# Vectorial Phase Retrieval for Linear Characterization of Attosecond Pulses

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The waveforms of attosecond pulses produced by high-harmonic generation carry information on the electronic structure and dynamics in atomic and molecular systems. Current methods for the temporal characterization of such pulses have limited sensitivity and impose significant experimental complexity. We propose a new linear and all-optical method inspired by widely used multidimensional phase retrieval algorithms. Our new scheme is based on the spectral measurement of two attosecond sources and their interference. As an example, we focus on the case of spectral polarization measurements of attosecond pulses, relying on their most fundamental property—being well confined in time. We demonstrate this method numerically by reconstructing the temporal profiles of attosecond pulses generated from aligned $CO_2$ molecules.

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Optical and extreme ultraviolet (XUV) pulses with durations significantly below one picosecond cannot be directly characterized in the time domain, since there are no suitable photodetectors. Therefore they are usually characterized in the frequency domain by measuring both the spectral amplitude and spectral phase of the pulse. The former may be determined straightforwardly by means of a spectrometer. The latter requires either a fast modulator, a detector, or a reference pulse with which the unknown pulse can be interfered. A modulator or gate of sufficiently rapid response is typically synthesized by means of nonlinear optical processes [1]. In the femtosecond regime, most measurement schemes are based on nonlinear light matter interactions. By exploiting media having a nonlinear response, various successful schemes have been developed for complete characterization of femtosecond pulses, most notably FROG [2] and SPIDER [3].

The ability to produce attosecond pulses has set new benchmarks in time-resolved measurements. Such pulses enable one to probe electron dynamics on the atomic time scale. Recently, it has been demonstrated that the attosecond production process carries information about both electron dynamics [4] and molecular structure [5]. However, full access to the information contained in the pulse requires its complete characterization. A direct implementation of the pulse characterization schemes developed for the femtosecond regime is challenging, due to the low signal levels and the absence of appropriate nonlinear media in the XUV.

Several characterization schemes have been developed for the attosecond XUV domain [6–11]. However, time-resolved measurement of attosecond pulses remains a major challenge. For example, certain important attosecond-scale processes such as plasma mirrors [12] and field-enhancement high harmonic generation (HHG) [13] have not yet been fully characterized.

In this Letter we propose a new approach for measuring attosecond pulses, which, in contrast with other methods, relies only on linear spectral measurements of the radiation generated by several sources. The key new feature of the method that enables this approach is the explicit utilization of a temporal support constraint in the retrieval algorithm. The fact that the pulse is limited in duration allows a unique solution to be extracted. To illustrate the approach, we consider the two polarization components of the attosecond pulse as two independent sources. We show that the spectral measurement of the two polarizations together with their relative phases, obtained by means of spectral interference, is sufficient to retrieve the spectral phase of each component individually. We numerically test the method on attosecond pulses calculated for aligned $CO_2$ molecules [14], in which the polarization varies nontrivially with frequency. We establish excellent reconstructions even for pulses that do not exactly satisfy the finite duration assumption, but rather have a sufficiently rapid decay.

Phase retrieval problems are common in many branches of physics, including astronomy [15], NMR [16], ultrafast optics [2], crystallography [17], and lensless imaging [18–20]. In each of these cases, the measured signal spectrum, together with some assumption on the signal itself, allows one to retrieve the phase without directly measuring it. In our case, the assumed property is that the pulse has a limited, but not necessarily known, ''time window'' in which the intensity is nonzero, usually referred to as compact support. Generic compact support phase retrieval

 

problems are known to have many solutions in the one-dimensional (1D) case, but usually a unique solution for higher dimensions [21]. As our measurements are of a one-dimensional Fourier transform, the compact support assumption is not enough to reconstruct the spectral phase. This is solved by measuring the relative phase between two (or more) spectra. Such a measurement enables a vectorial 1D phase retrieval problem. If the two components of the vector are nondegenerate (in a sense to be defined later on), then the problem has a unique solution for the spectral phase, up to a phase linear in frequency (i.e., an arbitrary delay that is not physically significant). It is useful to consider how the temporal support constraint enables a unique inversion of the data. We first develop a formal approach that illustrates this, and then describe a simpler inversion algorithm.

