Considerations for arbitrary rotation on $\boldsymbol{\chi}(-2\omega;\omega,\omega)$

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To take the components of $\boldsymbol{\chi}(-2\omega;\omega,\omega)$ from the crystallographic frame to the lab frame, we can simply apply a standard rotational matrix,

$$R = \begin{pmatrix} R_{Xx} & R_{Xy} & R_{Xz} \\ R_{Yx} & R_{Yy} & R_{Yz} \\ R_{Zx} & R_{Zy} & R_{Zz} \end{pmatrix} = \begin{pmatrix} \sin\Phi & -\cos\Phi & 0 \\ \cos\Phi & \sin\Phi & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

such that

$$\chi^{IJK} = \sum_{ijk} R_{Ii}R_{Jj}R_{Kk}\chi^{ijk},$$

where I, J, and K (i, j, k) cycle through X, Y, or Z (x, y, z). Thus, our χ^{IJK} components in terms of the original ijk coordinate system are

$$\begin{aligned}
\chi^{XXX} &= \sin^3\Phi\chi^{xxx} + \sin\Phi\cos^2\Phi\chi^{xyy} - 2\sin^2\Phi\cos\Phi\chi^{xxy} \\
&\quad - \sin^2\Phi\cos\Phi\chi^{yxx} - \cos^3\Phi\chi^{yyy} + 2\sin\Phi\cos^2\Phi\chi^{yxy}, \\
\chi^{XYY} &= \sin\Phi\cos^2\Phi\chi^{xxx} + \sin^3\Phi\chi^{xyy} + 2\sin^2\Phi\cos\Phi\chi^{xxy} \\
&\quad - \cos^3\Phi\chi^{yxx} - \sin^2\Phi\cos\Phi\chi^{yyy} - 2\sin\Phi\cos^2\Phi\chi^{yxy}, \\
\chi^{XZZ} &= \sin\Phi\chi^{xzz} - \cos\Phi\chi^{yzz}, \\
\chi^{XYZ} = \chi^{XZY} &= \sin^2\Phi\chi^{xyz} + \sin\Phi\cos\Phi\chi^{xxz} - \sin\Phi\cos\Phi\chi^{yyz} - \cos^2\Phi\chi^{yxz}, \\
\chi^{XXZ} = \chi^{XZX} &= -\sin\Phi\cos\Phi\chi^{xyz} + \sin^2\Phi\chi^{xxz} + \cos^2\Phi\chi^{yyz} - \sin\Phi\cos\Phi\chi^{yxz}, \\
\chi^{XXY} = \chi^{XYX} &= \sin^2\Phi\cos\Phi\chi^{xxx} - \sin^2\Phi\cos\Phi\chi^{xyy} + (\sin^3\Phi - \sin\Phi\cos^2\Phi)\chi^{xxy} \\
&\quad - \sin\Phi\cos^2\Phi\chi^{yxx} + \sin\Phi\cos^2\Phi\chi^{yyy} + (\cos^3\Phi - \sin^2\Phi\cos\Phi)\chi^{yxy},
\end{aligned} \tag{1}$$

for the χ^{XJK} components,

$$\begin{aligned}
\chi^{YXX} &= \sin^2\Phi\cos\Phi\chi^{xxx} + \cos^3\Phi\chi^{xyy} - 2\sin\Phi\cos^2\Phi\chi^{xxy} \\
&\quad + \sin^3\Phi\chi^{yxx} + \sin\Phi\cos^2\Phi\chi^{yyy} - 2\sin^2\Phi\cos\Phi\chi^{yxy}, \\
\chi^{YYY} &= \cos^3\Phi\chi^{xxx} + \sin^2\Phi\cos\Phi\chi^{xyy} + 2\sin\Phi\cos^2\Phi\chi^{xxy} \\
&\quad + \sin\Phi\cos^2\Phi\chi^{yxx} + \sin^3\Phi\chi^{yyy} + 2\sin^2\Phi\cos\Phi\chi^{yxy}, \\
\chi^{YZZ} &= \cos\Phi\chi^{xzz} + \sin\Phi\chi^{yzz}, \\
\chi^{YYZ} = \chi^{YZY} &= \sin\Phi\cos\Phi\chi^{xyz} + \cos^2\Phi\chi^{xxz} + \sin^2\Phi\chi^{yyz} + \sin\Phi\cos\Phi\chi^{yxz}, \\
\chi^{YXZ} = \chi^{YZX} &= -\cos^2\Phi\chi^{xyz} + \sin\Phi\cos\Phi\chi^{xxz} - \sin\Phi\cos\Phi\chi^{yyz} + \sin^2\Phi\chi^{yxz}, \\
\chi^{YXY} = \chi^{YYX} &= \sin\Phi\cos^2\Phi\chi^{xxx} - \sin\Phi\cos^2\Phi\chi^{xyy} + (\sin^2\Phi\cos\Phi - \cos^3\Phi)\chi^{xxy} \\
&\quad + \sin^2\Phi\cos\Phi\chi^{yxx} - \sin^2\Phi\cos\Phi\chi^{yyy} + (\sin^3\Phi - \sin\Phi\cos^2\Phi)\chi^{yxy},
\end{aligned}$$

