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Use of Electron Correlation to Make Attosecond Measurements without Attosecond Pulses

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We describe how correlations between electrons can be used to trace the dynamics of correlated two-electron ionization with attosecond precision, without using attosecond pulses. The approach is illustrated using the example of Auger or Coster-Kronig decay triggered by photoionization with an extreme ultraviolet pulse. It requires correlated measurements of angle-resolved energy spectra of both the photo- and Auger electrons in the presence of a laser pulse. To reconstruct the dynamics, we use not only classical time and energy correlation, but also entanglement between the two electrons.

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In conventional pump-probe spectroscopy, ultrafast time resolution is achieved by using pump and probe pulses much shorter than the duration of the process one wants to measure. A short pump is used to avoid the need for complex deconvolution of the process with the pulse that initiates it. Similarly, a short probe takes a snapshot of the process at a desired instant in time.

We show how, in a pump-probe measurement, particle correlation can be used to time-resolve a fast process triggered by a much slower pump pulse and probed with a much slower probe pulse. Correlation and/or entanglement can be exploited for enhanced measurements in a variety of situations [1], from photon down-conversion [2] to attosecond measurements [3].

Here, complete characterization of the process is done by reconstructing the amplitude and phase of the correlated two-electron spectrum. Phase information is obtained in a manner similar to the SPIDER (spectral shearing interferometry for direct electric field reconstruction) method of [4]: the phase is mapped onto an amplitude modulation of the spectral intensity by recording the interference of the original spectrum with its spectrally-shifted replica. Particle correlation allows us to effectively solve the deconvolution problem, uncovering the fast component of the correlated process. One essential requirement is temporal stability of the probe pulse relative to the pump: their relative jitter degrades time resolution. Fortunately, modern few-cycle infrared (IR) femtosecond pulses can be phase-stabilized with attosecond precision over long times, naturally leading to attosecond stabilization of XUV (extreme ultraviolet) pulses which they generate [5].

Our approach can be used for any process resulting in the emission of two charged particles with fixed total energy. Important examples are shakeoff in one-photon/two-electron ionization, photoinduced Auger or Coster-Kronig decay, etc. Ultrafast stages of particular interest in such processes can include Zeno and anti-Zeno stages of decay [6], core rearrangement, nonexponential decay due to structured continuum, etc.

As an illustration, we consider the case of Auger or Coster-Kronig decay induced by one-photon ionization. A schematic of the process is shown in Fig. 1. An XUV-photon with energy Ω is absorbed by an atom in the ground state (energy E_g), promoting a core electron to a continuum state with energy $E_X = k_X^2/2$ and leaving an ion in an autoionizing state with energy E_h (core vacancy). Triggered by the first ionization event, autoionization follows, filling the core vacancy. This creates a doubly charged ion and liberates the second electron with kinetic energy $E_A = k_A^2/2$.

Fast Auger decay implies that the autoionizing state E_h has a large width. Consequently, energy of the Auger electron E_A has a large uncertainty (broad spectral line). However, the total energy of both electrons is defined by the energy of the absorbed photon:

$$E_A + E_X = k_A^2/2 + k_X^2/2 = \Omega - I_p^{++}, \quad (1)$$

FIG. 1 (color online). (a) Energy diagram of the Auger process: absorption of XUV photon Ω creates a free photoelectron k_X and a hole state E_h which autoionizes to create the second free electron k_A . (b) Linear contour plot of the field-free correlated two-electron spectrum. Energy is measured in units of laser frequency ω ; $E_X^{(0)}$, $E_A^{(0)}$ are the central energies of the photoelectron and the Auger spectra.

where I_p^{++} is the energy of removing two electrons from the neutral atom. Equation (1) implies that the uncertainty in E_A translates into an equal uncertainty in E_X , Fig. 1. Correlated spectra of such shape appear whenever a two-particle conservation law is present; their general properties for enhanced measurements were studied in [7].

