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Author
Shen, Y.R.

Publication Date
1977-03-01

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To be published as a Chapter in
"Quantum Electronics", C. L. Tang,
ed., Academic Press Series of
"Methods of Experimental Physics"

LBL-6091
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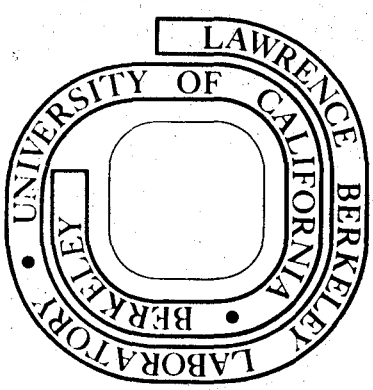
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An Article in the Book on "Quantum Electronics" in the Academic Press Series of "Methods of Experimental Physics"

LBL-6091

NONLINEAR OPTICAL EFFECTS

Y. R. Shen

Physics Department, University of California
Berkeley, California 94720

MARCH 1977

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I. Introduction

Nonlinear optics has long been one of the most important subfields in quantum electronics. Immediately after the laser was invented, it was recognized that the high field intensity of a laser beam could induce nonlinear responses in a medium. Franken et al.[1] first demonstrated in 1961 optical second-harmonic generation in a nonlinear crystal. Since then, many other nonlinear optical effects such as optical mixing, parametric oscillation, stimulated light scattering, self-focusing, multiphoton transitions, coherent transient phenomena, etc. have been observed.[2] Although most of the nonlinear optical effects were discovered in the earlier years, the rapid progress in the field has not been interrupted since 1961. First, it is on the physical understanding of the various nonlinear optical effects, and then, it is on the applications of these effects to various disciplines. More recently, the advent of tunable lasers, high-power lasers, and ultrashort laser pulses have brought new excitement to the field of nonlinear optics. As judged from the large number of papers on the subject in the recent literature, the field has apparently not yet lost its vigor in making advances.

It is clearly impossible for us to have any sort of comprehensible review of nonlinear optics in a short chapter. We should therefore be satisfied here with only a rather brief and qualitative description of the subject. Our emphasis is on physical concepts and potential applications. We will not venture into serious discussion on any topic, nor will be provide an extensive list of references at the end. The readers are referred to other review articles or books for more details.[2]

In the following sections, we shall first give a brief summary on

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the fundamentals of nonlinear optics, followed by a short discussion on nonlinear optical susceptibilities. We then describe as physically as possible the various nonlinear optical effects and their potential applications.

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II. Fundamentals of Nonlinear Optics

As is well known, all electromagnetic phenomena, linear or nonlinear, are governed by the Maxwell equations or the resulting wave equations

$$\left[\nabla \times (\nabla x) + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\vec{E}(\vec{r},t) = -\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\vec{P}(\vec{r},t)$$

$$\nabla \cdot \vec{E}(\vec{r},t) = -4\pi\,\nabla \cdot \vec{P}(\vec{r},t) \qquad (1)$$

where $\vec{P}(\vec{r},t)$ is a generalized electric polarization which includes all multiple contributions.[3,4] This polarization $\vec{P}$ as a function of $\vec{E}$ describes the response of the medium to the incoming field. In the weak field limit, $\vec{P}$ is linearly proportional to $\vec{E}$, but in general, $\vec{P}$ can be a complicated nonlinear function of $\vec{E}$. In some cases dealing with transients, $\vec{P}$ is an integral response function of $\vec{E}(\vec{r},t)$ and obeys a certain equation of motion. Knowing $\vec{P}$ as a function of $\vec{E}$, one can in principle solve Eq. (1) and hence fully describe the corresponding optical phenomenon. Unfortunately, this is usually not the case in practice. First, it is sometimes difficult to write down a simple correct expression for $\vec{P}(\vec{E})$. Second, it is often not possible to obtain a complete solution for Eq. (1) even with the largest computer available. One must resort to various approximations in order to circumvent the difficulties. This is actually the work of theoretical research on nonlinear optics.

In many cases of nonlinear optics, we can approximate the problem as one with several quasi-monochromatic light beams interacting in a nonlinear medium. It is then more convenient to decompose $\vec{E}$ and $\vec{P}$ into their Fourier components.

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$$\vec{E}(\vec{r},t) = \sum_i \vec{E}(\omega_i) = \sum_i \vec{\mathcal{E}}(\omega_i)\exp(i\vec{k}_i \cdot \vec{r} - i\omega_i t)$$

$$\vec{P}(\vec{r},t) = \sum_i \vec{P}(\omega_i) \qquad (2)$$

where $\vec{\mathcal{E}}(\omega_i)$ is a slowly varying spatial function. If the field is not excessively intense, then the polarization $\vec{P}(\omega_i)$ can be expanded into a power series

$$\vec{P}(\omega_i) = \overleftrightarrow{\chi}^{(1)}(\omega_i) \cdot \vec{E}(\omega_i)$$

$$+ \sum_{j,k} \overleftrightarrow{\chi}^{(2)}(\omega_i = \omega_j + \omega_k) : \vec{E}(\omega_j)\vec{E}(\omega_k) \qquad (3)$$

$$+ \sum_{j,k,\ell} \overleftrightarrow{\chi}^{(3)}(\omega_i = \omega_j + \omega_k + \omega_\ell) : \vec{E}(\omega_j)\vec{E}(\omega_k)\vec{E}(\omega_\ell)$$

$$+ - - - .$$

In Eq. (3), $\overleftrightarrow{\chi}^{(n)}$ are the nth-order susceptibility tensors. They actually govern the nth-order nonlinear optical effects. In the electric-dipole approximation, $\overleftrightarrow{\chi}^{(n)}$ are independent of the wave vectors $\vec{k}$.

