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Nicholas Kotov

kotov@umich.edu

University of Michigan-Ann Arbor <https://orcid.org/0000-0002-6864-5804>

Joseph Daafour

Rensselaer Polytechnic Institute

Bing Ni

College of Chemistry, Beijing Normal University <https://orcid.org/0000-0001-9657-6933>

Pascal Bassene

Rensselaer Polytechnic Institute

Ciu Law

University of Wisconsin - Milwaukee

Moussa N'gom

Rensselaer Polytechnic Institute

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Joseph Daafour, Bing Ni, Pascal Bassène, Chiu Tai Law, Moussa N’Gom^{*}, and Nicholas Kotov^{*}

Dr. Bing Ni, Prof. Nicholas Kotov

Department of Chemical Engineering and Department of Materials Science and Engineering, University of Michigan, Ann Arbor, MI 48109, USA

E-mail: kotov@umich.edu

Prof. Chiu Tai Law

Electrical Engineering and Computer Science Department, University of Wisconsin, Milwaukee, WI 53201, USA

Joseph Daafour, Dr. Pascal Bassène, Prof. Moussa N’Gom

Department of Physics, Applied Physics, and Astronomy, Rensselaer Polytechnic Institute (RPI), 110 8th St., Troy, NY 12180, USA

The Shirley Ann Jackson, PhD. Center for Biotechnology and Interdisciplinary Studies, RPI, Troy, NY 12180, USA

Center for Materials, Devices and Integrated Systems (CMDIS), RPI, Troy, NY 12180, USA

Corresponding authors: ngomm@rpi.edu; kotov@umich.edu

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Orbital angular momentum (OAM) has enabled linear chiroptical discrimination, but its potential to probe nonlinear optical responses in chiral nanoparticles remains largely untapped. Here we introduce the vortex offset technique, employing asymmetric Laguerre–Gaussian beams with a controlled phase-singularity displacement δ , to drive two-photon luminescence from individual twisted gold nanorods. Breaking the cylindrical symmetry of conventional vortex beams generates a crescent-shaped intensity profile that selectively couples to chiral nanostructures, yielding single-particle chiral sensitivity 10^5 -fold greater than conventional circular dichroism (CD). We further define momentum dichroism that captures the combined effect of spin and orbital angular momentum in two-photon absorption. We discover a δ -dependent coupling parameter q that governs angular momentum transfer, a new observable capturing combined spin and orbital angular momentum effects in two-photon absorption. Opposite momentum-dichroism trends for left- and right-handed enantiomers enable unambiguous chirality identification. This work transforms structured light from a passive probe into an active participant in angular momentum exchange at the nanoscale – and does so in the nonlinear regime, achieving single-particle chiral sensitivity that outperforms both conventional CD and symmetric OAM methods.

1 Introduction

Chirality, the property of an object that cannot be superimposed on its mirror image, is fundamental to molecular biology, pharmacology, and photonics [1–3]. Circularly polarized light, carrying spin angular momentum (SAM), interacts differentially with left- and right-handed media to produce circular dichroism (CD) [4, 5], the cornerstone technique for chiroptical characterization. Nanoscale chirality of plasmonic assemblies and particles amplifies these weak molecular chiroptical signals by orders of magnitude, enabling polarization-selective responses from the ultraviolet to terahertz frequencies [6–9]. Their intense near-field enhancement additionally boosts nonlinear optical processes such as second-harmonic

generation (SHG) and two-photon luminescence (TPL), suggesting that nonlinear chiroptical effects could provide far richer information than their linear counterparts [10–12]. Despite this promise, nonlinear chiroptical detection at the single-nanoparticle level has remained out of reach. Linear CD has been measured for individual nanostructures across multiple spectroscopic windows from UV to THz [8], using photothermal [13], dark-field scattering [14], and scanning microscopy [15] approaches. However, nonlinear analogs, two-photon circular dichroism (TPCD) [16–18], chiral SHG [10, 19], and multiphoton optical rotation, have required ensemble averaging due to inherently weaker signals [20–22]. This gap has prevented the extension of nonlinear chiroptical spectroscopy to single-molecule biosensing, chiral drug screening at the nanoscale, and chirality-selective optoelectronics. The discovery that light beams can carry orbital angular momentum (OAM) in addition to spin angular momentum introduced a fundamentally new probe of chirality [23–25]. Laguerre–Gaussian (LG) beams possess a helical phase front $\exp(i\ell\phi)$ with topological charge ℓ , yielding $\ell\hbar$ of OAM per photon and a characteristic donut-shaped intensity profile with a central phase singularity [26]. Theoretical work predicted that OAM could engage chiral matter through higher-order multipole transitions [27–29], but early experiments produced only weak helical dichroism (HD) of $\sim 0.6\%$ even with plasmonic enhancement [30]. Brulot *et al.* improved this to $\sim 120\%$ by matching OAM beam dimensions to chiral microstructures [31], and Ni *et al.* demonstrated $\sim 20\%$ HD from single planar-chiral nanostructures [32]. The concept was further validated in hard X-rays [33], electron beams [34, 35], and theoretically formalized through explicit OAM-exchange frameworks [36–39]. A breakthrough came in 2022 when Rouxel *et al.* demonstrated nonlinear helical dichroism in molecular media using multiphoton ionization with vortex beams [40], proving that OAM-based chirality probing extends beyond the linear regime. Forbes *et al.* subsequently developed the theory of nonlinear vortex dichroism (NVD) for two-photon absorption by chiral molecules [41], and OAM-controlled third-harmonic generation was explored in isotropic media [42]. However, a fundamental limitation persisted across all these studies: the use of *symmetric* vortex beams with centered phase singularities. The perfect cylindrical symmetry of conventional LG modes averages out local chiral coupling when the beam is centered on a nanoparticle [32, 43], explaining why helical dichroism signals remained modest for nanoscale targets and why no single-particle nonlinear chiroptical detection had been achieved.

