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On a new quasi-classical expansion for multiphoton processes in solids in intense far-infrared laser fields

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Abstract. In this paper a new quasi-classical expansion is defined. It consists of the asymptotic expansion of wavefunctions with respect to the Planck constant, which multiplies the time derivative in the Schrödinger equation. It is proved that such an approach provides a systematic and unequivocal scheme for studies of multiphoton processes in which a large number of photons are exchanged with a far-infrared laser field. The domain of validity of this approach is also discussed.

1. Introduction

The appearance of lasers capable of generating high-power far-infrared coherent radiation has revealed new aspects of the multiphoton interaction of electromagnetic radiation with solids. Subsequently, on the basis of these researches, new properties of many materials have been investigated, and new radiation detectors have been developed (see e.g. Dornhause and Nimtz 1976, Richter 1976, Vedenov *et al* 1982, Kaminskii 1981). Experimental investigations of multiphoton transitions in crystals have usually been made in the case of radiation with wavelengths in visible, near-infrared and middle-infrared ranges. For such frequencies the transition amplitude for an n-photon process is much higher than the transition amplitude for an $(n+1)$-photon process right up to light intensities corresponding to the damage threshold of investigated crystals. The application of far-infrared lasers has changed this situation to a large extent. This is due to the fact that the 'effective expansion parameter' for multiphoton transitions is proportional to $I\omega^{-3}$ (as will follow shortly), where I and ω are the intensity and the photon energy of the laser beam in atomic units (the atomic unit of intensity is 3.51×10^{16} W cm^{-2} and the atomic unit of energy is 27.2 eV). To be more specific, one can apply perturbation theory provided that the 'effective expansion parameter' is much smaller than 1 (this is in fact the definition of this parameter); in this case the transition amplitude for an n-photon process dominates the transition amplitude for an $(n+1)$-photon process. In the case of submillimetre radiation of wavelength in the order of 100 μm ($\omega \sim 0.5 \times 10^{-3}$ atomic units) the 'effective expansion parameter' approaches unity already for intensities of the order of MW cm^{-2}. For such intensities a new type of

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non-linear absorption of light has been reported for p-type Ge crystals subjected to high-power pulses from an NH_3 laser (Ganichev *et al* 1983). Also a general approach to multiphoton intraband transitions has recently been developed (Ganichev *et al* 1986).

Although multiphoton processes have been studied in solids for many years (see e.g. Ridley 1982), the invention of powerful far-infrared lasers has changed this subject to a large extent. This is due to the fact that in this case *multi* does not stand for two or even three, but rather 10 or more. A rigorous treatment of these processes would require solving numerically the Schrödinger equation describing a many-electron system coupled to an intense laser field. It appears, however, that such an approach is still very difficult to perform and that the only possibility of attacking this problem would consist of the development of a systematic and efficient expansion scheme. The aim of this paper is to develop such a scheme for absorption/emission of high-power far-infrared radiation which induces multiphoton transitions of free carriers between different conduction bands of a crystal. Some preliminary results have already been presented in Kamiński (1988a) where I have calculated the leading contribution to the so-called $1/n$ expansion for n-photon processes. However, it is not clear how to calculate systematically the next orders of this expansion and how to estimate its domain of validity. This is the subject of this paper. The notation used here is taken from my previous paper (Kamiński 1988a).

Other topics of the interaction of radiation with solids and the scattering of charged carriers in crystals are discussed for instance in an interesting monograph of Ridley (1982) and in the review article of Chattopadhyay and Queisser (1981).

2. Quasi-classical expansion

For simplicity we shall limit our discussion to scattering processes of electrons by impurities in the presence of intense far-infrared laser fields. Let me emphasise, however, that the non-perturbative scheme developed here can be applied to an arbitrary multiphoton process in which a large number of photons are exchanged between the laser field and matter; for instance, it can be used in studies of multiphoton ionisation of excitons.

Since we are interested in the interaction of matter with radiation generated by lasers, i.e. with a special kind of radiation that is characterised by its very high intensity and its coherence properties, the quasi-classical approximation of a laser field is adequate (see e.g. Manakov *et al* 1986). For a single-mode field this approach consists of treating the electromagnetic vector potential not as an operator but as a function that fulfils the classical Maxwell equations. For a multimode field, however, such an approximation is not further applicable because the quantum character of radiation implies that, in the limit of large intensities, the electromagnetic vector potential has to be treated as a stochastic process, an ensemble average over which should be performed at the end (Białynicki-Birula and Białynicka-Birula 1976, Mittleman 1982). This means that it suffices to determine different kinds of cross sections for a laser field and afterwards to average them over stochastic changes of laser-field parameters. This paper deals with the first part of this procedure. The second part is usually much more difficult to perform, especially if one wants to account for the interaction of radiation with matter in a non-perturbative manner; for a low-frequency radiation field a general procedure was presented in Kamiński (1988b). For more information about the quasi-classical approximation for the interaction of an intense radiation field with matter, one should read Białynicki-Birula and Białynicka-Birula (1976), Mittleman (1982), Manakov *et al* (1986) or

Ehlotzky (1985), whereas scattering processes in the presence of the quantised radiation field are discussed for instance in Rosenberg (1982) and Klinskikh and Rapoport (1985). Let me also note that both the quasi-classical and the quantum formalisms in the limit of large intensities give equivalent results and the notion of 'multiphoton processes' can be used as well in the quasi-classical formalism.

