

Coherent phase control of free–free transitions in bichromatic laser fields

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Abstract. We consider the coherent phase control of the nonlinear cross sections of induced and inverse bremsstrahlung in a powerful bichromatic laser field. The two field components have frequencies $r\omega$ and $s\omega$, where r and s are small positive integers, and the relative phase φ of both components can be changed arbitrarily. Amending our earlier work on this problem (1993 *Phys. Rev. A* 47 715, 1993 *Opt. Commun.* 99 177) we show that for the ratios $r : s = 1 : 2$, $1 : 3$ and $2 : 3$ the phase-dependent changes of the free–free cross sections are quite appreciable, whereas for increasing values of s the phase dependence gradually disappears, assuming both field components have the same intensity. If, however, the intensity of the harmonic field component is considerably increased, phase dependent effects again show up even for larger values of s .

1. Introduction

Free–free transitions in a powerful, monochromatic laser field have been considered by many authors since the fundamental paper of Bunkin and Fedorov appeared [1]. Surveys on this subject can be found in the books by Mittleman [2] and by Faisal [3]. Recently, it has become feasible experimentally to coherently control the phase φ between a fundamental laser field component of frequency ω and one of its harmonics. This possibility stimulated a considerable body of research work on the coherent phase control of multiphoton processes. In particular, phase-dependent effects in multiphoton ionization and harmonic generation [4–10] and the phase control of molecular reactions [11–13] have been investigated. Moreover, the phase-dependent modulation of the line-shape of autoionizing resonances [14] and the phase control of resonance scattering in a bichromatic field [15] have been considered very recently.

Amending our earlier work [16], we reconsider in the present paper potential scattering of electrons in a powerful, bichromatic laser field. The two field components will have the same direction ϵ of linear polarization and will have the frequencies $r\omega$ and $s\omega$, respectively, where r and s are small, positive integers. Both fields will be out of phase by an arbitrary angle φ . As we shall show below, for the ratios $r : s = 1 : 2$, $1 : 3$ and $2 : 3$ the cross sections for induced and inverse bremsstrahlung become strongly phase-dependent, whereas for increasing values of s the phase dependence gradually disappears. These findings are in accord with the results of corresponding calculations of multiphoton ionization by Potvliege and Smith [7]. In the case of free–free transitions, these general features of the phase dependence essentially rest on the particular properties of the generalized Bessel functions,

which determine the electron scattering spectrum. As in our previous papers [16], we shall describe the atomic field, in the low-frequency limit, by a static potential $U(x)$ and we shall treat the laser-assisted scattering of electrons by $U(x)$ in the first Born approximation. As has been shown for scattering in a single laser field by Trombetta and Ferrante [17] and by Trombetta [18], the second Born term only yields considerable contributions to the nonlinear cross sections in close-to-forward scattering directions, whereas for larger scattering angles the results of their calculations agree with the predictions of the Kroll–Watson formula [19]. Unfortunately, in the case of a bichromatic laser field, no Kroll–Watson-type scattering formula can be derived in a satisfactory manner [20].

2. Theory

We describe the bichromatic laser field of linear polarization ε and frequencies $r\omega$ and $s\omega$ by a plane wave in the dipole approximation whose vector potential reads

$$A(t) = \varepsilon[(cF_r/r\omega)\cos(r\omega t) + (cF_s/s\omega)\cos(s\omega t + \varphi)] \quad (1)$$

where F_r and F_s are the two field strengths, respectively. The Schrödinger equation for an electron moving in this field has the well known Gordon–Volkov solutions [21, 22]

$$\Psi_p(x, t) = V^{-1/2} \exp[-(i/\hbar)(Et - p \cdot x)] \exp\{i(p \cdot \varepsilon/\hbar)[\alpha_r \sin(r\omega t) + \alpha_s \sin(s\omega t + \varphi)]\} \quad (2)$$

for an electron of initial momentum p and energy $E = p^2/2m$. Adiabatic decoupling from the field has been assumed for $t \rightarrow -\infty$ and the A^2 -part of the electromagnetic interaction has been dropped since it yields no contribution to scattering. For scattering from an initial state Ψ_p , to a final state Ψ_{p_f} of the form (2), the transition matrix element in the first Born approximation reads

$$T_{fi} = -(i/\hbar) \int_{-\infty}^{+\infty} dt \int d^3x \Psi_{p_f}^*(x, t) U(x) \Psi_p(x, t). \quad (3)$$

