

Time-domain interference in photo-double-ionization revealing nonseparable two-electron wavefunctions

Y. Hikosaka ^{1,*} T. Kaneyasu ^{2,3} S. Wada ⁴ H. Ota,^{5,2} M. Katoh ^{6,2} and F. Koike⁷

¹*Institute of Liberal Arts and Sciences, University of Toyama, Toyama 930-0194, Japan*

²*Institute for Molecular Science, Okazaki 444-8585, Japan*

³*Sokendai (The Graduate University for Advanced Studies), Okazaki 444-8585, Japan*

⁴*Graduate School of Advanced Science and Engineering, Hiroshima University, Higashi-Hiroshima 739-8526, Japan*

⁵*Japan Synchrotron Radiation Research Institute, Sayo 679-5198, Japan*

⁶*Research Institute for Synchrotron Radiation Science, Hiroshima University, Higashi-Hiroshima 739-0046, Japan*

⁷*Department of Materials and Life Sciences, Sophia University, Chiyoda, Tokyo 102-8554, Japan*



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We reveal interference patterns in photo-double-ionization spectra, produced by the interaction of argon atoms with an extreme-ultraviolet light wave packet in the form of a double pulse with variable pulse separation. These interference features provide direct evidence for nonseparable two-electron wavefunctions, an insight previously deemed beyond the reach of electron spectroscopy. This advance leverages the recently uncovered longitudinal coherence of undulator radiation, opening new avenues for attosecond-scale quantum interference experiments. The present undulator-based methodology is straightforwardly extendable into the x-ray regime, potentially offering a platform for investigating multielectron correlations associated with core-hole creation and decay.

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I. INTRODUCTION

One-photon double ionization of atoms and molecules, in which absorption of a single photon leads to the emission of two electrons, provides a direct manifestation of electron correlation in the initial states, a long-standing central topic in atomic and molecular physics. This process is commonly termed photo-double-ionization (PDI), although double photoionization also appears in the literature; we use the former term throughout this paper. The two photoelectrons leaving the dication field share the available energy and influence each other's angular distributions as well as the mutual orientation of their spins. Consequently, the state of the two photoelectrons must be described by a genuine two-electron wavefunction that cannot be factorized into independent single-electron components. Such nonseparable two-electron states have often been discussed in the context of entanglement [1] and may, in principle, serve as resources for tests of nonclassical correlations, as originally highlighted in the Einstein-Podolsky-Rosen argument [2] and formalized through Bell inequities [3].

While the nonseparable nature of the two photoelectrons generated by PDI has been extensively examined in theoretical studies [1,4–6], experimental confirmation of this

nonseparability primarily relies on measurements of the angular correlation between the photoelectrons [7]. The visualization of interference patterns in the electron angular distributions measured in the molecular frame of a hydrogen molecule serves as a representative demonstration of two-electron wavefunction describing the emitting photoelectrons [8,9]. On the other hand, the spectral features related to the kinetic energy of the photoelectrons do not directly exhibit an imprint of nonseparability, and, to date, such spectroscopic experiments have not been regarded as being capable of providing clear evidence of this aspect in PDI.

In this study, we demonstrate that direct signatures of nonseparable two-electron states in PDI are exhibited in the electron spectrum resulting from the interaction of argon atoms with an extreme-ultraviolet light wave packet in the form of a double pulse. To generate double-pulse wave packets, we employ a tandem-undulator system installed in a straight section of a synchrotron. Individual relativistic electrons traveling through the tandem undulator each emit wave packets of radiation with a common double-pulse form [10–12], fully determined by the magnetic field generated by the undulators and the delay introduced to the electron beam by a chicane situated between the two undulators. The longitudinal coherence of such light wave packets is a recently uncovered property, enabling time-domain interference experiments in the extreme-ultraviolet regime [12–16], which remain rather challenging to achieve even with state-of-the-art free-electron lasers and laser-generated high-order harmonic sources.

Alternative interactions of the two components of each light wave packet with an atom or a molecule give rise to two distinct PDI transition amplitudes separated by a time delay.

