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Non-Linear Absorption in Solids

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Abstract Multiphoton transitions in direct-gap crystals are studied considering a "non-perturbative" approach, similar to Keldysh approximation. We consider a simple two parabolic band model in the effective mass approximation, so that exact solutions associated to the full, crystal plus field, Hamiltonian may be approximated by Volkov wavefunctions. In this manner we construct an S-matrix which incorporates the electromagnetic field to all orders in an approxirnated way, leadlng to analytical solutions for the multiphoton transition rates. We discuss the use of the saddle-polnt method and its limitations as well as the connectlon between multiphoton absorption and tunnelinq. We show that fcir near threshold excitations the Keldysh approach is fairly accurate as long as terms arising from the saddle-point integration are consistently accounted.

1. INTRODUCTION

We have recently used a "non-perturbative" approach to study multiphoton transitions in direct-gap crystals[1] by considering a simple two parabolic band model in the effective mass approximation. Theexact soiution for the Schrödinger equation for crystal plus the electromagnetic field is approximated by a Volkov wavefunction[2]. This leads toan S-matrix which contains approximately the electromagnetic field in all orders and which may be analytically evaluated. This approach is an extension of the work of Jones and Reiss (JR)[3] assuming the field linearly polarized, which is the experimental situation for most of the experiments concerning multiphoton absorption in crystals.

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The most famous alternative approach to high orderperturbatlon theory Is the scheme suggested by Keldysh[4] almost two decades ago which throughout this paper we denominate Keldysh approximatlon (KA).

Although the present approach is similar to KA, there exist three basic differences among them: the first concerns the choice of the gauge $\vec{\nabla}.\vec{A} = 0$, $\phi = 0$ instead of Keldysh's $\vec{\nabla}.\vec{A} = 0$, $\phi = -e\vec{E}.\vec{r}$. Of course, if we consider an exact S-matrix formalism, any results derived from it is gauge independent, but since all "non-perturbative" approaches are approximated, different schemes usually lead to a gauge dependence. The second difference is that in KA, both initial and final states are assumed as modified by the electromagnetic field, which in principle cannot by justified from an S-matrix approach. The third (only apparent) is that, within the approach used by Keldysh, the non--parabolic form, for the energy dispersion relation, is its initial assumption. However, it was shown by Bychkov and Dykhene[5] that, in order to obtain analytical expresslons, Keldysh performed approximatlons whlch turned his calculations equivalent to the one derived from a parabolic model.

The purpose of this work is to dlscuss the class of "non-perturbative" approaches, originally suggested by Keldysh, which has been frequently used in the study of multiphoton absorptlon In solids. Since KA ís not justified from first principles, it is of basic interest to investigate its results by comparison with those arising from others models. In this connection, there has been many criticisms[6-10] concerning the applicablIity of the analytical expressions of KA to processes involving few photons absorption, which are those most studied experimentally. We must comment that these criticisms are basically due to the behavlor of its analytical expressions (obtained after the use of the saddle-point technique) in the limiting weak field case, as compared with other theoretical models. We address part of our paper to this last point, since our Initial assumption of parabolic bands allows us to derive analytical results prior and after the use of the saddle--point technlque. This provides a criterion to determine the accuracy of thls technique, and the valldity of the analytical results derlved from it as it is done in KA. From this later method, it is not possible to make the same type of analysis, because in this case no analytical results are obtained prior the application of the saddle-point due to the initical non-parabolic character of the dispersion relation.

We show that the main difficulties in interpreting the results of KA as compared with perturbation theory, lies in the fact that after performing the saddle-point integration the KA neglects oscillating terms which are dominant in the weak field limit.

In Sec. 2 we obtain the multiphoton transition rates in a manner similar to (JR), and discuss the weak field limit. Sec. 3 is dedicated to discuss the connection between multiphoton absorption and tunneling in the present approach, and to study the implications of the use of the saddle-point method as used in the KA. In Sec. 4 we present our conclusions.

2. THE MULTIPHOTON TRANSITION RATES

In this work we assume plane polarized light and the dipole approximation for the electromagnetic field.

