ARPA ORDER NO. 189-1

MEMORANDUM
RM-5595-ARPA
MARCH 1968

OPTICAL HARMONIC GENERATION AT A METAL SURFACE

R. F. Lutomirski

PREPARED FOR:

ADVANCED RESEARCH PROJECTS AGENCY

The RAND Corporation
SANTA MONICA • CALIFORNIA

48

ARPA ORDER NO. 189-1

MEMORANDUM
RM-5595-ARPA
MARCH 1968

OPTICAL HARMONIC GENERATION AT A METAL SURFACE

R. F. Lutomirski

This research is supported by the Advanced Research Projects Agency under Contract No. DAHC15 67 C 0141. RAND Memoranda are subject to critical review procedures at the research department and corporate levels. Views and conclusions expressed herein are nevertheless the primary responsibility of the author, and should not be interpreted as representing the official opinion or policy of ARPA or of The RAND Corporation.

DISTRIBUTION STATEMENT
Distribution of this document is unlimited.

The RAND Corporation
1700 MAIN ST • SANTA MONICA • CALIFORNIA • 90406

PREFACE

This Memorandum was prepared for the Advanced Research Projects Agency's high energy laser program. It examines the interaction of a high intensity laser and a metal, and discusses the heating and second harmonic generation. The discussion should be of interest to those concerned with high energy laser devices and with the propagation of intense laser beams through bounded media.

SUMMARY

This Memorandum presents a calculation of the classical interaction between laser light and a metal. Using a simple model for the statistics of the conduction electrons and their interaction with the surface, Maxwell's equations and the Boltzmann equation are solved self-consistently for the fields in the metal, yielding the usual Fresnel solutions plus correction terms which, at optical frequencies and above liquid helium temperatures, are shown to be of the order of one percent.

When laser light of sufficient intensity is used, a nonlinear polarization is induced and waves of twice the fundamental frequency are produced. For the indicated model and for normal incidence, the second harmonic waves in the metal are calculated and the magnitude of the relative heating is computed. The method for solving the general problem for an arbitrary incidence angle is outlined and the ratio of the average energy flux reflected in the second harmonic to the incident flux is estimated. The relevance to a high-intensity laser experiment is considered and an error in the computation of the reflection coefficient as obtained by Jha[1,2] is discussed.

CONTENTS

PRECEDING PAGE BLANK

I. INTRODUCTION

When a laser beam of sufficient strength interacts with a metal, a nonlinear polarization is induced in the metal and waves of twice the frequency of the incident radiation are produced. The amplitude of these waves decreases rapidly with distance in the direction of propagation and, except for very thin foils, there is no transmission of waves through the metal. However, for oblique incidence, second harmonic waves will be reflected from the surface. While it is difficult to perform high-intensity experiments without damaging the metal, claims of detecting a frequency doubling of the reflected light have been made.(3)

A number of calculations of the nonlinear conductivity tensor have been made based on a free-electron gas model.(4) In the usual theory it is assumed that the current density at any point in the metal is determined entirely by the value of the electric field at that point. With this assumption Maxwell's equations have solutions representing simple harmonic plane waves which are exponentially damped in the direction of propagation.(5) These expressions for both the fundamental and second harmonic solutions will be referred to as the classical results.

The basic assumption of the above theory is not valid when the electric field varies appreciably over distances of the order of the conduction electron free path, i.e., when the free path is of the order of the penetration depth of the electric field. In general the expression for the current density takes the form of a definite integral involving the electric field at all points in the metal, and

Maxwell's equations lead to an integro-differential equation from which the electric field has to be determined.(6)

For the fundamental wave the present theory predicts a propagating disturbance in the metal which is characterized essentially by the classical index of refraction with a small correction, plus a "transient" term (from a branch cut) which is of small amplitude and decreases rapidly with distance from the surface. However, for the second harmonic the theory predicts, in addition to the propagating second harmonic component of classical theory (with a small correction) and a small transient, a propagating disturbance approximately equal in magnitude and opposite in phase to the classical result at the surface, but more rapidly attenuated. Hence the new term is important in computing surface effects. The magnitude of these terms at the surface is shown to be approximately $eE_i^2/mc\omega_p \approx (4.2 \times 10^{-14})\ E_i^2$ (mks units), where e and m are the electronic charge and mass, ω_p is the plasma frequency $(ne^2/m\epsilon_o)^{\frac{1}{2}} \simeq 1.4 \times 10^{16}$ rad/sec, c is the vacuum speed of light, and E_i is the amplitude of the incident electric field.

For the present treatment, the metal is assumed to be semi-infinite in extent and the problem is formulated as a boundary value problem. The relevant equations and approximations are discussed in Section II. In the third section solutions for the fundamental wave (frequency ω) are found using Fourier transforms, and the inversion involves a contribution from a pole and a branch cut. The contribution from the pole approaches the classical result as $v_f/c \rightarrow 0$ (where v_f is the Fermi velocity), with the correction term a function of both frequency and the relaxation time of the conduction electrons.

This correction to the classical refractive index is small except at liquid helium temperatures; for light in the infrared shining on silver at room temperature the correction is of the order of one percent. The contribution from the branch cut is also discussed and its magnitude is shown to be a small fraction of the pole contribution at the surface (e.g., less than one percent for light at 10^3 cm^{-1} shining on silver) and to attenuate rapidly with distance. Finally, the correction to the linear reflection coefficient is considered, and is shown to be negligibly small.

The second harmonic problem for normal incidence is considered in Section IV, and an analysis similar to that of Section III yields the contributions from the poles and the branch cut. It is demonstrated that for normal incidence no second harmonic components are reflected from the surface. In Section V the second harmonic problem for oblique incidence is discussed and the nonlinear reflection coefficient is estimated. The relevance to a high-intensity laser experiment is also discussed, and, in the light of the present analysis, results obtained by Jha[1] are demonstrated to be in error.

II. PHYSICAL MODEL

For the purpose of this analysis the penetration depth of the field is assumed to be small compared with the linear dimensions of the specimen, so that it is permissible to regard the metal as occupying the half-space $z \geq 0$. The physical properties of the metal can be adequately described in terms of the free electron model, according to which the valence electrons are able to move about freely through the volume of the specimen. In the absence of the light wave the zero-order energy distribution of the electrons is taken to be that of a Fermi gas at absolute zero.(7) Under the combined action of the applied electromagnetic field and the collisions of the electrons with the lattice, a steady state is set up, and the distribution function is determined by the Boltzmann equation. All of the details of the collision processes are summarized for the present purpose by specifying the relaxation time τ, or, equivalently, the collision frequency $\nu = \tau^{-1}$. The collisions are caused by thermal or structural imperfections in the lattice; for example, in silver ν is of the order 10^{14} sec^{-1}. The amplitude of the incident light (assumed monochromatic) is then treated as a perturbation parameter in the Boltzmann equation.

It is further necessary to make an assumption concerning the reflection of the electrons from the surface. The surface potential varies from a value of the order of the Fermi energy at the surface to zero at a distance $d \approx 10^{-8}$ cm in from the surface. The average time taken by an electron to enter and leave the barrier region is then of the order d/v_f. Hence, provided $\omega \ll v_f/d \simeq 10^{16}$ sec^{-1}, it is permissible to replace the surface potential barrier by a step potential--i.e., to assume the conduction electrons are specularly reflected from the surface.

In discussing the anomalous skin effect in metals, Reuter and Sondheimer[8] have shown their results to be relatively insensitive to the fraction of electrons assumed specularly reflected from the surface, and reasonable results can therefore be expected at higher frequencies. The mathematical formulation is presented in the next section.

III. THE LINEAR PROBLEM

The electron distribution $f(\underline{r},\underline{v},t)$ satisfies the Boltzmann equation[9]

$$\frac{\partial f}{\partial t} + (\underline{v}\cdot\nabla) f - \frac{e\underline{E}}{m}\cdot\frac{\partial f}{\partial \underline{v}} = -\nu(f-f_o) \tag{1}$$

Let

$$f = f_o + \sum_q f_q\, e^{-iq\omega t} \tag{2}$$

where f_o is the distribution of a degenerate Fermi gas at 0^oK. The fundamental electric field in the metal may be written as

$$\underline{E} = E(z)e^{-i\omega t}\,\underline{e}_x \tag{3}$$

and is proportional to the amplitude of the incident electric field E_i. Treating E_i as a small parameter in a perturbation expansion,

$$f_1 = O(E_i), \qquad f_2 = O(E_i^2)$$

If we linearize Eq. (1) and seek solutions which vary sinusoidally with time and depend only on the coordinate z, we obtain

$$\frac{\partial f_1}{\partial z} - \frac{i\omega'}{v_z} f_1 = \frac{eE(z)}{mv_z}\frac{\partial f_o}{\partial v_x} \tag{4}$$

where $\omega' = \omega + i\nu$.

