Ramsey’s method for Auger electron interference induced by an attosecond twin pulse

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We examine the archetype of an interference experiment for Auger electrons: two electron wave packets are launched by inner-shell ionizing a krypton atom using two attosecond light pulses with a variable time delay. This setting is an attosecond realization of Ramsey’s method of separated oscillatory fields. Interference of the two ejected Auger electron wavepackets is predicted indicating that the coherence between the two pulses is passed to the Auger electrons. For the detection of the interference pattern an accurate coincidence measurement of photo- and Auger electrons is necessary. The method allows one to control inner-shell electron dynamics on an attosecond time scale and represents a sensitive indicator for decoherence.

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Ramsey’s method of separated oscillatory fields [1, 2] represents a paradigm of precision measurement for various physical quantities. In its original conception for the measurement of nuclear magnetic moments, the scheme uses two coherent radiation fields, which are separated by a field-free propagation interval. The signature of the coherent interaction is the appearance of interference fringes when the physical quantity under consideration is measured at the exit of this experimental setup. Since Ramsey’s seminal studies, his method has been extended and modified extensively, e.g., by considering multiple fields with varying phase and amplitude and by applying it to masers and lasers [1].

In this paper, we propose a Ramsey scheme for attosecond science assuming two coherent pulses with a FWHM duration of \( \tau_X = 500 \) as each, which are separated by a variable delay of \( \tau \) [Fig. 1]. With an essential-states model [4, 5], we investigate the situation where a twin pulse that ionizes the 3d shell of krypton atoms which subsequently decay in terms of an \( M_{4,5},N_1,N_2 \),3 Auger process. In fact one of the first applications in attosecond science was the determination of the time constant of this Auger decay channel—a well-known datum from frequency-domain spectroscopy \( \tau_{3d} = 7.5 \) fs—with a single attosecond pulse in the presence of an optical streaking laser [6]. Unlike this first investigation, our proposal of an attosecond Ramsey method [Fig. 1] represents a fundamental experiment which is only feasible with attosecond science and has no frequency domain equivalent. Furthermore, Auger decay [8] is a pure manifestation of electron correlations: it cannot be understood in terms of an effective one-electron model. Additionally, the method of separated oscillatory fields is an interferometric approach which has the advantage over pump-probe schemes that it does not depend on an intense pump pulse.

The twin pulse shown in Fig. 1 induces two outgoing photoelectron and Auger electron waves. Interference between two photoelectron wavepackets was examined in Ref. [9]. In that study, 5p Rydberg electrons of potassium atoms were subjected to two identical time-delayed laser pulses with a FWHM duration of 30 fs each at 790 nm wavelength and the resulting interference pattern in the photoelectron spectrum was analyzed. In contrast to the experiment in Ref. [9], we ask whether the coherence of the light is also transferred to the Auger electrons and what kind of Auger electron spectrum can we expect if it is? Clearly, for a time delay between the two pulses of \( \tau = 0 \) and of \( \tau \to \infty \), we observe no interference fringes. What happens in between the two limiting cases? Clearly, our proposal of an attosecond Ramsey scheme represents an important experimental test of our understanding of Auger decay. Further, the scheme also represents a versatile tool for measurements. It enables one to precisely determine the position of Auger lines and it is a measure of coherence. Such an experiment would also address the following questions: how much decoherence is caused by the Auger process and what is the coherence length? Is our understanding of Auger
decay complete?

The simplest way to describe Auger decay is a rate-equation model for the krypton 3d hole population [3]. The solution at time $t$ is the convolution of exponential Auger decay with a width of $\Gamma_A$ with the XUV intensity

$$\rho(t) = \frac{\sigma}{\omega_X} \int_{-\infty}^{t} I_X(t') e^{-\Gamma_A(t-t')} \, dt'.$$

Here, the cross section $\sigma$ is taken to be constant over the bandwidth of the XUV pulse with a central angular frequency of $\omega_X$ and an intensity of $I_X(t')$ at time $t'$. The population induced by the twin pulse and its decay is also shown in Fig. 1. The model does not honor the phase relationship between the two ejected Auger electron wave packets and thus does not take into account account phase relations, which are crucial for the description of interference effects [4].

