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I. INTRODUCTION

W. E. Spicer

In Part II of this report, a summary is given of the method developed to measure electron-phonon scattering in insulators by photoinjection. This is closely related to prevent charging in insulating films by photoinjecting electrons into the films from a metal substrate while photocurrent is being drawn from the insulator. These studies of alkali halides are part of the over-all program to study alkali halides and their alloys.

In the third section of the report, a description is given of new techniques developed in the Kramers-Krönig analysis of optical reflectivity data. This work was done as part of the study of the silver halides.

II. HOT ELECTRON TRANSPORT IN THE ALKALI HALIDES: LITHIUM FLUORIDE

T. H. DiStefano

Hot electron scattering is being studied in the alkali halides by the technique of photoejection.^{1,2} In the photoejection experiment, developed by the author, electrons are photoemitted from the substrate of an insulating thin film, through the film itself, and into vacuum. The scattering properties of the film can be determined from the energy distribution of those electrons which have propagated through the film.

The major scattering mechanism in the alkali halides is due to interaction with the longitudinal optical phonon. Since the energy gain or loss in each collision is small, the motion of the electron is well modeled by the random walk process. For more than a few scattering events, the random walk reduces to diffusion with a characteristic length, λ , which is $1/\sqrt{3}$ times the average path length l .³

The energy of the longitudinal optical phonon is largely independent of the phonon wavelength in the alkali halides. Thus, in each collision the electron will either gain or lose energy $\hbar\omega_p$. The probability of energy gain and of energy loss in each scattering event is,

$$\text{Prob. (gain)} = \frac{n}{2n + 1} \quad (\text{phonon absorption})$$

$$\text{Prob. (loss)} = \frac{n + 1}{2n + 1} \quad (\text{phonon emission})$$

$$\text{where, } n = \frac{1}{e^{-\hbar\omega_p/KT} - 1}$$

On the average, a stream of electrons moving through the crystal will lose energy Δe in each collision:

$$\Delta e = \frac{\hbar\omega_p}{2n + 1} .$$

Since the hot electron motion is essentially a random walk, the average number of these collisions made in diffusing a distance L is $N(L)$:

$$N(L) = (L/\lambda)^2 .$$

And, by this elementary model, the average energy loss suffered by an electron beam in traversing a crystal of length L is ΔE . In the light alkali halides at room temperature there is little chance of phonon absorption, simply because there are not many excited phonon modes in the crystal. At these temperatures there is a near certainty of a loss of energy $\hbar\omega_p$ per collision, which corresponds to the case for which $n = 0$.

Now, it is seen that if the energy loss of a stream of hot electrons were measured, then the scattering length of electrons of that particular energy could be determined. The photoinjection experiment, which was designed to measure this loss of energy, has been tried and preliminary results have been obtained for the case of LiF.

Electron transport in LiF was studied by analyzing the energy distribution of a beam of electrons which has propagated through a thin film of the material. Hot electrons were photoemitted from the substrate of the film into the film itself; the energy distribution of the electrons transmitted through the film and into vacuum were measured by the standard photoemission techniques.⁴ Photoexcitation in the substrate was produced by light which has propagated through the alkali halide at less than the band gap energy.

The highest energy photon which will pass through an insulator of band energy E_g is, of course, E_g . These photons can excite electrons in the metallic substrate to an energy of E_g above the Fermi energy. For most

metal-insulator interfaces, the conduction band of the insulator lies about four volts above the Fermi level in the metal. Thus, this technique can inject electrons into the insulator with kinetic energies ranging from zero to $E_g - 4$ eV. For a film of CsI on copper the yield is similar to that of the copper below the band gap of the CsI; however, the threshold of the copper has been lowered by the CsI coating which causes it to have a higher yield than the yield for pure copper. At Photon energies greater than the threshold of 6.5 eV for the CsI, photoemission from the CsI itself dominated the yield.