Written in terms of the Fourier components, Eq. (1) now splits into a set of nonlinearly coupled equations

$$\left[\nabla \times (\nabla x) - \frac{\omega_i^2}{c^2}\right]\vec{E}(\omega_i) = \frac{4\pi\omega_i^2}{c^2}\vec{P}(\omega_i)$$

$$\nabla \cdot \vec{E}(\omega_i) = -4\pi \nabla \cdot \vec{P}(\omega_i) \qquad (4)$$

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where $\vec{P}(\omega_i)$ is given in Eq. (3). It is then the solution of these coupled equations one must strive for. As an example, let us consider the problem of three-wave interaction with $\omega_1 + \omega_2 = \omega_3$. Assuming all fields transverse, $\vec{E}(\omega_i) \perp \vec{k}_i$, we obtain from Eq. (4)[4]

$$\left[\nabla^2 + \omega_1^2 \overleftrightarrow{\varepsilon}(\omega_1)/c^2\right] \vec{E}(\omega_1) = - (4\pi\omega_1^2/c^2) \overleftrightarrow{\chi}^{(2)}(\omega_1 = -\omega_2 + \omega_3) : \vec{E}^*(\omega_2)\vec{E}(\omega_3) \tag{5a}$$

$$\left[\nabla^2 + \omega_2^2 \overleftrightarrow{\varepsilon}(\omega_2)/c^2\right] \vec{E}(\omega_2) = - (4\pi\omega_2^2/c^2) \overleftrightarrow{\chi}^{(2)}(\omega_2 = \omega_3 - \omega_1) : \vec{E}(\omega_3)\vec{E}^*(\omega_1) \tag{5b}$$

$$\left[\nabla^2 + \omega_3^2 \overleftrightarrow{\varepsilon}(\omega_3)/c^2\right] \vec{E}(\omega_3) = - (4\pi\omega_3^2/c^2) \overleftrightarrow{\chi}^{(2)}(\omega_3 = \omega_1 + \omega_2) : \vec{E}(\omega_1)\vec{E}(\omega_2) \tag{5c}$$

where $\overleftrightarrow{\varepsilon}(\omega_1) = 1 + 4\pi \overleftrightarrow{\chi}^{(1)}(\omega_i)$ are the linear dielectric constants. The above equations show explicitly that the three fields are nonlinearly coupled through the $\overleftrightarrow{\chi}^{(2)}$ terms. With $\overleftrightarrow{\chi}^{(2)}$ given, Eq. (5) with appropriate boundary conditions can in principle be solved. The solution can be greatly simplified if depletion of the incoming pump fields is negligible as is fairly often the case.

Physically, nonlinear coupling leads to energy transfer between waves. Then, as one would expect, the maximum rate of energy transfer should occur when both energy and momentum matching conditions are satisfied. In the steady (or quasi-steady) state case, energy matching $\omega_1 + \omega_2 = \omega_3$ is automatically satisfied. The momentum matching condition suggests that $\Delta\vec{k} = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$ should vanish where $\Delta\vec{k}$ is often called the phase mismatch between the interacting waves. This can be seen formally from Eq. (5) if we transform for example Eq. (5c) into the form

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$$(\vec{k}_3 \cdot \nabla)\vec{\mathcal{E}}(\omega_3) = (i\, 2\pi\omega_3^2/c^2)\overleftrightarrow{\chi}^{(2)}(\omega_3 = \omega_1 + \omega_2) : \vec{\mathcal{E}}(\omega_1)\vec{\mathcal{E}}(\omega_2)\exp(i\Delta\vec{k} \cdot \vec{r}) \tag{6}$$

assuming $\mathcal{E}(\omega_3)$ is a slowly varying function of distance. As the above equation stands, we see clearly that the spatial growth of $\mathcal{E}(\omega_3)$ at the expense of $\mathcal{E}(\omega_1)$ and $\mathcal{E}(\omega_2)$ should be a maximum when $\Delta\vec{k} = 0$. This phase (or momentum) matching condition can be somewhat relaxed in an absorbing medium since $\vec{k}_i$ will then have an imaginary part.

We will not proceed further on the discussion of nonlinear wave interaction here. We should however note that the coupled wave approach described above is extremely powerful in helping our understanding of a large number of nonlinear optical phenomena. This is even more so if we will generalize the waves to include also the excitational waves in the medium. In a later section, we will discuss several well-known nonlinear optical effects along this line.

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III. Nonlinear Susceptibilities

The role of nonlinear susceptibility tensors in nonlinear optics is the same as that of linear dielectric constants or refractive indices in linear optics. It is absolutely necessary to know $\overleftrightarrow{\chi}^{(n)}$ in order to prescribe the relevant nonlinear optical effects. For many purposes, one would like to be able to predict $\overleftrightarrow{\chi}^{(n)}$ theoretically even though $\overleftrightarrow{\chi}^{(n)}$ can also be measured experimentally.

How do we find $\overleftrightarrow{\chi}^{(n)}$ theoretically? As an nth-rank tensor, $\overleftrightarrow{\chi}^{(n)}$ has 3^n elements, but governed by the symmetry of the medium, not all of them are independent or nonvanishing. In evaluating $\overleftrightarrow{\chi}^{(n)}$, we should first find out from symmetry argument the nonvanishing and independent elements of $\overleftrightarrow{\chi}^{(n)}$, and then from a microscopic theory, the magnitudes of those nonvanishing elements.

