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THE OPTICAL ANALYSIS OF PHOTOEMISSION

by Stephen V. Pepper
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Cleveland, Ohio

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • JANUARY 1970

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1. Report No.
NASA TN D-5610

2. Government Accession No.

3. Recipient's Catalog No.

4. Title and Subtitle
THE OPTICAL ANALYSIS OF PHOTOEMISSION

5. Report Date
January 1970

6. Performing Organization Code

7. Author(s)
Stephen V. Pepper

8. Performing Organization Report No.
E-5321

9. Performing Organization Name and Address
Lewis Research Center
National Aeronautics and Space Administration
Cleveland, Ohio 44135

10. Work Unit No.
124-09

11. Contract or Grant No.

12. Sponsoring Agency Name and Address
National Aeronautics and Space Administration
Washington, D.C. 20546

13. Type of Report and Period Covered
Technical Note

14. Sponsoring Agency Code

15. Supplementary Notes

16. Abstract

The role of classical optics in the process of photoemission is analyzed. Rigorous expressions are obtained for the quantum yield of a uniaxially anisotropic thin film for arbitrary values of angle of incidence, film thickness and optical constants of the film. Some applications to emission from metal films are discussed.

17. Key Words *(Suggested by Author(s))*
Photoemission
Vacuum ultraviolet radiation
Anisotropic thin films
Photoelectric detection

18. Distribution Statement
Unclassified - unlimited

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price*
Unclassified	Unclassified	23	$3.00

*For sale by the Clearinghouse for Federal Scientific and Technical Information
Springfield, Virginia 22151

THE OPTICAL ANALYSIS OF PHOTOEMISSION

by Stephen V. Pepper

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SUMMARY

An analysis of the role of classical optics in the process of photoemission from thin films is presented. The generation of excited electrons throughout the film is assumed to be proportional to the divergence of the Poynting vector in the film. The film and substrate are allowed uniaxial optical anisotropy with the optic axis normal to the plane of the film. The main results of the analysis, expressions for the reflectance, transmittance, and divergence of the Poynting vector in the film, are valid for all angles of incidence and arbitrary values of the dielectric functions of the initial medium, film, and substrate. Expressions for the quantum yield are obtained with the aid of a simple function that describes the diffusion and escape of the excited electrons. Some applications to photoemission from metals in the vacuum ultraviolet region are discussed.

INTRODUCTION

It is now generally accepted that photoemission of electrons is a volumetric process. Electrons are excited in the solid beneath the irradiated surface and then diffuse to the surface, where some of them have sufficient energy to penetrate the potential barrier and escape. The electrons are emitted from either the irradiated surface or the one opposite to it in the case of a film type of emitter.

Since the electrons are excited by the electromagnetic field in the interior of the solid, a description of the absorption of the field is necessary for any quantitative theory of photoemission. The analysis of the absorption of the fields in the interior of the emitter constitutes the optical analysis of photoemission. For radiation obliquely incident on an optically thin film, the optical analysis can be rather complicated. For example, the presence of multiple reflections and standing waves in the interior of the film can cause wide departures from simple exponential decay of the fields. An optical analysis for thin films was discussed by Fry (ref. 1) almost 40 years ago in connection with the work of Ives and Briggs (ref. 2) on photoemission from thin films of the alkali metals. In this early work, Fry gave a complicated prescription for obtaining an approximate value for the energy density of the electromagnetic field at any point in the interior

of a metal (ref. 3) and the absorptance of thin metal films.

The analysis of photoemission from thin films, however, requires an analysis of the absorption of radiation throughout the interior of the film, and in this respect Fry's analysis is incomplete. Since there has recently been a revival of interest in photoemission from thin films, it is desirable to have a complete optical analysis available. This report presents a useful and rigorous optical analysis of photoemission from thin films for arbitrary angles of incidence and arbitrary values of the dielectric functions of the incident medium, photoemitter, and substrate. All multiple reflections of waves in the interior of the film are taken into account and wave interference is treated. The main results of the analysis - expressions for the absorption of radiation throughout the film - have the virtue that, besides being mathematically rigorous (within the domain of classical optics), all the terms have a direct physical interpretation.

The photoemitting medium and its substrate are allowed uniaxial optical anisotropy with the optic axis normal to the plane of the film. Thus, the results of the analysis may be useful in the study of epitaxial films and optically anisotropic single crystals. Since expressions for the reflectance and transmittance of anisotropic films are not generally known, they are also presented.

The diffusion and escape aspects of the photoemission analysis are treated by a simple model attributable to Spicer (refs. 4 and 5). Such a model, together with the optical analysis, is sufficient to serve as a phenomenological framework for the analysis of most photoemission experiments.

First, the photoemission process is formulated in precise terms and rigorous optical formulas are derived. Then, a specific example illustrating the utility of the optical formulation is discussed briefly.

FORMULATION OF THE MODEL

Consider first the excitation of electrons. The basic assumption made herein is that the number of electrons excited at any point in the solid is proportional to the number of photons absorbed per unit volume at that point. Denote the number of photons absorbed per unit volume at any point in the photoemitter divided by the number of photons incident per unit area of the irradiated surface as η. The quantity η is equal to the ratio of the negative of the divergence of the Poynting vector of the electromagnetic field in the photoemitter to the incident flux.

The quantum mechanical factor $\hbar\omega$ is divided out in the ratio of fluxes, and η is determined by only classical electromagnetic considerations. The negative sign is necessary because the divergence of the Poynting vector yields the energy flux out of a unit volume whereas η is defined as the flux into a unit volume. The function η is a purely

optical quantity that contains all the optical information necessary for the photoemission analysis, and explicit expressions will be obtained for it.

