Interferences of Ultrashort Free Electron Wave Packets

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Interferences of free electron wave packets generated by a pair of identical, time-delayed, femtosecond laser pulses which ionize excited atomic potassium have been observed. Two different schemes are investigated: threshold electrons produced by one-photon ionization with parallel laser polarization and above threshold ionization electrons produced by a two-photon transition with crossed laser polarization. Our results show that the temporal coherence of light pulses is transferred to free electron wave packets, thus opening the door to a whole variety of exciting experiments.

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After the pioneering electron diffraction experiments of Davisson and Germer [1] which demonstrated the wave properties of matter, interferences of matter waves have been among the most successful confirmation of the wave-particle duality [2]. The first evidence of spatial electron interferences is given in a Young's double slit experiment [3]. Besides the fundamental aspect of emphasizing the wave character of a particle, an interference experiment provides a tool of ultrahigh sensitivity which is often used to probe matter properties. Interferences are, for instance, used in experiments such as Ramsey fringes [4] to measure the refraction index of atomic waves [5] or to detect the gravitational acceleration [6]. It has recently been shown that a pair of ultrashort laser pulses can be used to generate wave packet interferences in bound states of an atomic or molecular system. These interferences have been observed in molecular vibrational wave packets [7], where a direct mapping of vibrational wave packets over the complete range of energetically allowed internuclear distances was demonstrated with the help of femtosecond pump-probe photoelectron spectroscopy [8]. Likewise interference studies were carried out on Rydberg wave packets [9], spin-orbit wave packets [10], and excitons in quantum wells [11]. In each case interferences provide a tool to control coherently the excited state population. Extension to quasicontinuum states was achieved [12–14]. On the other hand, ultrashort electron pulses have been used to supplement the tools provided by combinations of light pulses. They were used to follow the dissociation dynamics of a polyatomic molecule through time-resolved electron diffraction [15]. In the atomic streak camera, ultrafast electrons provide details of the wave function of a Rydberg state [16].

We present in this Letter a direct extension of the techniques developed to generate and manipulate wave packets in bound states to studies of free electron wave packets. We start by showing that a pair of two ultrashort laser pulses can be used to generate free electron wave packets in the ionization continuum. We discuss how interferences between these wave packets are formed and observed. Subsequently, experimental demonstrations are given by the example of photoionization of potassium atoms, using threshold ionization and additionally above threshold ionization (ATI).

A pair of two time-delayed ultrashort laser pulses [Fig. 1(a)] generates free electron wave packets in the ionic continuum. From a quantum mechanical point of view these wave packets have to be considered as one

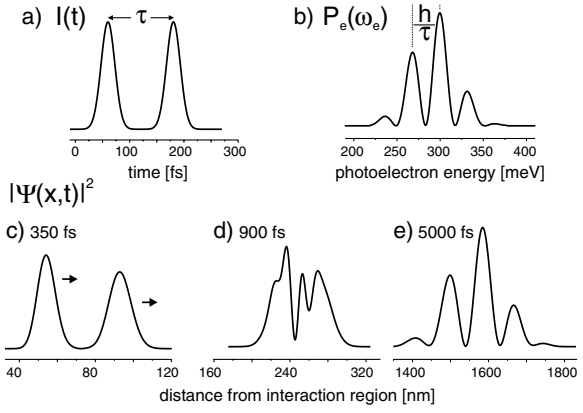

FIG. 1. Principle of the experiment. (a) Intensity of a pair of identical 30 fs Gaussian laser pulses separated by a time delay τ. (b) $P_e(\omega_e)$ corresponding photoelectron spectrum for threshold electrons. Spatial distribution of the electron wave packets: (c) after creation by the pulse pair, (d) free motion of the wave packets leading to an overlap due to dispersion, and (e) long term evolution of the interference pattern.

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double peaked free electron wave packet. So far the free interfering electrons are originating neither from double ionization of one atom nor from single ionization of different atoms. For simplicity, the ionic continuum beyond the ionization potential (IP) is described as an unstructured continuum, i.e., by a homogeneous set of levels characterized by their frequencies ω_k (see Fig. 2). During ionization photoelectrons of the kinetic energy $\hbar\omega_e = \hbar\omega_k - \hbar\omega_{\mathrm{IP}}$ are released. The laser electric field $E(t)$ couples the initially prepared $5p$ state to the kth level of the continuum, which may be labeled according to ω_e. In the weak field regime the expansion coefficients $c(\omega_e, t)$ for the ionic states may be written as

$$c(\omega_e, t) \propto \int_{-\infty}^{t} e^{-i(\omega_{5p}-\omega_k)t'} E(t')dt', \tag{1}$$

and after the pulse, when $c(\omega_e) = c(\omega_e, \infty)$, the photoelectron distribution $P_e(\omega_e) \propto |c(\omega_e)|^2$ is given by

