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IN THE NON-PERTURBATIVE APPROACH

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MULTIPHOTON TRANSITIONS IN SEMICONDUCTORS  
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ABSTRACT

Transition rates for multiphoton absorption via direct band-to-band excitation have been calculated using a non-perturbative approach due to Jones and Reiss<sup>1)</sup>, based on the Volkov type final state wave functions. Both cases of parabolic and non-parabolic energy bands have been included in our calculations. Absorption coefficients have been obtained for the cases of plane polarized and circularly polarized light. In particular, two-photon absorption coefficients are derived for the two cases of polarization for the parabolic band approximation as well as for non-parabolic bands and compared with the results based on perturbation theory. Numerical estimates of the two-photon absorption coefficients resulting from our calculations are also provided.

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1. INTRODUCTION

Multiphoton absorption represents an important tool for probing the electronic structure of solids. Although such absorption processes have been extensively studied theoretically, perturbation theory seems to provide the main framework for analysis, so far<sup>1)</sup>. However, with the recent advent of high power lasers the regime where the optical field no longer provides only a weak perturbing influence has become accessible. This clearly calls for non-perturbative approaches to the problem of the interaction of the high intensity radiation field with the solid media. Keldysh<sup>2)</sup> developed such a non-perturbative method based on the philosophy that the strong radiation fields change the initial and the final state wave-functions in a direct manner thus invalidating the use of the unperturbed solid state (Bloch) wave-functions. Such wave-functions in the case of a free electron are known to be the Volkov functions which represent the exact solution of the Schrödinger equation. Keldysh assumed that the same functions could be reliably used to represent the wave-functions of the electrons in the energy bands of a solid. Keldysh's results were expectedly in conflict, in the low intensity limit, with the results obtained from the perturbation approach. Recently, Jones and Reiss<sup>3)</sup> removed the high intensity and low photon-frequency assumptions inherent in the Keldysh theory to obtain results which approached the perturbation theory results in the low intensity limit. They used this modified Keldysh approach to obtain the transition rates for circularly polarized light incident on a direct gap semiconductor or insulator with energy bands approximated by a parabolic E-k relationship. Later, Brandi and Malta<sup>4)</sup> extended their treatment to the case of linearly polarized light but still restricted to parabolic band materials.

In this paper we report the application of the Volkov-approximation non-perturbative approach to non-parabolic band materials for the first time. Like Jones and Reiss<sup>3)</sup> and Brandi and Malta<sup>4)</sup> we work within the two-band model of multiphoton transitions using the effective mass description of the solid. However, a convenient prescription due to Kane<sup>5)</sup> has been used to account for the non-parabolicity effects. Both cases of linearly polarized light and circularly polarized light are included in our calculations. Further, since we found some errors in the results of earlier works for parabolic bands<sup>3),4)</sup> we have recalculated the multiphoton transition rates for the two cases of polarization. We thus present a complete analytical description of multiphoton absorption in direct gap materials within the framework of the Volkov approximation. Numerical estimates of the effects

of non-parabolicity of energy bands are then presented for the specific case of two-photon absorption. Comparisons are also made with the perturbation theory results for two-photon absorption for both parabolic and non-parabolic two-band models.

## 2. FORMALISM

The band-to-band multiphoton transition in a semiconductor can be viewed as a general scattering event taking the electron from an initial valence band state to a final conduction band state described in S-matrix formalism by

$$S_{fi} = -\frac{i}{\hbar} \int dt d^3r \psi_f^*(r,t) H_{int} \phi_i(r,t) \quad (1)$$

The interaction hamiltonian is given in the dipole approximation by

$$H_{int} = -\frac{e}{mc} \underline{A} \cdot \underline{p} \quad (2)$$

with  $\underline{A}$  the vector potential of the field and  $\underline{p}$  representing the electron momentum. The initial state is assumed to be the usual field-free valence band state

$$\phi_i(r,t) = \frac{1}{\sqrt{N_0}} u_{v\mathbf{k}}(r) e^{i\{\mathbf{k}\cdot\mathbf{r} - \frac{1}{\hbar} E_v t\}} \quad (3)$$

while the final conduction band state is taken to be a field dependent wave function approximated by a Volkov <sup>6)</sup>-type function

$$\psi_f(r,t) = \frac{1}{\sqrt{N_0}} u_{c\mathbf{k}}(r) e^{i\{\mathbf{k}\cdot\mathbf{r} - \frac{1}{\hbar} \int E_c[\mathbf{k}(t)] dt\}} \quad (4)$$

where the  $\mathbf{k}$  dependence of  $E_v$  and  $E_c$  is different for the parabolic and non-parabolic bands.