For a given medium with a certain class of symmetry, there is a set of symmetry operations such as inversion, rotation, etc., under which the properties of the medium remain invariant. Let S be such an operation. Then under the transformation of S, the tensor $\overleftrightarrow{\chi}^{(n)}$ should remain unchanged. We have for example

$$(\overleftrightarrow{S}^{\dagger} \cdot \hat{i}) \cdot \overleftrightarrow{\chi}^{(2)} : (\overleftrightarrow{S} \cdot \hat{j})(\overleftrightarrow{S} \cdot \hat{k}) = \hat{i} \cdot \overleftrightarrow{\chi}^{(2)} : \hat{j}\,\hat{k}. \qquad (7)$$

There is one such equation for each symmetry operation. These equations make some elements of $\overleftrightarrow{\chi}^{(n)}$ vanish and some depend on others. As an example, Eq. (7) leads immediately to the familiar result that $\overleftrightarrow{\chi}^{(2)}$ of a medium with inversion symmetry has no nonvanishing element. The reduced forms of $\overleftrightarrow{\chi}^{(2)}$ and $\overleftrightarrow{\chi}^{(3)}$ for various classes of materials have been tabu-

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lated in the literature.[5]

The formal microscopic expression for $\overleftrightarrow{\chi}^{(n)}$ can always be obtained from the nth-order quantum mechanical perturbation calculation. Numerical evaluation of $\overleftrightarrow{\chi}^{(n)}$ from such an expression is however difficult since the transition frequencies and matrix elements of a given medium are usually not very well known. Thus, except in some simple cases such as alkali vapors, one would like to resort to simple theoretical models for evaluation of $\overleftrightarrow{\chi}^{(n)}$.

One such simplifying model which has been quite successful in calculating $\overleftrightarrow{\chi}^{(2)}$ and $\overleftrightarrow{\chi}^{(3)}$ for molecules and crystals is the so called bond model. This model was used in the early 1930's to calculate linear polarizabilities of molecules and linear dielectric constants of crystals.[6] According to the model, the induced polarization on a molecule or crystal is the vector sum of the induced polarizations on all the bonds between atoms in the molecule or crystal. This is known as the bond additivity rule.[6] We can write

$$\overleftrightarrow{\chi}^{(n)} = \sum_i \overleftrightarrow{\beta}_i^{(n)} \tag{8}$$

where $\overleftrightarrow{\beta}_i^{(n)}$ is the nth-order polarizability tensor of the ith bond and the summation is over all bonds in a unit volume. Thus, knowing the crystal structure, we need only to calculate $\overleftrightarrow{\beta}^{(n)}$.

Let us consider here only $\overleftrightarrow{\beta}^{(n)}$ due to electronic contribution. For optical frequencies far below the absorption bands, the calculation of $\overleftrightarrow{\beta}^{(n)}$ is made simple by the recent development of the bond theory.[7] Following the Penn approximation,[8] we can write

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$$4N\left(\beta_{\parallel}^{(1)} + 2\beta_{\perp}^{(1)}\right)/3 = \hbar^2\Omega_p^2/4\pi\ \bar{E}_g^2 \qquad (9)$$

where $\beta_{\parallel}^{(1)}$ and $\beta_{\perp}^{(1)}$ are the linear polarizabilities parallel and perpendicular to the cylindrically symmetric bond respectively, and Ω_p is the plasma frequency of the N valence electrons per unit volume. The average energy gap $\bar{E}_g$ between bonding and anti-bonding states is composed of a homopolar gap E_h and a heteropolar gap C.

$$\bar{E}_g^2 = E_h^2 + C^2. \qquad (10)$$

Approximate expressions of E_h and C in terms of the bond length, convalent radii and valences of the bond atoms etc. can be obtained from the bond theory.[7]

Knowing $\overleftrightarrow{\beta}^{(1)}$, we can then calculate $\overleftrightarrow{\beta}^{(n)}$ (and hence $\overleftrightarrow{\chi}^{(n)}$) from the derivative

$$\overleftrightarrow{\beta}^{(n)} = \partial^{(n-1)}\ \overleftrightarrow{\beta}^{(1)}/(\partial\vec{E})^{(n-1)}. \qquad (11)$$

As suggested by Eq. (9), the effect of the external field $\vec{E}$ on $\overleftrightarrow{\beta}^{(1)}$ is through $\bar{E}_g$. Two different models have been used to calculate the dependence of $\bar{E}_g$ on $\vec{E}$. One is the bond-charge model.[9] It assumes that the bond charges along the bond can be approximated by a point charge located at a distance equal to the covalent radii from the two bond atoms, and that the external field $\vec{E}$ induces a shift in the position of this point charge. Using this model, Levine has successfully calculated $\overleftrightarrow{\chi}^{(2)}$ and

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and $\overleftrightarrow{\chi}^{(3)}$ for a large number of liquids and crystals in good agreement with measured values. The other is the charge-transfer model.[10] It assumes that $\vec{E}$ induces a transfer of a definite amount of valence charges from one bond atom to the other. Tang and coworkers[10] have calculated $\overleftrightarrow{\chi}^{(2)}$ using the charge-transfer model for a number of semiconductors. Their results also agree well with the measured values. Both models are in fact only crude approximations of the real picture. In reality, there is a broad distribution of valence electrons along and around the bonds. The external field simply induces a slight redistribution of the valence electron distribution.