In the time domain, the process is a convolution of the Auger decay with temporal dependence $F_A(t)$ and the XUV pulse $\mathcal{E}_X(t)e^{-i\Omega t}$ that creates the core vacancy. Consequently, the correlated two-electron spectral amplitude $C^{(0)}(k_X, k_A)$ of the process is the product of the spectral amplitude of the Auger decay $\tilde{F}_A$ and the Fourier image $\tilde{E}_{\text{XUV}}$ of the envelope $\mathcal{E}_{\text{XUV}}(t)$ of the XUV pump with carrier frequency Ω [8]:

$$C^{(0)}(k_X, k_A) \propto \tilde{E}_{\text{XUV}}\left(\frac{k_X^2}{2} + \frac{k_A^2}{2} - \Omega + I_p^{++}\right) \tilde{F}_A\left(\frac{k_A^2}{2} - E_A^0\right) \quad (2)$$

where E_A^0 is the central energy of the Auger line. The argument of $\tilde{E}_{\text{XUV}}$ expresses energy conservation Eq. (1).

The bandwidth of the XUV pulse, which determines the uncertainty of the sum energy of both electrons, can be significantly narrower than the spectral line for each of the two electrons measured separately, i.e., integrating over all final states of the other electron [see Fig. 1(b)]. While we may know the energy Ω put into the system, without the correlated measurement we do not know how it was shared between the two liberated electrons. On the other hand, measuring the energy of the photoelectron E_X defines the energy of the Auger electron E_A within the bandwidth of the XUV pulse.

To find the law of the Auger decay $F_A(t)$ we need to find both the amplitude $|\tilde{F}_A|$ and phase Φ_A of its Fourier transform $\tilde{F}_A$. While the Auger spectrum can be measured with conventional spectroscopy, the measuring phase is less trivial. To find Φ_A we need to reconstruct the spectral phase $\Phi(k_X, k_A)$ of the two-electron amplitude $C^{(0)}(k_X, k_A)$. Then, knowing the spectral phase Φ_{XUV} of the XUV pulse we will be able to recover Φ_A since $\Phi = \Phi_{\text{XUV}} + \Phi_A$. Finding Φ_A in the full range of the Auger electron energies implies deconvolution of the spectrally wide Auger process from a spectrally narrow pump. In practice, outside the narrow pump bandwidth, the convolution spectrum is buried under noise. The correlated spectrum allows us to circumvent this problem.

Interference permits us to record the phase by mapping it onto an amplitude modulation. Let us apply a linearly polarized low-frequency laser field $\mathcal{E}_L \cos(\omega t)$ which does not affect the Auger decay itself but induces absorption of laser photons once an electron appears in the continuum. Absorption of a photon by one of the electrons creates a replica of the original spectrum shifted by the photon energy $\hbar\omega$. Its interference with the original spectrum maps the spectral phase onto amplitude modulation. We restrict the laser intensity to the perturbative regime

where the laser-dressed spectrum is dominated by the original (unshifted) spectrum and two replicas corresponding to absorption and/or emission of one laser photon, similar to the regime used in [9,10] for reconstructing attosecond pulse trains from one-electron spectra.

To simplify the phase reconstruction, we select a geometry in which only one of the two electrons has non-negligible probability of absorbing a photon from the laser field. We minimize the effect of the probing laser field on the photoelectron and maximize its effect on the Auger electron. This is done by detecting the photoelectron in the direction perpendicular to the field and the Auger electron in the direction parallel to the field. In this geometry, the energy of the photoelectron interaction with the laser field is $\mathcal{A}_L^2/4$, where $\mathcal{A}_L = \mathcal{E}_L/\omega$ is the amplitude of the field vector potential. The same energy for the Auger electron is $k_A \mathcal{A}_L \gg \mathcal{A}_L^2/4$. Selecting laser intensity $I \sim 10^{10}$ W/cm² and wavelength $\lambda = 800$ nm ensures $N_X = \mathcal{A}_L^2/4\omega \sim 4 \times 10^{-4}$, making absorption of photons by the photoelectron negligible. The resulting amplitude is [8]

$$C(k_X, k_A) = \sum_m (-1)^m e^{-im\omega\Delta t} J_m\left(\frac{k_A \mathcal{A}_L}{\omega}\right) \times C^{(0)}(k_X, \sqrt{k_A^2 - 2m\omega}), \quad (3)$$

