Strong versus Weak Excitation in Frequency Mixing and Harmonic Generation by Two-Level Systems

R. Parzyński* and T. Kopyciuk

Faculty of Physics, A. Mickiewicz University, Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland

Received: 22.07.2021 & Accepted: 07.10.2021

Doi: 10.12693/APhysPolA.140.407

*e-mail: parzynsk@amu.edu.pl

In the limits of weak and strong excitation, we solve analytically the Riccati-type differential equation for a two-level system in the field of two different lasers and then use these solutions to find easy to handle formulae for the induced electric-dipole moment of the system. We apply these formulae to describe the effects of strong excitation in both the higher harmonics generation and multiphoton frequency mixing, namely the diminishing of the generation of harmonics of a given laser when an additional laser is turned on and the diminishing of the radiation at mixed frequencies with increasing strength of lasers under the condition of multiphoton excitation.

topics: extreme nonlinear optics, frequency conversion

1. Introduction

One of the theoretical methods of nonlinear optics and multiphoton physics of two-level systems exploits the Riccati-type differential equation as its basis [1, 2]. It is a time-dependent nonlinear differential equation for the ratio of the population amplitudes in the system exposed to electromagnetic field. This single Riccati equation replaces the standard pair of coupled differential equations for the population amplitudes [3]. Recently, we have solved analytically this Riccati equation along approximate lines for a single-laser field [1, 4, 5] and used the solutions to study, e.g., field-dependent corrections to refractive index [5], bunch-type dipole spectrum of harmonics generated [4], and ionization effects in these harmonics [1]. However, due to the single-beam assumption, these solutions did not cover the phenomenon of frequency mixing of different laser beams. In the practice of spectroscopic measurements, e.g., this phenomenon is of great importance since by mixing different frequencies one obtains a new radiation of a frequency better suited to a given transition frequency.

In the present paper, we remove this deficiency giving general solutions of the Riccati-type equation for a many-colour field produced by several different-frequency lasers. Two limiting cases will be considered for which approximate analytical solutions are accessible. The first case is that of weak excitation of a two-level system and the other concerns its strong excitation. The former case is realized when the standard field-dependent Rabi frequency is much smaller than the frequency of separation between the levels in the system while in the latter case the above inequality is reversed. The general solutions found in these two excitation limits are then adapted to the representative case of the two-colour (two-laser) field and the appropriate formulae for the dipole moment induced by such a field will be derived. From these formulae we will finally conclude about the effect of strength of the two-colour field in the spectrum of the radiation emitted by the induced dipole moment.

2. Fundamentals

Let $\hbar\Delta$ be the separation energy between two states of the opposite parity, the lower state $|1\rangle$ and the upper state $|2\rangle$. The assumed opposite parity means that this paper does not concern the so-called asymmetric two-level systems with broken inversion symmetry [6]. When exposed to an electromagnetic field, a two-state system is described by the superposition $\Psi(t) = C_1(t)\,|1\rangle + C_2(t)\,|2\rangle \exp(-\mathrm{i}\,\Delta t)$ with the state population amplitudes, $C_1(t)$ and $C_2(t)$, governed by the set of differential equations [3]

$$\mathrm{i}\,\dot{C}_1 = -Q(t)C_2, \tag{1}$$

and

$$\mathrm{i}\,\dot{C}_2 = -Q^*(t)\,C_1, \tag{2}$$

where the dots over $C_1(t)$ and $C_2(t)$ stand for the time derivatives, and $Q(t)$ is determined by the matrix element of the interaction Hamiltonian between the system and the electromagnetic field. In the present paper, the electromagnetic field is assumed to come from the combination of a number of different laser beams. A given laser beam is treated classically (see [7, 8] for the approaches with quantized field) and characterized by its frequency ω_j, electric field amplitude $\boldsymbol{\varepsilon}_{0j}$ and smooth laser-pulse envelope $0 \leq f_j(t) \leq 1$, where $j = 1, 2, 3, \ldots$ numbers the beams. For all beams, we take the linear polarization $\boldsymbol{\varepsilon}_j(t) = \boldsymbol{\varepsilon}_{0j} f_j(t) \cos(\omega_j t)$ and the electric-dipole approximation, but no rotating-wave approximation, resulting in

$$Q(t) = \sum_j Q_j(t) = \mathrm{e}^{-\mathrm{i}\Delta t} \sum_j \Omega_j f_j(t) \cos(\omega_j t), \tag{3}$$

where $\Omega_j = \boldsymbol{\mu}_{21} \cdot \boldsymbol{\varepsilon}_{0j}/\hbar$ is the standard Rabi frequency for the j beam and $\boldsymbol{\mu}_{21} = \langle 2 | e\boldsymbol{r} | 1 \rangle$ — the dipole matrix element.

In this paper, we replace the ordinary set of (1) and (2) by a single equation of motion for the variable $R(t) = C_2(t)/C_1(t)$. The equation for $R(t)$ is the known [1, 2] quadratically nonlinear Riccati-type differential equation

$$\mathrm{i}\dot{R} = Q(t) R^2 - Q^*(t). \tag{4}$$

In the limits of weak and strong excitation, we will solve the last equation analytically in order to derive the formulae for the induced electric-dipole moment of the system, $\boldsymbol{d}(t)$, defined as

$$\boldsymbol{d}(t) = \langle \Psi(t) | e\boldsymbol{r} | \Psi(t) \rangle = 2\boldsymbol{\mu}_{21} \mathrm{Re}\left(C_1 C_2^* \mathrm{e}^{\mathrm{i}\Delta t}\right) = 2\boldsymbol{\mu}_{21} \frac{\mathrm{Re}\left(R\mathrm{e}^{-\mathrm{i}\Delta t}\right)}{1 + |R|^2}. \tag{5}$$