The expressions to be obtained for η may be used with any theory that describes the passage of the electrons from the point of excitation to the emitting surface. Since the purpose herein is to elucidate only the optics of photoemission, an elementary model (refs. 4 and 5) of the transport and escape processes will be used. Although the model is elementary, it has nevertheless proved quite capable of describing most features of the transport of excited electrons in solids and is widely used. For more detail in this area, the articles by Berglund and Spicer (ref. 6) should be consulted.

The probability that an electron excited at a depth y below the irradiated surface will reach the surface and enter the collecting medium is denoted by $C \exp(-y/L)$. The electron "attenuation length" or "escape depth" L is independent of y but depends on the energy of the photon and the different physical mechanisms that can deexcite the electron. Such mechanisms are, for example, inelastic collisions with photons and other electrons. The factor C is a lumped parameter that takes into account the probability of excitation of the electron into energy states above the vacuum level and the probability of penetration of the surface potential barrier. In principle, L and C may depend on the direction of the electric field at the point of excitation. Such "vectorial" effects are not considered here, since in most situations the direction of the electron is randomized soon after excitation. The elementary diffusion model thus assumes that the mean free path for randomizing collisions is small relative to the thickness of the film. For the present purposes both L and C will be regarded simply as empirical constants which characterize the behavior of excited electrons in a particular solid for a given incident photon energy.

The number of electrons emitted per incident photon Y from the illuminated surface of a film of thickness d is then given by integration over the depth of the film:

$$Y = C \int_0^d e^{-y/L} \eta(y)dy = CF(L) \tag{1}$$

The quantity Y is usually known as the quantum yield. The function F depends on the nonoptical part of the photoemission process only through the attenuation length L.

For emission from the surface opposite to that being illuminated, the quantum yield is

$$Y = C \int_0^d e^{-(d-y)/L} \eta(y)dy = C e^{-d/L} F'(L) \tag{2}$$

Obviously $F'(L) = F(-L)$ and only one function $F(L)$ needs to be calculated. Note, however, that since C is characteristic of a given interface, emission into the final medium generally requires a different C than emission into the initial medium.

Since there is often an increase in yield for oblique angles of incidence, it is convenient to define the "relative gain" for angle of incidence θ as

$$G(\theta) = \frac{Y(\theta)}{Y(\theta = 0)} = \frac{F(\theta)}{F(\theta = 0)} \tag{3}$$

Aside from the optical constants of the various media, the relative gain depends only on the electron attenuation length. Results presented herein show that besides being a useful method to obtain an increased yield, illumination from oblique angles might be used to determine L since G does not depend upon C. Such a possibility is pointed out in a particular example calculation.

ANALYSIS

In this section expressions for η and F (and also the thin film transmittance and reflectance) are obtained. In particular, a new form for the volumetric absorptance η, appropriate for the study of photoemission, is presented.

These quantities involve the Poynting vector of the electromagnetic field in the photoemitting medium. Although the theory of electromagnetic plane waves in absorbing media is discussed in many places (refs. 7 to 9), many differences in sign conventions and nomenclature have appeared. In the present case there is the additional complication of optical anisotropy. Thus, for the sake of clarity and completeness, a sketch of the derivation leading to the final expressions is presented. The general procedure and notation follow Stern (ref. 8), while the treatment of uniaxially anisotropic media is similar to that of Mosteller and Wooten (ref. 10).

To establish the notation, let monochromatic linearly polarized light of frequency ω be incident on a photoemitter of thickness d (see fig. 1). The reflectance and transmittance of the film are denoted by R and T, respectively. The plane of incidence is defined by the direction of the incident light and the normal to the surface of the film. Standard notation for the polarized fields in the three media is used: subscript s denotes quantities associated with waves whose electric vector is perpendicular to the plane of incidence, and subscript p denotes quantities associated with waves whose electric vector lies in the plane of incidence.

The initial medium 0 is assumed to be a nonabsorbing, homogeneous, isotropic dielectric characterized by a real dielectric function $\epsilon_0(\omega) = n_0^2(\omega)$, where $n_0(\omega)$ is the

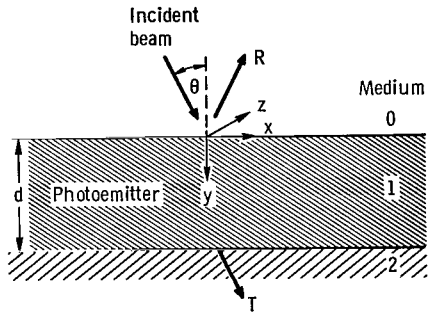

Figure 1. - Diagram of optical system under consideration.

index of refraction of the medium for radiation of frequency ω. Media 1 and 2 are assumed to be homogeneous, uniaxially anisotropic absorbing media with their axes of symmetry (optic axes) directed along the y-axis. With this choise of the optic axis the dielectric tensor is diagonal and has only two independent complex components ϵ_x and ϵ_y, which respectively describe the response of the media to electric fields parallel and perpendicular to the plane of the film. For isotropic media, $\epsilon_x = \epsilon_y$. These complex dielectric functions are given in terms of the index of refraction $n(\omega)$ and extinction coefficient $k(\omega)$ as $\epsilon(\omega) = [n(\omega) + ik(\omega)]^2$ for each x and y component and for each medium 1 and 2.

