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Effect of target dressing in free–free transitions in a bichromatic laser field

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Abstract. The influence of laser dressing of the atomic target on free–free transitions in a bichromatic laser field of variable relative phase is investigated. For simplicity, hydrogen is taken as target atom and only the elastic, non-resonant case is considered. Exchange effects are neglected and the scattering process is treated to the lowest-order Born approximation. It is found that at very small scattering angles the dressing effects have some influence on the phase dependence of the scattering cross sections in a bichromatic field.

1. Introduction

Free–free transitions in a laser field have been considered by many authors. Surveys can be found in the books by Mittleman [1] and Faisal [2] and in the review by Mason [3].

Recently, it has become of interest to investigate multiphoton processes in bichromatic laser fields. First, it has been suggested that the rates of laser-assisted and laser-induced molecular reactions can be considerably enhanced or modified if the process takes place in a laser field of two components of frequencies ω and 2ω or ω and 3ω, where both components are out of phase by an angle φ. It was found that for particular values of φ the molecular reaction rates can be suppressed or enhanced and the angular distributions modified. This effect was coined coherent phase control. An overview of such investigations can be found in the work of Giusti-Suzor *et al* [4] and in a series of recent papers with references to earlier work [5–8]. These investigations were soon followed by similar considerations of the coherent phase control of multiphoton ionization in the experimental work of Muller *et al* [9], Chen and Eliott [10] and Yin *et al* [11], which initiated a considerable body of theoretical papers [12–23]. Similarly, phase-dependent effects in autoionization in a bichromatic field were considered [24, 25] and the most recent work has been on the coherent phase control of higher harmonic generation in laser fields of two frequency components [26–31].

Stimulated by such investigations, a few years ago we began to consider free–free transitions in a bichromatic laser field [32, 33] and we found that the variation of the relative phase φ considerably modifies the nonlinear scattering spectrum. It was also shown that the phase-dependent effects are particularly pronounced if the ratio of the two frequencies $r : s = 1{:}2$, 1:3 and 2:3 [34]. In order to go beyond the first-order Born approximation with respect to the scattering potential, generalizations of the Kroll–Watson scattering formula for a bichromatic laser field were derived [35–37]. Finally, these three-dimensional investigations were amended by one-dimensional model calculations to consider off-shell effects in a bichromatic field [38] and to study resonance phenomena [39, 40].

All the above considerations of free–free transitions in a bichromatic laser field were performed by assuming that the atomic target may be described by a static potential. This assumption is certainly justified as long as low laser frequencies are contemplated, as is true for the series of experiments performed by Weingartshofer and co-workers [41–44]. If, however, the laser frequency is increased, it will become necessary to consider the influence of the dressing of the target atom by the radiation field. For a single laser frequency, such investigations were performed in the early work by Gersten and Mittleman [45] and by Mittleman [46–49], and at about the same time by Zon [50] and Beilin and Zon [51]. More recently, very detailed calculations were presented by Joachain and co-workers [52–55]. It is the purpose of the present paper to extend such investigations to the problem of elastic electron–atom scattering in a bichromatic laser field. We shall assume that the field has the frequency components ω and 2ω of the same linear polarization $\vec{\varepsilon}$ and the field will be treated in the dipole approximation. Moreover, we consider the simplest case of electron–hydrogen scattering and treat the laser–atom interaction by lowest order of perturbation theory neglecting, for simplicity, exchange effects and resonance phenomena. In this case it is well known from the work mentioned above that the laser–atom interaction can essentially be described by a polarization potential which is induced by the radiation field, the effect of which on the nonlinear scattering cross sections becomes dominant at small momentum transfer. As we shall show below, this polarization potential has only an appreciable influence on the coherent phase control of electron–atom scattering in a bichromatic radiation field under very extreme parameter conditions. In our derivation of the essential scattering formula we shall closely follow the work of Zon [50] and Byron *et al* [54].