The formal solution of Eq. (4) may be written as

$$f_1(z,\underline{v}) = \frac{e}{mv_z}\frac{\partial f_o}{\partial v_x} e^{i\omega' z/v_z}\left[\int_0^z E(\xi)\, e^{-i\omega'\xi/v_z}\, d\xi + A(\underline{v})\right] \qquad (5)$$

Then $e^{i\omega' z/v_z}$ increases indefinitely with z for $v_z < 0$. In order that $f(\infty,\underline{v})$ remain finite, f_1 must be written as

$$f_1^-(z,\underline{v}) = -\frac{e}{mv_z}\frac{\partial f_o}{\partial v_x} e^{i\omega' z/v_z}\int_z^\infty E(\xi) e^{-i\omega'\xi/v_z}\, d\xi$$

$$\text{for } v_z < 0 \qquad (6)$$

For $v_z > 0$, write

$$f_1^+ = \frac{e}{mv_z}\frac{\partial f_o}{\partial v_x} e^{i\omega' z/v_z}\left[\int_0^z E(\xi) e^{-i\omega'\xi/v_z} d\xi + A(v)\right] \qquad (7)$$

The specular reflection condition at z = 0 implies that

$$f_1^+(0,v_x,v_y,v_z) = f_1^-(0,v_x,v_y,-v_z) \qquad (8)$$

Substituting Eqs. (6) and (7) into Eq. (8) yields

$$A(\underline{v}) = \int_0^\infty E(\xi)\, e^{-i\omega'\xi/v_z} d\xi$$

and therefore

$$f^+ = \frac{e}{mv_z}\frac{\partial f_o}{\partial v_x} e^{i\omega' z/v_z}\left[\int_0^z E(\xi)\, e^{-i\omega'\xi/v_z} d\xi + \int_0^\infty E(\xi)\, e^{i\omega'\xi/v_z} d\xi\right] \qquad (9)$$

The current density is given by

$$\underline{j} = -e \int \underline{v} \; f \; d^3v \tag{10}$$

If the functions $P_i(\mu)$ are defined by

$$P_i(\mu) = \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_0^{\infty} dv_z \left[\frac{v_i}{v_z} \frac{\partial f_o}{\partial v_x} \, e^{i\omega'\mu/v_z}\right] \tag{11}$$

then $P_i(\mu)$ vanishes unless $i = x$ because $f_o = 0$ at $v_x = \pm\infty$. Dropping the subscript x, note that

$$\int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} dv_z \left[\frac{v_x}{v_z} \frac{\partial f_o}{\partial v_x} \, e^{i\omega'\mu/v_z}\right] = P(\mu) - P(-\mu) \tag{12}$$

The total current density can then be written

$$\begin{aligned} j_x &= -\frac{e^2}{m}\left[\int_0^z E(\xi)\; P(z-\xi)d\xi + \int_0^{\infty} E(\xi)\; P(z+\xi)d\xi + \int_z^{\infty} E(\xi)\; P(\xi-z)d\xi\right] \\ &= -\frac{e^2}{m}\left[\int_0^{\infty} E(\xi)\; P(|z-\xi|)d\xi + \int_0^{\infty} E(\xi)\; P(z+\xi)d\xi\right] \end{aligned} \tag{13}$$

The electromagnetic fields are related to the current through Maxwell's equations

$$\nabla \times \underline{E} = i\omega\underline{B} \tag{14}$$

and

$$\nabla \times \underline{B} = \mu_o \underline{j} - \frac{i\omega E}{c^2} \tag{15}$$

Because of the convolution form of Eq. (13), it is desirable to solve the system by means of Fourier transforms. Equation (13) is valid only in the half-space $z \geq 0$, however, and while several techniques exist for treating this type of problem, perhaps the simplest is a mathematical artifice first used by Shafranov.[7] The technique, as applied to this problem, is to assume that the metal occupies all space, i.e., that Eqs. (13), (14) and (15) hold for $z < 0$ as well as for $z > 0$, and to introduce a Dirac delta function current source at $z = 0$ to yield

$$\nabla \times \underline{B} + \frac{i\omega}{c^2}\underline{E} = \mu_o[\underline{j} + J\delta(z)\underline{e}_x] \tag{16}$$

Equations (3), (14) and (16) yield

$$\frac{\partial^2 E}{\partial z^2} + \frac{\omega^2 E}{c^2} + i\omega\mu_o j = -i\omega\mu_o J\delta(z) \tag{17}$$

The general solution of Eq. (17) is a valid solution in the half-space $z > 0$ of the original problem. The constant J must then be chosen such that the fields connect properly to the vacuum field at $z = 0^-$, so that the tangential electric and magnetic fields are continuous at the boundary. The result of this procedure is a solution which satisfies both the correct differential equation for $z > 0$ and the proper boundary conditions at the surface of the metal.

Then, substituting Eq. (13) into Eq. (17) yields

$$\frac{\partial^2 E}{\partial z^2} + \frac{\omega^2}{c^2} E - \int_0^\infty K(z-\xi)\ E(\xi)d\xi - \int_0^\infty K(z+\xi)\ E(\xi)d\xi = -i\omega\mu_o J\delta(z) \tag{18}$$

where

$$K(\mu) = \frac{i\omega\,\omega_p^2}{n_o c^2}\int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_0^{\infty} dv_z\, \frac{v_x}{v_z}\frac{\partial f_o}{\partial v_x}\, e^{i\omega'|\mu|/v_z} \tag{19}$$

and $\omega_p = (n_o e^2/m\epsilon_o)^{\frac{1}{2}}$, the plasma frequency of the electron gas. From the symmetry of the "new" problem, $E(z) = E(-z)$; hence Eq. (18) may be written as

$$\frac{\partial^2 E}{\partial z^2} + \frac{\omega^2}{c^2}E - \int_{-\infty}^{\infty} K(z-\xi)\; E(\xi)d\xi = -i\omega\mu_o J\delta(z) \tag{20}$$

Equation (20) can now be solved by a Fourier transformation. Defining the Fourier transform of the function $f(z)$ by

$$\tilde{f}(k) = \int_{-\infty}^{\infty} f(z)e^{-ikz}\, dz \tag{21}$$

the transformed equation becomes

$$[-k^2 + (\omega/c)^2 - \tilde{K}(k)]\, \tilde{E}(k) = -i\omega\mu_o J \tag{22}$$

where

$$\tilde{K}(k) = \int_{-\infty}^{\infty} K(z)\; e^{ikz}\, dz = \frac{\omega}{k}\frac{\omega_p^2}{n_o c^2}\int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} dv_z \left[\frac{\partial f_o}{\partial v_x}\,\frac{v_x}{v_z - \omega'/k}\right] \tag{23a}$$

$$= \frac{\omega}{\omega'}\frac{\omega_p^2}{n_o c^2}\int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} dv_z \left[\frac{f_o}{1 - kv_z/\omega'}\right] \tag{23b}$$

and where Eq. (23b) follows from integrating Eq. (23a) by parts.

By the Fourier inversion theorem, the inverse transform of the function $\tilde{f}(k)$ defined by Eq. (21) is

$$f(z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{f}(k)\, e^{ikz}\, dk \qquad (24)$$

and therefore the solution of Eq. (20) is

$$E(z) = \frac{i\omega\mu_o J}{2\pi}\int_{-\infty}^{\infty} \frac{e^{ikz}dk}{k^2-(\omega/c)^2+\tilde{K}(k)} \qquad (25)$$

where the integration, as in Eq. (24), is to be carried out along the real axis in the k plane. The usual procedure for evaluating such an integral is to close the contour in the upper half-plane and to apply the theory of residues. Now assume f_o will vanish for v_z greater than some value v_f. Equation (23) then reveals that $\tilde{K}(k)$ is not defined whenever kv_f/ω' is both real and has magnitude greater than unity, i.e., whenever $\arg k = \pm\,\delta \equiv \pm \arg \omega' = \pm \tan^{-1}(\nu/\omega)$ and $|k| \geq \Omega/v_f$ where $\Omega = |\omega'| = (\omega^2+\nu^2)^{\frac{1}{2}}$. These conditions define the branch cuts around which the contour must be deformed. The deformed contour and the branch lines are indicated in Fig. 1, where E(z) is recovered in the limit as $R \rightarrow \infty$.

The integral over the infinite semicircle is easily shown to vanish. Then the residue theorem applied to the contour yields

$$E(z) = -\frac{i\omega\mu_o J}{2\pi}\int_{Br} \frac{e^{ikz}dk}{k^2-(\omega/c)^2+\tilde{K}(k)} - \omega\mu_o\, J \sum_j \mathrm{Res}\left[\frac{e^{ikz}}{k^2-(\omega/c)^2+\tilde{K}(k)}\right]_{k=k_j} \qquad (26)$$

where each k_j is an isolated pole of the integrand of Eq. (25) in the upper half-plane, the integral is along the branch cut and denoted by Br, and Res is the residue.

