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Intense-field effects in solids*

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The interaction of solids with electromagnetic radiation is investigated using nonconventional theoretical techniques. Interband transitions in solids are investigated with a Volkov-type wave function used to represent the conduction-band electron in a calculation of the multiphoton interband transition rate induced by circularly polarized electromagnetic radiation of arbitrary intensity. The behavior of the transition rate is shown to be identical to that obtained from perturbation theory in the low-intensity domain, but deviates sharply from conventional weak-field characteristics for high intensities. Intense-field behavior in solids is found to have an earlier onset than in atoms.

I. INTRODUCTION

Multiphoton absorption has been studied in solids both experimentally[1-4] and theoretically.[5-7] In most cases the intensity of the irradiating field of laser was no more than 10^{10} W/cm^2. In this intensity range, where low-order processes dominate, perturbation theory generally agrees with the experimental data. Pulsed laser systems are now readily available which can produce focused intensities of 10^{14} W/cm^2 and more. Little theoretical or experimental work has been reported with solids in this intensity domain. Although a power-series expansion in intensity is exact if summed to all orders, the practical difficulties which arise are formidable, and, in general, assumptions which are appropriate only in the low-intensity domain must be applied to perform the calculation.

The multiphoton transition rate for excitation of a valence electron into the conduction band of a solid initiated by a linearly polarized electromagnetic field has been reported in the often-quoted work of Keldysh.[8] A Volkov-type wave function[9] was used for the final state. A Volkov wave function is the exact solution of the Schrödinger equation for a free electron in a plane-wave electromagnetic field. The transition rates were obtained with the assumption of low frequency and high photon numbers. Therefore, the disagreement of Keldysh's results in the low-intensity limit with perturbation theory, in most instances, is not surprising. As expected, for high intensities and the frequency of electric field approaching zero, Keldysh's result reduces to an expression describing the tunnel effect initiated by a static field.

The essential difference between the work of Keldysh and the investigation reported here is that, by using a different gage than Keldysh for the electromagnetic field, we have been able to achieve manageable closed-form analytical results without making assumptions about low frequencies or high photon order. Thus we obtain results which reduce to lowest-order perturbation theory when the low-intensity limit is taken.

Excitation of a valence electron into the conduction band of a wide-band-gap semiconductor or an insulator in the presence of a circularly polarized electromagnetic field of arbitrary intensity is investigated here. An electron in a solid in the effective-mass representation has properties which resemble those of a free electron. This is particularly true for conduction-band electrons which are not strongly localized to the atomic sites of the crystal. Therefore, it is assumed that a conduction-band electron in an electromagnetic field can be represented by a Volkov-type[9] wave function. The dipole approximation is invoked for the radiation field. For a description of the electron states in the solid, the single-particle representation is used with the effective-mass approximation. The energy surfaces of the valence and conduction bands are assumed to be spherical.

S-matrix formalism,[10] where the final state is the field-dependent state, is used in Sec. I to calculate the transition rate for N-photon excitation of a valence electron into the conduction band. Closed-form analytical results are obtained for arbitrary values of the intensity. The limiting cases for both low and high intensities are discussed. Numerical computations are also presented.

Although there have been many publications on interband-transition-rate calculations, there is a scarcity of work which employs circularly polarized light. Therefore, in Sec. II, the transition

rate for multiphoton excitation is calculated using standard perturbation theory for a circularly polarized electromagnetic field. Again, the radiation field is assumed to be independent of position, and Bloch functions are used to represent the electron states of the solid. The infinite sum over intermediate states is carried out to obtain closed-form results for the perturbative multiphoton transition rate. This is compared with the low-intensity limit obtained from the Volkov approximation in Sec. I.

Natural units ($\hbar = c = 1$) are used throughout this investigation unless another system of units is explicitly specified.