Employing the generating function of generalized Bessel functions

$$\exp[ia_r \sin(r\omega t) + ia_s \sin(s\omega t + \varphi)] = \sum_{n=-\infty}^{+\infty} B_n(a_r, a_s; \varphi) \exp(in\omega t) \quad (4)$$

the matrix element (3) can be decomposed into an infinite, incoherent sum of elements T_n , describing induced ($n > 0$) and inverse ($n < 0$) bremsstrahlung of the nonlinear order $|n|$,

$$T_n = -2\pi i V^{-1} \delta(E_f - E_i - n\hbar\omega) U(Q) B_n(a_r, a_s; \varphi) \quad (5)$$

where V is the normalization volume and $U(Q)$ the Fourier transform of the scattering potential as a function of the momentum transfer $Q = p_f - p_i$. The arguments a_r and a_s of the generalized Bessel functions B_n follow from (2) and (3) to be given by

$$a_r = -\alpha_r Q \cdot \varepsilon/\hbar \simeq -\alpha_r K_i(n_f - n_i) \cdot \varepsilon \quad (6)$$

$$a_s = -\alpha_s Q \cdot \varepsilon/\hbar \simeq -\alpha_s K_i(n_f - n_i) \cdot \varepsilon. \quad (7)$$

In equations (6) and (7) we made the low-frequency approximation $|p_f| \simeq |p_i| = \hbar K_i$ and n_i and n_f are the unit vectors of the directions of propagation of the ingoing and scattered electrons, respectively. The parameters α_r and α_s are the classical amplitudes of the electron oscillations in the two fields, namely

$$\alpha_r = \mu_r c/r\omega \quad \mu_r = eF_r/mcr\omega \quad (8)$$

$$\alpha_s = \mu_s c / s \omega \quad \mu_s = e F_s / m c s \omega \tag{9}$$

where μ_r and μ_s are the corresponding intensity parameters of the two fields.

From equation (5) we easily obtain the various differential scattering cross sections of free-free transitions normalized with respect to the Born cross section of elastic scattering in the low-frequency limit

$$d\sigma_n / d\sigma_B = |B_n(a_r, a_s; \varphi)|^2 \tag{10}$$

which is our generalization for a bichromatic field of the Bunkin-Fedorov formula [1]. The explicit representations of the generalized Bessel functions B_n in terms of ordinary Bessel functions J_λ depend on the specific values of the parameters r and s .

(i) If $r = 1$ and $s = 2, 3, \dots$ we find

$$B_n(a_1, a_s; \varphi) = \sum_{\lambda=-\infty}^{+\infty} J_{n-\lambda s}(a_1) J_\lambda(a_s) \exp(i\lambda\varphi) \tag{11}$$

which is 2π periodic in φ . Moreover, we get the symmetry properties in n : for even $s = 2, 4, \dots$

$$B_{-n}(a_1, a_s; \varphi) = (-1)^n B_n^*(a_1, a_s; \varphi + \pi) \tag{12}$$

and for odd $s = 3, 5, \dots$

$$B_{-n}(a_1, a_s; \varphi) = (-1)^n B_n^*(a_1, a_s; \varphi). \tag{13}$$

Hence, in the first case, the spectrum (10) of scattered electrons will be symmetric with respect to $n > 0$ and $n < 0$ for particular values of φ , like $\varphi = \pm\pi/2, \pm3\pi/2$, and will be asymmetric for other φ , as $\varphi = 0, \pm\pi$, where these latter two spectra show, however, mirror symmetry with respect to $n = 0$ (see, for example, our first paper of [16] for illustrative figures). In the second case, on the other hand, the electron spectrum (10) will always be symmetric for $n > 0$ and $n < 0$ irrespective of the specific value of the phase φ (see, for example, the figures for $r : s = 1 : 3$ in our second paper of [16]).

(ii) If $r = 2$ and $s = 3, 4, \dots$ we have to distinguish the following two cases in the representations of B_n :

For even $s = 2\sigma, \sigma = 2, 3, \dots$, we obtain the same formulae for B_n as in (i) and we get the same spectra, if only ω is replaced by 2ω . Therefore, this case is of little interest.