Where necessary, the real and imaginary parts of a complex quantity are denoted by primes and double primes, respectively. For example,

$$\epsilon = \mathcal{Re}\,\epsilon + i\,\mathcal{Im}\,\epsilon = \epsilon' + i\epsilon''$$

The complex dielectric functions for media 1 and 2 are allowed to depend on the frequency of the plane wave but not on its wave vector. Plasma phenomena associated with the free barriers of the media are thus incorporated in so far as they can be described by such a dielectric function. Although the optical properties of solid state plasmas have recently been discussed (refs. 8 and 11) in terms of dielectric functions depending on the propagation vector, the need for such a dependence to describe photoemission has not been established and is not discussed herein.

Let $\psi(\vec{r}, t)$ denote a component of one of the fields of the plane waves in one of the media

$$\psi(\vec{r}, t) = \psi^+ e^{i\vec{K}^+ \cdot \vec{r} - i\omega t} + \psi^- e^{i\vec{K}^- \cdot \vec{r} - i\omega t} \tag{4}$$

where ψ^+ and $\vec{K}^+$ (ψ^- and $\vec{K}^-$) denote, respectively, the complex amplitude and propagation vector of the waves propagating in the positive (negative) y-direction. For oblique

incidence $\vec{K}'$ is not parallel to $\vec{K}''$ in the absorbing media and the wave is termed inhomogeneous. The boundary conditions on the fields show that the x components of the propagation vectors of the plane waves in all the media are equal:

$$K_x^+ = K_x^- = \frac{2\pi}{\lambda_v} n_0 \sin(\theta) = \frac{\omega}{c} n_0 \sin(\theta) \tag{5}$$

where c is the velocity of light and λ_v is the vacuum wavelength of the incident radiation. The generalized law of reflection states that

$$K_y^+ = -K_y^- \equiv \xi \tag{6}$$

where the y component of the propagation vector of a positively propagating plane wave is denoted by ξ, the functional form of which is given subsequently.

The amplitudes of the electric and magnetic fields of the plane waves are governed by Maxwell's equations in the form

$$\vec{K} \times \vec{H} = -\omega \varepsilon_v \tilde{\epsilon} \cdot \vec{E} \tag{7}$$

$$\vec{K} \times \vec{E} = \omega \mu_v \vec{H} \tag{8}$$

where $\vec{E}$ and $\vec{H}$ are the amplitudes of the electric and magnetic fields, ε_v and μ_v are the permitivity and permeability of the vacuum ($\varepsilon_v \mu_v = c^{-2}$), and the media are assumed to be nonmagnetic ($\mu = \mu_v$). The quantity $\tilde{\epsilon}$ is the dielectric tensor, and the tensor product $\tilde{\epsilon} \cdot \vec{E}$ is a vector with components $\epsilon_x E_x$, $\epsilon_y E_y$, and $\epsilon_z E_z$. For the initial medium $\epsilon_x = \epsilon_y = \epsilon_z = \epsilon_0$ and for media 1 and 2, $\epsilon_x = \epsilon_z$.

The time-averaged Poynting vector in the various media and its divergence in medium 1 are given in terms of the complex amplitudes of the fields as

$$\vec{S}_j = \frac{1}{2} \mathcal{R}e\, (\vec{E}_j^* \times \vec{H}_j) \tag{9}$$

$$-\vec{\nabla} \cdot \vec{S}_1 = \frac{1}{2} \omega \varepsilon_v \epsilon_1'' \vec{E}_1^* \cdot \vec{E}_1 \tag{10}$$

where the subscript j refers to the three media and the star superscript denotes a complex conjugate. The incident, reflected, and transmitted energy fluxes are then given by S_{0y}^+, S_{0y}^-, and S_{2y}^+ (y = d), where the superscripts + and - refer to the waves traveling in the positive and negative y-directions, respectively. In terms of these quantities

the following expressions hold for the reflectance R, transmittance T, and volumetric absorptance η:

$$R = -\frac{S_{0y}^{-}}{S_{0y}^{+}} \tag{11}$$

$$T = \frac{S_{2y}^{+}\,(y = d)}{S_{0y}^{+}} \tag{12}$$

$$\eta = -\frac{\vec{\nabla} \cdot \vec{S}_1}{S_{0y}^{+}} \tag{13}$$

In order to obtain explicit expressions for R, T, and η, it is convenient to treat parallel and perpendicular polarization separately.

Perpendicular Polarization

The s-wave electric fields are perpendicular to the plane of incidence in all media, and the response of the system to the incident radiation depends only on ϵ_x and not on ϵ_y. The normal (y) components of the propagation vectors of the positively going waves in the three media are

$$\xi_{j,s} = \frac{2\pi}{\lambda_v}\left[\epsilon_{j,x} - \epsilon_0 \sin^2\theta\right]^{1/2} \tag{14}$$

The positive square root is specified in order to yield decaying and not growing waves in the absorbing media. With this choice of sign, the complex quantity ξ always lies on the real axis or in the first quadrant of the complex plane.

The incident radiation is specified by the amplitude of the electric field, denoted by E_0^+. Then the amplitude of the other electric fields in the three media are obtained by requiring continuity in the tangential components of $\vec{E}$ and $\vec{H}$ at the two interfaces. From the "method of resultant waves" (ref. 12),

$$E_0^- = \frac{r_{1,s} + r_{2,s} e^{2id\xi_{1,s}}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} E_0^+ \tag{15}$$

$$E_1^+ = \frac{t_{1,s}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} E_0^+ \tag{16}$$

$$E_1^- = \frac{t_{1,s} r_{2,s} e^{2id\xi_{1,s}}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} E_0^+ \tag{17}$$

$$E_2^+ = \frac{t_{1,s} t_{2,s} e^{id(\xi_{1,s} - \xi_{2,s})}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} E_0^+ \tag{18}$$

$$E_2^- = 0 \tag{19}$$

where the s-wave amplitude reflectance $r_{j,s}$ and transmittance $t_{j,s}$ coefficients at the (j-1, j) interface are defined by

$$r_{j,s} = \frac{\xi_{j-1,s} - \xi_{j,s}}{\xi_{j-1,s} + \xi_{j,s}} = t_{j,s} - 1 \qquad j = 1,\ 2 \tag{20}$$