The S-matrix, thus calculated, is related to the T-matrix by

$$S_{fi} = S_{fi} - 2\pi i T_{fi} \delta(E_f - E_i) \quad (5)$$

which in turn can be used to calculate the transition rate  $W_{fi}(\mathbf{k})$  as

$$W_{fi}(\mathbf{k}) = 4\pi^2 |T_{fi}|^2 \delta(E_f - E_i) \quad (6)$$

for a particular order of the multiphoton process. Summation over  $\mathbf{k}$  over a Brillouin zone will subsequently lead to the absorption coefficient  $\alpha$  for the corresponding process;

$$\alpha = \frac{2\hbar\omega}{I} W_{fi} \quad (7)$$

where  $I$  is the intensity of the radiation and  $\omega$  its frequency.

## 3. PARABOLIC BANDS

For parabolic energy bands,

$$E_v(\mathbf{k}) = -\frac{\hbar^2 k^2}{2m_v^*} \quad (8)$$

and

$$E_c(\mathbf{k}) = E_g + \left[ \hbar k - \frac{e}{c} \underline{A}(t) \right]^2 / 2m_c^* \quad (9)$$

where  $m_v^*$  and  $m_c^*$  are the valence band and conduction band effective masses, respectively. With this dispersion relation the final state wave function for the conduction electrons assumes the Volkov form,

$$\psi_f(t) = \frac{1}{\sqrt{N_0}} u_{c\mathbf{k}}(r) \exp\left[i\left\{\mathbf{k}\cdot\mathbf{r} + \int \sin\omega t - \eta \sin 2\omega t - \frac{E_c t}{\hbar}\right\}\right] \quad (10)$$

for the plane polarized light and

$$\psi_f(t) = \frac{1}{\sqrt{N_0}} u_{c,\mathbf{k}}(r) \exp\left[i\left\{\mathbf{k}\cdot\mathbf{r} + \int \sin(\omega t - \rho) - \frac{E_c t}{\hbar}\right\}\right] \quad (11)$$

for circularly polarized light, where

$$\begin{aligned} \eta &= \frac{eA_0}{m_c^* \omega c} k_z, \quad \eta' = \frac{e^2 A_0^2}{8m_c^* \hbar c \omega}, \quad \rho = \frac{eA_0}{m_c^* \omega c \sqrt{2}} k_\rho \\ E_c^0 &= E_g + \frac{\hbar^2 k^2}{2m_c^*} + \frac{e^2 A_0^2}{2m_c^* c^2}, \quad E_c' = E_g + \frac{\hbar^2 k^2}{2m_c^*} + \frac{e^2 A_0^2}{4m_c^* c^2} \\ k_\rho &= (k_x^2 + k_y^2)^{1/2}, \quad \rho = \tan^{-1}\left(\frac{k_y}{k_x}\right) \end{aligned} \quad (12)$$

With these wave functions, using the Bessel function expansion of

$$e^{i\zeta \sin \theta} = \sum_{s=-\infty}^{\infty} (i)^s J_s(\zeta) e^{is(\frac{\pi}{2} - \theta)} \quad (13)$$

one obtains the following expressions for the S-matrix:

$$S_{fi} = i\pi \delta_{\underline{k}, \underline{k}'} \frac{eA_0}{mc} M_{\underline{z}} \sum_m \sum_s (-1)^s i^{m+s} J_s(\zeta) J_m(\eta) \times e^{i(m+s)\frac{\pi}{2}} \left[ \delta\left\{\frac{1}{\hbar}(E_c^0 - E_v) - (s+2m-1)\omega\right\} + \delta\left\{\frac{1}{\hbar}(E_c^0 - E_v) - (s+2m+1)\omega\right\} \right] \quad (14)$$