for the χ^{YJK} components, and lastly

$$\chi^{ZXX} = \sin^2\Phi\chi^{zxx} + \cos\Phi\cos\Phi\chi^{zyy} - 2\sin\Phi\cos\Phi\chi^{zxy},$$

$$\chi^{ZYY} = \cos^2\Phi\chi^{zxx} + \sin^2\Phi\chi^{zyy} + 2\sin\Phi\cos\Phi\chi^{zxy},$$

$$\chi^{ZZZ} = \chi^{zzz},$$

$$\chi^{ZYZ} = \chi^{ZZY} = \sin\Phi\chi^{zyz} + \cos\Phi\chi^{zxz},$$

$$\chi^{ZXZ} = \chi^{ZZX} = -\cos\Phi\chi^{zyz} + \sin\Phi\chi^{zxz},$$

$$\chi^{ZXY} = \chi^{ZYX} = \sin\Phi\cos\Phi\chi^{zxx} - \sin\Phi\cos\Phi\chi^{zyy} - \cos 2\Phi\chi^{zxy},$$

for the χ^{ZJK} components. Fortunately, the intrinsic permutation symmetry of SHG is also present in the new coordinate system, such that $\chi^{IJK} = \chi^{IKJ}$; therefore, there are only 18 unique components in either system. Setting $\Phi = \pi/2$ signifies that there is no rotation, and thus $\chi^{IJK} = \chi^{ijk}$.

It should also be clear that the crystal symmetries do **not** follow into the rotated system. For instance, the C_{3v} symmetry satisfies the following,

$$\begin{aligned}\chi^{xxx} &= -\chi^{xyy} = -\chi^{yxy},\\ \chi^{yxx} &= \chi^{yyy} = 0.\end{aligned}$$

In the rotated system, the top relationship holds true such that $\chi^{XXX} = -\chi^{XYY} = -\chi^{YXY}$. However, we also obtain that

$$\chi^{YYY} = \cos 3\Phi\chi^{xxx},$$

which is most definitely not zero. Fortunately, we can simply apply the crystal symmetry to the non-rotated system before transforming to the rotated system.

As an example case, we present χ^{XXX} for three values of Φ for a system with C_{3v} symmetry in Fig. 1. The nonzero components in the original coordinates are presented in Fig. 2, multiplied by the appropriate prefactors from Eq. (1). We can see how we recover the component in the original coordinates when $\Phi = \pi/2$.