where m is the number of absorbed photons and Δt is the delay between the peak of the XUV pump and the instantaneous maximum of the laser field $\mathcal{E}_L \cos\omega t$. Equation (3) shows that the probing laser field creates interference of the replicas of the field-free spectrum Eq. (2) spaced by ω ; see Fig. 2. For this illustrative figure we used a transform-limited Gaussian XUV pulse with full width at half maximum (FWHM) duration of 1.77 fsec and a single exponential Auger decay $F(t) = \exp(-\Gamma t)$ with $1/\Gamma = 210$ asec. Reconstruction of indirect double-exponential decay will be considered below. To make the interference more visible, in Fig. 2 we have used a higher intensity of $I = 10^{11}$ W/cm². For the actual phase reconstruction we use $I = 10^{10}$ W/cm², so that the probability $J_1^2(k_A \mathcal{A}_L/\omega)$ of absorbing one photon from the IR laser field is about 1% for $k_A^2/2 \approx 40$ eV.

The width of each sideband is defined by the width of the XUV pulse [see Eqs. (2) and (3)]. For the sidebands to overlap, the spectral width of the XUV pulse should be about $\hbar\omega$. For a single slice of the two-electron spectrum with fixed E_X [Fig. 2(b)] the region where the signal is nonzero is much smaller than the width of the Auger line for this slice. We are facing the standard difficulty of the deconvolution—reconstructing information from a low signal, which in practice is buried under noise.

The correlated two-electron distribution allows one to circumvent this problem. By selecting a different energy E_X of the photoelectron, one takes a different slice of the correlated spectrum, where the signal is nonzero in a different interval of energies. Concatenating slices [ladder

FIG. 2 (color online). (a) Correlated two-electron spectrum $|C(k_x, k_A)|^2$ in the presence of the laser field. E_X^0, E_A^0 are the central energies of the field-free photoelectron spectrum and the Auger line. (b) Slice for fixed energy of the photoelectron.

in Fig. 2(a)], one reconstructs the spectral phase in the full region of energies.

For each slice, the problem of phase reconstruction is no different than for a one-electron spectrum generated by a short XUV pulse in the presence of an IR laser field. Many methods have now been developed, including those suited for the perturbative regime, see, e.g., [9,10]. They typically use a sequence of interference patterns recorded while changing the delay of the XUV pump relative to the IR probe. Given that correlated measurements require long data collection times, we need a method which uses only few interferograms. We have used a version of the two-sideband reconstruction method [11] which requires only four time delays ($\omega\Delta t = 0, \pi/2, \pi, 3\pi/2$). This method uses interference in the region where the original line overlaps with only one of the two sidebands. For example, the region near $E_A - E_A^0 = \omega$ in Fig. 2(b) contains interference between the main line and the right sideband, with negligible contribution from the left sideband. There Eq. (3) yields the spectrum:

$$|C|^2 = J_0^2|C_0|^2 + J_1^2|C_1|^2 + J_0J_1C_0^*C_1e^{-i\omega\Delta t} + \text{c.c.} \quad (4)$$

Here arguments of Bessel functions are the same as in Eq. (3) and $C_m \equiv C^{(0)}(k_x, \sqrt{k_A^2 - 2m\omega})$. Equation (4) shows the analogy of this method to SPIDER [4]: information about phase differences between the two sidebands is recorded in the interference term $C_0^*C_1 \propto \exp(i\Delta\Phi)$, $\Delta\Phi = \Phi(E_A - \omega) - \Phi(E_A)$. Using four different delays we retrieve the phase difference in this spectral region:

$$\tan(\Delta\Phi) = \frac{|C(k_X, k_A, 0)|^2 - |C(k_X, k_A, \frac{\pi}{\omega})|^2}{|C(k_X, k_A, \frac{3\pi}{2\omega})|^2 - |C(k_X, k_A, \frac{\pi}{2\omega})|^2}. \quad (5)$$

For one-electron spectra, the two-sideband method yields reliable phase reconstruction only in a narrow spectral interval. For correlated spectra, this is not a problem—adjacent spectral slices for different E_X give access to adjacent spectral regions. For each slice we reconstruct the phase only in the best-suited spectral region, improving

the stability of the method with respect to noise in the data. For the Auger line, stability of phase reconstruction is further improved by the redundancy of the data on Φ_A in the two-electron spectrum. Indeed, we can choose different “ladders” of slices leading us from top to bottom of the spectrum in Fig. 2.

