Atomic Spectroscopy with Twisted Photons

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About George Washington University

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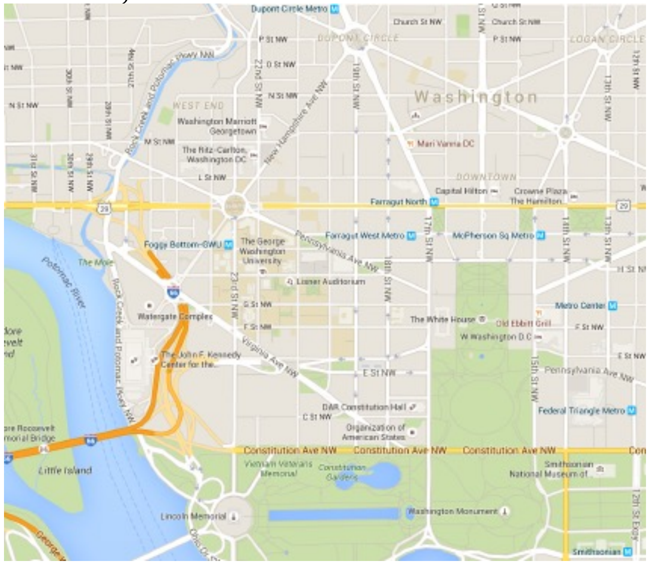

Collaborators: C.E. Carlson (William and Mary), A. Mukherjee (Mumbai), V. Serbo (Novosibirsk)
Experiment/theory collab: Ferdinand Schmidt-Kaler (Mainz), Christian Schmiegelow (Buenos Aires),

<u>Students:</u>

Maria Solyanik  Hao Wang 

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Questions Addressed

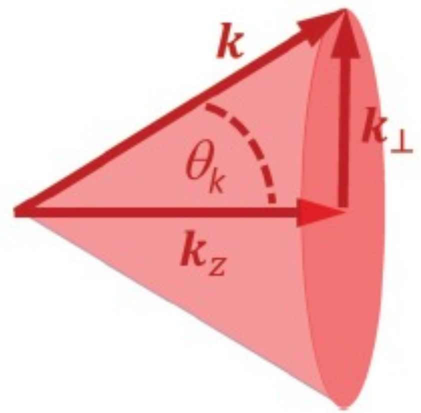
- ***Primary objective:*** Analysis of the interaction of optical vortices (or twisted light) with quantum systems (atoms, ions, atomic nuclei and quantum dots).
  - Are there any differences in atomic excitation of higher-angular momentum states (compared to plane waves)?
  - Excitation properties:
    - Singular cross sections
    - Circular vortex dichroism
    - Separation of mixed-multipole transitions
    - Non-diffractive (non-divergent) polarization features:
      - L/T field scaling

Twisted Photon State

- For Bessel beam vector potential and plane-wave expansion we use formalism from Jaregui PRA 70, 033415 (2004) and Jentschura&Serbo, PRL 106, 013001 (2011)
- Use plane-wave expansion

$$|\kappa m_\gamma k_z \Lambda\rangle = \int \frac{d^2 k_\perp}{(2\pi)^2} a_{\kappa m_\gamma}(\vec{k}_\perp) |\vec{k}, \Lambda\rangle$$
$$= \sqrt{\frac{\kappa}{2\pi}} \int \frac{d\phi_k}{2\pi} (-i)^{m_\gamma} e^{i m_\gamma \phi_k} |\vec{k}, \Lambda\rangle$$

$$a_{\kappa m_\gamma}(\vec{k}_\perp) = (-i)^{m_\gamma} e^{i m_\gamma \phi_k} \sqrt{\frac{2\pi}{\kappa}} \delta(\kappa - |\vec{k}_\perp|)$$



- Plane wave: $\langle 0 | A^\mu(x) | \vec{k}, \Lambda\rangle = \varepsilon^\mu_{\vec{k},\Lambda} e^{-ikx}$
- Twisted wave: $\mathcal{A}^\mu_{\kappa m k_z \Lambda}(x) = \langle 0 | A^\mu(x) | \kappa m k_z \Lambda\rangle$

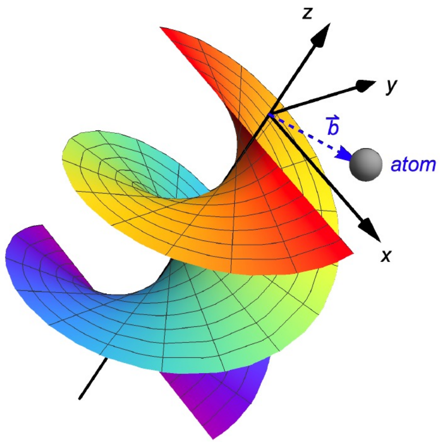
$$= \sqrt{\frac{\kappa}{2\pi}} \int \frac{d\phi_k}{2\pi} (-i)^{m_\gamma} e^{i m_\gamma \phi_k} \varepsilon^\mu_{\vec{k},\Lambda} e^{-ikx}$$

Twisted-photon factorization and forbidden transitions

- Plane-wave selection rules (from ground state):
  $|l_i = 0, m_i = 0 > \to |l_f = any; m_f = \pm 1 = (photon\ helicity) >$
- What is the new effect from twisted wave?
  - The quantization axis is tilted by an angle θ_k
  - A single eigenstate of J_z becomes a sum over all m_f' expressed via Wigner functions

$$R(\theta_k)|l_f, m_f = +1 >= \sum_{m_f'} |l_f, m_f' > d^{(l_f)}_{m_f' m_f}(\theta_k)$$

  - Each transition amplitude receives a factor $e^{i(m_f' - m_\gamma)\phi_k}$
  - ϕ_k-integration gives Bessel factors $J_{m_f' - m_\gamma}(\kappa b)$
  - $J_{m_f' - m_\gamma}(0) = 0$ if $m_f' \neq m_\gamma$ => on-axis "selection rules"



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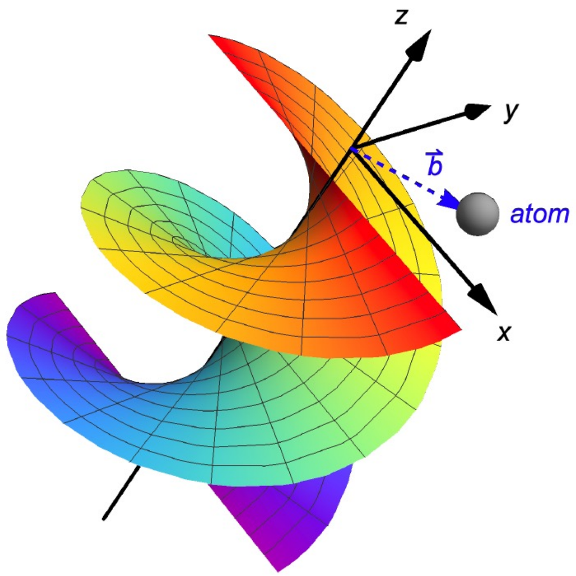
Factorization Property and Selection Rules