Here, we resolve this fundamental limitation by introducing the vortex offset technique (VOT). By displacing the phase singularity of an LG beam by a controlled offset δ from the beam center, we generate asymmetric LG (aLG) modes whose crescent-shaped intensity distribution breaks cylindrical symmetry and geometrically matches the chiral morphology of twisted nanostructures, as illustrated in Fig.1–ii. We apply this technique to twisted gold nanorods, synthesized via chiral seeded growth with L- or D-cysteine [44, 45], which possess a four-fold twisted morphology ($C_{n=4}$ symmetry) and strong plasmonic resonances ideally suited for nonlinear excitation [46, 47].

Our approach achieves four advances, each opening new capabilities for probing and utilizing non-linear effects in chiral nanoparticles. *First*, we demonstrate the first nonlinear chiroptical detection at the single-nanoparticle level using structured light, achieving chiral sensitivity 10^5 -fold greater than conventional single-nanoparticle CD. *Second*, we discover that angular momentum conservation in TPL from chiral nanostructures follows $J_{\text{TPL}} = n_s(\ell + \sigma) + qN$, where the coupling parameter q depends on the vortex offset δ —a degree of

freedom entirely absent in symmetric-beam experiments (see Supplementary Information A for first principle calculations of q). *Third*, we introduce momentum dichroism (MD), as a new chiroptical observable that captures the combined effect of spin and orbital angular momentum in two-photon absorption—superseding both CD (SAM-only) and HD (OAM-only). *Fourth*, we prove that beam asymmetry, not merely OAM, is the essential ingredient: symmetric LG beams ($\delta = 0$) produce no chiral discrimination regardless of topological charge, while controlled offsets ($|\delta| \geq 5$ nm) unlock strongly enantiomer-selective responses.

These results transform structured light from a passive diagnostic tool into an active participant in angular momentum exchange at the nanoscale [48–50]. The VOT concept is immediately generalizable to other nonlinear processes (SHG, coherent Raman), other chiral platforms (DNA origami assemblies [51, 52], perovskite nanocrystals [53], metamaterials [54]), and time-resolved measurements of angular momentum dynamics on femtosecond timescales [55], opening a broad new frontier for chiroptical science and technology.

