

## 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  $\infty$ . 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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