Second Harmonic Generation Optical Rotation Solely Attributable to Chirality in Plasmonic Metasurfaces

Supporting Information

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Supporting Information 1. Identifying Chirality Tensor Components

A reflection in the *x*-*z* plane can be described by the transformation matrix given in equation (1).

$$A_{ij}^{y} = \begin{bmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} \qquad (1)$$

This reflection can then be applied to the nonlinear susceptibility tensor $\chi_{ijk}^{(2)}$ by equation (2).

$$\chi_{ijk}^{\prime(2)} = A_{i\alpha}^{y} A_{j\beta}^{y} A_{k\gamma}^{y} \chi_{\alpha\beta\gamma}^{(2)} \qquad (2)$$

Under mirror symmetry in the *y*-*z* plane, the nonlinear susceptibility tensor $\chi_{ijk}^{(2)}$ should remain unchanged under this transformation, and so we impose the equality given in equation (3).

$$A_{i\alpha}^{y} A_{j\beta}^{y} A_{k\gamma}^{y} \chi_{\alpha\beta\gamma}^{(2)} = \chi_{ijk}^{(2)} \qquad (3)$$

For a general second-order susceptibility tensor with no rotational symmetry, this equality forces components to zero, so that $\chi_{ijk}^{(2)}$ with *y*-*z* mirror symmetry reduces to equation (4).

$$\chi_{ijk}^{(2)} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & \chi_{xzx} & \chi_{xxz} & \chi_{xxy} & \chi_{xyx} \\ \chi_{yxx} & \chi_{yyy} & \chi_{yzz} & \chi_{yyz} & \chi_{yzy} & 0 & 0 & 0 & 0 \\ \chi_{zxx} & \chi_{zyy} & \chi_{zzz} & \chi_{zyz} & \chi_{zzy} & 0 & 0 & 0 & 0 \end{bmatrix} \qquad (4)$$

In the case of indistinguishable incident fields, permutation symmetry applies, and this reduces further to equation (5)

$$d_{il} = \begin{pmatrix} 0 & 0 & 0 & 0 & \chi_{xzx} & \chi_{xyx} \\ \chi_{yxx} & \chi_{yyy} & \chi_{yzz} & \chi_{yzy} & 0 & 0 \\ \chi_{zxx} & \chi_{zyy} & \chi_{zzz} & \chi_{zyz} & 0 & 0 \end{pmatrix} \qquad (5)$$

This reveals the susceptibility tensor components that exclusively appear in chiral structures.