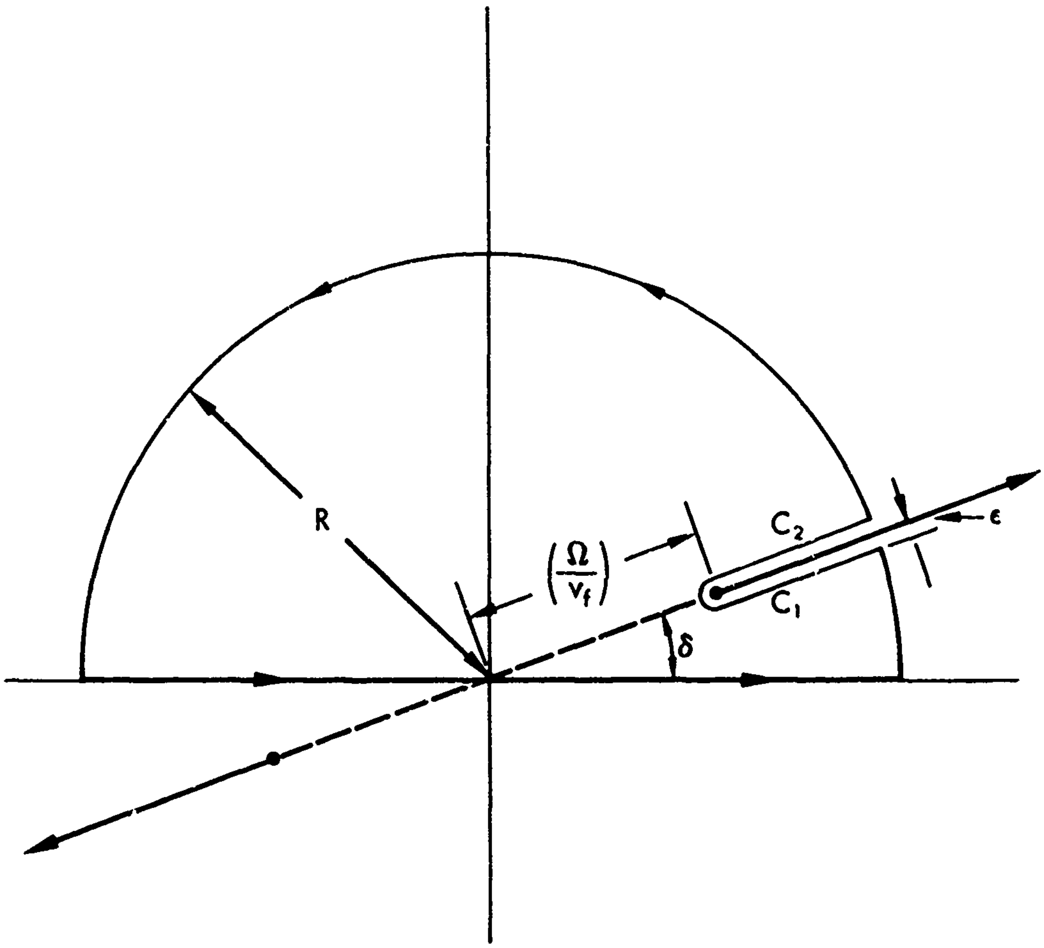

Fig. 1—Integration contour in the complex k plane

CONTRIBUTION TO THE ELECTRIC FIELD FROM THE POLE

In this section the contribution from the residues to the electric field are calculated. To locate the poles in the k plane, the roots of the dispersion relation

$$D(k) = k^2-(\omega/c)^2+\tilde{K}(k) = 0 \tag{27}$$

must be found. These roots are discussed in Appendix A. In the limit $\omega \gg \nu$ it is shown that there always exists one and only one zero of this expression, provided f_o is a symmetric function of v_z, and it occurs when $\left|\frac{kv_f}{\omega'}\right| \ll 1$. The integrand of Eq. (23) is an analytic function in the "cut" plane and $\tilde{K}(k)$ can be evaluated analytically for $\omega \gg \nu$ by expanding the denominator to yield:

$$\tilde{K}(k) = \frac{\omega}{\omega'} \frac{\omega_p^2}{c^2}\left[1 + \frac{k^2\overline{v_z^2}}{\omega'^2} + \cdots\right] \tag{28}$$

The index of refraction $n = (kc/\omega)$ is then given by the positive square root of

$$n^2 = \frac{1 - \frac{\omega_p^2}{\omega\omega'}}{1 + \frac{\omega\omega_p^2}{\omega'^3}\frac{\overline{v_z^2}}{c^2}} \tag{29}$$

Equation (29) describes the wave propagation characteristics of a metal as a function of frequency, including the effect of a finite velocity spread in the distribution function for the free electrons. As the ratio $\overline{v_z^2}/c^2$ tends to zero, the above expression continuously approaches the usual formula for a "cold" electron gas

$$n_c^2 = \left(\frac{k_c c}{\omega}\right)^2 = 1 - \frac{\omega_p^2}{\omega(\omega+i\nu)} \tag{30}$$

The magnitude of the thermal correction in n^2, $\Delta = \frac{n^2-n_c^2}{n_c^2}$, is

$$\Delta \cong \left(\frac{\omega_p}{c}\right)^2 \overline{v_z^2} \, \frac{\omega}{(\omega^2+\nu^2)^{3/2}} \tag{31}$$

The maximum value of the correction occurs at $\omega = \nu/\sqrt{2}$. Substituting in Eq. (30) reveals that

$$\Delta \le \frac{2}{3^{3/2}} \left(\frac{\omega_p}{\nu}\right)^2 \frac{\overline{v_z^2}}{c^2} \tag{32}$$

Hence for $\omega \gg \nu$, only the first few moments of f_o need be estimated to compute the dispersion relation. For lower frequencies it is necessary to approximate the shape of the Fermi surface by an analytic expression or to numerically integrate the expression for $\mathcal{K}(k)$. In general, the energy surface can be complicated, and its shape for certain metals has been estimated from measurements of various optical constants, soft x-ray emission, Knight shift, and various transport properties.(11) For rough order-of-magnitude calculations we can assume $\overline{v_z^2}$ is $O(v_f^2)$ where v_f is calculated from the free electron model and depends only upon the valence electron density. A spherical energy surface of the free electron model is expected to be a good approximation in a number of cases, including sodium, silver, and certain alloys of copper.(12) While it is not necessary to assume a spherical distribution to proceed with parts of the analysis, if some of the integrations are performed the formulas become simpler and permit us to readily estimate the magnitudes of the various terms. Then with

$$f_o = \frac{n_o}{(4/3)\pi\ v_f^3} H(v_f - v) \tag{33}$$

where

$$H(x) = 1, \qquad x \geq 0$$

$$= 0, \qquad x < 0$$

we have $\overline{v_z^2} = \frac{1}{5} v_f^2$. For silver at 0^oC, $v_f \approx 1.4 \times 10^8$ cm/sec, $\omega_p \approx 1.4 \times 10^{16}$ rad/sec, and $\nu \approx .5 \times 10^{14}$ sec^{-1}.[(7)] Then with $c = 3 \times 10^{10}$ cm/sec, the maximum value of Δ is .14. For poorer conductors, Δ_{max} will be correspondingly less.

The root of Eq. (27) can then be written approximately as

$$k_o = k_c\left\{1 - \frac{\Delta}{2} e^{i\varphi}\right\} \tag{34}$$

where $\varphi = \tan^{-1}\left[\frac{\nu(\nu^2 - 3\omega^2)}{\omega(\omega^2 - 3\nu^2)}\right]$, k_c is the classical propagation vector, and $\Delta/2 \leq .07$. For example, for light at $10^3 cm^{-1}$ shining on silver, $\omega \cong 2 \times 10^{14} sec^{-1}$, and from Eq. (31), $\Delta/2 \approx .01$.

Returning to Eq. (26), the contribution to the electric field from the pole, E_p, becomes

$$E_p = -\omega\mu_o J \times \text{Res}\left[\frac{e^{ikz}}{D(k)}\right]_{k=k_o} = \frac{-\omega\mu_o J\ e^{ik_o z}}{\left[1 + \frac{1}{5}\left(\frac{\omega_p v_f}{c}\right)^2 \frac{\omega}{\omega'^3}\right] 2k_o} \tag{35}$$

The magnetic field associated with E_p is given by

$$\underline{B}_p(z) = \frac{1}{i\omega}\frac{\partial E_p}{\partial z}\,\underline{e}_y = -\frac{\mu_o J\, e^{ik_o z}}{2\left[1+\frac{1}{5}\left(\frac{\omega_p v_f}{c}\right)^2\left(\frac{\omega}{\omega'^3}\right)\right]}\,\underline{e}_y \qquad (36)$$

The constant J cannot be evaluated until the contribution from the branch cut is calculated. However, in most practical cases, the propagation characteristics of E_p (and B_p) will be shown to be given approximately by the classical electron theory (i.e., $k_o \approx k_c$), with a correction of at most a few percent above liquid helium temperatures.

CONTRIBUTION TO THE ELECTRIC FIELD FROM THE BRANCH CUT

In the integration along the branch cut in Eq. (26), the contribution from the small semicircle is easily seen to approach zero as $\epsilon \to 0$. Then the contribution from the branch cut is

$$E_{Br} = -\frac{i\omega\mu_o J}{2\pi}\int_{c_1+c_2}\frac{e^{ikz}\,dk}{k^2-(\omega/c)^2+\tilde{K}(k)} \qquad (37)$$

Along c_2, i.e., on top of the branch cut, $k \approx \rho\, e^{i\delta}\, e^{i\epsilon}$. Then $\omega'/k = (\Omega e^{i\delta})/(\rho\, e^{i\delta}e^{i\epsilon}) = \frac{\Omega}{\rho}\, e^{-i\epsilon}$. Therefore, the pole in the integrand in Eq. (23) lies just below the real axis in the complex v_z plane. However, the pole can be considered to lie on the real ax. and the path of integration can then be deformed to lie just above the pole. The new path will be denoted by c_+, and the integral so defined by $\tilde{K}^+(\rho)$. Similarly, along c_1 the path in the v_z plane can be deformed to c_- (see Fig. 2), and the corresponding integral will be denoted by $\tilde{K}^-(\rho)$. Then