- In the transition matrix element for atomic photoexcitation by OAM Bessel beam can be presented as a plane-wave matrix element times factors *independent of the atomic* structure AA, Carlson, Mukherjee, Phys. Rev. A **88**, 033841 (2013), J. Opt. **18** (2016) 074013; Scholz-Margraf, Fritzsche, Serbo, AA, Surzhykov, Phys. Rev. A **90**, 013425 (2014)

$$|\mathcal{M}_{n_f l_f m_f \Lambda}(b)| = \left| \sqrt{\frac{\kappa}{2\pi}} J_{m_f - m_\gamma}(\kappa b) d^{l_f}_{m_f \Lambda}(\theta_k) \mathcal{M}^{(\mathrm{pw})}_{n_f l_f \Lambda \Lambda}(\theta_k = 0) \right|$$



It results in new selection rules near optical vortex center when off-axis b≲λ

The ratio of high-multipole amplitudes to E1 transition is

$$\frac{\mathcal{M}(m_\gamma \geqslant l_f > 1)}{\mathcal{M}(l_f = 1)} \propto \left(\frac{a_0}{b}\right)^{(l_f - 1)}$$

The formalism was applied to photo-disintegration of a deuteron in A Afanasev, VG Serbo, M Solyanik - 2018 J. Phys. G: Nucl. Part. Phys. 45 055102

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Ion-Trap Test for On-Axis Selection Rules

(Experiment in QUANTUM Center, Mainz U.)

nature COMMUNICATIONS

ARTICLE

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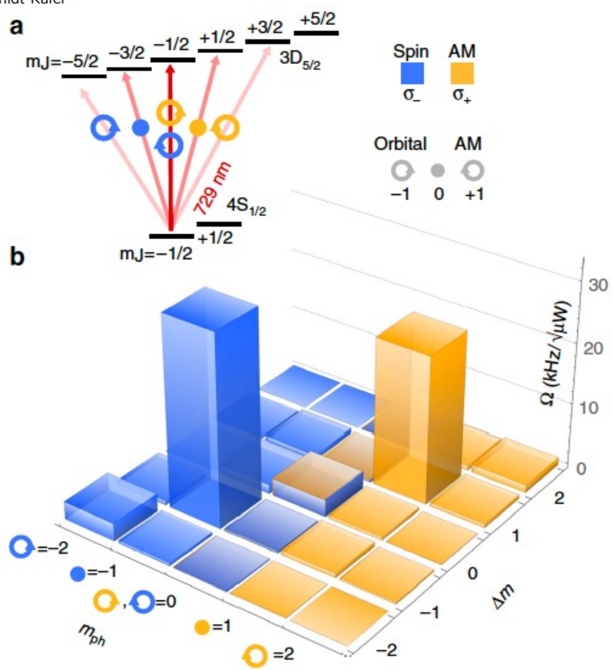
Transfer of optical orbital angular momentum to a bound electron

Christian T. Schmiegelow[1,†], Jonas Schulz[1], Henning Kaufmann[1], Thomas Ruster[1], Ulrich G. Poschinger[1] & Ferdinand Schmidt-Kaler[1]



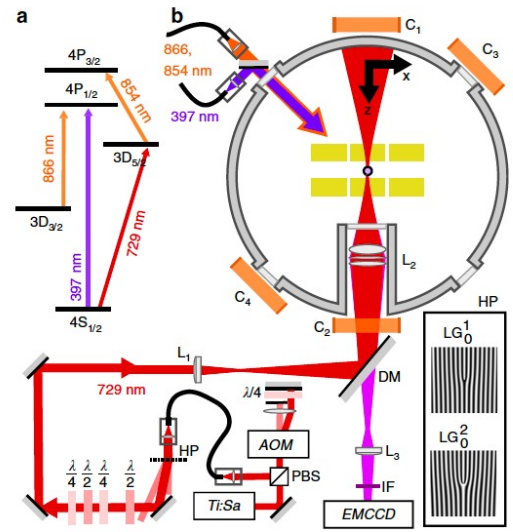


Figure 1 | Energy levels and experimental set-up. (**a**) Energy levels in $^{40}Ca^+$. The quadrupole transition at 729 nm is used to investigate the transfer of OAM from a photon to a single ion, the dipole transitions near 397, 866 and 854 nm are used for cooling, initialization and detection. (**b**) Experimental set-up. A single ion is trapped in a linear segmented Paul trap (yellow) inside an UHV chamber (gray). Delivered through fibres (top-left), light resonant with the dipole transitions is used for Doppler cooling, detection (397 and 866 nm) and state reset (854 nm). Resonance fluorescence near 397 nm is imaged on an EMCCD camera (bottom-right) with lenses $L_{2,3}$, passing a dichroic mirror and an interference filter. To excite the quadrupole $4^2S_{1/2} \leftrightarrow 3^2D_{5/2}$ transition, coherent light from a Ti:Sa laser is transmitted through an acousto-optic modulator for frequency and timing control, filtered by a polarization maintaining fibre and converted to the desired vortex beam with a holographic phase plate. The laser beam polarization is set by a series of quarter- and half-wave plates, and focused onto the ion by lenses L_1 ($f=50$ mm) and L_2 ($f=67$ mm). The magnetic field is controlled by coils C_{1-4} plus an additional coil (not shown) in the vertical direction.

Selection rules verified both on-axis and off-axis

ARTICLE

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PAPER

Experimental verification of position-dependent angular-momentum selection rules for absorption of twisted light by a bound electron

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Journal of the Optical Society of America B
OPTICAL PHYSICS

Excitation of E1-forbidden atomic transitions with electric, magnetic, or mixed multipolarity in light fields carrying orbital and spin angular momentum [Invited]