The bond theoretical study of nonlinear susceptibilities has led to the following conclusion. For highly nonlinear optical materials, the bond polarizabilities must have large nonlinearity. For materials with large $\overleftrightarrow{\chi}^{(2)}$, the crystal structure should also be as asymmetric as possible so that in the summation of Eq. (8), the vectorial cancellation of $\overleftrightarrow{\beta}^{(2)}$ from different bonds is as small as possible.

The approximations used in the bond theory discussed above does not hold when the optical frequencies are close to or in the absorption bonds. We must then use the full microscopic expression to calculate $\overleftrightarrow{\chi}^{(n)}$. Recent empirical calculations[11] suggest that in order to obtain the correct dispersion of $\overleftrightarrow{\chi}^{(2)}$, one cannot make any drastic approximation on the matrix elements and density of states. As in the linear case, the effect of critical points dominate the dispersion of $\overleftrightarrow{\chi}^{(2)}$. In principle, the measurements of the dispersion of $\overleftrightarrow{\chi}^{(2)}$ should compliment the linear optical measurements of $\overleftrightarrow{\chi}^{(1)}(\omega)$ or $\varepsilon(\omega)$ in the study of electronic structures of solids. Unfortunately, limited by the tunability of lasers and by the

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usually poor signal-to-noise ratio of nonlinear optical experiments, such measurements cannot yet be considered as a useful technique to study band structures of solids.

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IV. Nonlinear Optical Processes

In this section, we will describe briefly the various nonlinear optical processes. We can divide them into two groups: those in which a nonlinear susceptibility can be used to describe the nonlinear response of the medium laser and those cannot. The former includes harmonic generation, optical mixing, parametric amplification and oscillation, two-photon absorption, stimulated light scattering, optical-field-induced birefringence, and self-focusing. The latter include coherent transient effects, multiphoton ionization, laser breakdown, and multiphoton dissociation of molecules. Because of limitation of scope, we will not discuss here the highly nonlinear optical effects in plasmas, in the formation of laser -induced plasmas, and in laser-induced fusion.

A. Optical Mixing and Harmonic Generation

Among all nonlinear optical effects, optical mixing is probably the most important and well understood. Its basic physics is simple. Incoming pump beams interact nonlinearly in a nonlinear medium. They beat with one another and induce in the medium a nonlinear polarization oscillating at the combination (sum and difference) frequencies. This nonlinear polarization then acts as a source in generating an output beam at the combination frequencies. The rate of energy transfer from the pump beams to the output beam is maximum when phase matching between input and output waves is realized. Optical harmonic generation is a special case of optical mixing where an incoming pump wave beats with itself and generates a harmonic wave.

The theoretical description of optical mixing follows Eq. (4). We

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discuss here sum-frequency generation by optical mixing as an example. Consider only the equation for the sum-frequency field $\vec{E}(\omega_3)$. Assuming $\vec{E}(\omega_3) \perp \vec{k}_3$, we have

$$\left[\nabla^2 + \omega_3^2 \overleftrightarrow{\varepsilon}(\omega_3)/c^2\right] \vec{E}(\omega_3) = -\left(4\pi\omega_3^2/c^2\right)\vec{P}^{NL}(\omega_3) \tag{12}$$

which is the same as Eq. (5c) with the nonlinear polarization given by $\vec{P}^{NL}(\omega_3) = \overleftrightarrow{\chi}^{(2)}(\omega_3 = \omega_1 + \omega_2) : \vec{E}(\omega_1)E(\omega_2)$. Noting that we can write $\vec{P}^{NL}(\omega_3) = \vec{\mathcal{P}}^{NL}(\omega_3)\exp(i\vec{k}_s \cdot \vec{r} - i\omega_3 t)$, we can transform the corresponding Eq. (6) into

$$(\vec{k}_3 \cdot \nabla)\vec{\mathcal{E}}(\omega_3) = \left(i2\pi\omega_3^2/c^2\right)\vec{\mathcal{P}}^{NL}(\omega_3)\exp(i\Delta\vec{k} \cdot \vec{r}) \tag{13}$$

with $\Delta\vec{k} = \vec{k}_s - \vec{k}_3$ being the phase mismatch. Let $\Delta\vec{k}$ be along $\hat{z}$. The solution of Eq. (13) can then be expressed into an integral from

$$|\mathcal{E}(\omega_3)|^2 = \left(2\pi\omega_3^2/c^2\vec{k}_3 \cdot \hat{z}\right)^2 \left|\int_0^{\ell} dz\left[\hat{\mathcal{E}}(\omega_3) \cdot \vec{\mathcal{P}}^{NL}(\omega_3)\right]\exp(i\Delta kz)\right|^2 \tag{14}$$

assuming $\mathcal{E}(\omega_3) = 0$ at $z = 0$. From Eq. (14), we see immediately that if depletion of the pump fields is negligible, then $P^{NL}(\omega_3)$ is independent of z and the sum-frequency output is given by[4]

$$|\mathcal{E}(\omega_3)|^2 = \left(2\pi\omega_3^2/c^2\vec{k}_3 \cdot \hat{z}\right)^2 |\hat{\mathcal{E}}(\omega_3) \cdot \vec{\mathcal{P}}^{NL}(\omega_3)|^2 [\sin^2(\Delta k\ell/2)/(\Delta k\ell/2)^2] \tag{15}$$

which is a maximum at $\Delta k = 0$. The maximum output is therefore proportional to the square of the nonlinear susceptibility, the product of the

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pump field intensities, and the square of the crystal length ℓ. Depletion of the pump fields is of course non-negligible when the output conversion efficiency is high. Then in order to solve the integral in Eq. (14), we must first find the pump field variation as a function of distance. This must be done by solving the set of coupled equations in Eq. (5). The theoretical description here can be easily extended to other cases of optical mixing when $\vec{P}^{NL}(\omega_3)$ is taken as the nonlinear polarization at the output frequency.