The scattering matrix element S_{fi} for the transition $(\boldsymbol{p}_i, n_i) \to (\boldsymbol{p}_f, n_f)$, where n_i and n_f label conduction bands, is equal to (Kamiński 1988a)

$$S_{fi} = \delta_{n_i n_f}\delta(\boldsymbol{p}_i - \boldsymbol{p}_f) - \frac{\mathrm{i}}{\hbar}(2\pi)^{-3}\int_{-\infty}^{+\infty} \mathrm{d}t\,\mathrm{d}^3r\,\psi^{(0)*}_{p_f,n_f}(\boldsymbol{r},t)V_{\mathrm{I}}(\boldsymbol{r})\psi^{(+)}_{p_i,n_i}(\boldsymbol{r},t) \tag{2.1}$$

where $\psi^{(0)}_{p_f,n_f}$ and $\psi^{(+)}_{p_i,n_i}$ are the time-dependent Bloch wavefunction and the outgoing-wave scattering wavefunction. These wavefunctions fulfil the following equations:

$$\mathrm{i}\hbar\partial_t\psi^{(0)}_{p_f,n_f} = \left(-\frac{\hbar^2}{2m}\Delta + V_{\mathrm{C}}(\boldsymbol{r}) + \frac{\mathrm{i}e\hbar}{m}\boldsymbol{A}(t)\boldsymbol{\nabla}\right)\psi^{(0)}_{p_f,n_f} \tag{2.2}$$

$$\mathrm{i}\hbar\partial_t\psi^{(+)}_{p_i,n_i} = \left(-\frac{\hbar^2}{2m}\Delta + V_{\mathrm{C}}(\boldsymbol{r}) + V_{\mathrm{I}}(\boldsymbol{r}) + \frac{\mathrm{i}e\hbar}{m}\boldsymbol{A}(t)\boldsymbol{\nabla}\right)\psi^{(+)}_{p_i,n_i} \tag{2.3}$$

where $V_{\mathrm{C}}(\boldsymbol{r})$ is a periodic-in-space potential, which describes the crystal lattice, whereas $V_{\mathrm{I}}(\boldsymbol{r})$ is a short-range potential, which approximates an impurity. The final state of electrons is described by (2.2), whereas the initial state, together with the whole Born series with respect to V_{I}, is described by (2.3). Let me emphasise that the impurity potential V_{I} can appear only in one of these equations, and that in the case of scattering one can arbitrarily choose one of these possibilities. Note that I have removed the term quadratic in $\boldsymbol{A}(t)$ by a phase transformation; although the $\boldsymbol{A}^2$ term in the Schrödinger equation is irrelevant for scattering processes, it has to be accounted for in photo-ionisation (see e.g. Kamiński 1988a, 1990a).

For optical fields of moderate intensities perturbation theory to the lowest non-vanishing order is an appropriate approach to the calculation of transition amplitudes. However, in the case of intense far-infrared fields the application of the standard perturbation theory to lowest non-vanishing order becomes insufficient (*even* for non-resonant processes), because higher-order terms become comparable to the lowest-order term and should be included (see e.g. Ganichev *et al* 1986), independently of uncertainties concerning the convergence of the summation. The alternative is to devise a direct, non-perturbative approach for solving the Schrödinger equation in the presence of a strong far-infrared laser field. The aim of this section is to propose a new quasi-classical expansion, which can effectively describe multiphoton processes with a large number of absorbed or emitted photons. In these investigations the frequency of the alternating field is supposed to be non-resonant with respect to eigenfrequencies of the quantum system. This gives us the possibility to restrict our considerations to the purely monochromatic fields without accounting for the non-monochromaticity effects, which essentially determine the character of resonances. Owing to this limitation I shall denote the time dependence of electromagnetic potential (and also other related quantities) interchangeably by $\boldsymbol{A}(t)$ or $\boldsymbol{A}(\omega t)$, where ω is the laser frequency.

Guided by the results obtained in Kamiński (1988a) I am seeking solutions of (2.2) and (2.3) in the form

$$\psi^{(0)}_{p_f,n_f} = \psi^{(0)}_f = \exp\left(-\frac{\mathrm{i}}{\hbar}E_f t - \frac{\mathrm{i}}{\hbar}\boldsymbol{q}_f\boldsymbol{\alpha}(\omega t)\right)\phi^{(0)}_f \tag{2.4}$$

and

$$\psi^{(+)}_{p_i,n_i} = \psi^{(+)}_i = \exp\left(-\frac{\mathrm{i}}{\hbar}E_i t - \frac{\mathrm{i}}{\hbar}\boldsymbol{q}_i\boldsymbol{\alpha}(\omega t)\right)\phi^{(+)}_i \tag{2.5}$$

where $E_i = E_{n_i}(\boldsymbol{p}_i)$, $E_f = E_{n_f}(\boldsymbol{p}_f)$, $\boldsymbol{q}_i = \boldsymbol{q}_{n_i}(\boldsymbol{p}_i) = m\partial E_{n_i}(\boldsymbol{p}_i)/\partial \boldsymbol{p}_i$, and $\boldsymbol{q}_f = \boldsymbol{q}_{n_f}(\boldsymbol{p}_f) = m\partial E_{n_f}(\boldsymbol{p}_f)/\partial \boldsymbol{p}_f$. The function $\boldsymbol{\alpha}(t)$ is determined by the vector potential $\boldsymbol{A}(t)$,