2. Theory

The calculations required to derive the corresponding scattering cross section formula are rather straightforward. We start with the Schrödinger equation for a single atomic electron embedded in the bichromatic laser field

$$[-(\hbar^2/2m)\Delta' + V(\vec{x}') + eF_0\vec{x}'\cdot\vec{\varepsilon}(\sin\omega t + \sin(2\omega t + \varphi))]\psi(\vec{x}', t) = \mathrm{i}\hbar\partial_t\psi(\vec{x}', t) \tag{1}$$

where it is assumed that both field components have the same amplitude of field strength F_0 and polarization $\vec{\varepsilon}$ in which case we expect a particularly large effect. To solve (1), we make an ansatz in terms of the unperturbed eigenstates $\psi_n(\vec{x}', t) = |n\rangle\exp(-\mathrm{i}E_n t/\hbar)$

$$\psi(\vec{x}', t) = |0\rangle\exp(-\mathrm{i}E_0 t/\hbar) + \sum_n{}' c_n(t)|n\rangle\exp(-\mathrm{i}E_n t/\hbar) \tag{2}$$

to find by lowest-order time-dependent perturbation theory with adiabatic decoupling of the radiation field at $t \to -\infty$

$$c_n(t) = -\frac{eF_0}{2i}\langle n|\vec{\varepsilon}\cdot\vec{x}'|0\rangle\left\{\frac{\exp[\mathrm{i}(E_n - E_0 + \hbar\omega)t/\hbar]}{E_n - E_0 + \hbar\omega + \mathrm{i}\varepsilon} - \frac{\exp[\mathrm{i}(E_n - E_0 - \hbar\omega)t/\hbar]}{E_n - E_0 - \hbar\omega + \mathrm{i}\varepsilon}\right.$$
$$\left. + \frac{\exp[\mathrm{i}(E_n - E_0 + 2\hbar\omega)t/\hbar + \mathrm{i}\varphi]}{E_n - E_0 + 2\hbar\omega + \mathrm{i}\varepsilon} - \frac{\exp[\mathrm{i}(E_n - E_0 - 2\hbar\omega)t/\hbar - \mathrm{i}\varphi]}{E_n - E_0 - 2\hbar\omega + \mathrm{i}\varepsilon}\right\} \tag{3}$$

which yields with (2) as the expression for the laser-dressed ground state

$$\psi_{\mathrm{g}}(\vec{x}',t)=\Bigg\{|0\rangle - eF_0\sum_n{}'|n\rangle\langle n|\vec{\varepsilon}\cdot\vec{x}'|0\rangle\Bigg[\frac{(E_n-E_0)\sin\omega t+\mathrm{i}\hbar\omega\cos\omega t}{(E_n-E_0)^2-(\hbar\omega)^2} + \frac{(E_n-E_0)\sin(2\omega t+\varphi)+\mathrm{i}2\hbar\omega\cos(2\omega t+\varphi)}{(E_n-E_0)^2-(2\hbar\omega)^2}\Bigg]\Bigg\}\exp(-\mathrm{i}E_0t/\hbar). \qquad (4)$$

The effective potential for scattering of electrons by a laser-dressed hydrogen atom is then given by [56]

$$V_{\mathrm{eff}}(\vec{x},t)=-\frac{e^2}{|\vec{x}|}+e^2\int\frac{|\psi_{\mathrm{g}}(\vec{x}',t)|^2}{|\vec{x}-\vec{x}'|}\,\mathrm{d}^3x'. \qquad (5)$$

Using (4), we obtain from (5) up to first order in F_0

$$e^2\int\frac{|\psi_{\mathrm{g}}(\vec{x}',t)|^2}{|\vec{x}-\vec{x}'|}\,\mathrm{d}^3x' = e^2\int\frac{|\psi_0(\vec{x}',t)|^2}{|\vec{x}-\vec{x}'|}\,\mathrm{d}^3x' - \frac{e^3F_0}{|\vec{x}|^3}\sum_n{}'\Bigg\{(\langle 0|\vec{x}\cdot\vec{x}'|n\rangle\langle n|\vec{\varepsilon}\cdot\vec{x}'|0\rangle + \langle 0|\vec{\varepsilon}\cdot\vec{x}'|n\rangle\langle n|\vec{x}\cdot\vec{x}'|0\rangle)\Bigg[\frac{(E_n-E_0)\sin\omega t}{(E_n-E_0)^2-(\hbar\omega)^2}+\frac{(E_n-E_0)\sin(2\omega t+\varphi)}{(E_n-E_0)^2-(2\hbar\omega)^2}\Bigg] + \mathrm{i}(\langle 0|\vec{x}\cdot\vec{x}'|n\rangle\langle n|\vec{\varepsilon}\cdot\vec{x}'|0\rangle - \langle 0|\vec{\varepsilon}\cdot\vec{x}'|n\rangle\langle n|\vec{x}\cdot\vec{x}'|0\rangle)\Bigg[\frac{\hbar\omega\cos\omega t}{(E_n-E_0)^2-(\hbar\omega)^2} + \frac{2\hbar\omega\cos(2\omega t+\varphi)}{(E_n-E_0)^2-(2\hbar\omega)^2}\Bigg]\Bigg\}. \qquad (6)$$