The T-matrix may be written as

$$T = -\frac{i}{\hbar}\int_{-\infty}^{\infty} dt \int d^3x \, \psi_f^{(-)*}(\vec{x},t) H_1 \phi_i(\vec{x},t) \qquad (2.1)$$

where the interaction Hamiltonian H_1 is given by

$$H_1 = -\frac{e}{mc}\vec{A}.\vec{p} + \frac{e^2}{2mc^2}A^2 \qquad (2.2)$$

and in the dipole approximation $\vec{A} = A_0\hat{z}\cos\omega t \equiv cE_0/\omega \ \hat{z}\cos wt$.

In our model the wavefunction of the initial state is given by the solution of the zero field Schrödinger equation associated to the valence band

$$\phi_i(\vec{x},t) = u_{\vec{k}}^{v}(x)\, e^{i\left(\vec{k}.\vec{x} - \frac{E_v}{\hbar}t\right)} \qquad (2.3)$$

The final state is approximated by a Volkov type of solution given by

$$\psi_f^{(-)}(\vec{x},t) = u_{\vec{k}}^{c}(\vec{x})\, e^{i\{\vec{k}.\vec{x} - \frac{1}{\hbar}\int_0^t E_c[\vec{k}(\tau)]d\tau\}} \qquad (2.4)$$

Assuming the parabolic band model, we take $E_V = \hbar^2k^2/2m_0$ and $E_c[\vec{k}(\tau)] = E_g + [\hbar\vec{k} - (e/c)\vec{A}(\tau)]^2/2m_1$, where m_0 and m_1 are the effective masses of the valence and of the conduction band respectively.

Taking eqs. (2.3) and (2.4) into (2.1) we get for the T-matrix

$$T = \frac{i}{\hbar}\frac{eA_0}{mc} P_{vc}\, \delta_{\vec{k},\vec{k}'} \int_{-\infty}^{\infty} dt \; e^{\frac{i}{\hbar}\left\{\frac{\hbar^2 k^2}{2m_1} + \tilde{E}_g - \frac{\hbar^2 k'^2}{2m_0}\right\} t} \cos \omega t$$
$$\times\, e^{-\frac{i}{\hbar}\left[\frac{e\hbar\vec{k}.\vec{A}_0}{} \sin \omega t - \frac{e^2 A_0^2}{8m_1 \omega c^2} \sin 2\omega t\right\}} \tag{2.5}$$

where we have defined the following quantities

$$P_{vc} = \int d^3x \; e^{-i\vec{k}.\vec{x}} \; u^{c*}_{\vec{k}}(\vec{x})\, \vec{z}.\vec{p}\; u^{v}_{\vec{k}'}(\vec{x})\, e^{i\vec{k}'.\vec{x}} \tag{2.6}$$

and

$$\tilde{E}_g = E_g + e^2 A_0^2/4m_1 c^2 \tag{2.7}$$

To evaluate the time integral in eq. (2.5) we use the relations involving the Bessel functions J_n

$$e^{i\alpha \sin n\omega t} = \sum_{n=-\infty}^{\infty} J_n(\alpha)\; e^{in\omega t} \tag{2.8}$$

$$\cos\omega t \; e^{i\alpha \sin n\omega t} = \frac{1}{\alpha} \sum_{n=-\infty}^{\infty} n\, J_n(\alpha)\; e^{in\omega t} \tag{2.9}$$

to obtain

$$T = \frac{i 2\pi m_1 \omega}{\vec{k}.\hat{z}} \frac{P_{vc}}{m} \delta_{\vec{k},\vec{k}'} \sum_{Nn} \left[(N+2n) J_{N+2n}\left(\frac{e\vec{k}.\vec{A}_0}{m_1 \omega c}\right) \right.$$
$$\left. \times\, J_n\left(\frac{e^2 A_0^2}{8 m_1 \hbar \omega c^2}\right)\right] \delta\left\{\frac{\hbar^2 k^2}{2m^*} + \tilde{E}_g - N\hbar\omega\right\} \tag{2.10}$$

In eq. (2.10) we have defined the reduced effective mass $(m^*)^{-1} = m_1^{-1} + |m_0|^{-1}$, and from energy conservation we easily obtain the transition rate (per unit of volume) associated to N-photon processes

$$W^{(N)} = \int \frac{d^3k}{(2\pi)^3}\; W^{(N)}(\vec{k}) \tag{2.11}$$

where

$$W^{(N)}(\vec{k}) = \left(\frac{2\pi m_1\omega}{m}\right)^2 \frac{p_{vc}^2}{(\vec{k}.\hat{z})^2} \left| \sum_n (N+2n) J_{N+2n}\left(\frac{e\vec{k}.\hat{z}A_0}{m_1\omega c}\right) \times \left(\frac{e^2A_0^2}{8m_1\hbar\omega c^2}\right) \right|^2 \frac{1}{2\pi\hbar}\,\delta\left(\frac{\hbar^2k^2}{2m^*} + \tilde{E}_g - N\hbar\omega\right) \tag{2.12}$$

