7 eV when additional uncertainty was introduced from reading published graphic data.

However, the situation is not as clear for the larger discontinuity near 15 eV (the plasmon energy). Here the ϵ_2 data fall off significantly faster below 15 eV than in the interval between 15 eV and the $L_{II,III}$ edge. This is contrary to the simple expectation, based on single-particle excitations, that at photon energies high compared to the principal interband transition energies, the valence electrons should behave almost as free electrons so that their contribution to ϵ_2 should be approximately Drude-like; i.e., $\epsilon_2(\omega) \sim \omega^{-3}$. A possible explanation is that the change in slope is an experimental artifact: below 15 eV, the $\epsilon_2(\omega)$ data are based primarily on samples prepared in

uhv, whereas from 15 eV to the $L_{II,III}$ edge, the samples were prepared in conventional vacua and the measurements were made in air. Over the latter range, surface contamination could lead to significant errors in the optical constants reported by Ditchburn and Freeman [55]. Such an error cannot be detected as a violation of the f sum rule to within the accuracy of present data, since the contribution to the f sum rule over the region in question is of the order of a few tenths of an electron per atom.

A second and more intriguing possibility is that there is a deviation of the optical properties from the random-phase-approximation values above the plasmon energy. Just such an effect has been proposed by Hopfield [132] as a result of the dynamic screening of the phonon—or disorder—contribution to the optical absorption. A third possibility [133] is the excitation of bulk plasmons by oscillating charges induced at the metal surface by the exciting light; however, studies [134] of the surface photoelectric effect suggest the latter process is negligible. New measurements on well-characterized uhv samples in this energy range are needed to clarify this situation. Such experiments should be possible with the increasing availability of synchrotron radiation sources. Recent inelastic electron-scattering studies by Livins *et al.* [134a] confirm a deviation from Drude behavior near the plasmon energy that is consistent with Hopfield's suggestion.

Temperature Dependence **D**

The effect of temperature on the optical properties of aluminum has been studied directly with ellipsometry by Liljenvall *et al.* [101] and by Mathewson and Myers [5], who found $\sigma(\omega)$ as a function of energy and temperature as shown in Fig. 17. As the temperature rises, the 1.5-eV interband transition shows a pronounced broadening, shifts toward lower energies, and becomes weaker. The free-electron contribution also shows a broadening but becomes stronger with rising temperature. This is in line with the fact that in the liquid metal interband transitions are not observed [97], all the conduction electron oscillator strength having been transferred to the Drude-like intraband term. The ellipsometric results have been analyzed [5] within the framework of the Ashcroft–Sturm [18] two-band model to obtain the temperature dependence of the Drude parameters, the Fourier components of the crystal field, and the interband relaxation time (see Table XI and Mathewson and Myers [5]).

As a result of the high values of $n(\omega)$ and $k(\omega)$ in the visible and infrared, even a large variation of the optical constants with temperature produces only a small change in the reflectance R . This effect has been investigated qualitatively in thermomodulation studies by Rosei and Lynch [135]. The results are rather featureless except near the “1.5-eV” interband absorption. They show that dR/dT is negative below approximately 1.3 eV (0.95 μm) and positive at higher energies. This is consistent with the ellipsometric results [5]

and may be understood as arising primarily from (1) a broadening and shift toward lower energy of the “1.5-eV” interband transition with rising temperature, combined with (2) a decrease in reflectance in the Drude region (below ~ 1 eV) as a result of a shortening of the intraband lifetime at higher temperatures. Although the temperature modulation involved was not determined, the scale of the effect is given by Pudkov’s measurements [136] at 1.96 eV ($0.633\text{ }\mu\text{m}$) that yield a value for dR/dT of approximately $3 \times 10^{-5}\text{ }(^{\circ}\text{C})^{-1}$.

The temperature dependence of the reflectance and absorptance is of particular interest for high-power laser applications (see, for example, Pudkov [136] and Konov and Tokarev [137]). A detailed study of the wavelength and temperature dependence of the absolute reflectance of metallic Al (as well as Ag and Cu) from $4000\text{ }\text{\AA}$ to $10\text{ }\mu\text{m}$ has been made by Decker and Hodgkin [138]. Measurements were made to a precision of a few parts in 10^5 and are in agreement with the qualitative discussion given above. The reader is referred to Decker and Hodgkin [138] for numerical results.

E Pressure Dependence

The effect of pressure on the reflectance of aluminum has been investigated at room temperature by Tups and Syassen [139] in both the infrared and the visible (0.5–3.0 eV). Polycrystalline samples were studied by using the gas-gated diamond-anvil technique [140] with the reflectance measured relative to the diamond windows of the pressure cell. Representative results are shown in Fig. 18. Note that the reflectance of the diamond–aluminum interface shown in Fig. 18 for a pressure of 1 kbar is significantly less than that shown in Fig. 12 for the air–aluminum interface at 1 atm because of the higher refractive index of diamond ($n \approx 2.4$). Tups and Syassen [139] found that as pressure is increased, the reflectance drop arising from the strong interband transitions associated with the square (200) faces of the Brillouin zone shifts toward higher energies. From a value of ~ 1.47 eV at 1 atm, the transition energy rises monotonically to ~ 2.34 eV at 310 kbar. This shift is indicated by the upper set of arrows in the figure. The initial rate of change of the transition energy is $4.35 \pm 0.3\text{ meV/kbar}$.

Near pressures of 100 kbar, a shoulder developed on the reflectance curve at about 1 eV. With increasing pressure this shoulder moved toward higher energies, and a well-defined reflectance minimum develops at low energies. This feature corresponds to the second strong interband transition associated with the hexagonal (111) faces of the Brillouin zone. At normal pressure and temperature, this transition lies at ~ 0.4 eV and is obscured by the intraband absorption, but it shifts toward higher energies with increasing pressure reaching ~ 1.32 eV at 310 kbar, where it is well resolved from lower-energy intraband processes. The peak positions of this low-energy interband transition are indicated by the lower set of arrows.

In a detailed analysis, Tups and Syassen [139] show that both the interband transitions become stronger at the expense of the oscillator strength of the intraband absorption. This exchange of strength reflects the partial f sum rule for conduction-electron absorption [147], which enters the theory of Ashcroft and Sturm [18]. The weakening of intraband processes is evident in the reflectance spectra as the retreat of the high-reflectance almost-free-electron region that lies below ~ 1 eV under normal conditions, toward lower energies with increasing pressure. A fitting of the data to the theory of Ashcroft and Sturm [18] indicates that the optical mass, which is inversely proportional to the intraband strength, increases by approximately a factor of 1.4 in going from atmospheric pressure to 310 kbar. This corresponds to a transfer of $\sim 28\%$ of the intraband strength of approximately 0.5 e/at. to the interband transitions.

