APPENDIX C: APPLICATION OF **RPA SCREENING**

Because of our lack of faith in the linear screening of strong potentials,³² we first isolate the long-range part of the ionic potential $v_{LR}(r)$ which is defined as follows:

$$v_{\rm LR}(r) = -4r^{-1}\{1 - e^{-r/r_c}\}$$

where r_c is the average of the $\{r_{lc}\}$ introduced in

³² N. H. March and A. D. Boardman, J. Phys. Soc. Japan Suppl. II, 18, 80 (1963).

Eq. (37). $v_{LR}(r)$ was Fourier transformed to obtain $v_{LR}(q)$ which was then divided by the exchangecorrected RPA dielectric constant¹⁶ $\epsilon_{RPA}(q)$ to obtain the screened long-range potential $v_{SLR}(q)$. Using the fact that only values of $v_{SLR}(q)$ for **q** equal to nonzero reciprocal-lattice vectors are relevant to the band structure the irrelevant values of $v_{SLR}(q)$ were adjusted to further reduce the range of $v_{SLR}(r)$ which was finally obtained by Fourier transform. $v_{SLR}(r)$ was then recombined with the short-range part to produce the curve labeled RPA in Fig. 2.

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Free-Carrier Optical Nonlinearity Due to Carrier Scattering and Nonparabolicity

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Nonlinear mixing of electromagnetic waves in low-temperature degenerate semiconductors has been analytically investigated. Usual kinetic-theory techniques have been employed for evaluating the mixedfrequency components in the current density. Two types of free-carrier nonlinearity are considered, one arising from carrier scattering processes and the other due to nonparabolicity of conduction band. Numerical results, comparing the two nonlinearities for the special case of mixing of two CO2 laser beams in indium antimonide, have been presented at the end.

1. INTRODUCTION

YONLINEAR harmonic generation and mixing of electromagnetic waves in semiconductors has been extensively studied both experimentally¹⁻⁵ and theoretically.⁶⁻¹⁴ It is well established now that at microwave and lower frequencies the free-carrier contribution to nonlinearity dominates the contribution arising from polarization of background lattice; the latter contribution seems to become important, in general, only at optical frequencies.¹⁵ Recent experi-

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- (1966). ¹² V. V. Paranjape and H. C. Law, Phys. Letters 25A, 146 (1967).
- ¹³ P. K. Kaw, Phys. Rev. Letters 21, 539 (1968).
- ¹⁴ M. S. Sodha and S. Sharma, J. Phys. C 2, 914 (1969).
 ¹⁵ N. Bloembergen and Y. R. Shen, Phys. Rev. 141, 298 (1966).

ments with indium antimonide and indium arsenide,⁵ however, indicate that for these materials free-carrier nonlinearity is the dominant nonlinearity, even at CO_2 laser frequencies (wavelengths 9.6 μ and 10.6 μ). Although this conclusion has been challenged,¹⁶ there can be no doubt that for many III-V compounds the freecarrier contribution to nonlinear phenomena is very important even at optical frequencies. In this paper we devote our attention exclusively to this type of nonlinearity in semiconductors.

Nonlinear phenomena of free carriers arise because of two different effects: (a) energy dependence of carrier relaxation time^{6-10,13,14} and (b) nonparabolicity of conduction band.^{11,12,17,18} For a semiconductor with a nonparabolic conduction band, both of these effects occur; however, the current theories⁶⁻¹³ of nonlinear harmonic generation and mixing seem to be restricted to either one or the other of these effects. It is the explicit purpose of the present paper to remove this restriction. Following Matz,17 the Boltzmann transfer equation for free carriers in a uniform isotropic semiconductor with spherical nonparabolic energy bands has been set up and solved by the usual Legendre-poly-

¹⁶ S. S. Jha and N. Bloembergen, Phys. Rev. 171, 891 (1968).
¹⁷ D. Matz, J. Phys. Chem. Solids 28, 373 (1967).
¹⁸ I. Licea, Phys. Status Solidi 25, 461 (1968); 26, 115 (1968).

nominal expansion methods; an explicit expression has been obtained for the mixed-frequency component $\omega_3 = 2\omega_1 - \omega_2$ in the current density when two electric vectors of frequencies ω_1 and ω_2 are acting on the semiconductor. The theory is restricted to relatively low temperatures ($\leq 80^{\circ}$ K) and ionized impurity scattering has been considered to be the sole mechanism of carrier scattering; the experiments of Kinch¹⁹ on electron mobilities in *n*-type indium antimonide clearly demonstrate the dominance of ionized impurity scattering at these low temperatures.

