PHYS2202 Nonlinear Optics
Problem Set 2
Due at the beginning of class on Tuesday, March 17, 2020

1. (20 points) **Local field factors**

We noted in lecture that for the general case of non-spherical symmetry, the local electric field, $\vec{E}^{(\text{loc})}$, can be written in terms of the macroscopic electric field, $\vec{E}$ and the polarization, $\vec{P}$ as

$$\vec{E}^{(\text{loc})} = \vec{E} + \frac{1}{\epsilon_0}\overleftrightarrow{L} \cdot \vec{P}.$$

(*Note that the ppt handout has an error; $1/\epsilon_0$ is replaced in the notes by 4π, a relic from writing the equation in my notes in gaussian units rather than SI units.) We determined the value of $\overleftrightarrow{L}$ by a *mean field* approximation in which we represent the medium, which in reality consists of discrete atomic or molecular units, by a continuous medium. If we assumed that the system has spherical symmetry (or more generally is isotropic), we were able to calculate that the tensor $\overleftrightarrow{L}$ reduces to $\overleftrightarrow{L} = L\mathbf{1} = \frac{1}{3}\mathbf{1}$ ($\mathbf{1}$ is the identity matrix). The local field factor is then the same in all directions and given by

$$f(\omega) = 1 + (\epsilon_r - 1) L,$$

where ϵ_r is the relative permittivity.

Given the dramatic implications of the local field factor in nonlinear optics, it is important to consider systems that are not isotropic. Indeed, in the case of anisotropic systems, the $\overleftrightarrow{L}$ takes different forms. (Fortunately, in the case of a linear relationship between the polarization and the local electric field, $\overleftrightarrow{L}$ can always be written in a diagonal form.) In general, for molecules with elongated or oblate shape, one can use ellipsoidal cavities with semiaxes a_1, a_2, and a_3, and one finds that

$$L_i = \frac{a_1 a_2 a_3}{2} \int_0^\infty \frac{1}{\left(s + a_i^2\right)\sqrt{\left(s + a_1^2\right)\left(s + a_2^2\right)\left(s + a_3^2\right)}} ds,$$

which can be evaluated numerically.

Here we will just investigate molecular crystals of closely packed molecules of simple symmetry.

(a) Suppose that the we are dealing with molecules that are rod-like (having a cylindrical form with radius r much smaller than the length l).

i. Calculate the elements of the matrix $\overleftrightarrow{L}$.

ii. To realize the largest local field factor, in what direction should the polarization of the electric field be?

iii. What are the values of the largest and smallest local field factor if $\epsilon_r = 4$.

(b) Answer the preceding questions for molecules that have a disk-like form (having a cylindrical form but with $r >> l$).

(c) Everything else being equal (for example, molecules that pack with the same number density), for what shape molecule (spherical, rod-like, or disk-like) do we expect to yield a molecular crystal with the largest local field factor?