To simplify our calculations, we now introduce the closure approximation by replacing $E_n - E_0$ in the square brackets by some average value $\bar{E}$ and by performing the summation over n, using $\sum_n |n\rangle\langle n| = 1$. Thus we obtain from (6) by means of a Fourier decomposition of $|\vec{x}-\vec{x}'|^{-1}$

$$e^2\int\frac{|\psi_{\mathrm{g}}(\vec{x}',t)|^2}{|\vec{x}-\vec{x}'|}\,\mathrm{d}^3x' = (2\pi\hbar)^{-3}\Bigg[e^2\int V(\vec{q})F(\vec{q})\exp(\mathrm{i}\vec{q}\cdot\vec{x}/\hbar)\,\mathrm{d}^3q + \alpha_{\mathrm{s}}a_0^3eF_0\vec{\varepsilon}\cdot\nabla\int V(\vec{q})\exp(\mathrm{i}\vec{q}\cdot\vec{x}/\hbar)\,\mathrm{d}^3q\Bigg[\frac{\sin\omega t}{1-(\hbar\omega/\bar{E})^2}+\frac{\sin(2\omega t+\varphi)}{1-(2\hbar\omega/\bar{E})^2}\Bigg]\Bigg] \qquad (7)$$

where $V(\vec{q})$ is the Fourier transform of the atomic potential and $F(\vec{q})$ the atomic form factor. α_{s} is the static polarizability of hydrogen in its ground state in units of the Bohr radius a_0. Using (7), the effective scattering potential of (5) becomes

$$V_{\mathrm{eff}}(\vec{x},t) = -\frac{e^2}{(2\pi\hbar)^3}\int V(\vec{q})\exp(\mathrm{i}\vec{q}\cdot\vec{x}/\hbar)\,\mathrm{d}^3q\,\Bigg\{1-F(\vec{q})-\mathrm{i}\frac{\alpha_{\mathrm{s}}a_0^3F_0}{e\hbar}\vec{\varepsilon}\cdot\vec{q}\Bigg[\frac{\sin\omega t}{1-(\hbar\omega/\bar{E})^2} + \frac{\sin(2\omega\hbar+\varphi)}{1-(2t\omega/\bar{E})^2}\Bigg]\Bigg\}. \qquad (8)$$

We treat the scattering of electrons by this potential in the bichromatic laser field to the first-order Born-approximation, since we shall consider fast electrons of about 100 eV kinetic energy. The laser-dressed in-going and out-going electron of momenta $\vec{p}$ and $\vec{p}'$, respectively, will be represented by corresponding Gordon–Volkov states [57]

$$\psi_{\vec{p}}(\vec{x},t) = V^{-1/2}\exp\{-\mathrm{i}[Et-\vec{p}(\vec{x}-\alpha_0\vec{\varepsilon}\sin\omega t-(\alpha_0/4)\vec{\varepsilon}\sin(2\omega t+\varphi)]/\hbar\} \qquad (9)$$

$$\psi^*_{\vec{p}'}(\vec{x},t) = V^{-1/2}\exp\{\mathrm{i}[E't-\vec{p}'(\vec{x}-\alpha_0\vec{\varepsilon}\sin\omega t-(\alpha_0/4)\vec{\varepsilon}\sin(2\omega t+\varphi))]/\hbar\} \qquad (10)$$

where $\alpha_0 = eF_0/mc\omega^2$ and the contribution of the A^2 part of the electromagnetic interaction has been neglected, since it drops out in the end. With (8)–(10) we find the transition matrix elements

$$T_{\mathrm{fi}} = -(\mathrm{i}/\hbar)\int \mathrm{d}^3x\,\mathrm{d}t\,\psi^*_{\vec{p}'}(\vec{x},t)V_{eff}(\vec{x},t)\psi_{\vec{p}}(\vec{x},t) = \sum_n T_n \tag{11}$$

where we obtain, after Fourier decomposition of (9) and (10) into Bessel functions,

$$\begin{aligned} T_n = 2\pi\mathrm{i}\frac{4\pi(e\hbar)^2}{VQ^2}\Bigg\{&[1-F(\vec{Q})]B_n(a;a/4;\varphi) + \frac{\alpha_s a_0^3 k^2}{2r_0}a\Bigg[\frac{1}{1-(\hbar\omega/\bar{E})^2}(B_{n+1}(a;a/4;\varphi) \\ &- B_{n-1}(a;a/4;\varphi)) + \frac{1}{1-(2\hbar\omega/\bar{E})^2}(B_{n+2}(a;a/4;\varphi)\exp(\mathrm{i}\varphi) \\ &- B_{n-2}(a;a/4;\varphi)\exp(-\mathrm{i}\varphi))\Bigg]\Bigg\}\delta(E'-E-n\hbar\omega). \end{aligned} \tag{12}$$