$$m\partial_t\boldsymbol{\alpha}(t) = -e\boldsymbol{A}(t). \tag{2.6}$$

The vectors $\boldsymbol{v}_i = \boldsymbol{q}_i/m$ and $\boldsymbol{v}_f = \boldsymbol{q}_f/m$ are the group velocities of initial and final wavepackets, whereas $\boldsymbol{\alpha}(t)$ describes the classical motion of a charged particle in the presence of an electromagnetic plane wave. As follows from the Floquet theorem, $\phi^{(0)}_f$ and $\phi^{(+)}_i$ are periodic functions of time, with period equal to $2\pi/\omega$. These functions fulfil the following equations (note that owing to the time periodicity one can replace the time derivative ∂_t by $\omega\partial_\varphi$):

$$\left(E_f - H_{\mathrm{C}} + \frac{e}{m}\boldsymbol{A}(\omega t)(\boldsymbol{P} - \boldsymbol{q}_f) + \mathrm{i}\hbar\omega\partial_\varphi\right)\phi^{(0)}_f(\boldsymbol{r},\varphi) = 0 \tag{2.7}$$

$$\left(E_i - H_0 + \frac{e}{m}\boldsymbol{A}(\omega t)(\boldsymbol{P} - \boldsymbol{q}_i) + \mathrm{i}\hbar\omega\partial_\varphi\right)\phi^{(+)}_i(\boldsymbol{r},\varphi) = 0 \tag{2.8}$$

where $H_{\mathrm{C}} = -\hbar^2\Delta/2m + V_{\mathrm{C}}(\boldsymbol{r})$, $H_0 = H_{\mathrm{C}} + V_{\mathrm{I}}(\boldsymbol{r})$, $\boldsymbol{P} = -\mathrm{i}\hbar\boldsymbol{\nabla}$ and $\varphi = \omega t$. I shall use further the Dirac notation. The substitution of (2.4) and (2.5) into (2.1) leads to the scattering matrix, which I write down in the form

$$S_{fi} = \sum_{n=-\infty}^{\infty} S^{(n)}_{fi} \tag{2.9}$$

where

$$S^{(n)}_{fi} = \delta_{n_i n_f}\delta(\boldsymbol{p}_i - \boldsymbol{p}_f)\delta_{n,0} + \frac{\mathrm{i}}{2\pi m}\delta(E_f - E_i - n\hbar\omega)\mathcal{F}^{(n)}_{fi} \tag{2.10}$$

and

$$\mathcal{F}^{(n)}_{fi} = -\frac{m}{4\pi^2\hbar^2}\int_{-\pi}^{\pi}\mathrm{d}\varphi\,\exp\left(\mathrm{i}n\varphi + \frac{\mathrm{i}}{\hbar}\boldsymbol{\Delta}_{fi}\boldsymbol{\alpha}(\varphi)\right)\langle f,0;\varphi|V_{\mathrm{I}}|i,+;\varphi\rangle. \tag{2.11}$$

In the above equation $\langle\boldsymbol{r}|f,0;\varphi\rangle = \phi^{(0)}_f(\boldsymbol{r},\varphi)$, $\langle\boldsymbol{r}|i,+;\varphi\rangle = \phi^{(+)}_i(\boldsymbol{r},\varphi)$ and $\boldsymbol{\Delta}_{fi} = \boldsymbol{q}_f - \boldsymbol{q}_i$. This result is derived by using the Fourier decomposition of periodic functions $\phi^{(+)}_i$ and $\phi^{(0)}_f$ and performing the integration over t in (2.1), which leads to the δ function in (2.10) expressing the condition of the conservation of energy. Note that the space integration is present in the scalar product $\langle f, 0; \varphi | V_{\rm I} | i, +; \varphi \rangle$,

$$\langle f, 0; \varphi | V_{\rm I} | i, +; \varphi \rangle = \int_{-\infty}^{\infty} \mathrm{d}^3 r\, \phi_f^{(0)*}(\boldsymbol{r}, \varphi) V_{\rm I}(\boldsymbol{r}) \phi_i^{(+)}(\boldsymbol{r}, \varphi).$$

We now define a quasi-classical expansion,

$$|i, +; \varphi\rangle = \sum_{l=0}^{\infty} \hbar^l |i, +, l; \varphi\rangle \tag{2.12}$$

$$|f, 0; \varphi\rangle = \sum_{l=0}^{\infty} \hbar^l |f, 0, l; \varphi\rangle \tag{2.13}$$

in such a way that

$$\left(E_i - H_0 + \frac{e}{m}\boldsymbol{A}(\varphi)(\boldsymbol{P} - \boldsymbol{q}_i)\right) |i, +, 0; \varphi\rangle = 0 \tag{2.14}$$

$$\left(E_f - H_{\rm C} + \frac{e}{m}\boldsymbol{A}(\varphi)(\boldsymbol{P} - \boldsymbol{q}_f)\right) |f, 0, 0; \varphi\rangle = 0 \tag{2.15}$$

and

$$|i, +, l+1; \varphi\rangle = -\mathrm{i}\omega\left(E_i - H_0 + \frac{e}{m}\boldsymbol{A}(\varphi)(\boldsymbol{P} - \boldsymbol{q}_i)\right)^{-1} \partial_\varphi |i, +, l; \varphi\rangle \tag{2.16}$$

$$|f, 0, l+1; \varphi\rangle = -\mathrm{i}\omega\left(E_f - H_{\rm C} + \frac{e}{m}\boldsymbol{A}(\varphi)(\boldsymbol{P} - \boldsymbol{q}_f)\right)^{-1} \partial_\varphi |f, 0, l; \varphi\rangle. \tag{2.17}$$