*Contact author: hikosaka@las.u-toyama.ac.jp

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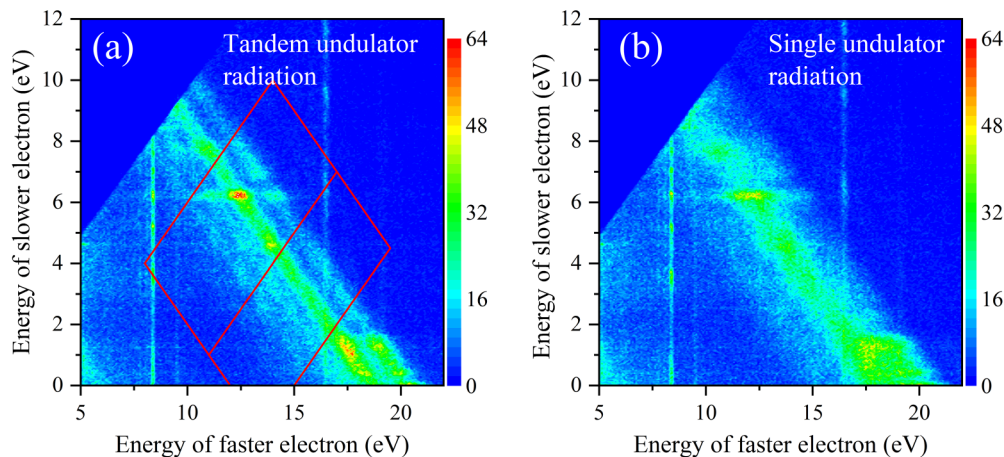


FIG. 1. Two-dimensional energy-correlation maps of photoelectron pairs emitted from argon atoms irradiated with undulator radiation tuned to a central energy of approximately 63.5 eV. Panel (b) corresponds to irradiation by a single rectangular wave packet comprising 30 optical cycles (~ 1.8 fs duration), whereas panel (a) shows the result for a pair of such wave packets separated by a delay of 2.45 fs. The red-outlined regions shown in panel (a) were used to extract the PDI spectra in Figs. 4(b) and 4(c).

The coherent superposition of the two transition amplitudes can be observed in the electron spectra for PDI, providing evidence for the two-electron wavefunction, which cannot be decomposed into independent single-electron components. Although analogous interference patterns are observed in two-photon sequential double ionization, where two independent single-photon ionization events occur consecutively [17], their origin is entirely different from the present one-photon double ionization. In the two-photon sequential double ionization case, the interference arises from entanglement of the two-electron spin state in the initial state [17].

II. EXPERIMENTAL

Electron coincidence measurements for PDI of Ar were conducted at the undulator beamline BL1U [18] of the UVSOR-III synchrotron [19] operated in single-bunch mode. A tandem undulator [20] comprising two identical APPLE-II undulators placed in series with a three-pole electromagnetic phase shifter inserted between them was used for the measurements. Both undulators were operated in a horizontally linear polarization mode. A 0.4-mm-diameter pinhole cut the spatial center of the tandem-undulator radiation, placed 9.5 m downstream from the midpoint of the two undulators. The third harmonic of the undulator radiation, whose central photon energy was set to be around 63.5 eV, was utilized to induce PDI of gas-phase argon atoms introduced effusively. The bandwidth of the radiation was about 3% of the central photon energy. Orders other than the third harmonic components of the undulator radiation were effectively suppressed using a combination of Al (300 nm thickness) and polyimide (110 nm thickness) filters. The undulator radiation consisted of light wave packets of a pair of rectangular 30-cycle oscillations with a duration of ~ 1.8 fs.

A magnetic bottle time-of-flight electron spectrometer [21,22] was employed to achieve efficient electron coincidence measurements. Pulsed-field extraction of electrons [23] was applied to measure electron times of flight in the microsecond range using synchrotron light pulses of 178-ns

repetition period. In the current measurements, while applying a dc retardation electric field of -60.2 V to the drift tube, the potential at the interaction region was periodically dropped by applying negative voltage pulses of -65.2 V at 100 kHz, synchronized to the incident light pulses. The electron energy resolution and detection efficiency were nearly constant at $\sim 3\%$ of kinetic energy and 70%, respectively, for kinetic energies up to 30 eV.