I. TRANSITION RATES FROM VOLKOV APPROXIMATION

Since the conduction electron has more free-electron character than the valence electron, the S matrix[10] which describes a transition between states i and f in the presence of an electromagnetic field is taken as

$$S_{fi} = -i \int_{-\infty}^{\infty} dt\, (\psi_f(t), H'(t)\phi_i(t)) , \tag{1}$$

where $\psi_f(t)$ is the field-dependent wave function for the conduction electron, $\phi_i(t)$ is the field-free wave function for the valence electron, and

$$\begin{aligned} H'(t) &= -e\vec{A}\cdot\vec{P}/m \\ &= ea(P_x \cos\omega t + P_y \sin\omega t)/m\sqrt{2} \end{aligned} \tag{2}$$

is the interaction Hamiltonian arising from a circularly polarized electromagnetic field. Standard notation is employed in Eq. (2). The $(e\vec{A})^2$ term nominally in $H'(t)$ does not make a contribution to the transition rate in the dipole approximation.

The wave function for the valence electron is simply the standard Bloch function

$$\phi_i(t) = (1/\sqrt{N_0}) u_{v,\vec{k}'}(\vec{r}) e^{i(\vec{k}'\cdot\vec{r} - E_v t)} , \tag{3}$$

where N_0 is the number of unit cells in the crystal, $\vec{k}'$ is the wave vector and

$$E_v = -E_g - k'^2/2m_v \tag{4}$$

is the energy in the effective-mass approximation with a spherical band. The quantities E_g and m_v are the energy gap and the effective mass, respectively.

A Volkov-type wave function is used to represent the conduction electron in the presence of a circularly polarized monochromatic electromagnetic field and is given by

$$\begin{aligned} \psi_f(t) &= (1/\sqrt{N_0}) u_{c,\vec{k}}(\vec{r}) \\ &\quad \times \exp\{i[\vec{k}\cdot\vec{r} + \zeta \sin(\omega t - \rho) - E_c t]\} , \end{aligned} \tag{5}$$

where

$$E_c = k^2/2m_c + (ea)^2/4m_c , \tag{6}$$

$$\zeta = ea(k_x^2 + k_y^2)^{1/2}/\omega m_c \sqrt{2} , \tag{7}$$

and

$$\rho = \tan^{-1}(k_y/k_x) . \tag{8}$$

The energy of the conduction electron in Eq. (6) includes a mass-shift energy. Energy is no longer a good quantum number because of the explicit time dependence of the Volkov Hamiltonian. This approximate wave function is satisfying on physical grounds; the electromagnetic field introduces the Volkov factor, and the Bloch function accounts for the potential of the crystal.

Using a Bessel function expansion for $\exp[i\zeta \sin(\omega t - \rho)]$, the time integration for the S matrix can be performed to yield

$$\begin{aligned} S_{fi} &= -i(\pi ea/m\sqrt{2})\delta_{\vec{k},\vec{k}'} \\ &\quad \times \sum_{l=-\infty}^{\infty} (-)^l J_l(\zeta) e^{-il\rho} \\ &\quad \times \{M_- \delta(E_c - E_v + (l+1)\omega) \\ &\quad + M_+ \delta(E_c - E_v + (l-1)\omega)\} , \end{aligned} \tag{9}$$

where

$$M_\pm = \langle u_c(\vec{r}) \,|\, P_x \pm i P_y \,|\, u_v(\vec{r})\rangle , \tag{10}$$

and the integration for the M-matrix elements is over a single unit cell of the crystal. Momentum conservation was obtained from the standard summation relation[11] in conjunction with periodicity of the $u_{\vec{k}}$ functions.

The S matrix of Eq. (9) is now specialized to describe the absorption of N photons. Therefore,

$$l \pm 1 = -N. \tag{11}$$

With this constraint the T matrix can be written in cgs units as

$$\begin{aligned} T_{fi}^{(N)} &= (ea/2^{3/2}mc)\delta_{\vec{k},\vec{k}'} e^{iN\rho}[J_{N+1}(\zeta)\, \exp(i\rho) M_- \\ &\quad + J_{N-1}(\zeta)\, \exp(-i\rho) M_+] . \end{aligned} \tag{12}$$

Using the relationship

$$\delta^2_{\vec{k},\vec{k}'} = \delta_{\vec{k},\vec{k}'} , \tag{13}$$

$|T_{fi}^{(N)}|^2$ is given by

$$\begin{aligned} |T_{fi}^{(N)}|^2 &= (ea/mc)^2 \delta_{\vec{k},\vec{k}'} \\ &\quad \times \tfrac{1}{8}\{|M_-|^2 J_{N+1}^2(\zeta) \\ &\quad + |M_+|^2 J_{N-1}^2(\zeta) + 2J_{N+1}(\zeta) J_{N-1}(\zeta) \\ &\quad \times [(M_x^2 - M_y^2)\cos 2\rho + 2M_x M_y \sin 2\rho]\} , \end{aligned} \tag{14}$$

where the $M_{x,y}$ matrix elements are related to those in Eq. (10) in the obvious manner.