Using Maxwell's equations to eliminate H in the expressions for the Poynting vector yields the following expressions for R_s, T_s, η_s, and F_s:

$$R_s = \left| \frac{r_{1,s} + r_{2,s} e^{2id\xi_{1,s}}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} \right|^2 \tag{21}$$

$$T_s = \left| \frac{t_{1,s} t_{2,s} e^{id\xi_{1,s}}}{1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}} \right|^2 \mathcal{R}e \frac{\xi_{2,s}}{\xi_0} \tag{22}$$

$$\eta_s(y) = \frac{\left(1 - |r_{1,s}|^2\right) \alpha_s}{\left|1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}\right|^2} \left\{ e^{-\alpha_s y} + |r_{2,s}|^2 e^{-\alpha_s(2d-y)} + 2e^{-\alpha_s d} \mathcal{R}e \left[r_{2,s} e^{-2i\xi'_{1,s}(y-d)} \right] \right\} \tag{23}$$

$$F_s(L) = \frac{\left(1 - |r_{1,s}|^2\right) \alpha_s}{\left|1 + r_{1,s} r_{2,s} e^{2id\xi_{1,s}}\right|^2} \left\{ \frac{1 - e^{-[\alpha_s + (1/L)]d}}{\alpha_s + \frac{1}{L}} + |r_{2,s}|^2 e^{-2\alpha_s d} \frac{1 - e^{-[(1/L) - \alpha_s]d}}{\frac{1}{L} - \alpha_s} + 2e^{-\alpha_s d} \mathcal{R}e \left[\frac{r_{2,s}\left(e^{2i\xi'_{1,s} d} - e^{-d/L}\right)}{2i\xi'_{1,s} + \frac{1}{L}} \right] \right\} \tag{24}$$

where

$$\alpha_s = 2\xi''_{1,s} \tag{25}$$

The quantity α_s is an absorption coefficient that depends on the angle of incidence that is discussed subsequently in more detail.

Parallel Polarization

The electric fields of the plane waves lie in the plane of incidence and have components in both the x- and y-directions. The response of media 2 and 3 to electric fields

now depends on both ϵ_x and ϵ_y. The normal components of the propagation vectors of the positively going waves in the three media are

$$\xi_{j,p} = \frac{2\pi}{\lambda_v}\left(\frac{\epsilon_{j,x}}{\epsilon_{j,y}}\right)^{1/2}\left[\epsilon_{j,y} - \epsilon_0 \sin^2(\theta)\right]^{1/2} \tag{26}$$

For isotropic media, $\epsilon_{j,x} = \epsilon_{j,y}$ and the propagation vectors are independent of the wave polarization. Note also that $\xi_{j,s}(\theta = 0) = \xi_{j,p}(\theta = 0)$ as expected since for normal incidence the distinction between s and p polarization disappears. Again, the correct root to be taken is that which puts $\xi_{j,p}$ in the first quadrant of the complex plane.

It is standard practice to specify the incident radiation by the amplitude of the magnetic field which is perpendicular to the plane of incidence in the initial medium. Let this amplitude be denoted as H_0^+. Then the amplitudes of the other magnetic fields in the three media (all of which are perpendicular to the plane of incidence) are determined by requiring continuity in the tangential components of the electric and magnetic fields at the two interfaces. Therefore,

$$H_0^- = \frac{r_{1,p} + r_{2,p}e^{2id\xi_{1,p}}}{1 + r_{1,p}r_{2,p}e^{2id\xi_{1,p}}} H_0^+ \tag{27}$$

$$H_1^+ = \frac{t_{1,p}}{1 + r_{1,p}r_{2,p}e^{2id\xi_{1,p}}} H_0^+ \tag{28}$$

$$H_1^- = \frac{t_{1,p}r_{2,p}e^{2id\xi_{1,p}}}{1 + r_{1,p}r_{2,p}e^{2id\xi_{1,p}}} H_0^+ \tag{29}$$

$$H_2^+ = \frac{t_{1,p}t_{2,p}e^{id(\xi_{1,p}-\xi_{2,p})}}{1 + r_{1,p}r_{2,p}e^{2id\xi_{1,p}}} H_0^+ \tag{30}$$

$$H_2^- = 0 \tag{31}$$

where the p wave amplitude reflectance $r_{j,p}$ and transmittance $t_{j,p}$ coefficients at the (j-1, j) interface are given by

$$r_{j,p} = \frac{\epsilon_{j,x}\xi_{j-1,p} - \epsilon_{j-1,x}\xi_{j,p}}{\epsilon_{j,x}\xi_{j-1,p} + \epsilon_{j-1,x}\xi_{j,p}} = t_{j,p} - 1 \qquad j = 1,\ 2 \tag{32}$$