These equations follow from the substitution of (2.12) and (2.13) into (2.8) and (2.7) respectively, and from the comparison of terms multiplied by a given power of $\hbar\omega$. Let me note in passing that this is the asymptotic expansion with respect to the Planck constant which multiplies the time derivative in the Schrödinger equation, whereas the Planck constant which multiplies the space derivative is accounted for exactly; this is the reason why this expansion can be called as a quasi-classical one. From now on units in which $\hbar = 1$ are used. The scattering amplitude $\mathcal{F}_{fi}^{(n)}$ now adopts the form

$$\mathcal{F}_{fi}^{(n)} = \sum_{k=0}^{\infty} \mathcal{F}_{fi,k}^{(n)} \tag{2.18}$$

where

$$\mathcal{F}_{fi,k}^{(n)} = -\frac{m}{2\pi}\sum_{l=0}^{k}\frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{d}\varphi\, \exp[\mathrm{i}n\varphi + \mathrm{i}\boldsymbol{\Delta}_{fi}\boldsymbol{\alpha}(\varphi)] \langle f, 0, l; \varphi | V_{\rm I} | i, +, k-l; \varphi\rangle. \tag{2.19}$$

The aim of this paper is to prove that in the limit of a large number of absorbed or emitted photons (i.e. for $n \gg 1$) and for given $\boldsymbol{\alpha}_0$ (defined by (3.2)) and $n\omega = E_f - E_i$, the term $\mathcal{F}_{fi,k}^{(n)}$ behaves like n^{-k}. Hence, the leading contribution to the scattering amplitude $\mathcal{F}_{fi}^{(n)}$ is supplied by $\mathcal{F}_{fi,0}^{(n)}$, and the first correction to it is given by $\mathcal{F}_{fi,1}^{(n)}$. Further corrections can also be accounted for in the unequivocal manner by the inclusion of $\mathcal{F}_{fi,k}^{(n)}$ for $k = 2, 3$, etc. This statement is valid for an arbitrary polarisation of light.

It is difficult to say how large the number of exchanged photons should be in order to apply effectively the quasi-classical expansion developed above. For instance, the model calculation shows (Shakeshaft and Robinson 1982) that even the case of $n = 2$ seems to give reasonable agreement with the Kroll–Watson formula. This problem, however, has to be studied more carefully and I hope that recently developed one-dimensional models (Kamiński 1990b, 1990c) will shed more light on the applicability of this expansion.

The physical meaning of the quasi-classical expansion can be understood qualitatively on the basis of the condition of the conservation of energy, $n\hbar\omega = E_f - E_i$. Indeed, for a given $E_f - E_i$ the limit $\hbar \to 0$ corresponds to the limit $|n| \to \infty$. Let me emphasise, however, that this condition does not provide any limitations on attainable intensities of laser beams; these limitations follow from an analysis presented in the next sections.

3. Asymptotic behaviour of quasi-classical expansion for large n

For an elliptically polarised electromagnetic plane wave the vector potential in the dipole approximation adopts the form

$$A(\varphi) = \tfrac{1}{2}(\boldsymbol{a}\,\mathrm{e}^{-\mathrm{i}\varphi} + \boldsymbol{a}^*\,\mathrm{e}^{\mathrm{i}\varphi}) \tag{3.1}$$

where a constant vector $\boldsymbol{a}$ has in general complex coordinates; for linearly polarised light $\boldsymbol{a}$ is a real vector, hence $A(\varphi) = \boldsymbol{a}\cos\varphi$. Let me now define the following quantities:

$$\boldsymbol{\alpha}_0 = -\frac{e}{m\omega}\boldsymbol{a} \tag{3.2}$$

$$\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi} = |\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|\,\mathrm{e}^{-\mathrm{i}\delta_{fi}} \tag{3.3}$$

$$Q_{\mathrm{C}i,f} = \tfrac{1}{2}(\boldsymbol{\alpha}_0\boldsymbol{Q}_{i,f}\,\mathrm{e}^{\mathrm{i}\delta_{fi}} + \boldsymbol{\alpha}_0^*\boldsymbol{Q}_{i,f}\,\mathrm{e}^{-\mathrm{i}\delta_{fi}}) \tag{3.4}$$

$$Q_{\mathrm{S}i,f} = \frac{1}{2\mathrm{i}}(\boldsymbol{\alpha}_0\boldsymbol{Q}_{i,f}\,\mathrm{e}^{\mathrm{i}\delta_{fi}} - \boldsymbol{\alpha}_0^*\boldsymbol{Q}_{i,f}\,\mathrm{e}^{-\mathrm{i}\delta_{fi}}) \tag{3.5}$$

where $\boldsymbol{Q}_{i,f} = \boldsymbol{P} = \boldsymbol{q}_{i,f}$. As follows from (3.2) and (3.3), $\boldsymbol{\Delta}_{fi}\boldsymbol{\alpha}(\varphi)$ is equal to $-|\boldsymbol{\alpha}_0\boldsymbol{\Delta}^{fi}|\sin(\varphi + \delta^{fi})$. Moreover, the quasi-classical expansions (2.14)–(2.17) now adopt the form

$$[E_i - H_0 - \omega Q_{\mathrm{C}i}\cos(\varphi + \delta_{fi}) - \omega Q_{\mathrm{S}i}\sin(\varphi + \delta_{fi})]\,|i, +, 0; \varphi\rangle = 0 \tag{3.6}$$

