Schemes for the generation of circularly polarized high-order harmonics by two-color mixing

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Tong and Chu [Phys. Rev. A **58**, R2656 (1998)] recently suggested a scheme of how to produce circularly polarized high harmonics. It is compared to an alternative scheme, and some characteristic differences between the two are discussed. [S1050-2947(99)09608-0]

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Recently, Tong and Chu (TC) [1] proposed a scheme for the production of circularly polarized high-harmonic radiation. They suggest a crossed-beam experiment focusing into the gas chamber a linearly polarized laser field with frequency 2ω and at a right angle to the former a circularly polarized field with frequency ω. The setup implies that the linear polarization is perpendicular to the plane of the circular polarization. Provided the ground state has zero angular momentum (as is the case for rare gases), dipole transition selection rules dictate that linearly polarized harmonics with frequency $(2n+1)2\omega$ are radiated in the direction of the incident linearly polarized field and circularly polarized harmonics with frequency $(2n+1)\omega$, in the direction of the incident circularly polarized field. TC confirmed these ideas with calculations employing time-dependent density-functional theory. The results displayed the expected circularly polarized harmonics. However, within the plateau their intensities come out significantly weaker than the (linearly polarized) harmonics of the pure one-color 2ω field.

A different color-mixing scheme was suggested and implemented some time ago and predicted to produce circularly polarized harmonics [2]. In this experiment two parallel circularly polarized laser beams of frequencies ω and 2ω were used such that the electric-field vectors of the two beams rotate in the same plane, either in the same or in the opposite direction. In the corotating case, all harmonic frequencies $n\omega$ are emitted. Their intensities were observed to drop quickly with increasing harmonic order without any indication of a plateau. Theory predicts their polarization to be elliptic. In the counter-rotating case, on the other hand, selection rules only allow for the emission of harmonics with frequencies $(3n\pm1)\omega$ and purely circular polarization of alternating helicity. These harmonics were observed and found to exhibit a plateau. In fact, out of various polarization schemes that were being considered the counter-rotating polarizations provided the most intense signal. These experiments were modeled by calculations based on the zero-range model [3], which produced qualitative agreement with the data. In the experiment, only the harmonic intensities were recorded. No attempt was made to confirm the circular polarization of the harmonics.

It is illuminating to write down the field-induced dipole moments of either scheme. In what follows we will assume infinite plane waves for all driving fields. In TC's scheme the driving field has the vector potential

$$\mathbf{A}(t)=\left[\frac{a_L}{2}\mathbf{e}_3 e^{-2i\omega t}+\frac{a_c}{\sqrt{2}}\mathbf{e}_+ e^{-i(\omega t+\phi)}\right]+\text{c.c.}, \tag{1}$$

where $\mathbf{e}_\pm=(\mathbf{e}_1\pm i\mathbf{e}_2)/\sqrt{2}$. It produces a time-dependent dipole moment with frequency components (at frequency Ω)

$$\begin{aligned}\mathbf{d}(\Omega)=&\frac{a_L}{2}\mathbf{e}_3\sum\ \delta(\Omega-(2n+1)2\omega)d_{L,n}(a_L^2,a_c^2)\\&+\frac{a_c}{\sqrt{2}}\mathbf{e}_+e^{-i\phi}\sum\ \delta(\Omega-(4n+1)\omega)d_{+,n}(a_L^2,a_c^2)\\&+\frac{a_c}{\sqrt{2}}\mathbf{e}_-e^{i\phi}\sum\ \delta(\Omega-(4n-1)\omega)d_{-,n}(a_L^2,a_c^2).\end{aligned} \tag{2}$$

The functions $d_{i,n}$ depend on the model used for the description of the atom [4]. For a zero-range atomic potential they can be written down in the form of one-dimensional quadratures.

On the other hand, the two parallel incident counter-rotating circular polarizations, specified by the vector potential

$$\mathbf{A}(t)=\frac{1}{\sqrt{2}}[a_1\mathbf{e}_-e^{-i(\omega t+\phi)}+a_2\mathbf{e}_+e^{-2i\omega t}]+\text{c.c.}, \tag{3}$$

generate the dipole components [3]

$$\begin{aligned}\mathbf{d}(\Omega)=&\frac{a_1}{\sqrt{2}}\mathbf{e}_-e^{-i\phi}\sum\ \delta(\Omega-(3n+1)\omega)e^{-in\phi}d_{n11}\\&+\frac{a_1}{\sqrt{2}}\mathbf{e}_+e^{i\phi}\sum\ \delta(\Omega-(3n-1)\omega)e^{-in\phi}d_{n12}\\&+\frac{a_2}{\sqrt{2}}\mathbf{e}_+\sum\ \delta(\Omega-(3n+2)\omega)e^{-in\phi}d_{n21}\\&+\frac{a_2}{\sqrt{2}}\mathbf{e}_-\sum\ \delta(\Omega-(3n-2)\omega)e^{-in\phi}d_{n22}.\end{aligned} \tag{4}$$