The transition rate in cgs units is given by

$$w_{fi}^{(N)} = \frac{2\pi}{\hbar}\sum_{\vec{k},\vec{k}'} |T_{fi}^{(N)}|^2 \delta(E_f - E_i - N\hbar\omega) . \quad (15)$$

From Eqs. (14) and (15) the transition rate per unit volume is given by

$$\frac{W^{(N)}}{V} = \frac{(ea/mc)^2}{8(2\pi)^2\hbar}\int_{BZ} \{|M_-|^2 J_{N+1}^2(\zeta) + |M_+|^2 J_{N+1}^2(\zeta) + 2J_{N+1}(\zeta)[(M_x^2 - M_y^2)\cos 2\rho + 2M_x M_y \sin 2\rho]\} \times \delta(E_c - E_v - N\hbar\omega)\, d^3k \quad (16)$$

where the integration over the Brillouin zone represents a sum over the final states. To obtain Eq. (16) the momentum-conservation constraint has been utilized to sum over the initial states. Evaluation of the integral in Eq. (16) is conveniently performed in cylindrical coordinates. With the assumption that the M_i matrix elements are independent of $\vec{k}$, the angular integration removes the cross terms of $|T_{fi}^{(N)}|^2$; therefore,

$$\frac{W^{(N)}}{V} = \frac{(ea/mc)^2}{16\pi\hbar} \times \int [|M_-|^2 J_{N+1}(\zeta) + |M_+|^2 J_{N-1}^2(\zeta)] \times \delta\left(\hbar^2\frac{(k_\rho^2 + k_z^2)}{2m_r} + E_g + \frac{(ea/c)^2}{4m_c} - N\hbar\omega\right) \times k_\rho\, dk_\rho\, dk_z , \quad (17)$$

where the $\vec{k}$ dependence of the argument of the δ function was taken from Eqs. (4) and (6), and m_r is the reduced mass. The k_z integration can be performed with the δ function, so that the transition rate for a unit volume becomes

$$W^{(N)} = \frac{(ea/mc)^2 m_r}{8\pi\hbar}\int_0^y [|M_-|^2 J_{N+1}^2(\zeta) + |M_+|^2 J_{N-1}^2(\zeta)] \times k_\rho (y^2 - k_\rho)^{-1/2}\, dk_\rho , \quad (18)$$

where

$$y = [2m_r(N\hbar\omega - E_g - (ea/c)^2/4m_c)]^{1/2}/\hbar . \quad (19)$$

The parameter y is the amplitude of the wave vector of the conduction electron. The radicand in Eq. (19) is non-negative as a consequence of the δ function in Eq. (17). It is convenient to change the variable of integration in order that Eq. (18) can be written as

$$W^{(N)} = \frac{(ea/mc)^2 m_r}{8\pi\hbar} y \int_0^{\pi/2} \sin\theta [|M_-|^2 J_{N+1}^2(\lambda\sin\theta) + |M_+|^2 J_{N-1}^2(\lambda\sin\theta)]\, d\theta \quad (20)$$

where

$$\lambda = eay/\omega c m_c\sqrt{2} . \quad (21)$$

The intensity parameter λ^2 is the free-electron intensity parameter times the square of the ratio of the wave vector of the conduction electron to the wave vector of the electromagnetic field. Utilization of the tables in Luke[12] furnishes the result

$$\int_0^{\pi/2} \sin\theta J_M(\lambda\sin\theta)\, d\theta = \left(\frac{\lambda}{2}\right)^{2M}\left[\frac{\sqrt{\pi}}{2}\right]\Gamma(1+M)\Gamma(M+\tfrac{3}{2}) \times {}_1F_2(M+\tfrac{1}{2}; 2M+1, M+\tfrac{3}{2}; -\lambda^2) , \quad (22)$$