$$[E_f - H_{\mathrm{C}} - \omega Q_{\mathrm{C}f}\cos(\varphi + \delta_{fi}) - \omega Q_{\mathrm{S}f}\sin(\varphi + \delta_{fi})]\,|f, 0, 0; \varphi\rangle = 0 \tag{3.7}$$

and

$$|i, +, l+1; \varphi\rangle = -\mathrm{i}\omega[E_i - H_0 - \omega Q_{\mathrm{C}i}\cos(\varphi + \delta_{fi}) - \omega Q_{\mathrm{S}i}\sin(\varphi + \delta_{fi})]^{-1} \times \partial_\varphi |\mathrm{i}, +, \mathrm{l}; \varphi\rangle \tag{3.8}$$

$$|f, 0, l+1; \varphi\rangle = -\mathrm{i}\omega[E_f - H_{\mathrm{C}} - \omega Q_{\mathrm{C}f}\cos(\varphi + \delta_{fi}) - \omega Q_{\mathrm{S}f}\sin(\varphi + \delta_{fi})]^{-1} \times \partial_\varphi |f, 0, l; \varphi\rangle. \tag{3.9}$$

It is now clear that for a given α_0 (hence, for given $Q_{Ci,f}$ and $Q_{Si,f}$) the lth term of the quasi-classical expansion can be represented as follows:

$$|i,+,l;\varphi\rangle = \sum_{j=0}^{\infty} \omega^{l+j} \sum_{k=0}^{j} \cos^k(\varphi+\delta_{fi})|i, ljk, C\rangle + \sum_{j=1}^{\infty} \omega^{l+j} \sum_{k=0}^{j} \cos^{k-1}(\varphi+\delta_{fi})\sin(\varphi+\delta_{fi})|i, ljk, S\rangle \tag{3.10}$$

and similarly for $|f,0,l;\varphi\rangle$. Important in these representations is that the kets $|i, ljk, C\rangle$, etc., are independent of φ and depend on the laser parameters only through $\boldsymbol{\alpha}_0$ and δ^{fi}, i.e. through $Q_{Ci,f}$ and $Q_{Si,f}$, but not on ω separately. Having this in mind we arrive at the following expression for $\mathscr{F}^{(n)}_{fi,k}$:

$$\mathscr{F}^{(n)}_{fi,k} = \mathrm{e}^{-\mathrm{i}n\delta_{fi}} \sum_{j=0}^{\infty} \omega^{k+j} \sum_{l=0}^{j} C^l_n(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)C_{kjl}(\boldsymbol{p}_i, n_i; \boldsymbol{p}_f, n_f; \boldsymbol{\alpha}_0) + \mathrm{e}^{-\mathrm{i}n\delta_{fi}} \sum_{j=1}^{\infty} \omega^{k+j} \sum_{l=0}^{j} S^l_n(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)S_{kjl}(\boldsymbol{p}_i, n_i; \boldsymbol{p}_f, n_f; \boldsymbol{\alpha}_0) \tag{3.11}$$

where the functions $C^l_n(z)$ and $S^l_n(z)$ are defined in the appendix (equations (A1) and (A28)). Explicit forms of C_{kjl} and S_{kjl} are not necessary for our further discussion; important, however, is that these functions do not depend on n and ω.

Now we can perform the limit $n\to\infty$ for a given energy transfer (from the laser field to matter) and for a given α_0. Taking into account asymptotic expansions of C^l_n and S^l_n (equations (A31) and (A32)) we conclude that the leading term of $\mathscr{F}^{(n)}_{fi,k}$ behaves as

$$\mathscr{F}^{(n)}_{fi,k} = \mathrm{e}^{-\mathrm{i}n\delta_{fi}} n^{-k} J_n(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|) \sum_{j=0}^{\infty} (E_f - E_i)^{k+j} (|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)^{-j} \times C_{kjj}(\boldsymbol{p}_i, n_i, \boldsymbol{p}_f, n_f, \boldsymbol{\alpha}_0) + \mathrm{O}(n^{-k-1}). \tag{3.12}$$

Let me note in passing that although in the above sum we have singular terms for $\alpha_0 = 0$ (i.e. $(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)^{-j}$), these singularities are cancelled, because functions C_{kjj} are proportional to α_0^j.

Hence, we see that in the above mentioned limit ($n\to\infty$, $n\omega$ and α_0 fixed),

$$\mathscr{F}^{(n)}_{fi,k} = \mathrm{e}^{-\mathrm{i}n\delta_{fi}} J_n(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)[n^{-k}F_{fi,k} + \mathrm{O}(n^{-k-1})] + \mathrm{e}^{-\mathrm{i}n\delta_{fi}} J'_n(|\boldsymbol{\alpha}_0\boldsymbol{\Delta}_{fi}|)[\mathrm{O}(n^{-k-1})] \tag{3.13}$$

where $F_{fi,k}$ depends only on $E_f - E_i$ and α_0. This result proves our previous statement that the leading contribution to the scattering amplitude $\mathscr{F}^{(n)}_{fi}$ with a large number of absorbed (or emitted) photons from (or to) the laser field is given by the zeroth-order term of the quasi-classical expansion (2.18) and that corrections to this approximation can be accounted for unequivocally by calculating next orders of the quasi-classical expansion (2.18).