Using Maxwell's equations to find E in terms of H yields the following expressions for R_p, T_p, η_p, and F_p:

$$R_p = \left| \frac{r_{1,p} + r_{2,p} e^{2id\xi_{1,p}}}{1 + r_{1,p} r_{2,p} e^{2id\xi_{1,p}}} \right|^2 \tag{33}$$

$$T_p = \left| \frac{t_{1,p} t_{2,p} e^{id\xi_{1,p}}}{1 + r_{1,p} r_{2,p} e^{2id\xi_{1,p}}} \right|^2 \mathcal{R}e \frac{\epsilon_0 \xi_{2,p}}{\epsilon_{2,x}\xi_0} \tag{34}$$

$$\eta_p(y) = \frac{\left(1 - |r_{1,p}|^2\right)\alpha_p}{\left|1 + r_{1,p} r_{2,p} e^{2id\xi_{1,p}}\right|^2} \left\{ e^{-\alpha_p y} + |r_{2,p}|^2 e^{-\alpha_p(2d-y)} \right.$$

$$\left. + 2e^{-\alpha_p d} \mathcal{R}e\left[r_{2,p} e^{-2i\xi'_{1,p}(y-d)}\right] \frac{\xi'_{1,p}\, \mathcal{I}m\left(\epsilon^*_{1,x}\xi_{1,p}\right)}{\xi''_{1,p}\, \mathcal{R}e\left(\epsilon^*_{1,x}\xi_{1,p}\right)} \right\} \tag{35}$$

$$F_p(L) = \frac{\left(1 - |r_{1,p}|^2\right)\alpha_p}{\left|1 + r_{1,p} r_{2,p} e^{2id\xi_{1,p}}\right|^2} \left\{ \frac{1 - e^{-[\alpha_p + (1/L)]d}}{\alpha_p + \frac{1}{L}} + |r_{2,p}|^2 e^{-2\alpha_p d} \frac{1 - e^{-[(1/L) - \alpha_p]d}}{\frac{1}{L} - \alpha_p} \right.$$

$$\left. + 2e^{-\alpha_p d} \mathcal{R}e\left[\frac{r_{2,p}\left(e^{2i\xi'_{1,p}d} - e^{-d/L}\right)}{2i\xi'_{1,p} + \frac{1}{L}}\right] \times \frac{\xi'_{1,p}\, \mathcal{I}m\left(\epsilon^*_{1,x}\xi_{1,p}\right)}{\xi''_{1,p}\, \mathcal{R}e\left(\epsilon^*_{1,x}\xi_{1,p}\right)} \right\} \tag{36}$$

where

$$\alpha_p = 2\xi''_{1,p} \tag{37}$$

is the p wave absorption coefficient.

The theory has, thus far, been developed for linearly polarized radiation. However, the incident radiation is often unpolarized. As discussed by Bell (ref. 9), the Poynting vector for unpolarized radiation in a solid cannot, in general, be resolved into a simple sum of s and p wave components. For inhomogeneous waves in absorbing media the Poynting vector has cross terms between the s and p polarized fields that result in energy flow parallel to the plane of the film. Such cross terms, however, do not contribute to the divergence of the Poynting vector since they depend only on y and not on x and z. The proper absorption per unit volume can thus be obtained for incident radiation of arbitrary polarization by the usual weighted sum $\eta = a_s\eta_s + a_p\eta_p$ and $a_s + a_p = 1$, where a_s and a_p are the relative amounts of s and p polarized radiation.

Discussion of Formulas

Equations (23) and (35) are rigorous expressions for the absorption per unit volume of energy from the electromagnetic field in a thin film. The expressions are somewhat complicated but readily lend themselves to physical interpretation.

For example, the coefficient containing $1 - |r_1|^2$ is the fraction of the incident radiant energy that is refracted across the (0-1) interface into medium 1. The absorption of this energy is proportional to the absorption coefficient α and to the intensity of the radiation field, the relative magnitude of which is determined by the remaining factors. The terms in braces refer successively to the relative intensity of the wave refracted into medium 1, the wave reflected from the (1-2) interface, and the interference between these two waves. The term $\left|1 + r_1 r_2 e^{2id\xi_1}\right|^2$ takes into account all further reflections. Thus, although full account was taken of the inhomogeneity of the electromagnetic waves and of the anisotropy of media 1 and 2, the rigorous expressions for η are, in fact, what would be expected from an intuitive treatment of the problem.

The major modification introduced by allowing optical anisotropy is the appearance of different forms for the propagation vectors of s and p waves in media 1 and 2. Aside from this difference, the expressions obtained for the simple, uniaxially anisotropic media considered herein are formally identical with those for isotropic media.

The attenuation of the wave in the film is determined by the product of $\alpha(\theta)$ and y,

the perpendicular distance from the surface. However, one usually thinks of attenuation as being related to the distance along the direction of wave propagation. These two notions may be reconciled by considering the limiting case of an isotropic weakly absorbing material. Since the rate of absorption is determined by the imaginary part of the propagation vector (eqs. (25) and (37)), the condition for weak absorption is (see eqs. (14) and (26))

$$\epsilon_1'' << \epsilon_1' - \epsilon_0 \sin^2\theta \tag{38}$$

If this inequality is satisfied, $\alpha(\theta) \approx \alpha(0)/\cos\theta_1$, where $\alpha(0) = 4\pi k/\lambda_v$ is the usual absorption coefficient (ref. 13) and θ_1 is the angle of refraction given by Snell's Law:

$$n_0 \sin\theta = n_1 \sin\theta_1 \tag{39}$$

This is the expected result: The attenuation is determined by the product of $\alpha(0)$ and the distance along the direction of energy flow, the latter being given by $y/\cos\theta_1$ for weak absorbers. Although such an interpretation can also be given if the medium is strongly absorbing, the direction of energy flow no longer coincides with the classical angle of refraction given by equation (39) because of the strongly inhomogeneous nature of the refracted wave for oblique incidence. In such cases no simple expression for the direction of energy flow is available, and there seems to be no simple physical interpretation for the angular dependence of the absorption coefficient.

