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Space-Dependent Photoelectron Dynamics Driven by Intense Vector Vortex Beams

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Vortex beams are exotic states of classical light fields that contain orbital angular momentum (OAM) due to their space-dependent phases. While the experimental realization of intense, low-frequency optical vortices has led to the discovery and application of OAM conservation in high-harmonic generation, there is not a comparable signature of light's OAM on photoelectrons. Here, we propose using a linear combination of vortex modes, referred to as a vector vortex beam, for strong field ionization of atoms. We then demonstrate that the input vortex beam's position-dependent phase can be obtained from the resulting electron momenta distribution following strong field ionization. We validate this method by performing classical trajectory Monte-Carlo simulations and comparing momentum distributions corresponding to different ionization locations within the beam profile, demonstrating that one can unambiguously determine the ionizing radiation's OAM index. These results lend way to the added control offered by vortex light compared to Gaussian or plane-wave radiation in intense light-matter interactions.

I. INTRODUCTION

In recent decades, there has been growing interest in the study and application of vortex light [1]. Since the discovery that classical light can contain orbital angular momentum (OAM) in addition to spin angular momentum (SAM) [2], these spatially structured beams have found applications in many diverse fields, including astronomy [3], optical tweezing [4], communications [5], and condensed matter physics [6]. Light's OAM can be transferred to atoms located along the optical axis, leading to modified selection rules in atomic transitions [7–10]. Current research efforts emphasize the search for conditions under which light's OAM is both relevant and useful.

In the context of intense ($\sim 10^{13} - 10^{14}$ W/cm²) radiation fields, vortex light beams have led to new discoveries and added control in gas phase high-harmonic generation (HHG). Since the first creation of harmonics equipped with OAM [11], there has been theoretical [12] and experimental [13, 14] demonstration of OAM conservation within the HHG process, including the creation of an extreme ultraviolet (XUV) pulse with OAM index $\ell = 100$ from a fundamental $\ell = 4$ vortex beam [15]. Employing two beams equipped with OAM to perform strong-field ionization has led to the creation of attosecond pulses with time-dependent OAM [16], influence over the polarization and vortex charge of individual harmonics [17], and HHG with tunable divergence and line spacing [18]. In short, light's OAM has demonstrated newfound influence over the polarization, spatial profile, and energy spectrum of attosecond pulse trains generated by intense vortex beams.

Despite the success in demonstrating and using OAM

conservation in HHG, there is no apparent transfer of OAM to individual electrons in tunneling or multiphoton ionization [19, 20]. This is because the ionized electronic wavepacket is too small and borne too far from the vortex center to experience spatial variation in the electric field during the pulse's duration. However, individual electrons do store phase information from the pulse that gets passed on to resulting harmonics, manifesting in global OAM conservation with harmonic order [21, 22]. Still, there is growing interest in designing experimental conditions that lead to novel ultrafast electron dynamics [23–25], including the search for fingerprints of light's OAM on electron momenta [26, 27], as typical low-intensity, all-optical methods for classifying vortex beams can lead to destruction of the input pulse before it can be used for applications [26]. In this paper, we propose a new method for determining the OAM index of a relatively intense, low-frequency vortex beam similar in setup to that proposed in [24, 28, 29] that involves excitation of target gas atoms by an extreme ultraviolet pulse into an excited bound state at a particular location within the vortex beam profile. The excited electrons are then tunnel ionized by a vector vortex beam consisting of a circularly polarized Laguerre-Gaussian field plus a mirror image copy of itself, transforming the beam's space-dependent phase into a space-dependent linear polarization. By performing classical trajectory Monte Carlo simulations for helium and comparing momentum distributions corresponding to different ionization locations within the beam profile, we demonstrate that one can unambiguously determine the OAM index of the ionizing radiation from photoelectron momenta.

II. LAGUERRE-GAUSSIAN VORTEX BEAMS

Before elucidating the proposed setup, it is insightful to first discuss the vortex beam field profile. Under the paraxial approximation, the electric field for classical

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light propagating in a vacuum can be written as

$$\mathbf{E}(\mathbf{r}, t) = E_0 e^{i(kz - \omega t)} u(\mathbf{r}) g(\mathbf{r}, t) \frac{\hat{\mathbf{x}} + i\epsilon \hat{\mathbf{y}}}{\sqrt{1 + \epsilon^2}}, \quad (1)$$

where $|E_0|^2$ is the electric field peak intensity, ϵ is the light's ellipticity, and $e^{i(kz - \omega t)}$ describes the pulse's plane-wave behavior. $g(\mathbf{r}, t)$ is the pulse envelope, which can be approximated as

$$g(\mathbf{r}, t) = \begin{cases} \cos^2\left(\frac{\omega t - kz}{2N}\right), & |\omega t - kz| \leq N\pi \\ 0 & \text{otherwise.} \end{cases} \quad (2)$$