By a careful selection of the substrate material, the stream of injected electrons has been made to be quite monoenergetic. This was accomplished by exciting electrons from just below the Fermi level to the conduction band states of the metal substrate. Yttrium was chosen for its exceptionally high density of filled $4d^1$ states, as well as for its low work function. Eastman has found⁵ that electrons photoemitted from this band have an energy spread of only .4 eV, which is narrow enough to provide an excellently monoenergetic source of electrons.

Initially, there was some difficulty in obtaining a clean surface of yttrium because of its high chemical activity and its affinity for hydrogen. However, any metal with the low work function necessary to inject electrons high into the conduction band of the insulator deposited on itself would be very electropositive, and hence chemically active.

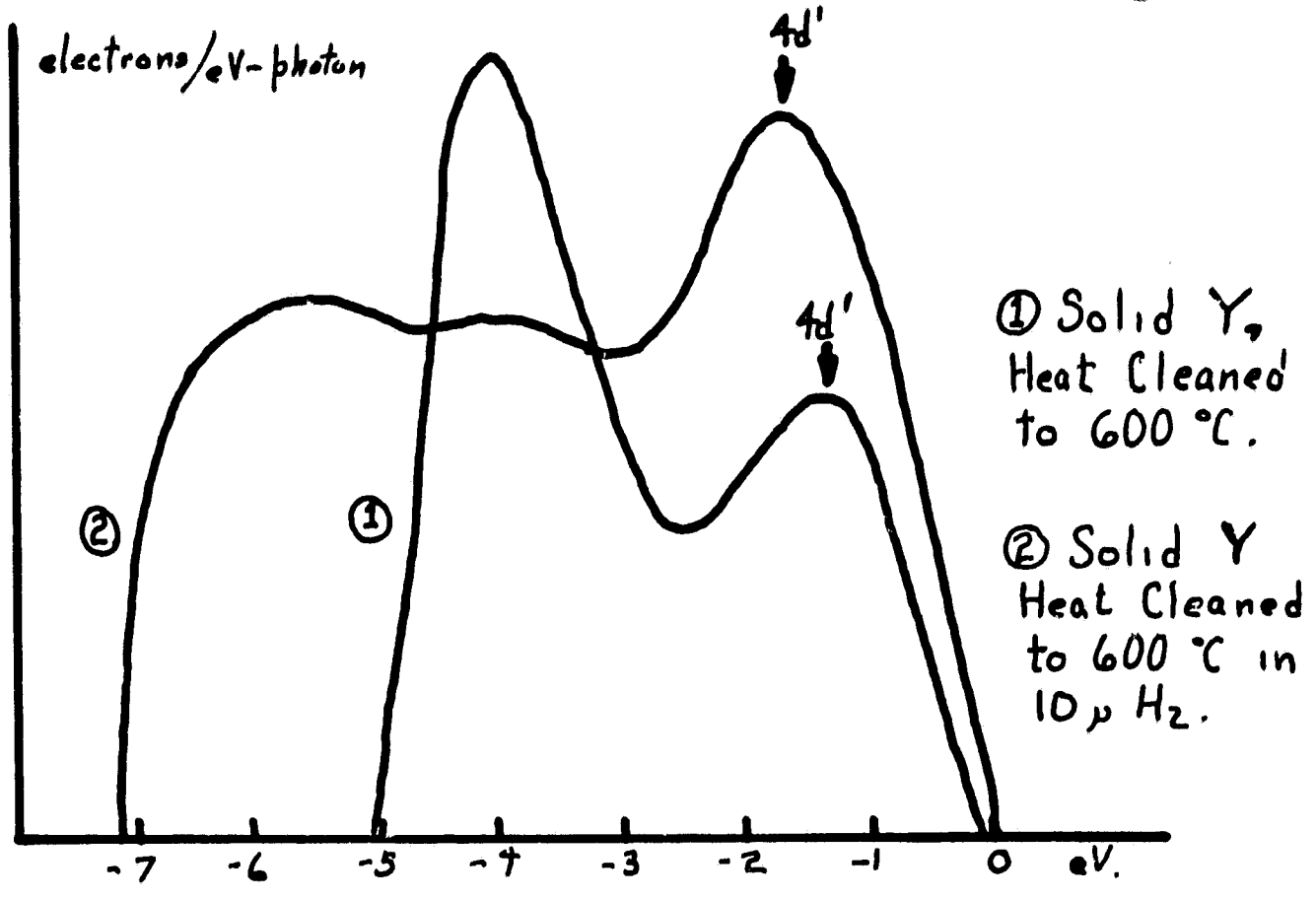
A solid substrate of yttrium was tried and found to be unsatisfactory. This substrate was polished with 20% anhydrous nitric acid in glycerine, suspended in kerosene; the more usual polishing techniques⁶ were unsuccessful. Transfer to the photoemission apparatus was done in dry nitrogen. The solid sample yielded abysmally poor results after being heat cleaned in a vacuum