4. Domain of validity of the quasi-classical expansion

Although we have considered so far the limit of large n for given $n\omega$ and α_0, the conclusion of the previous section also applies to the limit of large n for given $n\omega$ and

$\tilde{\alpha}_0$, where $\tilde{\alpha}_0 = \alpha_0/n$. Indeed, in this case equation (3.11) can be written down as follows (we consider for simplicity positive values of n):

$$\mathscr{F}^{(n)}_{fi,k} = \mathrm{e}^{-in\delta_{fi}} \sum_{j=0}^{\infty} \omega^k (n\omega)^j \sum_{l=0}^{j} \mathrm{C}^l_n(n|\tilde{\boldsymbol{\alpha}}_0 \boldsymbol{\Delta}_{fi}|) \tilde{C}_{kjl}(\boldsymbol{p}_i, n_i; \boldsymbol{p}_f, n_f; \tilde{\boldsymbol{\alpha}}_0)$$
$$+ \mathrm{e}^{-in\delta_{fi}} \sum_{j=1}^{\infty} \omega^k (n\omega)^j \sum_{l=0}^{j} \mathrm{S}^l_n(n|\tilde{\boldsymbol{\alpha}}_0 \boldsymbol{\Delta}_{fi}|) \tilde{S}_{kjl}(\boldsymbol{p}_i, n_i; \boldsymbol{p}_f, n_f; \tilde{\boldsymbol{\alpha}}_0). \tag{4.1}$$

Proceeding in the similar manner as before and applying the asymptotic expansions (A27) and (A30) we arrive at the same conclusion. However, now the domain of validity of the quasi-classical expansion is larger. Indeed, the qualitative estimation of it can be based on the applicability of the asymptotic expansions of $C^l_n(z)$ and $S^l_n(z)$ for large n, where $z = |\boldsymbol{\alpha}_0 \boldsymbol{\Delta}_{fi}| = n|\tilde{\boldsymbol{\alpha}}_0 \boldsymbol{\Delta}_{fi}|$. As follows from the analysis performed in the appendix these expansions are valid provided that $n \gg 1$ and $|\boldsymbol{\alpha}_0 \boldsymbol{\Delta}_{fi}| \lesssim n$, where the symbol $\lesssim$ means less than or in the vicinity of. Qualitatively, these conditions can be written down as

$$I \lesssim I_0 \omega^4 n^2 / \boldsymbol{\Delta}^2_{fi} \qquad n \gg 1 \tag{4.2}$$

where ω and $\boldsymbol{\Delta}^2_{fi}$ are in the atomic unit of energy, $I_0 = 3.51 \times 10^{16}$ W cm^{-2} and the intensity of radiation I is in W cm^{-2}. For instance, for $n \sim 10$, $\omega \sim 0.5 \times 10^{-3}$ au and $\boldsymbol{\Delta}^2_{fi} \sim 5 \times 10^{-2}$ au (i.e. for $\boldsymbol{\Delta}^2_{fi}$ of the order of 1 eV) the intensity I can reach 10^{11} W cm^{-2}, whereas for $\boldsymbol{\Delta}^2_{fi}$ of the order of ω, the intensity can be of the order of 10^{13} W cm^{-2} or less, hence much larger than the intensity corresponding to the damage threshold of the investigated crystal. Let me note, however, that conditions (4.2) are necessary ones; sufficient conditions certainly depend on a particular process. For instance, one can expect that intermediate resonances can change these estimations significantly.

The applicability of standard perturbation theory can be characterised by the condition $|\boldsymbol{\alpha}_0 \boldsymbol{\Delta}_{fi}| \ll 1$, under which one can expand the Bessel functions in power series. Approximating now $\boldsymbol{\Delta}^2_{fi}$ by ω and knowing that $\boldsymbol{\alpha}^2_0$ is of the order of $I\omega^{-4}$, where I and ω are in atomic units, one obtains the following estimation:

$$I\omega^{-3} \ll 1. \tag{4.3}$$

This is exactly the condition mentioned in the introduction and also obtained in Ganichev *et al* (1986).

5. Conclusions

The aim of this paper was to introduce a new quasi-classical expansion and to discuss its domain of validity. It appears, in particular, that this approach provides a systematic and unequivocal expansion scheme for multiphoton processes, in which the number of absorbed or emitted photons is large. The domain of validity of this expansion has also been discussed, showing that it can be applied to free–free transitions in solids in the presence of an intense far-infrared laser field.

The quasi-classical approximation defined in this paper can immediately be applied to photoionisation processes (see e.g. Kamiński (1988a), in which only the leading term has been considered). In this way one can give an analytical justification for the Keldysh-type theory (Keldysh 1965) and estimate its domain of validity, i.e. to treat problems that have not been studied before. These problems are considered elsewhere (Kamiński 1990a).

Considerably more work is needed before one can obtain (by performing numerical calculations) practical results. Let me, however, emphasise that the quasi-classical expansion developed in this paper suggests in fact in which direction such a calculation should go, i.e. which approximations should be applied and what are their limitations; other Keldysh-type approaches do not give such possibilities.

At the end of this paper let me make a short historical note. It follows from equations (2.14) and (2.15) that the leading terms of our quasi-classical expansion are given by the wavefunctions defined by equations (29) and (30) in Kamiński (1988a), provided that we further apply the low-temperature approximation (equation (26) in Kamiński (1988a)). Similar approximations, although in a different context and without rigorous discussions of their domains of validity, appeared many years ago in Houston (1940). Applications of such an approximation in solids to the so-called Bloch oscillations have recently been discussed (Krieger and Iafrate 1986), where also a brief historical account of some controversies is given; for further applications see e.g. Chalbaud and Gallinar (1989).