N is the number of laser cycles contained within the pulse. $u(\mathbf{r})$ provides the light's spatial structure. In the trivial case of purely plane-wave radiation, $u(\mathbf{r}) = 1$. The paraxial approximation restricts the electric field to the plane transverse to the beam's propagation axis, and therefore does not include longitudinal field components. This fact is captured in the polarization of the field proportional to $(\hat{\mathbf{x}} + i\epsilon \hat{\mathbf{y}})$. For instances when light's longitudinal fields become relevant, see References [30, 31]. More generally, a complete orthogonal basis set of solutions to Maxwell's equations in vacuum under the paraxial approximation is given by the Laguerre-Gaussian modes [2, 32]. These structured light pulses take on the spatial profile

$$u_{p\ell}(r, \phi, z) = \mathcal{N} \frac{w_0}{w(z)} \left[\frac{\sqrt{2}r}{w(z)} \right]^{| \ell |} L_p^{|\ell|} \left(\frac{2r^2}{w^2(z)} \right) \cdot \exp \left[-\frac{r^2}{w^2(z)} - ik \frac{r^2}{2R(z)} + i\phi_0(z) \right] \cdot e^{i\ell\phi}. \quad (3)$$

Here, ℓ and p are the orbital angular momentum (OAM) and radial indices for the Laguerre-Gaussian beams, respectively; $\mathcal{N}$ is a normalization constant; $w(z) = w_0 \sqrt{1 + (z/z_0)^2}$ is the beam waist, which takes on its minimum value of the beam waist radius w_0 at $z = 0$; $z_0 = \pi w_0^2 / \lambda$ is the Rayleigh length; $L_p^{|\ell|}(\cdot)$ is the associated Laguerre polynomial [33, 18.5.12]; $k = 2\pi/\lambda$ is the wave number; $R(z) = z[1 + (z_0/z)^2]$ is the beam's radius of curvature, and $\phi_0 = (2p + \ell + 1) \tan^{-1}(z/z_0)$ is the Gouy phase shift of the pulse.

In the special case where $p = \ell = 0$, the equation for Gaussian beam propagation is recovered. When $p = 0$ but $\ell = \pm 1, \pm 2, \dots$, $L_p^{|\ell|}(\cdot) = 1$ and the electric field takes on a spiraling phase structure around the azimuthal coordinate ϕ and a spherically symmetric ring or “doughnut” of intensity $\propto |u_{p\ell}|^2$ with zero electric field along the center of the light's propagation ($r = 0$). Setting $p = 1, 2, 3, \dots$ increases the number of peaks and troughs in the radial intensity distribution.

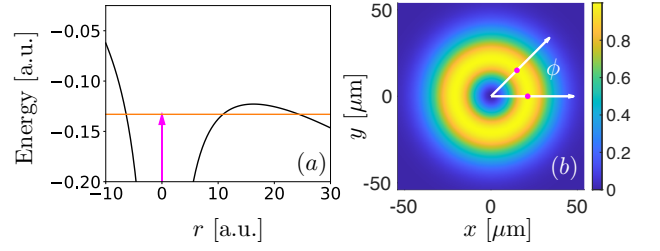


Figure 1. Schematic of the orbital angular momentum (OAM) extraction procedure. Simulated photoelectrons are first excited to a $2p$ state in helium (left) at a particular location within the vortex beam ring of maximum intensity, indicated by the magenta dots (right). Excited electrons are then strong-field ionized by the infrared vortex pulse of unknown orbital angular momentum index ℓ . The ionizing vortex field's space-dependent linear polarization depends on ℓ and leads to a rotation of photoelectron momentum distributions generated at different ionization locations.

III. VORTEX BEAMS IN STRONG-FIELD IONIZATION

The aim of this work is to extract the orbital angular momentum value ℓ from a low frequency vortex beam of the form of Equations (1) and (3) from strong-field ionized electrons. One might expect angular momentum conservation in HHG [12–14] to imply detectable angular momentum transfer in photoelectrons. However, when Laguerre-Gaussian beams are used to perform strong-field ionization in place of Gaussian beams, resulting photoelectrons are mostly unaffected by the light pulse's added spatial structure [19, 27].