Consider next the last term in η_p. If equation (38) is satisfied, then the last term of equation (35) can be reduced to

$$\frac{\xi_1' \mathcal{Im}(\epsilon_1^* \xi_1)}{\xi_1'' \mathcal{Re}(\epsilon_1^* \xi_1)} \approx -\cos 2\theta_1 \tag{40}$$

This result may be understood by referring to figure 2 where the electric vectors of the refracted and internally reflected p waves are shown. The angle between these two

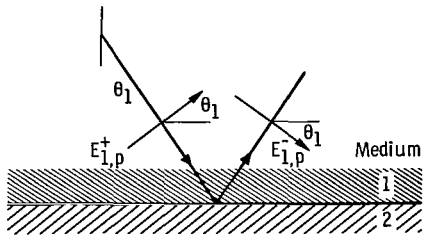

Figure 2. - Electric field vectors of p wave internally reflected at film-substrate interface.

vectors is simply $2\theta_1$, so that the amplitude of the p-wave interference term is modulated by the scalar product of the electric field vectors of the two waves. Only the components of the electric fields parallel to each other interfere. Since the s-wave electric fields are always parallel to each other, such a scalar product does not appear in the interference term of η_s.

Two limiting cases of F are also of interest. The limit $L \to \infty$ corresponds to all excited electrons having an equal chance of being emitted, regardless of the point of excitation in the photoemitting medium. In this case the quantum yield Y is just proportional to the number of photons absorbed by the photoemitter. In fact, if the absorptance of the film is denoted by A, it can be shown that

$$\lim_{L \to \infty} F(L) = A \tag{41}$$

Thus in addition to the standard expression for the absorptance A of the film, $A = 1 - R - T$, an equivalent expression is available in which the contributions of the refracted, internally reflected and interference terms have been explicitly separated out.

The limit $d \to \infty$ corresponds to emission from a semi-infinite medium. In this limit,

$$F(L) = \frac{\left(1 - |r_1|^2\right)\alpha}{\alpha + \frac{1}{L}} \tag{42}$$

where the polarization subscripts have been omitted. An identical expression has been obtained by Spicer (refs. 4 and 5) for normal incidence on an isotropic medium. This widely used expression is also valid for oblique incidence and materials with the simple form of anisotropy considered herein provided that the expression for the reflectance r_1 (eqs. (20) and (32)) and absorption coefficient α (eqs. (25) and (37)) are used.

There is an important generalization of the preceding formulation if medium 2 is replaced by a multilayer structure. Following the method of Rouard as outlined by Heavens (ref. 14) makes it necessary only to replace r_{2s} and r_{2p} in the expressions for R, η, and F by the amplitude reflectance coefficient for the composite medium 2. For example, if medium 2 is decomposed into a film of thickness d' (medium 2) and a substrate (medium 3), then

$$r_2 \to \frac{r_2 + r_3 e^{2id'\xi_2}}{1 + r_2 r_3 e^{2id'\xi_2}} \tag{43}$$

where equations (20) and (32) hold for the amplitude reflectance coefficients at the (1-2) and (2-3) interfaces. This method is useful for dealing with spaced-reflective photocathodes (ref. 15) and may be employed for calculating the total optical properties of any multilayer system.

ILLUSTRATION

The responses of metals to vacuum ultraviolet radiation are currently being studied to determine both the band structure (ref. 16) and collective behavior (ref. 17) of the electrons in the solids. The optical studies are frequently performed on thin films since the purest and most specular metal specimens are fabricated by vapor deposition of films in the submicrometer thickness range. This section illustrates the utility of the preceding analysis by considering, in some detail, the optical physics of the angular dependence of photoemission from a thin metal film.

Aluminum exhibits optical properties in this wavelength region that are typical of many metals and therefore will be used for the illustrative example. In addition, for $\lambda_v < 837$ Å, its "plasma wavelength," aluminum acts as a weakly absorbing dielectric with an index of refraction less than 1. Therefore, it is also well suited for illustrating the approximations for weak absorbers discussed previously.

Consider a 1000-Å-thick isotropic aluminum film on a glass substrate. The aluminum surface is assumed to be illuminated by radiation of wavelength $\lambda_v = 584$ Å at an angle of incidence θ and the emitted electrons are collected in a vacuum. This particular wavelength was chosen because the optical constants of aluminum and glass are well known for this very strong line of the helium spectrum. The optical constants of aluminum and glass are $\epsilon_1 = (0.71 + 0.018\ i)^2$ and $\epsilon_2 = (0.83 + 0.45\ i)^2$, respectively (ref. 18). The thickness of the film (1000 Å) is comparable to the wavelength of the incident radiation, and thus interference effects are expected to appear.

For $\theta < 45^\circ$ the inequality of equation (38) is satisfied, so that the behavior of the waves in the film may be described in the familiar terms of waves in a dielectric film. The absorption of radiation η within the film is plotted in figure 3 for $\theta = 30^\circ$ and 60°. For the s-wave incident at $\theta = 30^\circ$, the usual exponential attenuation of the absorption is modified by the standing wave produced by interference between the refracted wave and the wave reflected from the aluminum-glass interface. The wavelength of the standing wave is obtained from the argument of the exponential in the interference term of η_s as $\pi/\xi'_{1,s} \approx 600$ Å.

A standing wave is not produced by the p-wave incident at 30°, because the radiation is refracted into the aluminum at an angle $\theta_1 \approx 45^\circ$. The electric fields of the two waves in the film are at right angles and do not interfere. Mathematically this suppres-

sion of interference is expressed by the vanishing of the coefficient of the p-wave interference term (eq. (40)), since $\cos 2\theta_1 = 0$ for $\theta_1 = 45^\circ$.