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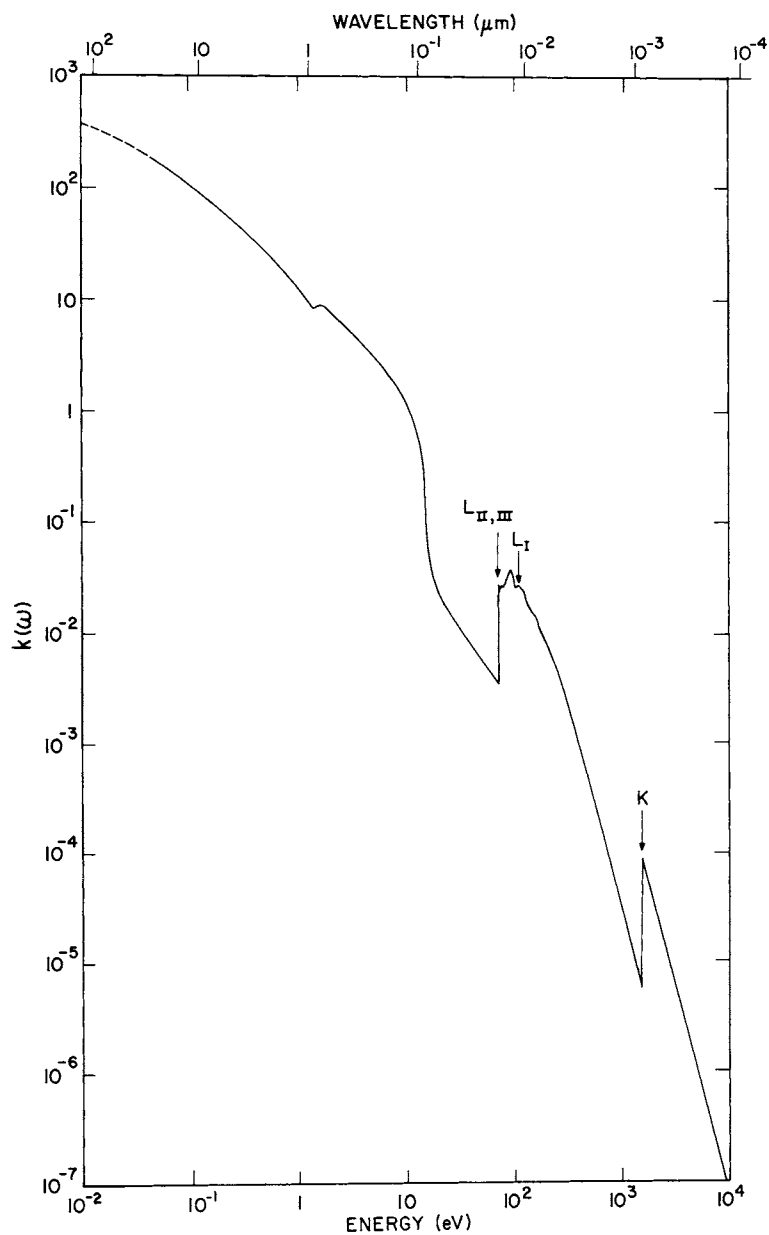


Fig. 11. The extinction coefficient $k(\omega)$; i.e., the imaginary part of the refractive index, for metallic aluminum at room temperature. (After Shiles *et al.* [7].)

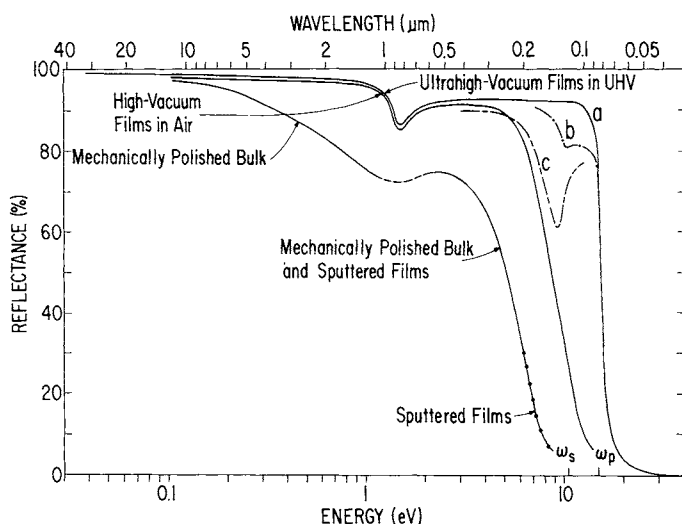


Fig. 12. The reflectance of metallic aluminum at room temperature. The uppermost curve gives the reflectance of opaque evaporated aluminum films prepared in ultrahigh vacuum. These films are presumed to be free of bulk and surface oxide contamination. The effect of light scattering and coupling to surface plasmons (at $\omega_s \approx 10.6$ eV) is illustrated by curves b and c. Curve b was obtained by Vehse *et al.* [90] for a film on a microscope slide cleaned by vapor decreasing and evaporated in a vacuum of $\sim 3 \times 10^{-8}$ torr; the “oxide-free” reflectance was obtained by extrapolation of the reflectance back to the time that the evaporation was completed. Curve c is for a uhv film with rms surface roughness σ of 18 Å, as reported by Endriz and Spicer [53]. The curve labeled high vacuum is an average for films evaporated in vacuum of the order of 10^{-5} torr. At this pressure surface and bulk oxidation occurs. The values reported for such films vary widely, especially in the ultraviolet, presumably because of surface roughness. The third curve is representative of mechanically polished samples, but the reported range of measurements is extremely large. Data for sputtered films are available from 380 nm well into the UV [44, 45]; curiously enough, they track the data for polished samples very closely.

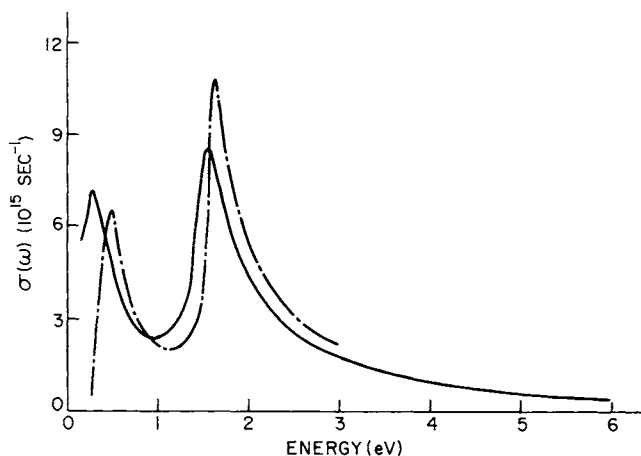


Fig. 13. The real part of the interband conductivity for aluminum obtained by subtracting the intraband contribution in the Drude model from the measured optical conductivity. The solid curve is for room temperature and was calculated by Smith and Segall [15], who employed Shiles *et al.*'s [7] analysis of uhv reflectance data. The dashed curve is taken from Benbow and Lynch [14] and applies to electropolished samples at 4.2 K.