This apparent discrepancy can be understood when one considers the relevant length scales associated with the beam profile and electron motion [20]. The beam waist w_0 defines the light pulse's spatial extent, and for $p = 0$ LG beams, the radius of maximum intensity within the plane of focus ($z = 0$) occurs at $w_0 \sqrt{|\ell|}/2$. While w_0 is typically on the order of tens of microns, photoelectrons exhibit oscillatory motion in response to the driving field which is characterized by the quiver radius E_0/ω^2 , or the maximum excursion distance of a classical electron inside the AC electric field [34]. For typical strong-field experiments, this parameter is on the order of tens-to-hundreds of angstroms. Therefore, the majority of photoelectrons do not experience any spatial variation of the laser field, and effects due to dipole breakdown [35, 36] including OAM transfer are greatly suppressed.

Furthermore, in the absence of a localized pump-pulse, the electron momenta distribution sums up ionization events over different locations of the vortex beam, hence averaging over the localized phase information.

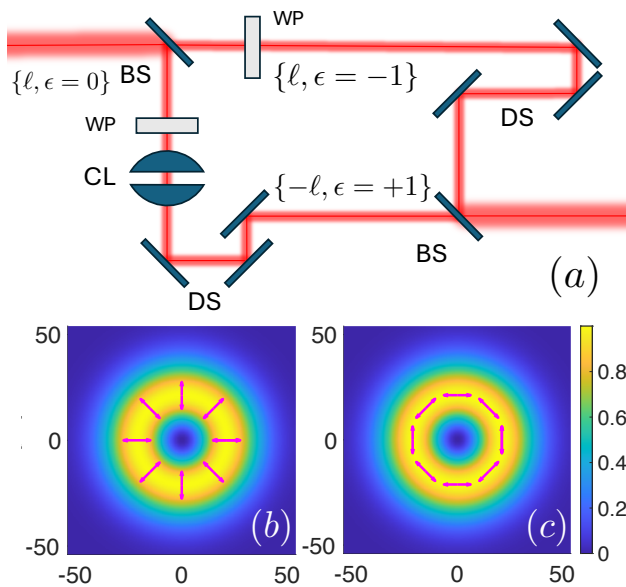


Figure 2. Creating polarization vortices out of phase vortices. (a) Possible beamline for transforming a linearly polarized Laguerre-Gaussian mode into a linearly polarized vector vortex beam. (b-c) Vector vortex beam intensity and polarization configurations for an $\ell = 1$ input mode, determined by Equation (4). (b) Radial polarization. (c) Azimuthal polarization. Legend: BS = beam splitter; WP = quarter wave plate; CL = cylindrical lenses; DS = Delay Stage.

IV. OAM EXTRACTION PROCEDURE

Our proposal to overcome these obstacles can be understood from Figure 1 and involves first exciting bound-state electrons from the ground to $2p$ state with a leading extreme-ultraviolet (XUV) pulse at a particular location within the beam profile. The experimental feasibility of this step comes from the fact that the XUV can be more tightly focused than the IR pulse [37]. Furthermore, previous experiments have involved aiming this more tightly focused XUV at the vortex center [10]. Excited state electrons are then strong-field ionized by a linear superposition of the input, left-circularly polarized ($\epsilon = -1$) vortex beam with OAM index ℓ and its mirror-image counterpart: a right-circularly polarized ($\epsilon = +1$) pulse with OAM of $-\ell$. Such a beam profile can be created using the beamline shown in Figure 2 (a): a linearly polarized ($\epsilon = 0$) Laguerre-Gaussian mode with OAM index ℓ is split into left and right circularly polarized components with the use of a beam splitter and quarter wave plates. Then, the positive spin photons pass through a pair of cylindrical lenses which flip the OAM index from ℓ to $-\ell$ [2, 38]. The recombined pulses may then be classified as a vector vortex beam [39] which exhibits a space-dependent linear polarization as opposed to a

space-dependent phase:

$$\mathbf{E}(r, \phi, z = 0, t) \propto e^{-i(\omega t + \Phi/2)} \cdot [\cos(\ell\phi + \Phi/2)\hat{\mathbf{x}} + \sin(\ell\phi + \Phi/2)\hat{\mathbf{y}}]. \quad (4)$$

Here, the phase offset Φ between the two constituent beams can be achieved by variably lengthening or shortening one arm of the beamline. For an $\ell = 0$ Gaussian beam, the polarization is uniform throughout the beam profile. For $\ell = 1$, the laser polarization rotates as one moves around the vortex center. Examples of such vector vortex beams are visualized in Figure 2 (b-c): displayed are possible polarizations (magenta) at different points along the vortex beam's ring of maximum intensity (blue-yellow color scale). The OAM index $\ell = 1$ in Equation (4) for both cases, while the phase shift Φ is set to 0 and π in panels (b) and (c), respectively, leading to polarization states that point either radially or in the azimuthal direction. In both cases, the polarization vector performs one complete rotation as one travels around the optical axis.