Numerical results have been presented for the mixing of two CO_2 laser beams in indium antimonide and special emphasis is given to a quantitative comparison of the two types of free-carrier nonlinearity.

2. MIXED-FREQUENCY COMPONENTS IN CURRENT DENSITY

The Boltzmann transfer equation for electrons in a homogeneous semiconductor may be written as

$$\partial f/\partial t - (e\mathbf{E}/\hbar) \cdot \nabla_k f = (\partial f/\partial t)_c,$$
 (1)

where $\mathbf{E} = \mathbf{E}_1 e^{i\omega_1 t} + \mathbf{E}_2 e^{i\omega_2 t}$ is the applied electric vector and the rest of the symbols have their usual meanings. We are ignoring the $v \cdot \nabla f$ term in Eq. (1) even though we know that the wave field is inhomogeneous in space; the assumption that this term can be neglected is justified if the amplitude of the field varies only slightly over the so-called energy relaxation length $(\bar{v}/v\delta^{1/2})$, where \bar{v} is the average carrier speed, ν is the collision frequency, and δ is the fraction of carrier energy lost in each collision. When the dominant form of carrier scattering is ionized impurity scattering, one can always define a carrier momentum relaxation time τ and replace the right side of Eq. (1) by

$$(\partial f/\partial t)_c = -(f - f^0)/\tau, \qquad (2)$$

where f^0 is the distribution to which f relaxes. An explicit expression for τ is given later.

Instead of denoting the energy-momentum relation by $\mathcal{E}(k)$ as usual, we use the more convenient relation¹⁷ of \mathcal{E} in terms of k; viz.,

$$\frac{\hbar^2 k^2}{2m_n = \gamma(\mathcal{E})}, \qquad (3)$$

where m_n is the carrier effective mass at the band edge and $\gamma(\mathcal{E}) \to \mathcal{E}$ as $\mathcal{E} \to 0$. The momentum effective mass for the carriers, appearing in the Boltzmann equation, is then defined by

$$m^* = \hbar k / v, \qquad (4a)$$

and for a spherical band is related to m_n by the relation

$$m^* = m_n \gamma', \qquad (4b)$$

where γ' denotes $d\gamma/d\mathcal{E}$.

We assume that the electric vectors \mathbf{E}_1 and \mathbf{E}_2 are oriented along the same direction (the *x* axis). When the disturbance in the semiconductor is in one direction only, there exists an axis of symmetry and one can expand the distribution function of carrier wave vectors in terms of Legendre polynomials. Thus, we write

$$f(\mathbf{k}) = \sum_{l=0}^{\infty} f^l(k) P_l(\cos\theta),$$

where $\cos\theta = k_x/k$. Substituting this expansion in Eq. (1) and using the orthogonality and normalization properties of Legendre polynomials, one obtains a set of coupled equations for f^0 , f^1 , f^2 , ..., etc. In the present analysis we shall only retain terms up to f^2 ; earlier workers^{8,20} have established that this is necessary as well as sufficient for deriving correct lowest-order expressions for the mixed-frequency component $\omega_3 = 2\omega_1 - \omega_2$ in the current density. The coupled equations for f^0 , f^1 , and f^2 are

$$\frac{\partial f^{0}}{\partial t} - \frac{2}{3} \frac{eE}{(2m_{n})^{1/2}} \frac{1}{\gamma' \sqrt{\gamma}} \frac{\partial}{\partial \mathcal{E}} (\gamma f^{1}) = \left(\frac{\partial f^{0}}{\partial t}\right)_{c}, \quad (5a)$$

