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Reply to "Comment on 'Ultrafast terahertz-field-driven ionic response in ferroelectric $BaTiO_3$' "

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In this reply to S. Durbin's comment on our original paper "Ultrafast terahertz-field-driven ionic response in ferroelectric $BaTiO_3$," we concur that his final equations 8 and 9 more accurately describe the change in diffracted intensity as a function of Ti displacement. We also provide an alternative derivation based on an ensemble average over unit cells. The conclusions of the paper are unaffected by this correction.

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We thank Durbin for his comment on the paper, "Ultrafast terahertz-field-driven ionic response in ferroelectric $BaTiO_3$" [1]. We are in agreement that his final Eqs. (8) and (9) more accurately describe the change in diffracted intensity as a function of the THz-driven Ti atom displacement. We first emphasize that this result does not change the conclusions of the paper, only slightly modifying the estimated incoherent Ti atom displacement required to explain the observed results; the rms time-dependent displacement is still 0.03 Å within experimental resolution.

Since extraction of the THz-driven time-dependent displacement is a somewhat subtle calculation which has not appeared previously, we explain briefly in the following an alternative explanation that we believe more transparently leads to the correct result, consistent with Durbin's final equations. We begin from Eq. (2) of Ref. [1] for the time-dependent structure factor with

$$F = f_{\mathrm{Ba}} - f_{\mathrm{Ti}} e^{-6\pi i\delta} e^{-6\pi i A(t)}, \tag{1}$$

where f_{Ba} and f_{Ti} are the scattering factors for the Ba and Ti atoms, δ is the static displacement of the central Ti atom, and A(t) is the induced time-dependent Ti atom displacement. One needs to first average this structure factor over many unit cells in the case of an incoherent response. This is analogous to adding the scattered electric fields from each unit cell first to allow for potential interference. In this case, one then finds

$$\langle F(t)\rangle = f_{\mathrm{Ba}} - f_{\mathrm{Ti}} e^{-6\pi i\delta} \langle e^{-6\pi i A(t)}\rangle. \tag{2}$$

If the time-dependent displacements A(t) are distributed spatially in a roughly Gaussian manner [2], then $\langle e^{-6\pi i A(t)}\rangle = e^{-36\pi^2\langle A^2(t)\rangle}$ and one obtains for the unit cell structure factor averaged over all unit cells:

$$\langle F(t)\rangle = f_{\mathrm{Ba}} - f_{\mathrm{Ti}} e^{-6\pi i\delta} e^{-M_{\mathrm{Ti}}}, \tag{3}$$

where $M_{\mathrm{Ti}} = 18\pi^2\langle A^2(t)\rangle$, consistent with Eq. (6) of Durbin. We note, however, that no time average comes into this

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calculation. Given the time-resolved nature of the experiment, it is much more straightforward to consider this as an incoherent spatial average over unit cells. From Eq. (3), one then obtains the final result for the induced time-dependent scattered intensity, i.e., the measured signal from $I = |\langle F(t)\rangle|^2$.

In conclusion, the final result of Durbin more precisely quantifies the induced Ti atom displacement. In the derivation above, the ensemble average is performed over the spatial distribution of atomic displacements rather than over time as is required for the time-resolved measurements described.

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