$$P_e(\omega_e) \propto |\tilde{E}(\omega_e - \omega_{5p} + \omega_{\mathrm{IP}})|^2 = \mathrm{PSD}(\omega_{ph}), \tag{2}$$

i.e., the photoelectron spectrum $P_e(\omega_e)$ is proportional to the power spectral density (PSD) of the driving electric field at the photon frequency $\omega_{ph} = \omega_e - \omega_{5p} + \omega_{\mathrm{IP}}$ (Fig. 2). If a pair of two time-delayed ultrashort laser pulses characterized by the electric field $E'(t) = E(t) + E(t - \tau)$ [Fig. 1(a)] is used to generate a coherent double peaked free electron wave packet in the ionic continuum,

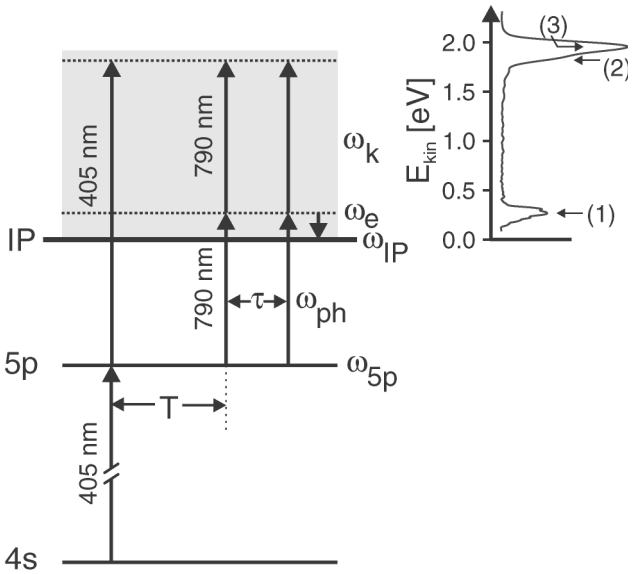

FIG. 2. Excitation scheme in potassium atoms. With a 405 nm femtosecond laser pulse the atom is prepared in its $5p$ state. After a fixed delay (T) of 3 ns, the pair of two identical pulses generates a coherent double peaked free electron wave packet. The experimental photoelectron spectrum shows the threshold electrons (1) and the first ATI electron peak produced by the 790 nm femtosecond laser pulse (2). The ATI peak (2) overlaps with a strong contribution from the photoionization of the $5p$ state by the 405 nm pulse (3). For frequency labels see text.

we may write for the coefficients according to Eqs. (1) and (2)

$$c'(\omega_e) \propto (1 + e^{i\omega_{ph}\tau})\tilde{E}(\omega_{ph}) \tag{3}$$

and obtain for the photoelectron spectrum of the pulse pair

$$P_e(\omega_e) \propto [1 + \cos(\omega_{ph}\tau)]\mathrm{PSD}(\omega_{ph}). \tag{4}$$

From Eq. (4) we expect the photoelectron signal at a given photoelectron energy to sinusoidally oscillate as a function of the pulse delay time τ with the photon frequency [see Fig. 3(b)]. Likewise, at fixed delay time τ fringes in the photoelectron spectrum with an energy separation of h/τ should be visible [compare Fig. 1(b) for calculated and Fig. 3(d) for measured data]. Hence, varying the temporal delay between the pair of ionizing laser pulses changes the interference pattern. By recording the energetically resolved photoelectron signal as a function of the time delay, a bending structure of tilted ellipses similar to Figs. 3(a) and 3(c) shows up. For the feasibility of this experiment two conditions should be fulfilled: (i) the energy resolution of the spectrometer E_{res} should resolve the fringes, i.e., $E_{\mathrm{res}} < h/\tau$ and (ii) the spectral width of the laser $\Delta\omega_l$ should support several fringes, $h/\tau < \hbar\Delta\omega_l$. This leads to

$$\Delta E_{\mathrm{res}} < h/\tau < \hbar\Delta\omega_L. \tag{5}$$

Otherwise, the interferences vanish because of (i) averaging over the broad distribution of final continuum states or (ii) not enough fringes being visible. In