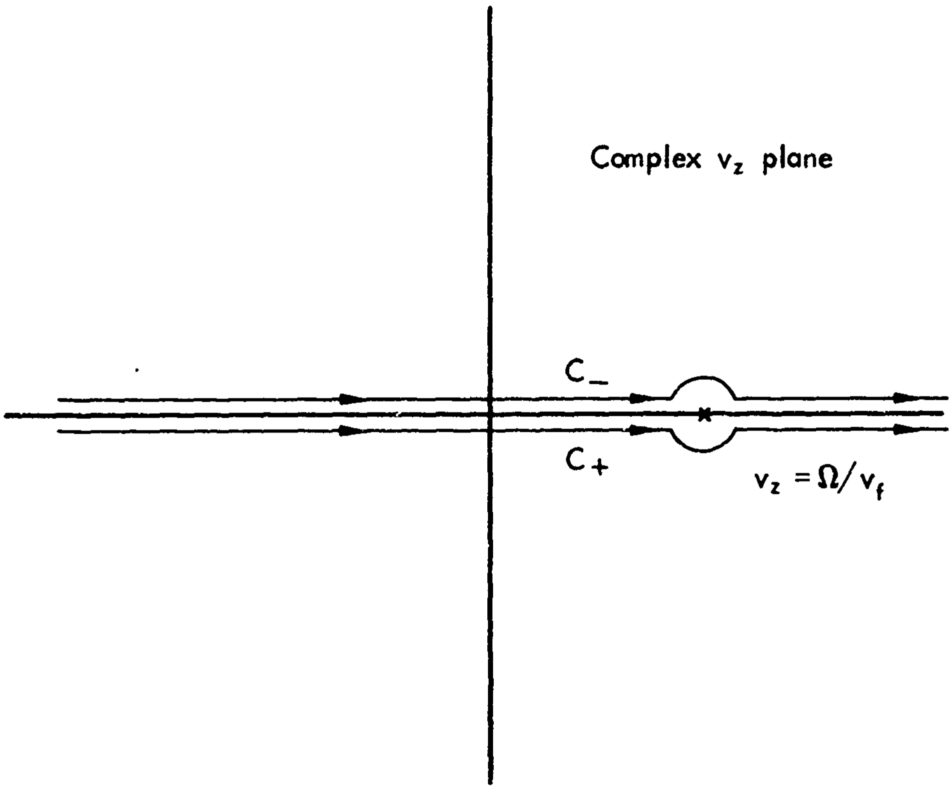

Fig. 2—Paths of integration for $\tilde{k}^+(k)$ and $\tilde{k}^-(k)$ in the second harmonic

$$-\frac{2\pi E_{Br}}{i\omega\mu_o J} = \int_{\Omega/v_f}^{\infty} \frac{e^{iz\rho e^{i\delta}} e^{i\delta}\, d\rho}{\rho^2 e^{2i\delta} - \omega^2/c^2 + \tilde{K}^+(\rho)} + \int_{\infty}^{\Omega/v_f} \frac{e^{iz\rho e^{i\delta}} e^{i\delta}\, d\rho}{\rho^2 e^{2i\delta} - \omega^2/c^2 + \tilde{K}^-(\rho)} =$$

$$e^{i\delta} \int_{\Omega/v_f}^{\infty} e^{iz\rho e^{i\delta}} \frac{[\tilde{K}^-(\rho) - \tilde{K}^+(\rho)]\, d\rho}{[\rho^2 e^{2i\delta} - \omega^2/c^2 + \tilde{K}^+(\rho)][\rho^2 e^{2i\delta} - \omega^2/c^2 + \tilde{K}^-(\rho)]} \quad (38)$$

where

$$\tilde{K}^{\pm}(\rho) = \frac{\omega}{\rho} \frac{\omega_p^2}{n_o c^2} \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{c_\pm} dv_z \left[\frac{\partial f_o}{\partial v_x} \frac{v_x}{v_z - \frac{\Omega}{\rho}}\right] \quad (39)$$

$[\tilde{K}^-(\rho) - \tilde{K}^+(\rho)]$ can also be expressed by Eq. (39), except that the integration is now performed along a closed contour enclosing the pole (in the positive direction). Consequently, according to the theory of residues,

$$\tilde{K}^- - \tilde{K}^+ = 2\pi i \frac{\omega}{\rho} \frac{\omega_p^2}{n_o c^2} \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \left(v_x \frac{\partial f_o}{\partial v_x}\right)_{v_z = \Omega/\rho} \quad (40)$$

From Eq. (33),

$$\frac{\partial f_o}{\partial v_x} = -\frac{n_o}{\frac{4}{3}\pi v_f^3} \frac{v_x}{\sqrt{v_x^2 + v_y^2 + v_z^2}} \delta\left(\sqrt{v_x^2 + v_y^2 + v_z^2} - v_f\right) \quad (41)$$

Then, changing to cylindrical coordinates and substituting $v^2 \rightarrow v_\perp^2 + (\Omega/\rho)^2$ yields

$$\tilde{K}^- - \tilde{K}^+ = -\frac{3}{2} \pi i \frac{\omega}{\rho} \frac{\omega_p^2}{c^2 v_f^3} [v_f^2 - (\Omega/\rho)^2]\, H[v_f^2 - (\Omega/\rho)^2] \quad (42)$$

Therefore

$$E_{Br} = -\frac{3}{4}\mu_o J \frac{\omega^2\omega_p^2}{c^2 v_f^3} e^{i\delta} \int_{\Omega/v_f}^{\infty} \frac{[v_f^2-(\Omega/\rho)^2]\, e^{iz\rho e^{i\delta}}\, d\rho}{\rho[\rho^2 e^{2i\delta}-\omega^2/c^2+\tilde{K}^+(\rho)][\rho^2 e^{2i\delta}-\omega^2/c^2+\tilde{K}^-(\rho)]} \qquad (43)$$

First note that neglecting the ω^2/c^2 terms in the denominator in Eq. (46) is equivalent to neglecting $(v_f/c)^2$ compared to unity. Then the magnetic field from the branch cut is given by

$$B_{Br} = \frac{1}{i\omega}\frac{\partial E_{Br}}{\partial z} = -\frac{3}{4}\omega\mu_o J \frac{\omega_p^2}{c^2} e^{-2i\rho} \int_{\Omega/v_f}^{\infty} \frac{[v_f^2-(\Omega/\rho)^2]\, e^{iz\rho e^{i\delta}} d\rho}{[\rho^2+\tilde{K}^+ e^{-2i\delta}][\rho^2+\tilde{K}^- e^{-2i\delta}]} \qquad (44)$$

To evaluate the fields at the surface, substitute $u = \Omega/v_f\rho$ and $z = 0$ in Eq. (43) and Eq. (44) to yield

$$E_{Br}(0) = -\frac{3}{4}\mu_o J \frac{\omega^2\omega_p^2}{c^2\Omega^4} v_f^3 e^{-3i\delta} \int_0^1 \frac{(1-u^2)u^3\, du}{\left[1+\left(\frac{uv_f}{\Omega}\right)^2 \tilde{K}^+ e^{-2i\delta}\right]\left[1+\left(\frac{uv_f}{\Omega}\right)^2 \tilde{K}^- e^{-2i\delta}\right]} \qquad (45)$$

$$B_{Br}(0) = -\frac{3}{4}\mu_o J \frac{\omega\,\omega_p^2}{c^2\Omega^3} e^{-2i\delta} \int_0^1 \frac{(1-u^2)u^2\, du}{\left[1+\left(\frac{uv_f}{\Omega}\right)^2 \tilde{K}^+ e^{-2i\delta}\right]\left[1+\left(\frac{uv_f}{\Omega}\right)^2 \tilde{K}^- e^{-2i\delta}\right]} \qquad (46)$$

But $\left|\left(\frac{v_f}{\Omega}\right)^2 \tilde{K}^{\pm}\right|$ is $O(\Delta)$, and therefore, to lowest order in Δ

$$E_{Br}(0) = -\frac{5}{16}\omega\mu_o J\,\Delta \frac{v_f}{\Omega} e^{-3i\delta} \qquad (47)$$

$$B_{Br}(0) = -\frac{1}{2}\mu_o J\,\Delta\, e^{-2i\delta} \qquad (48)$$

The magnitude of the ratio (R_o) of the contribution to the electric field from the branch cut to that from the pole at the surface of the metal, to lowest order in Δ, is

$$R_o(\omega) = \left|\frac{E_{Br}(0)}{E_p(0)}\right| = \frac{5}{8}\left(\frac{k_c v_f}{\Omega}\right)\Delta = \frac{1}{8}\,\omega_p^2\left(\frac{v_f}{c}\right)^3\left|\frac{k_c c}{\omega}\right|\frac{\omega^2}{(\omega^2+\nu^2)^2} \tag{49}$$

Using Eq. (30) to express the ratio as a function of ω, the maximum value of the above expression can be shown to occur at $\omega = \nu/\sqrt{2}$ (provided $\nu^2 \ll \omega_p^2$, which is well satisfied for most metals). The maximum value of R_o is $\approx .03\ (\omega_p v_f/\nu c)^3$. For silver, $(R_o)_{max} = R_o(3.6 \times 10^{13}) \approx .06$. For poorer conductors, $(R_o)_{max}$ will be considerably less. The relative contribution from the branch cut also falls off rapidly with frequencies far from $\nu/\sqrt{2}$. Again, for light at $10^3\ cm^{-1}$ shining on silver, $R_o(2 \times 10^{14}) \approx .005$.

A complete evaluation of $E_{Br}(z)$ requires integration of Eq. (43), which can only be performed numerically. However, it is possible to find an asymptotic representation for $E_{Br}(z)$ valid for $(\Omega z/v_f) \gg 1$ by successively integrating by parts ($v_f/\Omega < v_f/\nu$; for silver this representation is always valid for $z \gtrsim 10^{-6}$ cm).

$$E_{Br}(z) \sim \frac{15}{2}\,\omega\mu_o J\,\Delta\,\frac{v_f}{\Omega}\,e^{-5i\delta}\,\frac{e^{iz\omega/v_f}}{(\Omega z/v_f)^2}\,e^{-z\nu/v_f} \tag{50}$$

The ratio of the electric fields in the metal, R_z, to lowest order in Δ, is given by

$$R_z(\omega) = \left|\frac{E_{Br}(z)}{E_p(z)}\right| \sim \frac{24R_o(\omega)e^{-z\nu/v_f}\,e^{Im(k_c)z}}{(\Omega z/v_f)^2} \tag{51}$$

provided $\Omega z/v_f >> 1$. It is interesting to observe that for optical frequencies, $\mathrm{Im}(k_c) \sim \omega_p/c \approx .5 \times 10^6\ \mathrm{cm}^{-1}$, and with exception of the best conductors, $(\omega_p v_f/\nu c) < 1$. Hence, for z sufficiently large, $R_z(\omega)$ can be greater than unity. However, this occurs when both $|E_p|$ and $|E_b|$ are negligibly small. For $\omega/\nu << 1$, $R_z(\omega) << 1$, and the electric field falls off exponentially with z.

