

**1. Classical Model for Nonlinear Response:** The nonlinear optical response of a medium can be understood in terms of a simple classical model. Suppose a medium contains classical particle-like electrons that move in a 1-dimensional potential

$$V(x) = V_0 \left( \frac{1}{2} \frac{x^2}{a^2} + \frac{1}{3} \frac{x^3}{a^3} \right)$$

where  $V_0$  is a characteristic atomic energy scale and  $a$  is a characteristic atomic length scale. Assuming each electron has an electric dipole moment  $-ex$ , the macroscopic polarization of the medium will be given by  $P = -exN$  for electron density  $N$ .

(a) Write out the equation of motion for an electron of mass  $m$  in this potential which is also driven by an electric field  $E(t) = E_0 \exp(i\omega t)$ . What is the resonant frequency  $\omega_0$  in the limit of small excursion  $x$ ?

(b) Assuming a steady-state solution of the form

$$x(t) = x_1 e^{i\omega t} + x_2 e^{2i\omega t} + x_3 e^{3i\omega t} + \dots$$

solve for the amplitudes of the first three terms  $x_1$ ,  $x_2$ , and  $x_3$ .

(c) For most optical materials, the lowest resonant frequencies are in the ultraviolet, so we can take  $\omega \ll \omega_0$ . Evaluate the amplitudes in this limit, and find expressions for the linear susceptibility  $\chi$  and the second and third order nonlinear coefficients, defined by

$$P(t) = \epsilon_0 \chi E + 2dE^2 + 4\chi^{(3)} E^3$$

Note that for  $eE_0 \ll V_0/a$ , the terms in the expression for  $P$  decrease in magnitude as their order increases.

**2. Nearly Degenerate Three-Wave Mixing:** The fundamental equation for second-order nonlinear response is

$$P(t) = 2dE(t)^2$$

for real electric field  $E$  and polarization  $P$ . (Assume here that the fields can be treated as scalars.) In class, we showed that for a single applied field oscillating at frequency  $\omega$ , the complex amplitude of the polarization component at  $2\omega$  is

$$P(2\omega) = dE(\omega)^2,$$

but for a applied field containing components at distinct frequencies  $\omega_1$  and  $\omega_2$ , the polarization component at  $\omega_1 + \omega_2$  is

$$P(\omega_1 + \omega_2) = 2dE(\omega_1)E(\omega_2).$$

Now suppose that the frequencies  $\omega_1$  and  $\omega_2$  are nearly identical, with  $\omega_1 = \omega_2 + \epsilon$ . Then on time scales  $t \ll 1/\epsilon$ , the frequencies  $2\omega_1$ ,  $2\omega_2$ , and  $\omega_1 + \omega_2$  cannot be distinguished. Show that the net polarization at these frequencies satisfies  $P = dE^2$  for  $E = E(\omega_1) + E(\omega_2)$ , just as would be obtained if  $\omega_1 = \omega_2$ . This should illustrate the continuity between the degenerate and non-degenerate cases.

3. **SHG in Te:** Design a second-harmonic generation experiment in Te using an input at  $\lambda = 10.6 \mu\text{m}$ . Te is a uniaxial crystal with indices of refraction

$\lambda$	$n_o$	$n_e$
$5.3 \mu\text{m}$	4.856	6.307
$10.6 \mu\text{m}$	4.794	6.243

It has symmetry class 32, which gives three non-zero second-order coefficients,  $d_{11} = -d_{12} = -d_{26} = 5.7 \times 10^{-21} \text{ C/V}^2$ .

Find the phase-matching angle and decide on the proper beam polarization and crystal orientation for maximum power output at  $5.3 \mu\text{m}$ . Find the effective value of the nonlinear coupling parameter,  $d'$  (including angle effects).

4. **Properties of BBO:** Look up the properties of the nonlinear crystal  $\beta$ -BaB<sub>2</sub>O<sub>4</sub>, commonly called BBO. Find the range of wavelengths over which it is transparent, whether it is uniaxial or biaxial, and values for as many of its nonzero second-order coefficients as you can find (including crystal symmetry effects). Also find its various indices of refraction at 1064 nm and 532 nm. Cite the sources you use.

*822 students only:*

5. **Electro-optic and Nonlinear Optic Coefficients:** In Section 19.2-B, Saleh and Teich explain the relationship between the electro-optic and second-order nonlinear optical coefficients, for scalar (rather than vector) fields. Show that in the vector case, the relationship is

$$r_{ijk} = -\frac{4\epsilon_0 d_{ijk}}{\epsilon_{ii}\epsilon_{jj}}$$

where  $\epsilon_{ij}$  is the dielectric tensor. Problem 19.6-3 in the text has a hint you may use, but I'd like you to derive the relation given there.