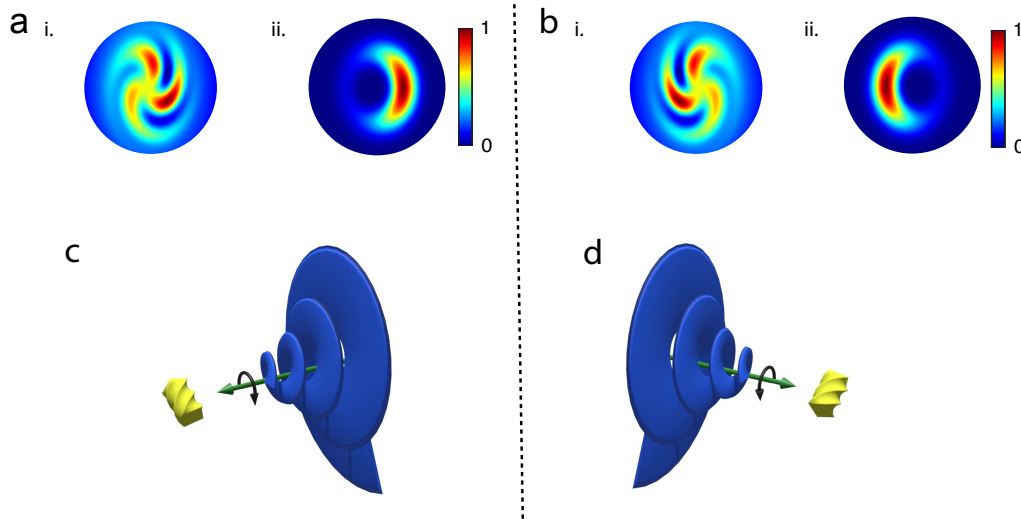


Fig. 1. Asymmetric vortex beam. (Top) (i) Interference pattern of a Gaussian beam with an asymmetric Laguerre–Gaussian beam. (ii) Spatial intensity distribution of an aLG beam with (a) negative ($\ell < 0$) and (b) positive ($\ell > 0$) topological charge. (Bottom) Illustration of a vortex wavefront incident on (c) left-handed and (d) right-handed twisted gold nanorods.

2 Results

2.1 Twisted gold nanorods

Left- and right-handed twisted gold nanorods were synthesized via seed-mediated growth [56] (Fig. 2a–d). For nonlinear measurements, NRs were drop-cast onto glass slides with sufficient spacing to avoid interparticle chiroptical effects. Individual NRs were identified by localized plasmon resonance. Scanning electron microscopy (SEM) shows uniform dimensions: left-handed NRs average 110.6 ± 9.9 nm by 64.6 ± 6.88 nm; right-handed NRs average 109.2 ± 7.5 nm by 61.4 ± 4.7 nm. Three-dimensional electron tomography (Fig. 2e)

reveals a four-fold twisted rod morphology consistent with prior reports [56]. This four-fold symmetry is critical for the angular momentum selection rules discussed below, as it imposes constraints on the allowed q parameter in the conservation of total angular momentum. Optical activity was characterized by circular dichroism (CD) and UV-vis spectroscopy (Fig. 2f–h). The extinction spectra show two plasmonic resonances: a transverse mode near 600 nm and a longitudinal mode near 860 nm. The CD g -factor exhibits sign-dependent peaks at 614 nm (left-handed, $g = +0.062$) and 620 nm (right-handed, $g = -0.058$), red-shifted from the transverse extinction peak. The emission spectrum (Fig. 2h) shows two peaks around 540 nm and 610 nm, characteristic of two-photon-induced luminescence from d -band to sp -band transitions in gold [57, 58].

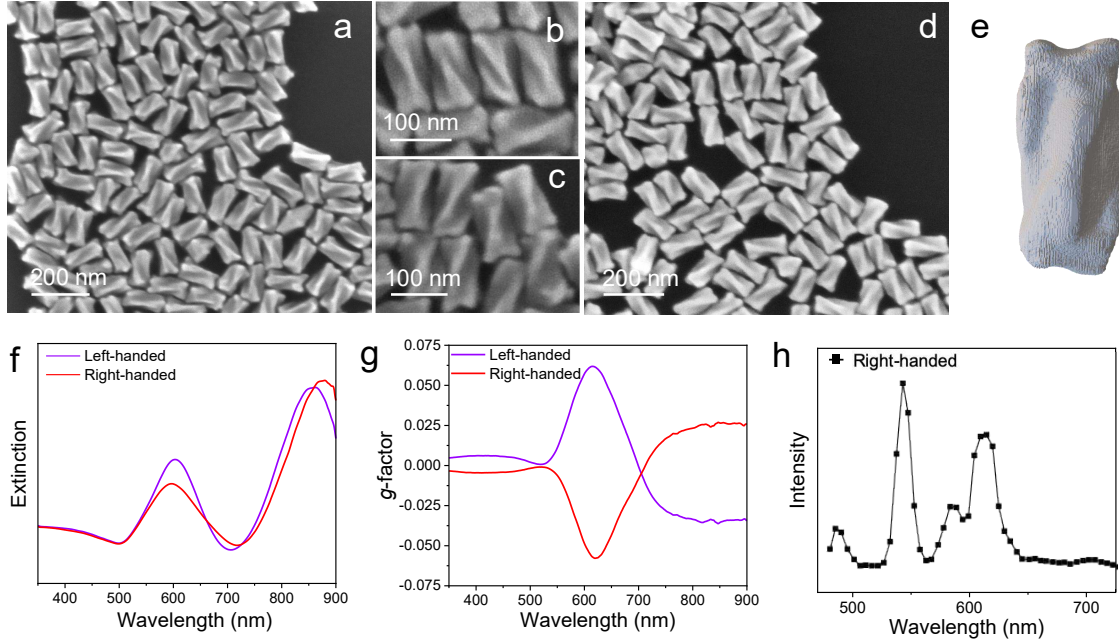


Fig. 2. Structural and optical characterization. (a,b) SEM images of left-handed and (c,d) right-handed twisted gold nanorods. (e) 3D electron tomography reconstruction of a right-handed NR highlighting the four-fold twisted morphology. (f) Circular dichroism dissymmetry factor (g -factor) showing peaks at 614 nm (+0.062) for left-handed and 620 nm (−0.058) for right-handed NRs. (g) Extinction spectra with transverse and longitudinal plasmon resonances. (h) Emission spectrum of right-handed NRs.