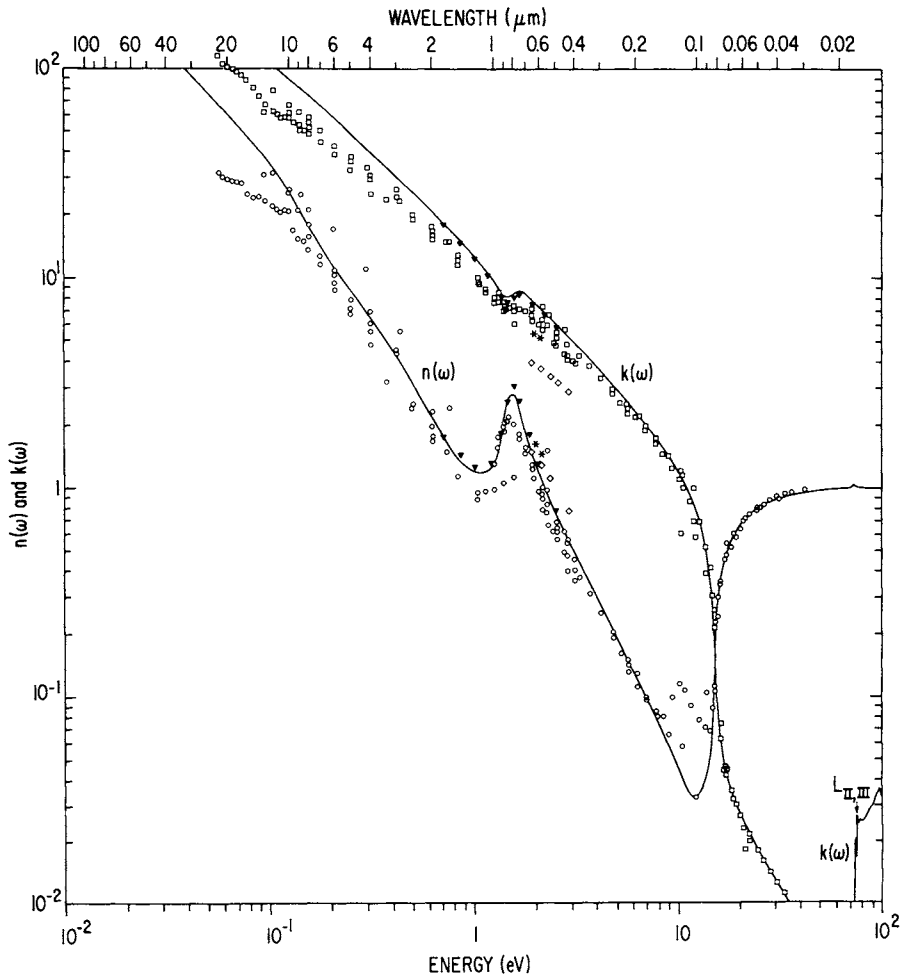


Fig. 14. The complex refractive index $n(\omega) + ik(\omega)$ for aluminum. The curves are taken from Shiles *et al.*'s [7] analysis of uhv reflectance data. A portion of the uhv ellipsometric data of Mathewson and Myers [100] is given for comparison. Quincke's [33] and Drude's [34] results for polished bulk samples are shown for historical interest; considering the materials and techniques available, these early measurements are remarkably good, especially for $n(\omega)$. The remainder of the data points are given to show the range of values of $n(\omega)$ ($\circ$) and $k(\omega)$ ($\square$) reported in the literature. Most refer to evaporated films prepared in conventional or high vacuum and measured by using polarimetric, interferometric, and like methods. The sources of these data are given in the references [10, 11, 25, 26, 27, 51, 55–56, 65, 66, 73, 75, 91, and 121–129]. Note that the curves for $n(\omega)$ and $k(\omega)$ curve cross each other at roughly 15 eV, the plasmon energy. This corresponds to the plasmon condition $\epsilon_1(\omega_p) = n^2(\omega_p) - k^2(\omega_p) \approx 0$. The onset of the L-shell absorption appears in the lower-right-hand corner. The corresponding dispersion is visible as a little "pimple" on the index curve near 72 eV. (Mathewson and Myers [100], $\blacktriangledown$; Quincke [33], $\diamond$; Drude [34], $*$.)

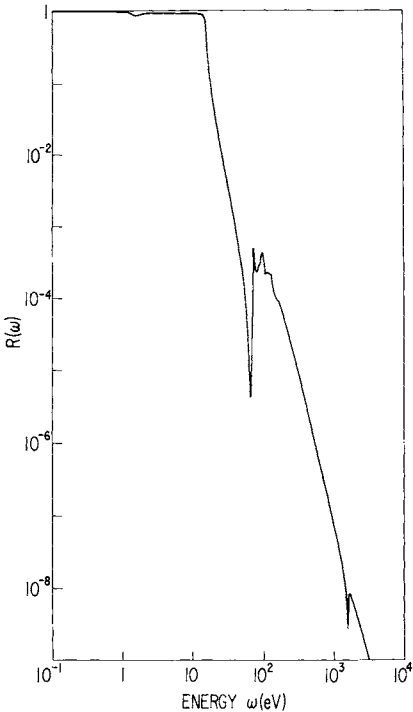


Fig. 15. The reflectance $R(\omega)$ at normal incidence of a smooth oxide-free metallic aluminum surface in vacuum. (After Shiles *et al.* [7].)

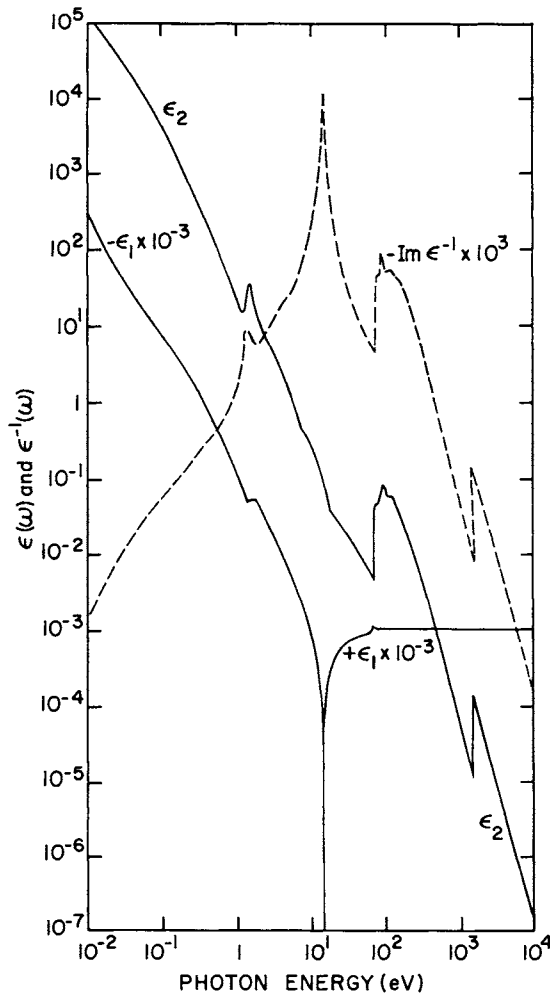


Fig. 16. The dielectric-response function of metallic aluminum. The real part $\epsilon_1(\omega)$ is negative with large absolute values at low energies, vanishes at $\hbar\omega_p \approx 15.0$ eV (the plasmon energy for conduction electrons), and is positive and approaches unity at higher energies. The imaginary part $\epsilon_2(\omega)$ is also largest at low energies and decreases with energy except at thresholds for newer modes of excitation (first at the beginning of interband excitation of valence electrons, next at the L-shell threshold, and finally at the K-shell threshold). The quantity $\text{Im}[-1/\epsilon(\omega)]$, which governs the energy transfer from fast charged particles, shows a prominent maximum near $\hbar\omega_p$ and is small at both lower and higher energies. (After Shiles *et al.* [7].)

