

7-1970

Low-Energy Optical Absorption Peak in Aluminum and Al-Mg Alloys

L. W. Bos
Iowa State University

David W. Lynch
Iowa State University, dlynch@iastate.edu

Follow this and additional works at: http://lib.dr.iastate.edu/physastro_pubs

 Part of the [Atomic, Molecular and Optical Physics Commons](#)

The complete bibliographic information for this item can be found at http://lib.dr.iastate.edu/physastro_pubs/24. For information on how to cite this item, please visit <http://lib.dr.iastate.edu/howtocite.html>.

This Article is brought to you for free and open access by the Physics and Astronomy at Iowa State University Digital Repository. It has been accepted for inclusion in Physics and Astronomy Publications by an authorized administrator of Iowa State University Digital Repository. For more information, please contact digirep@iastate.edu.

Low-Energy Optical Absorption Peak in Aluminum and Al-Mg Alloys

Abstract

Measurements of the absorptivity of Al and dilute Al-Mg alloys were made at 4.2 K in the 0.2- to 5-eV energy region by a calorimetric technique. In addition to the well-known conductivity peak around 1.6 eV, another peak has been found near 0.5 eV. These peaks are present in the alloys, broadened, but unshifted, as well as in Al. The peaks are at about $2|V_{200}|$ and $2|V_{111}|$, respectively, where the V 's are Fourier coefficients of the pseudopotential.

Keywords

pseudopotential, conductivity, Ames Laboratory

Disciplines

Atomic, Molecular and Optical Physics | Physics

Comments

This article is from *Physical Review Letters* 25 (1970): 156, doi:[10.1103/PhysRevLett.25.156](https://doi.org/10.1103/PhysRevLett.25.156). Posted with permission.

LOW-ENERGY OPTICAL ABSORPTION PEAK IN ALUMINUM AND Al-Mg ALLOYS*

L. W. Bos† and D. W. Lynch

Department of Physics and Institute for Atomic Research, Iowa State University, Ames, Iowa 50010

(Received 14 May 1970)

Measurements of the absorptivity of Al and dilute Al-Mg alloys were made at 4.2 K in the 0.2- to 5-eV energy region by a calorimetric technique. In addition to the well-known conductivity peak around 1.6 eV, another peak has been found near 0.5 eV. These peaks are present in the alloys, broadened, but unshifted, as well as in Al. The peaks are at about $2|V_{200}|$ and $2|V_{111}|$, respectively, where the V 's are Fourier coefficients of the pseudopotential.

The optical absorption of aluminum is rather featureless in the region of interband absorption. The only reported structure¹ is a reflectivity minimum at 1.5 eV, although suspicions of an absorption peak at lower energy have been reported in some experimental work.²⁻⁵ The strength of the 1.5-eV absorption peak was once a controversial subject,⁶ but recent calculations agree well with experiment.⁷⁻¹⁰ This peak was originally attributed to transitions near W in the Brillouin zone,¹¹ but now it appears to be the result of transitions occurring in a larger volume of the zone, near, but not including W . Harrison¹² first pointed out that for many polyvalent metals, a pseudopotential approach leads to parallel bands, partly empty, partly occupied, throughout appreciable regions of k space. Such bands lead to a sharp edge in the conductivity at a photon energy of $2|V_G|$, where V_G is a Fourier coefficient of the pseudopotential. At higher energy, each edge is followed by a decaying tail. Golovashkin, Kopeliovich, and Motulevich¹³ carried out similar calculations. Any pseudopotential calculation of the band structure for Al should give bands that will yield roughly the above results when the optical properties are calculated from them. Recent calculations,⁷⁻¹⁰ including calculated dipole matrix elements, predict two absorption peaks, corresponding to $2|V_{200}|$ (~ 1.5 eV) and $2|V_{111}|$ (~ 0.5 eV). The lower energy peak has not been reported previously. The high-energy peak has recently been remeasured.⁸