of 10^{-9} Torr. After being heat cleaned in 10 microns of hydrogen and then heat cleaned in vacuum, this solid sample gave the poor photoemission results shown in Fig. 1.

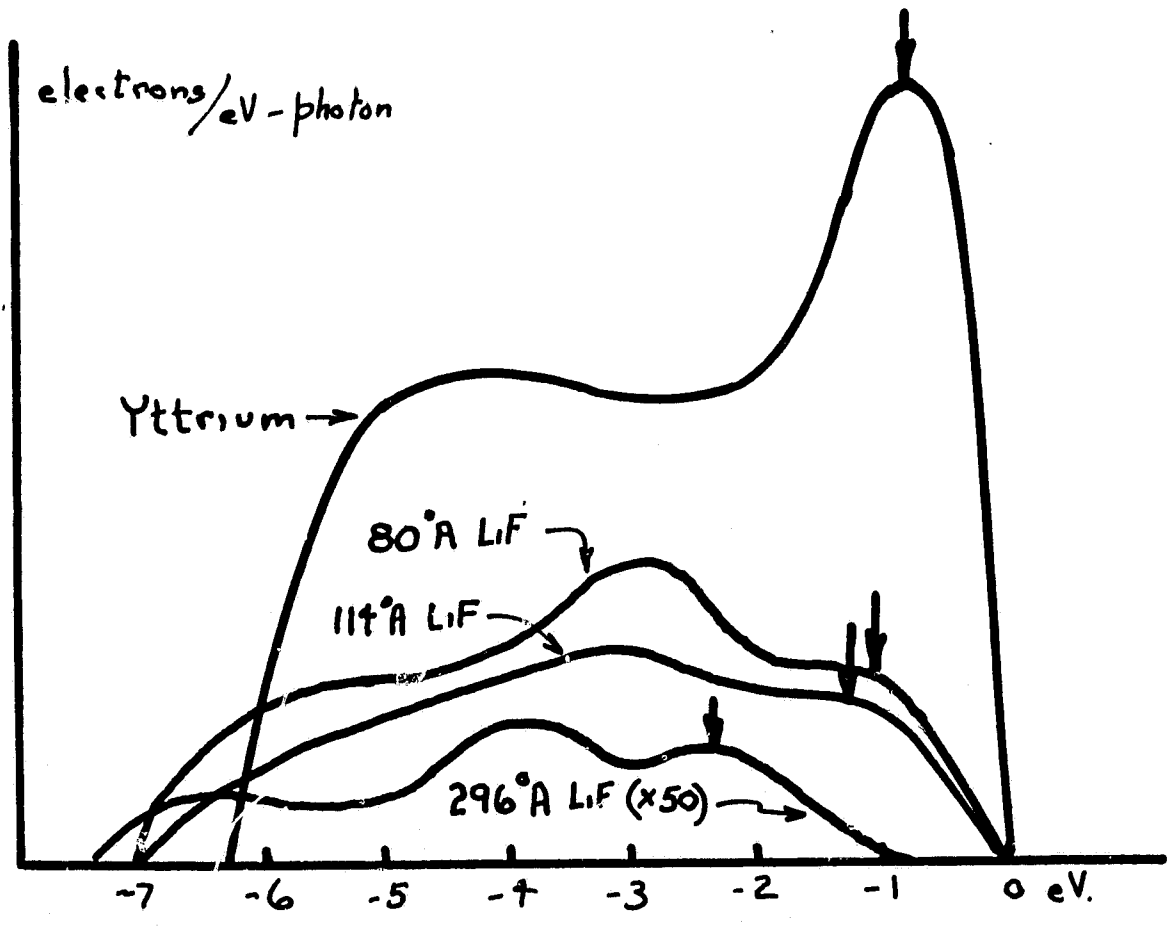
Vacuum evaporated yttrium yielded photoemission energy distribution curves which were comparable to those of Eastman. The yttrium was evaporated at 1150°C from a pre-fired tantalum boat. At this temperature the tantalum acts as a getter, as well as being completely insoluble in the yttrium.⁷ The results shown in Fig. 1 indicate that this yttrium provides a well-resolved peak of injected electrons, with a width of less than a volt. This peak from the yttrium is identifiable in the energy distributions of electrons photoinjected through alkali halide thin films. In the preliminary results for LiF, using yttrium as a monochromatic source, the scattering lengths found vary from 95 \AA to 120 \AA for electrons with kinetic energies in the range 5 eV to 10 eV.