Photoelectrons ionized by such a pulse are sensitive to the local polarization at their respective ionization locations. The relationship between electron momenta and the ionizing radiation's OAM can be understood within the classical trajectory picture of strong-field ionization. According to the semiclassical model for strong-field ionization [34], bound-state electrons in the presence of an intense electric field are ionized into the continuum at some classically forbidden tunnel exit, then propagate as classical charged particles interacting with the AC laser electric field. For linearly polarized light, the most likely electron trajectories involve ionization at the peak of the laser pulse with zero momentum transverse to the electric field. The probability of ionization is exponentially suppressed for deviations from these peak conditions [40–42]. Accounting for the Coulomb potential, electrons borne with nonzero transverse momenta are subsequently squeezed by the ion in a process called Coulomb focusing [43]. At the end of the laser pulse, the most likely electron trajectories are those with zero momentum transverse to the linear electric field. Therefore, one can extract the local field direction from electrons ionized at a particular point within the vector vortex beam profile by radially integrating the momentum distribution and identifying the angle at which electron counts are peaked. By comparing peak momentum angles at nearby ionization sites within the beam profile, one can determine the orbital angular momentum index ℓ of the input pulse.

V. RESULTS

The proposed characterization scheme involves the following setup: helium atoms are confined to the vortex beam plane of focus ($z = 0$) and excited at a particular location within the light's intensity profile using an

extreme-ultraviolet (XUV) photon tuned to the $1s \rightarrow 2p$ transition frequency. We assume within the simulation that the XUV excites helium atoms at a single point of maximum intensity $\mathbf{r}_0 = (r_{\text{peak}} \cos \phi, r_{\text{peak}} \sin \phi, 0)^T$, where $r_{\text{peak}} = w_0 \sqrt{|\ell|/2}$. This ionization location is chosen to maximize the electron yield, but in principle, the laser polarization is independent of the radial distance within the beam profile. We consider a linear combination of Laguerre-Gaussian vortex beams described by Equations (1)-(4) confined to the plane of laser focus ($z = 0$) with trivial radial index ($p = 0$) and peak intensity $2E_0^2$. The normalization $\mathcal{N} = |\ell|^{-|\ell|/2} e^{|\ell|/2}$ is chosen such that $|u_{p=0,\ell}(r_{\text{peak}}, \phi, z = 0)| = 1$. Furthermore the phase offset Φ is set to zero. The electric field takes on an envelope (2) lasting $N = 8$ cycles.

The excitation step is modeled by simply simulating strong-field ionization of the $2p$ electrons (ionization potential $I_p = 3.62$ eV) at their specified ionization locations. These excited state electrons are ionized by the $\omega = 1600$ nm infrared vector vortex field whose space-dependent polarization is described by Equation (4). The vector vortex beam peak intensity $I = 2E_0^2 = 0.5 \cdot 10^{11}$ W/cm² is chosen to be strong enough to tunnel ionize $2p$ electrons yet too weak to ionize the more deeply bound $1s$ electrons or induce over-the-barrier ionization [44] from the $2p$ state. Therefore, the upper bound for intensity within this configuration is $6.9 \cdot 10^{11}$ W/cm². Under these conditions, the Keldysh parameter $\gamma = \omega \sqrt{(2I_p)/I} \approx 3.90$. Therefore, we employ the non-adiabatic ionization theory of Li *et al.* [45] to generate initial conditions for electrons at the tunnel exit. Compared to adiabatic models, this one captures the electron's non-zero initial longitudinal velocity [46] as well as non-adiabatic modifications to the tunnel exit position. After ionization, electrons propagate as classical particles interacting with the atomic Coulomb potential as well as the laser electric field according to Newton's equation:

$$\ddot{\mathbf{r}} = -\mathbf{E}(\mathbf{r}, t) - \nabla \left(-\frac{1}{\sqrt{r^2 + SC}} \right), \quad (5)$$

where $SC = 0.01$ provides a soft-core modification to the Coulomb potential, aiding numerical integration of the equations of motion. At the end of the laser pulse, Equation (5) resembles that of a Kepler orbit [47–49] and asymptotic electron momenta are calculated analytically.