2.2 Characterization of gold NR chirality with vortex offset technique (VOT)

Structured light fields were generated using a spatial light modulator (SLM) to impart OAM, with circular polarization controlled by a quarter-wave plate. Asymmetric vortex beams (aLG) were produced by shifting the phase mask, breaking cylindrical symmetry (Fig. 3a). The nonlinear response was quantified via power-law fits to TPL intensity. As shown in the Supplementary Information B, the nonlinear slope is enhanced under SAM-matched conditions: left-handed NRs show higher slopes under left-circular (σ^-) light, and right-handed NRs under right-circular (σ^+) light, indicating spin-to-orbital angular momentum transfer.

We define helical dichroism (HD) to quantify the differential absorption of helical light. For linearly polarized light:

$$\text{HD}_{\sigma^0} = \frac{I(\ell, \sigma^0) - I(-\ell, \sigma^0)}{\frac{1}{2}[I(\ell, \sigma^0) + I(-\ell, \sigma^0)]}, \quad (1)$$

where $I(\pm\ell, \sigma^0)$ is the TPL intensity for aLG $_{\pm\ell,0}$ beams. For circularly polarized light:

$$\text{HD}_{\sigma^\pm} = \frac{I(\ell, \sigma^-) - I(-\ell, \sigma^+)}{\frac{1}{2}[I(\ell, \sigma^-) + I(-\ell, \sigma^+)]}. \quad (2)$$

The TPL intensity as a function of offset δ (Fig. 3b,d) shows maxima at $\delta = \pm 30$ nm for left-handed and ± 20 nm for right-handed NRs. At $\delta = 0$, no chiral dependence is observed. HD becomes measurable only for $|\delta| \geq 5$ nm (Fig. 3c,e). Compared to conventional CD g -factors, VOT enhances chirality signals by factors of 1.06 (linear) and 1.21 (circular), establishing it as a more sensitive method. This enhancement arises from the asymmetric