Standing waves are usually presented in textbooks on optics in terms of Wiener's classical experiments on standing waves in air in front of a highly reflecting surface (refs. 19 and 20). The preceding discussion extends the elementary analysis to the standing waves in an absorbing thin film.

The phenomenon of total reflection exhibited by transparent media is also exhibited (ref. 21), in modified form, by aluminum for wavelengths below the plasma wavelength. Since the index of refraction is less than 1, a critical angle for total reflection exists for incidence from the vacuum. For $\theta > \theta_c$, where $\theta_c \equiv \sin^{-1} n_1$, the reflectance of the vacuum-aluminum interface approaches 1, and the wave in the film no longer propagates with weak attenuation but instead is strongly attenuated (evanescent). The absorption length is given approximately by

$$\alpha^{-1}(\theta) = \frac{\lambda_v}{4\pi\sqrt{n_0^2 \sin^2 \theta - n_1^2}} \tag{44}$$

It may be verified that the inequality of equation (38) is not satisfied for $\theta > \theta_c$, since the attenuation of the refracted wave is so strong.

For $\theta = 60^\circ$, $\alpha^{-1} = 95$ Å and the usual exponential decrease for η_s is depicted in figure 3. The behavior of η_p is similar. In the example chosen here the evanescent

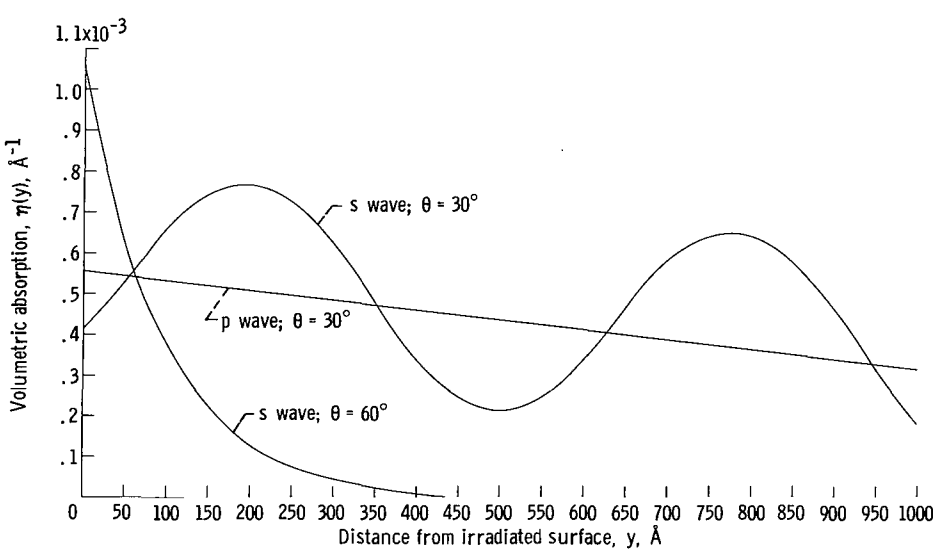

Figure 3. - Absorption per unit volume through a 1000-Å-thick aluminum film on a glass substrate. Incident wavelength, 584 Å.

wave in effect never reaches the substrate, there is no wave reflected at the aluminum-glass interface, and the absorption is well described by exp $(-\alpha y)$. However, for film thicknesses of the order of 100 Å, the evanescent refracted wave does generate a reflected wave (also evanescent) at the aluminum-glass interface that interferes with the refracted wave, and the full expression for η should be used. The interference of these waves, however, would not result in absorption periodic in the y-coordinate (as in η_s (30°)), since their dependence on y is that of a decaying exponential.

In order to evaluate the relative gain in photoemission for oblique incidence $G(\theta)$, a numerical value of L, the electron attenuation length, must be chosen. There are indications (ref. 22) that the attenuation length of electrons excited by light below the plasma wavelength of metals may be less than 50 Å. The relative gain for both p and s polarization is thus plotted in figure 4 for L = 5, 15, 25, and 35.