An obvious application of optical mixing is in constructing coherent sources at new frequencies. This is described in Chapter in detail. So far, optical mixing has already been used to generate coherent radiation throughout the spectral range from microwave or far infrared below 1 cm^{-1} [12] to extreme ultraviolet at 512 Å.[13] In selecting nonlinear materials for such applications, one must take several factors into consideration. 1) The material should have a large nonlinearity. 2) The material should be more or less transparent to all frequencies involved. 3) Phase matching should be achievable in the material. 4) The material should have a sufficiently large dimension. 5) The laser breakdown threshold should be sufficiently high.

For efficient optical mixing, we often need to focus the pump beams in order to increase the pump field intensities and to use phase-matchable collinear beam geometry in order to increase the active interaction length. Research in this respect has been concentrated in finding materials which can have non-critical collinear phase matching for focused beams or in devising methods to circumvent the difficulty such as using waveguide or periodic structure. The non-critical collinear phase matching can occur,

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for example, in gas media and in crystals along a crystal axis.

The requirement of large nonlinearity is, of course, crucial for most nonlinear optical applications. We have already seen in Sec. II how a molecule or crystal can have large nonlinearity at frequencies well in the transparent region. Physically, a nonlinear susceptibility can also become much larger as a result of resonance enhancement when one or several optical frequencies approach resonances. The resonance is particularly strong in atomic vapors where the spectral lines are narrow. In addition, unlike solids, an atomic vapor can have wide transparent regions between sharp spectral lines, a very high laser breakdown threshold, and a cell length longer than we need. Thus, with a resonant nonlinear susceptibility and somewhat higher pump laser intensities, we can find high-order optical mixing in atomic vapors almost as efficient as the non-resonant lower-order optical mixing in solids.[14,15] Third-order optical mixing in atomic vapors has already been used to generate tunable radiation both in the infrared and in the ultraviolet.[14] In fact, because of the transparency requirement, atomic vapors are the only media one can use to generate extreme ultraviolet radiation by optical mixing.

The resonance enhancement of nonlinear susceptibilities can also be used as a spectroscopic means to study excitations in a medium.[16,17] This is especially useful for one-photon-forbidden but two-photon-allowed transitions. Such transitions can be easily detected from resonance enhancement in optical mixing when the sum or difference of two pump frequencies approaches these transitions. As an example, consider the use of four-wave mixing to study the Raman-allowed vibrations. The three pump fields are at ω_1, ω_2, and ω_3, and the output is at $\omega_1 - \omega_2 + \omega_3$. When $\omega_1 - \omega_2$

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approaches a particular vibrational frequency, the output gets strongly resonance-enhanced. Since the output is coherent, it can be easily discriminated from incoherent scattering or flourescence background. This method therefore has great advantage over spontaneous Raman scattering technique in cases where sample fluorescence is a nuisance. With ultrashort pump pulses, the technique can also be used to measure the vibrational relaxation times. Usually, we can let $\omega_1 = \omega_3$. This four-wave mixing technique to probe low-frequency excitations is often called coherent anti-Stokes Raman spectroscopy (CARS) and is described in detail in Chapter . As is clear from our general discussion, the technique is certainly not limited to the probing of only low-frequency excitations.[17]

B. Parametric Amplification and Oscillation

In second-order sum-frequency mixing, two pump fields at ω_1 and ω_2 generate an output field at $\omega_3 = \omega_1 + \omega_2$. One would expect that the inverse process in which one pump field at ω_3 generates two output fields at ω_1 and ω_2 should also be possible. The latter is known as parametric generation.[18] The splitting of ω_3 into ω_1 and ω_2 is clearly not unique, but the output has a sharp maximum near phase matching. Thus, one can tune the frequencies of the output by tuning the phase matching condition, e.g., by varying the orientation or the temperature of the nonlinear crystal. The difference between parametric amplification and oscillation is that the latter occurs in a cavity so that feedback effectively increases the active length. However, if the pump field is sufficiently intense, then the single-path parametric amplification from noise can already yield intense output. This is sometimes known as parametric superfluor-

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escence and has been used to generate tunable ultrashort optical pulses.[19] The formal theoretical derivation of parametric amplification and oscillation follows the coupled equations in Eq. (5) with proper boundary conditions. The solution is particularly simple if depletion of the pump field $E(\omega_3)$ is negligible, since then the equations of $E(\omega_1)$ and $E(\omega_2)$ become linearly coupled.

Parametric oscillators are important optical devices for generating tunable coherent sources, in particular in the infrared range.[18] They are discussed in detail in Chapter . Presently, with different nonlinear crystals, they have already been used to cover a tuning range from 0.42 μm to 10.5 μm.

C. Two-Photon Absorption

Two photon processes were well-known even long before the laser was invented,[20] but they are more readily detectable with laser beams. The rate of two-photon absorption in a medium is given by

$$\partial|E(\omega_1)|^2/dz = -\left(4\pi\omega_1^2/c^2k_1\right)\left[\text{Im } \chi^{(3)}(\omega_1 = \omega_1 + \omega_2 - \omega_2)\right]|E(\omega_1)|^2|E(\omega_2)|^2 \quad (16)$$

and a similar equation for $E(\omega_2)$. Here, the imaginary part of the third-order nonlinear susceptibility, Im $\chi^{(3)}$, is directly proportional to the two-photon absorption cross-section derived by the golden rule.[21] It is seen that the absorption rate is proportional to the product of the two incoming beam intensities. Therefore with sufficiently intense laser beams, two-photon absorption can be quite appreciable.