Here, the explicit form of the Fourier transform of the Coulomb potential $V(\vec{Q}) = 4\pi\hbar^2/Q^2$ has been introduced with $\vec{Q} = \vec{p} - \vec{p}'$ being the momentum transfer. $B_n(a;a/4;\varphi)$ are generalized Bessel functions which are defined by

$$B_n(a;a/4;\varphi) = \sum_{\lambda=-\infty}^{+\infty} J_{n-2\lambda}(a)J_\lambda(a/4)\exp(-\mathrm{i}\lambda\varphi) \tag{13}$$

where the argument of the ordinary Bessel functions is given by $a = \alpha_0\vec{Q}\cdot\vec{\varepsilon}/\hbar$. Moreover, we have used in (12), $r_0 = e^2/mc^2$ and $k = \omega/c$. Finally, we obtain from (12) by standard methods the nonlinear differential scattering cross sections with emission or absorption of n photons of the laser field

$$\begin{aligned} \frac{\mathrm{d}\sigma_n}{\mathrm{d}\Omega} = \frac{(2r_0)^2(mc)^4}{Q_n^4}\frac{p_n'}{p}\Bigg|\Bigg\{&[1-F(\vec{Q}_n)]B_n(a_n;a_n/4;\varphi) \\ &+ \frac{\alpha_s a_0^3 k^2}{2r_0}a_n\Bigg[\frac{1}{1-(\hbar\omega/\bar{E})^2}(B_{n+1}(a_n;a_n/4;\varphi) - B_{n-1}(a_n;a_n/4;\varphi)) \\ &+ \frac{1}{1-(2\hbar\omega/\bar{E})^2}(B_{n+2}(a_n;a_n/4;\varphi)\exp(\mathrm{i}\varphi) \\ &- B_{n-2}(a_n;a_n/4;\varphi)\exp(-\mathrm{i}\varphi))\Bigg]\Bigg\}\Bigg|^2. \end{aligned} \tag{14}$$

For hydrogen, the form factor can be easily evaluated to be

$$F(\vec{Q}_n) = \frac{1}{[1+(Q_n a_0/2\hbar)^2]^2} \qquad \vec{Q}_n = \vec{p} - \vec{p}_n' \qquad p_n = [2m(E+n\hbar\omega)]^{1/2}. \tag{15}$$

Expression (14) is the generalization for a bichromatic field of the corresponding formula presented by Zon [50]. In the following numerical example, we choose for the average transition energy $\bar{E}$ the value used in the work of Byron *et al* [54], $\bar{E} = (8/9)|E_0|$, where E_0 is the ground-state energy of hydrogen.

3. Numerical example

For our numerical example we have chosen nearly the same parameter values as in the paper by Byron *et al* [54]. However, we had to use a considerably higher intensity for the fundamental field component such that the second field would contribute appreciably. This

