Quantum control of entangled photon-pair generation in electron-atom collisions driven by multiphoton-tailored free-electron wave packets

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Two-pathway coherent control of photoionization is an example of control of matter waves using light. Here, we analyze the opposite and investigate the control of quantum light using matter waves. We report coherent control of entangled photon-pair emission in electron-atom collisions mediated by transfer of the temporal and atomic coherence carried by the incident electron wave packet. The latter is engineered by resonantly-enhanced multiphoton ionization exploiting interfering ionization pathways. We show that both sources of coherence can be used to control the angular distributions of photons emitted by radiative cascade upon optical decay in the target atom, offering the possibility of coherent control driven by sculpted matter waves via transfer of optical and atomic coherence.

Introduction.— Atomic coherence [1] and quantum interferences [2, 3] are the cornerstone of coherent control of quantum phenomena. Control over the constructive and destructive interferences involving the different angular momentum components of the wave packets originating from commensurable interfering photoionization pathways play a pivotal role in the development of coherent control of light over matter waves and are at the heart of two-pathway coherent control of photoionization.

With the advent of bichromatic laser sources [4, 5], free-electron wave-packet interferometry made it possible to exploit such interferences by generating and engineering free-electron wave packets [6, 7]. Typically, coherent control is achieved by manipulating the temporal coherence of the ionizing field, e.g., by modifying the relative phase [8–14] or, alternatively, the time-delay [6, 15] between the different frequency components contributing to the interfering photoionization pathways. The optical and atomic coherence inherited from the interferometric process by the released photoelectron wave packet is imprinted on the photoelectron momentum distribution [6, 7, 16]. Polarization-tailored bichromatic laser fields and high-order intrapulse frequency mixing have also been applied to tailor matter waves [7].

In the above cases, the optical coherence of a light source is devised to exploit atomic coherence and quantum interferences to ultimately control the properties of matter waves. In this Letter, we adopt the opposite approach and investigate the control of quantum light by matter waves, mediated by the temporal and atomic coherences carried by a tailored photoelectron wave packet.

Coherent control of single- and entangled photon-pair states produced by parametric down-conversion using coherent light sources has become a current topic of intense and active research with major impact in photonic quantum information sciences [17–20]. It is thus of paramount importance for the development of new spectroscopy techniques, such as quantum-optical coherence tomography [21, 22], virtual-state spectroscopy [23–26], quantum light-based pump-probe spectroscopy [27–31], and entangled two-photon absorption spectroscopy [32, 33].

For instance, control of fluorescence in single-photon emission has been reported in a four-level system by varying the relative phase between two pump-laser sources with equal frequencies but adjustable relative phase [34]; in a three-level system by adjusting the relative phase between a pump (excitation) and control (driving optical transitions) pulse [35]; by varying the relative phase and amplitude of a trichromatic pump-laser field [36]; and by controlling the Rabi frequency and the phase of a microwave field coupling the two outermost levels in a four-level system [37]. More recently, high degrees of entanglement between parametric-down photons were reported by exploiting the twist phase of a twisted Gaussian pump beam with partial transverse spatial coherence [38]. Control over spontaneous parametric-down photons using chirped pump pulses [39] and the cross section of entangled photon pairs using classical light sources has also been reported [40].

In this Letter, we propose to use matter waves, rather
than light, in the form of photoelectron wave packets pulsed by coherent ionizing fields from a manifold of interfering photoionization pathways as the carrier of the coherence required to control two-photon cascade emission after electron-impact excitation. In addition to optical (temporal) coherence, atomic coherence inherited by the tailored photoelectron wave packet from a pump-probe spectroscopic preparation may also be exploited as a source of control. The purpose of this work is to elucidate the extent to which both sources of coherence can be transmitted to the tailored electron wave packet to ultimately manipulate entangled photon-pair generation in electron-atom collisions.