Two-photon absorption is useful as a spectroscopic technique compli-

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menting the single-photon absorption spectroscopy because of the different selection rules.[21] Recently, two-photon absorption with oppositely propagating beams has also been developed into a high-resolution spectroscopic technique for gases to beat Doppler broadening. This is discussed in detail in Chapter .

D. Stimulated Light-Scattering

Light scattering is also a two-photon process in which one incoming photon at ω_1 is absorbed, one photon at ω_2 is emitted, and the material system makes a transition from the initial state $|i\rangle$ to the final state $|f\rangle$ with a transition frequency $\omega_{fi} \cong \omega_1 - \omega_2$ (See Fig. 1). The final state can correspond to any material excitation, namely, electronic excitation, vibration, rotation, magnon, plasmon, etc. Spontaneous light scattering is of course well known to most scientists. If, however, the incoming pump field at ω_1 is very intense, then, the scattering probability can become so high that stimulated emission by the scattered photons dominates and leads to an avalanche build-up of the emitted radiation at ω_2. This is known as stimulated scattering. For each type of spontaneous scattering, there should be a corresponding stimulated scattering process.[22] Formally, Eq. (16) with ω_1 and ω_2 interchanged can be used to describe the growth of the Stokes field at ω_2 in the stimulated scattering. The quantity Im $\chi^{(3)}(\omega_2)$ in this case is negative and is directly proportional to the spontaneous scattering cross section.

The following stimulated scattering processes have been observed: phonon-Raman, electronic-Raman, spin-flip-Raman, polariton, Brillouin, Rayleigh, Rayleigh-wing, and concentration scattering. Among these, the

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Raman and polariton scattering have found useful applications in generation of tunable radiation, particularly in the infrared. Stimulated Raman scattering in atomic and molecular vapors pumped by a dye laser can generate tunable radiation in the range between 670 cm^{-1} and ~ 10000 cm^{-1} with maximum peak power as high as 60 MW.[14] Stimulated spin-flip Raman scattering pumped by a molecular laser can tune over the ranges 590 - 1100 cm^{-1} and 1540 - 2000 cm^{-1}. Finally, stimulated polariton scattering pumped by a ruby laser can generate tunable far-infrared radiation between 14 and 200 cm^{-1}.[12]

E. Optical-Field-Induced Birefringence

A polarized laser beam of high intensity can induce a birefringence in a medium. Formally, this can be seen from the expression for the induced polarization $\vec{P}(\omega)$ at the frequency ω of a weak probing beam. Let the intense beam be at ω'. Then following Eq. (3), we can write

$$\begin{aligned}\vec{P}(\omega) &= \left[\overleftrightarrow{\chi}^{(1)}(\omega) + \overleftrightarrow{\chi}^{(3)}(\omega = \omega' - \omega' + \omega) : \vec{E}(\omega')\vec{E}^{*}(\omega')\right]\vec{E}(\omega) \\ &\equiv \left(\overleftrightarrow{\chi}^{(1)} + \Delta\overleftrightarrow{\chi}\right) \cdot \vec{E}(\omega)\end{aligned} \tag{17}$$

assuming $\overleftrightarrow{\chi}^{(2)} = 0$ and higher order terms negligible. The induced susceptibility $\Delta\overleftrightarrow{\chi}$ proportional to $|E(\omega')|^2$ has in general a different tensorial symmetry than $\overleftrightarrow{\chi}^{(1)}$. In particular, $\Delta\overleftrightarrow{\chi}$ is usually anisotropic even if the medium is originally isotropic.

Physically, the induced $\Delta\chi$ can arise from various different mechanisms. The intense optical field can perturb the electron distribution

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around atoms or molecules, modifies the density of the medium through electrostriction, induces molecular reorientation, redistribution, libration, and population change among energy states, etc. All these mechanisms can effectively cause a change in the linear response of the medium. The optical-field-induced $\Delta\overleftrightarrow{\chi}$ gets resonantly enhanced when $\omega \pm \omega'$ approaches a two-photon allowed transition. This fact has recently been used as a high-resolution nonlinear optical spectroscopic technique to study Raman transitions in liquids[23] and optical transitions in gases.[24] The induced birefringence at resonance in an atomic vapor is so large that it may find application in the construction of a fast optical switching device.[25]

F. Self-Focusing

The optical-field-induced $\Delta\overleftrightarrow{\chi}$, or equivalently the optical-field-induced refractive index $\Delta\overleftrightarrow{n}$, changes the propagation velocity of a laser beam in a medium. In most media, Δn is positive. Then, if the beam has a Gaussian-shaped profile, its central portion is more intense and should propagate more slowly than the edge portion. Consequently, as described in Fig. 2a, the wavefront of the beam gets distorted, and since ray propagation is always perpendicular to the wavefront, the beam tends to focus by itself. This of course happens only when the self-focusing action is strong enough to overcome the usual diffraction action.[26]

Relatively high intensity laser pulses ($\gtrsim$ 10 MW/cm^2) are often necessary for self-focusing experiments. The self-focusing dynamics varies according to the ratio of the laser pulse width T_p to the response time τ of Δn. If the ratio is much larger than 1, we have the quasi-steady-state case. The focusing depends only on the instantaneous incoming laser power