Eq. (2.12) is very similar to the one obtained in the treatment of atomic ionization by strong laser field[10,11]. We mention briefly here how to relate the present treatment to KA. In order to make the equivalente between both formalisms, it is necessary to express the interband momentum matrix element (p_{vc}) in terms of the Keldysh constant values $-\hbar\omega/3$ (due to different choice of gauge), and to replace m_1 by m^* in all equations throughout our paper (due to different choice of initial state).

Using the properties of Bessel functions, the weak field limit for the multiphoton rates is easily obtained from eq.(2.12). In fact, keeping all terms proportional to $(A_0)^{2N}$, we get

$$W^{(N)}_{\text{w.f.}} = \frac{1}{2\pi\hbar}\left(\frac{m_1\omega p_{vc}}{m}\right)^2 \left(\frac{eA_0}{2m_1\omega c}\right)^{2N} \times \sum_{n,n'=0}^{N_0} \frac{(-1)^{n+n'}\left(\frac{m_1\omega}{4\hbar}\right)^{n+n'}\left(\frac{2m^*}{\hbar^2}\right)^{N+\frac{1}{2}-(n+n')}}{n!\,n'!\,(N-2n-1)!\,(N-2n'-1)!} \frac{[N\hbar\omega-E_g]^{N-\frac{1}{2}-n-n'}}{[2N-2n-2n'-1]} \tag{2.13}$$

where N_0 is the largest integer smaller than $N/2$. From eq. (2.13) we can easily verify that the frequency behavior for the multiphoton transition rate (or the multiphoton absorption coefficient), near the excitation threshold ($N\hbar\omega \simeq E_g$), is $(N\hbar\omega-E_g)^{3/2}$ for even N ($n=n'=(N-2)/2$) and $(N\hbar\omega-E_g)^{1/2}$ for odd N ($n=n'=(N-1)/2$). These are the dominant terms in that region for the weak field regime.

At this point we notice that the main difference between the analytical results for the multiphoton transitions rate given by eqs. (2.12), (2.13) and those of KA is that we are using a parabolic band model, while the non-parabolic band model adopted by Keldysh requires a saddle-point integration in order to obtain analytical expressions for the transition rate.

Since most of the work concerning non-linear absorption has been done for two photon processes, we use our results to obtain the two-photon absorption coefficient for semiconductors where the parameters required in the calculations are well known. Noticing that $A_0^2 = 8\pi c I/n\omega^2$, where I is the intensity of the radiation and n is the Index of refraction, we may obtain from eq. (2.13) the two-photon absorption coefficient

$$\beta^{(2)} = 2\left(\frac{2\hbar\omega}{I^2}\right) = \frac{2^{11/2}\pi e^4}{3n^2c^2}\left(\frac{p_{vc}^2}{m^2}\right)\frac{(m^*)^{5/2}}{m_1^2}\frac{(2\hbar\omega-E_g)^{3/2}}{(\hbar\omega)^5} \qquad (2.14)$$

If we use the $\vec{k}.\vec{p}$ approximation[12], we have for the momentum matrix element $p_{vc}^2/m^2 = E_g/4m^*$. We may relate our expression for $\beta^{(2)}$ with the result of eq. (1) of Vaidyanathan et al[13] for the Basov[14] two -photon absorption coefficient.

$$\frac{\beta^{(2)}}{\beta_{BASOV}} = \left(\frac{m^*}{m_1}\right)^2 \qquad (2.15)$$

It is possible to show that the "non-perturbative" approach discussed here, for low fields, is equivalent to perturbation theory in all orders, if it is assumed a model for which the intermediate states are all conduction band states. This model is an extension, to higher multiplicity, of the one proposed by Braunstein and Ockman[15] for two-photon absorption and that of Hassan and Raouf[16] for four--photon absorption where the intermediate states are chosen to be conduction band and energy higher than the final state. The difference is that, in the present, all intermediate states are degenerate to the final state. Basov *et al*[14], for 2-photon absorption considered intermediate states valence and conduction bands degenerate to the initial and final states respectively. We may interpret our results as a consequence of a model which can be understood as a hybrid extension of Braunstein[15], Hassan[16] and Basov[14] models.