If, however, s is odd, i.e. $s = 2\sigma + 1, \sigma = 1, 2, \dots$, we find for B_n by considering $n = 2m$ and $n = 2m + 1, m = 0, \pm 1, \pm 2, \dots$, separately

$$B_{2m}(a_2, a_s; \varphi) = \sum_{\lambda=-\infty}^{+\infty} J_{m-\lambda s}(a_2) J_{2\lambda}(a_s) \exp(i2\lambda\varphi) \tag{14}$$

$$B_{2m+1}(a_2, a_s; \varphi) = \sum_{\lambda=-\infty}^{+\infty} J_{m-\lambda s-\sigma}(a_2) J_{2\lambda+1}(a_s) \exp[i(2\lambda + 1)\varphi]. \tag{15}$$

Replacing φ in (14) and (15) by $\varphi + \pi$, we get $B_{2m}(\varphi + \pi) = B_{2m}(\varphi)$ and $B_{2m+1}(\varphi + \pi) = -B_{2m}(\varphi)$. Hence, the spectrum $|B_n(\varphi)|^2$ will be π -periodic. Moreover, we obtain the following symmetry relations:

$$B_{-2m}(a_2, a_s; \varphi) = (-1)^m B_{2m}^*(a_2, a_s; \varphi + \pi/2) \tag{16}$$

$$B_{-(2m+1)}(a_2, a_s; \varphi) = i(-1)^{m+\sigma} B_{2(m+2\sigma)+1}^*(a_2, a_s; \varphi + \pi/2) \tag{17}$$

and therefore the spectrum $|B_n(a_2, a_s; \varphi)|^2$ will always be asymmetric in relation to $n \geq 0$, if φ is varied from 0 to π , as will be shown in our examples below.

Considering in cases (i) and (ii) increasing values of s , ordinary Bessel functions J_λ of rather different orders will be coupled in the expressions (11), (14), (15) and then, apparently, one particular term dominates the sums. Hence, with growing s the phase dependence of the nonlinear cross sections gradually washes out. This same behaviour of the data has been found in multiphoton ionization and molecular reactions in a phase-dependent bichromatic laser field and will be recognized for the present process in the numerical examples presented in the following section. However, this general behaviour only holds true as long as both field components have about the same intensity. If, on the other hand, the intensity of the harmonic field component is increased considerably, phase-dependent effects will again show up even for higher values of s . For all cases considered, we obtain in the low-frequency limit, on which we are concentrating, the sum rule $\sum_n |B_n(a_r, a_s; \varphi)|^2 = 1$, as can be easily demonstrated by means of the generating function (4).

3. Numerical example

As in our previous work [16], we base our calculations on the experimental setup of Weingartshofer *et al* [23]. For the kinematics and parameters chosen in this experiment, our cross section formula (10) should reasonably well apply. In our case of a bichromatic