THE REFLECTION COEFFICIENT

The analysis has shown that when an electromagnetic wave is incident upon a metal, energy is transmitted in the metal as a complicated electromagnetic disturbance. The contribution from the branch cut is dissipated within a distance of the order v_f/Ω. The contribution from the pole has a longer range, and its behavior is essentially determined by the classical model, with a small correction (above liquid helium temperatures) due to the nonzero Fermi velocity. The fraction of energy reflected from the surface can be calculated by applying the usual boundary conditions at the metal surface:

$$E_i + E_r = E(0) = E_p(0) + E_B(0) \tag{52a}$$

$$E_i - E_r = cB(0) = cB_p(0) + cB_B(0) \tag{52b}$$

where E_i and E_r are the incident and reflected electric fields, respectively. The constant J can now be computed by adding Eqs. (52a) and (52b):

$$J = -\frac{4E_i}{c\mu_o\left(1 + \frac{1}{n_c}\right)} \left\{1 + \frac{\Delta}{2}\left[\left(1 + \frac{1}{n_c}\right)e^{i\varphi} + 2e^{-2i\delta}\right]\right\} \tag{53}$$

As $\Delta \to 0$, Eqs. (35) and (53) yield

$$|E_p(0)| \to \frac{2E_i}{1+n_c} \tag{54}$$

the classical result for the amplitude of the transmitted field at the metal surface.

The reflection coefficient, $r = \left|\frac{E_r}{E_i}\right|^2$, to lowest order in Δ, is given by

$$\frac{r}{r_c} = 1 + 2\Delta\beta n_i \left[\frac{1}{(1-n_r)^2 + n_i^2} + \frac{1}{(1-n_r)^2 + n_i^2}\right] \tag{55}$$

where $r_c = \left|\frac{1-n_c}{1+n_c}\right|^2$, $\beta = -[\sin(2\delta) + \frac{1}{2}\sin\omega]$, and n_r and n_i are the real and imaginary parts of n_c, respectively.

For $\omega_p^2 \gg \omega^2 \gg \nu^2$, the correction term is $0(v_f^2/c^2) \sim 10^{-5}$. For $\omega_p^2 \gg \nu^2 \gg \omega^2$

$$r/r_c \approx 1 + \Delta \frac{\sqrt{2\omega\nu}}{\omega_p} \tag{56}$$

which again gives a negligible correction. Reuter and Sondheimer,[8] using a similar analysis, have discussed the reflectivity of silver at liquid helium temperatures (corresponding to much larger Δ's in this analysis) and have concluded that the correction to the classical result would still be very difficult to measure.

IV. SECOND HARMONIC GENERATION

In this section, the formalism developed in Section III is utilized with the new assumption that terms of order E_1^2 are no longer negligible. The electromagnetic fields are expanded in a Fourier series,

$$\underline{E} = \sum_{n=-\infty}^{\infty} \underline{E}_n \, e^{-in\omega t}, \qquad \underline{B} = \sum_{n} \underline{B}_n \, e^{-in\omega t} \tag{57}$$

and solutions are sought for which the series converge rapidly. These representations are valid when E_1 oscillates with frequency ω and has a sufficiently small amplitude. In this case each of the n^{th}-order Fourier coefficients contains a term which is the n^{th} power of the incident wave, and the series converges quickly for small amplitude waves.

Substituting Eqs. (2) and (57) into Eq. (1), the second-order component of the transport equation becomes

$$(\nu-2i\omega)f_2 + v_z \frac{\partial f_2}{\partial z} - \frac{e}{m} \underline{E}_2 \cdot \frac{\partial f_o}{\partial \underline{v}} = \frac{e}{m} [\underline{E}_1 + (\underline{v} \times \underline{B}_1)] \cdot \frac{\partial f_1}{\partial \underline{v}} \tag{58}$$

Let $\omega'' = \omega + \frac{1}{2}i\nu$ and define g_i by

$$g_i \equiv \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \, v_i f_2 \tag{59}$$

Then multiplying Eq. (58) by v_i and integrating yields

$$-2i\omega'' g_i + v_z \frac{\partial g_i}{\partial z} - \frac{e}{m} E_{2j} \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \, v_i \frac{\partial f_o}{\partial v_j} = D_i \tag{60}$$

where

$$D_i = \frac{e}{m} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left[E_1 \frac{\partial f_1}{\partial v_x} + B_1 \left(v_x \frac{\partial f_1}{\partial v_x} - v_z \frac{\partial f_1}{\partial v_x} \right) \right] v_i \, dv_y dv_x \tag{61}$$

and summation over j is implied. From the relation $\partial f_o/\partial v_j = (v_j/v)(\partial f_o/\partial v)$, it follows that the third term in Eq. (60) vanishes unless $i = j$. Further, since there is no second harmonic component in the incident wave, the boundary conditions (Eq. (55)) require that the second harmonic electric field in the metal be longitudinal, or $j = z$. For the z component of Eq. (60), the third term integrates to

$$\frac{3}{2} \frac{n_o e}{m} \frac{v_z^2}{v_f^3} E_2 \tag{62}$$

Similarly, f_1 (given by Eq. (5)) is an even function of v_y and an odd function of v_x, from which it can be shown that $D_x = D_y = 0$, and D_z reduces to

$$D_3 = \frac{eB_1}{m} v_z \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} v_x \frac{\partial f_1}{\partial v_z} dv_x dv_y \tag{63}$$

The equation for g_z seems intractable as it now stands since the exact forms of the fields and distribution function of the fundamental are complicated by boundary effects. Fortunately it is possible to make reasonable approximations which will render Eq. (60) tractable. In the last section it was shown that the branch cut contribution to the fundamental field has a small amplitude and short range, so only the contribution from the pole will be included. A second assumption, which is implicit in the classical theory and its conclusions, is that the electric field may be regarded as spatially constant for the purpose of calculating the current at a point. This assumption is valid when the field does not change appreciably over a distance of the order of the mean free path of the conduction electrons, that is to say, when the mean free path is small compared to the effective field penetration depth. This condition is equivalent to setting $\Delta = 0$ and is satisfactory for all frequencies of interest except at very low temperatures. The solution of Eq. (4) can therefore be written

$$f_1 \approx \frac{ieE_1(z)}{m\omega'} \frac{\partial f_o}{\partial v_x} \tag{64}$$

which yields

$$j_1 \approx - \frac{ie^2}{m\omega'} E_1(z) \int v_x \frac{\partial f_o}{\partial v_x} d^3\underline{v} \tag{65}$$

for the current density. When Eq. (64) is substituted into Maxwell's equations, the classical dispersion relation, Eq. (30), is recovered. Then, using Eqs. (33) and (64) and carrying out the indicated integration results in the following equation for g_z:

$$\frac{\partial g_z}{\partial z} - \frac{2i\omega''}{v_z} g_z = v_z \psi(z) \tag{66}$$

where

$$\psi(z) = \frac{3}{2} \frac{n_o e}{m v_f^3} \left[F(z) - E_2(z)\right] \tag{67}$$

and

$$F(z) = \frac{ie}{m\omega'} E_1(z) B_1(z) \tag{68}$$

The solution of Eq. (66) that satisfies the specular reflection condition at $z = 0$ is

$$g_2^- = -v_z\, e^{2i\omega'' z/v_z} \int_z^\infty e^{-2i\omega''\xi/v_z}\, \psi(\xi)\,d\xi \tag{69a}$$

for $z < 0$ and

$$g_2^+ = v_z\, e^{2i\omega'' z/v_z} \left[\int_0^z e^{-2i\omega\xi/v_z}\, \psi(\xi)\,d\xi - \int_0^\infty e^{2i\omega\xi/v_z}\, \psi(\xi)\,d\xi\right] \tag{69b}$$

for $z > 0$.

Following the same procedure used to derive Eq. (13) yields for the second harmonic current density

$$j_2 = \left(\frac{2mv_f^3}{3n_o e}\right) 2i\omega\epsilon_o \left[\int_0^\infty K_2(z+\xi)\ \psi(\xi)\,d\xi - \int_0^\infty K_2(z-\xi)\ \psi(\xi)\,d\xi\right] \tag{70}$$

where

$$K_2(x) = -\frac{3i\omega_p^2}{\omega v_f^3} \int_0^{v_f} e^{2i\omega''|x|/v_z}\, v_z\,dv_z \tag{71}$$

With this definition of $K_2(x)$, and introducing Maxwell's equation for the longitudinal harmonic

$$E_2 - \frac{1}{2i\omega\epsilon_o}\, j_2 = 0 \tag{72}$$

results in

$$E_2(z) + \left(\frac{2mv_f^3}{3n_o e}\right)\left[\int_0^\infty K_2(z-\xi)\ \psi(\xi)\,d\xi - \int_0^\infty K_2(z+\xi)\ \psi(\xi)\,d\xi\right] = 0 \tag{73}$$

or, using Eq. (68),

$$E_2(z) - \left[\int_0^\infty K_2(z-\xi)\ E_2(\xi)\,d\xi - \int_0^\infty K_2(z+\xi)\ E_2(\xi)\,d\xi\right] = W(z) \tag{74}$$

where

$$W(z) = \int_{-\infty}^{0} K_2(z-\xi)\ F(-\xi)d\xi - \int_{0}^{\infty} K_2(z-\xi)\ F(\xi)d\xi \tag{75}$$