3. Weak-excitation limit

3.1. Linearization technique

If the system was initially ($t = t_0$) in its lower state $|1\rangle$, the domain of weak excitation means $|R(t)|^2 \ll 1$ for arbitrary t. In this limit, we will adapt the linearization method of approximate solution of (4) presented previously [1, 2] in the context of the single-laser field. Shortly, the method consists in, first, neglecting the quadratic term $Q(t)R^2$ in (4) and finding the zero-order solution

$$R_0(t) = \mathrm{i} \int_{t_0}^{t} \mathrm{d}t'\, Q^*(t') \tag{6}$$

and, then, postulating the corrected solution in the form $R(t) = R_0(t) + R_1(t)$ with the restriction that $|R_1(t)| \ll |R_0(t)|$. With this restriction, the Riccati equation for R_1, obtained from (4), is linearized to the ordinary differential equation

$$\mathrm{i}\dot{R}_1 = 2QR_0R_1 + QR_0^2, \tag{7}$$

whose formal solution is [9]

$$R_1(t) = -\frac{1}{2} \int_{t_0}^{t} \mathrm{d}t'\, R_0(t') \frac{\mathrm{d}Z(t,t')}{\mathrm{d}t'} \tag{8}$$

with

$$Z(t,t') = \exp\left(2 \int_{t'}^{t} \mathrm{d}t''\, \dot{R}_0^*(t'') R_0(t'')\right). \tag{9}$$

To calculate $R_0(t)$ from (6), with $Q(t)$ given by (3), we assume long laser pulses in the sense that each pulse-shape function $f_j(t)$ has its full width at half maximum much greater than the optical cycle $2\pi/\omega_j$. Also, the laser frequencies ω_j are assumed to be far from the transition frequency Δ. Under these assumptions, the j component of (6) includes the time integral of the product of the slowly time-varying pulse-shape function $f_j(t)$ and the fast-varying function $\exp(\mathrm{i}\Delta t)\cos(\omega_j t)$. We calculate (6) by parts for an arbitrary smooth pulse-shape function $f_j(t)$, applying the boundary condition $f_j(t_0) = 0$ and finally retaining only the leading part $f_j(t) \int \mathrm{d}t \exp(\mathrm{i}\Delta t)\cos(\omega_j t)$, resulting in the approximate

$$R_0(t) = \mathrm{e}^{\mathrm{i}\Delta t} \sum_j \frac{x_j f_j(t)}{y_j^2 - 1} \times \Big(y_j \cos(\omega_j t) - \mathrm{i}\sin(\omega_j t)\Big) = \sum_j R_{0j}(t), \tag{10}$$

where

$$x_j = \frac{\Omega_j}{\omega_j}, \quad y_j = \frac{\Delta}{\omega_j} \tag{11}$$

are dimensionless strength and frequency parameters, respectively, related to a given laser beam. To fulfil the condition of weak excitation, i.e., $|R(t)|^2 \approx |R_0(t)|^2 \ll 1$, each pair of the parameters needs to be chosen so that $|R_{0j}(t)|^2 \ll 1$. This will lead to the restriction

$$\frac{x_j^2}{2} \frac{y_j^2 + 1}{\left(y_j^2 - 1\right)^2} \ll 1, \tag{12}$$

obtained by replacing $\cos^2(\omega_j t)$ and $\sin^2(\omega_j t)$ by their time average values, and $f_j^2(t)$ by its maximum value equal to 1. Obviously, (10) and (12) can be used far from one-photon resonance only ($y_j \neq 1, \omega_j \neq \Delta$).

3.2. Low-frequency case

From now on we focus on a physically important case of low laser frequencies, meaning $y_j = \Delta/\omega_j \gg 1$. Alternatively, this frequency limit is also called the multiphoton excitation one. It is usually met when typical lasers are applied to excite the majority of atoms, molecules and ions from their ground states to their first excited states. In this limit, we are allowed to simplify (10) for $R_0(t)$ by rejecting the term proportional to $\sin(\omega_j t)$. With the so simplified $R_0(t)$, the integrand in (9) for $Z(t,t')$ takes the form

$$2\dot{R}_0^*(t'') R_0(t'') = -\mathrm{i}2Q(t'') R_0(t'') = -\frac{\mathrm{i}}{\hbar} \sum_j \sum_{j'} \boldsymbol{d}_{0j}(t'') \cdot \boldsymbol{\mathcal{E}}_{j'}(t''), \tag{13}$$

where $\boldsymbol{\mathcal{E}}_{j'}(t'') = \boldsymbol{\mathcal{E}}_{0j'} f_{j'}(t'') \cos(\omega_{j'} t'')$ is the electric field of the j' laser beam, and

$$\boldsymbol{d}_{0j}(t'') = 2\boldsymbol{\mu}_{21} \frac{x_j y_j}{y_j^2 - 1} f_j(t'') \cos(\omega_j t'') \tag{14}$$

is the electric dipole moment induced by the j laser beam in the lowest-order approximation, i.e., by substituting $R(t) = R_{0j}(t)$ in (5) and then neglecting the term $|R_{0j}(t)|^2$. In (13), the double summation is recognized as the total interaction energy between all induced partial dipole moments $\boldsymbol{d}_{0j}(t)$ and all laser fields engaged $\boldsymbol{\mathcal{E}}_{j'}(t)$.

