

Correction to “Basic Theory of Surface Sum-Frequency Generation”

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The definition of $\Delta\vec{k}$ in eq 1 in the paper should be $\Delta\vec{k} = \vec{k} - (\vec{k}_1 + \vec{k}_2)$ instead of $\Delta\vec{k} = (\vec{k}_1 + \vec{k}_2) - \vec{k}$. There is also a mistake in the paper inherited from a previous publication (ref 20): Derivation of eqs 1, 14, and 15, involves an integration of the bulk sum-frequency response, $\vec{P}_B^{(2)}(\omega, z) dz$ or $\vec{\chi}_B^{(2)}(\omega, z) dz$, over medium II from $z = 0^-$ to ∞ . The integration, properly done, should yield an effective surface susceptibility given by

$$\begin{aligned}\vec{\chi}_{S,\text{eff}}^{(2)} &\equiv \vec{\chi}_{SS}^{(2)} + \frac{\vec{\chi}_B^{(2)}(\vec{k} \neq \vec{k}_1 + \vec{k}_2)}{i\Delta k_z^{\text{II}}} \\ &= \vec{\chi}_S^{(2)} + \frac{\vec{\chi}_{BB}^{(2)}(\vec{k} = \vec{k}_1 + \vec{k}_2)}{i\Delta k_z^{\text{II}}}\end{aligned}$$

instead of eq 20 in the paper, where $\vec{\chi}_B^{(2)}$, $\vec{\chi}_{BB}^{(2)}$, $\vec{\chi}_S^{(2)}$, and $\vec{\chi}_{SS}^{(2)}$ are defined by eqs 16 and 17 in the paper. The conclusion in the paper must be revised accordingly. We can separately deduce $\vec{\chi}_S^{(2)}$ and $\vec{\chi}_{BB}^{(2)}$ from measurement, but we have $\vec{\chi}_S^{(2)} \equiv \vec{\chi}_{SS}^{(2)} - \vec{\chi}_{BS}^{(2)}$, where $\vec{\chi}_{BS}^{(2)} \equiv \vec{\chi}_q^{(2)}\hat{z}$ in the paper is from the bulk-like electric-quadrupole contribution, intrinsically inseparable from $\vec{\chi}_{SS}^{(2)}$. The inseparability originates from well-known nonuniqueness in defining multipole expansion of a polarization (eq 5 in the paper), first pointed out previously in ref 31 of the paper.

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