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Chapter 3

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Supplement: Frequency Dependence And Symmetry Properties Of The Nonlinear Susceptibility

In Section 2.2 of "Compact Blue–Green Lasers," we have described the second-order nonlinear susceptibility as a frequency domain quantity, and have also suggested that it may possess certain symmetry properties (e.g., $\chi^{(2)}(-2\omega_1) = \left[\chi^{(2)}(2\omega_1)\right]^*$). In this supplementary note, we examine the nature of $\chi^{(2)}$ in more detail. The frequency dependence of $\chi^{(2)}$ can be more complicated that we have implied thus far. The value of $\chi^{(2)}$ may depend on all the frequencies involved in the interaction; that is, on both the *applied* and *generated* frequencies; thus, in describing the sum-frequency component on page 40 of the text, we should really have written $\widetilde{P}_{NL}^{(\omega_1+\omega_2)} = 2\epsilon_o\chi^{(2)}(\omega_1+\omega_2;\omega_1,\omega_2)\widetilde{E}_1^{(\omega_1)}\widetilde{E}_2^{(\omega_2)}$. We have already suggested, in our second harmonic generation example, that $\chi^{(2)}$ might depend on the *generated* frequency. Compare second-harmonic generation with optical rectification using the simple "electron on a spring" model described earlier. In both cases, the same electric fields are applied; however, in the former case the response of the electron is at an optical frequency ($\omega = 2\omega_1$), whereas in the latter case, the response of the electron is at $\omega = 0$. It does not seem unreasonable to anticipate that the electron might oscillate differently at 0 Hz than at 10^{15} Hz. Thus, we might expect that the value of $\chi^{(2)}$ depends on the frequency we are trying to generate.

Similarly, the value of $\chi^{(2)}$ might depend on the *applied* frequencies, even if the generated frequency is the same. For example, suppose that we compare generation of 490-nm light by frequency doubling of a 980-nm laser with generation of the same wavelength by frequency mixing of two lasers: one at 1300 nm and the other at 786 nm. Even though the generated wavelength is the same in both cases, we might expect that the response of the electron is different when excited at a wavelength of 980 nm than when simultaneously excited at both 1300 and 786 nm.

In the text, we mentioned, but did not elaborate, on the fact that the nonlinear polarization and the applied electric fields are vectors, and are represented by three scalar components in a $x - y - z$ cartesian coordinate system. On page 40 of the text, we wrote that the sum-frequency component of induced polarization could be written as $\widetilde{P}_{NL}^{(\omega_1+\omega_2)} = 2\epsilon_o\chi^{(2)}(\omega_1+\omega_2)\widetilde{E}_1^{(\omega_1)}\widetilde{E}_2^{(\omega_2)}$, and mentioned that $\widetilde{P}_{NL}^{(\omega_1+\omega_2)}$, $\widetilde{E}_1^{(\omega_1)}$ and $\widetilde{E}_2^{(\omega_2)}$ are all vectors, each potentially having three vector components. A particular vector component of $\widetilde{P}_{NL}^{(\omega_1+\omega_2)}$ —the x-component $\widetilde{P}_{NL,x}^{(\omega_1+\omega_2)}$, for example—can receive contributions from nine separate terms: $\widetilde{E}_x^{(\omega_1)}\widetilde{E}_x^{(\omega_2)}$, $\widetilde{E}_x^{(\omega_1)}\widetilde{E}_y^{(\omega_2)}$, $\widetilde{E}_x^{(\omega_1)}\widetilde{E}_z^{(\omega_2)}$; $\widetilde{E}_y^{(\omega_1)}\widetilde{E}_x^{(\omega_2)}$, $\widetilde{E}_y^{(\omega_1)}\widetilde{E}_y^{(\omega_2)}$, $\widetilde{E}_y^{(\omega_1)}\widetilde{E}_z^{(\omega_2)}$; $\widetilde{E}_z^{(\omega_1)}\widetilde{E}_x^{(\omega_2)}$, $\widetilde{E}_z^{(\omega_1)}\widetilde{E}_y^{(\omega_2)}$, $\widetilde{E}_z^{(\omega_1)}\widetilde{E}_z^{(\omega_2)}$, (where we have dropped the 1,2 subscript from $\widetilde{E}$, and distinguish the two separate applied signals through the ω_1, ω_2 notation). Each of these product terms contributes to $\widetilde{P}_x^{(\omega_1+\omega_2)}$ through a component of the $\chi^{(2)}$ tensor. Each of these components may be different, corresponding to the different polarizations involved. This polarization dependence should not be too surprising if we again consider the simple-minded "mass-on-a-spring" model. If we contemplate the extension of this analogy to three dimensions, we might imagine three springs governing the response of the electron cloud, each accounting for the restoring force which arises when the cloud is displaced along spatial *x, y*, and *z* axes. Each of these springs may have a different stiffness—thus, the response of the electron may be different when it is displaced in the x-direction compared to its response when displaced along the y- or z- axes. (This simple model does not account very well for "cross-coupling", in which, for example, displacement along the y-axis results in motion along the x -axis, but this behavior occurs—and is of great practical importance—in the polarization response of nonlinear crystals.) Hence, to account for the polarization dependence in the nonlinear interaction, we can write something like:

$$\widetilde{P}_i^{(\omega_1+\omega_2)} = 2\epsilon_o \sum_{j,k} \chi_{ijk}^{(2)} \widetilde{E}_j^{(\omega_1)} \widetilde{E}_k^{(\omega_2)} \tag{3.1}$$

where, in principle, 27 $\chi^{(2)}$ components are required to determine $\widetilde{P}^{(\omega_1+\omega_2)}$ from $\widetilde{E}^{(\omega_1)}$ and $\widetilde{E}^{(\omega_2)}$.

In actuality, a number of factors reduce the number of independent components of the $\chi^{(2)}$ tensor. We briefly review them here; a more extended discussion may be found in books by Boyd [**?**] or Shen [**?**].

1. Reality of signals. Since $P(z,t)$ is a real signal, we would expect its Fourier transform to have the property that $\mathcal{P}(z,\omega) = \mathcal{P}^*(z,-\omega)$. The real-valued electric field has the same property. Thus, if $\widetilde{P}_i^{(\omega_3)} = 2\epsilon_o \sum_{j,k} \chi_{ijk}^{(2)}(\omega_3;\omega_2,\omega_1)\widetilde{E}_j^{(\omega_1)}\widetilde{E}_k^{(\omega_2)}$, this must be equal to $\widetilde{P}_i^{*(-\omega_3)} = 2\epsilon_o \sum_{j,k} \chi_{ijk}^{(2)*}(-\omega_3;-\omega_2,-\omega_1)\widetilde{E}_j^{*(-\omega_1)}\widetilde{E}_k^{*(-\omega_2)}$, which requires that $\chi_{ijk}^{(2)}(\omega_3;\omega_2,\omega_1) = \chi_{ijk}^{(2)*}(-\omega_3;-\omega_2,-\omega_1)$. Furthermore, it can be shown that for lossless media with no externally applied magnetic fields, all components of $\chi^{(2)}_{ijk}$ are real-valued.

2. Intrinsic permutation symmetry. In Eq. 3.1, the ω_1 signal is associated with the j index, and the ω_2 signal is associated with the k index. The order in which j and k are written in describing $\chi^{(2)}_{ijk}$ is a matter of convention. They can be interchanged, as long as ω_1 continues to be associated with the j index, and the ω_2 signal continues to be associated with the k index. Thus, $\chi^{(2)}_{ijk}(\omega_1 + \omega_2; \omega_1, \omega_2) = \chi^{(2)}_{ikj}(\omega_1 + \omega_2; \omega_2, \omega_1)$.

3. Overall permutation symmetry. This symmetry arises from the argument that the nonlinear medium cannot "tell the difference" between *generated* and *generating* field—all it "knows" is that three waves with frequencies ω_1, ω_2, and ω_3 are present within it, but not which were applied and which were produced. Thus, the same nonlinear susceptibility that applies to sum-frequency generation of a signal at $\omega_3 = \omega_1 + \omega_2$ from applied fields at ω_1 and ω_2 applies to difference frequency generation of a signal at ω_1 from applied fields at ω_2 and ω_3, e.g., $\chi^{(2)}_{ijk}(\omega_3; \omega_1, \omega_2) = \chi^{(2)}_{jki}(\omega_1; \omega_2, \omega_3)$. Thus, *any* of the ijk indices can be interchanged, as long as the corresponding frequencies are also interchanged.

4. Kleinman symmetry. If all the frequencies involved in the interaction lie sufficiently far away from any resonance frequencies of the nonlinear material, the $\chi^{(2)}_{ijk}$ are not strongly dependent on frequency. Hence, as far as the medium is concerned, the frequencies ω_1, ω_2, and ω_3 are virtually indistinguishable. Thus, the indices can be interchanged *without* interchanging the corresponding frequencies, as would be otherwise be required by overall permutation symmetry.

5. Spatial symmetry. If the nonlinear medium is a crystal, its constituent atoms are arranged in an orderly lattice. The orderliness of this arrangement may lead to spatial symmetries, such that if the crystalline structure were rotated about an axis, inverted through a point, or reflected across a plane, an identical arrangement of atoms would be obtained. Since the interacting fields would encounter the same arrangement of atoms in both the original crystal and the transformed version obtained through such a symmetry operation, the form of the $\chi^{(2)}$ tensor must be the same in both cases. By applying the symmetry operations valid for the crystal to the $\chi^{(2)}$ tensor and requiring it to remain unchanged, certain elements can be shown to be zero or related to others.