III. RESULTS AND DISCUSSION

Figure 1 presents two-dimensional energy-correlation maps of the photoelectron pairs emitted via PDI under two different undulator configurations. For the measurement shown in Fig. 1(a), both undulators in the tandem setup were operated, resulting in radiation composed of light wave packets in the form of a double pulse—specifically, a pair of rectangular (constant amplitude) pulses consisting of 30 oscillatory cycles, each pulse having a total duration of approximately 1.8 fs. The time delay τ between the two rectangular components, controlled by the length of the chicane electron trajectory formed by the phase shifter, was set to be approximately 2.45 fs. This time delay was calibrated by measuring the interference patterns observed for the He 1s photoelectron peak [16]. The energy-correlation map in Fig. 1(b) was measured using the radiation from a single undulator, with the pole gap of the other undulator opened widely (200 mm), rendering its radiation negligibly weak. The light wave packets generated by the single undulator were thus of a single rectangular shape consisting of 30 oscillatory cycles.

The two photoelectrons emitted in PDI share the available energy, which is defined as (photon energy) $-$ (Ar^{2+} binding energy). Therefore, signal due to coincidences between the two photoelectrons necessarily lies on the diagonal line $X + Y = (\text{available energy})$ on the two-dimensional plots, where X and Y correspond to the energies of the faster and slower electrons, respectively. Both maps shown in Fig. 1 are dominated by such diagonal structures, which can be associated with signal arising from PDI. The vertical stripes are due to

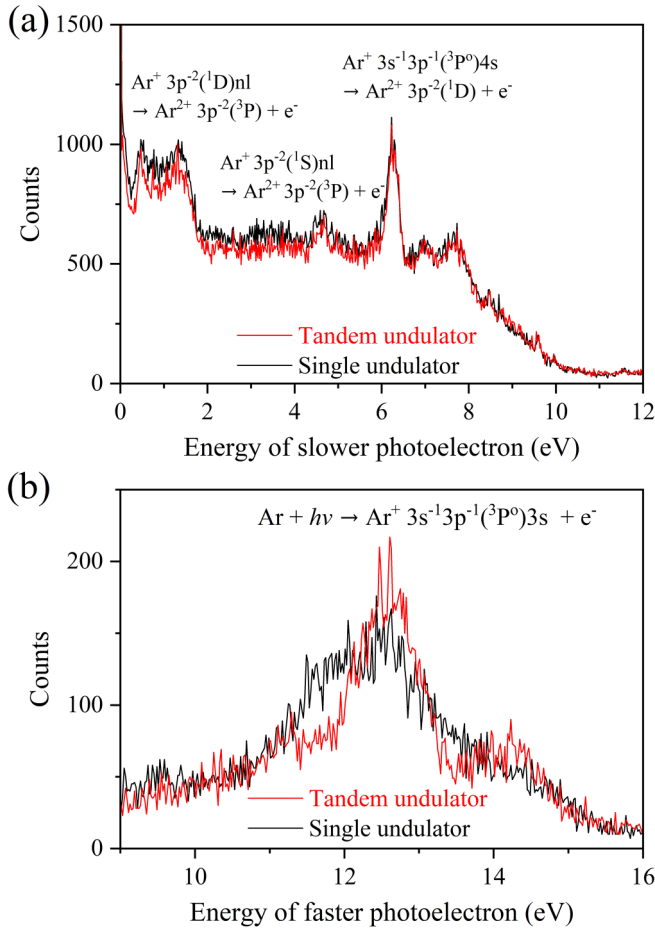


FIG. 2. (a) Spectra of slower photoelectrons resulting from argon double photoionization, corresponding to vertical projections of the diagonal features within the range $14 \text{ eV} < X + Y < 22 \text{ eV}$ observed in the maps of Fig. 1. (b) Spectra of photoelectrons emitted in the first step of sequential PDI via intermediate formation of $\text{Ar}^+ 3s^{-1} 3p^{-1} ({}^3\text{P}^0) 4s$, corresponding to horizontal slices in Fig. 1 obtained by integrating the intensity over the range of $Y = 5.8\text{--}6.5 \text{ eV}$. In both panels, data acquired with the tandem undulator are shown in red, while those obtained with the single undulator are shown in black.

