Crossed-beam experiment: High-order harmonic generation and dynamical symmetry

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The selection rules for the harmonic generation within the two-color crossed-beam scheme proposed by Tong and Chu [Phys. Rev. A **58**, R2656 (1998)] can be readily proved on the basis of the dynamical symmetry of the Floquet Hamiltonian by Alon, Averbukh, and Moiseyev [Phys. Rev. Lett. **80**, 3743 (1998)], represented within the dipole approximation. [S1050-2947(99)04609-0]

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In a recent rapid communication [1], Tong and Chu have proposed a two-color crossed laser-beam scheme for the production of circularly and linearly polarized high harmonics by atoms. The circularly polarized harmonic frequencies, $\Omega_n = n\omega$, generated within the authors' scheme obey the unusual selection rule: $n = 4k \pm 1$. It was rationalized by the authors on the basis of the dipole selection rules with the reference only to the leading channel of the high-order harmonic generation by the multiphoton mechanism. The purpose of the present paper is to show that this selection rule can be proved in a straightforward manner by regarding the dynamical symmetry of the Floquet Hamiltonian of the crossed beam setup. In fact, the $4k \pm 1$ selection rule is a specific case of the $Nk \pm 1$ selection rule derived by Alon, Averbukh, and Moiseyev for a general many-electron system possessing a dynamical symmetry of the order N [2]. While the original work of Tong and Chu reveals several aspects of the *dynamics* of He atoms interacting with the crossed laser beams, our paper is meant to complement only the *symmetry* aspect of the problem.

Consider the Floquet Hamiltonian of a He atom interacting with crossed laser beams (the circularly polarized beam of angular frequency ω and the other beam linearly polarized along the propagation direction of the first one and possessing the angular frequency 2ω) within the dipole approximation,

$$\hat{\mathcal{H}}_f = \frac{\hbar}{i}\frac{\partial}{\partial t} + \sum_{j=1,2}\left[\frac{\hat{P}^2_{\rho_j} + \dfrac{\hat{P}^2_{\varphi_j}}{\rho_j^2} + \hat{P}^2_{z_j}}{2m} - \frac{2e}{\sqrt{\rho_j^2 + z_j^2}} + eE_\omega \rho_j \cos(\varphi_j - \omega t) + eE_{2\omega} z_j \cos(2\omega t)\right] + \frac{e^2}{\sqrt{\rho_1^2 + \rho_2^2 - 2\rho_1\rho_2\cos(\varphi_1 - \varphi_2) + (z_1 - z_2)^2}}. \quad (1)$$

The above Floquet Hamiltonian is invariant under the following fourth-order dynamical symmetry operator:

$$\hat{P}_4 = \left(\varphi_{1,2} \to \varphi_{1,2} + \frac{\pi}{2}, z_{1,2} \to -z_{1,2}, t \to t + \frac{\pi}{2\omega}\right), \quad \hat{P}_4^4 = \hat{I} \quad (2)$$

(note that the Floquet Hamiltonian of an atomic system interacting with two electric fields of frequencies ω and 2ω and phase difference ϕ possesses a similar fourth-order symmetry if regarded as a function of ϕ [3]).

As follows from the general discussion in Ref. [2], the nth harmonic is emitted by the system if the operator of its amplitude is invariant under the dynamical symmetry operator [Eq. (2)].

The harmonic amplitude operators chosen to commute with dynamical symmetry operator [Eq. (2)] are

$$\hat{f}_{\pm}^{(n)} = \sum_{j=1,2} \rho_j e^{i(\pm\varphi_j - n\omega t)}, \quad \hat{f}_z^{(n)} = \sum_{j=1,2} z_j e^{-in\omega t}. \quad (3)$$

An elementary calculation shows that $f_{\pm}^{(n)}$ is invariant under $\hat{P}_4$ *iff* $n = 4k \pm 1$, while $f_z^{(n)}$ is invariant under $\hat{P}_4$ *iff* $n = 4k + 2$. Thus the selection rules found by Tong and Chu are proved.

The proof of selection rules given above relies on the exact symmetry of the Floquet Hamiltonian and is not limited to any specific region of the parameter values, e.g., the value of the Keldysh parameter. It should be noted, however, that in the experiments employing high-intensity lasers one deals with pulses of finite length. If the pulse supports a sufficient number of the field oscillations, Floquet theory can be still applicable even for the calculation of the relative harmonic intensities and not only for the derivation of the selection rules (see Refs. [4,5] for the numerical examples). In the numerical calculations reported by Tong and Chu the laser pulse was assumed to be 90 optical cycles long. Apparently, for such a pulse length the symmetry analysis based on Floquet theory is perfectly valid.

In conclusion, the two-color crossed-beam setup suggested by Tong and Chu brings about the formation of the atom—time-dependent fields system possessing (within the dipole approximation and in the infinitely long pulse limit) the fourth-order dynamical symmetry. This dynamical symmetry accounts for the selection rules found by the authors. One can consider the more general case in which the frequency of the linearly polarized field is $N\omega$. In such a case the Floquet Hamiltonian is invariant under the following $2N$th-order dynamical symmetry operator:

$$\hat{P}_{2N}=\left(\varphi_{1,2}\rightarrow\varphi_{1,2}+\frac{\pi}{N},z_{1,2}\rightarrow -z_{1,2},t\rightarrow t+\frac{\pi}{N\omega}\right),\quad \hat{P}_{2N}^{2N}=\hat{I}. \tag{4}$$

The symmetry analysis of the harmonic amplitude operators Eq. (3) gives in this case the following selection rules for the emitted harmonics: $n=2Nk\pm 1$ and $n=N(2k+1)$, corresponding to $f_{\pm}^{(n)}$ and $f_{z}^{(n)}$, respectively. It implies that for $N>2$ the harmonics generated as a sum of both circularly and linearly polarized fields (see Fig. 2 of Tong and Chu [1]) are not all the odd harmonics of ω. On the other hand, the harmonic generated by the linearly polarized field would still only contain all the odd harmonics of the incident frequency $N\omega$.

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