Since $W(z) = -W(-z)$, $E_2(z)$ is antisymmetric, and Eq. (74) becomes

$$E_2(z) - \int_{-\infty}^{\infty} K_2(z-\xi)\ E_2(\xi)d\xi = W(z) \tag{76}$$

Fourier transforming Eq. (76) yields

$$\tilde{E}_2(k) = \frac{\tilde{W}(k)}{1-\tilde{K}_2(k)} = \tilde{K}_2(k)\ \frac{[\tilde{F}_+(-k) - \tilde{F}_+(k)]}{1-\tilde{K}_2(k)} \tag{77}$$

where

$$\tilde{K}_2(k) = \int_{-\infty}^{\infty} \tilde{K}_2(x)\ e^{-ikx}dx = -\frac{3\omega_p^2\omega''}{v_f^3\omega\ k^2} \int_0^{v_f} \frac{v_z^2 dv_z}{v_z^2 - (2\omega''/k)^2} \tag{78}$$

and

$$\tilde{F}_+(k) = \int_0^{\infty} F(z)\ e^{-ikz}dz \tag{79}$$

As in the linear case, the branch cuts in the k plane are those regions where $\tilde{K}_2(k)$ is not defined, i.e., where $kv_f/2\omega''$ is real and $\left|\frac{kv_f}{2\omega''}\right| > 1$. $\tilde{K}_2(k)$ is analytic in the rest of the plane and is given by

$$\tilde{K}_2(k) = -\frac{3\omega_p^2\omega''}{v_f^2\omega k^2}\left[1 - \frac{\omega''}{kv_f}\ \ln\left(\frac{1 + kv_f/2\omega''}{1 - kv_f/2\omega''}\right)\right] \tag{80}$$

where ln denotes that branch of the logarithm which vanishes for $k = 0$.

From Eqs. (68) and (79)

$$F_+(-k) - F_+(k) = \alpha \frac{2k}{k^2-(2k_c)^2} \tag{81}$$

where

$$\alpha = -\frac{e}{m\omega'} E_1^2 \frac{4nc}{c(1+nc)^2} \tag{82}$$

Referring to Eq. (79), it can readily be seen that $\tilde{K}_2(k)$ has no isolated singularities in the cut plane, and that the singularities of $\tilde{E}_2(k)$ are therefore the poles of $F_+(-k) - F_+(k)$ and the zeros of $1-\tilde{K}_2(k)$. The expression for $F_+(-k) - F_+(k)$ indicates that there is a pole of $\tilde{E}_2(k)$ which yields a signal with wave number $k = 2k_c$ which is twice that of the fundamental frequency, and since it is a second harmonic term, it therefore has the same phase velocity as that of the fundamental.

The roots of $1 - \tilde{K}_2(k)$ are discussed in Appendix B. In the limit $\omega \gg \nu$, it is shown that there always exists one and only one zero of this expression.

Finally, in Appendix C, the relative amplitudes of the residues are calculated, and the amplitude of the branch integral is estimated at $z = 0$. It is shown that the amplitudes of the waves from the residues are nearly equal at $z = 0$ but are approximately 180° out of phase. When $\omega \ll \omega_p$ the amplitudes at $z = 0$ are $\approx \alpha$ and $-\alpha$ from the residues at $1 - \tilde{K}_2(k) = 0$ and $k = 2k_c$, respectively. While both values of k correspond to evanescent waves for $\nu \ll \omega \ll \omega_p$, the latter has a much longer range ($\sim c/\omega_p$) than the former ($\sim v_f/\omega_p$) and is the only disturbance of interest away from the surface. In this frequency range

$$|\alpha| = \left| \frac{4ieE_i^2}{mc\omega_p} \sqrt{\frac{\omega}{\omega'}} \right| \approx 1.67 \times 10^{-13}\ E_i^2 \text{ (mks units)} \tag{83}$$

for $\omega \gg \nu$. The propagating wave with $k = 2k_c$ is smaller in magnitude than the fundamental in the ratio $\approx \frac{2eE_i}{mc\omega'}$ ($\approx 10^{-10}\ E_i$ for ω in the infrared) and has half the range. The heating effect of the second harmonic is therefore down by approximately the square of this ratio. When the inequality $\omega \ll \omega_p$ is not satisfied (e.g., $\omega/\omega_p \gtrsim 1/3$) the expressions for α and for the residues are more complicated and the expressions derived in Appendices B and C must be used.

V. OBLIQUE INCIDENCE

For the case where the propagation vector of the incident wave makes an angle θ with respect to the normal to the metal surface (i.e., the z direction), the q^{th} harmonic of the distribution function and the components of the fields are proportional to

$$e^{-iq\omega z \sin\theta/c}\, e^{-i\omega t}$$

Assuming specular reflection and following the procedure outlined in Section III, the first-order distribution function is found to be

$$f_1^+ = \frac{e}{mv}\frac{\partial f_o}{\partial v}\frac{e^{i\omega z/v_z}}{v_z}\Bigg\{v_x\left[\int_0^z e^{-i\omega\xi/v_z} E_x(\xi)d\xi + \int_0^\infty e^{i\omega\xi/v_z} E_x(\xi)d\xi\right]$$

$$+ v_y\left[\int_0^z e^{-i\omega\xi/v_z} E_y(\xi)d\xi + \int_0^\infty e^{i\omega\xi/v_z} E_y(\xi)d\xi\right]$$

$$+ v_z\left[\int_0^z e^{-i\omega\xi/v_z} E_z(\xi)d\xi - \int_0^\infty e^{i\omega\xi/v_z} E_z(\xi)d\xi\right]\Bigg\} \tag{84a}$$

for $v_z > 0$ and

$$f_1^- = -\frac{e}{mv}\frac{\partial f_o}{\partial v}\frac{e^{i\omega z/v_z}}{v_z}\sum_{k=1}^{3} v_k \int_z^\infty e^{-i\omega\xi/v_z} E_k(\xi)d\xi \tag{84b}$$

for $v_z < 0$, where $v = (v_x^2 + v_y^2 + v_z^2)^{\frac{1}{2}}$. Using Eq. (10) and introducing Maxwell's equations yield a set of three integral equations for the components of the electric field which can all be solved by Fourier transforms. The results yield propagating components of the fields, which are given by the usual classical or Fresnel equations with small corrections of order Δ, plus transient components of short range and small amplitude.

In discussing the second harmonic, the spatial derivatives of $f_1^{\pm}$ may again be ignored in the "source terms" for f_2 which yield three additional integral equations for the components of the second harmonic fields. The boundary conditions for oblique incidence no longer require a reflected harmonic to vanish, and the amplitude may, in principle, be calculated by the method described. However, even using the Fresnel relations for the fundamental fields yields extremely tedious first-order differential equations for the second harmonic current generators. Again, in order to match boundary conditions, it is necessary to find the residues at $z = 0$ from the poles of the transformed kernels as well as those at twice the fundamental propagation vector components. These have been shown to be approximately equal in magnitude. (The branch cut contributions will again be small.)

Using Eq. (83), the ratio of the energy flux reflected with frequency 2ω from the surface to the incident flux can be estimated from the present analysis to be

$$\frac{P_r}{P_i} \sim \left(\frac{4eE_i}{mc\omega_p}\right)^2 F(\theta, \frac{\omega_p}{\omega'}) \tag{85}$$

where θ is the incidence angle and F is of order unity with $F(0, \omega_p/\omega') = F(\pi/2, \omega_p/\omega') = 0$. For an incident laser beam of $E_i \sim 2.5 \times 10^6$v/meter, the fraction of the incident power reflected in the second harmonic is $\sim 10^{-14}$. For order-of-magnitude estimates of P_r/P_i, a plot of $(4eE_i/mc\omega_p)^2$ versus incident electric field E_i is shown in Fig. 3. For electric fields $\gtrsim 10^{11}$ v/meter, the metal will probably be damaged.

Brown, Parks and Sleeper,[3] using a ruby laser with 1 MW peak power and < 50 nsec duration shining on silver have obtained reasonable agreement with $F \propto \cos^4\theta$. They point out that these observations are consistent with a second harmonic polarization proportional to $\underline{E}_1(\nabla\cdot\underline{E}_1)$. This expression has arbitrarily been extracted from the known nonlinear polarization for a free electron gas where boundary effects are <u>not</u> included, which contains an additional term proportional to $\underline{E}_1 \times \underline{B}_1$, and which yields a much more complicated dependence on θ.

Jha[1,2] has attempted to include the boundary effects for oblique incidence by using an iteration procedure, perturbing about the classical Fresnel solutions. However, for the fundamental wave, his correction term, to lowest order in v_f/c, is of the same order of magnitude as the Fresnel solution. This result shows that, first of all, the iteration procedure is invalid near the surface. Secondly, the results of the analysis presented here indicate that the total correction to the Fresnel solutions are of order Δ, and one should obtain only the classical result as $v_f/c \to 0$.