To find $Z(t,t')$ from (9), we integrate (13) over time along a similar line as that described before (10) and obtain $Z(t,t') = \exp(\mathrm{i}(u(t') - u(t)))$, where

$$u(\tau) = \sum_j \left[\left(a_j \int \mathrm{d}\tau\, f_j^2(\tau) \right) + b_j f_j^2(\tau) \sin(2\omega_j \tau) \right] + \sum_j \sum_{j'>j} f_j(\tau) f_{j'}(\tau) x_j x_{j'} \left(\frac{y_j^2}{y_j^2 - 1} + \frac{y_{j'}^2}{y_{j'}^2 - 1} \right) \times \left(\frac{\sin(\omega_j + \omega_{j'})\tau}{y_{j'} + y_j} + \frac{\sin(\omega_j - \omega_{j'})\tau}{y_{j'} - y_j} \right) \tag{15}$$

with

$$a_j = \frac{x_j^2 y_j}{y_j^2 - 1}\omega_j, \quad b_j = \frac{a_j}{2\omega_j}. \tag{16}$$

Above, the single summation over j originates in the interaction between the zero-order partial dipole moment $\boldsymbol{d}_{0j}(t)$, induced by the j laser beam, with the same j beam. On the other hand, the double summation over j and j' comes from the interaction between the partial dipole moment $\boldsymbol{d}_{0j}(t)$, induced by the j laser beam, with a different $j' \neq j$ beam. Moreover, the parameter a_j is simply the ordinary Stark shift (in a frequency scale) introduced to the transition frequency Δ by the j laser beam.

3.3. Application to two-colour field

As representative, we consider in detail the case of two laser beams ($j = 1,2$ and $j' = 1,2$) of different low frequencies such that $y_j^2 - 1 \approx y_j^2$. Using the Fourier–Bessel expansion [10, 11] to $\exp(\mathrm{i}u(t'))$ in $Z(t,t')$ (see Appendix A) we then get the dominant part of the integrand in (8). Its form is

$$R_0(t') \frac{\mathrm{d}Z(t,t')}{\mathrm{d}t'} = \frac{\mathrm{i}\Delta}{4} \mathrm{e}^{-\mathrm{i}u(t)} \sum_{n=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \sum_{k=-\infty}^{\infty} \sum_{l=-\infty}^{\infty} B_{n,m,k,l}(t') \times \exp\left(\mathrm{i} \left(\Big(\Delta + (2n+k+l+1)\omega_1 + (2m+k-l)\omega_2 \Big) t' + a_1 \int \mathrm{d}t'\, f_1^2(t') + a_2 \int \mathrm{d}t'\, f_2^2(t') \right) \right), \tag{17}$$

where the coefficients $B_{n,m,k,l}(t')$ are defined from (A1) in Appendix A. These coefficients are slowly varying in time through the pulse-shape functions $f_j(t')$ and also depend on strength of pulses (see (A2)).

Finally, (17) is integrated over time t' with the assumption that the low-frequency Stark shifts $a_j = (x_j/y_j)^2 \Delta$ are much smaller than the mixed multiphoton detunings $|\delta| = |\Delta + (2n+k+l+1)\omega_1 + (2m+k-l)\omega_2|$. The ratio $(x_j/y_j)^2$ is, however, restricted here by the weak-excitation condition $(x_j/y_j)^2 \ll 2$ resulting from (12) for the assumed low frequencies ($y_j \gg 1$). Consequently, for the weak excitation by a low-frequency field the two conditions, $a_j \ll |\delta|$ and $(x_j/y_j)^2 \ll 2$, are reconciled when

$$\left(\frac{x_j}{y_j} \right)^2 = \left(\frac{\Omega_j}{\Delta} \right)^2 \ll \min\left(2, \left| 1 + \frac{2n+k+l+1}{y_1} + \frac{2m+k-l}{y_2} \right| \right), \tag{18}$$

where $\min(u,v)$ means the smaller of the values u and v. Under the restriction given by (18), the only fast varying function in (17) is $\exp(\mathrm{i}\delta t')$. Thus, the dominant part of $R_1(t)$, obtained from integration of (17) by parts along the line described before (10), is

$$R_1(t) = -\frac{1}{8} \sum_{n,m,k,l=-\infty}^{\infty} \sum_{n',m',k',l'=-\infty}^{\infty} \frac{B_{n,m,k,l}^{n',m',k',l'}(t)}{1 + \frac{2n+k+l+1}{y_1} + \frac{2m+k-l}{y_2}} \times \exp\left(\mathrm{i} \Big(\Delta + \big(2(n-n') + k - k' + l - l' + 1\big)\omega_1 + \big(2(m-m') + k - k' - l + l'\big)\omega_2 \Big) t \right) \tag{19}$$

where the summation over n', m', k', l' comes from the additional expansion of $\exp\left(-\mathrm{i}\,u(t)\right)$ in the Fourier–Bessel series, and

$$B_{n,m,k,l}^{n',m',k',l'}(t) = B_{n,m,k,l}(t)\, J_{n'}(\alpha_1(t)) \times J_{m'}(\alpha_2(t))\, J_{k'}(\beta_+(t))\, J_{l'}(\beta_-(t)) \tag{20}$$

with $\alpha_j(t)$ and $\beta_\pm(t)$ given by (A2) in Appendix A.

3.4. Induced dipole moment in the case of weak excitation by two-colour field of low frequencies

Due to (5), (10), and (19), two low-frequency laser beams ($y_j = \Delta/\omega_j \gg 1$), weakly exciting a two-level system (see (18)), induce the electric dipole moment $\boldsymbol{d}(t) = \boldsymbol{d}_0(t) + \boldsymbol{d}_1(t)$, where

$$\boldsymbol{d}_0(t) = 2\boldsymbol{\mu}_{21}\mathrm{Re}\left(R_0(t)\,\mathrm{e}^{-\mathrm{i}\,\Delta t}\right) = 2\boldsymbol{\mu}_{21}\sum_{j=1}^{2}\frac{x_j}{y_j} f_j(t)\cos(\omega_j t) \tag{21}$$

and

$$\boldsymbol{d}_1(t) = 2\boldsymbol{\mu}_{21}\mathrm{Re}\left(R_1(t)\,\mathrm{e}^{-\mathrm{i}\,\Delta t}\right) = -\frac{\boldsymbol{\mu}_{21}}{4}\sum_{n,m,k,l=-\infty}^{\infty}\;\sum_{n',m',k',l'=-\infty}^{\infty} B_{n,m,k,l}^{n',m',k',l'}(t)$$
$$\times\frac{\cos\Big(\big(2(n-n')+k-k'+l-l'+1\big)\omega_1 + \big(2(m-m')+k-k'-l+l'\big)\omega_2\Big)t}{1+(2n+k+l+1)/y_1 + (2m+k-l)/y_2}. \tag{22}$$