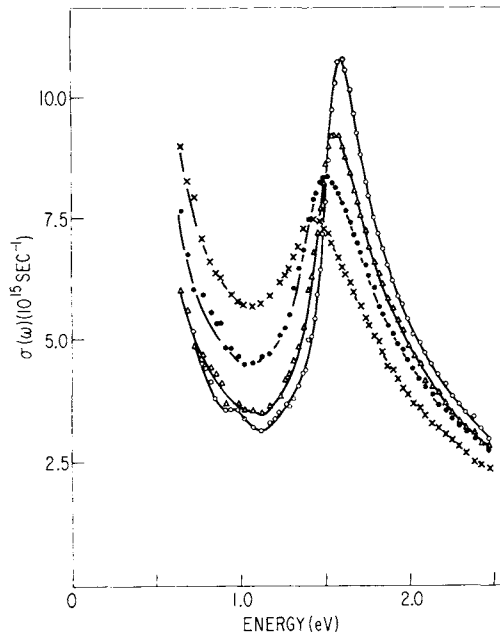


Fig. 17. The real part of the optical conductivity as a function of temperature for clean polycrystalline aluminum films deposited at room temperature. (198 K, $\circ$; 298 K, $\triangle$; 404 K, $\bullet$; and 552 K, $\times$.) (After Mathewson and Myers [5].) (Copyright © 1972 by the Institute of Physics).

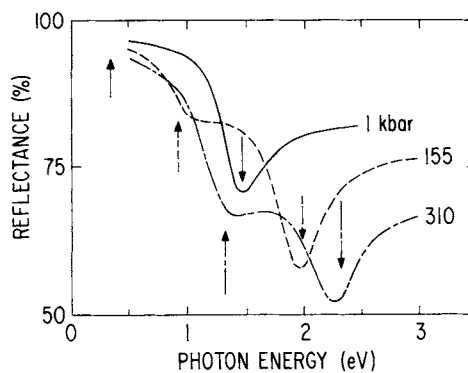


Fig. 18. The reflectance of aluminum at an interface with diamond as a function of pressure and photon energy. Pressures are given in kilobars to the right of each curve. The positions of the principal interband transitions, as determined by a fitting to the theory of Ashcroft and Sturm [18], are indicated by arrows coded to the corresponding reflectance curve. The position of the high-energy transition $2V_{200}$ is indicated by the upper set of arrows. That of the low-energy transition $2V_{111}$ is indicated by the lower set of arrows. (After Tups and Syassen [139].)

TABLE XI
Drude Parameters for the Intraband Absorption of Metallic Aluminum^a

Source	Temperature (K)	Strength ^b			Damping		DC conductivity $\sigma(0)$ (10^{17} sec^{-1})	
		$\hbar\Omega_p$ (eV)	n_{eff} (e/at.)	m_{opt} (m_e)	τ (10^{-14} sec)	$\hbar\gamma = \hbar\tau^{-1}$ (meV)	Optical	Electrical
Ehrenreich <i>et al.</i> [1]	RT	12.7	1.94	1.55	0.512	129	1.52	3.18 [16]–3.28 [17]
Bennett <i>et al.</i> [12]	RT	14.7	2.60	1.15	0.801	82.2	3.18 (input) ^c	
Powell [3]	RT	12.2	1.80	1.67	0.66	100	1.81	
Dresselhaus <i>et al.</i> [13]	RT	12.9 ± 0.7	2.0 ± 0.2	1.5 ∓ 0.15	0.5 ± 0.2	160 ∓ 60	1.60 ± 0.8	
Mathewson and Myers [5]	198	12.8	1.99	1.51^d	1.18	55.8	3.58	5.49 [5]
Mathewson and Myers [5]	298	13.0	2.03	1.48^d	1.02	64.5	3.14	3.21 [5]
Mathewson and Myers [5]	404	13.2	2.10	1.43^d	0.62	105	1.96	2.30 [5]
Mathewson and Myers [5]	552	13.3	2.13	1.41^d	0.52	128	1.65	1.63 [5]
Benbow and Lynch [14]	4.2	12.7	1.94	1.55 (input)^e	1.10	60	3.25	
Smith and Segall [15]	RT	12.5 ± 0.3	1.88 ± 0.09	1.60 ∓ 0.08	1.06 ± 0.12	63 ∓ 7	3.00 ± 0.3	3.18 [16]–3.28 [17]

^a See Eqs. (1)–(4) in the text.

^b The effective density of electrons n_{eff} is given in electrons per atom (e/at.). In aluminum the actual density of conduction electrons is 3 e/at. The optical conductivity at $\omega = 0$ has been calculated from Eq. (5); the electrical values of $\sigma(0)$ are for dc measurements on bulk samples.

^c The measured bulk value of $\sigma(0)$ was used to fix the ratio Ω_p^2/γ via Eq. (5).

^d Evaluated by fitting the interband absorption and use of Eq. (42) of Ashcroft and Sturm [18].

^e Assumed on the basis of a fit by Ashcroft and Sturm [18].

TABLE XII

The Optical Constants n and k and Reflectivity of Evaporated Metallic Aluminum Films at Room Temperature from $32\ \mu\text{m}$ to the $L_{II,III}$ Edge^{a,b}

eV	cm^{-1}	μm	n	n	k	k	R	R	R	R_{hv}
75		0.01653	1.01 [7]		2.40×10^{-2} [7]		1.74×10^{-4} [7]			
74		0.01676	1.01		2.42×10^{-2}		1.97			
73		0.01698	1.02		1.91×10^{-2}		2.33×10^{-4}			
72		0.01722	1.02		3.46×10^{-3}		6.85×10^{-5}			
71		0.01746	1.01		3.46		2.75			
70		0.01771	1.01		3.52		1.27×10^{-5}			
65		0.01908	0.995		4.17		9.85×10^{-6}			
60		0.02066	0.987		4.41		4.64×10^{-5}			
55		0.02254	0.979		5.08		1.21×10^{-4}			
50		0.02480	0.969		5.87		2.57			
45		0.02755	0.957		6.82		5.01			
40		0.03100	0.940		8.16		9.89×10^{-4}			
38		0.03263	0.931		8.71		1.30×10^{-3}			
36		0.03444	0.921	0.96 [55]	9.32×10^{-3} [7]	9.5×10^{-3} [55]	1.73	4.40×10^{-4} [55]		
34		0.03647	0.909		1.02×10^{-2}		2.33			
33		0.03757	0.902	0.943	1.07	1.10×10^{-2}	2.71	8.93×10^{-4}		
32		0.03875	0.894		1.11		3.17			
31		0.04000	0.885		1.16		3.75			

(continued)

^a Columns labeled [7] are the composite data of Shiles *et al.* [7]; the values tabulated are based primarily on the Kramers-Kronig analysis of reflectance measurements of uhv films from 32 to $0.1\ \mu\text{m}$. The uhv reflectance data of Bennett *et al.* [28], are given in columns labeled [28]. The *in situ* uhv ellipsometric data of Mathewson and Myers [100] are given in columns labeled [100], and the vacuum-UV transmission and angular-dependence-of-reflection measurements of Ditchburn and Freeman [55] are given in columns labeled [55]. Composite reflectance values for films evaporated in high vacuum and measured in air, R_{hv} , are given in columns labeled [142]. These data are taken from Smith *et al.* [142] and are also shown as the “high vacuum” curve in Fig. 12. The reflectance of these oxidized films is not unique. However, films prepared by rapid evaporation in high vacuum and exposed to a normal laboratory environment are generally reported to have reflectances throughout the IR and visible which are roughly 1% below the values for smooth, unoxidized films prepared in uhv. At higher energies, the high-vacuum films show a sharp fall-off in reflectance because of surface-oxide absorption and because of the scattering and surface-plasmon absorption caused by surface roughness. A wide range of optical properties is generally observed in the ultraviolet unless special precautions are taken to control the film’s surface roughness.