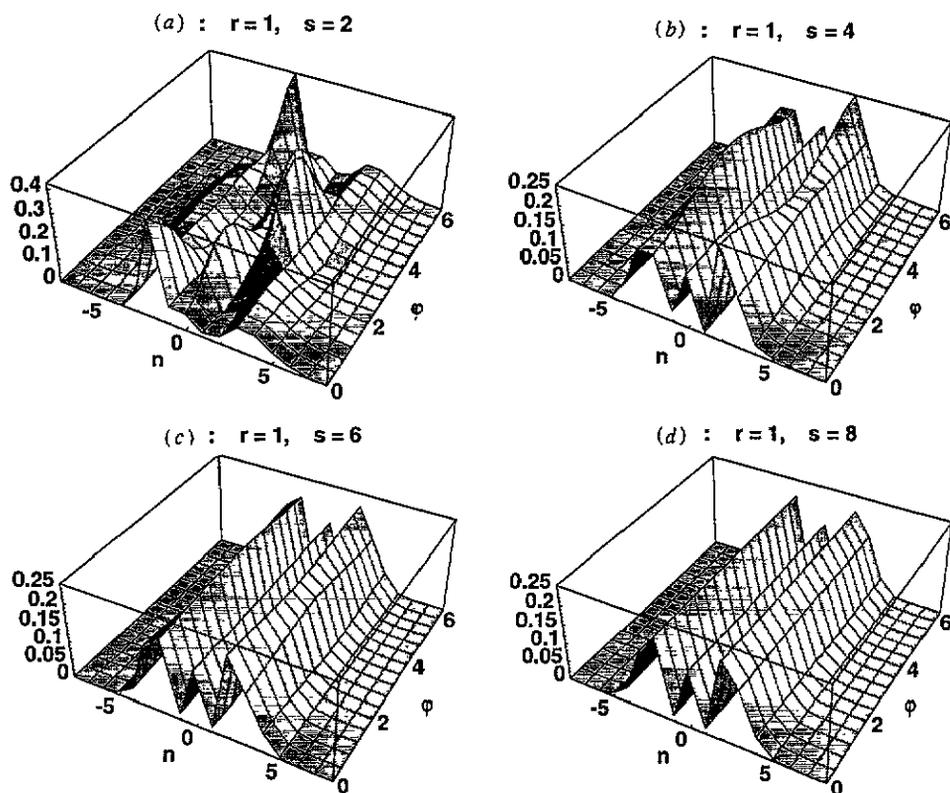


Figure 1. In this figure we show the cross section data of free-free transitions in a phase-dependent bichromatic field, evaluated for an intensity $I = 4 \times 10^7 \text{ W cm}^{-2}$, and the following ratios $r : s = 1 : 2\sigma$: (a) $\sigma = 1$; (b) $\sigma = 2$; (c) $\sigma = 3$ and (d) $\sigma = 4$. The symmetry of the spectra for $\varphi = \pi/2$ and $3\pi/2$ as well as the asymmetry for $\varphi = 0, \pi$ and 2π is clearly visible for $\sigma = 1, 2$. $|B_n|^2$ is plotted as a function of $n\hbar\omega$ and φ .

field, we take a CO₂-laser source and one of its harmonics. Both fields are assumed to have about equal intensity between $I = 4 \times 10^7$ and 3.6×10^8 W cm⁻² and the initial electron energy will be $E_i = 10$ eV. For the evaluation of the intensity parameters (8) and (9) we use $F_r = F_s = F$ evaluated from the above intensities, such that $\alpha_r = \mu c/r^2\omega = \alpha/r^2$ and $\alpha_s = \mu c/s^2\omega = \alpha/s^2$, where $\mu = eF/mc\omega$, and therefore, according to (6) and (7), $a_r = a/r^2$, $a_s = a/s^2$ with $a = -\alpha K_i(\mathbf{n}_f - \mathbf{n}_i) \cdot \boldsymbol{\varepsilon} = \alpha K_i [\cos \Psi_0 - \cos(\Psi_0 + \Theta)]$. Here $\boldsymbol{\varepsilon} \cdot \mathbf{n}_i = \cos \Psi_0$ and $\mathbf{n}_f \cdot \mathbf{n}_i = \cos \Theta$ define the angles Ψ_0 and the scattering angle Θ which, in accord with the work of Weingartshofer *et al* [23], are chosen as $\Psi_0 = 38^\circ$ and $\Theta = 155^\circ$.

For the ratios $r : s = 1 : 2\sigma$, $\sigma = 1-4$, we evaluated with the above parameter values and the laser intensity $I = 4 \times 10^7$ W cm⁻² from (10) and (11) the differential cross sections of free-free transitions depicted in figure 1, where (a) refers to $\sigma = 1$, (b) to $\sigma = 2$, (c) to $\sigma = 3$ and (d) to $\sigma = 4$. In these figures, the absorbed ($n > 0$) or emitted ($n < 0$) quanta are given by $n\hbar\omega$ and the phase φ is measured in units of $\pi/3$. As we can see, the phase dependence of the scattering pattern gradually fades away with increasing σ , where the main effects are found for $r : s = 1 : 2$, as discussed in detail in our earlier work [16]. Moreover, we recognize the 2π -periodicity and the fact that for $\varphi = 0, \pi, 2\pi$ the spectra are asymmetric with respect to $n = 0$ for $n \geq 0$ and that for $\varphi = \pi/2, 3\pi/2$ the spectra are symmetric, as has been analysed in section 2.

In figure 2 we reconsider the ratio $r : s = 1 : 2$ and present for a selected number of phases $|B_n|^2$ as a function of $n\hbar\omega$. In particular, we have taken in (a) $\varphi = \pi/4$, in (b) $\varphi = \pi/2$, in (c) $\varphi = 3\pi/4$ and in (d) $\varphi = \pi$. As indicated before, for $\varphi = \pi/2$ the