To address the issue of coherence, we use an ab initio formalism for the quantum dynamics of Auger decay by laser-dressed atoms, which are inner-shell ionized by extreme ultraviolet (XUV) light [5]. The attosecond pulses of present-day light sources have a low peak intensity and its interaction may be described perturbatively as a one-photon process [2]. The quantum dynamics of the inner-shell hole creation with subsequent Auger decay is given by equations of motion (EOM), which we simplify here in terms of an essential-states model [2, 3] and solve analytically. In doing so, we assume only one final dicatication state. Our theory yields the probability density amplitude $\tilde{c}_A^E \tilde{E}_A(\tau)$ to find a photoelectron with momentum $\vec{k}_P$, in coincidence with an Auger electron with momentum $\vec{k}_A$ for a delay of $\tau$ between the two pulses in Fig. 1. The probability density amplitude is adapted for no laser dressing from Eqs. (61) and (62) of Ref. [3]; it reads

$$\tilde{c}_A^E \tilde{E}_A(\tau) = \frac{1}{2} \tilde{d}(\vec{k}_P) \tilde{v}(\vec{k}_A) S(\tau, \vec{k}_P^2 / 2, \vec{k}_A^2 / 2),$$

with the rms dipole and rms Auger decay matrix elements $\tilde{d}(\vec{k}_P)$ and $\tilde{v}(\vec{k}_A)$, respectively. The line shape function in Eq. (2) is

$$S(\tau, \omega_P, \omega_A) = \frac{\tilde{\varepsilon}_X(\tau, \omega_P + \omega_A - \Omega_A - \Delta_R + \frac{i}{2})}{\omega_A - \Omega_A - \Delta_R + i \frac{\pi}{4}};$$

it depends only on the absolute values of the momenta $k_P = |\vec{k}_P|$ and $k_A = |\vec{k}_A|$. Further, it contains the nominal photoelectron and Auger electron energies, which are in our case $\Omega_A = 20 \, \text{eV}$ and $\Omega_A = 40 \, \text{eV}$, respectively [2]. In Eq. (3), $\Delta_R$ is the second-order energy shift and $\Gamma$ is the Auger decay width [3]. The spectral envelope of the XUV light for a twin pulse is given by

$$\tilde{\varepsilon}_X(\tau, \omega) = \sqrt{\frac{\pi}{2 \log 2}} \varepsilon_{X0} \tau \varepsilon e^{-\frac{\omega^2 \tau^2}{8 \varepsilon^2 \tau^2}} (1 + e^{i \omega \tau}),$$

with the peak electric field strength $\varepsilon_{X0}$.

The main result of this study, the probability density $|\tilde{c}_A^E \tilde{E}_A(\tau)|^2$ [Eq. (2)], is displayed in Fig. 2. We will return to it several times in what follows to discuss the Auger electron spectra induced by a twin pulse. The figure shows $|\tilde{c}_A^E \tilde{E}_A(\tau)|^2$ for no time delay between the two pulses of Fig. 1 i.e., a single pulse, and a time delay of 5 $\tau_{3d}$. We see clearly that the spectrum with the two separated pulses is structured, which indicates interference effects that we would like to analyze and characterize in the following. The shape of the plots in Fig. 2 is determined by the absolute square of the line shape function [3] in a non-trivial way. Horizontally, along the $k_P^2 / 2$ coordinate, the width of the line profile is determined by the FWHM of $|\tilde{\varepsilon}_X(\tau, \omega)|^2$, which in our case is 3.7 eV; it is maximal for $\vec{k}_A^2 / 2 = \omega_A = \Omega_A$ for which the white area in the plot has a roughly 10 eV spread, so about three times the FWHM in the chosen color scale. Vertically, along the $k_A^2 / 2$ coordinate, we have a more involved dependence, which will be discussed in conjunction with Eq. (4) below; overall, the contour has the shape of a skewed hyperbola with respect to $k_A$ caused by the denominator squared in Eq. (3).