for the plane polarized case and

$$S_{fi} = -i\pi \delta_{\underline{k}, \underline{k}'} \frac{eA_0}{\sqrt{2}mc} \sum_{l=-\infty}^{\infty} (-1)^l J_l(\zeta) e^{-il\varphi} \times \left[ M_- \delta\left\{\frac{1}{\hbar}(E_c' - E_v) + (l+1)\omega\right\} + M_+ \delta\left\{\frac{1}{\hbar}(E_c' - E_v) + (l-1)\omega\right\} \right] \quad (15)$$

for the circularly polarized light, where

$$M_{\underline{z}} = \langle u_c(\underline{r}) | p_{\underline{z}} | u_v(\underline{r}) \rangle \quad (16)$$

and

$$M_{\pm} = \langle u_c(\underline{r}) | p_x \pm ip_y | u_v(\underline{r}) \rangle \quad (17)$$

are the dipole matrix elements evaluated over one unit cell of the crystal.

We can obtain the S-matrix for N-photon absorption by simply restricting the coefficients of  $\omega$  in the  $\delta$ -functions to N in the summations in Eqs.(14) and (15). Subsequently using Eqs.(5) and (6) the N-photon transition rates per unit volume for a particular  $\underline{k}$  can be obtained as:

$$W^N(\underline{k}) = \frac{2\pi}{\hbar} \left(\frac{m_c^* \omega}{m}\right)^2 |M_{\underline{z}}|^2 \left| \sum_{n=-\infty}^{\infty} \frac{N+2n}{k_z} J_{N+2n}\left(\frac{ek_z A_0}{m_c^* \omega c}\right) \right|^2 \times J_n\left(\frac{e^2 A_0^2}{8m_c^* c^2 \hbar \omega}\right) \left| \delta\left(\frac{\hbar^2 k^2}{2m_c^*} + E_g - N\hbar\omega\right) \right|^2 \quad (18)$$

for the plane polarized case as obtained by Brandi and Malta<sup>4)</sup>. Here  $m_{cv}^* = \left(\frac{1}{m_c^*} + \frac{1}{m_v^*}\right)^{-1}$  is the reduced effective mass and

$$W^N(\underline{k}) = \frac{(eA_0/mc)^2}{(2\pi)^2 8\hbar} \left[ |M_-|^2 J_{N+1}^2(\zeta) + |M_+|^2 J_{N-1}^2(\zeta) + 2 J_{N+1}(\zeta) \left\{ (M_x^2 - M_y^2) \cos 2\rho + 2M_x M_y \sin 2\rho \right\} \right] \times \delta(E_c - E_v - N\hbar\omega) \quad (19)$$

for the circularly polarized case as obtained by Jones and Reiss<sup>3)</sup>. The total N-photon transition rates for the two cases can be obtained by summing over all  $\underline{k}$ 's,

$$W^N = \int \frac{1}{8\pi^3} W^N(\underline{k}) d^3\underline{k} \quad (20)$$

Noting that the dominant contribution in the plane polarized case comes from  $n=0$  term in the series and adopting the weak field limit, in which case the second Bessel function is unity, we find

$$W^N = \frac{m_c^{*2} \omega^2}{\pi \hbar m^2} \frac{N^2 |M_{\underline{z}}|^2}{2^{(2N+3)}} \left(\frac{2m_{cv}^*}{\hbar^2}\right)^{N+\frac{1}{2}} \left(\frac{eA_0}{m_c^* \omega c}\right)^{2N} (N\hbar\omega - E_g)^{N-\frac{1}{2}} \times \frac{1}{(N-\frac{1}{2})(N!)^2} {}_3F_4 \left( \begin{matrix} \frac{2N+1}{2}, N+1, N-\frac{1}{2} \\ 2N+1, N+1, N+1, N+\frac{1}{2} \end{matrix} \middle| -2\lambda^2 \right) \quad (21)$$

$$\text{where } \lambda = \frac{eA_0}{m_c^* c \hbar \omega} \left[ m_{cv}^* (N\hbar\omega - E_g) \right]^{\frac{1}{2}} \quad (22)$$

and  ${}_3F_4$  is the hypergeometric function.