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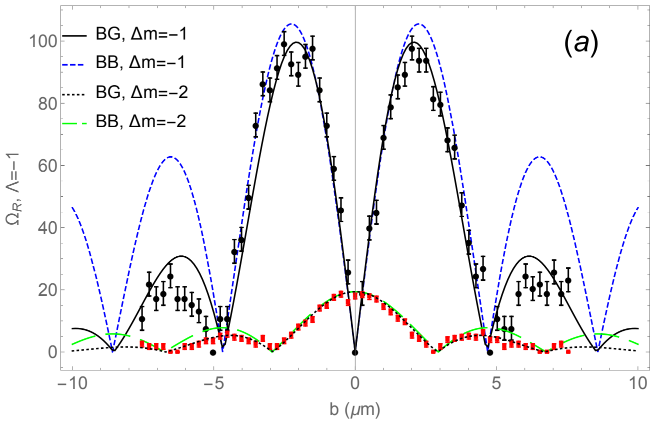
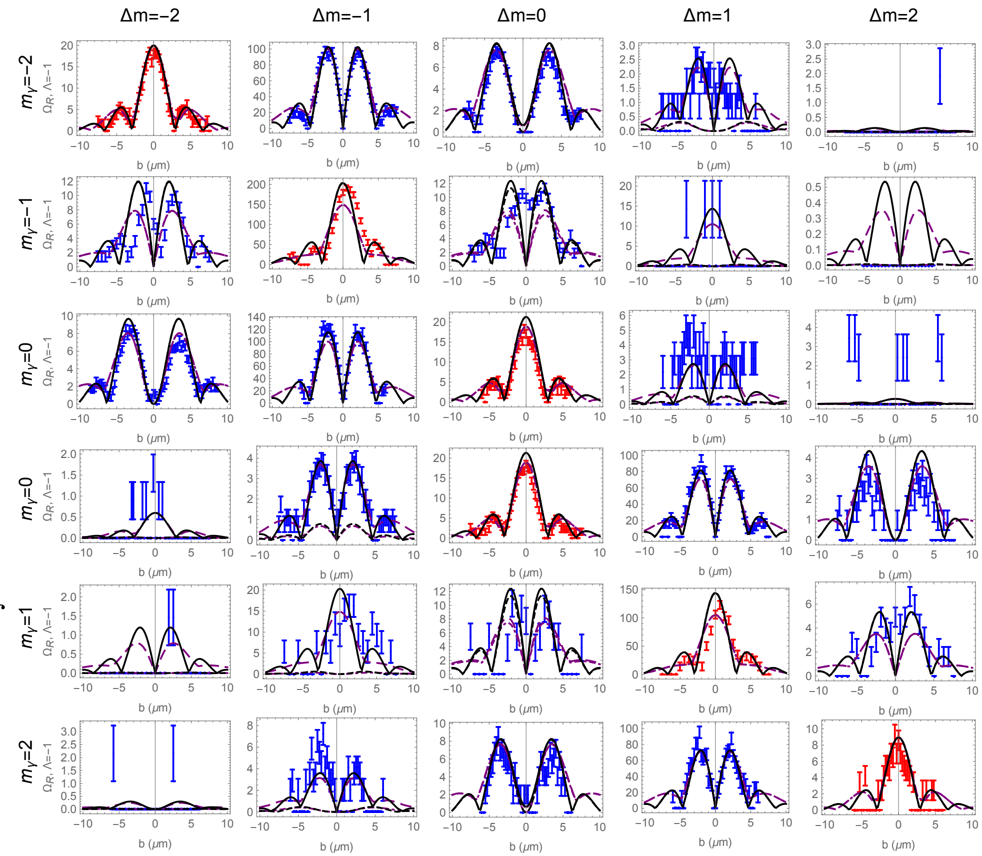
$^{40}Ca^{+}$ E2 Transitions (m_i=-1/2)



OAM=0,1



Validated theory for transitions driven by fields gradients, while the field itself is small

Implications: reduced AC Stark shift; circular & vortex dichroism; separation of atomic multipole transitions; superkick

More recent measurements of octupole (E3) transitions on 171Yb+ ion: Lange et al, PRL 129, 253901 (2022)

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Atomic spectroscopy with twisted photons

PHYSICAL REVIEW A **97**, 023422 (2018)

Atomic spectroscopy with twisted photons: Separation of $M1$-$E2$ mixed multipoles

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(Received 9 January 2018; published 26 February 2018)

We analyze atomic photoexcitation into the discrete states by twisted photons, or photons carrying extra orbital angular momentum along their direction of propagation. From the angular momentum and parity considerations, we are able to relate twisted-photon photoexcitation amplitudes to their plane-wave analogs, independently of the details of the atomic wave functions. We analyze the photoabsorption cross sections of mixed-multipolarity $E2$-$M1$ transitions in ionized atoms and found fundamental differences coming from the photon topology. Our theoretical analysis demonstrates that it is possible to extract the relative transition rates of different multipolar contributions by measuring the photoexcitation rate as a function of the atom's position (or impact parameter) with respect to the optical vortex center. The proposed technique for separation of multipoles can be implemented if the target's atom position is resolved with subwavelength accuracy; for example, with Paul traps. Numerical examples are presented for Boron-like highly charged ions.

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Formalism

$$\hat{A}_{k_z\kappa m_\gamma\Lambda}(\boldsymbol{r},t) = A\sqrt{\frac{2\pi}{\omega}}\sum_k \sum_{\Lambda=-1,1}\int \frac{d^2k_\perp}{(2\pi)^2}a_{\kappa m_\gamma} \times \{\hat{a}_{k\Lambda}\boldsymbol{e}_{\boldsymbol{k}\Lambda}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)} + \hat{a}^\dagger_{k\Lambda}\boldsymbol{e}^\dagger_{\boldsymbol{k}\Lambda}e^{-i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)}\}$$

Twisted amplitude = plane-wave amplitude x atomic-independent factors

$$\left|M^{(\mathrm{BB})}_{m_f,m_i}(b)\right| = \frac{A}{2\pi}\left|J_{m_f-m_i-m_\gamma}(\kappa b)\sum_{m'_f m'_i} d^{j_f}_{m_f,m'_f} \times (\theta_k) d^{j_i}_{m_i,m'_i}(\theta_k) M^{(\mathrm{pw})}_{m'_f m'_i}(\theta_k=0)\right|$$

$$\left|M^{(\mathrm{BG})}_{m_f,m_i}(b)\right| = e^{-b^2/\omega_0^2}\frac{A}{2\pi}\left|J_{m_f-m_i-m_\gamma}(\kappa b)\sum_{m'_f m'_i} d^{j_f}_{m_f,m'_f} \times (\theta_k) d^{j_i}_{m_i,m'_i}(\theta_k) M^{(\mathrm{pw})}_{m'_f m'_i}(\theta_k=0)\right|,$$

Spherical-harmonic expansion for the field | Multipole expansion for plane-wave transition amplitude

$$\boldsymbol{A}^M_{jm}(k,\boldsymbol{r}) = j_j(kr)\boldsymbol{Y}_{jjm}(\Omega),$$

$$\boldsymbol{A}^E_{jm}(k,\boldsymbol{r}) = \left(\sqrt{\frac{j+1}{2j+1}}j_{j-1}(kr)\boldsymbol{Y}_{j,j-1,m}(\Omega) - \sqrt{\frac{j}{2j+1}}j_{j+1}(kr)\boldsymbol{Y}_{j,j+1,m}(\Omega)\right)$$

$$M^{(\mathrm{pw})}_{m_f m_i}(0) = -\sqrt{4\pi}\sum_{j=1}^{\infty} i^{j+\mu}\sqrt{\frac{(2j+1)}{(2j_f+1)}}\Lambda^{\mu+1}C^{j_f,m_f}_{j_i,m_i,j,\Lambda}\, M_{j\mu}$$