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Appendix

The aim of this appendix is to find the asymptotic expansion for large n of the function $C_n^l(z)$ defined by the integral

$$C_n^l(z) = \frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{d}\varphi \cos^l(\varphi) \exp(\mathrm{i}n\varphi - \mathrm{i}z \sin\varphi) \tag{A1}$$

where n and l are integers and z is a real number. It follows from the definition (A1) that

$$C_n^0(z) = J_n(z) \tag{A2}$$

$$C_n^1(z) = \frac{n}{z} J_n(z) \tag{A3}$$

$$C_n^2(z) = \frac{n^2}{z^2} J_n(z) - \frac{1}{z} J_n'(z) \tag{A4}$$

where J_n and J'_n are the Bessel functions of order n and its derivative. These examples suggest that $C^l_n(z)$ can be looked for in the form

$$C^l_n(z) = A^l_n(z)J_n(z) + B^l_n(z)J'_n(z) \tag{A5}$$

where the asymptotic behaviour of unknown functions $A^l_n(z)$ and $B^l_n(z)$ is to be determined. To this end let me note that the functions $C^l_n(z)$ satisfy the following recurrence relation:

$$C^{l+1}_n(z) = \frac{n}{z} C^l_n(z) - \frac{l}{z}\frac{\mathrm{d}}{\mathrm{d}z} C^{l-1}_n(z). \tag{A6}$$

Taking into account the well known properties of the Bessel functions of integer order,

$$J_{n+1}(z) + J_{n-1}(z) = (2n/z)J_n(z) \tag{A7}$$

$$J_{n-1}(z) - J_{n+1}(z) = 2J'_n(z) \tag{A8}$$

$$J''_n(z) = (n^2/z^2 - \tfrac{1}{2})J_n(z) - \tfrac{1}{2}J'_n(z) \tag{A9}$$

one can immediately show that $A^l_n(z)$ and $B^l_n(z)$ fulfil the system of coupled recurrence relations, which can be written down as

$$\alpha^{l+1}_n(z) = \alpha^l_n(z) - \frac{l}{z}\left(1 - \frac{z^2}{2n^2}\right)\beta^{l-1}_n(z) + \frac{l(l-1)}{n^2}\alpha^{l-1}_n(z) - \frac{lz}{n^2}\alpha'^{l-1}_n(z) \tag{A10}$$

$$\beta^{l+1}_n(z) = \beta^l_n(z) - \frac{lz}{n^2}\alpha^{l-1}_n(z) + \frac{l^2}{n^2}\beta^{l-1}_n(z) - \frac{lz}{n^2}\beta'^{l-1}_n(z) \tag{A11}$$

where $\alpha^l_n(z)$ and $\beta^l_n(z)$ are defined by $A^l_n(z)$ and $B^l_n(z)$ by means of the equalities

$$A^l_n(z) = \left(\frac{n}{z}\right)^l \alpha^l_n(z) \tag{A12}$$

$$B^l_n(z) = \left(\frac{n}{z}\right)^l \beta^l_n(z). \tag{A13}$$

Introducing a new variable $x = z/n$ and defining $a^l_n(x)$ and $b^l_n(x)$ as

$$a^l_n(x) = \alpha^l_n(z) \tag{A14}$$

and

$$b^l_n(x) = \beta^l_n(z) \tag{A15}$$

one arrives at the following system of coupled recurrence relations for a^l_n and b^l_n:

$$a^{l+1}_n(x) = a^l_n(x) - \frac{l}{nx}\left(1 - \tfrac{1}{2}x^2\right) b^{l-1}_n(x) + \frac{l(l-1)}{n^2} a^{l-1}_n(x) - \frac{lx}{n^2} a'^{l-1}_n(x) \tag{A16}$$

$$b^{l+1}_n(x) = b^l_n(x) - \frac{lx}{n} a^{l-1}_n(x) + \frac{l^2}{n^2} b^{l-1}_n(x) - \frac{lx}{n^2} b'^{l-1}_n(x) \tag{A17}$$

where the prime now means the derivative with respect to x. As follows from these relations, for given l and x and for large n the functions $a_n^l(z)$ and $b_n^l(z)$ can be sought in the form of power series with respect to $1/n$,

$$a_n^l(x) = a_{nl}^{(0)}(x) + a_{nl}^{(1)}(x) + a_{nl}^{(2)}(x) + \ldots \tag{A18}$$

$$b_n^l(x) = b_{nl}^{(0)}(x) + b_{nl}^{(1)}(x) + b_{nl}^{(2)}(x) + \ldots \tag{A19}$$

where $a_{nl}^{(k)}(x)$ and $b_{nl}^{(k)}(x)$ are proportional to n^{-k}. In the leading approximation we have

$$a_{n,l+1}^{(0)}(x) = a_{nl}^{(0)}(x) \tag{A20}$$

$$b_{n,l+1}^{(0)}(x) = b_{nl}^{(0)}(x). \tag{A21}$$

These equations can easily be solved if we take into account that, according to (A2), $a_{n0}^{(0)}(x) = 1$ and $b_{n0}^{(0)}(x) = 0$. Hence,

$$a_{nl}^{(0)}(x) = 1 \tag{A22}$$

and

$$b_{nl}^{(0)}(x) = 0. \tag{A23}$$

In the next approximation we have

$$b_{n,l+1}^{(1)}(x) = b_{nl}^{(1)}(x) - lx/n. \tag{A24}$$

A general solution of this recurrence relation is of the form

$$b_{nl}^{(1)}(x) = B_n^{(1)}(x) - l(l-1)x/2n \tag{A25}$$

where $B_n^{(1)}(x)$ is an l-independent function. However, since $b_{n0}^{(1)}(x)$ is equal to zero for $l = 0$ (see equation (A2)), therefore, $B_n^{(1)}(x) = 0$ and

$$b_{nl}^{(1)}(x) = -\, l(l-1)x/2n. \tag{A26}$$

In a similar manner one can find that $a_{nl}^{(1)}(x) = 0$.