Part $\boldsymbol{d}_0(t)$ arises from the zero-order solution to the Riccati equation (see (6)) and describes the ordinary oscillations of the induced dipole moment at frequencies of the incoming laser beams. However, part $\boldsymbol{d}_1(t)$ results from the correction to the above solution (see (8)) and describes the additional oscillations at frequencies being different combinations of those of the incoming laser beams, i.e., the mixing of laser beams. Apart from the frequency denominator in (22), the amplitudes of the mixed oscillations are determined from (20) and (A1) for the field-dependent coefficients $B_{n,m,k,l}^{n',m',k',l'}(t)$. Obviously, the amplitudes of the mixed oscillations are much smaller than those of the oscillations given by (21), due to the restriction (18). This restriction causes that the arguments α_j and $\beta_\pm$ (see (A2)) of the Bessel functions in (20) and (A1) for the coefficients $B_{n,m,k,l}^{n',m',k',l'}(t)$ are generally much smaller than 1, if the laser frequencies are not too close to each other. Then, the greatest contribution to these coefficients comes from two sets of indices. Namely, $n', m', k', l', k = 0$, $l = 0, -1$ and the corresponding Bessel functions in (20) and (A1) can be approximated by 1. Consequently, the number of summations in (22) for $\boldsymbol{d}_1(t)$ is reduced to two only, namely over n and m, resulting in

$$\boldsymbol{d}_1(t) =$$
$$-\frac{\boldsymbol{\mu}_{21}}{4}\sum_{n,m=-\infty}^{\infty}\Bigg\{\Bigg[\frac{x_1^3}{y_1^3}f_1^3(t)\Big(3J_n\big(\alpha_1(t)\big)+3J_{n+1}\big(\alpha_1(t)\big)+J_{n-1}\big(\alpha_1(t)\big)+J_{n+2}\big(\alpha_1(t)\big)\Big)J_m\big(\alpha_2(t)\big)$$
$$+3\frac{x_1}{y_1}\frac{x_2^2}{y_2^2}f_1(t)f_2^2(t)\Big(J_n\big(\alpha_1(t)\big)+J_{n+1}\big(\alpha_1(t)\big)\Big)\Big(J_{m-1}\big(\alpha_2(t)\big)+J_{m+1}\big(\alpha_2(t)\big)+2J_m\big(\alpha_2(t)\big)\Big)\Bigg]$$
$$\times\frac{\cos\left((2n+1)\omega_1 t + 2m\omega_2 t\right)}{1+(2n+1)/y_1+2m/y_2} + (1\rightleftarrows 2, n\rightleftarrows m)\Bigg\}, \tag{23}$$

where $(1\rightleftarrows 2, n\rightleftarrows m)$ is the term obtained from the preceding one by a mutual interchange of the indices at x_j, y_j, f_j, α_j and J_q.

From (23) we can calculate simple formulae for the dipole components $d_1(t,\omega)$ oscillating at fixed frequencies ω. The dominant contribution to a given dipole component will arise from as small as possible summation indices in this equation due to the behaviour of the Bessel functions at small arguments. Approximating each Bessel function by its leading part in the series representation, we thus obtain from (23) the exemplifying single-beam formulae for the dipole components oscillating at the frequencies of odd harmonics

$$\boldsymbol{d}_1(t,3\omega_j) = -\frac{\boldsymbol{\mu}_{21}}{2}\left(\frac{\Omega_j}{\Delta}\right)^3\frac{y_j^2}{y_j^2-9} \times f_j^3(t)\cos(3\omega_j t), \tag{24}$$

$$\boldsymbol{d}_1(t,5\omega_j)=\frac{\boldsymbol{\mu}_{21}}{4}\left(\frac{\Omega_j}{\Delta}\right)^5\frac{y_j^4+15y_j^2}{\left(y_j^2-9\right)\left(y_j^2-25\right)}\times f_j^5(t)\cos(5\omega_j t)\,, \tag{25}$$

$$\boldsymbol{d}_1(t,7\omega_j)=-\frac{\boldsymbol{\mu}_{21}}{8}\left(\frac{\Omega_j}{\Delta}\right)^7\times\frac{y_j^6+71y_j^4}{\left(y_j^2-9\right)\left(y_j^2-25\right)\left(y_j^2-49\right)}f_j^7(t)\cos(7\omega_j t)\,, \tag{26}$$

$$\boldsymbol{d}_1(t,9\omega_j)=\frac{\boldsymbol{\mu}_{21}}{16}\left(\frac{\Omega_j}{\Delta}\right)^9\times\frac{y_j^8+206y_j^6+945y_j^4}{\left(y_j^2-9\right)\left(y_j^2-25\right)\left(y_j^2-49\right)\left(y_j^2-81\right)}\times f_j^9(t)\cos(9\omega_j t)\,, \tag{27}$$

and the two-beam formulae for the dipole components oscillating at mixed frequencies

$$\boldsymbol{d}_1(t,\omega_1\pm2\omega_2)=-\frac{3}{2}\boldsymbol{\mu}_{21}\frac{\frac{\Omega_1}{\Delta}\left(\frac{\Omega_2}{\Delta}\right)^2}{1-\left(\frac{1}{y_1}\pm\frac{2}{y_2}\right)^2}\times f_1(t)f_2^2(t)\cos(\omega_1\pm2\omega_2)\,t, \tag{28}$$