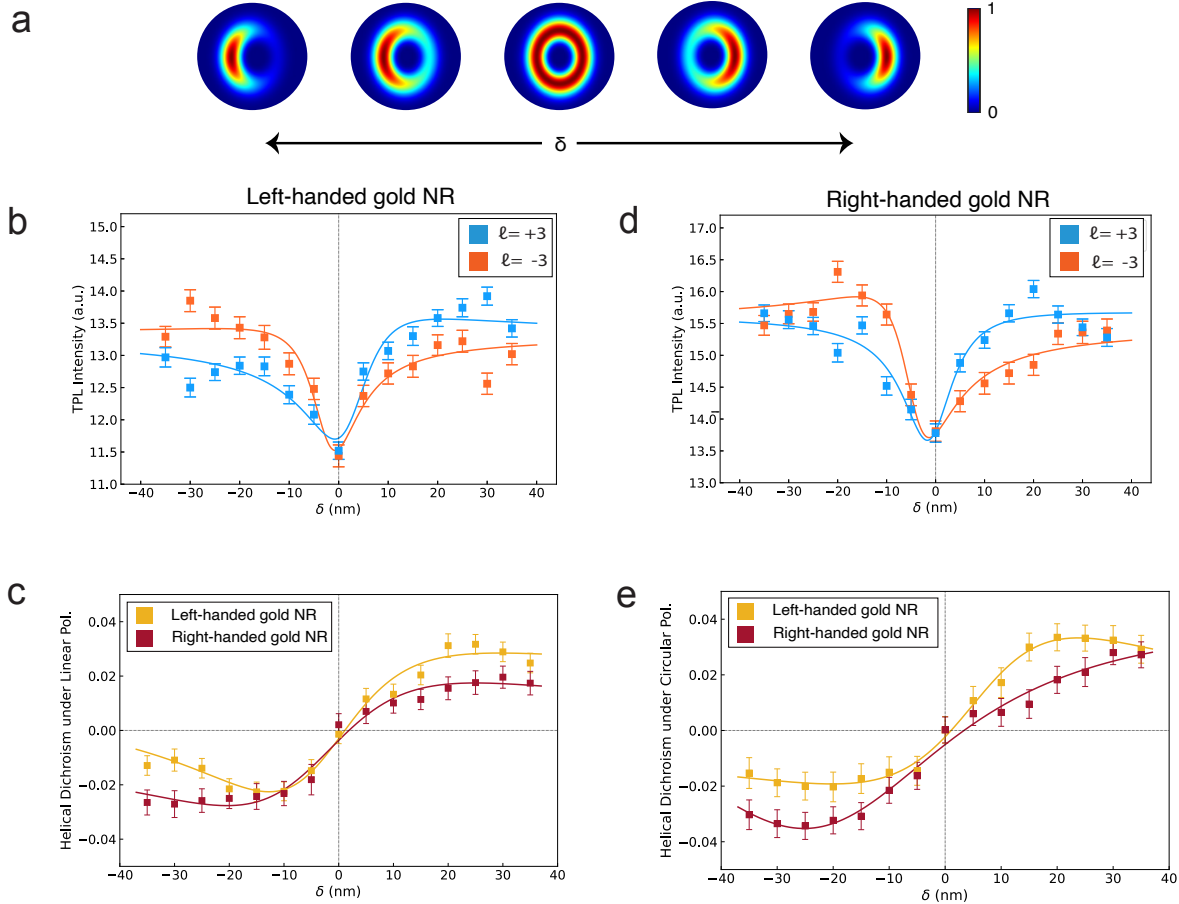


Fig. 3. Vortex offset technique (VOT). (a) Intensity profiles of aLG beams with varying phase singularity shift. (b,d) TPL intensity vs. vortex offset δ under linear polarization. (c,e) Helical dichroism (HD) for linearly polarized (c) and circularly polarized (e) aLG beams. Error bars represent standard error over three measurements.

intensity distribution of the aLG beam, which creates a local field gradient that couples more effectively to the chiral geometry of the twisted NRs than conventional circularly polarized light.

2.3 TPL polar plots for excitation with Gaussian, LG, and aLG excitation

Polarization-dependent TPL was measured by rotating incident polarization with a half-wave plate and detecting through a fixed analyzer. Under Gaussian beam excitation (Fig. 4a), TPL is strongly polarized. For LG beams with $\ell = \pm 3$ and $\delta = 0$ (Fig. 4b), polar plots for left- and right-handed NRs overlap, showing no chiral preference. With aLG beams (Fig. 4c,d), the TPL response becomes enantiomer-sensitive. For left-handed NRs with positive δ , TPL is stronger for $\ell > 0$; for right-handed NRs, the opposite trend occurs. This coupling between vortex handedness (sign of ℓ) and offset δ reveals a nonlinear response governed by structural chirality and beam asymmetry.

2.4 Total momentum dichroism

We define momentum dichroism (MD) to capture the combined effect of SAM and OAM:

$$\text{MD}(\delta) = \frac{I(J^+, \delta) - I(J^-, \delta)}{\frac{1}{2}[I(J^+, \delta) + I(J^-, \delta)]}, \quad (3)$$

where $J^\pm = 2(\ell + \sigma^\pm)$ and $\sigma^\pm = \pm 1$ correspond to left and right circular polarization. The factor of 2 accounts for the two-photon absorption process, as the total angular momentum transfer scales with the number of absorbed photons.