Only $\boldsymbol{\Delta}$m=±1 transitions allowed for plane-wave photons

$$M^{(\mathrm{pw})}_{3/2,1/2}(0) = i\sqrt{\pi}(E2-\sqrt{3}M1),$$
$$M^{(\mathrm{pw})}_{1/2,-1/2}(0) = -i\sqrt{\pi}(\sqrt{3}E2+M1).$$

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Results for amplitudes

- Consider highly-charged ions (HCI) as an example; M1 and E2 are mixed and similar in magnitude
- E2 and M1 enter the absorption rates weighted by twisted-photon factors

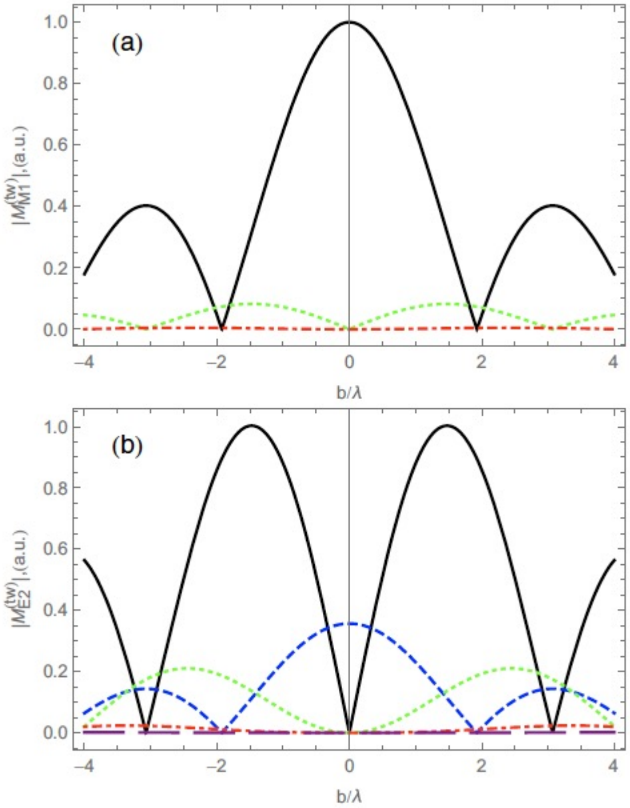

FIG. 1. Dependence of photoabsorption amplitudes of (a) $M1$ ($\lambda = 351$ nm) for $m_\gamma = 1$, and (b) $E2$ ($\lambda = 426$ nm) for $m_\gamma = 2$ transitions in Pr^{9+} HCI of OAM photons with Bessel profile for $\Delta m = 2$ (dashed blue curve), $\Delta m = 1$ (black solid curve), $\Delta m = 0$ (dotted green curve), $\Delta m = -1$ (dot-dashed red curve), $\Delta m = -2$ (long-dashed purple curve). $\Lambda = 1$ (RCP) in both plots.

Results for absorption rates

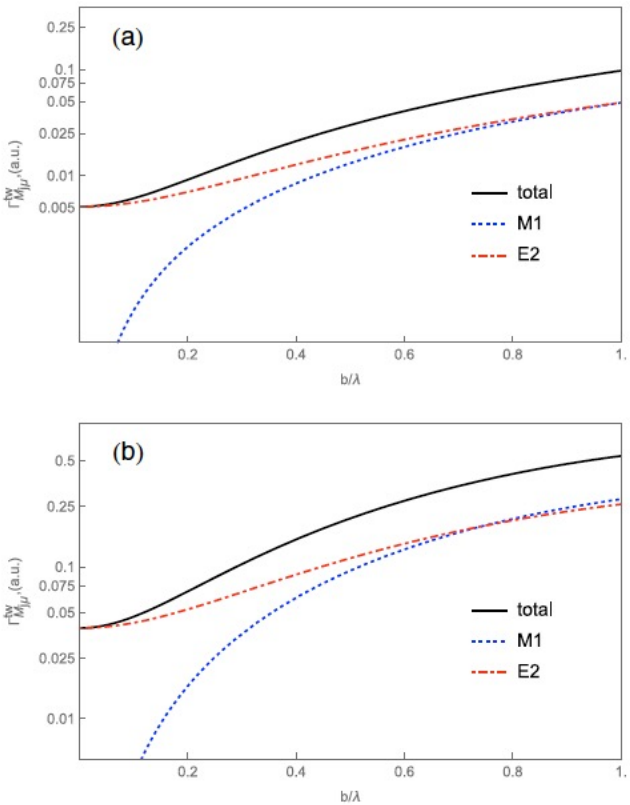

FIG. 3. Log plots of photoabsorption rates in Boron-like HCI for pitch angles (a) $\theta_k = 0.1$ and (b) 0.2. The transitions are excited by twisted photons with Bessel profile, $m_\gamma = 2$, and right-handed helicity ($\Lambda = 1$).

The absorption cross section (=rate/flux) has a singular term in ~1/b^2 *independent* of the pitch angle

$$\sigma^{(\mathrm{tw})}_{m_\gamma=2} \to \left(4\pi(E2^2 + M1^2) + \frac{4E2^2}{(b/\lambda)^2\pi}\right) + O(\theta_k^2),$$

$$\sigma^{(\mathrm{tw})}_{m_\gamma=3} \to \left(4\pi(E2^2 + M1^2) + \frac{16E2^2}{(b/\lambda)^2\pi}\right) + O(\theta_k^2),$$

$$\sigma^{(\mathrm{pw})} = 4\pi(E2^2 + M1^2),$$

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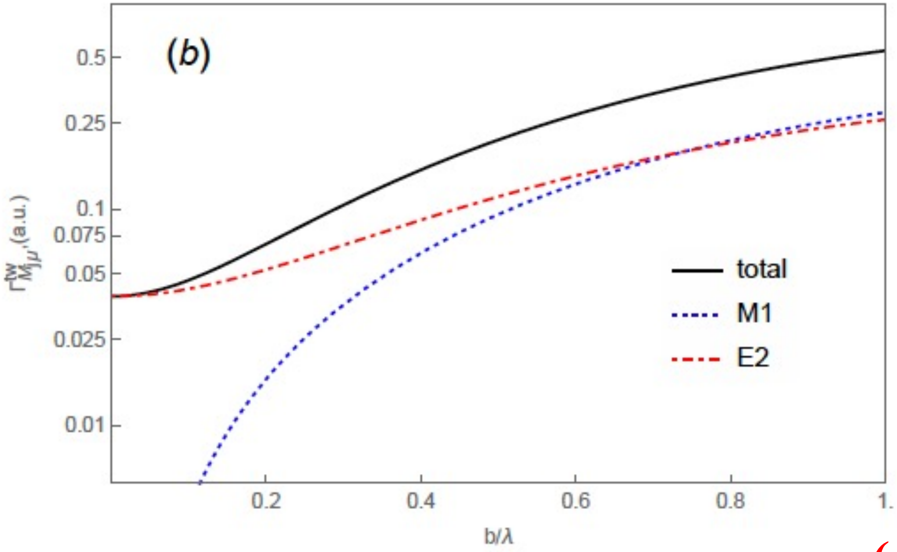
Separation of Multipolar Transitions with Twisted Photons

- Andrei Afanasev, Carl E. Carlson, and Maria Solyanik, Phys. Rev. A 97, 023422 (2018) arXiv:1801.03227, *Separation of M1-E2 Mixed Multipoles*

see also Schulz et al. PRA **102**, 012812 (2020) on E3 transitions

  - Theoretical analysis demonstrates that it is possible to extract the relative transition rates of different multipolar contributions by measuring the photo-excitation rate as a function of the atom's position (or the impact parameter) with respect to the optical vortex center.