$$\boldsymbol{d}_1(t,\omega_1\pm4\omega_2)=\frac{3}{8}\boldsymbol{\mu}_{21}\frac{\Omega_1}{\Delta}\left(\frac{\Omega_2}{\Delta}\right)^4\times\frac{y_2\left(\frac{4}{y_2}\pm\frac{1}{y_1}\right)}{1-\left(\frac{4}{y_2}\pm\frac{1}{y_1}\right)^2}f_1(t)f_2^4(t)\cos(\omega_1\pm4\omega_2)\,t, \tag{29}$$

$$\boldsymbol{d}_1(t,3\omega_1\pm2\omega_2)=\frac{\boldsymbol{\mu}_{21}}{8}\left(\frac{\Omega_1}{\Delta}\right)^3\left(\frac{\Omega_2}{\Delta}\right)^2\times\frac{(3y_1\pm y_2)\left(\frac{3}{y_1}\pm\frac{2}{y_2}\right)}{1-\left(\frac{3}{y_1}\pm\frac{2}{y_2}\right)^2}\times f_1^3(t)f_2^2(t)\cos(3\omega_1\pm2\omega_2)\,t. \tag{30}$$

In (24)–(30), the y_j-dependent fractions do not affect substantially the dipole components because $y_j \gg 1$ here. Thus, the range of magnitude of a given component is mainly determined by the appropriate power(s) of the ratio(s) Ω_j/Δ being much smaller than 1 according to (18). As a reference point, when considering the limit of strong excitation in Sects. 4 and 5, will serve (24)–(30).

4. Strong-excitation limit

4.1. General expression for induced dipole moment

To consider the case of strong excitation, it is convenient to transform (4) for the variable $R(t)$ into equation for $r(t)=R(t)\,\mathrm{e}^{-\mathrm{i}\,\Delta t}$ with the result

$$\mathrm{i}\dot{r}=\left(r^2-1\right)\Omega(t)+\Delta r, \tag{31}$$

where $\Omega(t)=\sum_j\Omega_j(t)$ with $\Omega_j(t)=\Omega_j f_j(t)\cos(\omega_j t)$ and $\Omega_j=\boldsymbol{\mu}_{21}\cdot\boldsymbol{\mathcal{E}}_{0j}/\hbar$. On the basis of (31), we define the limit of strong excitation by the condition $\Omega_j\gg\Delta$ being the opposite to the condition for weak excitation by a low-frequency field (see (18)).

For $\Omega_j\gg\Delta$, the first term generally dominates the second term on the right-hand side of (31). We thus neglect Δr in (31) and obtain the following zero-order solution in the case of strong excitation

$$r_0=\mathrm{i}\tan\left(\int\limits_{t_0}^{t}\mathrm{d}t'\,\Omega(t')\right). \tag{32}$$

In the next step, we calculate the effect of Δr on the above $r_0(t)$ postulating the corrected solution to (31) in the form $r(t)=r_0(t)+r_1(t)$ with the restriction that $|r_1(t)|\ll|r_0(t)|$. Under this restriction, (31) leads to the following linearized in $r_1(t)$ differential equation for the correction

$$\mathrm{i}\dot{r}_1=2\Omega(t)\,r_0r_1+\Delta r_0. \tag{33}$$

The last equation is a strong excitation analogue to (7) valid for weak excitation. As an ordinary linear equation, (33) has the exact solution [9]

$$r_1(t)=\int\limits_{t_0}^{t}\mathrm{d}t'\left(-\mathrm{i}\,\Delta r_0(t')\right)\times\exp\left(2\mathrm{i}\int\limits_{t}^{t'}\mathrm{d}t''\,\Omega(t'')\,r_0(t'')\right)=\frac{\Delta}{2}\frac{\int\limits_{t_0}^{t}\sin\left(2\int\limits_{t_0}^{t'}\mathrm{d}t''\,\Omega(t'')\right)\mathrm{d}t'}{\cos^2\left(\int\limits_{t_0}^{t}\mathrm{d}t'\,\Omega(t')\right)}. \tag{34}$$

Applying (5), (32), and (34), we finally find the general expression of the induced electric-dipole moment in the limit of strong excitation $\Omega_j/\Delta\gg1$, i.e.,

$$\boldsymbol{d}(t)=2\boldsymbol{\mu}_{21}\frac{r_1}{1+|r_0+r_1|^2}\approx2\boldsymbol{\mu}_{21}\frac{r_1}{1+|r_0|^2}=\boldsymbol{\mu}_{21}\Delta\int\limits_{t_0}^{t}\mathrm{d}t'\,\sin\left(2\int\limits_{t_0}^{t'}\mathrm{d}t''\,\Omega(t'')\right). \tag{35}$$

4.2. Two-colour field

As in Sect. 3.3, we now focus on the representative case of two different laser beams. In this case, the leading part of the argument of the sinus function in (35) is $2x_1f_1(t')\sin(\omega_1t')+2x_2f_2(t')\,sin\,(\omega_2t')$. Thus, using the Fourier–Bessel expansions to sin $(q\sin(\varphi))$ and $\cos(q\sin(\varphi))$ [10, 11], (35) gives the following dominant part of $\boldsymbol{d}(t)$ when two laser beams strongly excite a two-level system