We have measured the absorptivity of Al and several alloys of Al with Mg at 4.2 K using a calorimetric method.^{14-16, 4} The aluminum was a 99.999% pure Cominco polycrystalline bar. The alloys were produced by arc melting. The samples were spark cut, mechanically polished, electropolished, annealed in Ar at 550 C for at least 100 h, then electropolished again just before use. The samples were exposed to air for less than 2 h, and within another 3 h were at a pressure of 10^{-6} Torr or less. Data were taken

by a method similar to that of Biondi,^{4, 15, 16} but with higher sensitivity and resolution. A prism double monochromator was used to ensure high spectral purity in the infrared. Since structure in the 1.5-eV absorptivity peak has been reported⁸ for pure Al, points were taken every 0.02 eV (the spectral bandpass) in that region. The errors in the measurements of the absorptivity are estimated to be 2% in the 0.5- to 3-eV region, rising to 10% at 0.2 eV, not including any errors due to imperfect sample surfaces.

Figure 1 shows the absorptivities of Al and three alloys of Al with Mg. The absolute values of the absorptivities in this figure are difficult to guarantee. Several samples with noticeably poorer electropolished surfaces yielded absorptivity spectra like those shown in Fig. 1, but shifted upward by about 0.01 in the infrared and 0.02 around 3 eV. The structure near 0.5 eV was still discernible in these samples. Better surfaces might yield still lower absorptivities in the infrared. However, the absorptivities for pure Al shown in Fig. 1 are lower than any reported to date,⁴ indicating somewhat better surfaces on our samples.

A Kramers-Kronig inversion of the absorptivity was made to obtain the optical conductivity. Above 5 eV, published data¹⁷ for Al were used, while for the alloys above 3 eV, we used the Al data multiplied by a factor between 0.8 and 1 to make a smooth fit with the alloy data at 3 eV. From 0 to 0.05 eV the absorptivity of a free electron gas was used, and a smooth monotonic curve was drawn between 0.05 and 0.2 eV to connect to the data. Changes in the low-energy extrapolation had little effect on the computed conductivity, but changes in the high-energy extrapolation had a large effect on the magnitude, but not the shape, of the conductivity between 0.2 and 3 eV. The alloy data are particularly sensitive here because their poorer surfaces produced errors in the absorptivity. The conductivity spectra have only qualitative significance for the alloys, but that

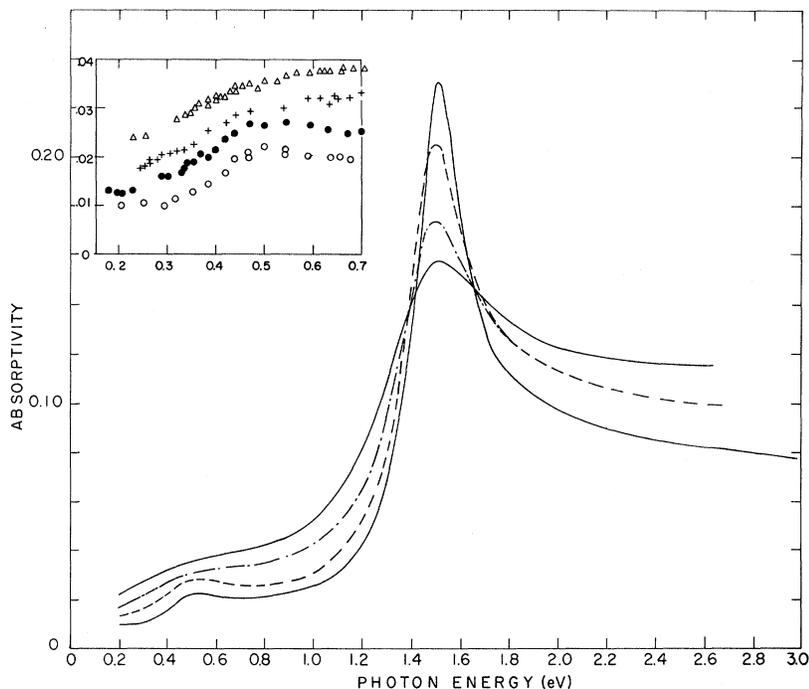


FIG. 1. Absorptivity at 4 K versus photon energy. Solid curve (lowest at low energy), Al; dashed curve, Al + 1.0% Mg; dot-dashed curve, Al + 2.5% Mg; solid curve, Al + 5.5% Mg. The inset shows the low-energy data on an expanded scale.

for Al should be reliable.