3. THE TRANSITION RATE AND TUNNELING

In this section we study the total transition rate which is obtained from eq. (2.11) by summing over all multiphoton processes, and discuss its relation with a tunneling-like process. It is necessary to evaluate the summation over the Bessel functions in eq. (2.12); this can be done approximately using the method of steepest descent as in Refs. 4, 10, 11. In order to evaluate the infinite summation we use the integral representation

$$\sum_n (N+2n) J_{N+2n}(a)\, J_n(b) = \frac{a}{2\pi} \int_{-\pi}^{\pi} d\theta \cos\theta \; e^{i(N\theta - a\sin\theta + b\sin 2\theta)} \tag{3.1}$$

Since one of the purposes of this work is to discuss some questions concerning the KA it is convenient to define the following quantities $\gamma = \omega\sqrt{2m_1 E_g}/eE_0$, $\nu = \tilde{E}_g/\hbar\omega$ and in analogy with the atomic problem we introduce a characteristic field $E_c = E_g\sqrt{2m_1 E_g}/e\hbar$. Using eq. (3.1) we may rewrite eq. (2.12) as

$$W^{(N)}(\vec{k}) = \left(\frac{eE_0}{m\omega}\right)^2 p_{vc}^2 \left| \int_{-\pi}^{\pi} d\theta \cos\theta \right.$$
$$\times \exp\left\{\frac{iE_g}{\hbar\omega}\left[\left(1 + \frac{1}{2\gamma^2} + \frac{\hbar^2 k^2}{2m^* E_g}\right)\theta - \frac{\hbar\vec{k}.\hat{z}}{\gamma}\left(\frac{2}{m_1 E_g}\right)^{1/2} \mathrm{sen}\theta\right.\right.$$
$$\left.\left.\left. + \frac{1}{4\gamma^2}\,\mathrm{sen}2\theta\right]\right\}\right|^2 \frac{1}{2\pi\hbar}\,\delta\left(\frac{\hbar^2 k^2}{2m^*} + \tilde{E}_g - N\hbar\omega\right) \tag{3.2}$$

The integral in the above equation may be obtained using the saddle--point method as in Refs. 4, 10, 11. It is important to comment that the integral is evaluated under the condition $E_g/\hbar\omega = \frac{1}{\gamma}\frac{E_c}{E_0} >> 1$, and that in the limit $\gamma\to 0$ the saddle points coalesce and the method cannot be applied to arbitrarily large electromagnetic fields. In fact the use of the saddle-point method restricts the applicability of the analytical expressions to field strengths satisfying the condition $E_0 << 9E_c$[10]. Furthermore, in the integration over θ we consider the fact that the effective values of $\hbar k_z$ and $\hbar k$ are much smaller than $\sqrt{2m_1 E_g}$ and $\sqrt{2m^* E_g}$, respectively, and therefore all quantities appearing in the argument of the exponential in eq. (3.2) should be expanded in powers of $\hbar k_z/\sqrt{2m_1 E_g}$ and $\hbar k/\sqrt{2m^* E_g}$ to include quadratic terms. We should mention that it is this approximation that makes the KA, which is apparently a non-parabolic theory, equivalent to a parabolic approach[5,8].

After the integration, we obtain an analytical approximation for eq. (3.2) which is

$$W^{(N)}(\vec{k}) = \frac{1}{A}\left[\frac{e\,E_0\,p_{vc}\gamma^2}{m\omega}\right]^2 \left(\frac{E_0}{E_c}\right) \frac{\exp\left[-\frac{2}{3}\frac{E}{E_0}\,g(\gamma)\right]}{(1+\gamma^2)^{1/2}} \left(1+\frac{\hbar^2k^2}{2m^*E_g}\right)$$

$$\times \exp\left\{-\frac{\hbar}{\omega}\left[\frac{k^2}{m^*}\sinh^{-1}\gamma - \frac{\gamma(\vec{k}.\hat{z})}{m_1(1+\gamma^2)^{1/2}}\right]\right\}(1+(-1)^N\cos 2f(\vec{k}.\hat{z},\gamma))$$

