High-Order Nonlinear Dipole Response Characterized by Extreme-Ultraviolet Ellipsometry

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Abstract: We demonstrate that polarization control and characterization of high-harmonic generation (HHG) in non-collinear geometry performs as an excellent ellipsometry that can fully retrieve the amplitude and phase of ultrafast dipole response, advancing high harmonic spectroscopy.

Characterization of the amplitude and phase of attosecond emissions, reflecting the single dipole response of high-harmonic generation (HHG), as a function of photon energy is essential for high harmonic (HH) spectroscopy. Moreover, the complex dipole response influences the wavefront, the pulse duration, and phase-matching of attosecond pulses. Very recently, we also found the HH dipole phase is the key to realize the polarization control of isolated attosecond pulses, while keeping the up-conversion efficiency in non-collinear HHG geometry [1]. However, the relation between the polarization control of HHG and its nonlinear complex dipole response is still unclear.

Our recent work shows that the polarization control scaling \( \left( \varepsilon_{\text{EUV}} \propto \varepsilon_{\text{IR}}^\sigma \right) \) varies differently depending upon not only the harmonic order but also the gas species [2]. The ellipticity scaling coefficient \( \sigma \) becomes much larger for higher-order harmonics and especially in high-Z atoms with low ionization potentials. Ellipsometry quantitatively points out the polarization control scaling vary differently because not only the effective order of nonlinearity \( q_{\text{eff}} \) is large, but also the intensity-dependent phase coefficients \( \alpha_q \) is larger in high harmonic orders, especially in low-ionization-potential atoms. The potential future applications of ellipticity-tunable attosecond XUV pulses, combined with characteristic polarization control should allow us gain insight into quantum dynamics, ultrafast chirality changes, and circular dichroism in atoms, molecules, and solids.

We generated HHG in Ar, Kr and Xe using 35 fs Ti:Sapphire laser in non-collinear geometry. To control polarization of high harmonic pulses, two elliptically polarized fundamental beams with the same ellipticity but opposite helicity are focused into a gas jet non-collinearly (see Fig. 1). On the focal plane, two fundamentals interferingly form local E-field vectors, inducing dipole oscillations rotating crossing the transverse direction. Each location with linear polarization acts as a high-order harmonics local emitter and superposes a pair of elliptically polarized UV beams in the far-field. As a result, the ellipticity of attosecond pulses \( \varepsilon_{\text{EUV}} \) can be well controlled by fundamental ellipticity \( \varepsilon_{\text{IR}} \), while keeping a good beam profile as well as the up-conversion efficiency. We found the tilt angle and \( \varepsilon_{\text{EUV}} \) of each harmonic order scale differently, as shown in Fig. 1(b), obtaining a harmonic-order dependent ellipticity scaling of \( \varepsilon_{15\text{th}} \propto \varepsilon_{12.5\text{th}} \), \( \varepsilon_{17\text{th}} \propto \varepsilon_{15\text{th}} \) and \( \varepsilon_{21\text{st}} \propto \varepsilon_{15\text{th}} \) in Kr, while keeping the up-conversion efficiency.

Fig. 1: (a) Schematic of the HHG ellipsometry that can fully retrieve the complex dipole response of HHG. (b) The polarimeter shows the tilt angle \( \tau \), and \( \varepsilon_{\text{EUV}} \) of each harmonic scale differently vs. \( \varepsilon_{\text{IR}} \). Here we only plot results driven by fundamentals with \( \varepsilon_{\text{IR}} = 0.9 \).
To get information of atomic dipole response from the polarimeter measurement, we developed a model which is known to provide good qualitatively results for HHG in thin gas jet. When focusing two identical elliptically polarized IR beams with equal ellipticity but opposite helicity, the local HH emission in x- and y-directions can be given by

\[
E_{EUV,x}(x,y) = I_{IR}(x,y)^{q_{eff}/2} e^{i[q_{eff}a^\Gamma(y)\alpha]} \frac{E_{IR,x}(x,y)}{\sqrt{I_{IR}(x,y)}} \theta^Z
\]

\[
E_{EUV,y}(x,y) = I_{IR}(x,y)^{q_{eff}/2} e^{i[q_{eff}a^\Gamma(y)\alpha]} \frac{E_{IR,y}(x,y)}{\sqrt{I_{IR}(x,y)}} \theta^Z
\]

where \(I_{IR}(x,y) = |E_{IR,x}(x,y)|^2 + |E_{IR,y}(x,y)|^2\) representing the local intensity. The Eq. (1) and (2) is based on the dipole response of q order harmonic as a function of the laser field having the form of \(E_{EUV}(I_{IR}) \propto A_q(I_{IR}) \exp\left(i\theta_q(I_{IR})\right)\), which can be well approximated as \(E_{EUV}(I_{IR}) \propto I_{IR}^{q_{eff}/2} \exp\left(-i\alpha a_q^\Gamma\right)\) where \(A_q(I_{IR}) \equiv I_{IR}^{q_{eff}/2}\) is the amplitude of the q order harmonic, \(q_{eff}\) is an effective order of nonlinearity, and \(\theta_q(I_{IR}) \equiv -I_{IR} a_q^\Gamma\) is the phase of the q order harmonic [2-4]. The coefficient \(a_q^\Gamma\) depends highly on the trajectory type (short or long), the harmonic order \(q\) and gas species. We found the short quantum path emission was predominant in this polarization control study, therefore \(a_q^\Gamma = a_q^\Gamma\). The far-field response is obtained by considering an infinitely thin medium and by summing the contributions of all atomic emission located on the generation plane as Huygens–Fresnel principle. The coherent sum of Equ.(1) and (2) for x- and y-polarizations on the focal plane results in two main diffraction EUV spots with the ellipticity \(\varepsilon_{EUV}\) and the tilt angle \(\tau\). In other words, two unknown microscopic parameters, \(q_{eff}\) and \(a_q^\Gamma\), can be extracted by the observed EUV ellipticity \(\varepsilon_{EUV}\) and the tilt angle \(\tau\) as presented in Fig. 1(b).

![Fig. 2: Dipole response information extracted from HH ellipsometry. (a) Power scaling, \(q_{eff}\) versus harmonic order \(q\), extracted from Ar (green), Kr (blue), and Xe (red). Green stars represent the calculated \(q_{eff}\) in Ar obtained from quantum SFA theory, as described in [5]. This calculation uses a peak intensity, \(1.2 \times 10^{14}\) W/cm², less than the \(2.8 \times 10^{14}\) W/cm² applied in HH ellipsometry. (b) Phase-intensity slope \(\alpha\) versus harmonic order \(q\). Light strips mark \(\alpha\) calculated from the semiclassical model (as indicated in [5]), using intensities 70% and 80% of their average peak intensities.](image)

In summary, we demonstrated that polarization control and characterization of HHG in a noncollinear geometry provides an excellent ellipsometry tool able to unveil the quantum dynamics of HHG samples under intense laser fields. HHG ellipsometry can measure two parameters—the ratio of intensities and phase difference information—compared with the single parameter of regular HH spectroscopic techniques, that is, only intensity without phase. Both the amplitude and phase of the HH dipole were precisely characterized using HHG ellipsometry, carrying detailed information about the evolution of an ionized electronic wave packet. This study also clearly demonstrated how the amplitude and phase of the nonperturbative HHG dipole influenced the power scaling of HHG and the achievable HH energy. Prospective future applications of HH ellipsometry should allow us to precisely quantify quantum dynamics, ultrafast chirality changes, and circular dichroism in HHG samples, as well as to enable critical comparisons with theory.

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