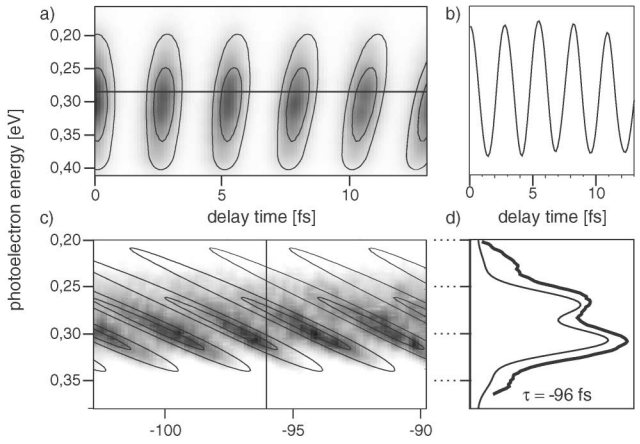

FIG. 3. Experimental results for threshold electrons with parallel polarizations, for zero (a) and negative (c) delay times τ. For comparison and to enhance the visibility of the observed ellipses, calculated contours are superimposed. (b) A section along the time delay axes at fixed energy indicates oscillatory behavior with varying time delay. (d) Section along the photoelectron energy axes at $\tau = -96$ fs showing fringes in the measured photoelectron spectrum (bold). A calculated spectrum convoluted with 25 meV spectrometer resolution is given for comparison.

order to describe the spatiotemporal evolution of the electron wave packet, the wave functions of the quasifree electrons are approximated by plane waves having the frequency ω_e,

$$\Psi_{\omega_e}(x, t) \propto e^{i(k_e x - \omega_e t)}, \tag{6}$$

with the corresponding wave number $k_e = \sqrt{2m\omega_e/\hbar}$. The approximation is justified since the interaction with the ionized K atoms is negligible for threshold electrons at 0.3 eV and has no effects on the long term evolution. The electron wave packet $\Psi(x, t)$ is therefore the superposition of $\Psi_{\omega_e}(x, t)$ with the expansion coefficients $c(\omega_e, t)$ from Eq. (1) as amplitudes:

$$\Psi(x, t) = \int_0^\infty c(\omega_e, t) e^{i(k_e x - \omega_e t)} d\omega_e. \tag{7}$$

The time development of the electron wave packet is depicted in Figs. 1(c)–1(e) for two 30 fs FWHM, 790 nm Gaussian laser pulses with a delay τ of 120 fs. At the end of the laser interaction the outward moving electron wave packet exhibits a double peaked structure similar to the exciting laser pulse pair slightly distorted due to dispersion. Progressing dispersion spreads the partial wave packets leading to interference structures as shown in Fig. 1(d). It can be shown analytically that the shape of the wave packet eventually converges to the photoelectron spectrum taken on a spatial scale [compare Figs. 1(b) and 1(e)]. Accordingly, the different arrival times at the detector translate into the spectrum measured in a time-of-flight photoelectron spectrometer. The experiments were carried out in a high vacuum chamber where a beam of atomic potassium K ($4s$) intersects perpendicularly with the femtosecond laser pulses leading to photoionization. The released photoelectrons are detected employing a magnetic bottle-type time-of-flight (TOF) electron spectrometer with an energy resolution of 25 meV for electrons with a kinetic energy of 1 eV. The laser beam at the fundamental wavelength—provided by an amplified 1 kHz Ti:sapphire laser system—is split into two beams. One beam is frequency doubled to provide 0.25 μJ of 100 fs, 405 nm photons perpendicularly polarized with respect to the TOF axes out of 810 nm being within the spectral width of our laser. This beam is used to prepare the K ($5p$) states as the initial state in our experiment (see Fig. 2). The other beam is delayed by 3 ns (T) and then coupled into a Mach-Zehnder type interferometer to generate a pair of two identical laser pulses (1 μJ, $<10^{12}$ W/cm^2, 30 fs at 790 nm) with a well defined temporal spacing τ. The laser intensity was kept rather low in order to avoid 790 nm multiphoton ionization from the K ($4s$) ground state. Furthermore, the polarization of one beam within the interferometer can be rotated by 90° employing a periscope, thus providing a pair of orthogonally polarized laser beams. Consequently, optical interferences are eliminated in that scheme.

In all experiments performed K ($5p$) serves as an initial state. This state is prepared via one-photon absorption at 405 nm. In the experiment on threshold electrons, the pair of 790 nm laser pulses has parallel polarization and the electrons are formed in a one-photon ionization process from the K ($5p$) state. The energy spectrum of the released photoelectrons is recorded as a function of the delay time τ between the two 790 nm laser pulses. Electrons at threshold energy show a periodic intensity modulation at the laser frequency with varying delay time resulting in the structures displayed in Fig. 3(a) for short delay times and in Fig. 3(c) for delay times at which the laser pulses are well separated in time so that no optical interferences occur. The results agree well with the calculated intensity profile also displayed in Fig. 3. For the calculation [using Eq. (4)] the measured PSD and time delay were adopted. To quantify the experimental results, the fringes in the photoelectron spectrum could be directly compared to simulated spectra as demonstrated in Fig. 3(d). In this comparison, the resolution of the spectrometer (25 meV) was taken into account. Because of the limited resolution of the spectrometer and the onset of fringes at large delay times [see Eq. (5)] only a small range of delay times would be available for analysis. Alternatively, the same information can be extracted from the periodic oscillations of the photoelectron signal with varying delay time [Fig. 3(b)]. In this case the time resolution can be adapted to experimental demands. In order to identify the frequency components with the largest modulation, a Fourier transform of the signal in a fixed time interval (25–50 fs) and also fixed energy interval of 12 meV normalized to the stationary signal is used for analysis. A narrow energy window is mandatory to avoid averaging over the interference structures. For parallel polarization an oscillation at the photon frequency ($\omega_{ph} = 12\,658$ cm^{-1}) with 100% modulation is observed [Fig. 4(a)]. Because of the sinusoidal shape [compare Eq. (4)] no higher harmonics of ω_{ph} are observed [see magnification in Fig. 4(a)]. However, in this one-photon ionization experiment the results could be interpreted in different ways. Even when the laser pulses are well separated in time so that no optical interferences in the time domain are present, the observed tilting ellipses might also be explained in terms of spectral interferences projected onto the ionic continuum. Even though such interferences can also be observed with incoherent light [17], distinction between optical and wave packet interferences can be achieved with multiphoton transitions [10,18].