For the second harmonic, Jha attributes the $\underline{E}_1(\nabla\cdot\underline{E}_1)$ term to a surface phenomenon, the $\underline{E}_1 \times \underline{B}_1$ expression to a volume contribution, and obtains a complicated expression for $F(\theta, \omega_p/\omega')$ in the absence of collisions. These correspond to the root of $1-\widetilde{K}_2(k) = 0$ and the residue at $k = 2k_c$, respectively. While such a partition enables one to estimate the correction in the metal fairly well, the procedure

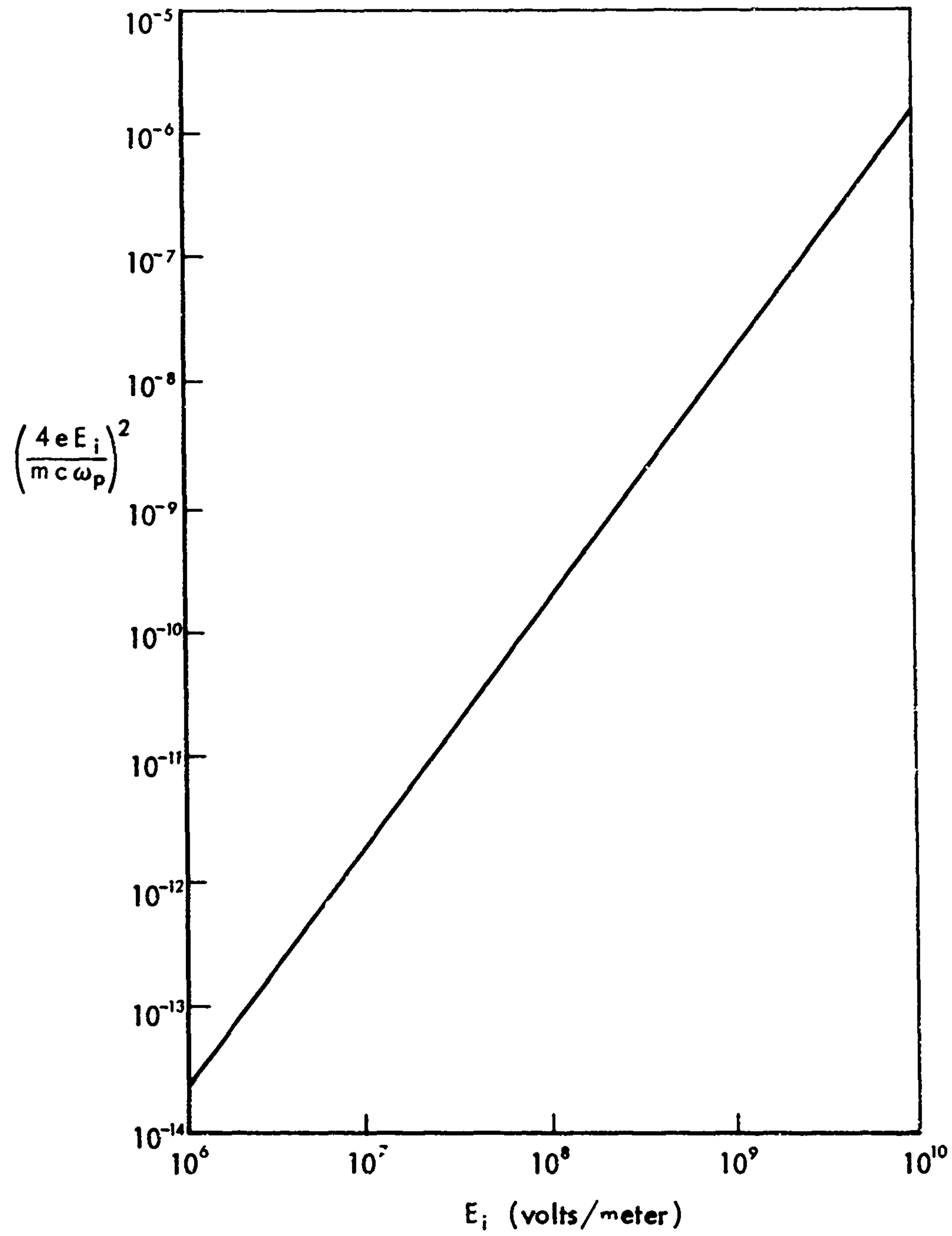

Fig. 3—Scale factor for the fraction of incident intensity reflected in the second harmonic

again breaks down near the surface. Here Jha has neglected terms in his iteration which are of the same order of magnitude as those he has retained. Basically, as has been shown, there are three terms to consider for the second harmonic. While the branch cut contribution is indeed negligible at the surface for $\omega \ll \omega_p$, it is necessary to include the residues from the transformed kernels in order to obtain better than the order-of-magnitude agreement predicted by Eq. (85).

The results presented are sufficient to obtain rough estimates of nonlinear heating and harmonic generation. While the amplitudes are small, it may still prove necessary to account for the second harmonic generation in applications where instabilities might be excited and grow to such amplitudes as to affect the operation of a device. The stability of a system when a wave of frequency 2ω is generated should therefore be considered and, when necessary, the growth rate should be estimated.

Appendix A

ROOTS OF THE LINEAR DISPERSION RELATION

With the restriction that f_o be a symmetric function of v_z, a valid representation for $K_T(k)$ with the same region of analyticity as Eq. (23b) is given by

$$K_T(k) = \frac{\omega}{\omega'} \frac{\omega_p^2}{n_o c^2} \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} dv_z \frac{f_o}{1 - (kv_z/\omega')^2} \qquad \text{(A-1)}$$

The zeros of D(k) are then roots of

$$1 - \left(\frac{kc}{\omega}\right)^2 = \frac{\omega_p^2}{n_o \omega \omega'} \iiint \frac{f_o}{1 - (kv_z/\omega')^2} dv_x dv_y dv_z \qquad \text{(A-2)}$$

Let $(kv_f/\omega')^2 = a + bi$, where a and b are real, and define $p = \nu/\omega$. Then $(kc/\omega)^2 = (c/v_f)^2 (a + bi)(1 - p^2 + 2ip)$. The general technique for locating the zeros of D(k) involves calculating the change in the argument of $(D'(k)/D(k))$ around a suitably chosen contour in the k plane. However, for all metals except at liquid helium temperatures, p is small for frequencies greater than $\sim 10^{13}$ cps. With this restriction, the real and imaginary parts of Eq. (A-2), for p = 0, are

$$1 - a(c^2/v_f^2) = \frac{\omega_p^2}{n_o \omega^2} \iiint f_o(v_x, v_y, \mu) \left[\frac{1 - a\mu^2}{(1 - a\mu^2)^2 + (b\mu^2)^2}\right] dv_x dv_y d\mu \qquad \text{(A-3)}$$

and

$$-b(c^2/v_f^2) = \frac{\omega_p^2}{n_o \omega^2} \iiint f_o(v_x, v_y, \mu) \left[\frac{\mu^2}{(1 - a\mu)^2 + (b\mu)^2}\right] dv_x dv_y d\mu \qquad \text{(A-4)}$$

respectively. The integrand in Eq. (A-4) is always positive, and this equation can then only be satisfied if b = 0. Equation (A-3) becomes

$$1 - a(c^2/v_f^2) = \frac{\omega_p^2}{n_o\omega^2} \iiint \frac{f_o(v_x,v_y,\mu)}{1-a\mu}\, dv_x dv_y d\mu \qquad \text{(A-5)}$$

which can readily be solved by sketching the left- and right-hand sides of the equation versus a. The left side is a straight line with y-intercept unity and a negative slope of $(c/v_f)^2$. The right-hand side is zero at $a = -\infty$ and has y-intercept $(\omega_p/\omega)^2$. Inspection of the denominator of Eq. (A-5) reveals that the slope is always positive for $a < 1$. (The region $a > 1$ corresponds to the branch cut.) Hence there exists one and only one intersection for $p = 0$.

For $(\omega_p/\omega)^2 < 1$, the root a_r is positive with $0 < a_r < (v_f/c)^2 \ll 1$. When $\omega < \omega_p$, the root is negative and $|a_r|$ can easily be seen to be less than 1 provided $\omega_p v_f/\omega c < 1$, or for frequencies greater than $\sim 10^{13}$ cps, which was our original assumption. (This is a sufficient, but not a necessary condition.)

For p nonzero but small, the root will have a small imaginary part, but its magnitude will remain < 1.

Appendix B

THE ZEROS OF $\tilde{K}_2(k) = 1$

The roots of $1 - \tilde{K}_2(k) = 0$ are the zeros of

$$\frac{4\omega\,\omega''}{\omega_p^2} = 3\int_0^1 \frac{x^2\,dx}{1 - \left(\frac{kv_f}{2\omega''}\right)^2 x^2} \tag{B-1}$$

For $|kv_f/2\omega''| << 1$ the integrand may be expanded and integrated term by term to yield

$$\left(\frac{kv_f}{2\omega''}\right)^2 \cong \frac{5}{3}\left[\frac{4\omega\,\omega''}{\omega_p^2} - 1\right] \tag{B-2}$$

The root given by Eq. (B-2) is not valid when $|\omega\,\omega''/\omega_p^2| \lesssim \frac{1}{10}$ or $>> 1$. The latter case is of little interest. A solution in the former case may be found by noting that a valid representation of Eq. (B-1) is

$$\frac{4\omega\,\omega''}{\omega_p^2} = 3p^2[1-p\tan^{-1}(1/p)] \tag{B-3}$$

where $p^2 = -(2\omega''/kv_f)^2$, and that branch of $\tan^{-1}(1/p)$ is chosen which equals $\pi/2$ at $p = 0$. Then for $|\omega\,\omega''/\omega_p^2| << 1$, Eq. (B-3) may be expanded about $p = 0$ to yield

$$\left(\frac{kv_f}{2\omega''}\right)^2 \cong -\frac{3\omega_p^2}{4\omega\,\omega''} \tag{B-4}$$

Let $(kv_f/2\omega'')^2 = a + bi$, where a and b are real, and consider the collisionless limit $\nu = 0$. Then $\omega'' = \omega$ and equating the imaginary parts yields $b = 0$. Sketching the left and right sides of

$$\frac{4\omega^2}{\omega_p^2} = 3\int_0^1 \frac{x^2dx}{1-ax^2} \tag{B-5}$$

reveals that one (and only one) intersection always exists whenever the right-hand side of Eq. (B-5) is defined (i.e., $-\infty < a < 1$).