Consider the electric field spectrum $E(\omega)$ of a pulse with a finite duration $T$, sampled at frequencies $\omega_j = 2\pi j/T$ for $j = 1, \ldots, N$. The spectrum can be written as $E(\omega) = \sum_t \hat{E}(t) e^{-i\omega t} = \sum_t \hat{E}(t) z^t$, where $z = e^{-i\omega}$. According to the fundamental theorem of algebra, we can write $E(z) = \sum_t E(t) z^t = \prod_j (z - z_j)$ where $z_j$ are the $N$ roots of the polynomial $E(z)$. A linear measurement of the spectrum measures $|E(\omega)|^2$, which at the sampled points $|z| = 1$ can be shown to equal

$$|E(z)|^2 = \frac{\prod \overline{z_j}}{(-z)^N} \prod_j (z - z_j)(z - \overline{z_j}^{-1}), \tag{1}$$

where $\overline{z_j}$ is the complex conjugate of $z_j$. Without any prior knowledge about the pulse, the spectral phase is, by definition, unrecoverable. This is because the polynomial representing the spectrum is undersampled: $E(\omega)$ corresponds to a polynomial of degree $N$ in $z$, but $|E(\omega)|^2$ corresponds to a polynomial of degree $2N$ in $z$. Therefore, the $N$ samples of $|E(z)|^2$ are insufficient to unambiguously determine $\hat{E}(t)$. If, however, a compact support constraint is assumed, meaning $\hat{E}(t) = 0$ for, say, the $N/2$ samples in the range $T/2 \le t \le T$, the degree of $|E(z)|^2$ is at most $N$. Therefore, it is well sampled and the $N$ roots of the polynomial ($z_j$ and $\overline{z_j}^{-1}$) are uniquely determined. This also agrees with the Nyquist criterion for sampling compact supported pulses. The compact support constraint, nevertheless, does not remove all the ambiguities. In order to retrieve the pulse $E(z)$, only one root from each pair of roots, $z_j$ and $\overline{z_j}^{-1}$, should be chosen. As there are $2^{N/2}$ possibilities to choose one root from each pair of $|E(z)|^2$, there still exist $2^{N/2}$ different pulses, all having both the measured spectrum and the correct compact support. This ambiguity in the 1D phase retrieval problem is well known [21]. Hence, additional information is required to unequivocally determine the ''correct'' choice.

The phase ambiguity can be overcome by using two (or more) spectra which we refer to as ''components,'' and the relative phase between them. We will denote such measurements as ''vectorial.'' As an example of a vectorial measurement, appropriate for the attosecond domain, we use polarization: when attosecond pulses are generated from an anisotropic media, such as aligned molecules by means of high-harmonic generation, nontrivial frequency-dependent polarization is expected [22]. Other examples are discussed in the Supplemental Material [23]. For the two component spectra, $|E_x(\omega)|^2$ and $|E_y(\omega)|^2$ with the same time domain compact support, we can find the $N$ roots $\{z_x\}_j$, $\{z_y\}_j$ corresponding to Eq. (1) and their complex conjugates. Without the relative phase, these are two independent 1D phase retrieval problems, each having many solutions. However, a complete polarization measurement, consisting of both the spectra of two orthogonal polarizations as well as the interference spectrum between them, provides sufficient information to eliminate the ambiguities. Noting that $E_{x,y}(\omega) = |E_{x,y}(\omega)| e^{i\phi_{x,y}(\omega)}$, the vectorial measurements allow us to establish $|E_x(\omega)|^2$, $|E_y(\omega)|^2$, and $E_x(\omega)\overline{E_y(\omega)} = |E_x||E_y| e^{i(\phi_x - \phi_y)}$. These quantities are represented by the factored polynomials