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Photoemission from Solid Yttrium



Photoemission Through LiF Films on Clean Yttrium

Fig. 1

III. EFFECTS OF THE EXTRAPOLATION OF THE REFLECTANCE
ON THE OPTICAL CONSTANTS DETERMINED BY THE
KRAMERS-KRÖNIG ANALYSIS

Robert S. Bauer

A. Introduction

If the complex normal incidence reflectivity $r = |r|e^{i\theta}$ is known, then all the optical properties can be determined from the Fresnel equation and the definitions of the constants. Further, if the reflectance $R = |r|^2$ is known at all photon energies E then the phase of the reflectivity at energy E_0 is determined uniquely by the Kramers-Krönig transform¹

$$\theta(E_0) = \frac{1}{\pi} \int_0^{\infty} \frac{d \ln |r|}{dE} \ln \left| \frac{E + E_0}{E - E_0} \right| dE. \quad (1)$$

Since the reflectance cannot be measured over an infinite photon energy range, the data must be extrapolated in some manner in the unknown regions. The necessity of using an extrapolation of the data requires an examination of the uniqueness of the calculated optical constants in the region of measurement.

This question has been studied by calculations of θ using the reflectance data of White and Straley² for AgCl. The Kramers-Krönig integral, Eq. (1), was performed using this laboratory's computer program³ which was modified to allow for the calculation to be performed using a high energy extrapolation which leads to the best agreement between the calculated low energy optical constants and known values in this region. In these calculations on AgCl, the phase shift was fit to zero in the transparent region (0 through 3.0 eV). Though calculated for a specific set of data and

fitting criterion, the results reported below should be characteristic of the calculation as performed using this laboratory's computer program.³

B. High Energy Extrapolation

The integral of Eq. (1) is performed by considering the reflectance as plotted on a log-log scale and straight lines drawn between the data points.⁴ The solution for the contribution to θ from each of these straight lines is known exactly.⁴ Thus, a natural extrapolation to high energies which is continuous with the last measured value is

$$R = R_f \left(\frac{E}{E_f}\right)^{-A}, \quad E_f \leq E \leq E_m \quad (2a)$$

where R_f is the reflectance at the highest energy of measurement, E_f . Being a straight line on a log-log scale, this extrapolation is uniquely specified by one point, R_m , calculated using Eq. (2a) at energy E_m .