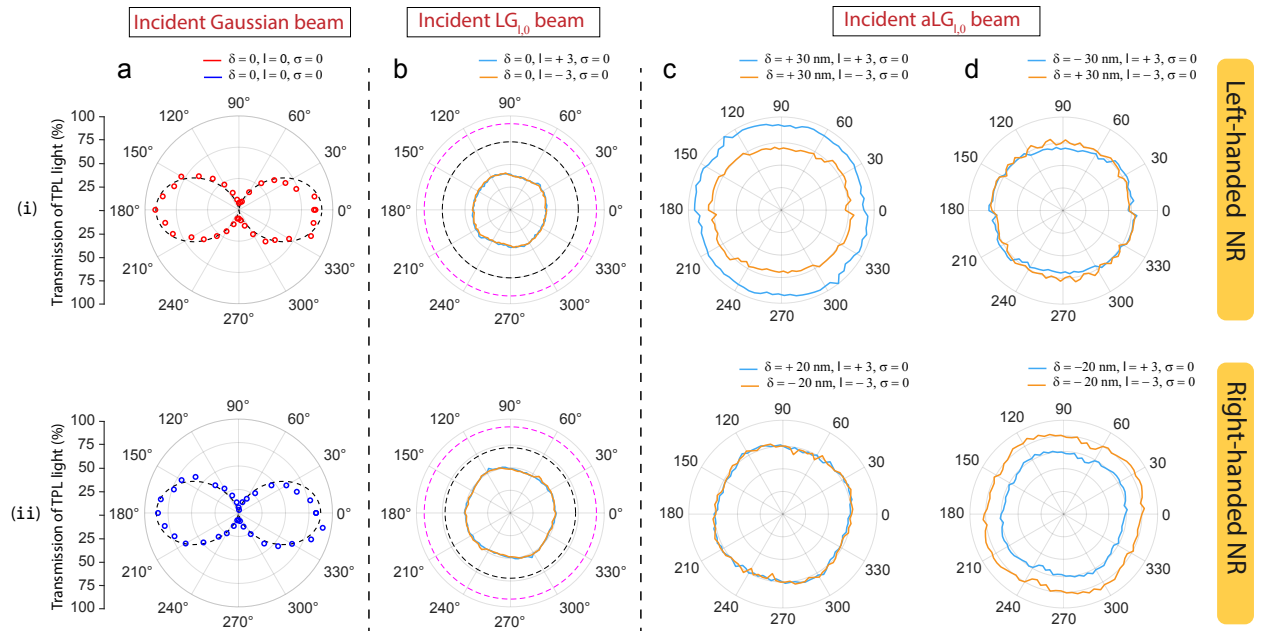


Fig. 4. Polarization-dependent TPL. (a) TPL polar plot under Gaussian beam. (b) TPL under $\text{LG}_{\pm 3,0}$ excitation. (c,d) TPL under $\text{aLG}_{\pm 3,0}$ excitation for (c) left-handed and (d) right-handed NRs with vortex offsets of ± 30 nm and ± 20 nm, respectively.

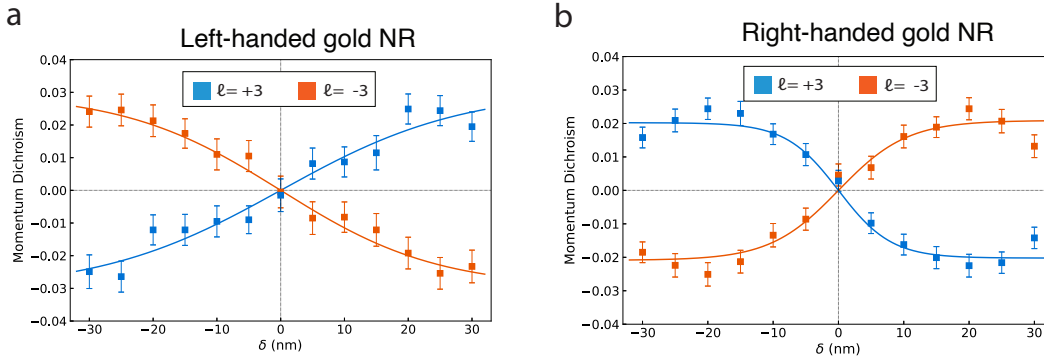


Fig. 5. Total momentum dichroism. MD as a function of offset δ for (a) left-handed and (b) right-handed gold nanorods under circularly polarized aLG beams. Opposite trends in the two enantiomers enable clear discrimination.

As shown in Fig. 5, MD exhibits opposite δ -dependent trends for left- and right-handed NRs, enabling clear enantiomer discrimination. The inversion of the response with both OAM sign and structural chirality provide unambiguous identification of the two enantiomers. The generated TPL carries angular momentum $J = n_s(\ell + \sigma) + qN$ [59], where $n_s = 2$ represents the two absorbed photons, $N = 1/4$ reflects the four-fold rotational symmetry of the twisted NRs, and q is an integer coupling parameter that depends on absorption efficiency, beam offset, and NR handedness. For aLG beams, q varies with δ , highlighting the role of beam asymmetry in angular momentum transfer. This first-principles derivation (see the supplementary Information A) confirms that the q -parameter is a genuine integer quantum number that quantifies the number of angular-momentum quanta exchanged with the discrete rotational modes of the chiral nanostructure. Its dependence on the vortex offset δ in the nonlinear regime is a unique feature of asymmetric vortex beams and has no analog in previous OAM dichroism studies [32, 60].

3 Discussion

The results presented here establish asymmetric vortex beams as a powerful route for probing nanoscale chirality in the nonlinear optical regime. By displacing the phase singularity of an LG beam from the optical axis, the vortex offset technique generates aLG fields whose crescent-shaped intensity profiles break the cylindrical symmetry of conventional centered vortex beams. This broken beam symmetry produces local intensity gradients and phase structures that couple efficiently to the four-fold twisted morphology of chiral gold nanorods. As a result, the VOT enhances chiral discrimination by factors up to 10^5 compared with conventional CD, enabling single-particle detection of TPL from individual chiral nanostructures.