false coincidences resulting from $3p$ photoelectrons whose detection cannot be prevented by the retardation potential applied. The diagonal structure in Fig. 1(a) includes several diagonal stripes that are not present in Fig. 1(b). These stripes are due to interference between PDI processes induced by the two components of the double-pulse light wave packet, and the interpretation of these will be discussed further below.

The diagonal structures in Figs. 1(a) and 1(b) both show several localized enhancements, resulting from sequential PDI paths via the intermediate formation of Ar^+ excited levels. The energy coordinates of these enhanced structures are defined by the discrete kinetic energies of the two electrons ejected in the first step (photoionization: $\text{Ar} + h\nu \rightarrow \text{Ar}^{+*} + e^-$) and second step (autoionization: $\text{Ar}^{+*} \rightarrow \text{Ar}^{2+} + e^-$). Projections of the diagonal structures (coincidence yields with $14 \text{ eV} < X + Y < 22 \text{ eV}$) onto Y axes are shown in Fig. 2(a). The projection curves, which are thus the spectra of the slower

electrons emitted in PDI, exhibit common peak structures corresponding to enhancements in the energy-correlation maps. All distinct structures are attributable to second-step transitions (autoionization), as indicated in the figure. Among them, the peak structures attributable to the autoionization of Ar^+ Rydberg states converging to $\text{Ar}^{2+} 3p^{-2} ({}^1\text{D})$ into $\text{Ar}^{2+} 3p^{-2} ({}^3\text{P})$, seen below a kinetic energy of 1.7 eV , were also clearly identified in electron coincidence measurements using an $\text{He II}\alpha$ line source [24]. The identical profiles of the autoionization structures in the two spectra, shown in Fig. 2(a) as black and red lines, demonstrate that the initial interaction with light wave packets of different waveforms does not influence autoionization processes that result from interelectron interaction.

The peak attributable to the autoionization process $\text{Ar}^+ 3s^{-1} 3p^{-1} ({}^3\text{P}^0) 4s \rightarrow \text{Ar}^{2+} 3p^{-2} ({}^1\text{D}) + e^-$ is clearly isolated in the spectra shown in Fig. 2(a), appearing around an electron energy of 6.3 eV . The spectra of photoelectrons emitted in the initial step of the corresponding sequential PDI pathway were extracted and are plotted in Fig. 2(b). They correspond to horizontal slices of the energy-correlation maps in Fig. 1, obtained by integrating the intensity over the range $Y = 5.8\text{--}6.5 \text{ eV}$. While a single broad peak is observed in the spectrum obtained with the single undulator configuration (black curve), a modulation appears in the spectrum measured with the double-undulator configuration (red curve). This modulation arises from the interference between photoionization induced by each of the two components of the double-pulse light wave packet [16]. Observation of this phenomenon is to be expected, since the initial photoionization in sequential PDI is driven by the interaction with light. The energy fringes arise because the two ionization events create coherent electron amplitudes separated in time. The temporal delay introduces an energy-dependent phase difference, which leads to the observed interference pattern.