where $\Gamma(z)$ and ${}_1F_2(a; b, c; z)$ are the γ and hypergeometric functions, respectively. Combination of Eqs. (20) and (22) yields

$$W^{(N)} = \frac{B(\lambda/\sqrt{2})^{2N}}{y(N-1)!\,(2N-1)!!} \times \left(|M_+|^2\, {}_1F_2(N-\tfrac{1}{2}; 2N-1, N+\tfrac{1}{2}; -\lambda^2) + \frac{(\lambda/\sqrt{2})^4 |M_-|^2}{N(N+1)(2N+1)(2N+3)} \times {}_1F_2(N+\tfrac{3}{2}; 2N+3, N+\tfrac{5}{2}; -\lambda^2)\right) , \quad (23)$$

where

$$B = (m_c\omega)^2 m_r/2\pi\hbar^3 m^2 . \quad (24)$$

For small values of λ, the transition rate per unit volume is described as

$$W^{(N)} \sim \lambda^{2N} . \quad (25)$$

However, inspection of Eq. (21) shows that small values of λ correspond to both the low- and high-intensity region. In the low-intensity domain the mass-shift energy can be neglected and

$$W^{(N)} \sim I^N , \quad (26)$$

where I is the intensity of the electromagnetic field. For large values of I, the mass-shift energy can attain magnitudes such that

$$0 \leq N\hbar\omega - E_g - \alpha I = \delta \ll 1 , \quad (27)$$

where

$$\alpha = 2\pi e^2/\omega^2 c m_c . \quad (28)$$

Negative values of δ are forbidden by energy conservation. The transition rate for intensities which satisfy the constraint of Eq. (27) is characterized by

$$W^{(N)} \sim \delta^{N-1/2} . \quad (29)$$

II. PERTURBATIVE CALCULATION OF INTERBAND TRANSITION RATES FOR CIRCULAR POLARIZATION

For jth order the S matrix which describes a transition between states i and f in standard nota-

tion is

$$S_{fi}^{(j)} = -i \int_{-\infty}^{\infty} dt\, (\Phi_f(t), H^1(t)\psi^{(j)}(t)) . \tag{30}$$

For a circularly polarized electromagnetic field in the dipole approximation, $H^1(t)$ is given by Eq. (2). However, for absorption, only part of $H^1(t)$ contributes so that

$$H^1(t) = H_1 e^{-i\omega t} , \tag{31}$$

where

$$H_1 = ea(P_x + iP_y)/2^{3/2} m . \tag{32}$$

Under these constraints the initial state in jth-order is

$$\psi_i^{(j)}(t) = \Phi_i(t) + \sum_{n_1,\ldots,n_j} \left(\Phi_{n_j}(t)\, \frac{\langle n_j | H_1 | n_{j-1}\rangle \cdots \langle n_1 | H_1 | i\rangle}{(E_{n_j} - E_i - j\omega)\cdots(E_{n_1} - E_i - \omega)} \exp[i(E_{n_j} - E_i - j\omega)t] \right) . \tag{33}$$

Inclusion of valence-band states in the sum over intermediate states in Eq. (33) is forbidden by the Pauli principle. Therefore, the $|n_j\rangle$'s in the above are conduction-band states with different $\vec{k}$ values. The wave functions are given by the Bloch functions of Eq. (3).

The T matrix for an Nth-order process is readily obtained from Eqs. (30) and (33) in the form

$$T_{fi}^{(N)} = \sum_{n_1,\ldots,n_{N-1}} \frac{\langle f | H_1 | n_{N-1}\rangle \cdots \langle n_1 | H_1 | i\rangle}{[E_{N-1} - E_i - (N-1)\omega]\cdots(E_{n_1} - E_i - \omega)} . \tag{34}$$