$$\times\,\delta\left(\frac{\hbar^2k^2}{2m^*} - (N\hbar\omega - \tilde{E}_g)\right) \qquad (3.3)$$

where

$$g(\gamma) = \frac{3}{\gamma}\left[1+\frac{1}{2\gamma^2}\sinh^{-1}\gamma - \frac{(1+\gamma^2)^{1/2}}{2\gamma}\right]$$

$$g(\gamma) = \begin{cases} 2\left(1-\frac{1}{10}\gamma^2+\frac{9}{280}\gamma^4+\ldots\right), & \gamma \ll 1 \\ \frac{3}{\gamma}(\ln 2\gamma - 1/2), & \gamma \gg 1 \end{cases} \qquad (3.4)$$

and

$$f(\vec{k}.\hat{z},\gamma) \equiv \frac{2\hbar(\vec{k}.\hat{z})}{(2m_1 E_g)^{1/2}}\left(\frac{E_g}{\hbar\omega}\right)\frac{(1+\gamma^2)^{1/2}}{\gamma} + \psi \qquad (3.5)$$

with

$$\psi \equiv \mathrm{tg}^{-1}\left[\frac{(2m_1E_g)}{(\hbar\vec{k}.\hat{z})^2}\left(1+\frac{\hbar^2k^2}{2m^*E_g} - \frac{\hbar^2(\vec{k}.\hat{z})^2}{2m_1E_g}\right)\right]^{1/2}$$

In deriving eq. (3.3) we have followed refs. 4,7,10 and 11 but did not neglect oscillating terms of the cosine type as in refs. 1,4,11. In fact for small values of γ, which corresponds to large fields, these terms oscillate rapidly giving negligible contribution, but this is not the case for large γ. As we show below it is due to the omission of these terms that the KA leads to a frequency dependence near the threshold $(N\hbar\omega - E_g)^{1/2}$ for all N's.

A tunneling-like expression is obtained from eq. (3.3), in the limit $\gamma \ll 1$. In fact after integration over $\vec{k}$ we obtain the following approximated expression for ey. (2.11)

$$W^{(N)} = W_0(\gamma)\exp\left[-\alpha(N-\nu)\right]S\left[\sqrt{\beta(N-\nu)}\right] \tag{3.6}$$

where

$$W_0(\gamma) = \frac{1}{(2\pi)^2}\left(\frac{e^2E_0^2}{\hbar\omega^2}\right)\left(\frac{p_{vc}^2}{m^2}\right)\left(\frac{E_0}{E_c}\right)^{3/2}\left(\frac{2m^*}{\hbar^3}\right)$$
$$\times\ (m_1E_g)^{1/2}\,\frac{\gamma^4}{(1+\gamma^2)^{1/4}}\,\exp\left[-\frac{2}{3}\frac{E_c}{E_0}g(\gamma)\right] \tag{3.7}$$

and

$$S(x) = \int_0^x e^{(u^2-x^2)}\,du = \frac{x}{2}\int_0^1 \frac{e^{-x^2t}}{(1-t)^{1/2}}\,dt \tag{3.8}$$

is the probability integral. We have also defined the following quantities

$$\beta = \frac{2m^*}{m_1}\,\frac{\gamma}{(1+\gamma^2)^{1/2}}$$
$$\alpha = 2\sinh^{-1}\gamma-\beta \tag{3.9}$$

The total transition rate is obtained by summing eq. (3.6) over all allowed processes, which corresponds to the absorptlon of an integer number of photons $N\geq\nu$; therefore

$$W = W_0(\gamma)\sum_{N\geq\nu}\exp\left[-\alpha(N-\nu)\right]S\left[\sqrt{\beta(N-\nu)}\right] \tag{3.10}$$

For the tunneling region which corresponds to $\gamma<<1$ there is a large number of important terms in the sum in eq. (3.10). For this reason it Is convenient to modify eq. (3.10) introducing the parameter $\delta = [\nu] + 1 - \nu$, where $[\nu]$ represents the integer part of ν. In this way we have

$$W = W_0(\gamma)\,\frac{\sqrt{\beta}}{2}\int_0^1 \frac{dx\; e^{-(\alpha+\beta x)}}{(1-x)^{1/2}}\,\phi(e^{-(\alpha+\beta x)},\,-1/2,\,\delta) \tag{3.11}$$

where $\phi(z,s,v)$ is the generalized Riemann zeta function[**] wtiich has the property

$$\lim_{z\to 1}(1-z)^{1-s}\phi(z,s,v) = \Gamma(1-s) \tag{3.12}$$

To evaluate the integral in eq. (3.10) in the limit $\gamma \ll 1$ we use eq. (3.12) to obtain

$$W = W_0(\gamma)\,\frac{\pi^{1/2}}{4\gamma}\,\frac{(m^*/m_1)^{1/2}}{\left[\left(1-\frac{m^*}{m_1}\right)+\frac{\gamma^2}{6}\left(\frac{3m^*}{m_1}-1\right)\right]^{1/2}} \tag{3.13}$$