We now turn our attention to the interference pattern appearing in the summed energy of the two photoelectrons. Figure 3(a) shows a histogram of the summed energy for photoelectron pairs detected in coincidence, generated by radiation from the single undulator setup. This spectrum corresponds to the projection of the coincidence counts in the energy-correlation map of Fig. 1(b) onto the line $X = Y$. While the possible final states following PDI are $\text{Ar}^{2+} 3p^{-2} ({}^3\text{P}, {}^1\text{D}, {}^1\text{S})$ states, the wide bandwidth of the undulator radiation prevents the individual Ar^{2+} states from being resolved. Fitting the spectrum with three Gaussian peaks (standard deviation σ of 0.95 eV) located at kinetic energies calculated using known binding energies [25] and the central photon energy of the undulator radiation well reproduces the observed structure. The fitting gives the relative populations of these states to be ${}^3\text{P} : {}^1\text{D} : {}^1\text{S} = 1:1.83:0.43$. It is worth noting that the actual undulator spectrum, shaped by the Fourier transform of the rectangular temporal profile of individual light wave packets, gradually deviates from a Gaussian function at the tails, but this is disregarded in the fitting.

The histogram of the sum energy of two photoelectrons for the double-pulse wave packet, obtained using the same projection method for the data shown in Fig. 1(a), is displayed in Fig. 3(b). In contrast to the diffuse structure seen in Fig. 3(a), the spectrum shown in Fig. 3(b) exhibits distinct peaks, which

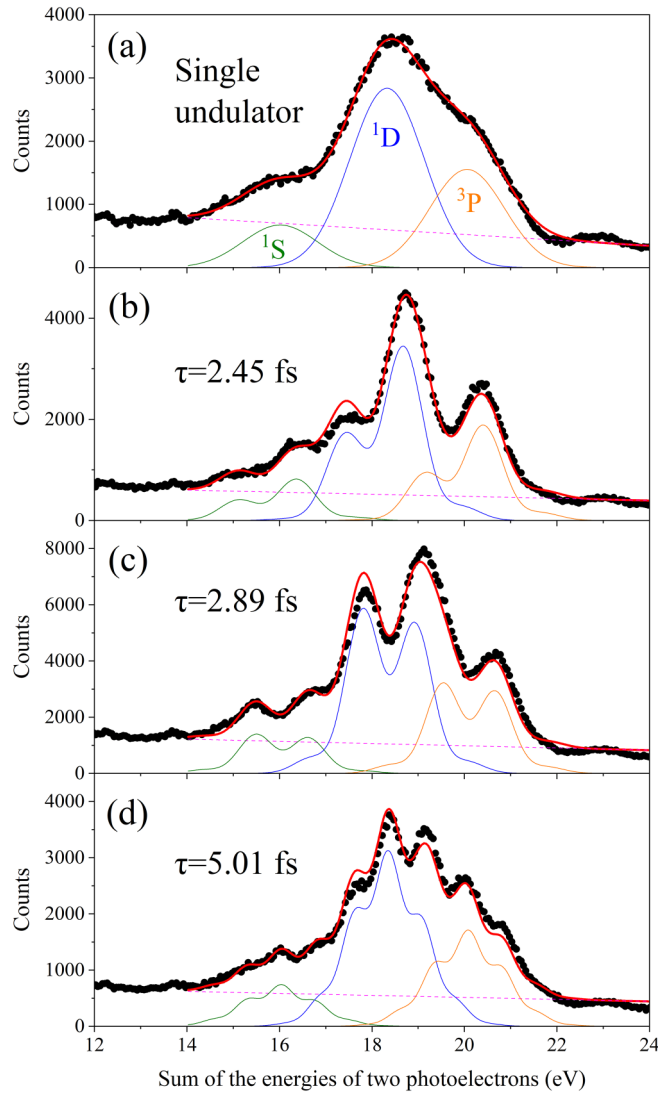


FIG. 3. Histograms of the energy sum of two coincident photoelectrons resulting from irradiation by a single undulator [panel (a)] and a tandem undulator [panels (b)–(d)]. Panels (a) and (b) show projections of the coincidence counts in the energy-correlation maps of Figs. 1(b) and 1(a), respectively, onto the diagonal direction ($X = Y$). Fitting results, indicating contributions from individual Ar^{2+} states and their total, are overlaid (see text for details). The time delays between the two rectangular components of the light wave packet pair are noted in panels (b)–(d).

correspond to the distinct stripes observed within the diagonal structure seen in Fig. 1(a). These fringe structures arise from interference in PDI driven by the two components of the light wave packet. Figures 3(c) and 3(d) present PDI spectra measured at larger τ , showing that fringe separation decreases with increasing τ . Although the autoionizing Ar^+ resonances contribute to the spectral intensities in Fig. 3, they do not influence the interference fringes, as demonstrated later.