$$\frac{\partial f^{1}}{\partial t} - \frac{2eE}{(2m_{n})^{1/2}} \frac{\sqrt{\gamma}}{\gamma'} \times \left[\frac{\partial f^{0}}{\partial \mathcal{E}} + \frac{2}{5} \frac{\partial f^{2}}{\partial \mathcal{E}} + \frac{3}{5} \frac{\gamma'}{\gamma} f^{2} \right] = -\frac{f^{1}}{\tau}, \quad (5b)$$

and

$$\frac{\partial f^2}{\partial t} - \frac{4}{3} \frac{eE}{(2m_n)^{1/2}} \frac{\gamma}{\gamma'} \frac{\partial}{\partial \mathcal{E}} \left(\frac{f^1}{\sqrt{\gamma}} \right) = -\frac{f^2}{\tau}.$$
 (5c)

 $(\partial f^0/\partial t)_c$ denotes the rate of change of f^0 due to scattering processes, and corresponds to some higher-order terms not included in Eq. (2). It can be shown that inclusion of these terms do not modify the expressions for the mixed components of current density to the lowest order in the present expansion scheme (which is essentially an expansion in the ratio of drift to thermal velocities²¹).

When the applied electric vector has two components at frequencies ω_1 and ω_2 , the time dependences of f^0 , f^1 , and f^2 are identical to those discussed by Sodha and Kaw²⁰ for mixing of waves in an ionized gas. Substituting these time dependences in Eqs. (5a)–(5c), equating the coefficients of various frequency terms on both sides of each equation, and making use of the assumption that

$$f^{1}, f^{2}, f^{0}(2\omega_{1}, \omega_{1}\pm\omega_{2}, \text{etc.}) \ll f_{0}^{0},$$

$$f^{1}(3\omega_{1}, 2\omega_{1}\pm\omega_{2}, \text{etc.}) \ll f^{1}(\omega_{1}) \text{ or } f^{1}(\omega_{2})$$

¹⁹ M. A. Kinch, Brit. J. Appl. Phys. 17, 1257 (1966).

²⁰ M. S. Sodha and P. K. Kaw, Proc. Phys. Soc. (London) 88, 373 (1966).

²¹ J. Yamashita and K. Inoue, J. Phys. Chem. Solids 12, 1 (1959).

(which corresponds to assuming that the amplitudes of generated harmonic and mixed-frequency components are much less than those of the fundamental frequency

components), one can derive an expression for the $(2\omega_1-\omega_2)$ frequency component of f^1 . The relevant expression is

$$f_{12}^{1-} = \frac{2e^{3}E_{1}^{2}E_{2}^{*}}{3(2m_{n})^{3/2}} \left(\frac{\tau\sqrt{\gamma}}{\gamma'[1+i\tau(2\omega_{1}-\omega_{2})]} \right) \left[\frac{\partial}{\partial\mathcal{S}} \left\{ \frac{1}{i(\omega_{1}-\omega_{2})\gamma'\sqrt{\gamma}} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\gamma^{3/2}}{\gamma'} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \left(\frac{1}{1+i\omega_{1}\tau} + \frac{1}{1-i\omega_{2}\tau} \right) \right] \right. \\ \left. + \frac{4}{5} \frac{\tau\gamma}{\gamma'[1+i(\omega_{1}-\omega_{2})\tau]} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\tau}{\gamma'} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \left(\frac{1}{1+i\omega_{1}\tau} + \frac{1}{1-i\omega_{2}\tau} \right) \right] + \frac{1}{2i\omega_{1}\gamma'\sqrt{\gamma}} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\gamma^{3/2}\tau}{\gamma'(1+i\omega_{1}\tau)} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \right] \right. \\ \left. + \frac{4}{5} \frac{\tau\gamma}{\gamma'(1+2i\omega_{1}\tau)} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\tau}{\gamma'(1+i\omega_{1}\tau)} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \right] \right\} + \frac{6}{5} \frac{\tau}{1+i(\omega_{1}-\omega_{2})\tau} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\tau}{\gamma'} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \left(\frac{1}{1+i\omega_{1}\tau} + \frac{1}{1+i\omega_{1}\tau} \right) \right] \\ \left. + \frac{6}{5} \frac{\tau}{1+2i\omega_{1}\tau} \frac{\partial}{\partial\mathcal{S}} \left[\frac{\tau}{\gamma'} \frac{\partial f_{0}^{0}}{\partial\mathcal{S}} \left(\frac{1}{1+i\omega_{1}\tau} \right) \right] \right], \quad (6)$$

where the * over E_2 denotes its complex conjugate.