Appendix C

THE RESIDUES AND BRANCH CUT CONTRIBUTION FOR THE SECOND HARMONIC

The residue of

$$\frac{2\alpha k\, \tilde{K}_2(k)}{[k^2-(2k_c)^2][1-\tilde{K}_2(k)]} \tag{C-1}$$

at $1-\tilde{K}_2(k) = 0$ is given by

$$\frac{2\alpha}{\left[1 - \left(\frac{2kc}{k}\right)^2\right] k \frac{\partial}{\partial k}\, [1-\tilde{K}_2(k)]} \tag{C-2}$$

First consider the range where Eq. (B-2) is valid. Then

$$1-\tilde{K}_2(k) \approx 1 - \frac{\omega_p^2}{4\omega\, \omega''}\left[1 + \frac{3}{5}\left(\frac{kv_f}{2\omega''}\right)^2\right]$$

Substituting from Eq. (B-2) for k^2 yields

$$k \frac{\partial}{\partial k}\, [1-\tilde{K}_2(k)] \approx - \frac{\omega_p^2}{4\omega\, \omega''} \cdot \frac{6}{5}\left(\frac{kv_f}{2\omega''}\right)^2 = - 2\left(1 - \frac{\omega_p^2}{4\omega\, \omega''}\right)$$

and the residue is

$$\frac{-\alpha}{\left[1 - \frac{3}{5}\left(\frac{\omega_p v_f}{\omega c}\right)^2 \left(\frac{\omega^3}{\omega'\omega''^2}\right)\left(\frac{\omega_p^2 - \omega\, \omega'}{\omega_p^2 - 4\omega\, \omega''}\right)\right]\left[1 - \frac{\omega_p^2}{4\omega\, \omega''}\right]} \simeq \frac{-\alpha}{1 - \frac{\omega_p^2}{4\omega\, \omega''}} \tag{C-3}$$

When $|\omega\, \omega''/\omega_p^2| \ll 1$, we use Eq. (B-4) which gives

$$1-\tilde{K}_2(k) \approx 1 + \frac{3\omega_p^2}{k^2 v_f^2}\frac{\omega''}{\omega}, \qquad k\frac{\partial}{\partial k}\left[1-\tilde{K}_2(k)\right] = 2$$

and the residue is

$$\frac{\alpha}{1 - \frac{4}{3}\left(\frac{v_f}{c}\right)^2 \frac{\omega^2}{\omega'\omega''}\left(1 - \frac{\omega\,\omega'}{\omega_p^2}\right)} \approx \alpha \qquad \text{(C-4)}$$

The residue of Eq. (C-1) at $k = 2k_c$ is

$$\frac{\alpha\tilde{K}_2(2k_c)}{1-\tilde{K}_2(2k_c)} \qquad \text{(C-5)}$$

When Eq. (B-2) is valid the residue is

$$\frac{\frac{\omega_p^2}{4\omega\,\omega'''}(1+s)\alpha}{1 - \frac{\omega_p^2}{4\omega\,\omega''}(1+s)} \approx \frac{\frac{\omega_p^2}{4\omega\,\omega''}\alpha}{1 - \frac{\omega_p^2}{4\omega\,\omega''}} \qquad \text{(C-6)}$$

where $s = \frac{3}{5}\left(\frac{\omega v_f}{\omega'' c}\right)^2\left(1 - \frac{\omega_p^2}{\omega\,\omega'}\right)$ can be neglected.

When Eq. (B-4) is valid the residue is

$$\frac{1 - 3\left(\frac{\omega'' c}{\omega v_f}\right)^2}{3\left(\frac{\omega''}{\omega}\frac{c}{v_f}\right)^2} \approx -\alpha \qquad \text{(C-7)}$$

In a manner identical to that used to derive Eq. (41), the contribution to the branch cut, E_{Br}, is

$$2\pi E_{Br}(z) = e^{i\delta'} \int_{2\Omega'/v_f}^{\infty} \frac{\gamma(\rho)[\tilde{K}_2^+ - \tilde{K}_2^-] e^{iz\rho e^{i\delta'}}}{(1-\tilde{K}_2^+)(1-\tilde{K}_2^-)} d\rho \qquad \text{(C-8)}$$

where

$$\tilde{K}_2^+ - \tilde{K}_2^- = \frac{6\pi i \omega_p^2}{v_f^3 \omega} e^{-i\delta'} \frac{\Omega'^2}{\rho^3} H[v_f - 2\Omega'/\rho] \qquad \text{(C-9)}$$

$\delta' = \tan^{-1}(\nu/2\omega)$, $\Omega' = (\omega^2+\nu^2/4)^{\frac{1}{2}}$ and $\gamma(\rho) = \dfrac{-2\alpha\rho e^{i\delta'}}{\rho^2 e^{2i\delta'} - (2k_c)^2}$. Then

$$E_{Br}(0) = -\frac{6i\alpha\omega_p^2\Omega'^2}{\omega v_f^3} e^{i\delta'} \int_{2\Omega'/v_f}^{\infty} \frac{d\rho}{\rho^2[\rho^2-(2k_c)^2 e^{-2i\delta}](1-\tilde{K}_2^+)(1-\tilde{K}_2^-)} \qquad \text{(C-10)}$$

With the substitution $\mu = 2\Omega'/v_f\rho$, Eq. (C-10) becomes

$$E_{Br}(0) = -\frac{3i\alpha\omega_p^2}{4\omega\Omega'} \int_0^1 \frac{\mu^2 \, d\mu}{\left[1 - \left(\frac{k_c v_f}{\Omega'}\right)^2 \mu^2 e^{-2i\delta'}\right] (1-\tilde{K}_2^+(\mu))(1-\tilde{K}_2(\mu))} \qquad \text{(C-11)}$$

But $|k_c v_f/\Omega'|^2 = 5\Delta\ (\omega^2+\nu^2)/(\omega^2+\nu^2/4) \ll 1$, and $K_2^{\pm} \propto \omega_p^2/\omega\Omega'$, which we have assumed $\gg 1$. Then $E_{Br}(0) \approx \alpha\omega\Omega'/\omega_p^2$. Hence the ratio of the contributions from the branch cut to the residue is $\sim \omega\Omega'/\omega_p^2$.

REFERENCES

1. Jha, S., "Theory of Optical Harmonic Generation at a Metal Surface," Phys. Rev., Vol. 140, 1965, p. A2020.

2. Jha, S., "Nonlinear Optical Reflection From a Metal Surface," Phys. Rev. Letters, Vol. 15, 1965, p. 412.

3. Brown, F., R. E. Parks, and A. M. Sleeper, "Nonlinear Optical Reflection From a Metallic Boundary," Phys. Rev. Letters, Vol. 14, 1965, p. 1029.

4. Cheng, H., and P. B. Miller, "Nonlinear Optical Theory in Solids," Phys. Rev., Vol. 134, 1964, p. A683.

5. Stratton, J., Electromagnetic Theory, McGraw Hill Book Co., Inc., New York, 1941.

6. Taylor, E. C., "Excitation of Longitudinal Waves in a Bounded Collisionless Plasma," Phys. Fluids, Vol. 6, 1963, p. 1305; also, E. C. Taylor, "The Theory of Wave Excitation in Bounded Rarefied Plasmas," PhD Thesis, University of California at Los Angeles, 1962.

7. Kittel, C., Introduction to Solid State Physics, John Wiley & Sons, Inc., New York, 1962.

8. Reuter, G., and E. Sondheimer, "The Theory of the Anomalous Skin Effect in Metals," Proceedings of the Royal Society, Vol. 195A, 1948, p. 336.

9. Wilson, A. H., The Theory of Metals, Cambridge University Press, Cambridge, 1936.

10. Shafranov, J. D., "Propagation of an Electromagnetic Field in a Medium with Spatial Dispersion," JETP (USSR), Vol. 6, 1958, p. 1019.

11. Ziman, J. M., Electrons and Phonons, Oxford University Press, London, 1962.

12. Cohen, M. H., and V. Heine, "Electronic Band Structures of the Alkali Metals and of the Noble Metals and Their α-Phase Alloys," Advances in Physics, Vol. 7, 1958, p. 395.

DOCUMENT CONTROL DATA

1 ORIGINATING ACTIVITY	2a. REPORT SECURITY CLASSIFICATION
THE RAND CORPORATION	UNCLASSIFIED
	2b. GROUP

3. REPORT TITLE

OPTICAL HARMONIC GENERATION AT A METAL SURFACE

4. AUTHOR(S) (Last name, first name, initial)

Lutomirski, R. F.

5. REPORT DATE	6a. TOTAL No. OF PAGES	6b. No. OF REFS.
March 1968	50	12

7. CONTRACT OR GRANT No.	8. ORIGINATOR'S REPORT No.
DAHC15 67 C 0141	RM-5595-ARPA

9a AVAILABILITY/LIMITATION NOTICES	9b. SPONSORING AGENCY
DDC-1	Advanced Research Projects Agency

10. ABSTRACT

An analysis of the interaction of a high intensity laser and a metal in terms of the optical harmonic generation at the metal surface. Using a simple model for the statistics of the conduction electrons and their interaction with the surface, Maxwell's equations and the Boltzmann equation are solved self-consistently for the fields in the metal, yielding the usual Fresnel solutions plus correction terms which, at optical frequencies and above liquid helium temperatures, are shown to be of the order of 1 percent. When laser light of sufficient intensity is used, a nonlinear polarization is induced and waves of twice the fundamental frequency are produced. For the indicated model and for normal incidence, the second harmonic waves in the metal are calculated and the magnitude of the relative heating is computed. The method for solving the general problem for an arbitrary incidence angle is outlined and the ratio of the average energy flux in the second harmonic to the incident flux is estimated.

11. KEY WORDS

Mathematical physics
Lasers
Materials
Wave propagation