Or

$$W^N = \frac{m_{cv}^{*2} \omega^2}{\pi k m^2} \frac{N^2 |M_2|^2}{2^{(2N+3)}} \left( \frac{2m_{cv}^*}{\hbar^2} \right)^{N+\frac{1}{2}} \left( \frac{eA_0}{m_{cv}^* c \omega} \right)^{2N} (N\hbar\omega - \bar{E}_g)^{N-\frac{1}{2}}$$

$$\times \sum_{m=0}^{\infty} \frac{(N+m+\frac{1}{2})! (2N)!}{(N-\frac{1}{2})! (2N+m)! (N+m)! N! (N-\frac{1}{2})} \times$$

$$\times \left[ \frac{2m_{cv}^*}{\hbar^2} \left( \frac{eA_0}{m_{cv}^* c \omega} \right)^2 (\bar{E}_g - N\hbar\omega) \right]^m \quad (23)$$

Here

$$\bar{E}_g = E_g + \beta \quad \text{and} \quad \beta = \frac{e^2 A_0^2}{4m_{cv}^* c^2} \quad (24)$$

It can clearly be seen that the above formula differs from the final expression obtained by Brandi and Malta<sup>4)</sup> as reported in the erratum to their paper. One has been made here of the following integral from Luke<sup>7)</sup> in performing integration over  $k_p = (k_x^2 + k_y^2)^{1/2}$  in (20) after resorting to cylindrical coordinates and integrating over  $k_z$ :

$$\int_0^{\frac{\pi}{2}} \int_N^2 (z \cos \theta) \cos^2 \theta \sin \theta d\theta = \frac{1}{2} \left( \frac{z}{2} \right)^{2N} \frac{1}{(N-\frac{1}{2}) \Gamma^2(N+1)} \times$$

$${}_3F_4 \left( \begin{matrix} 2N+1, N+1, N-\frac{1}{2} \\ 2N+1, N+1, N+1, N+\frac{1}{2} \end{matrix} \middle| -z^2 \right) \quad (25)$$

For the case of the circularly polarized light one gets

$$W^N = \frac{(m_{cv}^*)^{3/2}}{2^{1/2} \pi k^4} \left( \frac{eA_0}{m_{cv}^* c} \right)^2 (N\hbar\omega - \bar{E}_g)^{1/2} \times$$

$$\left[ \left( \frac{\lambda}{2} \right)^{2N-1} \frac{1}{\Gamma(2N)} {}_1F_2 \left( N-\frac{1}{2}; 2N-1, N+\frac{1}{2}; -\frac{\lambda^2}{4} \right) |M_+|^2 \right.$$

$$\left. + \left( \frac{\lambda}{2} \right)^{2N+3} \frac{1}{\Gamma(2N+4)} {}_1F_2 \left( 2N+\frac{5}{2}; 2N+3, N+\frac{5}{2}; -\frac{\lambda^2}{4} \right) |M_-|^2 \right]$$

$$= \frac{(m_{cv}^*)^{3/2}}{2^{7/2} \pi k^4} \left( \frac{eA_0}{m_{cv}^* c} \right)^2 (N\hbar\omega - \bar{E}_g)^{1/2} \times$$

$$\sum_{n=0}^{\infty} \left[ \frac{|M_-|^2}{n! (2N+n+2)! 2(N+n+\frac{5}{2})} (-1)^n \left( \frac{\lambda^2}{4} \right)^{n+N+1} + \right.$$

$$\left. \frac{|M_+|^2}{n! (2N+n-2)! 2(N+n-\frac{1}{2})} (-1)^n \left( \frac{\lambda^2}{4} \right)^{n+N-1} \right] \quad (26)$$

The expression obtained by Jones and Reiss<sup>3)</sup> for this case differs from the above in the Gamma function coefficients of the hypergeometric functions and also in the argument of the hypergeometric function where  $\lambda^2$  appears instead of  $\lambda^2/4$ .