Absorption rates with Boron-like Highly-charged ions: M1/E2=1.1 for plane-wave photoexcitation

The approach can be extended to separation of multipoles in molecules, nanoparticles or nuclei (see Kirschbaum et al, arXiv:2404.13023

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Circular Dichroism in Atomic Transitions

Afanasev, Carlson, Solyanik J.Opt. 19 (2017) 105401

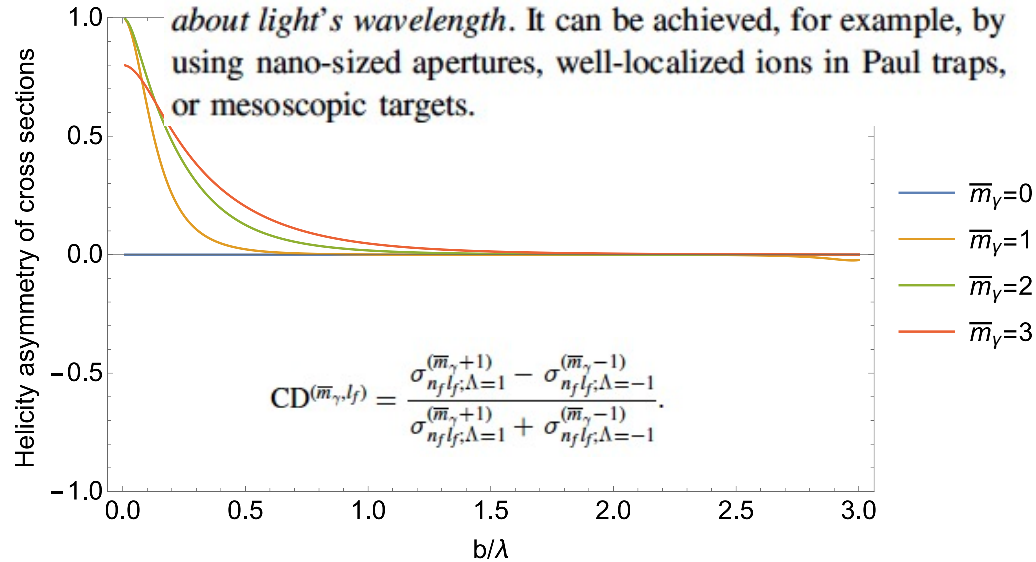
Photon states with aligned spin and OAM are more likely to be absorbed-> dichroism

However, opposite spin asymmetry at beam periphery =>net asymmetry is zero!

Different cross sections for opposite spin projections (while OAM is unchanged)

- No dichroism for E1-transitions
- Strong dichroism near vortex center for E2 transitions and higher

observation of spin-asymmetric absorption of twisted light by atomic matter requires localization of the target atoms within about light's wavelength. It can be achieved, for example, by using nano-sized apertures, well-localized ions in Paul traps, or mesoscopic targets.



$$\mathrm{CD}^{(\overline{m}_\gamma, l_f)} = \frac{\sigma^{(\overline{m}_\gamma+1)}_{n_f l_f;\Lambda=1} - \sigma^{(\overline{m}_\gamma-1)}_{n_f l_f;\Lambda=-1}}{\sigma^{(\overline{m}_\gamma+1)}_{n_f l_f;\Lambda=1} + \sigma^{(\overline{m}_\gamma-1)}_{n_f l_f;\Lambda=-1}}.$$

Implications: Parity-conserving birefringence and dichroism of an optical vortex in the *isotropic* medium

David Andrews, Kayn Forbes, Optics Letters (2018), on Circular Vortex Dichroism (CVD) in chiral matter; here we considered $(E2)^2$ contribution due to chirality of *the beam,* see also Phys. Rev. A 99, 023837 (2019), J. Phys. Photonics 3 022007 (2021)

- *NB:* No effect if integrated over the wavefront; largest effects for ~wavelength radius around vortex center

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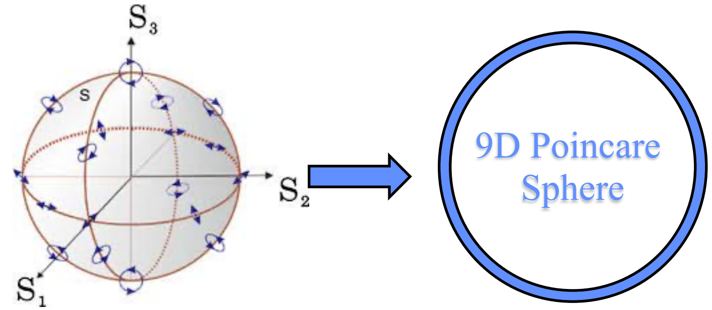
Spin-density matrix of a polarized vector vortex beam



- **AA, Carlson, Wang, J. Opt. 22 054001 (2020)**
- Spin wave function of spin-1 particle, a pure state

$$|\chi_1\rangle = a^+|\chi_{1+}\rangle + a^0|\chi_{10}\rangle + a^-|\chi_{1-}\rangle \qquad a^{\pm} = \frac{(\mp a_x + i a_y)}{\sqrt{2}},\ a_0 = a_z,\ \sum_i a_i a_i^* = 1$$

- Spin-density matrix for pure or non-pure states

$$\rho_{ij} = a_i a_j^*$$

where $\mathcal{P}_i$, $\mathcal{P}_{ij}$ are the operators of spin and quadrupole moment, and p_i, p_{ij} are corresponding vector and quadrupole polarizations that can be expressed in terms of the above amplitudes a_i and $a^{\pm,0}$,

$$p_i = i\epsilon_{ijk} a_i a_k^*;\ p_{ik} = -\frac{3}{2}(a_i a_k^* + a_k a_j^* - \frac{2}{3}\delta_{ik})$$

$$T_{20} = \frac{1}{\sqrt{2}} \frac{|a_+|^2 + |a_-|^2 - 2|a_0|^2}{|a_+|^2 + |a_-|^2 + |a_0|^2}$$

$$\rho = \frac{1}{3}\Big\{I + \frac{3}{2}(p_x\mathcal{P}_x + p_y\mathcal{P}_y + p_z\mathcal{P}_z) + \frac{2}{3}(p_{xy}\mathcal{P}_{xy} + p_{yz}\mathcal{P}_{yz} + p_{xz}\mathcal{P}_{xz}) + \frac{1}{6}(p_{xx} - p_{yy})(\mathcal{P}_{xx} - \mathcal{P}_{yy}) + \frac{1}{2}p_{zz}\mathcal{P}_{zz}\Big)$$