In performing the calculation of $\theta(E_0)$, E_m will be the largest energy in the numerical integration. Thus, in the calculation we are implicitly assuming that the contribution to θ for energies greater than E_m is zero. From inspection of Eq. (1) it is seen that this is equivalent to having assumed a constant reflectivity from E_m to infinity. Therefore, the numerical method inherently assumes

$$R = R_f \left(\frac{E_m}{E_f}\right)^{-A} = R_m = \text{constant}, \quad E \geq E_m. \quad (2b)$$

There are thus two adjustable parameters in the choice of such an extrapolation: A and E_m . It is found that variation of either of these quantities while the other is held fixed affects the magnitude but not the position of optical constant structure in the region of measurement. One

criterion that can be used in choosing the parameters is to obtain a fit of the calculated optical constants at low photon energies to known values. If A is chosen to satisfy this fitting criterion, then it is found that different choices of E_m will lead to different "optimum" values for A. For the AgCl data used² in which $E_f = 21.1$ eV, the absorption coefficient α is the same for all pairs of E_m and corresponding "optimum" A at photon energies below 15 eV if E_m is reasonably (at least 50%) larger than E_f .

To obtain a more accurate knowledge of the optical constants in the high energy region of measurement, a second criterion must be used in choosing the extrapolation.⁵ If the measured R is decreasing at high energies, a reasonable choice is to require that the slope of the reflectance be continuous at E_f . Using this criterion, the error in the optical constants can be estimated for a given R slope discontinuity. It is found that for all slope discontinuities

$$\Delta\alpha_f \approx \frac{1}{30} \Delta S \quad (3)$$

where $\Delta\alpha_f$ is the percent deviation of α at E_f from the corresponding value calculated with reflectance having a continuous slope at E_f and ΔS is the percent discontinuity in the slope of the reflectance at E_f .

The error of the optical constants is a maximum at E_f . It is found that the error in α at an energy E is given approximately by

$$\Delta\alpha \approx \Delta\alpha_f \left(\frac{E}{E_f}\right)^{8.6}, \quad E \leq E_f \quad (4)$$

As can be seen from Eq. (2a), the slope of the extrapolation at E_f depends only on A. Thus, using our second criterion of R slope continuity, A can be chosen unambiguously. Then E_m can be varied to satisfy the fitting

criterion at low photon energies. The computer program used for these calculations is currently being modified to perform this. In addition, the high energy errors of the other optical constants are being studied.

C. Conclusions

Except for energies near the end of the measurement range, the optical constants calculated by a Kramers-Krönig analysis of a set of reflectance data are insensitive to the extrapolation of the data to high photon energies when the extrapolation is chosen to make a few low energy calculated optical constants agree with known values. This very significant conclusion is further substantiated by calculations of the optical constants of amorphous Ge by T. M. Donovan using this program.⁶ By fitting θ to zero over the very narrow transparent region, the resulting value for the index of refraction at zero frequency was well within one percent of the measured value for amorphous Ge. In addition, the calculated ϵ_2 at the absorption edge fit the measured values quite well. Because of the reliability of the optical constants calculated in this manner, the Kramers-Krönig transform is a useful check of the measured reflectance data. In the case of AgCl,⁷ low photon energy negative dips in the calculated α , k , and ϵ_2 lead to an examination of the reflectance data in the corresponding energy region. It was found that by "smoothing" R in accordance with earlier measurements in this region, the calculated optical constants no longer exhibited these unphysical values. Thus, the Kramers-Krönig analysis revealed an error in the experimental data.

By requiring the extrapolation to have the same slope as the measured reflectance at the highest energy of measurement, an estimate of the error of the optical constants in this energy region is obtained. In addition, the extrapolation can be chosen unambiguously yielding more reliable high energy optical constants.

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5. It should be noted that since the optical constants depend on the energy that specifies the extrapolation of the data, E_m , the form of the extrapolation does not have any physical significance.
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