Pertinent matrix elements in the above are given in cgs units by

$$\langle n_1 | H_1 | i\rangle = (ea/2^{3/2} mc) M_+ \delta_{\vec{k}_{n_1}, \vec{k}_i} , \tag{35}$$

$$\langle n_1 | H_1 | n_j\rangle = (ea/2^{3/2} cm_c) k_+ \delta_{\vec{k}_{n_1}, \vec{k}_{n_j}} , \tag{36}$$

where M_+ is the matrix element of Eq. (10) and

$$k_+ = k_x + ik_y . \tag{37}$$

The spherical-band approximation has been invoked to obtain Eq. (36). Momentum conservation in Eqs. (35) and (36) follows from the summation relations[11] together with periodicity of the Bloch functions. With the momentum-conservation constraints the sum over the intermediate states can be done explicitly to yield

$$T_{fi}^{(N)} = (ea/2^{3/2} c)^N (\hbar/m_c)^{N-1} (M_+/m) k_+^{N-1} \delta_{\vec{k}_i, \vec{k}_f} \times \left[\prod_{j=1}^{N-1} \left(\frac{\hbar^2 k_f^2}{2m_r} + E_g - j\hbar\omega \right) \right]^{-1} . \tag{38}$$

From Eqs. (15) and (38) the transition rate can be written

$$W^{(N)} = (2\pi)^{-2} \hbar^{-1} (ea/2^{3/2} c)^{2N} (\hbar/m_c)^{2(N-1)} (|M_+|/m)^2 \times \int_{BZ} k_\rho^{2(N-1)} \delta \left(\frac{\hbar^2 k^2}{2m_r} + E_g - N\hbar\omega \right) \times \left[\prod_{j=1}^{N-1} \left(\frac{\hbar^2 k^2}{2m_r} + E_g - j\hbar\omega \right) \right]^{-2} d^3k , \tag{39}$$

where

$$k_\rho = |k_+| . \tag{40}$$

To obtain Eq. (39) the sum over the initial states was carried out using the momentum-conservation constraint and the $\vec{k}$ dependence of M_+ was neglected. It is advantageous to employ cylindrical coordinates in the evaluation of the integral in Eq. (39). Since there is no angular dependence in the integrand, a factor of 2π is immediately extracted, and for brevity the result of the δ function integration is anticipated. Thus

$$W^{(N)} = (2\pi\hbar)^{-1} (ea/2^{3/2} c)^{2N} (\omega m_c)^{-2(N-1)} (|M_+|/m)^2 \times [(N-1)!]^{-2} \int k_\rho^{2N-1} \delta \left(\frac{\hbar^2 (k_\rho^2 + k_z^2)}{2m_r} + E_g - N\hbar\omega \right) dk_\rho\, dk_z , \tag{41}$$

where use of the relationship

$$\prod_{j=1}^{N-1} (j\hbar\omega) = (\hbar\omega)^{N-1} (N-1)! \tag{42}$$

has been made. Application of the δ function for the k_ρ integration yields

$$W^{(N)} = (m_r/\pi\hbar^3)(ea/2^{3/2} c)^{2N} (\omega m_c)^{-2(N-1)} \times (|M_+|/m)^2 [(N-1)!]^{-2} Z^{2(N-1)} \times \int_0^Z (1 - k_z^2/Z^2)^{N-1} dk_z , \tag{43}$$

where

$$Z = [2m_r (N\hbar\omega - E_g)]^{1/2}/\hbar . \tag{44}$$

Use of the integral

$$\int_0^{\pi/2} \cos^{2N-1}\theta\, d\theta = \frac{2^{N-1}(N-1)!}{(2N-1)!!} \tag{45}$$

yields immediately

$$W^{(N)} = \frac{(2\pi)^{N-1}(|M_+|/m)^2(e^2/c)^N(2m_r)^{(2N+1)/2}(N\hbar\omega - E_g)^{(2N-1)/2}I^N}{2(N-1)!(2N-1)!!n^N\hbar^{2(N+1)}\omega^{2(2N-1)}m_c^{2(N-1)}}, \tag{46}$$

where

$$I = n\omega^2 a^2/8\pi c \tag{47}$$

is the intensity and n is the index of refraction.

Comparison of Eq. (46) and the low-intensity form of Eq. (23) demonstrates that the two results are identical for any value of N.

III. DISCUSSION OF RESULTS

In the low-intensity domain the transition rate obtained from the Volkov approximation is identical to the perturbative result for any value of N for circular polarization. This may be the only intense-field calculation to reproduce perturbative predictions exactly for arbitrary order. Obviously, the Volkov-type wave function employed here is an accurate description of the conduction-band state in the presence of an electromagnetic field, in the dipole approximation.