It would seem that for pure Al at 4 K such a dispersion analysis should not be correct. A small piece was cut from one Al sample used for optical measurements. It had a resistivity ratio of 5000, indicating that one cannot assume a local conductivity,¹⁸ but instead, one must analyze the absorptivity using the theory of the anomalous skin effect, including interband transitions, to obtain the (local) interband contribu-

tion to the conductivity. A Drude contribution to the conductivity for pure Al is thus meaningless, but both interband peaks should be about correct because the 1.6-eV peak agrees with room-temperature data on Al, free of nonlocality problems, and both peaks persist in Al-Mg alloys, similarly free of such problems.

The interband contribution to the conductivity is shown in Fig. 2 for Al and Al + 5.5% Mg. In order to make the low-energy peak more visible,

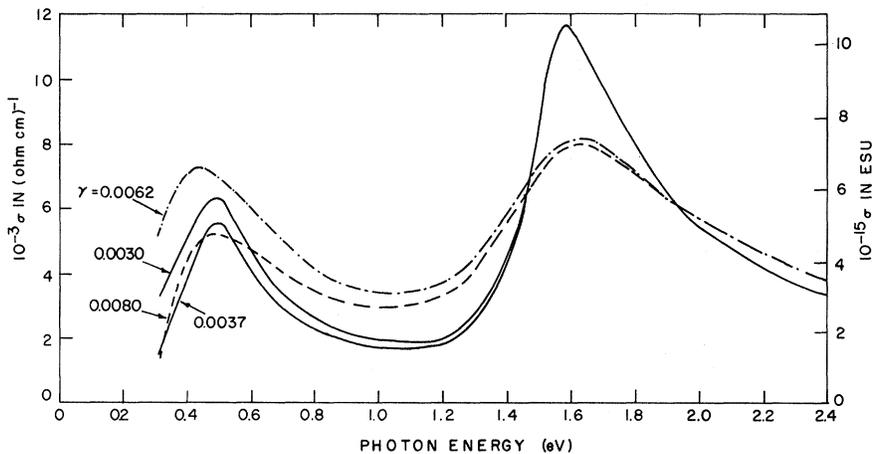


FIG. 2. Interband conductivity spectrum of Al (solid curve) and Al + 5.5% Mg (dashed curve) obtained by Kramers-Kronig inversion. The Drude term subtracted from the total conductivity was characterized by $\hbar\omega_p = 12.7$ eV and by the various values of $\gamma = (\omega_p\tau)^{-1}$ shown.

a Drude term has been subtracted from the total conductivity. Because interband absorption contributes everywhere in our data range and because of the anomalous skin effect, there is no way to obtain the Drude parameters from our data. For both samples we used $\hbar\omega_p = 12.7$ eV. For Al we used $(\omega_p \tau)^{-1} = \gamma = 0.0035$ and 0.0030 , while for the alloy, γ ranges from 0.0062 to 0.008 , the former value being that expected from the dc conductivity.¹⁹

It is clear that the main peak in Al seems structure free except for possible slope changes at 1.40 and 1.65 eV (better detected on plots with different scales). A recent set of measurements⁸ by an ac technique²⁰ reports structure on the low-energy side of the 1.6 -eV peak, structure large enough to be discernible in our data, but which is not found. The reason for the disagreement is not clear. It is either an artifact of the ac method or apparatus, or the result of inferior surfaces in our measurements.

The new peak at 0.50 eV has been predicted a number of times,⁷⁻¹⁰ but not observed before. According to Harrison's model, there should be sharp rises in the conductivity at $\hbar\omega = 2|V_G|$. Ashcroft's values²¹ of $|V_G|$, which yield a Fermi surface in substantial agreement with experiment, give $2|V_{200}| = 1.52$ eV and $2|V_{111}| = 0.485$ eV. The conductivity should then fall off as $\omega^{-1}(\hbar\omega - 2|V_G|)^{-1/2}$ beyond the sharp rise.¹³ Dresselhaus⁸ and Brust^{9,10} have recently calculated the conductivity more realistically, using the above pseudopotential coefficients. Their peak values of the conductivity are in substantial agreement with those of Fig. 2. They point out that the low-energy peak does not rise at $2|V_{111}|$ but begins at zero photon energy, a result of the crossing of two bands at the Fermi level on certain planes in the Brillouin zone. This low-energy absorption must be considered when obtaining the electron effective mass from infrared data on Al, as recognized by Golovashkin, Motulevich, and Shubin² and by Gurzhi and Motulevich.³