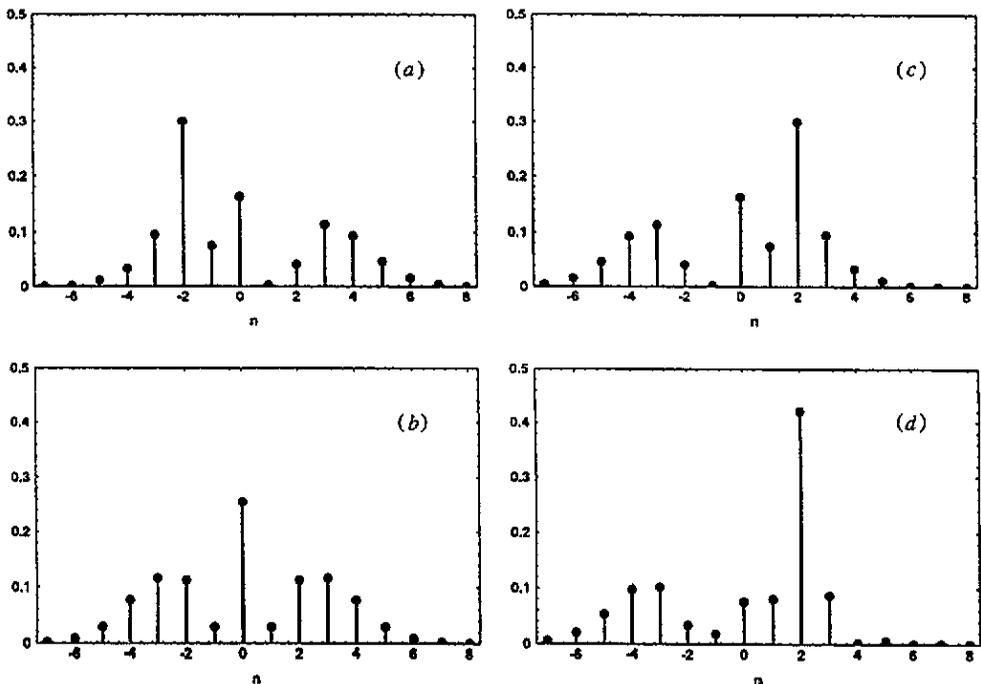


Figure 2. This figure shows the spectra $|B_n|^2$ as a function of $n\hbar\omega$ for the following set of phases: (a) $\varphi = \pi/4$, (b) $\varphi = \pi/2$, (c) $\varphi = 3\pi/4$ and (d) $\varphi = \pi$. Observe the symmetry for $\varphi = \pi/2$, the asymmetry for $\varphi = \pi$ and the mirror symmetry for $\varphi = \pi/4$ versus $\varphi = 3\pi/4$.

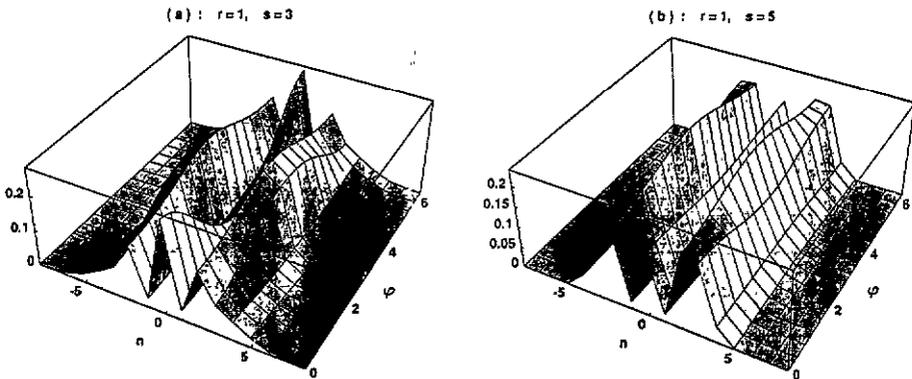


Figure 3. Here we show the corresponding data for $I = 4 \times 10^7 \text{ W cm}^{-2}$ and $r : s = 1 : (2\sigma + 1)$ with (a) $\sigma = 1$ and (b) $\sigma = 2$. These spectra are all symmetric irrespective of the value of φ and are 2π -periodic as in figure 1. For higher values of σ the φ dependence rapidly decreases.

spectrum is symmetric and for $\varphi = \pi$ it is asymmetric with respect to $n > 0$ versus $n < 0$. On the other hand, the spectra for $\varphi = \pi/4$ and $\varphi = 3\pi/4$ are mirror-symmetric with respect to $n = 0$.

Similarly, for the ratios $r : s = 1 : (2\sigma + 1)$, $\sigma = 1, 2$, and for the same intensity, we find the spectra of figure 3 with (a) $\sigma = 1$ and (b) $\sigma = 2$. Here, as predicted in section 2, all spectra are symmetric, irrespective of the value of the phase φ and the dominant phase dependences are found for the ratio $r : s = 1 : 3$, as analysed in detail previously [16]. Again, for higher values of σ the phase dependence gradually disappears.

In order to show the symmetry of the spectra for $r : s = 1 : 3$ more clearly, we show in figure 4, $|B_n|^2$ as a function of $n\hbar\omega$ for the following selected phases: (a) $\varphi = 0$, (b) $\varphi = \pi/3$, (c) $\varphi = \pi/2$ and (d) $\varphi = \pi$. Here we have the particular feature that for certain phases, like $\varphi = \pi$, a considerable number of nonlinear cross sections are strongly suppressed, while simultaneously others are very much enhanced.