For high intensity the mass-shift energy of the conduction increases rapidly. Physically, the electromagnetic field cannot supply enough energy to oscillate the electron and still effect a transition. As seen from Eq. (27) the cutoff intensity is linearly dependent on $N\hbar\omega - E_g$. Therefore, the transition rates for lower-order processes are zero for certain large values of I, and higher-order processes dominate. Reversal of dominance as a function of N for large I is also exhibited in the predictions of the momentum-translation approximation for atomic systems.[13] This intense-field behavior is substantially unlike the perturbative character of the low-intensity domain.

The variation of dimensionless transition rates $\bar{W}^{(N)}$ for Zns with irradiating intensity from a Nd-glass laser for $N=4$ and 5 are shown in Fig. 1. The dimensionless transition rate is defined from Eq. (23) as

$$\bar{W}^{(N)} = 6.87\times 10^{-14} W^{(N)}(2m_r)^{1/2}/B\hbar|M_+|^2 a_c, \tag{48}$$

where it is assumed that

$$|M_+| = |M_-| \tag{49}$$

and the lattice constant a_c is taken as 5.41×10^{-8} cm. The values $m_c = 0.3$, $m_r = 0.28$ are used.[7] The dimensionless intensity parameter is defined as

$$y = I/(10^{12}\ \mathrm{W/cm^2}). \tag{50}$$

A value of 3.8 eV is taken for the band gap of ZnS; therefore, $N=4$ is the lowest-order transition which can occur. Perturbative character is exhibited by the straight-line sections of the curves in the low-intensity region. The transition rates for $N=4$ and 5 are equal for $y=1.5$; and for $y>1.5$, it is found that $\bar{W}^{(5)} > \bar{W}^{(4)}$. This reversal of dominance is clearly nonperturbative behavior. For $y>2$ the mass-shift energy for the $N=4$ process is appreciable, and the rapid decline of the transition rate is governed by Eq. (29). At $y=2.8$ the transition rate for the 4th-order process is zero. All orders will display this behavior as the intensity approaches the cutoff value given by Eq. (29).

The deviation of the results from weak-field character is also exhibited in Fig. 2. Transition rates for the lower intensities agree with perturbative predictions; the transition rate for given intensity decreases as the order of the process increases. However, for $y=10$ there is a striking disparity. Here $\bar{W}^{(7)} > \bar{W}^{(6)}$ and the transition rates for the lower-order processes are zero. Transitions of order eight through twelve are about equally probable. This trend toward "flatness" of the $\bar{W}^{(N)}$ vs N curves at high intensities is similar to the results for hydrogen.[13]

These results are quite different from the results of Keldysh.[8] His low-intensity rates are generally lower than the corresponding perturbative prediction.[7,14,15] Keldysh's analytical results were obtained at the cost of requiring $N \gg 1$. This effectively severs him from any effective application to the most likely physical applications where $N \leq 10$. It has been found that the Keldysh approach successfully describes single photon absorption[16]; however, this appears to be a fortuitous result.

Observation of these intense field effects will require the use of picosecond pulses in order that the material not become an ionized plasma due to various absorption processes at these high intensities. Further, it is doubtful that the material can withstand intensities of 10^{13} W/cm^2 and beyond[17]; therefore, the observation of the deviation of the higher-order processes from perturbative behavior is quite improbable.

An interesting aspect of the results presented here is that they predict an onset of specific intensity effects at a relatively low intensity. From Figs. 1 and 2 it is seen that nonperturbative behavior should be clearly in evidence at intensities between 10^{12} and 10^{13} W/cm^2 with 1.06-μm laser radiation. By contrast recent experiments by Lompre *et al.*[18] do not show identifiable intensity effects even up to 10^{15} W/cm^2. The essential difference between the present work and the atomic ionization problem investigated by Lompre *et al.*

