PHYS2202 Nonlinear Optics
Problem Set 6
Due 17:00 on Thursday, May 14, 2020

1. **(20 points)**

   Context: Except for the final equation, this contextual information is not needed to solve the problem; it is just to explain why we would consider such a problem.

   Throughout the course, we have used the electric-dipole approximation. Namely, we have assumed that the interaction Hamiltonian between the optical fields and the medium is given by

$$\hat{H}_I = \hat{H}_{\mathrm{ED}} = -\sum_j q_j \hat{\vec{r}}_j \cdot \vec{E}(\vec{r}_j), \tag{1}$$

   where q_j is the charge of particle j located at position $\vec{r}_j$. This approximation is reasonable since the wavelengths of optical fields are long compared to the size of an atom, molecule, or crystal lattice period. We have seen, though, that for a crystal of given symmetry, there are certain combinations of input and output electric field polarizations for which the n^{th} order nonlinear response *in the electric-dipole approximation* (ED) is zero because $\hat{u} \cdot \overset{\leftrightarrow}{\chi}{}^{(n)}_{\mathrm{ED}} : \hat{a}\hat{b} \cdots \hat{n} = \chi^{(n)}_{\mathrm{ED},uab\ldots n} = 0$. For such combinations of polarizations, we might still see a non-zero n^{th} order nonlinear optical response (i.e., a response that depends on n interactions with the electric field), but this response will be due to typically weak but non-zero non-dipolar contributions.

   The leading order non-dipolar contributions to the optical response of a material are typically the electric quadrupole response and the magnetic dipole response. Let us consider the electric quadrupole response. Our elementary model for an electric quadrupole is two oppositely aligned electric dipoles of equal magnitude next to one another. To *induce* an electric quadrupole, the electric field cannot be constant across the material; the electric field must be spatially varying on a short enough timescale to push like charges in opposite directions. We can write the quadrupolar interaction Hamiltonian as

$$\hat{H}_{\mathrm{EQ}} = -\frac{1}{6}\sum_{i,j} Q_{ij} \frac{\partial}{\partial x_i} \vec{E}_j(\vec{r}), \tag{2}$$

   where the "molecular" (here we use the term in an extended sense to mean a microscopic object: atom, molecule, or unit cell) quadrupole operator $\overset{\leftrightarrow}{Q}$ for a collection of point charges is

$$\overset{\leftrightarrow}{Q}_{ij} = \frac{1}{6}\sum_n \left(3x_{n,i}x_{n,j} - r^2\delta_{ij}\right) q_n \tag{3}$$

Just as we saw that we can express the dipolar response in terms of a *local* susceptibility $\overset{\leftrightarrow}{\chi}^{(n)}$, the *nonlocal* electric quadrupole response (nonlocal because the response depends on the fact that the electric field has a spatial variation; there is a difference in electric field between two locations) can be expressed in terms of an n^{th} order nonlocal susceptibility $\overset{\leftrightarrow}{\chi}^{(n)}_{\text{EQ}}$. For example, the second-order electric quadrupole response can be written

$$P_u^{(2)}(\omega) = \sum_{a,b,c} \chi^{(2)}_{\text{EQ},uabc}(-\omega; \omega_1, \omega_2) E_a(\omega_1) \frac{d}{dx_b} E_c(\omega_2). \tag{4}$$

Note that the n^{th} order electric quadrupole contribution to the susceptibility is characterized by four indices instead of the three indices characterizing the dipolar $\overset{\leftrightarrow}{\chi}^{(2)}$, so although they are both labeled here by a (2) superscript they tensors are not the same rank.

Question: Consider the 3m crystal class whose stereogram is illustrated below. The filled circles represent atoms above the plane of the page. The only symmetry operations of this system are a three fold rotation about the z axis (out of the page), i.e., rotations of $2\pi/3$ and $4\pi/3$, and a mirror plane perpendicular to the x axis (in the yz plane). Note that the three-fold symmetry means that there must then be three mirror reflection planes. Find all combinations of input and output polarizations that could yield a non-zero polarization; that is, find all the non-zero quadrupole susceptibility elements $\chi^{(2)}_{\text{EQ},ijkl}$. Identify the dependences between elements that are not independent.

Note: You must demonstrate which elements are potentially non-zero and their dependencies. You cannot just give the result.