Retrieval of target photorecombination cross sections from high-order harmonics generated in a macroscopic medium

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Synopsis We investigate high-order harmonic generation (HHG) in a thin macroscopic medium by solving Maxwell's equation using microscopic single-atom induced dipole moment calculated from the recently developed quantitative rescattering (QRS) theory. We show that macroscopic HHG yields calculated from QRS compared well with those obtained from solving the single-atom time-dependent Schrödinger equation but with great saving of computer time. We also show that macroscopic HHG can be expressed as a product of a macroscopic wave packet and the photorecombination cross section of the target gas. The latter enables us to extract target structure from the experimentally measured HHG spectra, thus paves the way to use few-cycle infrared lasers for time-resolved chemical imaging of transient molecules with few-femtosecond temporal resolution.

In recent years high-order harmonic generation (HHG) by a strong infrared laser field interacting with a gas of atoms has been widely used for the production of subfemtosecond pulses in the extreme ultraviolet (XUV) radiation. The physical origin of the harmonics emission in a single atom can be easily described by a three-step model [1, 2]. Since HHG is generated by a focused laser beam over all the atoms in a macroscopic medium, the induced dipole moment on each atom should be inserted as a source term in the propagation equations of the harmonic field to obtain the macroscopic response of the excited nonlinear medium. Thus a typical HHG calculation consists of two parts: first, the calculation of single-atom response; second, the propagation of Maxwell's wave equation. Due to the limitation of strong field approximation (SFA) for the description of single-atom response, we have proposed a quantitative rescattering (QRS) theory recently [5]. The QRS is about as fast as the SFA, and the accuracy of it has been carefully tested against single-atom HHG spectra obtained by solving the time-dependent Schrödinger equation (TDSE). With QRS-based single-atom induced dipole moments, the macroscopic HHG spectra are nearly as accurate as those from TDSE based and are much better than SFA based. The macroscopic HHG yield can be expressed as the product of a "macroscopic wave packet" (MWP) and the single-atom recombination dipole moment [6].

In our simulation we assume that there is no ionization effect of the medium on the fundamental laser field. This condition can be satisfied when we consider low intensity laser, low density and short gas medium. The propagation equation of harmonic field in the moving coordinate frame is [3, 4]

$$\nabla_\perp^2 \widetilde{E}_h(r,z',\omega) - \frac{2i\omega}{c}\frac{\partial \widetilde{E}_h(r,z',\omega)}{\partial z'} = -\mu_0\omega^2 \widetilde{P}_{nl}(r,z',\omega). \quad (1)$$

The macroscopic HHG spectra are obtained by

$$S_h(\omega) \propto \int_0^\infty |\widetilde{E}_h(r,z',\omega)|^2 2\pi r dr. \quad (2)$$

Electric field of laser pulse in t'' frame is

$$E_1(r,z',t'') = |\varepsilon(r,z')|\cos^2(\frac{\pi t''}{\tau_p})\cos(\omega_0 t'' + \varphi_{CE}). \quad (3)$$

Nonlinear polarization term in moving coordinate frame can be obtained by

$$\widetilde{P}_{nl}(r,z',\omega) = \widehat{F}[P_{nl}(r,z',t'')]e^{-i\frac{\omega}{\omega_0}\varphi_{laser}(r,z')}. \quad (4)$$

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