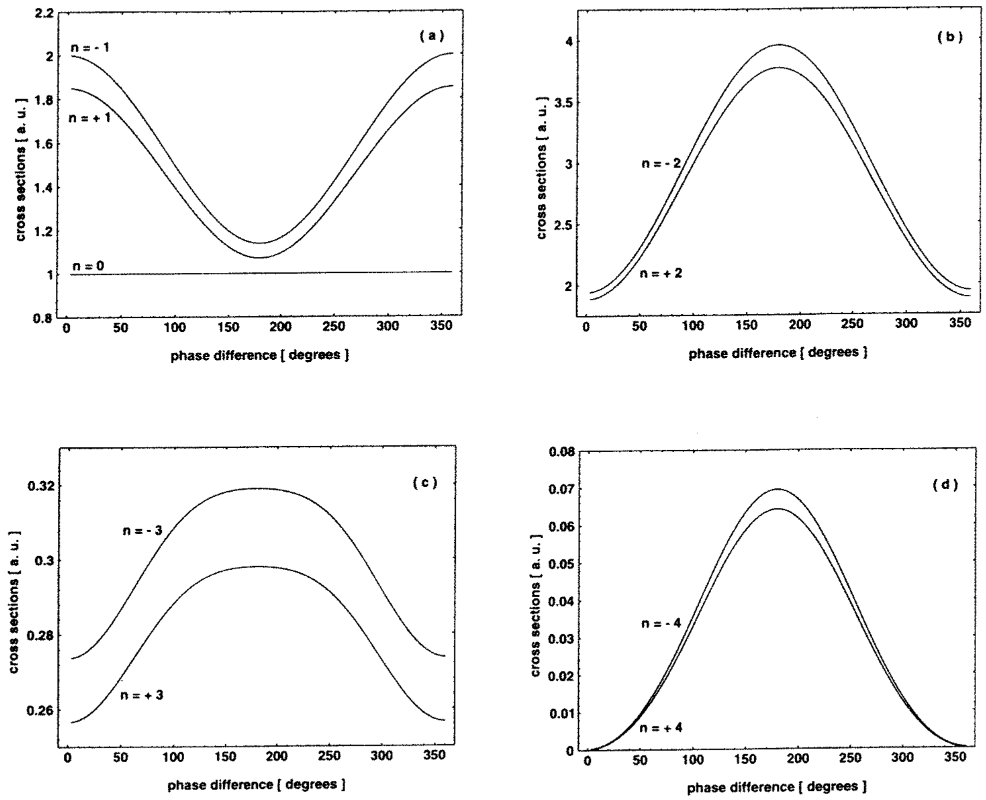

Figure 1. $a_0^{-2}\,\mathrm{d}\sigma_n/\mathrm{d}\Omega$ evaluated from (14) at $\theta = (2/3)^\circ$ for $0 \leqslant \varphi \leqslant 2\pi$ with (*a*) $n = 0, \pm 1$, (*b*) $n = \pm 2$, (*c*) $n = \pm 3$ and (*d*) $n = \pm 4$. Observe that there is no phase dependence for $n = 0$ and the largest cross sections are for $n = \pm 2$. There is a phase difference of π between the data for $n = \pm 1$ and those in (*b*)–(*d*). The dependence of Q_n on n yields a marked difference in the data for $n = \pm 3$.

automatically brought us into an intensity range where multiphoton ionization becomes a competing process. On the other hand, we had to choose the parameter values such that the momentum transfer Q_n is small in order to get an appreciable contribution from the laser-induced polarization potential describing the dressing effects and, on the other hand, we needed sufficiently large values of the argument a_n of the generalized Bessel functions (13) to be able to observe the coherent phase control of the nonlinear cross sections in a bichromatic laser field. Thus our necessary choice was as follows: laser intensity $I = 2 \times 10^{13}$ W cm^{-2}, laser frequency $\hbar\omega = 1.17$ eV, laser polarization $\vec{\varepsilon}$ parallel to the in-going electron momentum $\vec{p}$ and scattering angle θ in the range $0 \lesssim \theta \leqslant 45^\circ$. The exact static polarizability was taken from the tables of Radzig and Smirnov [59] to be $\alpha_s = 4.5a_0^3$ for the 1s ground state of hydrogen and the average excitation energy was taken to be $\bar{E} = (8/9)|E_0|$ [54]. Finally, the nonlinear scattering cross sections were expressed in units of a_0^2. As the following figures will show, the laser dressing of the target atom has only a considerable influence on the phase-dependent effects in electron–atom scattering in a bichromatic field for very small scattering angles $\theta \lesssim 1^\circ$, whereas for larger scattering angles there is hardly any difference whether the laser dressing of the target is included or