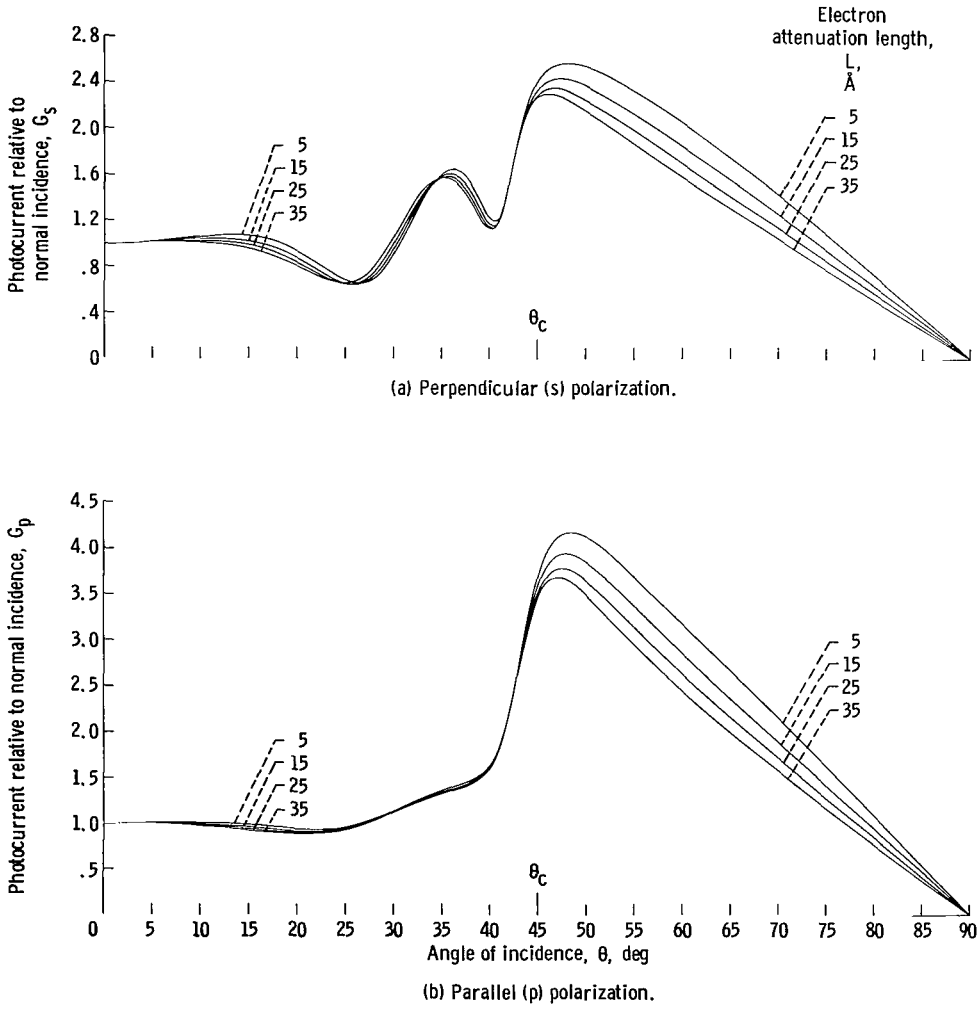

Figure 4. - Angular dependence of photoemission from 1000-Å-thick aluminum film on glass substrate. Wavelength of incident light, 584 Å.

Since most of the photoelectrons originate close to the surface, the yield is determined by the magnitude of the electric field in this region. The relative maximums and minimums exhibited by $G_s(\theta)$ for $\theta < \theta_c$ are respectively due to the appearance, near the surface, of antinodes and nodes of the electric field of the standing wave (and hence of the absorption through eq. (10)) in the interior of the film. The interference of p-waves within the film is suppressed for oblique incidence, so that oscillations are not exhibited by $G_p(\theta)$ to the extent that they are by $G_s(\theta)$.

The general increase in yield as θ approaches θ_c is due to refraction of radiation to angles away from the normal (since $n_1 < n_0$). Radiant energy is deposited closer to the surface and the excited electrons suffer less attenuation. For $\theta > \theta_c$ the reflectance at the vacuum-aluminum interface approaches 1, little radiation is refracted into the film and the emission drops to 0.

As shown in figure 4, the relative gain curves are rather sensitive to the value of the electron attenuation length for $\theta \geq \theta_c$. Thus, an experimental measurement of photocurrent as a function of angle of incidence and its comparison with the theoretical curves of figure 4 suggest a new method of determining the attenuation length of excited electrons.

Present methods of determining L require either (1) transparent substrates, or (2) the photoyield as a function of film thickness, or both (ref. 23). However, for $\lambda_v < 1050$ Å no transparent substrates exist and it is well known that the optical constants of very thin films are a function of film thickness. The relative gain method may employ an opaque substrate and has the advantage that optical and photoelectric properties may be determined for a single film, without the necessity of using some average value of ϵ_1 for films of different thickness.

Although the preceding discussion pertains to thin films, it should be noted that for very thick films the characteristic oscillations produced by interfering waves are absent. However, the increase of the yield to a maximum near θ_c continues to hold for thick films and is simply due to the fact that radiation is refracted away from the normal so that the electrons are excited closer to the surface. The existence of this maximum has been noted by many investigators and has been exploited to improve the response of metal photoelectric detectors to vacuum ultraviolet radiation of wavelength less than the plasma wavelength of the metal (ref. 24).

CONCLUSIONS

Equations for the quantum yield of a thin film photoemitter were formulated on the basis of classical thin film optics. The expressions are valid for all angles of incidence and arbitrary values of the dielectric functions of the initial medium, film, and sub-

strate. To aid in application to ordered structures such as epitaxial films and single crystals, both film and substrate were allowed uniaxial optical anisotropy with the optic axis normal to the plane of the film. The formulation is sufficiently general to be useful for wavelengths from the threshold of photoemission in the near infrared to the soft X-ray region and for both metals and semiconductors. Finally, a new method for determining the value of the electron attenuation length was suggested.

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National Aeronautics and Space Administration,
Cleveland, Ohio, October 8, 1969,
124-09.

APPENDIX - SYMBOLS

A absorptance of film

C parameter for excitation and escape probability

c velocity of light

d film thickness

E electric field

F function containing optical absorption and diffusion steps in photoemission process

G relative gain for oblique incidence

H magnetic field

K propagation vector of electromagnetic fields

k extinction coefficient

L attenuation length of excited electron

n index of refraction

p polarization parallel to incident plane

R reflectance of film

r radius vector

r_1 amplitude reflectance coefficient of 0-1 interface

r_2 amplitude reflectance coefficient of 1-2 interface

S Poynting vector

s polarization perpendicular to incident plane

T transmittance of film

t_1 amplitude transmittance coefficient of 0-1 interface

t_2 amplitude transmittance coefficient of 1-2 interface

Y quantum yield of film

y distance from irradiated surface

α absorption coefficient

ϵ dielectric function

ε_v permitivity of vacuum

η absorption per unit volume

θ angle of incidence

μ_v permeability of vacuum

ξ y component of propagation vector

ψ field component

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