^b A tabulation of optical constants at energies higher than those given here is available in Reference 142.

TABLE XII (Continued)

Metallic Aluminum

eV	cm ⁻¹	μm	n	n	k	k	R	R	R	R _{hv}
30		0.04133	0.876	0.912	1.25	1.25 × 10 ⁻²	4.44	2.16 × 10 ⁻³		
29		0.04275	0.865		1.35		5.28			
28		0.04428	0.854	0.880	1.45	1.41	6.30	4.13		
27		0.04592	0.841		1.55		7.58			
26		0.04769	0.826	0.838	1.65	1.59	9.20 × 10 ⁻³	7.84 × 10 ⁻³		
25		0.04959	0.809		1.77		1.13 × 10 ⁻²			
24		0.05166	0.789	0.785	1.90	1.82	1.40	1.46 × 10 ⁻²		
23		0.05391	0.766		2.05		1.77			
22		0.05636	0.740	0.718	2.22	2.13	2.26	2.71		
21		0.05904	0.707		2.42		2.96			
20		0.06199	0.668	0.635	2.68	2.67	3.98	5.01		
19.5		0.06358	0.646		2.84		4.67			
19		0.06526	0.620	0.580	3.02	3.07	5.54	7.10 × 10 ⁻²		
18.5		0.06702	0.591		3.24		6.64			
18		0.06888	0.558	0.520	3.48	3.55	8.09 × 10 ⁻²	0.10		
17.5		0.07085	0.520		3.81		0.100			
17		0.07293	0.474	0.445	4.23	4.24	0.128	0.148		
16.5		0.07514	0.419		4.87		0.168			
16		0.07749	0.351	0.345	5.95	6.32 × 10 ⁻²	0.233	0.239		
15.5		0.07999	0.258		7.77 × 10 ⁻²		0.350			
15		0.08266	0.125	0.225	0.153	0.220	0.612	0.419		0.050 [142]
14.5		0.08551	0.0616		0.301		0.798			0.057
14		0.08856	0.0481		0.416		0.849			0.064
13.5		0.09184	0.0409	0.104	0.517	0.390	0.879	0.697		0.074
13		0.09537	0.0376		0.609		0.896			0.086
12.5		0.09919	0.0344		0.700		0.912			0.102
12	96,790	0.1033	0.0328	0.033	0.791	0.580	0.922	0.906		0.127
11.5	92,750	0.1078	0.0331		0.883		0.928			0.165

11	88,720	0.1127	0.0356	0.978	0.930	0.215
10.5	84,690	0.1181	0.0396	1.08	0.929	0.270
10	80,660	0.1240	0.0442	1.18	0.929	0.328
9.5	76,620	0.1305	0.0495	1.29	0.928	0.388
9	72,590	0.1378	0.0557	1.40	0.928	0.448
8.75	70,570	0.1417	0.0592	1.46	0.927	0.478
8.5	68,560	0.1459	0.0630	1.53	0.927	0.510
8.25	66,540	0.1503	0.0671	1.59	0.927	0.543
8	64,520	0.1550	0.0716	1.66	0.927	0.579
7.75	62,510	0.1600	0.0765	1.74	0.927	0.616
7.5	60,490	0.1653	0.0820	1.81	0.927	0.656
7.25	58,480	0.1710	0.0880	1.90	0.926	0.698
7	56,460	0.1771	0.0946	1.98	0.926	0.735
6.75	54,440	0.1837	0.102	2.07	0.926	0.766
6.5	52,430	0.1907	0.110	2.17	0.926	0.793
6.25	50,410	0.1984	0.119	2.28	0.926	0.820
6	48,390	0.2066	0.130	2.39	0.926	0.841
5.75	46,380	0.2156	0.141	2.51	0.926	0.858
5.5	44,360	0.2254	0.155	2.64	0.926	0.873
5.25	42,340	0.2362	0.172	2.79	0.925	0.884
5	40,330	0.2480	0.190	2.94	0.924	0.894
4.75	38,310	0.2610	0.209	3.11	0.925	0.901
4.5	36,300	0.2755	0.233	3.30	0.925	0.905
4.25	34,280	0.2917	0.261	3.51	0.925	0.908
4.133	33,330	0.300	0.276	3.61	0.925	0.909
4	32,260	0.3100	0.294	3.74	0.925	0.910
3.9	31,460	0.3179	0.310	3.84	0.925	0.911
3.8	30,650	0.3263	0.326	3.95	0.925	0.912
3.7	29,840	0.3351	0.344	4.06	0.925	0.913
3.6	29,040	0.3444	0.364	4.17	0.925	0.913
3.542	28,570	0.350	0.375	4.24	0.925	0.914
3.5	28,230	0.3542	0.385	4.30	0.925	0.914

0.9208 [28]

0.9205

(continued)

TABLE XII (Continued)

Metallic Aluminum

eV	cm ⁻¹	μm	n	n	k	k	R	R	R	R _{hv}
3.4	27,420	0.3647	0.407		4.43		0.924			0.914
3.3	26,620	0.3757	0.432		4.56		0.924			0.915
3.2	25,810	0.3875	0.460		4.71		0.924			0.915
3.1	25,000	0.400	0.490		4.86		0.924	0.9194		0.915
3.0	24,200	0.4133	0.523		5.02		0.924			0.914
2.9	23,390	0.4275	0.558		5.20		0.924			0.914
2.8	22,580	0.4428	0.598		5.38		0.924			0.913
2.755	22,220	0.450	0.618		5.47		0.924	0.9175		0.913
2.7	21,780	0.4592	0.644		5.58		0.924			0.913
2.6	20,970	0.4769	0.695		5.80		0.924			0.912
2.5	20,160	0.4959	0.755	0.779 [100]	6.03	5.84 [100]	0.923		0.916 [100]	0.911
2.480	20,000	0.500	0.769		6.08		0.923	0.9162		0.911
2.45	19,760	0.5061	0.789	0.818	6.15	5.93	0.923		0.915	0.911
2.4	19,360	0.5166	0.826	0.852	6.28	6.10	0.923		0.916	0.910
2.35	18,950	0.5276	0.867	0.891	6.42	6.23	0.922		0.916	0.909
2.3	18,550	0.5391	0.912	0.944	6.55	6.36	0.922		0.915	0.909
2.254	18,180	0.550	0.958		6.69		0.921	0.9157		0.908
2.25	18,150	0.5510	0.963	1.00	6.70	6.52	0.921		0.914	0.908
2.2	17,740	0.5636	1.02	1.07	6.85	6.64	0.920		0.912	0.907
2.15	17,340	0.5767	1.08	1.15	7.00	6.77	0.919		0.909	0.906
2.1	16,940	0.5904	1.15	1.22	7.15	6.93	0.918		0.908	0.905
2.066	16,670	0.600	1.20		7.26		0.917	0.9117		0.904
2.05	16,530	0.6048	1.22	1.31	7.31	7.09	0.916		0.906	0.904
2.0	16,130	0.6199	1.30	1.40	7.48	7.21	0.915		0.903	0.903
1.95	15,730	0.6358	1.39	1.51	7.65	7.37	0.913		0.900	0.901
1.908	15,390	0.650	1.47		7.79		0.912	0.9057		0.899
1.9	15,320	0.6526	1.49	1.63	7.82	7.49	0.912		0.897	0.899