The peak profiles with fringe structures in Figs. 3(b)–3(d) are fitted by sums of contributions from the formation of the Ar^{2+} $3p^{-2}$ (^3P , ^1D , ^1S) levels, with relative populations set to the ratios determined by the fitting of the spectrum in Fig. 3(a). The contribution from the formation of each Ar^{2+} level is

assumed to be

$$I(E_1 + E_2) \propto \exp \left[\frac{-(E_1 + E_2 + E_B - h\nu_0)^2}{2\sigma^2} \right] \times \left[1 + \cos \left(\frac{E_1 + E_2 + E_B}{\hbar} \tau \right) \right], \quad (1)$$

where E_1 and E_2 are the kinetic energies of the two emitted photoelectrons, E_B is the binding energy of the Ar^{2+} level, and $h\nu_0$ is the central photon energy of the undulator radiation. This equation represents a straightforward extension of the formulation established for photoelectron interference in single photoionization [16,26,27], in which the kinetic energy of a single photoelectron is replaced by the summed kinetic energy of the two photoelectrons in PDI. In the fitting procedure, σ and τ were fixed at 0.95 eV and the values calibrated from the interference patterns of the He $1s$ photoelectron peaks, respectively. The only free parameters to the fits were those necessary to represent a linear background and an overall scaling factor.

The fringe structures observed in Figs. 3(b)–3(d) are well reproduced by the fittings, demonstrating that Eq. (1) appropriately characterizes the PDI spectra associated with the formation of individual Ar^{2+} levels. The observed interference in the PDI spectra can be interpreted as a manifestation of time-domain double-slit interference, analogous to that observed in single photoionization [16,26,27]. Notably, the patterns arising from time-domain double-slit interference differ significantly from those observed in spatial double-slit experiments of the Young type. While the intensity distribution in Young's experiment consistently exhibits a central maximum, the spectral center in time-domain interference does not necessarily correspond to a maximum. This is exemplified in Fig. 3(c), where the individual Ar^{2+} spectrum exhibits a minimum near the spectral center.

The argument of the cosine function appearing in Eq. (1) is proportional not to the kinetic energy sum $E_1 + E_2$, but rather to $E_1 + E_2 + E_B$. This underscores that the interference patterns observed in the PDI spectra cannot be interpreted solely by considering the wavefunction of the two emitted photoelectrons. Rather, a wavefunction that also incorporates the phase relationship between the electrons and the residual Ar^{2+} ion is also required. The formalism of Eq. (1) is based on the premise that the wavefunction of eigenenergy $E_1 + E_2 + E_B$ after the light-matter interaction in this investigation can be expressed in the asymptotic region as

$$\begin{aligned} \Psi(E_1 + E_2 + E_B) \\ = \phi_1 \phi_2 \Phi + \phi_1 \phi_2 \Phi \exp[i(E_1 + E_2 + E_B)\tau]. \end{aligned} \quad (2)$$

From now on, we will adopt atomic units. Here, the wavefunction of the two photoelectrons, $\phi_1 \phi_2$, and that of the residual Ar^{2+} ion, Φ , correspond to asymptotic states reached after sufficient time has elapsed following photoionization, such that mutual interactions have ceased. In the present coincidence measurement, we observe the population of $\Psi(E_1 + E_2 + E_B)$ by resolving the sum of the two electron energies, $E_1 + E_2$, whereas the total wavefunction is given by a linear combination of such $\Psi(E_1 + E_2 + E_B)$ states. An important point to note is that Eq. (2) cannot be factorized into

functions of the individual electrons and the residual ion. This fact implies that the two emitted electrons propagate outward while maintaining their phase relationship and preserving coherence.

