Comment on "Quantized Orbital Angular Momentum Transfer and Magnetic Dichroism in the Interaction of Electron Vortices with Matter"

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Lloyd et al. [1] (LBY) find that - contrary to the case of optical vortices - transfer of orbital angular momentum (OAM) from vortex electrons to the electronic degrees of freedom of an atom is possible, and thus explain the observed EMCD effect [2] [3].

However, the experimental consequences discussed by LBY deserve a comment. We refer to equations in the said paper by "L", followed by the Eq. number; LBY’s notation is adopted.

Concentrating on electric dipole transitions in EELS we note that they do not depend on the condition

\[ |q| \ll |r - R| \]  

used to derive L8, see e.g. [4]. In fact, the electric dipole scattering kernel peaks at \(|r - R| \sim |q|\), i.e. when the probe electron passes close to the atom electron [5]. Therefore we use the exact Hamiltonian L6. In the experimentally relevant case of rigidly fixed atoms that LBY assume after L13, the atom is approximated by a spatial eigenstate at coordinate \(R_0\) [6]

\[ \langle R|\psi_n\rangle \doteq \delta^3(R - R_0) \]

and we can integrate the nucleus contribution in Eq. L7, leading to a transition matrix element between orthogonal initial and final internal states \(|\psi_e\rangle\) and \(|\psi_f\rangle\)

\[ \mathcal{M}_{if} = \langle \psi_e^* \psi_B | H_{\text{int}} | \psi_e \psi_B \rangle. \]

The vortex \(|\psi_B\rangle\) Eq. L5 is a Bessel beam. The Hamiltonian depends on the electronic coordinate \(q\) in the center of mass system and the vortex coordinate \(r\) in the vortex centered system:

\[ H_{\text{int}} = \frac{e^2}{4\pi\varepsilon_0} \frac{1}{|r - R_0 - q|}. \]

The term in Eq. L6 containing the atom coordinate \(R_0\) vanished because \(\langle \psi_e^* | \psi_f' \rangle = 0\). The matrix element L7 reads

\[ \mathcal{M}_{if} = \int d^2r J_l(k_p r) J_{l'}(k_{p'} r) \int d^2q u(q) u^*(q) e^{i((m-m')\phi_q + (l-l')\Phi_{p'})} \frac{1}{|r - R_0 - q|}. \]  

\(\phi_q, \Phi_{p'}\) are the azimuthal angles of the respective vectors, \(u\) are the radial parts of the atom electron’s wave function. Substitution of atom-centered coordinates \(r' = r - R_0\) and the addition theorem for Bessel functions \(J_l\) yield

\[ \mathcal{M}_{if} = \sum_{p, p', \rho, \rho' } \mathcal{J}_p(k_p R_0) \mathcal{J}_{p'}(k_{p'} R_0) \int d^2r' J_{l+p}(k_p r') J_{l'+p'}(k_{p'} r') \int d^2q u(q) u^*(q) e^{i((m-m')\phi_q + (l+l' - p - p')\phi_{p'})} \frac{1}{|r' - q'|}. \]

Without loss of generality \(R_0\) points along the \(x\) direction of the reference frame. Both azimuthal angles \(\phi_q, \phi_{p'}\) refer to the center of the atom. Substituting \(\varphi = \phi_{p'} - \phi_q\) in the Coulomb term

\[ [r'^2 + q^2 - 2r'q \cos(\phi_{p'} - \phi_q)]^{-1/2} \]

\(LBY\)’s azimuthal component Eq. L10 reads

\[ \mathcal{M}_{az} = \int_0^{2\pi} d\varphi e^{i\lambda\varphi} F(r', q, \varphi) \int_0^{2\pi} d\phi_q e^{i(\lambda + \alpha)\phi_q} \]

with \(\lambda = l + p - l' - p'\) and \(\alpha = m - m'\). The second integral vanishes except for \(\lambda = -\alpha\), giving rise to selection rules for dipole transitions \(\alpha = \pm 1\)

\[ l' = l \pm 1 + p - p'. \]

Since \(p - p'\) spans the integer range, dipole transitions to any final vortex state \(l'\) are possible; the outgoing probe electron is not in an OAM eigenstate [7].

The conclusion of LBY must therefore be modified: electric dipole transitions mediate the transfer of OAM, but in general, the transfer is not quantized.

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To reconcile this result with the findings of LBY and with the experimental evidence \[2\] that vortices can be used to detect chiral electronic transitions it is sufficient to reconsider Eq. 4 in the limit \(k_p R_0 \to 0\): Since \(J_p(0)\) vanishes except for \(p = 0\) the sum over \(p, p'\) collapses into a single term, and the selection rule Eq. 5 reads \(l' = l - \alpha\) which is L12 for \(L = L'\). The matrix elements violating these selection rules will be small for small displacements \(|R_0|\) of the atom from the vortex core. This means that the larger the observed cluster the fainter is the EMCD effect.

Furthermore, an electron vortex can exchange OAM with the crystal lattice so that neither the assumption of an incident nor that of an outgoing OAM eigenstate are fulfilled in practice \[7\].

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[1] S. Lloyd, M. Babiker, and J. Yuan, Physical Review Letters 108, 074802 (2012)
[2] J. Verbeeck, H. Tian, and P. Schattschneider, Nature 467, 301 (2010)
[3] P. Schattschneider, B. Schaffer, I. Ennen, and J. Verbeeck, Physical Review B 85, 134422 (2012)
[4] S. T. Manson, Physical Review A 6, 1013 (1972)
[5] P. Schattschneider, M. Nelhiebel, and B. Jouffrey, Physical Review B 59, 10959 (1999)
[6] H. A. Bethe, Ann. Phys 5, 325 (1930)
[7] S. Löfler and P. Schattschneider, Acta Cryst. A68, 443 (2012)
[8] S. Franke-Arnold, S. M. Barnett, E. Yao, J. Leach, J. Courtial, and M. Padgett, New Journal of Physics 6, 1 (2004)
[9] This is a consequence of the uncertainty relation: The localisation of the atom electron to an angle \(\Delta \Phi_R \sim |q|/|R_0|\) as seen from the vortex center induces an uncertainty \(\Delta l' \geq 1/\Delta \Phi_R\), see \[8\].