Although the modulation is still observed for times large compared to the pulse duration [Fig. 3(c) and 3(d)], an additional experiment was performed on the ATI electrons employing ionizing laser pulses with crossed polarizations. In this case no optical interferences occur, neither in the spectral nor in the temporal domain. Because of the transition selection rules ($\Delta M = 0$ for

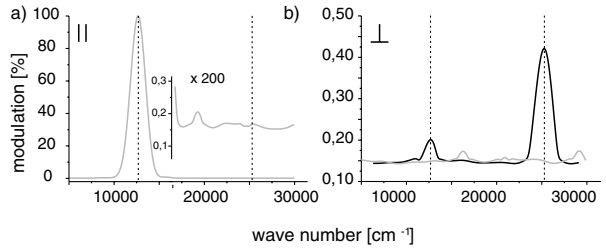

FIG. 4. Fourier transform of photoelectron signal employing a time window from 25–50 fs at threshold energy (0.29 eV, grey) and above threshold ionization (ATI) energy (1.86 eV, black). The photon frequency ω (12 658 cm^{-1}) and the second harmonic 2ω are indicated with vertical dashed lines. For parallel polarization (a) a dominant peak at ω is observed but no contribution at 2ω. For crossed polarization (b) the frequency spectrum of the threshold electrons does not show discernible features above the noise limit, whereas the ATI spectrum has a distinct maximum at 2ω due to quantum mechanical interferences.

the parallel polarized pulse and $\Delta M = \pm 1$ for the perpendicular polarized pulse), these laser pulses generate threshold electrons in orthogonal quantum states so that quantum interferences in these states are canceled out. However, in a nonlinear two-photon transition, the selection rules ($\Delta M = 0$ and $\Delta M = 0, \pm 2$ for the parallel and perpendicular polarized pulses, respectively) are such that ATI electrons can be produced in the same quantum state. Only quantum interferences of the free ATI electron wave packets are thus expected. The excitation energy is now twice the laser frequency ($2\times 12\,658$ cm^{-1}), and consequently, the modulation frequency should be $2\omega_{ph}$. The frequency spectrum for threshold electrons for crossed polarization indicates no discernible structures confirming the absence of optical interferences [Fig. 4(b), grey line]. By contrast, the ATI signal shows a pronounced peak at $2\omega_{ph}$ explicitly confirming the quantum mechanical interferences of the free electron wave packets [Fig. 4(b), black line]. Note that the ATI electron spectrum is strongly overlapped by the stationary one color 405 nm photoionization signal. Therefore the modulation depth is significantly underestimated. As in bound state interferometry, the nonlinearity of the two-photon transition removes the spectral interpretation [10,17].

The presented experiments demonstrate the coherence transfer from light pulses to free electron wave packets, thus opening the door to a whole variety of exciting new experiments. Our experiments prove that the coherence is preserved on a time scale much longer than the laser interaction. Shaped femtosecond laser pulses have been reported to create optimal wave packets in order to achieve evolution towards a preselected target [19]. Our results show that shaped free electron wave packets with a predetermined evolution can be generated [20]. This may significantly stimulate, for example, experiments on time-resolved electron diffraction [15] and the striking experiments on electron recollision [21]. If the free electron wave packet is generated by a bandwidth-limited laser pulse and a time-delayed shaped laser pulse, one can characterize the shaped laser pulse from the resulting interference pattern in the electron spectrum in analogy to spectral interferometry. This approach has the advantage to also work in the extreme ultraviolet spectral region and beyond where pulse characterization methods of complex formed laser pulses are strongly demanded. Applying the described technique towards the creation of shaped atomic wave packets using interference from molecular photodissociation was already pointed out in the stimulating work on molecular quantum picostructures by Stapelfeldt *et al.* [22].

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