For a more detailed discussion, we can explicitly consider the wavefunctions of each photoelectron ejected in PDI. The wavefunction ϕ_i of an electron with wave number k_i and orbital angular momentum quantum number l , leaving an ion with charge number Z , takes the following form [28] in the asymptotic region of the radial coordinate r_i :

$$\phi_i \propto \frac{1}{k_i r_i} \exp[i(k_i r_i + \xi_i)], \quad (3)$$

where

$$\xi_i = Zk_i \ln(2k_i r_i) - \frac{l\pi}{2} - \sigma_l + \delta_l. \quad (4)$$

Here, σ_l and δ_l denote the Coulomb phase shift and the phase shift associated with the short-range interaction with the residual ion, respectively. The latter may contain resonance phase shifts that arise from the correlative electron interactions involved in the autoionization decay of intermediate Ar^+ states.

Using the single-electron wavefunction, Eq. (2) can be expressed in the asymptotic region as follows:

$$\begin{aligned} &\Psi(E_1 + E_2 + E_B) \\ &\propto (\exp[i(k_1 r_1 + k_2 r_2 + \xi_1 + \xi_2)] \\ &\quad + \exp[i(k_1 r_1 + k_2 r_2 + \xi_1 + \xi_2 + (E_1 + E_2 + E_B)\tau)]) \\ &\quad \times \Phi. \end{aligned} \quad (5)$$

This expression demonstrates that the interference observed in the coincidence detection of two photoelectrons arises from the interplay between two distinct two-electron waves: $\exp[i(k_1 r_1 + k_2 r_2 + \xi_1 + \xi_2)]$ and $\exp[i(k_1 r_1 + k_2 r_2 + \xi_1 + \xi_2 + (E_1 + E_2 + E_B)\tau)]$. The former represents the electron pair emitted at a time delay of 0, and the latter represents the pair emitted at a time delay of τ . There are no cases in which the electron pair is emitted at different time delays (i.e., one electron at 0 and the other electron at τ). This feature indicates that the two electrons pass through the same time slit as a pair, which is a consequence of the single-photon double ionization, i.e., a single light-matter interaction involving the correlated two-electron wavefunction. Notably, the phase difference between these two-electron waves, $(E_1 + E_2 + E_B)\tau$, is defined by the energy sum of the two photoelectrons, $E_1 + E_2$, under a specific E_B , and is independent of how the available energy is shared by the two electrons. Consequently, the interference occurs in the two-electron states, rather than in the individual single-electron states, and thus appears as the spectral features of the individual dication states in the coincidence spectra shown in Fig. 3.

As mentioned above, the two-electron coincidence signal includes contributions from sequential PDI paths via Ar^+ resonances [see Fig. 2(a)], whose effects appear as resonant phase shifts in the asymptotic region. Although direct and indirect successive PDI mechanisms lead to different spectral features, as seen in Fig. 2(a), these differences do not affect the interference pattern in the PDI spectra in Fig. 3. This is