In a typical Auger electron spectroscopy experiment, the photoelectron is not observed. Hence we need to integrate the joint probability over the unobserved degree of freedom, which is in this case the photoelectron momentum $\int |\tilde{c}_A^E \tilde{E}_A(\tau)|^2 \, d^3k_P$. For each Auger electron momentum, this implies an integration along a horizontal line in Fig. 2. Following such a path in the right panel visually, we see that we average over many fringes with different energies leading to a washing out of the interference pattern. Indeed, the resulting Auger electron spectrum exhibits no noticeable fringes for an unknown photoelectron momentum; it resembles closely the green curve in Fig. 2.

Inspecting the line shape function [3], we find that the
xuv envelope is imprinted on the Auger electrons due to correlations between photoelectrons and Auger electrons \(4, 5, 10\). The finding that integrating over the photoelectron diminishes interference effects conforms to the general fact that summing over unobserved degrees of freedom generally comes with a loss of coherence. Consequently, to preserve the coherence of the Auger decay process, we need to take the photoelectron into account.

The other way round, however, does not hold true: it is not required to observe the Auger electron to see interference fringes in the photoelectron spectrum \(5\). To see why this is so, we derive the probability density to observe a photoelectron. Within our formalism \(5\) it is given by

\[
\tilde{P}_P(\tilde{k}_P) = \frac{|d(\tilde{k}_P)|^2}{2\pi} \int_{-\infty}^{\infty} \text{Im} \left[ \frac{\tilde{\varepsilon}_X(\tau, \omega)|^2}{\tilde{k}_P - \Omega_P + \Delta_R - \omega - i\frac{\Delta}{2}} \right] \text{d}\omega .
\]

(5)

Our analysis has revealed the interconnection of the photoionization and the subsequent Auger decay; the dependence of Eq. \(5\) on the Auger decay width represents the reciprocal connection. This can be understood as follows: the interference of the photoelectrons is caused by the envelop \(\tilde{\varepsilon}_X\) of the xuv light. The only impact of Auger decay on the photoelectron is due to the filling of the created hole which leads to a line broadening which leads for large Auger widths to a washing out of interference fringes.

To have a chance of observing interference between Auger electrons, we need to preserve the coherence of the Auger electron wavelets from the two xuv pulses. This can be achieved by realizing that the emission of an Auger electron with a momentum \(\tilde{k}_A\) enters in Eq. \(6\). Relation \(6\) is only exact for monochromatic xuv light with photon energy \(\omega_X\), i.e., a continuous wave source. Inspecting Eq. \(3\) we see that the energy balance \(6\) is reflected by the diagonal lines in the right panel.

Next, we integrate \(|\tilde{\varepsilon}_A(\tilde{k}_A)|^2\) over the full solid angle and a specified photoelectron momentum range \(\Delta k\). This yields the joint probability \(P_{A,\Delta k}(\tau, k_P, k_A)\) to observe an Auger electron with \(k_A\) —we look along the z axis—for a photoelectron in a solid angle of \(4\pi\) with a momentum magnitude in the range of \([\max\{0, k_P - \Delta k\}; k_P + \Delta k]\). By integrating Eq. \(6\) over the same angular and momentum range, we find the normalized probability distribution of the photoelectrons \(P_{A,\Delta k}(\tau, k_P)\): if we observe the photoelectron to lie in a chosen range, then the conditional probability to find an Auger electron with a specific momentum along the z axis \(P_{A,k_P,\Delta k}(\tau, k_A)\) follows from Bayes law \(11\):

\[
P_{A,\Delta k}(\tau, k_A) = \frac{P_{\tau, A, \Delta k}(\tau, k_P, k_A)}{P_{\tau, \Delta k}(\tau, k_P)} .
\]

(7)

In Fig. 3 we investigate the Auger electron spectrum from Eq. \(7\). For a very accurate measurement of the photoelectron momentum (small uncertainty in \(\Delta k\)), we find significant interference fringes. The interference effects are diminished with growing \(\Delta k\), i.e., we average over Auger waves in the energy range \([\max\{0, k_A - \Delta k\}^2; (k_A + \Delta k)^2]\). Furthermore, the first maximum off the main peak in the curves of Fig. 3 moves to higher energies with increasing \(\Delta k\).