$$|E_x(z)|^2 = \frac{\prod \overline{z_j^x}}{(-z)^N} \prod_j (z - z_j^x)(z - \overline{z_j^x}^{-1}) \tag{2}$$

$$|E_y(z)|^2 = \frac{\prod \overline{z_j^y}}{(-z)^N} \prod_j (z - z_j^y)(z - \overline{z_j^y}^{-1}) \tag{3}$$

$$E_x(z)\overline{E_y(z)} = \frac{\prod \overline{z_j^y}}{(-z)^N} \prod_j (z - z_j^x)(z - \overline{z_j^y}^{-1}) \tag{4}$$

The essential point is that from these equations the ''which root'' ambiguities of both components can be resolved: the correct roots for $E_x(z)$ are those that are common to $|E_x(z)|^2$ and $E_x(z)\overline{E_y(z)}$, and similarly for $E_y(z)$. In Fig. 1, we show an example of the method for a simple pulse. The time domain signals are shown in the inset. In the main figure, the roots generated by Eqs. (2) and (3) are shown as dots and crosses in the complex plane (the black line is the unit circle). As expected, the roots come in pairs: for example, the roots marked by $A$ and $B$ are related by $z_A = \overline{z_B}^{-1}$. The one-dimensional phase retrieval problem requires one to choose the correct root from each pair. In the vectorial case, this can be done using the roots generated by Eq. (4): these roots are marked by green squares. As seen in Fig. 1, each square coincides with only one of the roots, thus identifying the correct root of each pair. In the above example, $A$, rather than $B$, is the correct root, as it coincides with a root of Eq. (4).

Clearly, if $|E_x(z)|^2$ and $|E_y(z)|^2$ have $m$ common zeros, there is still an ambiguity among $2^m$ different pulse shapes. We will call such cases degenerate. In the absence of noise, the set of pulses which share common zeros is very small, unless they have been manipulated in a common manner to

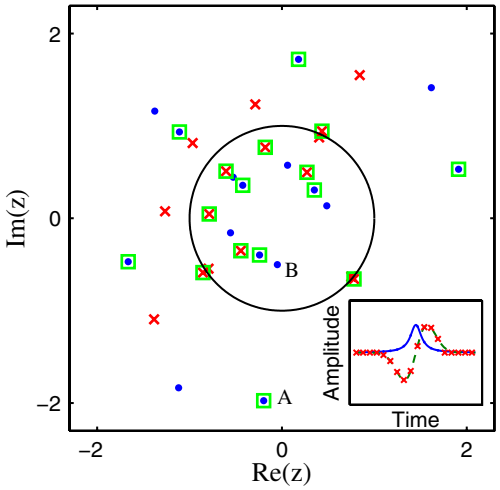


FIG. 1 (color online). An example for our reconstruction procedure for two artificially generated pulses with the same compact support. The blue dots, red crosses, and green squares are the roots, in the complex plane, of $|E_x(z)|^2$,$|E_y(z)|^2$, and $E_x(z)\overline{E_y(z)}$, respectively. A time domain plot of the two polarizations is shown in the inset.

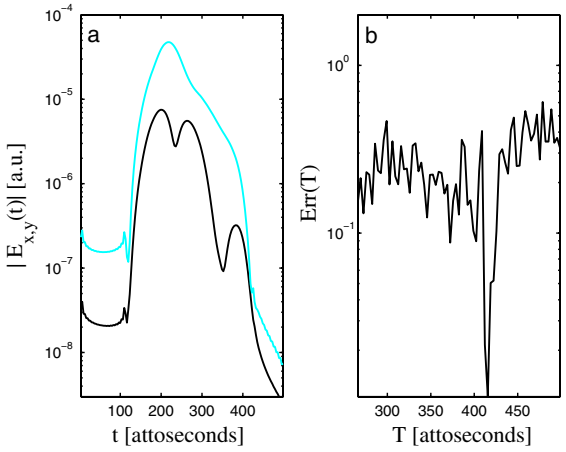


FIG. 2 (color online). (a) Two time domain polarization components of the attosecond pulse generated from $CO_2$ molecules at an alignment of 58° (between the molecule symmetry axis and the IR polarization) and IR laser intensity of 0.07 a.u. at 800 nm. (b) Err($T$) [defined in Eq. (8)] as a function of assumed compact supports.