The following relations hold between spin density matrices of the twisted photons and the excited $l_f = 1$ state:

$$p_{xx}^{(\gamma)} - p_{yy}^{(\gamma)} = p_{xx}^{(e)} - p_{yy}^{(e)},$$
$$p_{zz}^{(\gamma)} = p_{zz}^{(e)},\ p_{xy}^{(\gamma)} = p_{xy}^{(e)}$$
$$p_{xz}^{(\gamma)} = -p_{xz}^{(e)},\ p_{yz}^{(\gamma)} = -p_{yz}^{(e)}$$
$$p_z^{(\gamma)} = p_z^{(e)},\ p_{x,y}^{(\gamma)} = -p_{x,y}^{(e)}$$

In presence of quadrupole interaction each polarization component evolves, resulting in new types of birefringence and dichroism; formalism applies to mixed quantum states

Octofringence and octochroism?

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Examples of atomic alignment parameters (E1 transition)

E1-transitions:
Spin-density matrix of optically polarized atoms
in one-to-one correspondence with photon's

coherent superposition of states with ($m_\gamma = -2$, $\Lambda = 1$) and ($m_\gamma = 3$, $\Lambda = -1$).

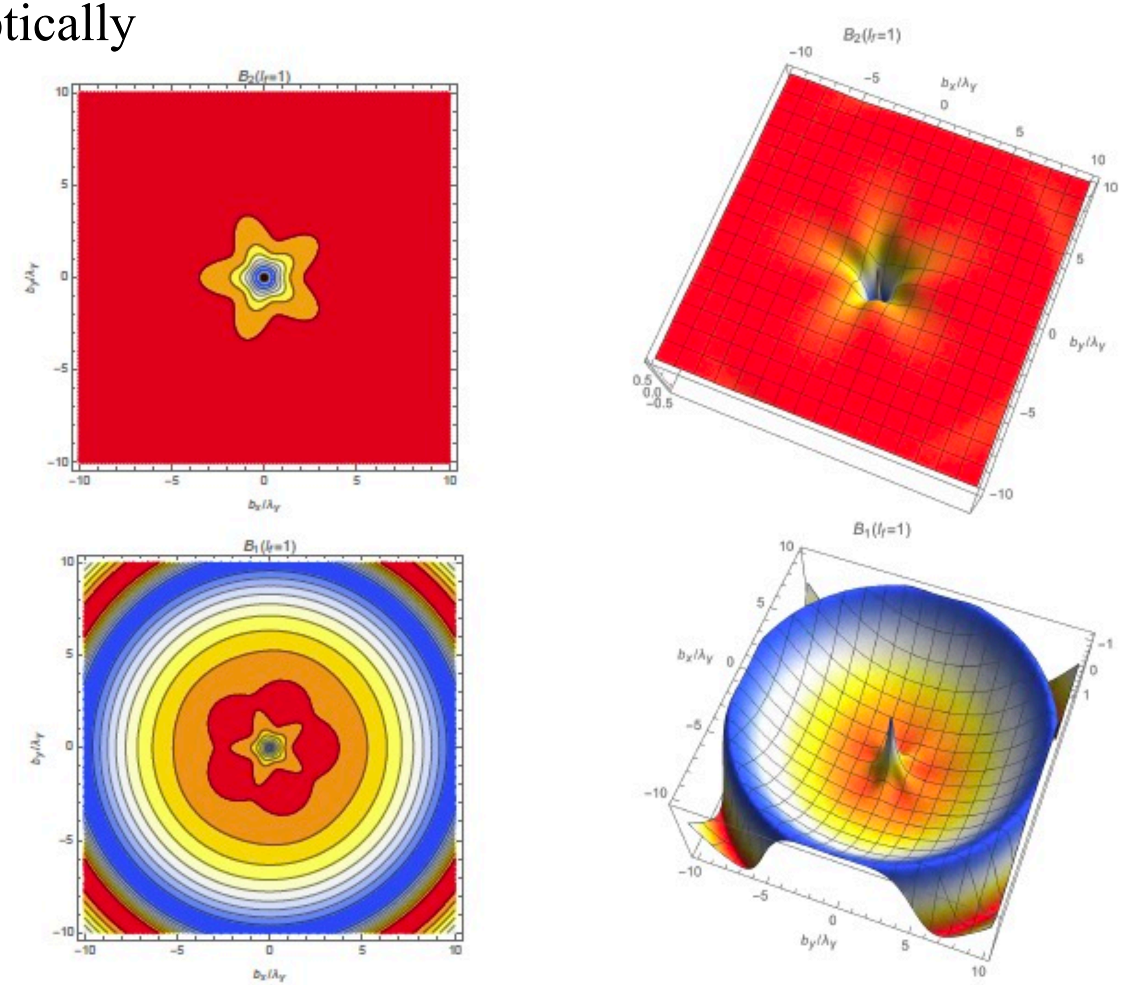


FIG. 2. Plots of the alignment parameters $B_2(1)$ and $B_1(1)$, top to bottom, for $S \to P$ transitions, with contour plots on the left and 3D versions of the same on the right. Each plot shows the B_k parameter as a function of the impact parameter components b_x and b_y measured in wavelengths of the incident light beam.

Examples of atomic alignment parameters (E2 transition)

coherent superposition of states with ($m_\gamma = -2$, $\Lambda = 1$) and ($m_\gamma = 3$, $\Lambda = -1$).