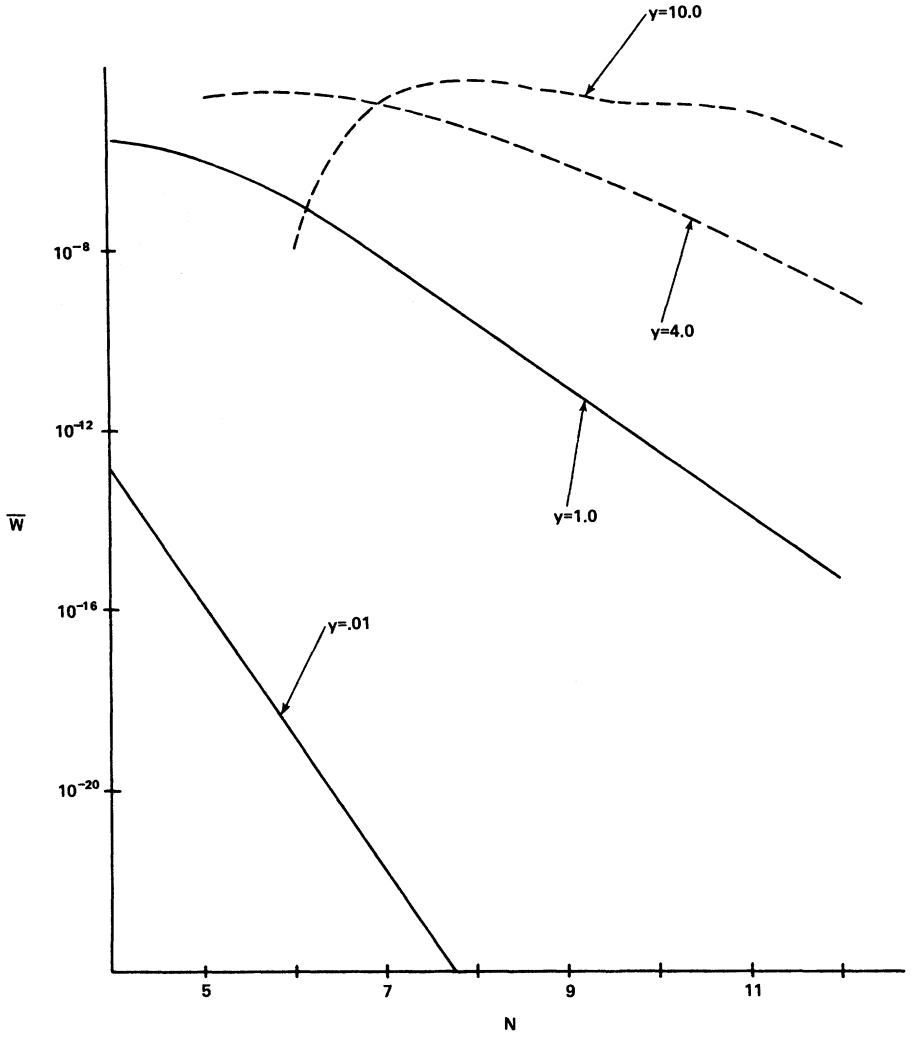

FIG. 1. Dimensionless multiphoton transition rate from Eq. (48) for ZnS as a function of intensity for Nd-glass laser radiation. The dimensionless parameter y is defined in Eq. (50). A value of $y=1$ corresponds to 10^{12} W/cm^2. The physical parameters for ZnS are given in Ref. 7.

arises from a difference in the intensity parameter appropriate to the problem. The intensity parameter which arises here is, from Eq. (23), λ^2. From Eqs. (21) and (19), this may be written

$$\lambda^2 = \frac{m_r}{m_c}\,\frac{e^2a^2}{\omega m_c}\left(N - \frac{E_g}{\omega} - \frac{e^2a^2}{4\omega m_c}\right) . \tag{51}$$

For the lowest-order process, the difference $N - E_g/\omega$ is less than unity. Therefore, if we set

$$z = e^2a^2/4\omega m_c , \tag{52}$$

then a value of z of order unity will mark a level of intensity which will have a significant effect in the problem. This parameter, z, is the simplest intensity parameter that can be identified for this physical problem. It is identical in form to the intensity parameter which arises in the negative atomic ion problem,[19] except that there m_c is replaced by the free-electron mass m. For ZnS subjected to 1.06-μm laser radiation, the value $z=1$ corresponds to $y=3.3$. The corresponding intensity parameter for the problem of the photoionization of atoms[18] is the inverse square of the Keldysh parameter[8], γ. The inverse is taken because γ decreases with intensity, rather than increases, and γ^{-1} is squared so as to be proportional to photon density as is z given in Eq. (52). This atomic intensity parameter is thus

$$\gamma^{-2} = e^2a^2/2mI_0 ,$$

where I_0 is the atomic ionization energy. For instance, if one were to substitute I_0 for hydrogen, the result would be

$$\gamma_H^{-2} = (eaa_0)^2 ,$$

where a_0 is the Bohr radius. A laser intensity of 10^{15} W/cm^2 gave values of γ^{-2} of the order of 10 in the experiments of Lompre *et al.*[18] The same