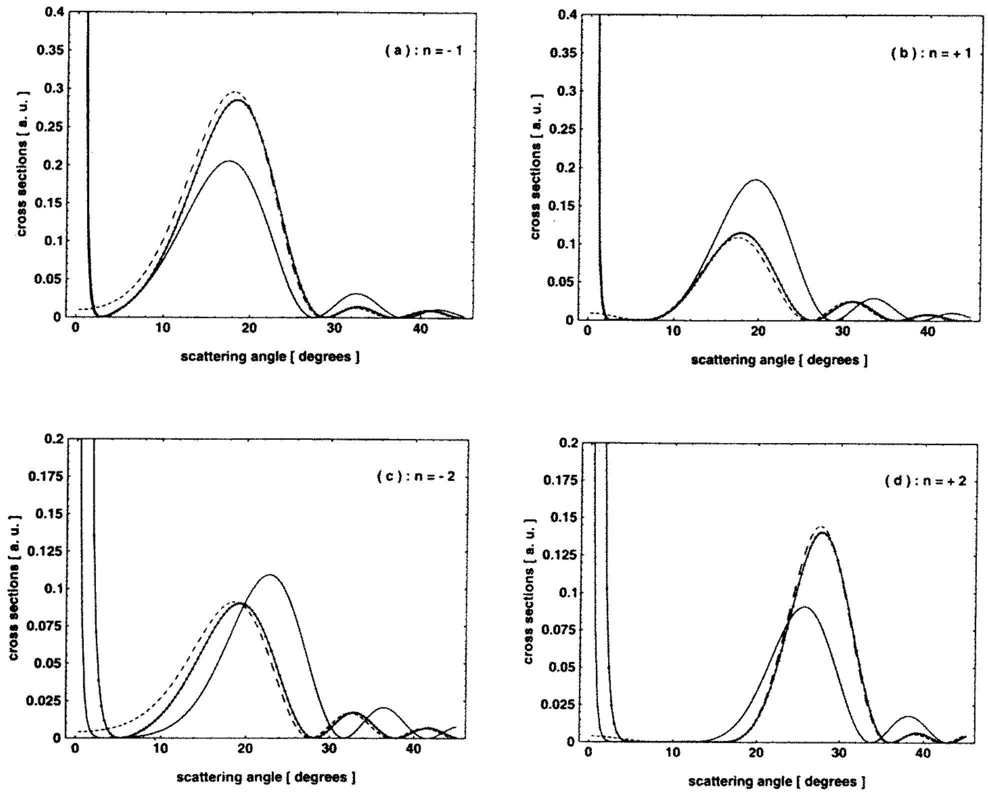

Figure 2. $a_0^{-2}\,\mathrm{d}\sigma_n/\mathrm{d}\Omega$ as a function of θ for fixed φ: (*a*) $n = -1(\varphi = 0)$; (*b*) $n = +1(\varphi = 0)$; (*c*) $N = -2(\varphi = \pi)$; and (*d*) $n = +2(\varphi = \pi)$. The values of φ are those yielding the largest cross sections in figure 1(*a*) and (*b*). Continuous curves refer to the data for a single frequency and target dressing, dashed curves to a bichromatic field and dotted curves to a bichromatic field and target dressing. Target dressing in a bichromatic field becomes prominent at very small θ, but at larger θ than for a single field.

not. Hence, for larger scattering angles the coherent phase control in a bichromatic field may be safely investigated by describing the atom by a static potential.

In figure 1 we show for $\theta = (2/3)^\circ$ and for $0 \leqslant \varphi \leqslant 2\pi$ the phase dependence of the nonlinear differential cross sections in au for (*a*) $n = 0, \pm 1$; (*b*) $n = \pm 2$; (*c*) $n = \pm 3$ and (*d*) $n = \pm 4$. Since the value of n roughly determines the number of terms which have to be taken into account in the sum over λ in (13), we observe no phase effects for $n = 0$ and we recognize that the phase effects increase with increasing $|n|$, although the cross sections get smaller. Moreover, for $\hbar\omega = 1.17$ eV the n-dependence of $\vec{Q}_n$ becomes appreciable and hence does the difference in the cross sections for $\pm|n|$.

Since, according to figure 1, the cross sections for $n = \pm 1$ are largest for $\varphi = 0$ and for $n = \pm 2$ have their largest values for $\varphi = \pi$, we show in figure 2 the angular dependence of the nonlinear cross sections for (*a*) $n = -1$ and (*b*) $n = +1$ both with $\varphi = 0$, and for (*c*) $n = -2$ and (*d*) $n = +2$ with $\varphi = \pi$. In all four figures, the continuous curves indicate the data obtained from (14) adapted to a single frequency corresponding to the formula of Zon [50], the dashed lines refer to the data obtained if in (14) the polarization effects are neglected and the dotted lines correspond to the cross section values evaluated