The use of eq. (3.4) shows that, in this case, the exponential dependence of $W_0(\gamma)$ has the tunneling behavior (similar to KA)

$$\exp\left[-\frac{4}{3}\,\frac{E_c}{E_0}\,(1-\frac{1}{10}\gamma^2+\ldots)\right]$$

The present scheme is very convenient to understand quantitatively the accuracy of the saddle-point method because it allows the determination of the multiphoton transition rate in a closed analytical form prior to the saddle-point integration in the weak field limit. Most of the experiments are performed in this region of field intensities which corresponds to $\gamma \gg 1$, and very simple results can be obtained from eq. (3.6) very near the excitation threshold ($E_g \cong N\hbar\omega$ or $N \cong \nu$). For small N this could be an unfavorable situation for the application of the steepest descent method ($E_g/\hbar\omega \gg 1$), nevertheless as we show below for the case $E_g = \hbar\omega$, which corresponds to one photon absorption, the method may lead to fairly accurate results. In fact if in eq. (3.8) we approximate $S(x) \cong x$ and $\exp[-\alpha(N-\nu)] \cong 1$ we may obtain an expression valid for odd N, noticing that the introduction of the oscillating term leads to a multiplication factor 2 (see eq. (3.3) with $\psi \to +\pi/2$)

$$W^{(N)}_{\text{odd}} = \frac{1}{2\pi^2\hbar}\left(\frac{m_1\omega p_{vc}}{m}\right)^2\left(\frac{eA_0}{2m_1\omega c}\right)^{2N}\frac{\omega^{N-\frac{1}{2}}}{2^{N-5/2}}\,\frac{(m_1)^{N-1}}{\hbar^{N+3/2}}\,(m^*)^{3/2}\,\frac{\exp(N)}{N^N}\,(N-\nu)^{1/2} \tag{3.14}$$

This result must be compared with the dominant term near the threshold for odd N in the weak field limit before the saddle-point integration (see eq.(2.12)). This term is obtained taking $n = n' = \frac{N-1}{2}$,

$$W^{(N)}_{\text{w.f.}}(\text{odd}) = \frac{1}{2\pi\hbar}\left(\frac{m_1\omega p_{vc}}{m}\right)^2\left(\frac{eA_0}{2m_1\omega c}\right)^{2N}\frac{\omega^{N-\frac{1}{2}}}{2^{2N-7/2}}\,\frac{(m_1)^{N-1}}{\hbar^{N+3/2}}\,(m^*)^{3/2}\,\frac{(N-\nu)^{1/2}}{\left[\left(\frac{N-1}{2}\right)!\right]^2} \tag{3.15}$$

It is clear that the use of the saddle-point method preserves the original frequency dependence $(N-\nu)^{1/2}$ for all multiphoton processes of odd multiplicity involving excitations close to the threshold.

The ratio $W^{(N)}$ to $W^{(N)}_{w.f.}$ is readily obtained from eqs. (3.14) and (3.15)

$$\frac{W^{(N)}_{odd}}{W^{(N)}_{w.f.}(odd)} = \frac{1}{\pi}\,\frac{\left[\left(\frac{N-1}{2}\right)!\right]^2}{N^N}\cdot \exp(N) \qquad (3.16)$$

which for $N=1$ gives

$$\frac{W^{(1)}}{W^{(1)}_{w.f.}} = \frac{e}{\pi} \simeq 0.82 \qquad (3.17)$$

and increases with increasing N. We should notice that if we had followed KA, neglecting the oscillating terms the expression (3.13) should be divided by 2. Most importantly is that also for even photon multiplicities the omission of those terms, in the saddle-point integration, leads to a frequency dependence $(N-\nu)^{1/2}$ and to a multiphoton transition rate: given also by that of eq. (3.14) divided by a factor of 2 (this could account for the smallness of the values obtained from KA for even photon processes). This result is not in agreement with the perturbation theories as developed, for instance, by Braunstein and Ockman[15] arid by Basov et $al.$[14], which predicts a "forbidden absorption" proportional to $(N-\nu)^{3/2}$, for two-photon absorption. This feature, concerning a theory of the KA type, may be corrected taking into account the above mentioned oscillating terms. The result for transition rate involving processes with an even number of photons, after the use of the saddle-point method is