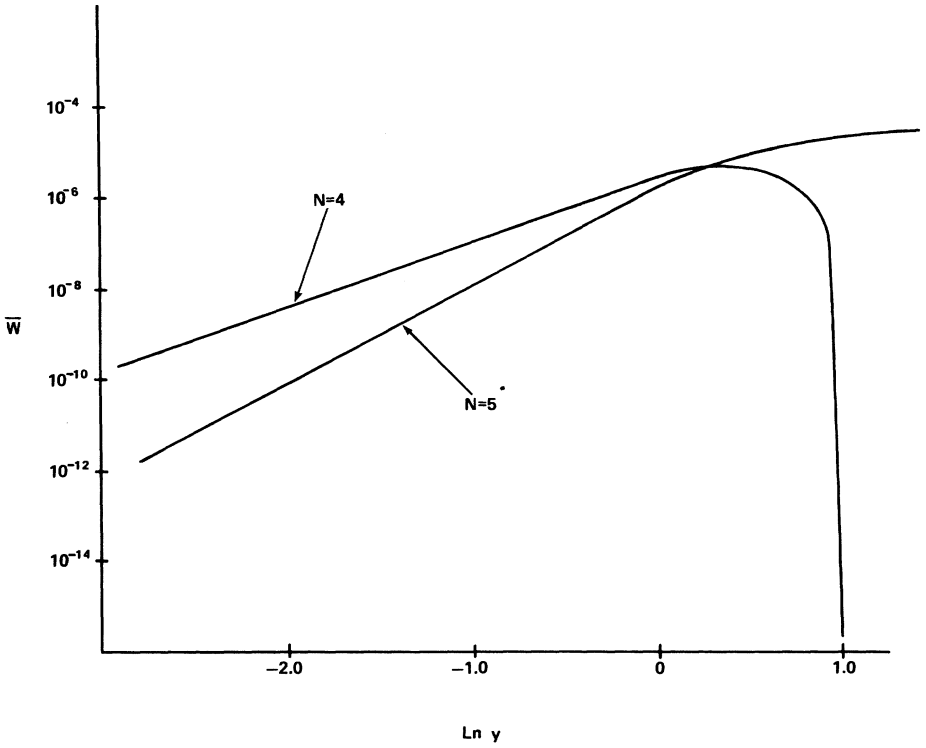

FIG. 2. Dimensionless multiphoton transition rate from Eq. (48) for ZnS as a function of photon multiplicity for Nd-glass laser radiation. The multiplicity N is regarded as a continuous parameter for convenience. Only integer values of N are physical.

laser intensity would lead to a value of z of about 300. Thus the problem of interband transition in insulators investigated here should show intensity effects much earlier than the photoionization of low-pressure atomic gases.

IV. SUMMARY

Multiphoton interband transition rates were calculated for an insulator in the presence of a circularly polarized electromagnetic field of arbitrary intensity by a Volkov approximation. All orders of the process deviated from the predictions of perturbation theory at a high intensity. There was also a reversal of dominance as a function of photon multiplicity, i.e., high-order processes can be more probable than lower-order processes when the intensity is sufficiently high. It was found that for high intensities the transition rate became nearly independent of the number of photons involved in the process.

For low intensity the transition rate with circular polarization from the Volkov approximation was identical to the perturbative result for all orders. This may be the only intense-field calculation which, in the low-intensity limit reproduces perturbative predictions exactly for arbitrary order.

It was found that specific intensity effects should make their appearance in multiphoton interband transitions in an insulator at much lower intensities than for multiphoton ionization of free atoms.

The Volkov approximation should be useful for investigation of other electromagnetic field interactions, such as photon absorption by free carriers in solids.

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