For f_0^0 we use its lowest-order value for degenerate semiconductors; viz., the usual Fermi-Dirac distribution

$$f_0^0 = \left[1 + \exp\left(\frac{\mathcal{E} - \mathcal{E}_F}{KT}\right)\right]^{-1},$$

where \mathcal{E}_F is the Fermi energy of carriers in the conduction band.

The density of energy states in the present representation is given by

$$N(\mathcal{E})d\mathcal{E} = \frac{(2m_n)^{3/2}}{2\hbar^3} \gamma' \gamma^{1/2} d\mathcal{E}.$$

Using this equation one can readily write down an expression for the current density in terms of f^1 . This expression is

$$J = -\frac{2en}{3(2m_n)^{1/2}} \int_0^\infty \gamma f^1 d\mathcal{E} \bigg/ \int_0^\infty \gamma' \gamma^{1/2} f_0^0 d\mathcal{E}, \quad (7)$$

where n is the carrier concentration in the semiconductor.

In order to carry out the relevant integrations, we must now choose a specific form for $\gamma(\mathcal{E})$. Let us investigate the form of $\gamma(\mathcal{E})$ for low-gap semiconductors like indium antimonide. Following Kane's model²² for low-gap semiconductors, the energy of an electron is related to its wave vector k by the relation

$$\mathcal{E}(k) = \hbar^2 k^2 / 2m_0 - \frac{1}{2} \mathcal{E}_g + \frac{1}{2} \left[\mathcal{E}_g^2 + (8/3) P^2 k^2 \right]^{1/2}, \qquad (8)$$

where m_0 is the free-electron mass, E_g is the band gap, and P is a matrix element defined by Kane as

$$P = -i(\hbar/m_0) \langle S | p_3 | X_3 \rangle,$$

²² E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957); H. Ehrenreich, *ibid*. 2, 131 (1957). in which S is the spatial part of conduction-band wave function, p_3 is the Z component of momentum operator, and X_3 is one of the triply degenerate valence-band functions. Ehrenreich²² has shown that P can be approximated by

$$P \simeq \frac{1}{2} \hbar \left(\frac{3 \mathcal{S}_{g}(m_{0} - m_{n})}{m_{0} m_{n}} \right)^{1/2}.$$
 (9)

When the effective mass $m^* \ll m_0$, one can neglect²³ the first term on the right side of Eq. (8) and m_n in comparison with m_0 in Eq. (9), and obtain

$$k^2 = (2m_n/\hbar^2)(\mathcal{E} + \mathcal{E}^2/\mathcal{E}_g),$$

which may be rewritten as

$$\gamma(\mathcal{E}) = \mathcal{E} + (\mathcal{E}^2/\mathcal{E}_g) . \tag{10}$$

In a more general case, one can retain this power-series representation of $\gamma(\mathcal{E})$ in terms of \mathcal{E} , with \mathcal{E}_g replaced by a general constant \mathcal{E}_1 .

We must also give an explicit expression for the carrier momentum relaxation time. For dominant ionized impurity scattering, one may write¹⁹

$$\tau = \frac{K_0^2 (2m^*)^{1/2} \mathcal{E}^{3/2}}{\pi e^4 N_{ig}(\mathcal{E})} = \tau_0 \epsilon^{3/2} (2\epsilon + \epsilon_g)^{1/2}, \qquad (11)$$

where $\epsilon = \mathcal{E}/KT$, K_0 is the dielectric constant of the material, N_i is the concentration of impurities, $g(\mathcal{E})$ is a slowly varying function of carrier energy and will be assumed to be constant in the present analysis and

$$\tau_0 = \frac{(KT)^2 (2m_n)^{1/2} K_0^2}{\pi e^4 N_{ig} \mathcal{E}_g^{1/2}}.$$

Substituting for f_{12}^{1-} in Eq. (7), using the definitions Eqs. (10) and (11), of γ and τ , and assuming that $\omega_1 \tau$,

²² T. C. Harman, J. M. Honig, and B. M. Tarmy, J. Phys. Chem. Solids 24, 835 (1963).