introduce them. Nevertheless, common zeros do arise when both pulses are passed through the same linear phase-only filter. The pulses then share a component of common spectral phase, leading to degeneracy. In this case the pulse fields cannot be retrieved by the above method (see Supplemental Material [23]). Further, most experimental data will have some noise, which creates uncertainty around the roots and, hence, might generate a degeneracy. As we show in the Supplemental Material [23], by sampling large enough bandwidth the uncertainty around the zeros gets smaller with respect to the distance between them; hence, degeneracy should not be commonplace.

Another consideration is that most physical pulses do not have exact compact support, but rather a sharp decay outside some temporal region. Our method is applicable even in such cases, as we now show by means of an alternative solution method. The compact support constraints can be expressed as a set of nonlinear equations for the spectral phase $\phi_{x,y}(\omega)$, which are the only unknowns in the problem. These equations, however, can be viewed as a linear set of equations for $X(\omega) = e^{i\phi_x(\omega)}$. To see this, we write $\hat{E}_x(t)$ and $\hat{E}_y(t)$ by using inverse discrete Fourier transform, as

$$\hat{E}_x(t) = \sum_\omega E_x(\omega)e^{i\omega t} = \sum_\omega |E_x(\omega)|e^{i\omega t}X(\omega), \quad (5)$$

$$\hat{E}_y(t) = \sum_\omega |E_y(\omega)|e^{i\omega t}e^{i(\phi_y(\omega)-\phi_x(\omega))}X(\omega). \quad (6)$$

Applying the compact support assumption gives

$$\hat{E}_x\left(t = 1, \ldots, \frac{N}{2}\right) = \hat{E}_y\left(t = 1, \ldots, \frac{N}{2}\right) = 0. \quad (7)$$

Using Eq. (7) in Eqs. (5) and (6) gives a set of $N$ linear homogenous equations for the $N$ unknowns $X(\omega)$. Exploiting the arbitrariness of the absolute phase, one can set $X(\omega_1) = 1$ and obtain an overdetermined set of inhomogeneous equations for the unknown phases $X(\omega)$, which for nondegenerate problems have a unique solution. By solving these equations, one can solve the phase problem provided $X(\omega)$ is a phase-only function, that is, $|X(\omega)| = 1$ for all $\omega$. This has two important consequences: (1) In cases where the solution to Eq. (7) is far from yielding a unimodular complex number, the compact support assumption is either wrong (as the case of assuming compact support smaller than the true one) or there is a degeneracy (as, for example, when the compact support domain is chosen smaller than the true one). As we later show, this gives us the means to search for the correct compact support without assuming it beforehand. (2) Since the problem boils down to solving linear equations, the sensitivity to noise is linear in the noise amplitude.

Our proposed algorithm seeks a domain of compact support choosing that which is most consistent with the above conditions as our constraint. For each assumed domain of support, $T$, we resample the spectral information at discrete frequencies with spacing $\Omega = 2\pi/T$, changing the number of sampled frequencies $N$. We then use Eq. (7) in Eqs. (5) and (6) to find $X(\omega)$. For each $T$ we calculate how far $X(\omega)$ are from being unimodular complex, by calculating the relative change in the pulse's energy when using $X(\omega)$ (which might not be unimodular) as the spectral phase:

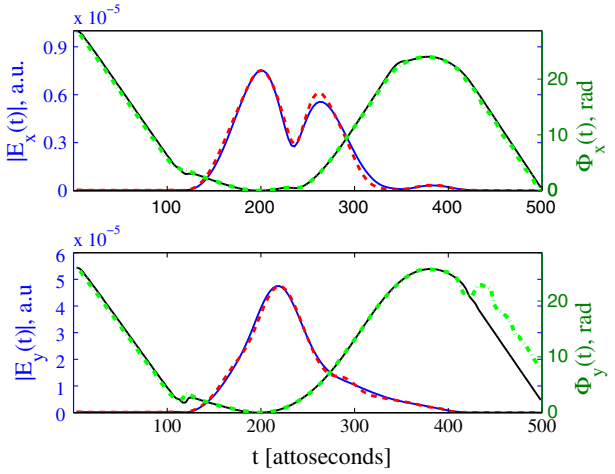


FIG. 3 (color online). Comparison between original (amplitude, blue; phase, black) and retrieved (amplitude, dashed red; phase, dash-dotted green) pulses, for the two polarizations of the HHG pulse generated from the $CO_2$ molecule at an alignment of 58° (between the molecule symmetry axis and the IR polarization) and IR laser intensity of 0.07 a.u. at 800 nm. The spectral resolution of the data was $3.9 \times 10^{14}$ Hz. The lower figure shows the radiation in the IR polarization, and the upper shows the perpendicular polarization. The assumed pulse duration $T$ which minimizes Err($T$) is 415 attoseconds (from $t = 0$ to $t = 415$), which corresponds to downsampling from $N = 769$ to $N = 250$.

$$\mathrm{Err}\,(T) = \frac{\sum_\omega |E(\omega)|^2 |(1 - |X(\omega)|)|^2}{\sum_\omega |E(\omega)|^2} \tag{8}$$

Err($T$) is the standard metric used in the Gerschberg-Saxton algorithm [15] for compact support phase retrieval problems. We then choose the value of $T$ that minimizes Err($T$) as the compact support domain and the argument of the corresponding $X(\omega)$ as the pulse's phase. We demonstrate our method by reconstructing simulated attosecond pulses generated from aligned $CO_2$ molecules (for HHG simulation details, see [14]). The simulated attosecond pulses do not have compact support, but exhibit a sharp Gaussian decay (see Fig. 2, where the two original pulse polarizations are plotted in a logarithmic scale). Figure 3 shows the temporal profiles of the two polarizations of the simulated pulse and their reconstruction. As can be seen, their agreement is excellent. The errors are typically of the order of Err($T$) $\sim 0.05$. A plot of Err($T$) is given in Fig. 2. For too small $T$ the solution is far from being unimodular as the compact support assumption cannot be fulfilled. For too large $T$ there are many solutions which correspond to lateral shifts. Hence, the system is degenerate, and the arbitrary solution found by the algorithm is not necessarily unimodular.

There are many experimental ways to implement the method developed here. So far we have discussed the spectrally dependent polarization components of HHG which can be measured, using an XUV polarizer, up to a sign ambiguity of the phase difference. The sign ambiguity can be resolved by means of a wave plate. For many cases this is not needed as by continuity the ambiguity is the same for all $\omega$, which means an overall time direction ambiguity. Other options to use the same idea (see Supplemental Material [23]) are to generate two XUV pulses from different sources, either spatially [4,24] or by using mixed gases [25]. By measuring the spectrum of each source alone and the interference between them, one can use our method to reconstruct the spectral phase. The spatial case can be generalized to spatiotemporal measurements by lateral shearing interferometry [26].

To conclude, we have proposed and demonstrated a novel characterization method for attosecond pulses using a vectorial phase retrieval algorithm. Our method presents a new class of solutions of phase retrieval problems, applicable to many other fields, such as lensless imaging and optical spectroscopy. The main strength of the method lies in the fact that it removes the requirement for a nonlinear interaction or photoionization to resolve attosecond processes. Extending our approach to characterize more complex electron dynamics proposes a new scheme of time-resolved measurements where attosecond-scale phenomena can be observed using linear, time-stationary, apparatus.

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