In figure 5 we show the corresponding cross section data for the ratio $r : s = 2 : 3$ for the somewhat higher laser field intensity $I = 3.6 \times 10^8 \text{ W cm}^{-2}$ in order to get sufficiently large values of $|B_n|^2$. We observe that here the period in the phase φ is π , as we have shown in section 2. The maximum phase-dependent effects occur for $r : s = 2 : 3$ and also here these effects gradually disappear for increasing s . Moreover, we see that in the present case, the spectra show no symmetries with respect to $n = 0$ for $n \gtrsim 0$ (see section 2). The present case has not yet been discussed by us.

The reason why with increasing s the phase dependence of the cross section data, discussed before, gradually disappears can be easily explained. We assumed that both fields have about equal intensity. As a consequence we found $a_r = a/r^2$ and $a_s = a/s^2$. Keeping r small, 1 or 2, but increasing s , immediately shows that a_s rather rapidly decreases and therefore in the formulae (11), (14) and (15) for $B_n(a_r, a_s; \varphi)$ the term with $\lambda = 0$ will begin to dominate and so the phase dependence gradually drops out. If, therefore, we want to have the phase dependence show up for higher values of s , we would have to choose the intensity of the second field $I_s = Is^4$, in order to compensate for the s dependence of a_s . This, however, will presently be impossible to achieve experimentally and, moreover, at these higher intensities tunnelling ionization will set in as a competing process. Physically speaking, the coupling of higher-order intermediate channels requires higher laser powers. One can expect that the same situation will hold true for multiphoton ionization and

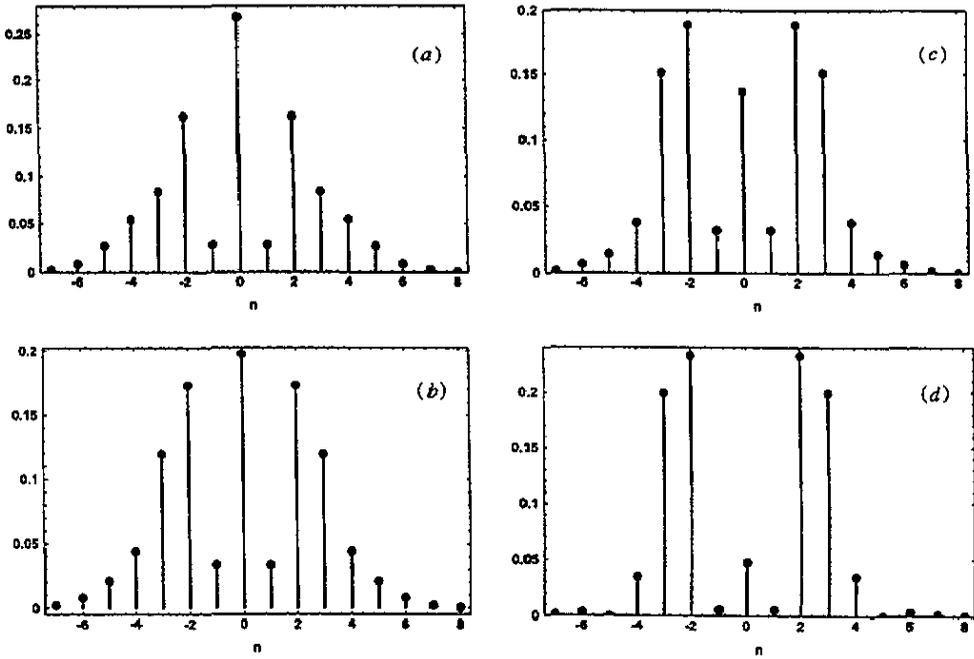


Figure 4. Here we present $|B_n|^2$ as a function of $n\hbar\omega$ for the following phases: (a) $\varphi = 0$, (b) $\varphi = \pi/3$, (c) $\varphi = \pi/2$ and (d) $\varphi = \pi$. Observe, in particular, the peak suppressions and peak enhancements for $\varphi = \pi$.