1.85	14,920	0.6702	1.60	1.78	8.01	7.66	0.910		0.893	0.896
1.8	14,520	0.6888	1.74	1.94	8.21	7.83	0.907		0.889	0.893
1.771	14,290	0.700	1.83		8.31		0.905	0.8977		0.890
1.75	14,120	0.7085	1.91	2.13	8.39	8.00	0.903		0.885	0.888
1.7	13,710	0.7293	2.14	2.34	8.57	8.15	0.897		0.880	0.882
1.653	13,330	0.750	2.40		8.62		0.888	0.8862		0.876
1.65	13,310	0.7514	2.41	2.57	8.62	8.25	0.888		0.873	0.876
1.6	12,905	0.7749	2.63	2.87	8.60	8.23	0.879	0.8773	0.861	0.868
1.55	12,500	0.7999	2.80	3.04	8.45	8.08	0.869	0.8676	0.851	0.860
1.503	12,120	0.825	2.75		8.31		0.868	0.8657		0.858
1.5	12,100	0.8266	2.74	2.94	8.31	7.76	0.868		0.845	0.858
1.459	11,770	0.850	2.61		8.22		0.870	0.8677		0.859
1.45	11,700	0.8551	2.58	2.53	8.21	7.61	0.872		0.856	0.860
1.417	11,430	0.875	2.38		8.18			0.878	0.8744	0.868
1.4	11,290	0.8856	2.24	2.14	8.21	7.75	0.885		0.878	0.874
1.378	11,110	0.900	2.06		8.30		0.895	0.8908		0.884
1.35	10,890	0.9184	1.86	1.82	8.44	8.14	0.906		0.902	0.897
1.340	10,810	0.925	1.77		8.49		0.911	0.9075		0.902
1.305	10,530	0.950	1.49		8.88		0.930	0.9243		0.916
1.3	10,490	0.9537	1.47	1.58	8.95	8.69	0.932		0.923	0.918
1.25	10,080	0.9919	1.37	1.41	9.49	9.19	0.943		0.937	0.932
1.240	10,000	1.000	1.35		9.58		0.944	0.9402		0.934
1.2	9,679	1.033	1.26	1.32	10.0	9.73	0.952		0.947	0.942
1.15	9,275	1.078	1.21	1.25	10.6	10.3	0.958		0.955	0.948
1.1	8,872	1.127	1.20	1.23	11.2	10.9	0.963		0.960	0.953
1.05	8,469	1.181	1.21	1.22	11.8	11.6	0.967		0.965	0.957
1.033	8,333	1.200	1.21		12.0		0.968	0.9637		0.958
1	8,066	1.240	1.21	1.25	12.5	12.2	0.970		0.967	0.960
0.95	7,662	1.305	1.23	1.30	13.2	12.9	0.973		0.970	0.963
0.9	7,259	1.378	1.26	1.35	14.0	13.7	0.975		0.972	0.964
0.85	6,856	1.459	1.33	1.43	14.9	14.5	0.977		0.974	0.966
0.8266	6,667	1.500	1.38		15.4		0.977	0.9742		0.967

(continued)

TABLE XII (Continued)

Metallic Aluminum

eV	cm ⁻¹	μm	n	n	k	k	R	R	R	R _{hv}
0.8	6,452	1.550	1.44	1.50	16.0	15.5	0.978		0.976	0.967
0.75	6,049	1.653	1.59	1.59	17.1	16.5	0.979		0.977	0.968
0.7	5,646	1.771	1.77	1.75	18.3	17.8	0.979		0.978	0.969
0.65	5,243	1.907	1.99		19.8		0.980			0.970
0.6199	5,000	2.0	2.15		20.7		0.980	0.9779		0.970
0.6	4,839	2.065	2.27		21.4		0.981			0.970
0.55	4,436	2.254	2.62		23.3		0.981			0.971
0.5	4,033	2.480	3.07		25.6		0.982			0.972
0.45	3,629	2.755	3.68		28.3		0.982			0.973
0.4133	3,333	3.0	4.24		30.6		0.982	0.9805		0.973
0.4	3,226	3.100	4.45		31.5		0.983			0.974
0.375	3,025	3.306	4.88		33.4		0.983			0.974
0.35	2,823	3.542	5.44		35.6		0.983			0.975
0.325	2,621	3.815	6.00		38.1		0.984			0.975
0.3100	2,500	4.0	6.43		39.8		0.984	0.9826		0.975
0.3	2,420	4.133	6.76		41.0		0.984			0.976
0.275	2,218	4.509	7.61		44.3		0.985			0.976
0.25	2,016	4.959	8.59		48.2		0.986			0.977
0.2480	2,000	5.0	8.67		48.6		0.986	0.9843		0.977
0.225	1,815	5.510	9.85		53.2		0.987			0.977
0.2066	1,667	6.0	11.1		57.6		0.987	0.9856		0.978
0.2	1,613	6.199	11.7		59.4		0.987			0.978
0.19	1,532	6.526	12.7		62.2		0.987			0.978
0.18	1,452	6.888	13.7		65.2		0.988			0.979
0.1771	1,429	7.0	14.0		66.2		0.988	0.9866		0.979
0.17	1,371	7.293	14.9		68.8		0.988			0.979
0.16	1,290	7.749	16.5		72.7		0.988			0.980
0.1550	1,250	8.0	17.5		74.9		0.988	0.9872		0.980

0.15	1,210	8.266	18.6	77.0	0.988		0.980
0.14	1,129	8.856	20.9	81.5	0.988		0.980
0.1378	1,111	9.0	21.5	82.6	0.988	0.9874	0.980
0.13	1,049	9.537	23.5	86.5	0.988		0.981
0.1240	1,000	10.0	25.3	89.8	0.988	0.9876	0.981
0.12	967.9	10.33	26.6	92.2	0.989		0.981
0.1127	909.1	11.0	29.2	96.6	0.989	0.9879	0.982
0.11	887.2	11.27	30.2	98.4	0.989		0.982
0.1033	833.3	12.0	33.0	103	0.989	0.9882	0.982
0.1	806.5	12.40	34.5	106	0.989		0.982
0.09537	769.2	13.0	36.6	109	0.989	0.9884	0.982
0.095	766.2	13.05	36.8	110	0.989		0.982
0.09	725.9	13.78	39.7	114	0.989		0.983
0.08856	714.3	14.0	40.5	116	0.989	0.9886	0.983
0.085	685.6	14.59	42.8	119	0.989		0.983
0.08	645.2	15.50	46.0	124	0.990		0.983
0.07749	625.0	16.0	47.7	127	0.990	0.9892	0.983
0.075	604.9	16.53	49.7	129	0.990		0.983
0.07	564.6	17.71	53.8	136	0.990		0.983
0.06888	555.6	18.0	54.7	137	0.990	0.9896	0.983
0.065	524.3	19.07	57.6	143	0.990		0.984
0.06199	500.0	20.0	60.7	147	0.990	0.9902	0.984
0.06	483.9	20.66	62.9	151	0.991		0.984
0.05636	454.6	22.0	66.9	157	0.991	0.9907	0.984
0.055	443.6	22.54	68.3	160	0.991		0.984
0.05166	416.7	24.0	72.2	168	0.991	0.9912	0.984
0.05	403.3	24.80	75.0	172	0.992		0.984
0.04769	384.6	26.0	79.1	178	0.992	0.9918	0.985
0.045	362.9	27.55	84.7	186	0.992		0.985
0.04428	357.1	28.0	86.3	189	0.992	0.9923	0.985
0.04133	333.3	30.0	94.2	199	0.992	0.9928	0.985
0.04	322.6	31.00	98.6	204		0.992	0.985
0.03875	312.5	32.0	103	208	0.992	0.9933	0.985