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The dipole moment contains two series of frequencies: $(3n+1)\omega$ and $(3n-1)\omega$, with negative and positive helicity, respectively. Closer inspection of Eq. (4) reveals that the spectral intensities do not depend on the phase ϕ. The dipole strengths d_{nij} depend on a_1^2, a_2^2, and $a_1 a_2$ [4]. The way we wrote the dipole component (4) illustrates the connection between harmonic frequency and helicity enforced by selection rules: If the atom absorbs, for counter-rotating circular polarizations, one 2ω photon and one ω photon, its energy is raised by 3ω while its magnetic quantum number does not change [cf. Eq. (3)]. In order to be able to emit the harmonic photon by a dipole transition, the atom has to absorb or emit one additional circularly polarized photon of either component of the driving field. If, for example, it emits an ω photon or absorbs a 2ω photon the harmonic frequency is $(3n-1)\omega=[3(n-1)+2]\omega$ and the helicity given by $\mathbf{e}_+$. The fact that pairs of photons of each field have to be absorbed is reflected in the pronounced dependence of the function d_{nij} on the product $a_1 a_2$.

An important practical question is the effect of imperfect circular polarization of the driving fields on the polarization of the harmonics. The scheme of TC involves the net absorption (or emission) of just one circularly polarized photon while the scheme with the incident counter-rotating circular polarizations depends on net absorption of n pairs of $(\omega, 2\omega)$ circularly polarized photons plus absorption or emission of one photon of either frequency, as discussed above. Therefore, one may expect that the TC scheme is less sensitive to imperfect incident circular polarization. However, this question can only be definitely settled by a calculation allowing for elliptic polarization for *all* incident fields or, even better, by experiment.

It is interesting to compare the presence or absence of a plateau structure in the spectra of the circularly polarized harmonics in either scheme. Owing to the fact that the two components of the field (1) are perpendicular, the functions d_{ni} in the TC dipole moment (2) only depend on the squares of the field amplitudes with no mixed term proportional to $a_L a_c$ [4]. Consequently, one expects the plateau of the linearly polarized harmonics also to dominate the circularly polarized spectrum, which indeed the calculations show it does. As pointed out by TC, there is a certain intensity range of the incident circularly polarized field, which leads to the highest intensity of the circularly polarized harmonics that can be achieved by their scheme. Raising the intensity of the incident circularly polarized field beyond this range is counterproductive. This is easily understood in terms of the simple man's model: the stronger the circularly polarized component of the driving field becomes, the less able is the electron to return to the ionic core and to recombine. The scheme of the two counter-rotating circular polarizations also produces a plateau, according to both the data and the accompanying calculations reported in Ref. [2]. The physical origin of this plateau is, however, not clear. The paradigm of high-harmonic emission—recombination of the returning electron with the parent ion—does not apply in this case. Inspecting the classical electronic orbits in the presence of the applied two-color field, one quickly realizes that only very few of them (provided they start with zero velocity from the position of the parent ion) will ever return. While the overlap of the electronic wave packet with the ion still allows for some harmonic generation, this process is known to be inefficient in the case of a one-color elliptically polarized laser field [5]. In contrast, harmonic generation in the counter-rotating case is very efficient. Further work is required in order to understand the mechanism of harmonic emission in this case.

The requirements for an experimental realization of the two schemes are quite different. The counter-rotating scheme requires that both driving fields are as close to plane waves as possible in order that both fields really rotate in the same plane. This implies weak focusing. Also, the admissible deviation from perfect circular polarization of the two driving fields is likely to be low. Realization of the TC scheme is hampered by the low spatiotemporal overlap of the crossed beams and by the lack of phase matching caused by the perpendicular wave vectors of the two beams. Moreover, in this case, the generation of a circularly polarized photon, as it follows from momentum conservation, requires a large momentum transfer to the atom.

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[4] The notation used in Eqs. (2) and (4), viz. $d_{ni}(a_L^2, a_c^2)$ or $d_{nij}(a_1^2, a_2^2, a_1 a_2)$ etc., actually refers to the dependence of the *classical action* that underlies the quantum calculation upon the indicated variables; see, e.g., Ref. [3] or M. B. Gaarde, A. L'Huillier, and M. Lewenstein, Phys. Rev. A **54**, 4236 (1996); D. B. Milošević and B. Piraux, *ibid.* **54**, 1522 (1996).

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