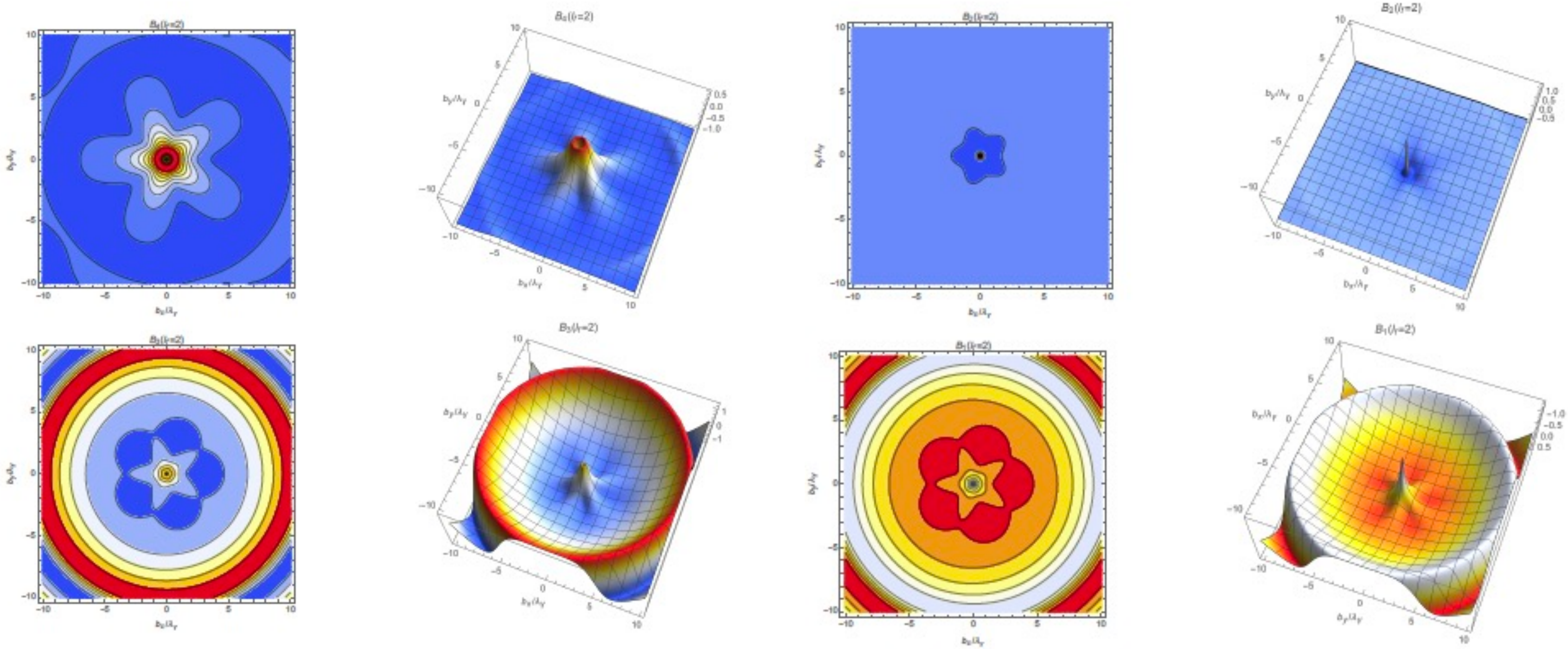
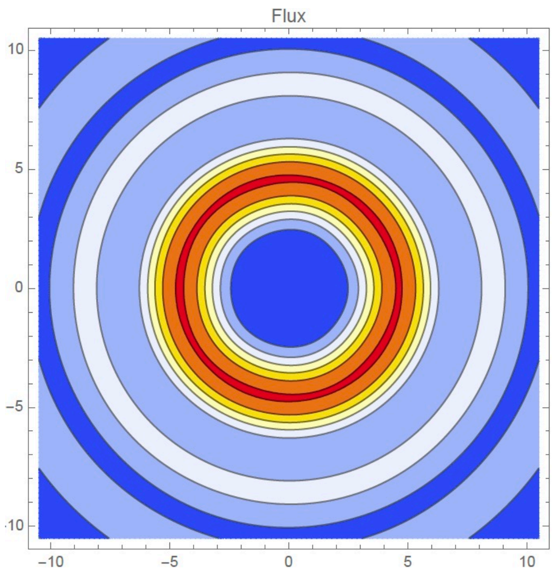
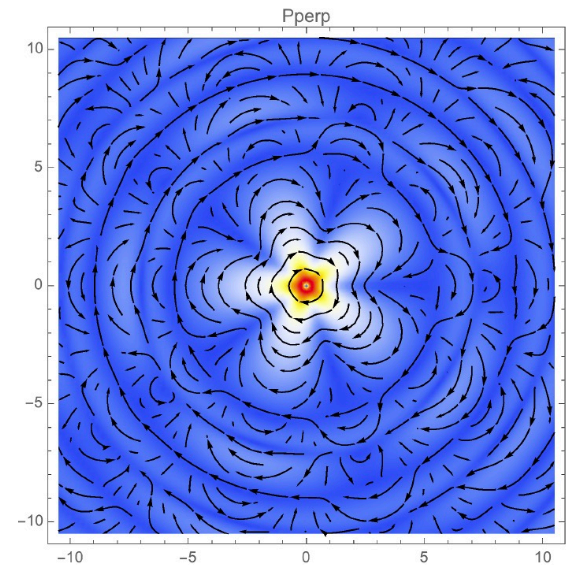


FIG. 3. Plots of the alignment parameters $B_4(2)$, $B_3(2)$, $B_2(2)$, and $B_1(2)$, top to bottom, for $S \to D$ transitions, with contour plots on the left and 3D versions of the same on the right. Again, each plot shows the B_k parameter as a function of the impact parameter components b_x and b_y measured in wavelengths of the incident light beam.

Transverse vector polarization of atoms by twisted light



Energy density



Transverse vector polarization (=zero for plane waves)

- one-to-one correspondence between photon's and atom's polarizations in E1-,M1-transitions
  - Different for higher multipoles
- spatial extent of transverse-polarization region is independent of beam waist (see next slide)

Independence of beam waist also shows in transition amplitudes near singularity

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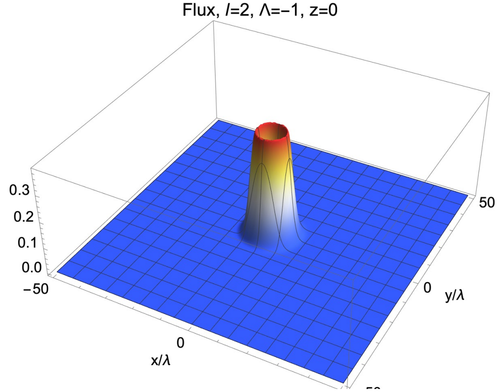
Non-Diffractive Polarization Features - L/T Ratio

- Consider 3D-polarization parameters (longitudinal fields included!)