TABLE XIII
The Dielectric Function for Evaporated Metallic Aluminum Films at Room Temperature from 0.04 to 10,000 eV^{a-d}

eV	Å	ϵ_1	ϵ_2	eV	Å	ϵ_1	ϵ_2
10,000	1.240	0.99999	1.42×10^{-7}	1,556	7.968	0.99982	6.209
9,000	1.378	0.99999	2.16	1,555	7.973	0.99983	5.909
8,000	1.550	0.99998	3.40	1,550	7.999	0.99978	1.200
7,000	1.771	0.99998	5.80×10^{-7}	1,540	8.051	0.99974	1.230
6,000	2.066	0.99997	1.046×10^{-6}	1,530	8.104	0.99971	1.260
5,000	2.480	0.99996	2.08	1,520	8.157	0.99969	1.290
4,000	3.100	0.99993	4.900	1,510	8.211	0.99968	1.324
3,500	3.542	0.99991	8.006×10^{-6}	1,500	8.266	0.99966	1.356
3,000	4.133	0.99988	1.420×10^{-5}	1,450	8.551	0.99961	1.560
2,500	4.959	0.99983	2.718	1,400	8.856	0.99957	1.780
2,000	6.199	0.99974	6.406	1,300	9.537	0.99947	2.319
1,900	6.526	0.99972	7.691	1,200	10.33	0.99936	3.119
1,800	6.888	0.99970	9.426×10^{-5}	1,100	11.27	0.99922	4.312
1,700	7.293	0.99968	1.159×10^{-4}	1,000	12.40	0.99904	6.222
1,650	7.514	0.99968	1.309	900	13.78	0.99879	9.355×10^{-5}
1,600	7.749	0.99970	1.476	800	15.50	0.99845	1.462×10^{-4}
1,590	7.798	0.99971	1.520	700	17.71	0.99796	2.363
1,580	7.847	0.99973	1.570	600	20.66	0.99717	4.408
1,570	7.897	0.99976	1.622	500	24.80	0.99590	8.695×10^{-4}
1,565	7.922	0.99979	1.686	400	31.00	0.99378	1.939×10^{-3}
1,564	7.927	0.99981	1.709	300	41.33	0.9898	4.684
1,563	7.933	0.99983	1.636	250	49.59	0.9862	8.325×10^{-3}
1,562	7.938	0.99985	1.437	200	61.99	0.9820	1.490×10^{-2}
1,561	7.943	0.99985	1.220	190	65.26	0.9810	1.680
1,560	7.948	0.99985	1.000×10^{-4}	180	68.88	0.9797	1.913
1,559	7.953	0.99985	8.229×10^{-5}	170	72.93	0.9776	2.174
1,558	7.958	0.99984	6.583	160	77.49	0.9785	2.718
1,557	7.963	0.99983	5.860	150	82.68	0.9791	2.925

145	85.51	0.9782	3.055×10^{-2}	88	140.9	1.010	5.823×10^{-2}
140	88.56	0.9772	3.225	86	144.2	1.011	5.680
135	91.84	0.9756	3.511	84	147.6	1.013	5.402
130	95.37	0.9746	4.070	82	151.2	1.014	5.129
125	99.19	0.9780	4.634	80	155.0	1.013	4.928
120	103.3	0.9814	4.769	79	156.9	1.013	4.933
115	107.8	0.9840	4.947	78	159.0	1.014	4.987
110	112.7	0.9871	5.059	77	161.0	1.018	5.082
105	118.1	0.9860	4.850	76	163.1	1.020	4.818
100	124.0	0.9804	5.927	75.5	164.2	1.021	4.841
98	126.5	0.9832	6.559	75	165.3	1.022	4.856
96	129.2	0.9915	7.150	74.5	166.4	1.025	4.928
94	131.9	0.9998	7.066	74	167.5	1.029	4.904
92	134.8	1.007	6.706	73.9	167.8	1.030	4.915
90	137.8	1.010	6.218	73.8	168.0	1.031	4.928

(continued)

^a Based on Shiles *et al.* [7].

^b These data are based primarily on the analysis of reflectance measurements below the $L_{II,III}$ edge and on transmission measurements above the edge. They are in close agreement with direct ellipsometric measurements on oxide-free films by Mathewson and Myers [100] in the range 0.7 to 2.5 eV (see Table XII). Suggested extrapolations are given in the footnotes *c* and *d*.

^c Low-energy extrapolations. The room-temperature dielectric function presented here may be extrapolated to lower energies by using the Drude model [Eq. (1)] with $\hbar\Omega_p = 11.5$ eV and $\hbar\gamma = 50.6$ meV [$\sigma(0) = 3.16 \times 10^{17}$ sec⁻¹]. This extrapolation fits on continuously at 0.04 eV, but with a small discontinuity in slope. However, it should be recognized that, while the dc conductivity is in excellent agreement with the measured value ($3.18 - 3.28 \times 10^{17}$ sec⁻¹) [16, 17], Ω_p and γ are somewhat outside the range recommended by Smith and Segall [5] (see Table XI). This represents a negligible conflict since, for the high reflectances (>99%) beyond 20 μ m, the experimental error ($\pm 0.1\%$) in the reflectance measurements used to obtain the dielectric function translates into an uncertainty of the order of $\pm 25\%$ in $\epsilon(\omega)$ even in favorable cases. The values of Smith and Segall [15] arise from fits over the range 0.04–3 eV, not just the lowest energy point, which is subject to considerable uncertainty.

^d High-energy extrapolations. For $\epsilon_1(\omega)$: at frequencies well above the K edge the real part of the dielectric function approaches $\epsilon_1 \approx 1 - \omega_{p,i}^2/\omega^2$, where $\omega_{p,i}$ is the plasma frequency for the *total* electron density (13 e/at.). For aluminum at room temperature $\omega_{p,i} \approx 32.86$ eV and the asymptotic region is achieved by $\omega \approx 3000$ eV. For $\epsilon_2(\omega)$: beyond the K edge $\epsilon_2(\omega)$ falls off very nearly as a power of energy $\epsilon_2(\omega) \sim \omega^{-\delta}$. The exponent varies slowly with energy. At 5000 eV, $\delta \sim 3.8$; at 10⁴ eV, $\delta \sim 4.0$; and from 10⁴ to 10⁵ eV, it increases to $\delta = 4.2$. An extrapolation from 10⁴ eV using $\delta = 4.1$ reproduces photoelectric data up to 10⁵ eV reasonably well. Theory predicts an asymptotic value of $\delta = 4.5$ [143].