The experimental investigation of the be propose is challenging because a coincident detection of two electrons is necessary. In a recent experimental study \(11\), electrons from the KVV Auger decay of a C 1s vacancy in a CO molecule were measured in coincidence with the angular distribution of O\(^+\) fragments. This technique can be used also in our case: the detection of the energy of the Auger electrons ejected along a specific direction in coincidence with the measurement of the momentum of the krypton ionic remnant offers an indirect pathway for a coincidence experiment for photo- and Auger electrons.

In order to analyze the origin of the interference effects, we assume an exactly known photoelectron momentum magnitude and an exact detection of the Auger electrons.
FIG. 4: (Color) Interference of the Auger electron wavelets (green) from an attosecond twin pulse for a precisely known photoelectron momentum $k^2/2 = \Omega_P$. The interference pattern is decomposed into the scaled line shape function (black) and the scaled xuv spectral envelope square (red, dashed).

FIG. 5: (Color) Interference of the Auger electron wavelets for different time delays $\tau$ between the two attosecond pulses in Fig. 1: $\tau = 0$ (black), $\tau = 3\tau_{3d}$ (red), and $\tau = 5\tau_{3d}$ (green) for a photoelectron momentum which is known with $(\Delta k)^2/2 = 10^{-5}$ eV.

For a system with decoherence, we assume that one will find a similar behavior of the interference pattern as for an inaccurately measured photoelectron momentum. In that case, in addition to averaging over wavelets with different wavelengths, also a jitter in the phase relation due to a coupling to other degrees of freedom in the system suppresses the interference fringes. Depending on the nature of the decoherence, a model of its impact on the signal can be made. Give a prediction of the signal the observed Auger electron interference pattern and its change, when the time delay $\tau$ in Fig. 1 is varied, can be used to identify and measure the amount of decoherence in a system. The impact of a variation of $\tau$ for our perfectly coherent case is revealed in Fig. 5. They resemble Fig. 3 in Ref. [9]. Thus an immediate application of the scheme discussed here is that it is a coherence meter.

In this letter, we have proposed a fundamental experiment for studying the attosecond science equivalent of Ramsey’s method of separated oscillatory fields. We have predicted the interference of Auger electrons from an attosecond twin pulse with variable time delay between the individual subpulses. The interference is only found when the photoelectron is observed in coincidence with the Auger electron. Our proposed experiment is very fundamental; it represents a crucial test for attosecond science: to what degree can we control an ultrafast electronic process in the time domain? The setting can be used as a meter for decoherence in a system. Studying Auger decay with attosecond science brings us closer to our goal of controlling electron motions on their natural time scale [7].

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[1] N. F. Ramsey, Phys. Today 33, 25 (1980).
[2] N. F. Ramsey, in Nobel Lectures in Physics 1981–1990, edited by G. Ekspan (World Scientific Publishing Company, Singapore, 1993), pp. 553–572, ISBN 981-02-0728-X, nobelprize.org/nobel_prizes/physics/laureates/1989/ramsey-lecture.html
[3] C. Buth and K. J. Schafer, J. Phys.: Conf. Ser. 194, 022038 (2009), arXiv:0905.2647.
[4] O. Smirnova et al., Phys. Rev. Lett. 91, 253001 (2003).
[5] C. Buth and K. J. Schafer, Phys. Rev. A 80, 033410 (2009), arXiv:0905.3756.
[6] M. Drescher et al., Nature 419, 803 (2002).
[7] P. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009).
[8] M. Wollenhaupt et al., Phys. Rev. Lett. 89, 173001 (2002).
[9] O. Smirnova et al., Phys. Rev. Lett. 94, 213001 (2005).
[10] M. L. Boas, Mathematical methods in the physical sci-
ences (John Wiley & Sons, New York, 1983), 2nd ed., ISBN 0-471-04409-1.

[12] G. Prümper et al., Phys. Rev. Lett. 101, 233202 (2008).