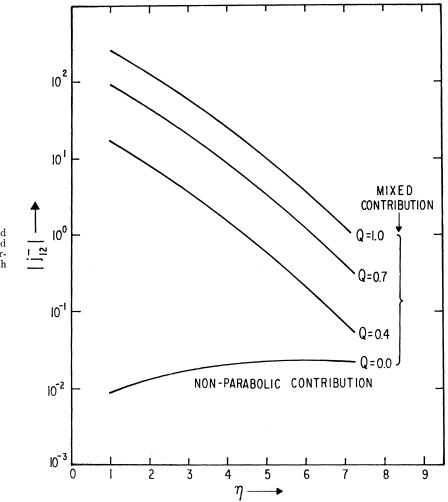


FIG. 1. Variation of $|j_{12}^{-}|$ [defined by Eq. (14)] with $\eta = (\mathcal{E}_F/KT)$ and $Q = (\omega_2 \tau_0)^{-1}$ for $(\omega_1/\omega_2) = 0.91$ (corresponding to CO₂ laser beams with wavelengths 10.6 μ and 9.6 μ).

 $\omega_2 \tau$, $(\omega_1 - \omega_2) \tau \gg 1$, one obtains

$$J_{12}^{-} = \alpha A E_2^* E_1^2 \exp[i(2\omega_1 - \omega_2)t], \qquad (12)$$

where $\alpha = (ne^4 \mathcal{E}_{\theta}^2/2m_n^2\omega_2^3K^3T^3)$, A is a dimensionless complex quantity involving definite integrals of the type

$$I_{r^{p,q}} = \int_{0}^{\infty} \frac{(e^{\epsilon - \epsilon_{F}})(\epsilon^{2} + \epsilon \epsilon_{g})^{p}}{[1 + \exp(\epsilon - \epsilon_{F})]^{2} \epsilon^{q} (2\epsilon + \epsilon_{g})^{r}} d\epsilon \quad (13)$$

(which result from integration over the energy-dependent carrier relaxation time and the carrier effective mass), p, q, and r may be integers or half-integers, and $\epsilon_F = \mathcal{E}_F/KT$. The explicit expression for A has not been presented because of its extremely cumbersome form.

3. DISCUSSION

The definite integrals defined by Eq. (13), for various values of p, q, and r were evaluated numerically. These integrals are required for the evaluation of A and hence that of J_{12}^{-} .

Figure 1 illustrates the variation of

$$|j_{12}^{-}| = |J_{12}^{-}/\alpha E_{1}^{2} E_{2}^{*} e^{i(2\omega_{1} - \omega_{2})t}|$$
(14)

with $\eta \equiv \epsilon_F = (\mathcal{E}_F/KT)$ for various values of $Q = (\omega_2 \tau_0)^{-1}$ and for the special case of the mixing of two CO2 laser beams (wavelengths 10.6μ and 9.6μ) in indium antimonide. The curve with Q=0 gives the contribution due to the band nonparabolicity (NP) alone; successively higher values of Q correspond to higher and higher contributions from energy-dependent scattering (EDS). It is noted that even for Q=0.1, EDS gives a large contribution to the mixed current density. For a temperature of 80°K and carrier concentrations in excess of 10¹⁶ cm⁻³ in indium antimonide (i.e., the regime where O is in excess of 0.1), one should thus be able to observe the EDS contribution to mixing (notice that this conclusion rests on the assumption that the dominant form of carrier scattering is the ionized impurity scattering). At lower temperatures, carrier concentrations less than 10¹⁶ cm⁻³ may be high enough for giving sizable EDS contributions.

The density and temperature dependences of the NP and EDS contributions to the mixed current density can be obtained from the variation of $|j_{12}|$ with $\eta = \mathcal{E}_F/KT$ in the two limiting cases, Q=0 and Q=1.0, respectively. We would like to emphasize however, that while looking at the temperature dependence of the EDS contribution, one should keep in mind the restricted temperature range ($\sim 30-80^{\circ}$ K) for which the theory is applicable.