TABLE XIII (Continued)
Metallic Aluminum

eV	Å	ϵ_1	ϵ_2	eV	Å	ϵ_1	ϵ_2
73.7	168.2	1.032	4.949×10^{-2}	55	225.4	0.9581	9.942×10^{-3}
73.6	168.5	1.033	4.976	50	248.0	0.9389	1.138×10^{-2}
73.5	168.7	1.035	5.048	45	275.5	0.9153	1.304
73.4	168.9	1.038	5.177	40	310.0	0.8827	1.533
73.3	169.1	1.043	5.308	35	354.2	0.8368	1.797
73.2	169.4	1.050	4.955	30	413.3	0.7665	2.197
73.1	169.6	1.052	3.998	25	495.9	0.6536	2.863
73	169.8	1.049	3.910	20	619.9	0.4460	3.582
72.9	170.1	1.051	4.089	19	652.6	0.3835	3.745
72.8	170.3	1.061	4.118	18	688.8	0.3103	3.885
72.7	170.5	1.069	2.549×10^{-2}	17	729.3	0.2232	4.013
72.6	170.8	1.061	8.139×10^{-3}	16	774.9	0.1195	4.174
72.5	171.0	1.050	7.264	15.8	784.7	0.09693	4.161
72.4	171.3	1.044	6.789	15.6	794.8	0.07327	4.081
72.3	171.5	1.040	6.871	15.4	805.1	0.04769	3.941
72.2	171.7	1.037	6.940	15.2	815.7	0.02019	3.855
72.1	172.0	1.035	6.949	15.0	826.6	-0.007825	3.834
72	172.2	1.033	7.042	14.8	837.7	-0.03727	3.613
71	174.6	1.020	6.982	14.6	849.2	-0.07038	3.644
70	177.1	1.012	7.086	14.4	861.0	-0.1033	3.802
68	182.3	1.002	7.339	14.2	873.1	-0.1365	3.928
66	187.9	0.9940	7.776	14.0	885.6	-0.1712	4.011
64	193.7	0.9876	8.204	13.5	918.4	-0.2656	4.229
62	200.0	0.9810	8.457	13.0	953.7	-0.3701	4.588
60	206.6	0.9746	8.709	12.5	991.9	-0.4887	4.814
eV	μm	ϵ_1	ϵ_2	eV	μm	ϵ_1	ϵ_2
12.0	0.1033	-0.6241	5.190×10^{-2}	1.90	0.6526	-58.96	23.28
11.5	0.1078	-0.7794	5.854	1.85	0.6702	-61.65	25.72

11.0	0.1127	−0.9562	6.971×10^{-2}	1.80	0.6888	−64.29	28.57
10.5	0.1181	−1.157	8.517×10^{-2}	1.75	0.7085	−66.68	32.00
10.0	0.1240	−1.386	0.1042	1.70	0.7293	−68.91	36.74
9.5	0.1305	−1.651	0.1273	1.65	0.7514	−68.52	41.61
9.0	0.1378	−1.962	0.1560	1.60	0.7749	−67.03	45.13
8.5	0.1459	−2.327	0.1922	1.58	0.7847	−66.27	46.67
8.0	0.1550	−2.762	0.2381	1.56	0.7948	−64.31	47.38
7.5	0.1653	−3.285	0.2974	1.54	0.8051	−62.95	47.06
7.0	0.1771	−3.923	0.3753	1.52	0.8157	−62.05	46.39
6.5	0.1907	−4.711	0.4791	1.50	0.8266	−61.50	45.61
6.0	0.2066	−5.700	0.6208	1.48	0.8377	−60.96	44.56
5.5	0.2254	−6.965	0.8192	1.46	0.8492	−60.71	43.08
5.0	0.2480	−8.617	1.120	1.44	0.8610	−60.94	41.51
4.5	0.2755	−10.83	1.538	1.42	0.8731	−61.07	39.28
4.0	0.3100	−13.90	2.203	1.40	0.8856	−62.42	36.74
3.8	0.3263	−15.47	2.575	1.38	0.8984	−64.45	34.50
3.6	0.3444	−17.29	3.035	1.36	0.9117	−66.70	32.54
3.4	0.3647	−19.42	3.606	1.34	0.9253	−69.01	30.03
3.2	0.3875	−21.95	4.327	1.32	0.9393	−72.93	27.48
3.0	0.4133	−24.97	5.258	1.30	0.9537	−77.92	26.28
2.9	0.4275	−26.71	5.800	1.28	0.9686	−82.27	26.11
2.8	0.4428	−28.64	6.440	1.26	0.9840	−86.20	26.10
2.7	0.4592	−30.78	7.188	1.24	0.9999	−89.97	25.92
2.6	0.4769	−33.15	8.066	1.22	1.016	−93.97	25.47
2.5	0.4959	−35.81	9.111	1.20	1.033	−98.61	25.23
2.4	0.5166	−38.79	10.38	1.15	1.078	−110.5	25.66
2.3	0.5391	−42.13	11.96	1.10	1.127	−123.6	26.86
2.2	0.5636	−45.83	13.94	1.05	1.181	−137.7	28.45
2.1	0.5904	−49.86	16.43	1.00	1.240	−153.9	30.21
2.0	0.6199	−54.24	19.50	0.95	1.305	−172.8	32.45
1.95	0.6358	−56.52	21.25	0.90	1.378	−195.0	35.45

(continued)

TABLE XIII (Continued)

Metallic Aluminum

eV	μm	ε_1	ε_2	eV	μm	ε_1	ε_2
0.85	1.459	-221.5	39.82	0.23	5.391	-2,622	994.9
0.80	1.550	-252.5	46.07	0.22	5.636	-2,845	1,104
0.75	1.653	-289.0	54.33	0.21	5.904	-3,096	1,231
0.70	1.771	-332.8	64.88	0.20	6.199	-3,387	1,393
0.65	1.907	-386.4	78.53	0.19	6.526	-3,703	1,578
0.60	2.066	-452.9	97.30	0.18	6.888	-4,065	1,791
0.55	2.254	-536.6	122.2	0.17	7.293	-4,512	2,054
0.50	2.480	-645.0	157.2	0.16	7.749	-5,016	2,407
0.45	2.755	-785.7	208.3	0.15	8.266	-5,578	2,859
0.40	3.100	-971.5	280.4	0.14	8.856	-6,205	3,410
0.39	3.179	-1,018	296.8	0.13	9.537	-6,930	4,065
0.38	3.263	-1,068	315.8	0.12	10.33	-7,787	4,899
0.37	3.351	-1,122	337.7	0.11	11.27	-8,769	5,940
0.36	3.444	-1,178	361.8	0.10	12.40	-9,964	7,279
0.35	3.542	-1,238	387.1	0.095	13.05	-10,720	8,080
0.34	3.647	-1,302	413.6	0.090	13.78	-11,450	9,049
0.33	3.757	-1,374	441.6	0.085	14.59	-12,260	10,160
0.32	3.875	-1,454	473.8	0.080	15.50	-13,210	11,380
0.31	4.000	-1,540	511.5	0.075	16.53	-14,290	12,860
0.30	4.133	-1,632	553.7	0.070	17.71	-15,470	14,580
0.29	4.275	-1,731	598.7	0.065	19.07	-16,990	16,420
0.28	4.428	-1,841	647.0	0.060	20.66	-18,790	18,960
0.27	4.592	-1,964	701.3	0.055	22.54	-20,840	21,810
0.26	4.769	-2,098	760.6	0.050	24.80	-24,030	25,830
0.25	4.959	-2,253	828.3	0.045	27.55	-27,580	31,590
0.24	5.166	-2,424	905.2	0.040	31.00	-31,770	40,170