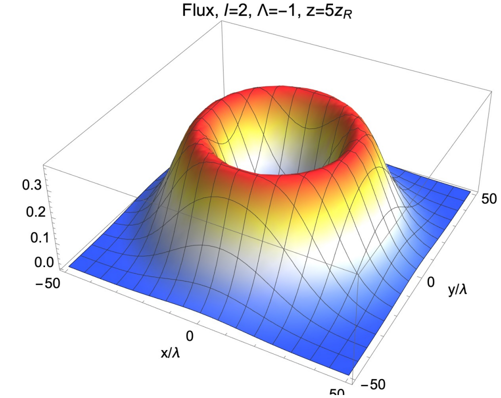
$$p_z = \frac{|E^{\mu}_{+1}|^2 - |E^{\mu}_{-1}|^2}{|E^{\mu}_{+1}|^2 + |E^{\mu}_{-1}|^2 + |E^{\mu}_{0}|^2}.$$

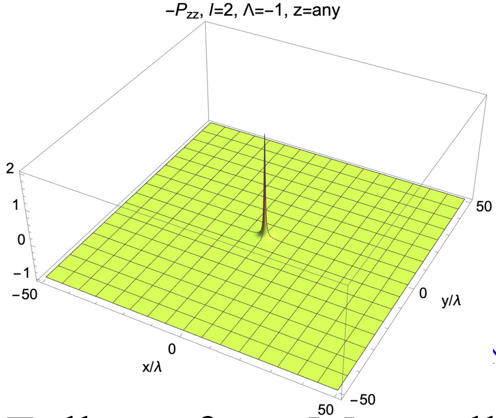
$$p_{zz} = \frac{|E^{\mu}_{+1}|^2 + |E^{\mu}_{-1}|^2 - 2|E^{\mu}_{0}|^2}{|E^{\mu}_{+1}|^2 + |E^{\mu}_{-1}|^2 + |E^{\mu}_{0}|^2}.$$

- *No transverse expansion due to propagation*, AA, Kingsley-Smith, Rodriguez-Fortuno, Zayats, Advanced Photonics Nexus **2**,026001 (2023)

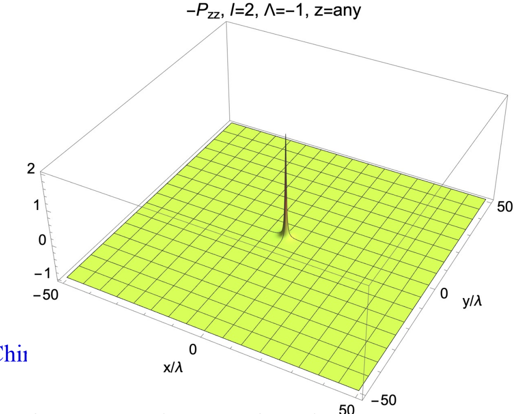


Propagation





Propagation



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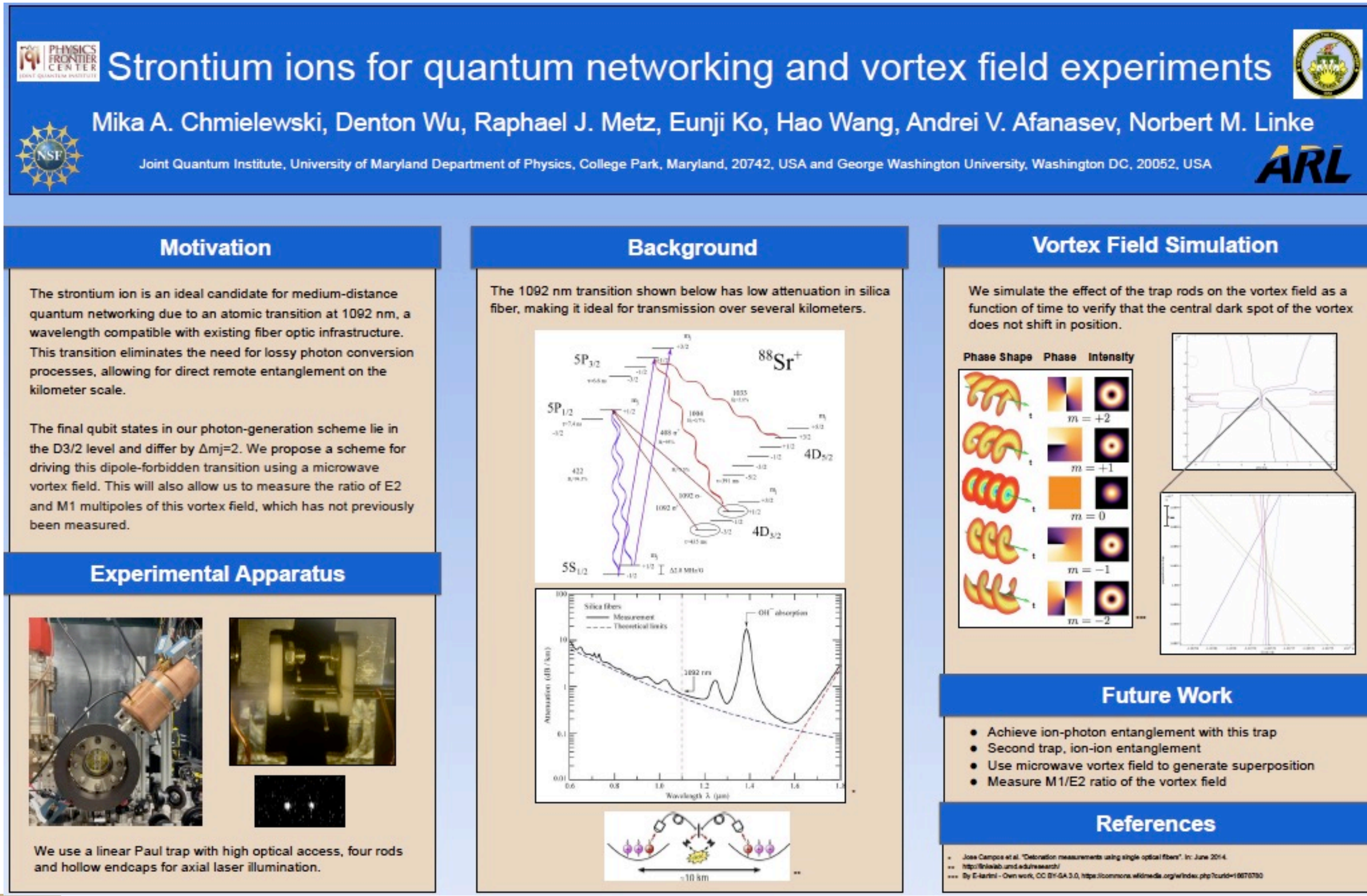
Follows from Maxwell's equations near phase singularity

Application considered for Quantum Networking

APS Division of Atomic and Molecular Physics Meeting 2023, abstract id.N01.065



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Summary

- Novel features of interaction between twisted photons and atomic matter
  - Excitation of states with a range of quantum numbers, different from plane waves
  - Modified quantum selection rules validated by Mainz measurements with trapped $^{40}Ca^{+}$
  - Can drive selected plane-wave forbidden $\Delta m=\pm2$ atomic transitions, while suppressing $\Delta m=\pm1$
  - Localization of the target atom is essential for modified twisted vs plane-wave selection rules
  - Circular dichroism of twisted photons takes place even in non-chiral matter
  - L/T field ratio and circular vortex dichroism show (non-diffractive) scaling against propagation – i.e., independence of beam waist
  - Possible applications include quantum computing and quantum networks
  - Nuclear transitions: Kirschbaum et al, arXiv:2404.13023 (todays talk)

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