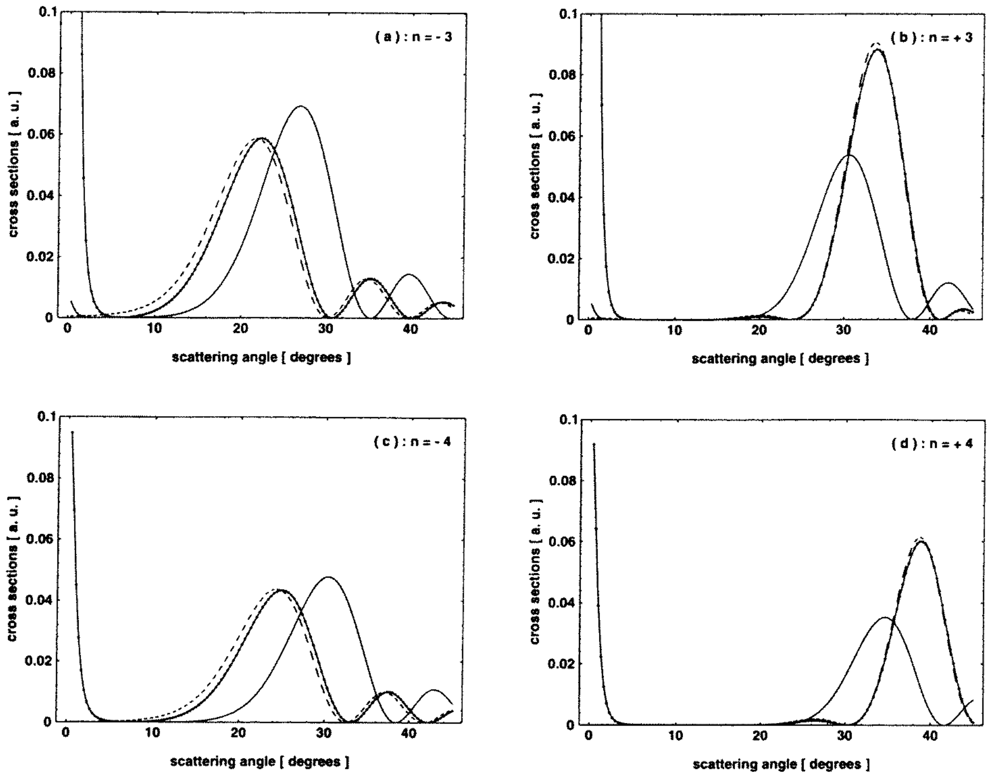

Figure 3. The same as in figure 2 but with (a) $n = -3$; (b) $n = +3$; (c) $n = -4$ and (d) $n = +4$ with $\varphi = \pi$ in all four cases.

from (14) including the target dressing. For $n = \pm 2$, in particular, there is a significant difference between the data for scattering in a single field and in a bichromatic field, but the target-dressing effects are only considerable for small scattering angles $\theta \lesssim 5°$. As we can see, the case $n = -2$ is one exception where target dressing in a bichromatic field has some influence on the cross section data for larger scattering angles, but even here the effect is rather minute.

In figure 3 similar angular dependences of the cross section data are shown for $\varphi = \pi$ in (a) for $n = -3$, (b) $n = +3$, (c) $n = -4$ and (d) $n = +4$. The continuous, dashed and dotted curves have the same meaning as in figure 2. At larger scattering angles, there are considerable differences between scattering in a single and a bichromatic laser field, but only at very small scattering angles a difference between target dressing in a single and a bichromatic field are indicated, where for $n = \pm 3$ we see that the target-dressing effects set in at larger scattering angles for a bichromatic field than for a single field.

Finally, we present in figure 4(a) the angular dependence of the differential cross section for $n = 0$ and $\varphi = 0$ for the three cases of scattering in a monochromatic field including laser dressing of the target atom (full curve), scattering in a bichromatic field without target dressing (dashed curve) and scattering in a bichromatic field with target dressing (dotted curve). Apparently, over the whole angular range there is hardly any difference between the cross sections for the three cases. In 4(b) we show the scattering spectra for $\theta = (2/3)°$ and $\varphi = 0$ where the bars correspond to scattering in the bichromatic field including