Theoretical model. — The atom-photon field interaction is treated at the level of the Weisskopf-Wigner theory for spontaneous emission [41]. De-excitation of atom $B$ occurring during and after collision with the incident electron wave packet yields the photon field in an excited multistate mode. The basis, $$|n_{k_1,\sigma_1}, n_{k_2,\sigma_2}, \ldots \rangle = \prod_{k_j,\sigma_j} |n_{k_j,\sigma_j},$$ represents $n_{k_j,\sigma_j}$ photons in mode $(k_j, \sigma_j)$: momentum $k_j$ and polarization $\sigma_j$, subject to the transversality conditions $\varepsilon_{\sigma_j} \cdot k_j = 0$. The target atom and photon field are coupled via the terms $\hat{A}_b(\mathbf{r}) \cdot \hat{\mathbf{P}}$ and $\hat{A}_A(\mathbf{r}) \cdot \hat{\mathbf{P}}$, with

$$\hat{A}_b(\mathbf{r}) = \sum_{k_j,\sigma_j} A_0(k_i) (\hat{a}_{k_j,\sigma_j} e^{i k_j \cdot \mathbf{r}} \varepsilon_{\sigma_j} + \hat{a}_{k_j,\sigma_j}^\dagger e^{-i k_j \cdot \mathbf{r}} \varepsilon_{\sigma_j}),$$

as the vector potential operator coupling the eigenstates of $B$ with the photon field. $\hat{a}_{k_j,\sigma_j}^\dagger$ (\hat{a}_{k_j,\sigma_j}) creates (annihilates) one photon in mode $(k_j, \sigma_j)$. The Hamiltonian,

$$\hat{H}_{AB}(t) = \hat{H}_A - \mathbf{e} \cdot \mathbf{E}(\mathbf{r}, t) \times (1 + \hat{V}_J)$$

$$+ 1 \sum_{k_j,\sigma_j} h \omega(k_i) + \frac{1}{2m} (\mathbf{P} - \mathbf{e} \hat{A}_b(\mathbf{r}, t))^2,$$

dictates the ionization dynamics of atom $A$, scattering of the resulting photoelectron wave packet by atom $B$, excitation of the latter due to collision, and photoemission upon de-excitation of atom $B$. The interaction,

$$\hat{V}_J = \frac{e^2}{\mathbf{r} - \mathbf{r}_0} + V_{nc}(\mathbf{r} - \mathbf{r}_0, B) \times (1 + \hat{V}_J) V_{nc}(\mathbf{r} - \mathbf{r}_0, B),$$

mediates the scattering as well as elastic and inelastic excitations of atom $B$ with no change in the distribution of the photon modes. Ionization of atom $A$ is controlled by the classical field $\mathbf{E}(r, t) = \mathbf{E}(t) f_{\Omega A}(t)$, with $f_{\Omega A}(t)$ a Heaviside function. The latter ensures a constant spatial distribution in the vicinity of atom $A$ and leaves the target atom $B$ unaffected. $V_{nc}(\mathbf{r} - \mathbf{r}_0, B)$ is the potential energy due to the effective charge distribution acting on the single active electron of atom $B$ in the absence of atom $A$, with $r_{0,B}$ the (fixed) origin of the coordinates of $B$ with respect to $A$. Equation (3a) is written in the basis $|\psi_\alpha^A \rangle \otimes |\psi_\beta^B \rangle \otimes |n_{k_1,\sigma_1}, n_{k_2,\sigma_2}, \ldots \rangle$, with $|\Phi_{\alpha\beta}^A \rangle$ and $|\Phi_{\alpha\beta}^B \rangle$ as the eigenvectors of the isolated Hamiltonians $\hat{H}_A$ and $\hat{H}_B$. They satisfy $\hat{H}_A |\Phi_{\alpha\beta}^A \rangle = \epsilon_{\alpha\beta}^A |\Phi_{\alpha\beta}^A \rangle$ and $\hat{H}_B |\Phi_{\alpha\beta}^B \rangle = \epsilon_{\alpha\beta}^B |\Phi_{\alpha\beta}^B \rangle$, respectively.

We solve the time-dependent Schrödinger equation

$$\frac{\partial}{\partial t} |\Psi_S(t)\rangle = \hat{H}_{AB}(t) |\Psi_S(t)\rangle,$$

and write $|\Psi_S(t)\rangle$ as a coherent superposition in the antisymmetrized tensor product space spanned by the eigenvectors of the isolated Hamiltonians and Eq. (1), i.e.,