In the general case, it is difficult to state the explicit dependences of the NP and EDS contributions on temperature. However, in the case when the EDS contribution is a small correction to the NP contribution, one may be able to distinguish between the two, by noticing from the analysis that the temperature dependence of EDS contribution is close to the temperature dependence of NP contribution multiplied by the temperature dependence of $(\omega_2 \tau_0)^{-1}$. Thus the dominant scattering mechanism determines the distinction between the temperature dependences of NP and EDS contributions.

Wynne⁵ has recently carried out an interesting experimental investigation on optical mixing in GaAs, Ge, Si, and InAs and has drawn some very useful conclusions about the nonparabolicity of the conduction band in GaAs. He has however, summarily rejected the EDS mechanism of optical mixing in his paper. It is quite conceivable that the parameter range of his experiments (especially the low carrier concentration) is such that the EDS contribution is actually negligible. However, it is quite clear that if one goes to a different parameter range (such as the one studied in this paper), then the EDS contribution may become comparable to the NP contribution and one will have to take it into account to draw correct conclusions about band nonparabolicity.

We conclude, finally, that optical mixing by free carriers in semiconductors may contain comparable contributions from conduction-band nonparabolicity and an energy-dependent scattering time.

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Photoelectric Emission from Silicon

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Effects of band structure on photoelectric yields from silicon have been determined from the interpretation of measurements which were made on surfaces oriented perpendicular to (111), (110), and (100). It was established that the yield spectrum for properly polished and annealed silicon is the same as that for cleaved and annealed silicon; low-energy electron diffraction patterns were obtained for all surfaces. Photoelectric thresholds obtained for (111), (110), and (100) silicon were 4.60, 4.73, and 5.11 eV, respectively. Improved experimental procedures show that yields well above threshold vary more rapidly with photon energy than those previously observed. Crystallographic differences in yields near threshold are ascribed to two sources: (1) the ionization energy ξ , which represents surface-barrier energy, and (2) the additional energy E_A required to emit electrons with crystal momentum (k) at an angle θ to the surface normal, beyond that for k at $\theta = 0$. It is shown that for transitions near the center of the Brillouin zone (BZ), E_A can be quite small (~0.15 eV); hence, electrons at large θ can be emitted for $h\nu - h\nu_0$ only a few tenths of an eV, where $h\nu_0$ is the threshold photon energy. For $(h\nu - h\nu_0) > 1.0$ eV, electrons are emitted for all θ , i.e., for all excitation within the inner half of the BZ. A quantitative estimate of crystallographic yield dependence gives good agreement for the assumption, suggested by the band structure, of transitions near threshold peaked for k along (111). These considerations lead to a more accurate interpretation of Y versus $(h\nu - h\nu_0)$, which depends on the position in the BZ of the operant optical transition. Such effects must be considered in analyzing all photoemission experiments, including both yield and energy distribution measurements.

I. INTRODUCTION

IN terms of one-electron solid-state theory, the volume photoelectric process consists of three stages: (1) excitation of an electron to an upper energy band by absorption of electromagnetic radiation; (2) motion of "hot carriers" in the upper band with elastic and inelastic scattering; (3) emission through the potential barrier at the surface of those electrons which have sufficient energy and the proper crystal momentum. Investigators interested in band structure information concentrate on stage 1 generally by measuring energy distribution of emitted electrons¹; those interested in surface barriers concentrate on stage 3 generally by measuring total photoelectron yield versus photon energy.²

¹ Typical recent papers are J. L. Shay and W. E. Spicer, Phys. Rev. 161, 799 (1967); T. A. Callcott, *ibid*. 161, 746 (1967); T. E. Fischer, *ibid*. 147, 603 (1966); F. G. Allen and G. W. Gobeli, *ibid*. 144, 558A (1966).

¹⁴⁴, 5384 (1960).
² Some typical papers are J. van Laar and J. J. Scheer, Surface Sci. 3, 189 (1965); F. G. Allen and G. W. Gobeli, Phys. Rev. 127, 150 (1962); J. van Larr and J. J. Scheer, Philips Res. Repts. 17, 101 (1962); J. A. Dillon, Jr., and H. E. Farnsworth, J. Appl. Phys. 29, 1195 (1958).