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and both focusing and subsequent diffraction are rather abrupt, leading to a sharp focal spot which moves along the axis as the input laser power varies with time. On a time-integrated photograph, it results in an intense filament of the order of 10 μm in diameter. This is the so-called moving focus model. If the ratio T_p/τ is close to or smaller than 1, we have the transient case. The field-induced Δn is now a transient cumulative response to the laser intensity variation over a period of $\sim \tau$ in the past. Focusing and subsequent diffraction become less abrupt for the more transient case. In the extreme transient limit, the front part of the laser pulse hardly focuses while the lagging part focuses more tightly, such that the longitudinal pulse profile first gets deformed into a horn shape and then propagates on without much further change (see Fig. 2b). As the horn-shaped pulse travels in the medium, its neck part again forms a track of intense filament on the integrated photograph. This is the so-called dynamic trapping model.

Because of the extremely high intesity ($\gtrsim 10$ GW/cm^2) in the focal region, self-focusing can readily initiate in the medium other nonlinear optical processes such as stimulated Raman and Brillouin scattering, multiphoton absorption, and optical breakdown. Optical breakdown is the most detrimental effect one must avoid in the design of high-power laser systems.

The field-induced Δn can sometimes be negative, leading to self-defocusing of light.[27] This often happens when the medium absorbs the laser beam and heating causes a decrease in n. Self-defocusing is extremely sensitive to the absorption coefficient. As a result, it has been used as a spectroscopic technique to study very weak absorption bands in

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materials.

G. Coherent Transient Effects

In magnetic resonance, it is well known that resonant coherent pulsed excitation leads to coherent mixing of initial and final states with a transverse relaxation time T_2. If the width of the exciting pulse or pulses is shorter than or comparable with T_2, then coherent transient effects result. There is a close analog between magnetic resonance and optical resonance. A system under resonant excitation can often be approximated by a two-level system. Then, as shown by Feynman et al.,[28] any two-level system can be treated as a spin - ½ system and hence its dynamic response to a rotating or cirularly polarized field can be described by the well-known Bloch equation. Physically, the Bloch equation is simply an equation of motion which describes the precession of a pseudodipole $\vec{p}$ around an effective field $\vec{E}$, subject to initial conditions and longitudinal and transverse relaxations. We define $\vec{p}$ and $\vec{E}$ as follows: $p_x\hat{x} + p_y\hat{y}$ is the real induced dipole; p_z is the product of the population difference and the transition matrix element γ between initial and final states; $E_x\hat{x} + E_y\hat{y}$ is the applied field; E_z is the quotient of the resonant energy and γ. Neglecting relaxation, the equation has the form

$$\partial\vec{p}/\partial t = -(\gamma/\hbar)\vec{E} \times \vec{p}. \qquad (18)$$

In magnetic resonance cases, $\vec{p}$ and $\vec{E}$ correspond to the induced magnetic dipole and the applied magnetic field respectively.

The Bloch equation coupled with the Maxwell equations essentially

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describe all the transient coherent phenomena in a two-level system.[29] There is however an important difference between optical and magnetic resonance cases. The former deal with propagating waves with a wavelength λ much smaller than the dimension d of the medium while the latter deal with standing waves with $\lambda \gtrsim d$. Thus, there are a number of observed coherent transient effects which are common for both optical and magnetic resonances. These include one and two-photon transient nutation and free-induction decay, photon or spin echoes, adiabatic following and inversion, and superradiance. There are also others which are unique for optical resonances. They include self-induced transparency, $n\pi$ pulse propagation, and some aspects of superradiance.

Applications of coherent transient effects are mainly in the measurements of relaxation times of excited states of a system. One hopes to learn how various interactions and external perturbations affect the relaxations. Recently, it has been proposed that adiabatic inversion[30] and π pulse propagation[31] can also be used for efficient isotope separation.

H. Multiphoton Ionization

Higher-order nonlinear optical effects can also occur if the laser field intensity in a medium is sufficiently high. Thus, in low-pressure gases, n-photon ionization processes with $n >> 1$ can readily be observed if the laser intensity is 10^{11} W/cm^2 or more. For example, Lecompte et al. have observed 11-photon ionization of Xe,[32] and Baravian et al. have observed 16-photon ionization of Ne.[33]

Theoretically, multiphoton ionization could presumably be described

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by a high-order perturbation calculation of transition probability. However, at the laser intensity necessary for multiphoton ionization, the optical-field-induced level shifts and level broadenings often become important. They can contribute significantly to the multiphoton ionization probability. This is particularly true when the laser combination frequencies are not far from intermediate resonances.[34] Being a very high-order nonlinear process, multiphoton ionization probability is very sensitive to the laser mode structure. For example, in the 11-photon ionization of Xe, it has been observed that a laser beam with 100 longitudinal modes yields an ionization probability $10^{6.9 \pm 0.3}$ times larger than a single-mode laser beam.[32]

Since ion detectors are extremely sensitive, two-photon ionization and two-photon transition with subsequent dc field-induced ionization have been used to study autoionization states[35] and high rydberg states of simple atoms.[36] Multiphoton ionization may also find potentially important applications in isotope separation[37] and in generating highly polarized electrons.[38]