#### 4. NON-PARABOLIC BANDS

As most real semiconductors have non-parabolic bands, we adopt the non-parabolic  $E - k$  relationship provided by Kane<sup>5)</sup> to obtain a more realistic picture of the multiphoton processes. According to Kane non-parabolic bands can be represented by

$$E_c - E_v \equiv E_{cv}(k) = E_g \left( 1 + \frac{\hbar^2 k^2}{m_{cv}^* E_g} \right)^{1/2} \quad (27)$$

Strictly speaking, new Volkov type wave functions should be obtained using Eq.(4) for this case. This unfortunately leads to an intractable expression for the S-matrix. However, we affect a simplifying approximation by noting that the  $\delta$ -function in the transition rate  $W^{(N)}(\underline{k})$  makes the strongest contribution to the integral of  $W^{(N)}(\underline{k})$  over  $\underline{k}$ . Therefore, we use the above non-parabolic expression for  $E_{cv}$  (Eq.(27)) in only the argument of the  $\delta$ -function, keeping the rest of the integrands the same as for the parabolic case. Within this approximation the  $N$ -photon transition rate  $W^N(\underline{k})$  can be written as

$$W^N(\underline{k}) = \left( \frac{2\pi m_c^* \omega}{m} \right)^2 \frac{|M_z|^2}{k_z^2} N^2 J_N^2 \left( \frac{eA_0 k_z}{m_c^* \omega c} \right) \times$$

$$\frac{1}{2\pi\hbar} \delta \left[ E_g \left( 1 + \frac{\hbar^2 k^2}{m_c^* E_g} \right)^{\frac{1}{2}} + \frac{e^2 A_0^2}{4m_c^* c^2} - N\hbar\omega \right]$$

(28)

For the plane polarized case, and

$$W^N(\underline{k}) = \frac{(eA_0/mc)^2}{16\pi\hbar} \left[ |M_-|^2 J_{N+1}^2(\xi) + |M_+|^2 J_{N-1}^2(\xi) + \right.$$

$$\left. 2 J_{N+1}(\xi) \left\{ (M_x^2 - M_y^2) \cos 2\varphi + 2M_x M_y \sin 2\varphi \right\} \right] \times$$

$$\delta \left[ E_g \left( 1 + \frac{\hbar^2 k^2}{m_c^* E_g} \right)^{\frac{1}{2}} + \frac{e^2 A_0^2}{4m_c^* c^2} - N\hbar\omega \right]$$

(29)

For the circularly polarized case,

Integration over  $\underline{k}$  using the procedure of Jones and Reiss<sup>3)</sup> as for the parabolic case now leads to the following expressions for the total transition rates:

$$W^N = \frac{1}{2^{2N+2}} \frac{m_c^* \omega^2}{\pi\hbar m^2} \cdot \frac{1}{E_g^2} \left( N\hbar\omega - \frac{e^2 A_0^2}{4m_c^* c^2} \right) \left( \frac{eA_0}{m_c^* \omega c} \right)^{2N} \times$$

$$\left( \frac{m_c^*}{2\hbar^2} E_g \right)^{N+\frac{1}{2}} \left[ \left\{ \left( N\hbar\omega - \frac{e^2 A_0^2}{4m_c^* c^2} \right)^2 - E_g^2 \right\} / E_g^2 \right]^{N-\frac{1}{2}} \times$$

$$\frac{|M_z|^2 N^2}{(N-\frac{1}{2})(N!)^2} \left[ \begin{matrix} 2N+1, N+1, N-\frac{1}{2} \\ 2N+1, N+1, N+1, N+\frac{1}{2} \end{matrix} \middle| -\lambda'^2 \right]$$

(30)

For the plane polarized case, where

$$\lambda' = \frac{eA_0}{m_c^* \omega c \sqrt{2}} K \quad (31)$$

and

$$K = \left( \frac{m_c^*}{2\hbar^2 E_g} \right)^{\frac{1}{2}} \left[ \left\{ N\hbar\omega - \frac{(eA_0/c)^2}{4m_c^*} \right\}^2 - E_g^2 \right]^{\frac{1}{2}} \quad (32)$$