Adding Mg, which simply lowers the Fermi energy according to a rigid-band model, has little effect on the positions of the peaks. The peaks simply broaden and become lower, an effect of the increased scattering in the alloys. Thus, the two values of $|V_G|$ change very little. A shift in the Fermi level is not expected to alter the position of the conductivity peak, because a lower Fermi level only makes the regions in k space where transitions occur at a particular

photon energy move slightly. Since the bands are nearly parallel in both regions, no appreciable change in the conductivity results.

Measurements further into the infrared are in progress, and a more detailed separation of the absorptivity into free-carrier and interband terms will be reported later. We wish to acknowledge profitable discussions and correspondence with Dr. D. Brust, Dr. G. Dresselhaus, and Professor K. L. Kliewer, and the sample preparation by H. H. Baker and F. A. Schmidt.

*Work performed at the Ames Laboratory of the U. S. Atomic Energy Commission.

†Present address: Western Electric Company, Engineering Research Center, Box 900, Princeton, N. J. 08540.

¹H. E. Bennett, M. Silver, and E. J. Ashley, *J. Opt. Soc. Amer.* **53**, 1089 (1963).

²A. I. Golovashkin, G. P. Motulevich, and A. A. Shubin, *Zh. Eksp. Teor. Fiz.* **38**, 51 (1960) [*Sov. Phys. JETP* **11**, 38 (1960)].

³R. N. Gurzhi and G. P. Motulevich, *Zh. Eksp. Teor. Fiz.* **51**, 1220 (1966) [*Sov. Phys. JETP* **24**, 818 (1967)].

⁴M. A. Biondi and A. I. Guobadia, *Phys. Rev.* **166**, 667 (1968).

⁵C. J. Powell, *J. Opt. Soc. Amer.* **60**, 78 (1970).

⁶J. C. Phillips, *Solid State Phys.* **18**, 55 (1966).

⁷A. J. Hughes, D. Jones, and A. H. Lettington, *J. Phys. C: Proc. Phys. Soc., London* **2**, 102 (1969).

⁸G. Dresselhaus, M. S. Dresselhaus, and D. Beaglehole, in *Proceedings of the Symposium on the Electronic Density of States*, Washington, D. C., 3-6 November 1969 (to be published).

⁹D. Brust, *Phys. Lett.* **31A**, 289 (1970).

¹⁰D. Brust, *Phys. Rev. B* (to be published).

¹¹E. Ehrenreich, H. R. Philipp, and B. Segall, *Phys. Rev.* **132**, 1918 (1963).

¹²W. A. Harrison, *Phys. Rev.* **147**, 467 (1966).

¹³A. I. Golovashkin, A. I. Kopeliovich, and G. P. Motulevich, *Zh. Eksp. Teor. Fiz.* **53**, 2053 (1967) [*Sov. Phys. JETP* **26**, 1161 (1968)].

¹⁴K. G. Ramanathan, *Proc. Phys. Soc. London, Sect. A* **65**, 532 (1952).

¹⁵M. A. Biondi, *Phys. Rev.* **102**, 964 (1956).

¹⁶M. A. Biondi and J. A. Rayne, *Phys. Rev.* **115**, 1522 (1959).

¹⁷R. P. Madden, L. R. Canfield, and G. Hass, *J. Opt. Soc. Amer.* **53**, 620 (1963).

¹⁸G. E. H. Reuter and E. H. Sondheimer, *Proc. Roy. Soc., Ser. A* **195**, 336 (1948); R. B. Dingle, *Physica* **19**, 311 (1953); R. Fuchs and K. L. Kliewer, to be published.

¹⁹A. T. Robinson and J. E. Dorn, *J. Metals* **3**, 457 (1951).

²⁰D. Beaglehole, *Appl. Opt.* **7**, 2218 (1968).

²¹N. W. Ashcroft, *Phil. Mag.* **8**, 2055 (1963).