$$W^{(N)}_{(even)} = \frac{1}{2\pi^2\hbar}\left(\frac{m_1\omega p_{vc}}{m}\right)^2 \left(\frac{eA}{2m_1\omega c}\right)^{2N} \frac{\omega^{N-\frac{1}{2}}}{2^{N-9/2}}\,\frac{(m_1)^{N-2}}{\hbar^{N+3/2}}\,\frac{(m^*)^{5/2}}{N^{N-1}}\,\frac{\exp(N)}{3}$$

$$\times\left(1-\frac{1}{2N}\right)^2 (N-\nu)^{3/2} \qquad (3.18)$$

This expression should be compared with the weak field limit obtained from eq. (2.13) prior to the saddle-point integration. The leading term, near threshold, is obtained taking $n = n' = \frac{N-2}{2}$, which gives

$$W^{(N)}_{w.f.}(\text{even}) = \frac{1}{2\pi\hbar}\left(\frac{m_1\omega p_{vc}}{m}\right)^2\left(\frac{eA_0}{m_1\omega c}\right)^{2N}\frac{\omega^{N-\frac{1}{2}}}{2^{2N-13/2}}\frac{(m_1)^{N-2}(m^*)^{5/2}}{\hbar^{N+3/2}}\frac{(N-\nu)^{3/2}}{3\left[\left(\frac{N-2}{2}\right)!\right]^2} \tag{3.19}$$

The frequency behavior $(N-V)^{3/2}$ is, therefore, kept after the steepest descent integration.

The ratio for the even multiplicity processes is

$$\frac{W^{(N)}_{(\text{even})}}{W^{(N)}_{w.f.}(\text{even})} = \frac{2^{N-2}}{\pi}\frac{\left[\left(\frac{N-2}{2}\right)!\right]^2 \exp(N)}{N^{N-1}}\left(1-\frac{1}{2N}\right)^2 \tag{3.20}$$

For N=2 this ration is $9e^2/32\pi \simeq 0.66$, for N=10 it is $\simeq$ 1.03, and it increases with increasing N.

4. CONCLUSIONS

We have used a "non-perturbative" approach which leads to closed analytical results for the multiphoton transition rates in direct-gap crystals that can be described by a two parabolic band model. We have compared our results with a Keldysh type of formulation having in mind two points: the connection between multiphoton absorption and tunneling, and the use and accuracy of the saddle-point method as applied in the KA. A tunneling-like expression, for the total transition rate, has been obtained by summing over all allowed multiphoton processes.

In the weak field limit, the use of the steepest descent technique does not introduce modifications on the expected frequency dependente, at least near threshold, provided that the oscillating terms neglected in the KA are properly considered. Concerning specific numerical values for the multiphoton transition rate, we must rnention that the saddle-point integratlon may lead to unreliable results. Furthermore if the oscillating terms are ignored, as in the KA, the frequency dependence may be incorrect.

As a final conclusion we emphasize that the main problems with a KA type of approach is not with the use of the saddle-point method[7] but with the neglection of important oscillating terms in the weak field region.

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Resumo

Estudamos transições multifotônicas em cristais de "gap" direto usando um método "não-perturbativo" do tipo sugerido por Keldysh. Consideramos um modelo de duas bandas parabólicas na aproximação de massa efetiva, de modo que as soluções exatas da Hamiltoniana total podem ser aproximadas por uma solução do tipo de Volkov. Assim construímos uma matriz S que inclui, de modo aproximado, o campo eletromagnético em todas as ordens, permitindo a obtenção de soluções analíticas para a taxa de transição multifotônica. Discutimos a aplicação do método de ponto de sela e suas limitações, bem como a relação entre absorção multi-

fotônica e tunelamento. Mostramos que, para excitações perto do limiar, o mêtodo de Keldysh pode ser aplicado desde que se leve em conta os termos adequados na integração pelo método ponto de sela.