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

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We develop explicit expressions for rotating the nonlinear susceptibility tensor $\boldsymbol{\chi}(-2\omega;\omega,\omega)$ through any arbitrary angle. Each component in the rotated frame of reference is a combination of different components from the non-rotated system; setting the angle of rotation to $\pi/2$ is equivalent to no rotation whatsoever.

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,

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

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). Fig. 1 depicts this rotation over any arbitrary angle γ. Since we only consider a rotation in the xy-plane along γ, the z and Z axes are the same.

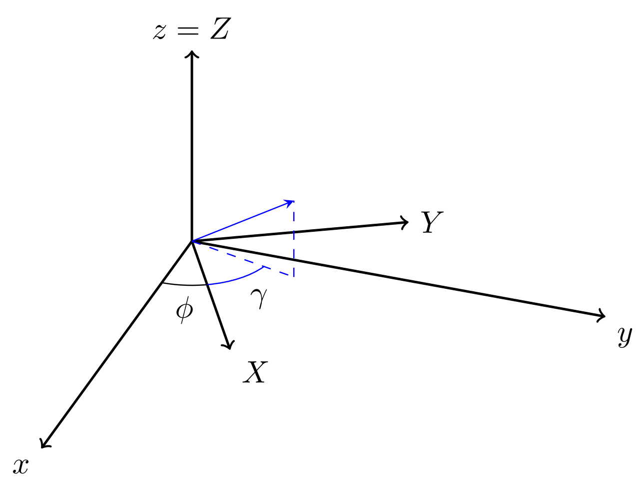

FIG. 1: The translation from the non-rotated xyz coordinates to the rotated XYZ system.

Therefore, our χ^{IJK} components in terms of the original ijk coordinate system are

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

for the χ^{XJK} components,

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

for the χ^{YJK} components, and lastly

$$\begin{aligned}
\chi^{ZXX} &= \sin^2\gamma\chi^{zxx} + \cos^2\gamma\chi^{zyy} - 2\sin\gamma\cos\gamma\chi^{zxy}, \\
\chi^{ZYY} &= \cos^2\gamma\chi^{zxx} + \sin^2\gamma\chi^{zyy} + 2\sin\gamma\cos\gamma\chi^{zxy}, \\
\chi^{ZZZ} &= \chi^{zzz}, \\
\chi^{ZYZ} = \chi^{ZZY} &= \sin\gamma\chi^{zyz} + \cos\gamma\chi^{zxz}, \\
\chi^{ZXZ} = \chi^{ZZX} &= -\cos\gamma\chi^{zyz} + \sin\gamma\chi^{zxz}, \\
\chi^{ZXY} = \chi^{ZYX} &= \sin\gamma\cos\gamma\chi^{zxx} - \sin\gamma\cos\gamma\chi^{zyy} - \cos 2\gamma\chi^{zxy},
\end{aligned}$$

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 $\gamma = \pi/2$ signifies that there is no rotation, and thus $\chi^{IJK} = \chi^{ijk}$.

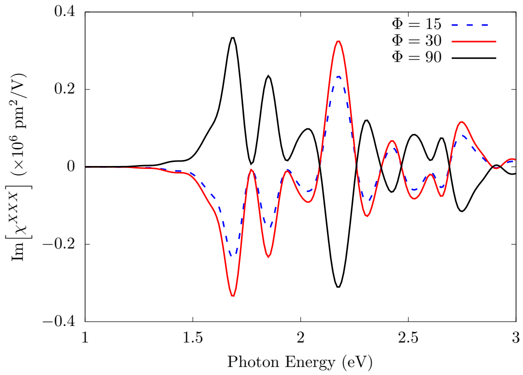

FIG. 2: χ^{XXX} for three values of γ calculated for a system with C_{3v} symmetry.

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,

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

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

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

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 γ for a system with C_{3v} symmetry in Fig. 2. The nonzero components in the original coordinates are presented in Fig. 3, multiplied by the appropriate prefactors from Eq. (1). We can see how we recover the component in the original coordinates when $\gamma = \pi/2$.

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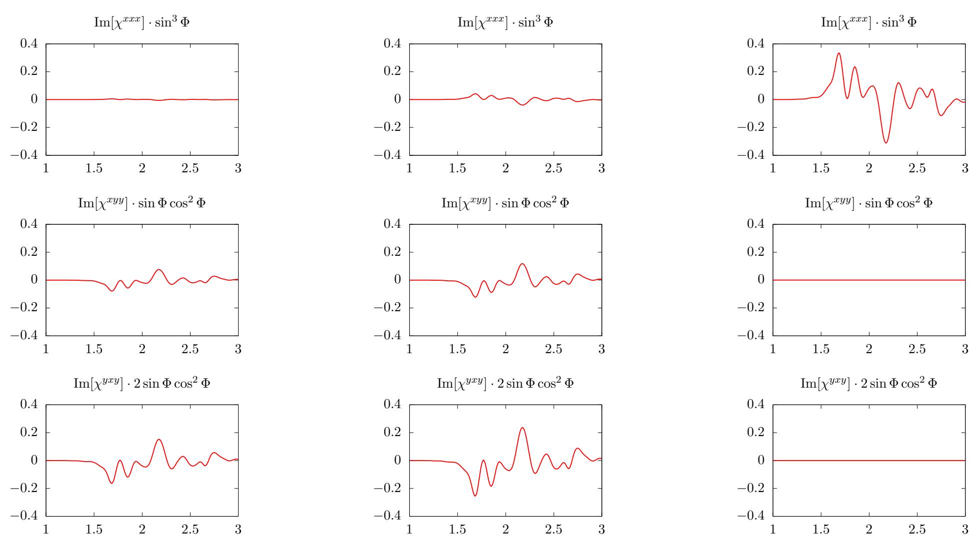

(a) $\gamma = \pi/12$. (b) $\gamma = \pi/6$. (c) $\gamma = \pi/2$.

FIG. 3: Nonzero components of χ^{ijk} multiplied by the appropriate prefactors, for three different values of γ.