A central outcome of this work is the observation that chiral discrimination is governed not only by the spin angular momentum of circularly polarized light or by the OAM of the vortex beam alone, but by their combined contribution together with an additional angular-momentum exchange channel mediated by the discrete rotational symmetry of the

nanostructure. This behavior is captured by the total angular momentum relation [59]

$$J = n_s(\ell + \sigma) + qN, \quad (4)$$

where $n_s = 2$ is the number of absorbed photons in the two-photon process, ℓ is the orbital angular momentum quantum number, $\sigma = \pm 1$ is the photon spin, $N = 1/4$ reflects the four-fold rotational symmetry of the twisted nanorods [56], and q describes angular momentum exchanged with the nanostructure.

The first-principles derivation of this parameter shows that q is not simply a phenomenological fitting constant. Rather, it is fixed by the angular-momentum selection rule imposed by the discrete rotational symmetry of the nanoparticle. For a nanostructure with C_n symmetry, conservation of angular momentum in a two-photon transition permits exchange with the rotational modes of the structure in integer multiples of $n\hbar$ [61]. For two identical absorbed photons, this gives

$$\frac{J_{\text{out}}}{\hbar} = 2(\ell + \sigma) + mn, \quad m \in \mathbb{Z}, \quad (5)$$

where m is an integer labeling the rotational angular-momentum channel coupled to the nanoparticle. For the twisted gold nanorods studied here, $n = 4$, and therefore

$$\frac{J_{\text{out}}}{\hbar} = 2(\ell + \sigma) + 4m. \quad (6)$$

Comparing Equation (6) with Equation (4), using $N = 1/4$, gives

$$2(\ell + \sigma) + \frac{q}{4} = 2(\ell + \sigma) + 4m, \quad \Rightarrow \quad q = 16m. \quad (7)$$

Thus, for the four-fold twisted nanorods, the allowed values of q are integer multiples of 16. The q -term therefore represents a quantized angular-momentum exchange with the discrete rotational modes of the chiral nanostructure. In this sense, the nanorod acts as an angular-momentum reservoir whose symmetry permits the optical field to exchange angular momentum modulo the rotational order of the structure.

This symmetry-based selection rule clarifies the physical origin of the measured momentum dichroism. In a centered, cylindrically symmetric vortex beam, the local chiral coupling averages over the azimuthal coordinate and the contribution from discrete rotational exchange is strongly suppressed. Consequently, symmetric LG beams do not produce pronounced enantio-selective signals, even though they carry orbital angular momentum. By contrast, an asymmetric vortex beam with nonzero offset δ contains azimuthal Fourier components that overlap with the chiral multipolar response of the twisted nanorods. The first-principles calculation (see supplementary A) shows that the relevant coupling channel can be expressed as an overlap between the fourth-power field distribution involved in two-photon absorption and a chiral correlation function of the nanostructure,

$$m \propto \int d^3r |E_{\text{inc}}(\mathbf{r}; \delta)|^4 \chi(\mathbf{r}), \quad (8)$$

where $\chi(\mathbf{r})$ encodes the handedness and multipolar asymmetry of the nanoparticle. Similarly, in a dipole-quadrupole description, the dichroic contribution arises from interference between the electric-dipole and electric-quadrupole transition amplitudes,

$$\Delta W \propto \text{Im}(\mathbf{d} \cdot \mathbf{Q} \cdot \nabla), \quad (9)$$

where $\mathbf{d}$ is the electric-dipole moment, $\mathbf{Q}$ is the electric-quadrupole moment, and the field gradient contains contributions controlled by both the vortex charge and the beam offset. The displacement δ therefore tunes the strength with which the incident structured field couples to the allowed rotational channel m , and hence to the corresponding value of q . The apparent continuous dependence of the measured helical and momentum dichroism on the vortex offset should therefore be understood as a variation in the coupling efficiency or statistical weight of the allowed angular-momentum channels, rather than as a violation of angular-momentum quantization. The allowed channels remain fixed by the C_4 selection rule, $q = 16m$, while the asymmetric field profile controls how strongly each channel contributes to the observed nonlinear response. For small offsets, the overlap coefficients are expected to vary approximately linearly with δ , explaining the smooth offset-dependent trends observed experimentally. Reversing the handedness of the nanorod reverses the sign of the chiral multipolar correlation function, which in turn reverses the sign of the preferred angular-momentum exchange channel. This accounts for the opposite momentum-dichroism trends observed for left- and right-handed twisted gold nanorods.