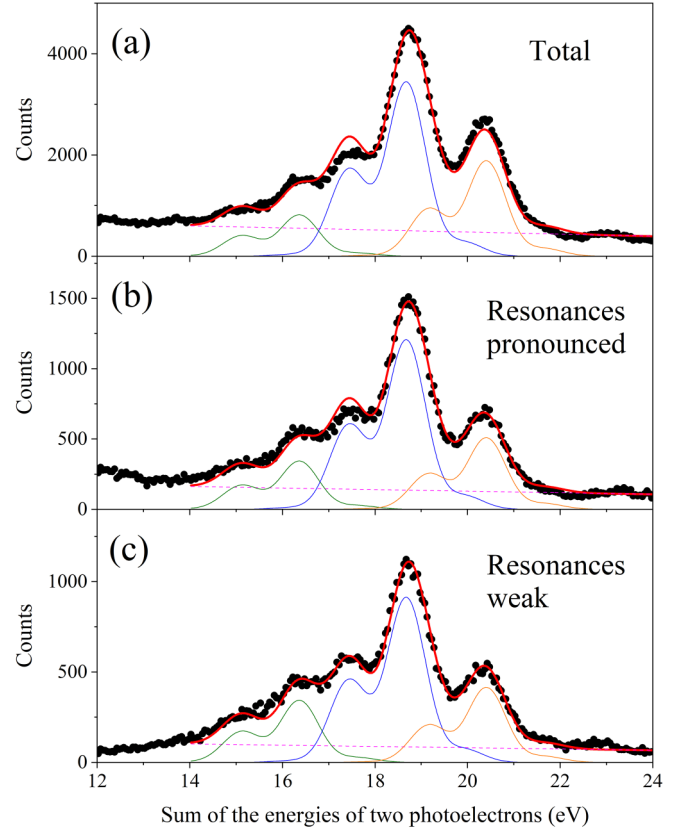


FIG. 4. Histograms of the energy sum of two coincident photoelectrons resulting from the interaction with wave packets separated by a delay of 2.45 fs. Panel (a) shows a replot of the total PDI spectrum from Fig. 3(b). The PDI spectra shown in panels (b) and (c) were extracted from the areas indicated in Fig. 1(a), where the Ar^+ resonances are pronounced and weak, respectively.

because, although the phase shifts vary rapidly with energy near the resonance points, the sum $\xi_1 + \xi_2$ is identical for the wavefunctions generated by the first and second components of the light wave packet, as shown in Eq. (5). Consequently, the contribution from sequential PDI paths does not influence the spectral fringe characteristics of the PDI spectra observed in this time-domain double-slit experiment. To make this point visible in the experimental data, we extracted PDI spectra from an area where Ar^+ resonances are pronounced and from an area where they are weak, as indicated in Fig. 1(a). The resulting spectra, plotted in Fig. 4 together with a replot of the total PDI spectrum from Fig. 3(b), show that the relative yields of the different Ar^{2+} final states are slightly modified by the contribution of the Ar^+ resonances. Nevertheless, the fringe structures remain essentially unchanged despite the different resonance contributions, confirming that the resonances do not affect the interference pattern.

IV. CONCLUSIONS

While the concept of two-electron wavefunction has been firmly established in theoretical frameworks of PDI, direct validation through experimental photoelectron spectra has so far been considered unavailable. However, the present study demonstrates that interference patterns in PDI spectra,

generated through the interaction with a light wave packet in the form of a double pulse, provide direct evidence supporting the existence of the two-electron wavefunction. While free-electron lasers and high-order harmonic generation laser sources have progressed into the extreme-ultraviolet and shorter-wavelength regions, generating a phase-locked double pulse with attosecond-level temporal precision at such wavelengths remains a challenge. The present achievement leverages the recently uncovered longitudinal coherence properties of tandem-undulator radiation.

Since the undulator-based method can be extended into the x-ray regime without technological difficulty, this approach enables the expansion of interference spectroscopy to the emission of two or more electrons associated with inner-shell photoionization and Auger decay. Whereas conventional interpretations are based on a two-step model, in reality inner-shell photoionization and Auger decay occur as nonstepwise processes [29,30], giving rise to correlated two-electron states [31]. In the absence of core-hole decoherence beyond Auger decay, such correlations between the two electrons emitted during core-hole creation and Auger decay should manifest in the energy-sum spectrum [32], as observed in the present study. In molecular systems, nonadiabatic couplings between electronic and nuclear degrees of freedom can induce decoherence in the core-hole wavefunction. Furthermore, in clusters or in the condensed phase, interactions such as

nonlocal energy transfer and intermolecular charge migration can significantly amplify this decoherence. This loss of coherence reduces the contrast of interference fringes in the two-electron spectra. Therefore, monitoring the fringe contrast as a function of time delay between the double pulses provides insight into the evolution of core-hole coherence. The present interference spectroscopy thus could offer a highly sensitive probe for observing and tracking decoherence phenomena associated with core-hole decay.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [33].

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