Example momentum distributions can be seen in Figure 3. For electrons ionized at $\phi = 0$ within an $\ell = 1$ vortex field (Figure 2 (b)), the photoelectron momentum distribution displays a well-defined peak in electron counts along the $\hat{\mathbf{x}}$ direction. When ionization takes place at $\phi = \pi/4$ within the same vortex field, electron counts are maximized along the $(\hat{\mathbf{x}} + \hat{\mathbf{y}})/\sqrt{2}$ direction, demonstrating the rotation of the PMD depending on the ionization location. From these PMDs, the “measured” primary axis angle is determined by radially integrating each momentum distribution and relating the laser primary axis

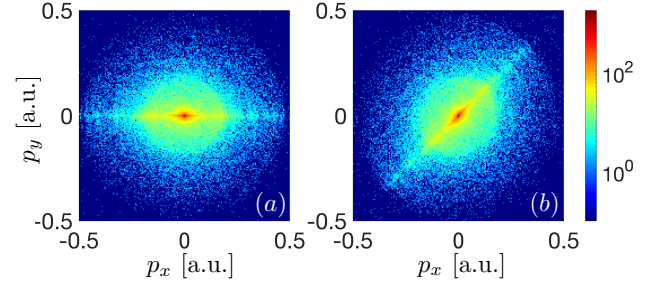


Figure 3. Comparing photoelectron momenta at different ionization locations. Electrons are ionized by an $\ell = 1$ vector vortex beam given by (Equation 4) at angles $\phi = 0$ (a) and $\phi = \pi/4$ (b) within the beam profile. The ionization locations are specified in Figure 1 (b).

angle to the angle of maximum electron counts. Comparing extracted primary axes at adjacent locations within the beam profile, one can then determine the input field's OAM.

The application of this analysis to vector vortex beams of arbitrary OAM value is demonstrated in Figure 4: simulations are performed along the ring of maximum intensity for ionizing Laguerre-Gaussian vector vortex fields with OAM indices $\ell = \pm 1, \pm 2$. For $\ell = 0$ beams, ionization takes place at radial distances $r = 0.230 \cdot w_0$, leading to an experienced intensity drop of $I(r)/I(0) = 90\%$. The measured primary axis angles are projected onto the $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ directions and compared to the known field profiles, shown as dotted and dashed lines. These curves describe the known polarization direction at each angle ϕ within the beam profile. In all cases there is excellent agreement between the extracted and known field directions without any use of free parameters or data fitting. Therefore, the results in Figure 4 validate this proposed scheme as a means of extracting the input beam's orbital angular momentum value.

While Figure 4 displays data for ionization from several azimuthal angles ϕ within each beam profile, in principle, extracting the infrared beam's OAM value can be accomplished by probing photoelectrons at just two closely spaced angles. Because the PMD's angle of rotation scales linearly with ℓ as a function of ϕ , it takes just two data points to determine this slope. However, the PMD's mirror symmetry leads to ambiguity when determining the local primary axis angle. That is, the PMD generated at an angle ϕ by a vortex field with OAM index ℓ is indistinguishable from that created by the same field at an angle $\phi + \pi$. Furthermore, as is seen in Figure 4, there can be degeneracy in the PMDs generated from the same ionization angle by beams with different ℓ values. Overall, if the maximum possible OAM value of the input field is ℓ_{max} , properly determining the input OAM index of a vector vortex beam of the form (4) with unknown ℓ requires that the angle spacing $\Delta\phi$ between adjacent ionization locations fall within the range $|\ell_{\text{max}}\Delta\phi| \leq \pi/2$.