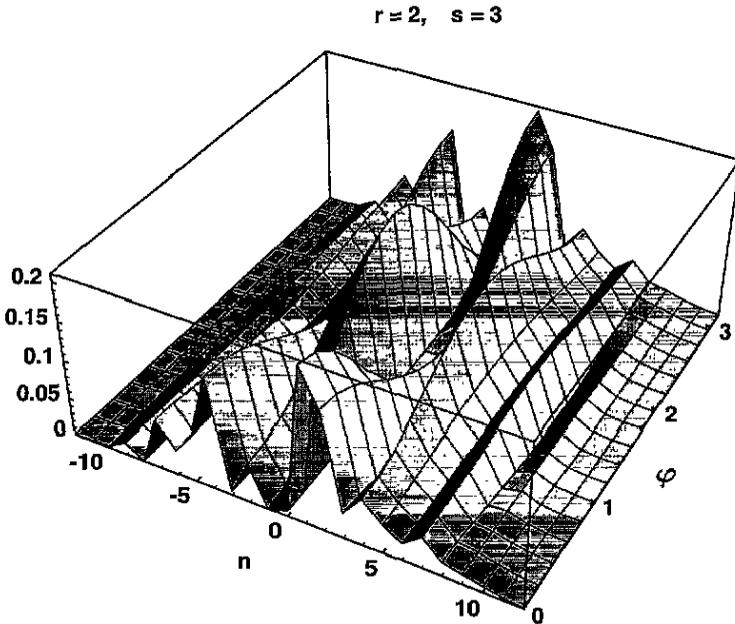


Figure 5. Now the intensity is $I = 3.6 \times 10^8 \text{ W cm}^{-2}$ and $r : s = 2 : 3$. Here the spectrum is asymmetric independent of the choice of φ and is π -periodic in φ . $|B_n|^2$ is again plotted as a function of $n\hbar\omega$ and φ . Higher values of s yield a rapidly decreasing dependence on φ .

molecular reactions in a bichromatic, phase-dependent laser field. Considering multiphoton ionization in the Keldysh–Faisal–Reiss approximation [24–26], the above conclusion is rather obvious, since, by neglecting the A^2 -part of the electromagnetic interaction, the spectrum of the ionized electrons will also be determined by generalized Bessel functions of the form (11), (14) and (15).

In order to demonstrate that with considerable increase of the intensity of the harmonic field component, phase-dependent effects in the nonlinear electron scattering spectra will reappear, we consider the case $r : s = 1 : 2\sigma$, $\sigma = 1-4$, of figure 1 but now choose for the intensity of the harmonic field component $I_s = Is^4$ in which case the arguments a_1 and a_s , in the generalized Bessel functions (11), will become equal, where $a_1 = a$ is taken to have the same value as in figure 1. The results of the corresponding evaluation of $|B_n(a, a; \varphi)|^2$ are shown in figure 6. Here (a) refers to $s = 2$, (b) to $s = 4$, (c) to $s = 6$ and (d) to $s = 8$. As we can see, with increasing s the phase effects are still appreciable and extend to higher nonlinear orders if compared with the corresponding data of figure 1, and, moreover, the values of the nonlinear cross sections gradually decrease on the average at the same time. This indicates that the coupling of higher-order intermediate channels in the sum over λ in (11) is less favoured even for higher intensities of the harmonic field component.

In the present work, the laser field has been described by a bichromatic, phase-dependent