$$
\begin{aligned}
\boldsymbol{d}(t) &= J_0\big(2x_2 f_2(t)\big)\boldsymbol{d}_1(t) + J_0\big(2x_1 f_1(t)\big)\boldsymbol{d}_2(t) \\
&\quad -2\boldsymbol{\mu}_{21}\sum_{m=0}^{\infty}\sum_{n=1}^{\infty}\Big[J_{2m+1}\big(2x_1 f_1(t)\big)J_{2n}\big(2x_2 f_2(t)\big) \\
&\quad \times\left(\frac{\cos\big((2m+1)\omega_1 + 2n\omega_2\big)t}{(2m+1)/y_1 + 2n/y_2}\right. \\
&\quad \left.+\frac{\cos\big((2m+1)\omega_1 - 2n\omega_2\big)t}{(2m+1)/y_1 - 2n/y_2}\right) + (1 \rightleftarrows 2)\Big],
\end{aligned} \tag{36}
$$

where $x_j = \Omega_j/\omega_j = (\Omega_j/\Delta)\, y_j$, $y_j = \Delta/\omega_j$, $(1 \rightleftarrows 2)$ denotes the term obtained from the preceding one by mutual interchange of the indices at x_j, y_j, f_j, and ω_j, while

$$
\begin{aligned}
\boldsymbol{d}_j(t) &= -2\boldsymbol{\mu}_{21}\sum_{m=0}^{\infty}\frac{J_{2m+1}\big(2x_j f_j(t)\big)}{(2m+1)/y_j} \\
&\quad \times\cos(2m+1)\,\omega_j t
\end{aligned} \tag{37}
$$

with j equal to either 1 or 2. The particular (37) is the known formula of Ivanov and Corkum [12] originally obtained along a different line for the single-beam case. In our approach, the formula of Ivanov and Corkum results from (36) for the two-beam case by neglecting in this equation one of the two beams and using the property of the Bessel functions at the zero-value argument.

5. Strong-excitation effects caused by two-colour field

Considering (36) for the two-beam strong-excitation case, two effects are observed when compared to (37) for the one-beam case. One effect is described by the first two terms on the right-hand side of (36). These terms show that the generation of odd-order harmonics by a given beam is diminished in the presence of an additional laser beam. This diminishing is governed by the zero-order Bessel function depending on the strength of the additional beam. The reason for the above is that the additional beam simply enhances the degeneration of the two-level system. We show this effect of diminishing graphically in Figs. 1 and 2 prepared for the time-coincident laser pulses of the same shape taken at the maximum ($f_1(t) = f_2(t) = 1$). On the ordinate, we put the absolute value of the amplitude A_N of the odd N-th harmonic of laser 1 in the absence ($\Omega_2/\Delta = 0$) and in the presence of laser 2. This amplitude (measured in units of $\boldsymbol{\mu}_{21}$) was calculated from $A_N = (-2y_1/N)\, J_N(2x_1)\, J_0(2x_2)$, where $x_j = (\Omega_j/\Delta)\, y_j$. Such a form of A_N results from the first term in (36) along with (37) for $j = 1$. Each figure consists of three parts corresponding to different frequency regimes: (a) low frequencies ($y_j \gg 1$), (b) moderate frequencies ($y_j \approx 1$), (c) high frequencies ($y_j \ll 1$). To guide the eye, the calculated values of A_N, corresponding to the successive odd N, have been connected by straight lines. All curves in a given figure are made for the same strength Ω_1/Δ of laser 1 and a few different strengths Ω_2/Δ of laser 2. We stress that Ω_1/Δ is the only parameter that makes a difference between Fig. 1 ($\Omega_1/\Delta = 15$) and Fig. 2 ($\Omega_1/\Delta = 90$). Irrespective of the frequency regime, the mentioned diminishing of A_N when switching on laser 2 and changing its strength is evident. For fixed parameters of laser 2, the general shape of the curves in Figs. 1 and 2 is determined by $(y_1/N)\, J_N(2x_1)$, i.e., the term resulting from the single-beam (37) of Ivanov and Corkum. If $2x_1 = 2y_1(\Omega_1/\Delta)$ is large enough (parts (a) and (b) of the figures), then three parts can be highlighted in each curve, generally. For $N < 2x_1$, a given curve first gets lower with increasing N, then reaches a kind of a plateau when N increases and, finally, descents sharply for $N > 2x_1$. In the plateau region, the amplitudes

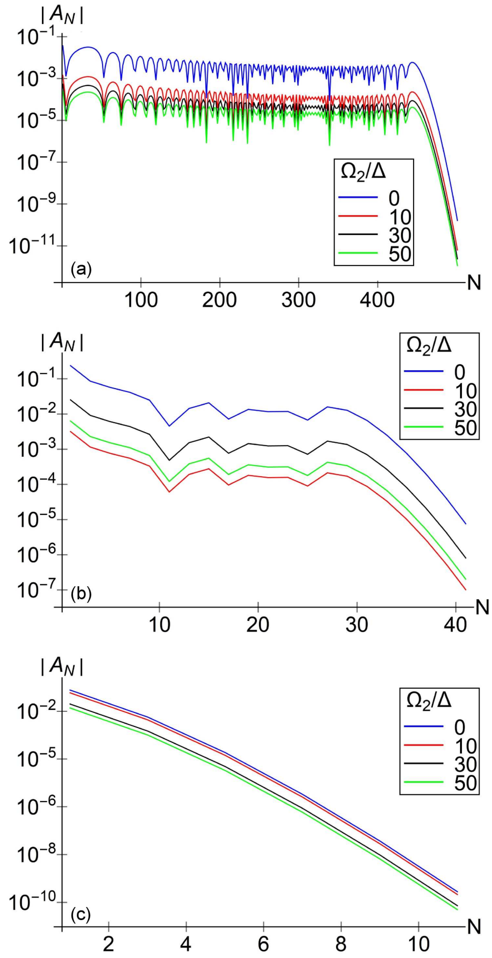

Fig. 1. The effect of strength of laser 2 ($\Omega_2/\Delta = 0$, 10, 30, 50) on the amplitude A_N of the N-th harmonic of laser 1 calculated from the strong excitation (36). For details, see the first paragraph of Sect. 5. (a) Low-frequency regime: $y_1 = 15$, $y_2 = 20$, (b) moderate-frequency regime: $y_1 = 1$, $y_2 = 0.9$, (c) high-frequency regime: $y_1 = 1/15$, $y_2 = 1/20$. The strength of laser 1 is kept fixed $\Omega_1/\Delta = 15$. Only the points on the curves corresponding to odd N have physical meaning.