These findings provide a mechanistic explanation for the advantages of the VOT over other nonlinear optical methods for probing chiral nanostructures. The asymmetric vortex beam does not merely supply OAM; it converts structured light into an active probe of the discrete rotational symmetry and handedness of the nanostructure. The nonlinear two-photon response is especially sensitive to this effect because the transition probability scales with the square of the optical intensity, amplifying spatial gradients and enhancing the overlap between the field asymmetry and the chiral multipolar modes of the particle. This explains why the VOT outperforms both conventional CD and centered OAM-based approaches for single-particle chiral detection.

From a broader perspective, the q -parameter provides a design principle for nonlinear chiroptical spectroscopies. For a nanostructure with C_n symmetry, the allowed angular momentum exchange channels are determined by the rotational order n , while their experimental visibility is controlled by the field geometry, vortex charge, spin state, beam offset, and the multipolar composition of the chiral object. This framework can therefore be generalized beyond four-fold twisted gold nanorods to other chiral plasmonic particles, DNA-origami assemblies [62], complex chiral ‘hedgehogs’ [56], metasurfaces, and molecular or hybrid nanostructures with different discrete symmetries [63]. By engineering both the optical field and the structural symmetry, one can selectively enhance or suppress specific angular-momentum transfer pathways.

The implications extend beyond TPL, of course. The same symmetry arguments should apply to other nonlinear optical processes, including SHG, coherent Raman scattering, sum-frequency generation, and multiphoton photoemission, where spin, orbital, and structural angular momentum can be exchanged. Combining vortex offset excitation with polarization-, spectral-, or time-resolved measurements could reveal how angular momentum flows between light, plasmons, and matter on femtosecond timescales. Such measurements would provide direct access to the microscopic dynamics underlying chiral light-matter interactions.

In conclusion, asymmetric vortex beams enable highly sensitive nonlinear chiral detection by coupling the total angular momentum of light to the discrete rotational modes of chiral nanostructures. The first-principles calculation shows that the coupling parameter q is a quantized angular-momentum exchange index constrained by the four-fold symmetry of

the twisted gold nanorods, with $q = 16m$. The experimentally observed dependence on vortex offset arises from the offset-controlled coupling strength to these allowed channels, while the sign reversal between enantiomers reflects the opposite handedness of their chiral multipolar response. This work therefore transforms structured light from a passive carrier of SAM and OAM into an active probe and controller of angular-momentum exchange at the nanoscale, laying the foundation for symmetry-selective nonlinear chiroptical spectroscopy and single-particle chiral photonics.

Supporting Information

Supplementary A and B

Acknowledgments

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Competing Interests

The authors declare no competing interests.

Data Availability

The data and code supporting this study are available upon reasonable request.

Experimental Section

Nanoparticle Preparation

Chemicals: $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$ (99.99%), L- and D-cysteine hydrochloride (98%), ascorbic acid, cetyltrimethylammonium chloride (CTAC, > 98.0%), cetyltrimethylammonium bromide (CTAB, 99%), silver nitrate ($\geq 99.0\%$), n-decanol ($\geq 99.0\%$), and sodium borohydride (99%) were purchased from Sigma-Aldrich. Hydrochloric acid (3% w/v) was sourced from Scharlau. All reagents were used as received. Milli-Q water was used throughout.

Twisted AuNRs were synthesized following a seed-mediated growth protocol [56]. Mini AuNR seeds were first prepared. For large achiral AuNRs: 3000 μL of 0.01 M AgNO_3 , 1000 μL of 0.05 M HAuCl_4 , 800 μL of 0.1 M ascorbic acid, and 7 mL of 1 M HCl were added

to 100 mL of 50 mM CTAB and 11 mM n-decanol. After stirring, 500 μ L of mini AuNRs were added and the mixture was left at 35 $^{\circ}$ C for 4 h, then centrifuged (8000 rpm, 15 min) and washed with 10 mM CTAB. For twisted AuNRs: 40 mL of 40 mM CTAC, 100 μ L of 3 mM HAuCl₄, and 4.8 mL of 0.1 M ascorbic acid were mixed. L-cys (5 μ M) was added for right-handed NRs (D-cys for left-handed), followed by 200 μ L of seed solution. HAuCl₄ additions were repeated 77 times, with temperature adjustments. After 9 additions, the product was centrifuged (6000 rpm, 15 min) and washed twice with H₂O.

Optical Characterization

CD and UV-vis spectra were acquired using a JASCO J-1500 spectrophotometer. Nonlinear optical measurements were performed on a custom-built microscope using a Ti:Sapphire laser (804 nm, 100 fs, 80 MHz). TPL signals were isolated using a 550 nm bandpass filter and detected with a photomultiplier tube. Structured light fields were generated using a spatial light modulator (Hamamatsu X13138) with phase masks designed for aLG modes. Polarization control was achieved using quarter-wave and half-wave plates.

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