$$|\Psi_S(t)\rangle = \sum_{\gamma_1,\gamma_2} \sum_{n_{k_1,\sigma_1}} \sum_{n_{k_2,\sigma_2}} \cdots |\Phi_{\gamma_1,\gamma_2}^A \rangle \otimes \prod_{k_j,\sigma_j} |n_{k_j,\sigma_j}\rangle \times \exp \left(-i \left(\epsilon_{\gamma_1}^A + \epsilon_{\gamma_2}^B + \sum_{k_j,\sigma_j} \omega(k_j) n_{k_j,\sigma_j} \right) t \right) \times S_{\gamma_1,\gamma_2}^B n_{k_1,\sigma_1}, n_{k_2,\sigma_2}, n_{k_3,\sigma_3}, \ldots \rangle.$$
defining the wave packets that arise from each of these ionization channels are coherently combined, carrying the temporal coherence of the classical field. The target atom $B$, initially in its ground state, is taken as the hydrogen atom. After excitation by the electron wave packet, optical decay may occur via different de-excitation pathways allowed by the selection rules, as epitomized in Fig. 1(c). The angular distribution of the emitted photons is obtained using the multipole expansion in Eq. (2),

$$e^{\pm \hat{k}_r} = 4\pi \sum_{\lambda,\mu} \left( \pm i \right)^\lambda j_\lambda (kr) Y_\lambda^\mu (\theta_r, \phi_r) Y_\lambda^{-\mu} (\theta_{k_j}, \phi_{k_j}), \quad (5)$$

with the spherical harmonics $Y_\lambda^\mu (\theta_{k_j}, \phi_{k_j})$ for the angles of photoemission defining the mode $(k_j, \sigma_j)$. For the photon energies considered in this work, the wavelength is several orders of magnitude larger than the extension of the highest bound state considered in our calculations. We therefore approximated the Bessel functions as $j_\lambda (kr) \approx (kr)^\lambda / (2\lambda + 1)!$ for $kr \ll 1$, which allows us to obtain the transition matrix elements as a power series in $r^\lambda$. The polarization components of the emitted photons are obtained according to $\mathbf{e}_{\sigma_j} = \mathbf{k}_j \times \mathbf{e}_0 / |\mathbf{k}_j \times \mathbf{e}_0|$ and $\mathbf{e}_{\sigma'} = \mathbf{k}_j \times \mathbf{e}_{\sigma_j} / |\mathbf{k}_j \times \mathbf{e}_{\sigma_j}|$, where

$$\mathbf{k}_j = (4\pi/3)^{1/2} |k_j| \sum_{q = 0, \pm 1} Y_1^q (\theta_{k_j}, \phi_{k_j}) \mathbf{e}_q^*,$$  

with $\mathbf{e}_q$ the covariant spherical unit vectors. Both polarization vectors are functions of the angles $(\theta_{k_j}, \phi_{k_j})$. The number of de-excitation pathways describing the radiative cascade emission is determined by the maximum order in the perturbation expansion. The results presented here were obtained by iterating the expansion coefficients up to $k = 6$, corresponding to the lowest order to describe the process of radiative two-photon cascade emission while considering feedback effects of the emitted photons on the scattered electron wave packet. The coincidence photodetection scheme is shown in Fig. 2(b): a first photodetector, fixed at $\theta_{k_2} = \pi/2$ and $\phi_{k_2} = -\pi/2$, measures the polarization component along the $z$-axis in Fig. 2 of a photon with energy $h\nu_2 = 12.078$ eV, corresponding to the transition $3p(m = \pm 1, 0) \rightarrow 1s(m = 0)$. The state of such photon is hereafter referred to as mode (2).

A second detector, fixed at $\phi_{k_1} = \pi/2$ but free to move along the polar coordinate $\theta_{k_1}$, is set to scan, along $\theta_{k_1}$, the direction of emission of its entangled peer: any photon of energy $h\nu_1 = 0.661$ eV, corresponding to the transition $4d(m) \rightarrow 3p(\pm 1, 0)$ with polarization component along $\mathbf{e}_{\sigma_j}$, as previously defined.

Figure 2 shows the angular probability distribution of measuring the correlated photon in coincidence with its entangled peer in mode (2) as a function of the relative phase between the frequencies $\omega_1$ and $\omega_3$ of Fig. 1(a). The direction of emission exhibits a noticeable dependence on the temporal coherence conveyed by the incident photon wave packet: the probability of entangled photon-pair detection is strongly affected by the relative phase between the interfering ionization pathways from which the photoelectron originates, controlled by the relative phase between the frequency components of the classical field probing the contributing photoionization pathways.

The angle-resolved occurrence of coincident photodetection is also sensitive to the parity of the photoionization pathways probed to engineer the incident photoelec-
tron wave packet. This is shown in Fig. 3, comparing, at the fixed emission angles \( \theta_{k_1} = 0 \) and \( \theta_{k_1} = \pi \), the probability of coincident photodetection already discussed in Fig. 2, this time using different photoionization schemes to engineer the incident electron wave packet: same- and opposite-parity photoionization pathways.