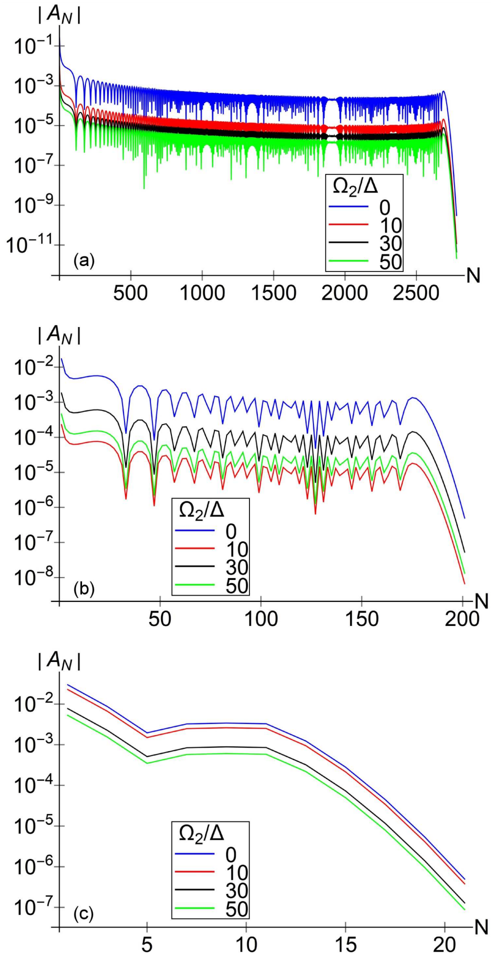

Fig. 2. Description is the same as given in Fig. 1, but for $\Omega_1/\Delta = 90$.

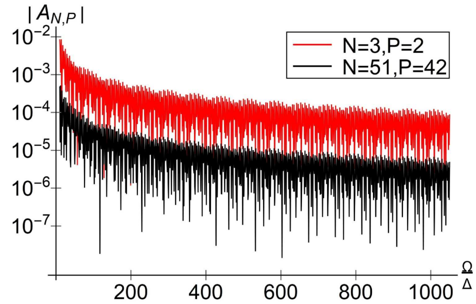

Fig. 3. The amplitude $A_{N,P}$ of the dipole component oscillating at a mixed frequency $N\omega_1 + P\omega_2$ versus the strength of lasers calculated from the strong-excitation given in (36). Both lasers are assumed to have the same strength ($\Omega_1/\Delta = \Omega_2/\Delta = \Omega/\Delta > 10$) but different low frequencies ($y_1 = 15$, $y_2 = 20$). For details, see par. 2 of Sect. 5..

corresponding to different N can have comparable values or even higher-N amplitudes can exceed lower-N amplitudes. If $2x_1$ gets increasingly smaller (parts (b) and (c) of the figures) then the plateau in Figs. 1 and 2 is shortened and even vanishes. Qualitatively, the shape of curves in parts (a) and (b) of Figs. 1 and 2 is similar to that known from higher-harmonic generation from different materials by a single laser in the so-called three-step mechanism involving laser ionization in the first step, then acceleration of the freed electron by the laser field and, finally, recombination of this electron with the parent ion [13–15]. The effect of an additional laser is to diminish this harmonics generation. No such effect has been encountered in the domain of weak excitation (see (24)–(27)).

The other effect is multiphoton mixing of frequencies of two laser beams, described by the part of (36) with double summation. The amplitudes of oscillations at mixed frequencies are shown to be determined by the products of the opposite-parity Bessel functions, depending on the strengths of different laser beams through the parameters x_j, as well as on the frequency denominators. These Bessel functions have their orders defined by the numbers of photons engaged in an elementary act of mixing. Obviously, the amplitudes of mixing in the strong-excitation case depend on the field strengths in a different way than those in the weak-excitation case. For evidence, let us focus on low frequencies ($y_j \gg 1$), i.e., the ones we have previously assumed in Sect. 3 when considering the case of weak excitation. This frequency regime leads, along with the strong-excitation condition ($\Omega_j/\Delta \gg 1$), to the strength parameters x_j being always much higher than 1. In the strong excitation (36), the appropriate Bessel functions of orders much smaller than their arguments can thus be well approximated by $J_k(q) \overset{q \gg k}{\approx} \sqrt{\frac{2}{\pi q}} \cos\left(q - \left(k + \frac{1}{2}\right)\frac{\pi}{2}\right)$ [10, 11]. As a result, the amplitudes of mixing described by these Bessel functions in (36) depend on the laser strengths through the factor $1/\sqrt{x_1 x_2}$, among others, and thus diminish with these strengths increasing. For low frequencies, such a behaviour in the strong-excitation case is quite the opposite to that in the weak-excitation case described in Sect. 3 (see, e.g., (28)–(30)). This strong-excitation effect is shown graphically in Fig. 3, where the absolute value of the amplitude $A_{N,P}$ of the dipole component oscillating at a mixed frequency $N\omega_1 + P\omega_2$ ($N = 1, 3, 5, \ldots$ and $P = 2, 4, 6, \ldots$) is plotted versus the strength parameter Ω/Δ assumed to be the same for both lasers, i.e., $\Omega_1/\Delta = \Omega_2/\Delta = \Omega/\Delta$. Generally, the expression for $A_{N,P}$ is $A_{N,P} = -2J_N(2x_1)\, J_P(2x_2) \,/\, (N/y_1 + P/y_2)$, where $x_j = (\Omega_j/\Delta)\, y_j$, and results from the third term in (36).