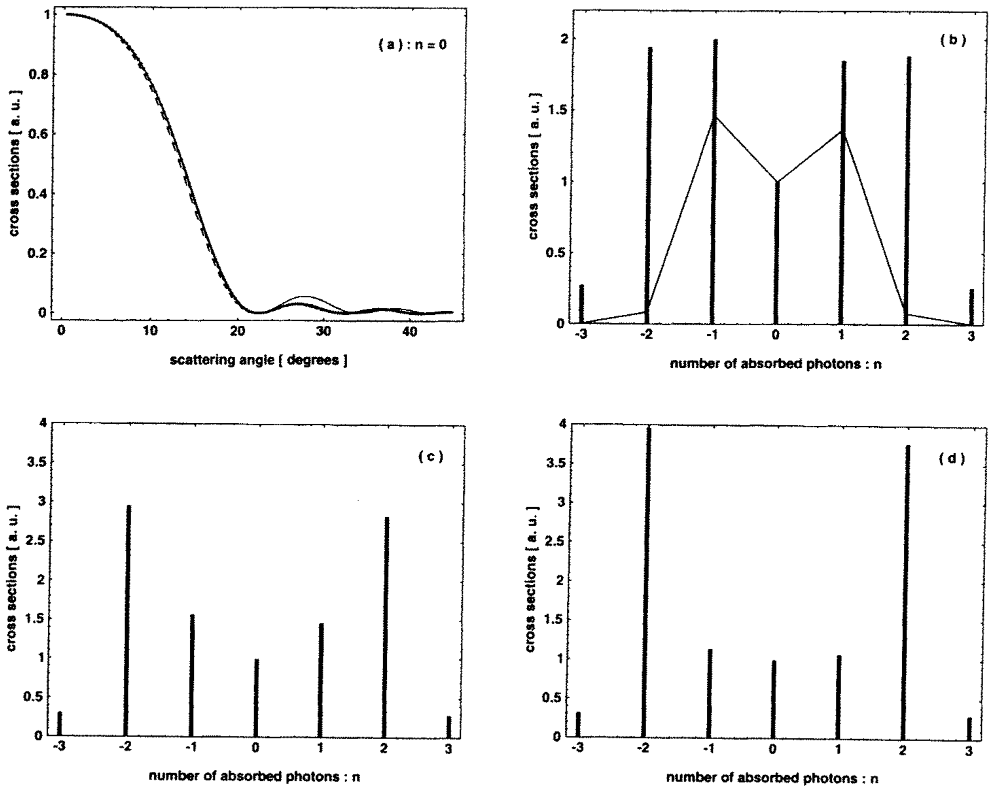

Figure 4. (*a*) $a_0^{-2}\,\mathrm{d}\sigma_0/\mathrm{d}\Omega$ for $(2/3)^\circ \leqslant \theta \leqslant 45^\circ$ as in figures 2 and 3 and (*b*) the spectrum evaluated from (14) for $\varphi = 0$ and $\theta = (2/3)^\circ$, where the thin line indicates the data for a single frequency and target dressing. (*c*) and (*d*) give the spectra at $\theta = (2/3)^\circ$ for $\varphi = \pi/2$ and $\varphi = \pi$, respectively, obtained from (14). The largest cross sections are at $n = \pm 1, \pm 2$ in accordance with figure 1.

the polarization effects, whereas the thin lines indicate the spectra for a single field with dressing effects. In the remaining figures 4(*c*) and (*d*) we depict the nonlinear cross sections at $\theta = (2/3)^\circ$ for $\varphi = \pi/2$ in (*c*) and for $\varphi = \pi$ in (*d*) evaluated from (14) in atomic units. Figures 4(*b*)–(*d*) demonstrate the considerable phase-dependent effects which are strongly modified by the laser dressing of the atom at very small scattering angles.

4. Summary and conclusions

In the investigation presented above, we considered electron–atom scattering in a bichromatic laser field taking into account the dressing of the atomic target by the radiation field. For simplicity we treated the dressing effects by first-order time-dependent perturbation theory, concentrating on the non-resonant case and using the closure approximation for evaluating the laser-induced polarization potential. The calculations were performed for hydrogen as the simplest target atom and exchange effects were neglected. The scattering of electrons in the bichromatic field by the effective laser-dressed potential was then evaluated to the first-order Born approximation yielding as the final result the nonlinear cross section formula (14) as the corresponding generalization of the expression derived by Zon [50] for a single field. The main conclusion drawn from our numerical example was that the dressing effects have only a considerable influence on the nonlinear cross section data in a bichromatic phase-dependent field under an extreme choice of parameters, namely at very small scattering angles $\theta \lesssim 1^\circ$ and for rather high laser field intensities of about 10^{13} W cm^{-2} such that multiphoton ionization becomes a concomitant process. Hence, the coherent phase control in scattering of electrons by atoms in a bichromatic radiation field can be best observed at larger scattering angles under less stringent parameter conditions and in this case the target atom can be safely described by a static potential. We do not think that these conclusions will be essentially modified if our calculations were amplified by taking into account exchange effects and higher-order Born terms or by treating the laser dressing of the target atom by means of Floquet techniques including resonance phenomena, as was done for a single frequency by Joachain and co-workers [52–55, 59–61].

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