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

Lloyd, Babiker, and Yuan (LBY) [1] 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 energy-loss magnetic chiral dichroism effect [2,3].

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

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

$$|\mathbf{q}| \ll |\mathbf{r} - \mathbf{R}| \tag{1}$$

used to derive (L8); see, e.g., Ref. [4]. In fact, the electric dipole scattering kernel peaks at $|\mathbf{r} - \mathbf{R}| \sim |\mathbf{q}|$, 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 $\mathbf{R}_0$ [6],

$$\langle R|\psi_n\rangle \doteq \delta^3(\mathbf{R} - \mathbf{R}_0),$$

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

$$\mathcal{M}_{if} = \langle \psi'_e \psi'_B | H_{\text{int}} | \psi_e \psi_B \rangle.$$

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

$$H_{\text{int}} = \frac{e^2}{4\pi\varepsilon_0} \frac{1}{|\mathbf{r} - \mathbf{R}_0 - \mathbf{q}|}. \tag{2}$$

The term in (L6) containing the atom coordinate $\mathbf{R}_0$ vanished because $\langle \psi_e | \psi'_e \rangle = 0$. The matrix element (L7) reads

$$\mathcal{M}_{if} = \int d^2 r J_l(k_\rho r) J_{l'}(k_{\rho'} r) \times \int d^2 q u(q) u'^*(q) e^{i[(m-m')\phi_q + (l-l')\Phi_r]} \frac{1}{|\mathbf{r} - \mathbf{R}_0 - \mathbf{q}|}. \tag{3}$$

ϕ_q, Φ_r 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, $\mathbf{r}' = \mathbf{r} - \mathbf{R}_0$, and the addition theorem for Bessel functions J_l yield

$$\mathcal{M}_{if} = \sum_{p,p',\rho,\rho'} J_p(k_\rho R_0) J_{p'}(k_{\rho'} R_0) \times \int d^2 r' J_{l+p}(k_\rho r') J_{l'+p'}(k_{\rho'} r') \times \int d^2 q u(q) u'^*(q) e^{i[(m-m')\phi_q + (l+p-l'-p')\phi'_r]} \frac{1}{|\mathbf{r}' - \mathbf{q}|}. \tag{4}$$

Without loss of generality, $\mathbf{R}_0$ points along the x direction of the reference frame. Both azimuthal angles ϕ_q, ϕ'_r refer to the center of the atom. Substituting $\varphi = \phi'_r - \phi_q$ in the Coulomb term

$$[r'^2 + q^2 - 2r'q\cos(\phi'_r - \phi_q)]^{-1/2} =: F(r', q, \phi_{r'} - \phi_q),$$

LBY's azimuthal component (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 $\alpha = \pm 1$:

$$l' = l \pm 1 + p - p'. \tag{5}$$

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.

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 $k_\rho R_0 \to 0$: 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 $|\mathbf{R}_0|$ of the atom from the vortex core. This means that the larger the observed cluster, the fainter the energy-loss magnetic chiral dichroism 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 is fulfilled in practice [9].

P. S. and S. L. acknowledge financial support by the Austrian Science Fund, Project No. I543-N20. J. V. acknowledges funding from the European Research Council under the 7th Framework Program (FP7), ERC Grant No. N246791-COUNTATOMS, and ERC Starting Grant No. 278510 VORTEX.

P. Schattschneider*
Institute of Solid State Physics
Vienna University of Technology
A-1040 Wien, Austria
and Ecole Centrale Paris
LMSSMat (CNRS UMR 8579)
F-92295 Châtenay-Malabry, France

S. Löffler
University Service Center for Transmission Electron Microscopy
Vienna University of Technology
A-1040 Wien, Austria

J. Verbeeck
EMAT, University of Antwerp
Groenenborgerlaan 171
2020 Antwerp, Belgium

Received 11 July 2012; published 1 May 2013
DOI: 10.1103/PhysRevLett.110.189501
PACS numbers: 41.85.−p, 42.50.Tx

*schattschneider@ifp.tuwien.ac.at

[1] S. Lloyd, M. Babiker, and J. Yuan, Phys. Rev. Lett. **108**, 074802 (2012).

[2] J. Verbeeck, H. Tian, and P. Schattschneider, Nature (London) **467**, 301 (2010).

[3] P. Schattschneider, B. Schaffer, I. Ennen, and J. Verbeeck, Phys. Rev. B **85**, 134422 (2012).

[4] S. T. Manson, Phys. Rev. A **6**, 1013 (1972).

[5] P. Schattschneider, M. Nelhiebel, and B. Jouffrey, Phys. Rev. B **59**, 10959 (1999).

[6] H. Bethe, Ann. Phys. (Weinheim) **397**, 325 (1930).

[7] This is a consequence of the uncertainty relation: The localization of the atom electron to an angle $\Delta\Phi_R \sim |\mathbf{q}|/|\mathbf{R}_0|$ as seen from the vortex center induces an uncertainty $\Delta l' \geq 1/\Delta\Phi_R$, see Ref. [8].

[8] S. Franke-Arnold, S.M. Barnett, E. Yao, J. Leach, J. Courtial, and M. Padgett, New J. Phys. **6**, 103 (2004).

[9] S. Löffler and P. Schattschneider, Acta Crystallogr. Sect. A **68**, 443 (2012).