Figure 3(a) displays the probability of coincident photodetection obtained when the incident photoelectron wave packet is engineered according to the resonantly-enhanced two-photon ionization scheme promoting even-parity pathways depicted in Fig. 3(b). As shown in Fig. 3(a), the probability for simultaneous photon-pair detection at a given direction \( \theta_{k_1} \) can be entirely suppressed or enhanced depending on the relative phase between the contributing photoionization pathways.

Likewise, as shown in Fig. 3(c), opposite-parity photoionization pathways, as depicted in Fig. 3(d), can also be exploited to engineer the photoelectron wave packet to ultimately suppress or enhance the probability of correlated photon-pair detection. In this case, control is achieved by manipulating the relative phase between the one- and two-photon ionization pathways through the relative phase between the frequencies \( \omega_0 \) and \( \omega_3 \).

As defined, the relative phases corresponding to \( \phi = 0 \) and \( \phi = \pi \) maximize (minimize) the probability of detection in the direction \( \theta_{k_1} = 0 \) (\( \theta_{k_1} = \pi \)) for both photoionization schemes depicted in Fig. 3(b) and (d) when a photon in mode (2) is simultaneously detected. Conversely, the relative phases corresponding to \( \phi = \pi/2 \) and \( \phi = 3\pi/2 \) result in the suppression of entangled photon-pair coincident detection at angles \( \theta_{k_1} = 0 \) and \( \theta_{k_1} = \pi \) for the case of odd-even parity photoionization pathway (cf. Fig. 3(d)), whereas its even-parity counterpart, shown in Fig. 3(c), results in an equal probability of coincidence photodetection.

Finally, we consider the case of coherent control of two-photon cascade emission by transfer of atomic coherence. For this scenario, we considered two pump-laser frequencies, \( \omega_1 \) and \( \omega_4 \), to resonantly excite the states \( (4s5p)^1P \) and \( (4s6p)^1P \) in atom \( A \), creating a superposition of states evolving according to the free-field Hamiltonian. After a delay \( \tau \), a probe field with frequencies \( \omega_2 \) and \( \omega_3 \) is introduced, ionizing the electron in the coherent superposition. The resulting photoelectron wave packet then carries the atomic coherence defined by the phase accumulated between the pump and probe pulses. Figure 4 shows the time-resolved probability of correlated photon-pair detection as a function of the delay between the pump and probe pulses. The probability for coincident photodetection is sensitive to the atomic coherence carried by the photoelectron wave packet. For a fixed direction \( \theta_{k_1} \), the photon yield can be controlled significantly. Compare, for example, the yields at \( \theta_1 = 45^\circ \) for the delays \( \tau = -50 \) fs and \( \tau = -40 \) fs.
When a photon in mode (2) is detected, several de-excitation pathways may contribute to the detected polarization: the first detector cannot determine the de-excitation pathway taken by the photon of energy $h\nu_2$ in the second step of the cascade, cf. Fig. 1(c), blue arrows. Consequently, it is not possible either to determine the pathway taken by the photon of energy $h\nu_1$ in the first step of the cascade, cf. Fig. 1(c), red arrows. The angular distribution of the photon of energy $h\nu_1$ therefore contains a superposition of such contributions. These depend on the population of the $4d(m)$ states and can be adjusted by controlling the ionization process generating the photoelectron wave packet.

Conclusions.— Motivated by the recent developments in free-electron wave packet interferometry and engineering, and the increasingly active research in coherent control of entangled photon pair generation, we investigated the generation and control of correlated photon pairs triggered by electron-atom collisions. In contrast to standard approaches, we demonstrated the possibility of controlling quantum light using engineered matter waves. Using calcium and hydrogen as prototypes to control two-photon cascade emission triggered by electron-atom collisions, we demonstrated that quantum interferences and atomic coherences can be manipulated to coherently control single-photon and correlated photon-pair generation based on the transfer of optical and atomic coherences. Our results can be extended to more complex cases, such as chiral molecules, with the potential to reveal new insights into the interaction of quantum light with chiral matter waves. We also foresee extending our approach to the case of electron-ion collisions to investigate the control of correlated photon-pair emission mediated by electron-trapping correlated decay.

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