Comment on "Observation of Attosecond Light Localization in Higher Order Harmonic Generation"

In a recent publication [1] the first measurement of attosecond (as) light structures created by high harmonic generation (HHG) was claimed, based on the following method. A Ti:S laser pulse ($\tau = 60$ fs and $I_0 = 3.4 \times 10^{14}$ W/cm^2) was split into two parts with a ratio of peak intensities of 1:0.95. Both pulses were focused into Ar gas, and the energy of the harmonics generated between orders $N = 27$ and 33 close to the cutoff was measured as a function of the delay between the two pulses. It was observed that the first derivative of the harmonic energy with respect to the delay exhibits structures on an attosecond time scale. The way the authors present their results suggests that the observed as-variation is a signature of the as-duration of the harmonic pulses. Here we show that the experiment [1] in its present form cannot prove the existence of as-pulses.

We performed a numerical analysis of the experiment of Papadogiannis *et al.* [1] for the above parameters. HHG is determined by a calculation of the single atom dipole moment $d(t)$ from Eqs. (34)–(39) in Ref. [2]. Our calculation reveals an as-modulation of the harmonic energy and of its first derivative qualitatively similar to the one reported in Ref. [1]. However, since the measurement of Papadogiannis *et al.* involves a complex, nonlinear interaction, it is not obvious whether, or in which way, the observed as-modulation is related to the duration of the harmonic pulse. In Ref. [1] no compelling answer to this crucial question is given.

The following simple test could clarify the origin of the observed sub-fs modulation. We propose to repeat the experiment of Ref. [1] with the only difference being the use of a narrower spectral window, e.g., between $N = 26$ and $N = 28$. As this spectral width cannot support an as-pulse, the as-variation of the harmonic energy must disappear, if it is connected to the pulse duration. In case the as-modulation does not vanish, it must arise from a mechanism insensitive to the harmonic pulse duration.

The importance of the experiment proposed above is supported by a numerical analysis. In Figs. 1(b) and 1(c) we have plotted the harmonic energy contained in the spectral interval between $N = 26$ and $N = 28$ and its first derivative versus delay between the two laser pulses. All other parameters were chosen equal to those of the experiment in Ref. [1]. Although the harmonic pulse is a few fs long, [see Fig. 1(a)] the derivative of the harmonic energy still exhibits attosecond features (periodic on the scale of half an optical period of the laser pulse) similar to the ones reported in Ref. [1]. This result raises concerns regarding the validity of the as-pulse measurement performed by Papadogiannis *et al.,* and emphasizes the necessity of further experiments putting the origin of the observed as-modulation on more solid grounds.

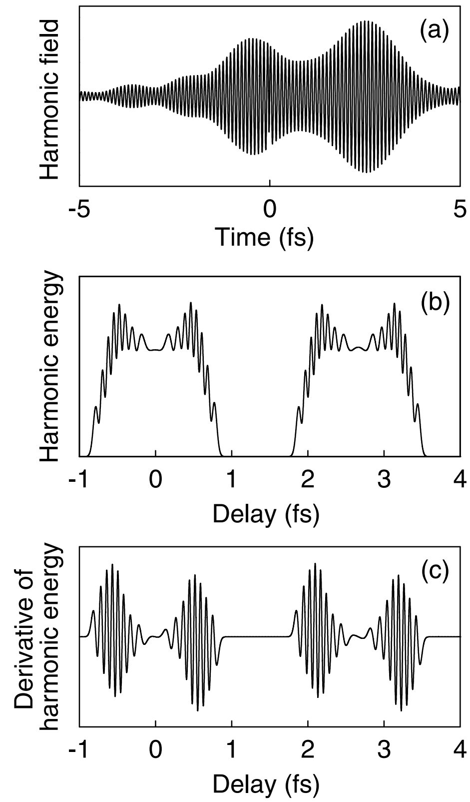

FIG. 1. Numerical calculation of HHG for the parameters used in the experiment of Papadogiannis *et al.,* with the difference that instead of the harmonic signal between $N = 27$ and $N = 33$ the narrower spectral band between $N = 26$ and $N = 28$ is used for our calculation of the harmonic energy and its first derivative. The laser pulses are modeled by a sech-pulse shape. (a) Harmonic field in time corresponding to the spectral window of the single atom dipole moment between $N = 26$ and $N = 28$. (b) Harmonic energy contained in the interval between $N = 26$ and 28, and (c) its first derivative as a function of delay between the two laser pulses.

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