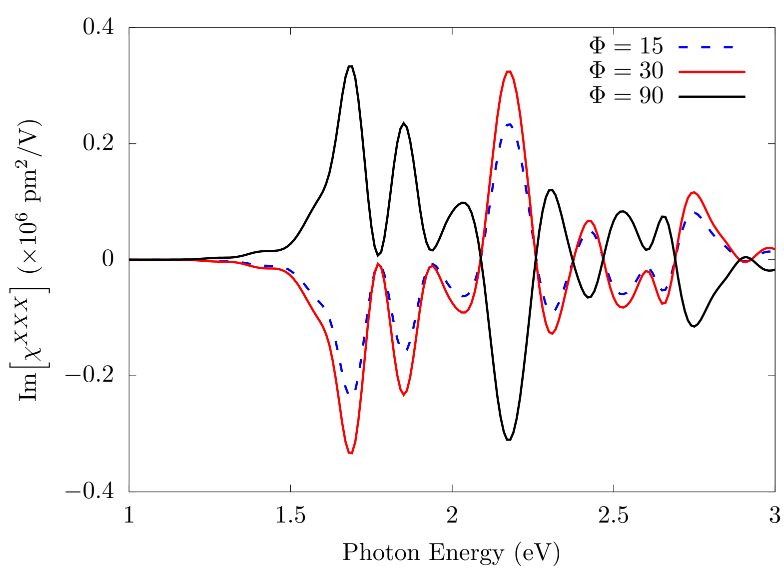
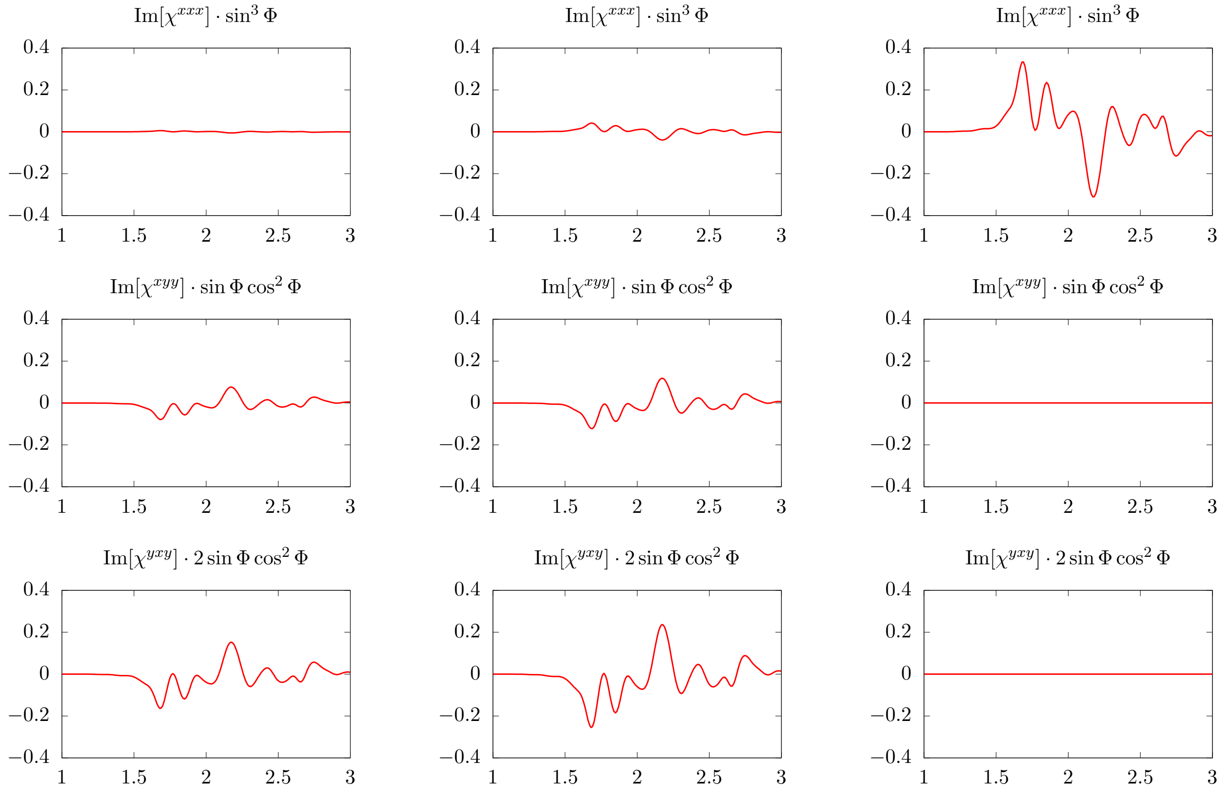


FIG. 1: χ^{XXX} for three values of Φ calculated for a system with C_{3v} symmetry.



(a) $\Phi = \pi/12$. (b) $\Phi = \pi/6$. (c) $\Phi = \pi/2$.

FIG. 2: Nonzero components of χ^{ijk} multiplied by the appropriate prefactors, for three different values of Φ.

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