Or

$$W^N = \frac{1}{2^{2N+2}} \frac{m_c^* \omega^2}{\pi\hbar m^2} \frac{1}{E_g^2} \left( N\hbar\omega - \frac{e^2 A_0^2}{4m_c^* c^2} \right) \left( \frac{eA_0}{m_c^* \omega c} \right)^{2N} \times$$

$$\left( \frac{m_c^*}{2\hbar^2} E_g \right)^{N+\frac{1}{2}} \left[ \left\{ \left( N\hbar\omega - \frac{e^2 A_0^2}{4m_c^* c^2} \right)^2 - E_g^2 \right\} / E_g^2 \right]^{N-\frac{1}{2}} \times$$

$$\sum_{n=0}^{\infty} \frac{(N+n-\frac{1}{2})! (2N)!}{(N-\frac{1}{2})(N-\frac{1}{2})! (2N+n)! (N+n)! N!} \times$$

$$\left[ \left( \frac{eA_0}{m_c^* \omega c} \right)^2 \left( \frac{m_c^* E_g}{2\hbar^2} \right) \left\{ \left( N\hbar\omega - \frac{e^2 A_0^2}{4m_c^* c^2} \right)^2 - E_g^2 \right\} / E_g^2 \right]^n$$

(33)

Similarly for the circularly polarized case,

$$W^N = \frac{(eA_0/mc)^2}{16\pi\hbar} \left( \frac{m_c^*}{\hbar^2 E_g} K^2 + \frac{m_c^*}{\hbar^4} \right)^{\frac{1}{2}} K \times$$

$$\left[ \left( \frac{\lambda'}{2} \right)^{2N+2} \frac{1}{\Gamma(2N+4)} {}_1F_2 \left( N+\frac{3}{2}; 2N+3, N+\frac{5}{2}; -\frac{\lambda'^2}{4} \right) |M_-|^2 \right.$$

$$\left. + \left( \frac{\lambda'}{2} \right)^{2N-2} \frac{1}{\Gamma(2N)} {}_1F_2 \left( N-\frac{1}{2}; 2N-1, N+\frac{1}{2}; -\frac{\lambda'^2}{4} \right) |M_+|^2 \right]$$

(34)

or

$$W^N = \frac{(eA_0/mc)^2}{16\pi\hbar} \left( \frac{m_{cv}^*}{\hbar^2 E_g} k^2 + \frac{m_{cv}^{*2}}{\hbar^4} \right) k \times \sum_{n=0}^{\infty} \left\{ \frac{1}{(2N+n+2)! n! 2(N+n+\frac{9}{2})} (-1)^n \left(\frac{\lambda^2}{4}\right)^{n+N+1} |M_-|^2 + \frac{1}{(2N+n+2)! n! 2(N+n-\frac{1}{2})} (-1)^n \left(\frac{\lambda^2}{4}\right)^{n+N-1} |M_+|^2 \right\} \quad (35)$$

## 5. TWO-PHOTON TRANSITIONS

The general expressions for N-photon transitions obtained above can now be specialized for the case of two-photon absorption for obtaining numerical estimates and comparisons. The two-photon transition rates obtained from the above formulae involve infinite sums over terms containing powers of  $A_0^2$ . The lowest order terms giving the dominant contributions will, therefore, be retained in expressions given below. The absorption coefficients for the respective cases obtained by using Eq.(7) are:

$$\alpha_{PP}^{(2)} = \frac{|M_z|^2}{3\sqrt{2}} B \frac{(2\hbar\omega - \bar{E}_g)^{3/2}}{(\hbar\omega)^5} I \quad (36)$$

-Plane polarized; parabolic bands

$$\alpha_{PNP}^{(2)} = \frac{|M_z|^2}{2\sqrt{\pi}} B E_g^{1/2} \frac{(2\hbar\omega - \beta)}{(\hbar\omega)^5} \left[ \left( \frac{2\hbar\omega - \beta}{E_g} \right)^2 - 1 \right]^{3/2} I \quad (37)$$

-Plane polarized; non-parabolic bands

$$\alpha_{CP}^{(2)} = \frac{|M_+|^2}{\sqrt{2\pi}} B \frac{(2\hbar\omega - \bar{E}_g)^{3/2}}{(\hbar\omega)^5} I \quad (38)$$

-Circularly polarized; parabolic bands

$$\alpha_{CNP}^{(2)} = \frac{|M_+|^2}{24} B E_g^{1/2} \frac{(2\hbar\omega - \beta)}{(\hbar\omega)^5} \left[ \left( \frac{2\hbar\omega - \beta}{E_g} \right)^2 - 1 \right]^{3/2} I \quad (39)$$

-Circularly polarized; non-parabolic bands

(36)

where

$$B = \frac{\pi e^4 m_{cv}^{*5/2}}{m^2 m_c^{*2} \hbar^2 c^2} \quad (40)$$

and  $n$  is the refractive index of the material. Here  $A_0^2 = \frac{2\pi I c}{n\omega^2}$  has been used to convert the vector potential amplitude into intensity  $I$  for the plane polarized case and  $A_0^2 = \frac{4\pi I c}{n\omega^2}$  for the circularly polarized case, taking account of the appropriate normalization factors.

