Lloyd, Babiker, and Yuan Reply: In the preceding Comment [1], Schattschneider, Löffler, and Verbeeck (SLV) raise some objections to the results and conclusions in our Letter [2]. We shall refer to equations in Ref. [1] by "S," followed by the equation number. SLV begin by emphasizing that electric dipole transitions in EELS do not depend on the condition $|\mathbf{q}| \ll |\mathbf{r} - \mathbf{R}|$. In fact, this is the standard definition of the electric dipole approximation which can be seen as arising from the expansion of the electric polarization field $\mathbf{P}(\mathbf{r})$ for a single bound electron in exact multipolar form [3,4]:

$$\mathbf{P}(\mathbf{r}) = -e \int_0^1 d\lambda \mathbf{q} \delta\{\mathbf{r} - \mathbf{R} - \lambda \mathbf{q}\}. \quad (1)$$

It is easy to see that under the condition $|\mathbf{q}| \ll |\mathbf{r} - \mathbf{R}|$ an expansion of $\mathbf{P}(\mathbf{r})$ in powers of $\mathbf{q}$ retaining only the leading (dipole) term yields

$$\mathbf{P}(\mathbf{r}) \approx -e\mathbf{q}\delta(\mathbf{r} - \mathbf{R}). \quad (2)$$

This is consistent with standard quantum treatments of an atomic transition where the dipole term involves a single power of $\mathbf{q}$ [5]. The higher multipoles arise from the higher order terms in the expansion of $\mathbf{P}(\mathbf{r})$.

The standard definition of the dipole interaction stands even under conditions in which $|\mathbf{q}|$ is of the order of $|\mathbf{r} - \mathbf{R}|$. The dipole term is then not necessarily the dominant term in the multipolar expansion, but this does not prevent exploration of the exchange of orbital angular momentum (OAM). The selection rules derived in Ref. [2] have now been confirmed using a different approach [6]. We therefore argue that the objection to the use of Eq. (1) in Ref. [2] is not warranted, and arises from a misunderstanding of the main objectives in Ref. [2], where we have shown that an exchange does indeed arise involving a single unit of OAM. This is an important finding in view of the earlier result that the dipole excitation of an atom by an optical vortex beam involves no exchange of OAM between the optical vortex and the internal dynamics of the atom. This distinguishes our approach from that of SLV. SLV's Comment is also more focused on the change of OAM of the vortex beam due to the interaction, which, albeit experimentally important, is a complex issue that was beyond the scope of Ref. [2]. However, we have examined this matter carefully and the analysis and the results are reported in Ref. [6]. Furthermore, we do have some further comments to make on SLV's analysis as presented in Ref. [1]. The matrix element in S(4) appears to have been derived without applying the addition theorem of Bessel functions properly in transforming the vortex beam wave function from the laboratory frame of reference (in terms of $\mathbf{r}$) to the atom center of mass frame (in terms of $\mathbf{r}'$). Work in hand [6] shows that a proper expansion of the Bessel functions yields, adopting the notation of SLV,

$$\mathcal{M}_{fi} \propto \sum_{p,p'=-\infty}^{\infty} e^{i(l-l'-p+p')\phi_R} \times \int_0^{2\pi}\int_0^{2\pi} \frac{1}{|\mathbf{r}' - \mathbf{q}|} e^{i(p-p')\phi'_r} e^{i(m-m')\phi_q} d\phi'_r d\phi_q. \quad (3)$$

The atomic electron now interacts with a series of vortex modes p, scattered into states p', but this fact suggests that the selection rule stated in S(5) cannot be correct for this interaction. Following SLV's approach leading to S(4), writing $\varphi = \phi'_r - \phi_q$ and integrating over φ first, followed by integration over ϕ_q, we find $\delta p = -\delta m$, so that, as SLV concluded, the p modes may exchange OAM with the atom. However, neither SLV's result nor $\delta p = -\delta m$ given here is a sensible selection rule. In order to properly arrive at a sensible selection rule one needs to eliminate p and p'. The problem has its roots in the assumption of ignoring the center of mass as a dynamical variable. The correct selection rules involving the change in the beam OAM δl can be derived only by reintroducing the centre of mass as a variable. This step, along with the identification of the dipole and higher multipole terms, is addressed in Ref. [6].

Finally, SLV's Comment questioned the suitability of the vortex beam description when interacting with atoms in a crystalline material. While this is an issue beyond the scope of Ref. [2], it is worth noting that the vortex beam description could be experimentally useful for crystalline materials oriented in kinematic conditions where Bloch waves are not strongly excited by the incident electron beam.

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