In femtosecond chemistry, the advent of ultrashort pulses has allowed the time-resolved multistage dynamics of dissipative processes (e.g., dissociation) characterized by the rising and falling parts in the decay law. In analogy, let us consider a model of an indirect Auger decay via an intermediate state [Fig. 3(a)]. The XUV pump excites a state $|1\rangle$ coupled to an intermediate state $|2\rangle$ (with the same energy) by the matrix element V_{12} . The state $|2\rangle$ is coupled to the continuum by the matrix element V_{2C} . For the system prepared in the state $|1\rangle$, the problem is mathematically identical to multiphoton ionization via exact resonance [12]. The decay law has rising and falling parts: $F_A(t) = [1 - \exp(-\Delta\Gamma t)]\exp(-\Gamma_2 t)$, where $\Gamma_{1,2} = \Gamma(1 \pm [1 - 4V_{12}^2/\Gamma^2]^{1/2})/2$, $\Gamma \equiv \pi|V_{2C}|^2$, and $\Delta\Gamma = \Gamma_1 - \Gamma_2$ [12]. If the system starts in state $|g\rangle$, the decay law is a convolution of $F_A(t)$ and the XUV pump. We set $1/\Delta\Gamma = 106$ asec

FIG. 3 (color online). (a) Model of the decay via an intermediate state $|2\rangle$. (b) Exact (solid line) and reconstructed decay for $N = 10^5$ counts (circles), $N = 10^6$ counts (empty circles), and 100 asec pump-probe jitter.

FIG. 4 (color online). Correlated spectra (a),(b) and reconstructed Auger lines in the frequency domain (c),(d) for $N = 10^5$ and $N = 10^6$ counts and 100 asec jitter. In (a),(b) shades of gray (color) code spectral intensity on a linear scale, with lighter color coding showing the higher signal (except for the high central spot). In panels (c),(d) the exact spectral phase is shown with a smooth solid line.

and $1/\Gamma_2 = 425$ asec. Using the XUV pulse with $\text{FWHM} = 1.2$ fsec we will resolve both the long tail of $F_A(t)$ and the fast rise associated with the presence of the intermediate state [2].

Let us show how reconstruction of this process works in the presence of measurement errors and noise associated with data statistics, finite resolution of the electron spectrometer, and jitter of the XUV pulse relative to the oscillation of the probe. To simulate the experiment, we set the jitter of the XUV pulse to 100 asec and the energy resolution of the electron spectrometer to 160 meV. Included in the simulation are finite count statistics leading to shot noise. The results are shown in Fig. 3(b).

For the total number of coincident counts $N = 10^5$, the reconstruction error is about 20–30 asec, better than the 100-asec jitter of the pump. The reduction of error is due to the random nature of the jitter and the redundancy of information about the Auger phase Φ_A in the two-electron spectrum. We utilize the redundancy by concatenating different ladders of slices such as in Fig. 2. For $N = 10^6$ counts, the error is below 10 asec [Fig. 3(b)]. The buildup of the electron spectrum is shown in Fig. 4 together with the frequency-domain reconstruction results for $N = 10^5$ and $N = 10^6$ counts. Note, that phase reconstruction at the edges is most important for the rising part of the time-dependent signal, since this rising part leads to 2π rather than π phase shift across the spectrum.

To some extent, our technique resembles the method of reconstructing a broad spectrum using its interference with a well characterized narrow spectral window which can be moved across the broad spectrum. In conventional optics, this technique has been used in [13].

In conclusion, correlated measurements enhance temporal resolution in pump-probe experiments, offering a route to solving the deconvolution problem and allowing one to use the redundancy of information recorded in the correlated spectrum. The intrinsic time resolution comes from the fast process itself. For a correlated spectrum, measuring both amplitude and phase implies recovering the entangled wave function. Using the interference of the wave function with its energy-shifted replica implies using the entangled wave function to characterize itself. In practice, achieving attosecond time resolution requires attosecond temporal stability of the XUV pump relative to the IR probe field. When a single XUV pump is generated by the same IR field which is used as a probe, as in the pioneering experiment [14], attosecond control over the generating IR field provides the required temporal stability. An attosecond temporal ruler is already present in the femtosecond pulse. Our approach shows how to access it.

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