## 6. DISCUSSION

The above calculations provide the multiphoton transition rates to arbitrary orders. This is a merit inherent in using the non-perturbative Volkov-type approach as against the commonly employed perturbative approach. A comparison can now be drawn between the present results and the corresponding results of the two-band model perturbation theory for the specific case of two-photon processes for which the latter theory has mostly been used. For the plane polarized light in the parabolic band approximation, the comparison between our formula and the well-known Basov<sup>8)</sup> formula provides

$$\frac{\alpha_{PP}^{(2)}}{\alpha_{Basov}^{(2)}} = \frac{1}{192} \left( \frac{m_{cv}^*}{m_c^*} \right)^2 \left[ \frac{2\hbar\omega - E_g - \beta}{2\hbar\omega - E_g} \right]^{3/2} \quad (41)$$

Even in the weak field limit ( $\beta \rightarrow 0$ ), this result is in marked contrast with that obtained by Brandi and Malta<sup>4)</sup> who do not seem to obtain the factor  $\frac{1}{192}$  in the ratio.

For plane polarized light with the non-parabolic bands, the comparison of our expression with the perturbation theory formula obtained by Hassan<sup>9)</sup> yields,

$$\frac{\alpha_{PNP}^{(2)}}{\alpha_{pert}^{(2)}} = \frac{\sqrt{2\pi}}{256} \left( \frac{m}{m_c^*} \right)^2 E_g \frac{2\hbar\omega - \beta}{(\hbar\omega)^2} \frac{\left[ \left( \frac{2\hbar\omega - \beta}{E_g} \right)^2 - 1 \right]^{3/2}}{\left[ \left( \frac{2\hbar\omega}{E_g} \right)^2 - 1 \right]^{3/2}} \quad (42)$$

This clearly implies a significant difference between the results obtained by the two approaches since even when  $\beta \rightarrow 0$  in the weak field limit, the ratio differs not only by the numerical factors but also has an inverse frequency dependence.



For the case of circularly polarized photons with parabolic bands the ratio of absorption coefficients given above and the perturbation theory result obtained by Jones and Reiss<sup>3)</sup> is,

$$\frac{\alpha_{cp}^{(2)}}{\alpha_{pert.}} = \frac{3}{4\sqrt{\pi}} \left( \frac{2\hbar\omega - E_g - \beta}{2\hbar\omega - E_g} \right)^{\frac{3}{2}} \quad (43)$$

which, like the corresponding plane polarized case, gives a difference of only the numerical factors in the low field limit ( $\beta \rightarrow 0$ ).

For the case of non-parabolic bands although no specific calculation exists for circularly polarized light, but on heuristic grounds the ratio can be expected to have the same form as obtained for the plane polarized case, Eq.(41), with different numerical factors.

Finally, to get estimates of the order of magnitude for the two-photon absorption coefficients in the framework of the non-perturbative approach used here we performed numerical calculations for the case of GaAs using reasonable values for the dipole matrix element obtained from previous perturbation theory fits to data<sup>10)</sup> and for the high intensities where such processes can be realistically detected. The results are given in Table 1.

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TABLE 1

Polarization Model	Plane polarized $\alpha^{(2)}(\text{cm}^{-1})$	Circularly polarized $\alpha^{(2)}(\text{cm}^{-1})$
Parabolic bands	22.65	12.69
Non-parabolic bands	101.6	15.0

Numerical estimates of the two-photon absorption coefficients  
for GaAs for photon density:  $10^{20} \text{ cm}^{-3}$  and  
 $|M_z|^2 = |M_+|^2 = 3.56 \times 10^{-38} \text{ erg}\cdot\text{gm}$ ,  $\hbar\omega = 0.9 \text{ eV}$ .