6. Conclusions

We have presented a fully analytical approach to multiphoton frequency mixing and high harmonics generation based on the Riccati-type differential equation for a two-level system in the field of several lasers. This approach has turned out to be very effective and led us to two main results given by (23) and (36) for the dipole moment induced in the system by the two-laser field under the conditions of weak and strong excitation, respectively. We have shown that both equations are easy in use when calculating the magnitude of the amplitudes of different dipole components oscillating at either high harmonics or mixed frequencies. Based on these equations, we have pointed to two strong-excitation effects. One effect, shown in Figs. 1 and 2, is the diminishing of the generation of odd-order harmonics of a given laser beam when an additional laser beam is turned on. The other effect, shown in Fig. 3, concerns low laser frequencies (multiphoton excitation) and consists in the diminishing of the radiation at mixed frequencies with increasing laser strengths. Obviously, both effects do not have their analogies in the weak-excitation limit (compare (24)–(30)).

Appendix A

To obtain (17), we have expanded $exp\left(\mathrm{i}u\left(t'\right)\right)$ in $Z\left(t,t'\right)$ into the Fourier–Bessel series, according to $\exp\left(\mathrm{i}q\sin\varphi\right) = \sum_{p=-\infty}^{\infty} J_p\left(q\right)\exp\left(\mathrm{i}pq\right)$, where $J_p\left(q\right)$ is the Bessel function of the first kind [10, 11]. As a result, the $B_{n,m,k,l}$ coefficients in (17) have been found as

$$B_{n,m,k,l}\left(t'\right) = \Bigg\{\Bigg[\left(\frac{x_1}{y_1}\right)^3 f_1^3\left(t'\right)\Big(3J_n\big(\alpha_1\left(t'\right)\big) + 3J_{n+1}\big(\alpha_1\left(t'\right)\big) + J_{n-1}\big(\alpha_1\left(t'\right)\big) + J_{n+2}\big(\alpha_1\left(t'\right)\big)\Big)J_m\big(\alpha_2\left(t'\right)\big)$$
$$+3\frac{x_1}{y_1}\left(\frac{x_2}{y_2}\right)^2 f_1\left(t'\right)f_2^2\left(t'\right)\Big(J_n\big(\alpha_1\left(t'\right)\big) + J_{n+1}\big(\alpha_1\left(t'\right)\big)\Big)$$
$$\times\Big(J_{m-1}\big(\alpha_2\left(t'\right)\big) + J_{m+1}\big(\alpha_2\left(t'\right)\big) + 2J_m\big(\alpha_2\left(t'\right)\big)\Big)\Bigg]J_l\big(\beta_-\left(t'\right)\big)$$
$$+\left(1 \rightleftarrows 2, n \rightleftarrows m, l \rightleftarrows l+1\right)\Bigg\}J_k\big(\beta_+\left(t'\right)\big) \tag{A1}$$

with

$$\alpha_j\left(t'\right) = b_j f_j^2\left(t'\right),$$
$$b_j = \frac{a_j}{2\omega_j} \approx \frac{y_j}{2}\left(\frac{x_j}{y_j}\right)^2,$$
$$\beta_\pm\left(t'\right) = \frac{2x_1x_2}{y_2 \pm y_1} f_1\left(t'\right)f_2\left(t'\right). \tag{A2}$$

Above, α_j is the dimensionless Stark-shift parameter for a given beam (see (16)), $\beta_\pm$ — the kind of dimensionless mixing parameter of two different beams, and $\left(1 \rightleftarrows 2, n \rightleftarrows m, l \rightleftarrows l+1\right)$ in (A1) denotes the term obtained from the preceding one by the indicated interchange of the indices at x_j, y_j, $f_j\left(t\right)$, α_j and J_q.

References

[1] A. Plucińska, R. Parzyński, *J. Mod. Opt.* **54**, 745 (2007).

[2] Y.V. Rostovtsev, H. Eleuch, A. Svidzinsky, H. Li, V. Sautenkov, M.O. Scully, *Phys. Rev. A* **79**, 063833 (2009).

[3] M.O. Scully, M.S. Zubairy, *Quantum Optics*, Cambridge University Press, Cambridge 1997.

[4] R. Parzyński, T. Kopyciuk, *Laser Phys. Lett.* **15**, 075206 (2018).

[5] R. Parzyński, T. Kopyciuk, *Acta Phys. Pol. A* **137**, 1080 (2020).

[6] F. Zamani, M. Miri, *Opt. Commun.* **423**, 207 (2018).

[7] A.V. Bogatskaya, A.M. Popov, *Laser Phys. Lett.* **17**, 096002 (2020).

[8] P. Földi, I. Magashegyi, Á. Gombköto, S. Varró, *Photonics* **8**, 263 (2021).

[9] D.G. Zill, *Differential Equations with Boundary-Value Problems*, Prindle, Weber and Schmidt, Boston 1986.

[10] L.S. Gradshteyn, I.M. Ryzhik, *Table of Integrals, Series and Products*, Academic Press, London 2000.

[11] M. Abramowitz, I.A. Stegun, *Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables*, Dover, New York 1964.

[12] M.Y. Ivanov, P.B. Corkum, *Phys. Rev. A* **48**, 580 (1993).

[13] N. Tancogne-Dejean, A. Rubio, *Sci. Adv.* **4**, eaao5207 (2018).

[14] Ó. Zurrón, A. Picón, L. Plaja, *New J. Phys.* **20**, 053033 (2018).

[15] L. Hareli, G. Shoulga, A. Bahabad, *J. Phys. B At. Mol. Opt. Phys.* **53**, 233001 (2020).