To recapitulate we have found the following asymptotic expansion of the function $C_n^l(nx)$ for large n:

$$C_n^l(nx) = [1 + \mathrm{O}(n^{-2})]x^{-1}J_n(nx) + [-\, l(l-1)/2n + \mathrm{O}(n^{-2})]x^{-l+1}J_n'(nx) \tag{A27}$$

valid for given l and x. The domain of validity of this expansion is limited to such values of x that are of the order of 1 or less, hence for $z \lesssim n$, where the symbol $\lesssim$ means less than or in the vicinity of. For x of the order of 1 the validity of the above asymptotic expansion is 'accelerated' by the Watson asymptotic expansion of the Bessel functions (see e.g. Erdelyi 1953); indeed, $J_n(nx)$ has a maximum for $x \sim 1$, hence $J_n'(nx)$ approaches zero and the corrections to the leading approximation of (A27) are proportional to $1/n^2$.

Knowing the asymptotic expansion of the integral (A1), one can easily arrive at a similar expansion for the following integral:

$$S_n^l(z) = \frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{d}\varphi \cos^{l-1}(\varphi) \sin\varphi \exp(\mathrm{i}n\varphi - \mathrm{i}z\sin\varphi). \tag{A28}$$

Indeed, to this end it is sufficient to note that

$$S_n^{l+1}(z) = \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}z} C_n^l(z). \tag{A29}$$

Hence, for given l and x, and for large n,

$$S_n^l(nx) = \tilde{a}_n^l(x)x^{-1}J_n(nx) + \tilde{b}_n^l(x)x^{-l+1}J'_n(nx) \tag{A30}$$

where $\tilde{a}_n^l(x)$ is of the order of $1/n$, whereas $\tilde{b}_n^l(x)$ is of the order of 1.

Proceeding in a similar manner as before, one can prove that for given z and l and in the limit of large n,

$$C_n^l(z) = \left(\frac{n}{z}\right)^l J_n(z) + \left(\frac{n}{z}\right)^l [\mathrm{O}(n^{-2})J_n'(z) + \mathrm{O}(n^{-2})J_n(z)] \tag{A31}$$

and similarly for $S_n^l(z)$,

$$S_n^l(z) = \left(\frac{n}{z}\right)^l [\mathrm{O}(n^{-1})J_n(z) + \mathrm{O}(n^{-1})J_n'(z)]. \tag{A32}$$

References

Białynicki-Birula I and Białynicka-Birula Z 1976 *Phys. Rev.* A **14** 1101
Chalbaud E R and Gallinar J O 1989 *J. Phys.: Condens. Matter* **1** 3325
Chattopadhyay D and Queisser H J 1981 *Rev. Mod. Phys.* **53** 747
Dornhaus R and Nimtz G 1976 *Springer Tracts in Modern Physics* (Berlin: Springer) vol 78, p 1
Ehlotzky F 1985 *Can. J. Phys.* **63** 907
Erdelyi A 1953 *Bateman Manuscript Project* (New York: McGraw-Hill) p 89
Ganichev S D, Emel'yanov S A, Ivchenko E L, Perlin E Yu, Terent'ev Ya V, Fedorov A V and Yaroshetskii I D 1986 *Sov. Phys.–JETP* **64** 729
Ganichev S D, Emel'yanov S A, Ivchenko E L, Perlin E Yu and Yaroshetskii I D 1983 *Sov. Phys.–JETP Lett.* **37** 568
Houston W V 1940 *Phys. Rev.* **57** 184
Kamiński J Z 1986 *Phys. Scr.* **34** 770
—— 1988a *J. Phys. C: Solid State Phys.* **21** 3983
—— 1988b *J. Phys. A: Math. Gen.* **21** 699
—— 1990a *Phys. Scr.* at press
—— 1990b *Phys. Rev.* A at press
—— 1990c *Z. Phys.* D at press
Kaminskii A A 1981 *Laser Crystals* (Berlin: Springer)
Keldysh L V 1965 *Sov. Phys.–JETP* **20** 1307
Klinskikh A F and Rapoport L P 1985 *Sov. Phys.–JETP* **61** 649
Krieger J B and Iafrate G J 1986 *Phys. Rev.* A **33** 5494
Manakov N L, Ovsiannikov V D and Rapoport L P 1986 *Phys. Rep.* **141** 319
Mittleman M H 1982 *Introduction to the Theory of Laser-Atom Interactions* (New York: Plenum)
Richter W 1976 *Springer Tracts in Modern Physics* (Berlin: Springer) vol 78, p 121
Ridley B K 1982 *Quantum Processes in Semiconductors* (Oxford: Clarendon)
Rosenberg L 1982 *Adv. At. Mol. Phys.* **18** 1
Shakeshaft R and Robinson E J 1982 *Phys. Rev.* A **25** 1977
Vedenov A A, Myl'nikov G D and Sobolenko D N 1982 *Sov. Phys.–Usp.* **25** 833