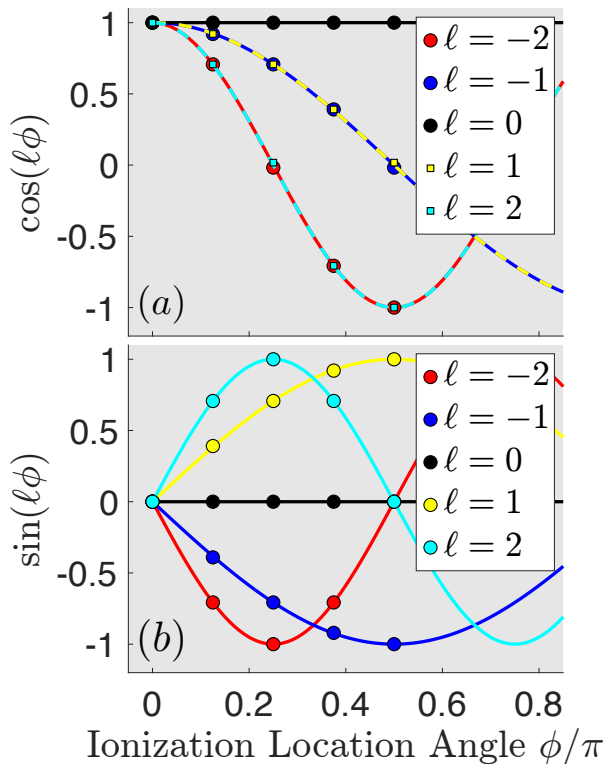


Figure 4. Determining vortex beam orbital angular momentum (OAM) from photoelectron momenta. Classical trajectory Monte-Carlo (CTMC) simulations are performed at different ionization angles ϕ within the vortex beam profile. From these simulations, local laser primary axis angles (circles and squares) are extracted from electron momentum distributions. Extracted laser primary axes angles are compared to the known field profile (solid and dashed lines) projected onto the x -axis (a) and y -axis (b), respectively.

VI. CONCLUSION

In this paper, we have outlined a technique for determining the orbital angular momentum of an intense Laguerre-Gaussian vector vortex beam from ionized electrons. By interfering a circularly polarized optical vortex with a mirror-image copy of itself, the light's space-dependent phase is transformed into a space-dependent linear polarization. Therefore, ionizing from a particular point in the vortex beam profile, assisted with a tightly focused extreme ultraviolet pulse, allows one to achieve a momentum distribution oriented along the local polarization direction. By comparing momentum distributions at different points within the beam, one is able to determine the ionizing radiation's OAM index ℓ . The technique's validity is supported by CTMC simulations for strong-field ionization by vector vortex beams with various OAM indices.

In general, light's orbital angular momentum is not transferred to strong-field ionized photoelectrons, as their wavepackets are typically very small compared to the

beam profile [19, 20]. However, by superimposing a linear combination of Laguerre-Gaussian beams with equal and opposite angular momentum components, the proposed laser configuration allows for space-dependent electron dynamics that can be traced back to the light's OAM index ℓ . This proposed scheme complements a growing list of intense vortex pulse characterization techniques [26, 27, 29, 50] that study induced electron dynamics to determine ℓ of the driving laser field. Compared to other all-optical approaches [13, 51–53], these methods do not require experimentally detecting, and therefore destroying, the beam itself. Furthermore, these methods describe experimental setups where photoelectrons are significantly influenced by vortex light's spatial profile, demonstrating added control over electrons compared to typical experiments with intense Gaussian beams.

We have demonstrated that light's spatial structure can predictably influence photoelectron dynamics, and that these dynamics can be used to determine the ionizing radiation's OAM index ℓ . In reality, one may wish to characterize a mixed-mode vortex field profile, for example, not just a single LG mode, but some linear combination. If such a beam passed through the apparatus proposed in Figure 1 (a), the resulting vector vortex beam would exhibit constructive and destructive interference from the different OAM modes as well as a space-dependent linear polarization. Therefore, the technique outlined in this manuscript could be adapted to determine the relative contributions of different LG modes by extracting both the local polarization direction and the peak field amplitude at different points within the transverse beam profile. This could be accomplished by comparing the final-state momentum distributions to those expected by ionization rate formulas which depend on the field strength [40, 41, 45]. Equipped with data detailing the local field direction and peak amplitude at different points within the vortex beam profile, one could effectively perform a 1-D Fourier transform of this data with respect to azimuthal angle ϕ to achieve the relative contributions of each OAM index ℓ . We believe this outlined procedure could be the subject of a future investigation.

This paper has emphasized performing strong-field ionization of atoms at specific points inside a vector vortex beam. However, variations of this field configuration can also be employed to drive collective circular currents in plasmas or metals, as has been investigated in theoretical studies of the inverse Faraday effect [54–56]. There has also been recent interest in optical angular momentum conservation from crystal-based HHG [57], as well as the generation and control of condensed phase skyrmions with vortex light [58, 59], as well as experimental transfer of optical OAM to excitons [6, 60]. These results demonstrate that light's orbital angular momentum is yet to be fully utilized as a tool for the study and control of ultrafast dynamics both in matter and the continuum.

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