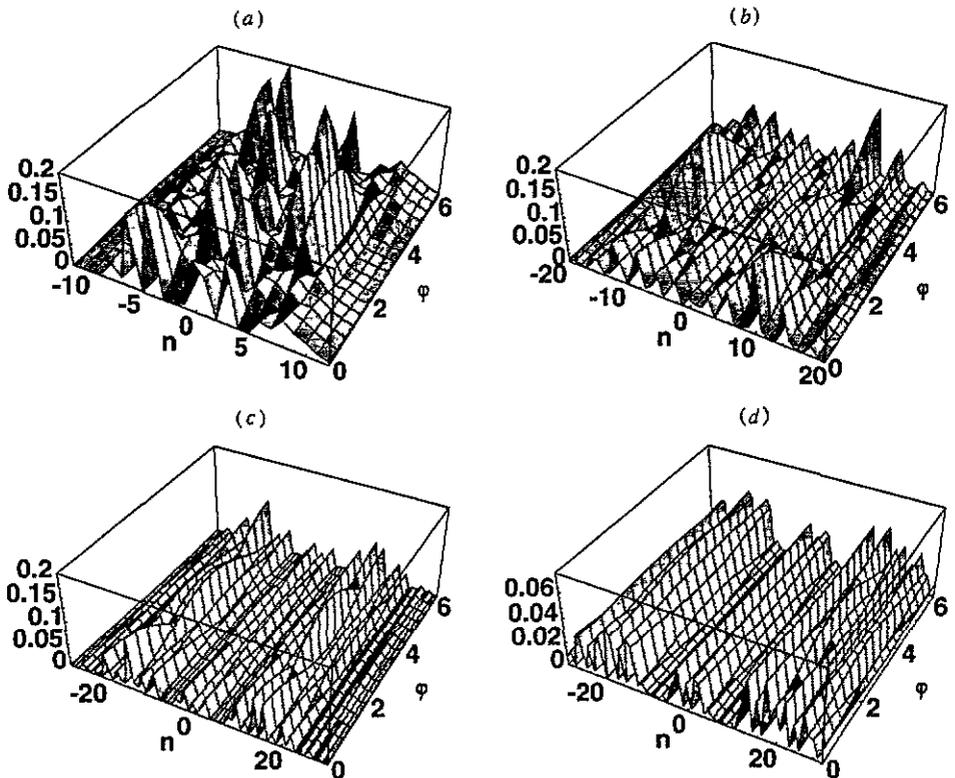


Figure 6. Here we present cross section data for $I = 4 \times 10^7 \text{ W cm}^{-2}$ with $r : s = 1 : 2\sigma$ for (a) $\sigma = 1$, (b) $\sigma = 2$, (c) $\sigma = 3$ and (d) $\sigma = 4$ as in figure 1, however, the second field has intensity $I_s = Is^4$ such that the arguments in the generalized Bessel functions (11) are equal, namely, $a_1 = a_s = a$.

and linearly polarized plane wave in the dipole-approximation, corresponding roughly to a single-mode laser operation and one of its harmonics. However, in the experiments of Weingartshofer *et al* [23], a multimode laser was employed. In later experiments of the Weingartshofer group [27, 28] it has been demonstrated that their cross section data are located between the data evaluated theoretically for a monochromatic plane wave field and for a chaotic field, for which the corresponding cross section formula has been derived by Zoller [29] and by Daniele and Ferrante [30]. If, therefore, we contemplate an experiment on free-free transitions in a multimode bichromatic laser field, depending on a phase φ , theoretical cross section data for a chaotic field with frequencies $r\omega$ and $s\omega$ should also be available. While the derivation of Zoller's expression for $d\sigma_n/d\Omega$ is rather simple [29, 30], we have not yet succeeded, to derive an appropriate formula for a bichromatic field, since it appears to require the evaluation of an integral of the form [30]

$$\varepsilon_0^{-2} \int_0^{\infty} d\xi |B_n(\lambda_r \sqrt{\xi}, \lambda_s \sqrt{\xi}; \varphi)|^2 \exp(-\xi/\varepsilon_0^2) \quad (18)$$

where $\varepsilon_0^2 = \langle F^2 \rangle$ is the average value of the field strength F of the fundamental laser field and $\lambda_r = \alpha_r/F_r$; $\lambda_s = \alpha_s/F_s$ are defined by (8) and (9), respectively. In our expression (18), it is assumed that both fields have the same field strength $F_r = F_s = F$ with the same chaotic fluctuations. While equation (18) can be easily evaluated analytically for a single field with $B_n \rightarrow J_n$, it looks hopeless to do so for the present case and a numerical integration will be rather cumbersome.

4. Summary and conclusions

In the present paper, we have reconsidered the coherent phase control of free-free transitions in a bichromatic laser field. In the low-frequency limit, the atomic target has been described by a static potential $U(x)$ and the electron scattering process has been treated in the first Born approximation in $U(x)$. The complications, arising from the higher-order Born terms have also been mentioned. The bichromatic laser field has been represented by a plane wave in the dipole approximation having linear polarization ε and the two frequencies $r\omega$ and $s\omega$ ($r = 1, 2; s = 2, 3, \dots$), where both components are out of phase by an angle φ . In section 2, the differential cross sections of induced and inverse bremsstrahlung have been evaluated for this field configuration and the arising symmetries and asymmetries of the electron spectra as a function of the phase φ have been discussed in 2(i) and 2(ii). In section 3, a numerical example was given based on the parameter values of an experiment by Weingartshofer *et al* [23], assuming both field components to have the same field strength F , evaluated from two laser field intensities $I = 4 \times 10^7$ or 3.6×10^8 W cm⁻² of a CO₂ laser source. The calculated cross section data are depicted in figures 1–5. As one can see, considerable phase-dependent effects can be expected for the ratios $r : s = 1 : 2, 1 : 3$ and $2 : 3$, whereas with increasing s the phase dependence of the data gradually disappears. This behaviour is simply a consequence of our choice of equal intensities of the two field components. As discussed in the same section, if the intensity of the harmonic field was taken as $I_s = Is^4$, then, also for larger s , phase effects would show up, but these conditions are beyond the limits of present experiments. Examples, referring to this latter case, are shown in figures 6(a)–(d). Finally, we indicated the complications which arise in evaluating the cross sections of free-free transitions in a chaotic bichromatic field. Such data would be useful for comparison with the possible experimental results for phase effects in a bichromatic, multimode CO₂ laser field.

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