I. Optical Breakdown

In the study of multiphoton ionization, one must keep the gas pressure low ($\lesssim 10^{-3}$ Torr), the laser pulse short ($\lesssim 10^{-8}$ sec.), and the focal dimension small ($\lesssim 10^{-2}$ cm.), so that electron-atom and atom-atom collision effects can be neglected. At higher pressures, free electrons released from atoms by multiphoton ionization can absorb photons by inverse bremsstrahlung during their collisions with atoms. An electron can then acquire enough energy capable of ionizing an atom to release another

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electron. This starts the electron multiplication process and leads to avalanche ionization. As soon as there is appreciable ionization, the incoming photons can be readily absorbed by electrons via free-free transitions in the field of ions. It causes intense heating of the electron plasma and consequently a rapid hydrodynamic expansion of the plasma in the form of a spherical shock wave with a visible spark. The phenomenon is known as optical breakdown. After the breakdown, the laser beam is preferentially absorbed by the part of the shock wave moving towards the laser, and hence the spark appears to propagate against the laser beam.[39]

The theory of optical avalanche ionization is similar to that of microwave avalanche ionization. However, in the optical case, the photon energy is not negligible compared to the electron energy. This is a quantum effect which must be taken into account in the correct treatment of optical avalanche ionization.[40] Interaction of the laser beam with the expanding plasma is another difficult theoretical problem. Besides direct optical absorption, other nonlinear optical processes can occur in the plasma. How efficiently a high-power laser beam can heat up a plasma in a very short period is presently of great importance for the understanding and realization of laser-induced fusion.

Optical breakdown can also occur in solids and is an extremely important problem for laser optical technology. In transparent solids, the physical mechanisms for optical breakdown are again multiphoton and avalanche ionization.[41] However, in solids with small energy gaps or appreciable impurity concentrations, the number of free electrons in the conduction bands resulting from thermal ionization can be fairly high. Then, multiphoton ionization is no longer needed to provide the first few elec-

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trons to start the avalanche ionization. Multiphoton ionization becomes important only when the energy gap is much larger than kT, but not much larger than the laser photon energy. The above description also applies to surface optical damage even though the surface contamination may significantly lower the breakdown threshold.

J. Multiphoton Dissociation of Molecules

Collisionless multiphoton dissociation of polyatomic molecules is presently the most interesting topic in nonlinear optics.[42] It deals with the problem of how a single molecule can absorb more than 30 or 40 infrared photons and get dissociated, and what the subsequent dynamics of dissociation is. The process is technically very important because of its potential application to isotope separation. It has been demonstrated by Ambartzumian et al. and others[42] that multiphoton molecular dissociation under quasi-monochromatic laser excitation is isotopically selective. In the case of a cell filled with 0.18-torr natural SF_6 and 2-torr H_2 as the scavanger, the observed enrichment factor of $^{34}S/^{32}S$ after irradiation of 2000 2-J CO_2 laser pulses was as high as 2800. Isotope separation of heavy elements such as ^{181}Os and ^{192}Os by multiphoton molecular dissociation has also been observed.[43]

Scientifically, multiphoton dissociation of molecules is also an extremely interesting process. In normal circumstances, even the occurance of a two- or three-photon absorption process would require a laser intensity of ~ 1 MW/cm^2. One would expect that for absorption of more than 30 or 40 photons by a single molecule, the laser intensity needed should be close to the optical breakdown threshold ≥ 10 GW/cm^2. In reality, the

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observed threshold for multiphoton dissociation of SF_6 was only ~ 10 MW/cm^2. The reason for this low dissociation threshold is now qualitatively understood[42] and is briefly sketched here.

For a single molecule to absorb several tens of infrared photons with high probability from a reasonably intense laser beam, two conditions must be satisfied. First, each single-photon step in the absorption must be at resonance or near resonance. Second, the density of states must increase rapidly with increase of energy so that even with a thermal distribution, up-pumping can dominate over down-transitions. For a polyatomic molecule, the second condition is usually satisfied because of the large number of vibrational degrees of freedom. That the first condition can also be satisfield is however not obvious. Consider the case of SF_6. Its ν_3 vibrational mode at 948 cm^{-1} can be excited by a CO_2 laser beam tuned to the ν_3 absorption band. Around 29 photons must be absorbed by a single SF_6 molecule in order to reach the dissociation level of $SF_6 \rightarrow SF_5 + F$ [44] at 27600 cm^{-1}. If we treat the problem as interaction of monochromatic laser radiation with a ν_3 anharmonic oscillator having an anharmonic shift of $\Delta\nu_{anh}$ = 2.88 cm^{-1}, then as shown in Fig. 3a, the laser frequency will quickly get out of steps with the ν_3 ladder in the up-excitation process. This picture is clearly not realistic since an SF_6 molecule should be far more complicated than an anharmonic oscillator. Associated with each vibrational state, there is a set of rotational levels. Each vibrational transition now has three branches P($\Delta J = -1$), Q($\Delta J = 0$), and R ($\Delta J = +1$) with frequencies given by

$$\nu(v,J \rightarrow v+1,J) \cong \nu_3 + v(\Delta\nu_{anh}) + 2\tilde{B}M$$

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where M = - J for $\Delta J = -1$, M = 0 for $\Delta J = 0$, and M = J + 1 for $\Delta J = 1$. The constant $\tilde{B}$ is 0.027 cm^{-1} for SF_6. With a proper J value, the anharmonic shift can be nearly compensated by the rotational shift. Thus, in the up-excitation, if it starts with a proper rotational state in the ground vibrational manifold ($J \sim 50$ in the case of SF_6), the first three or four single-photon steps can all be nearly resonant. The situation can be further improved when one considers in reality that the degeneracy of vibrational states is also split by Coriolis coupling and anharmonic coupling. Therefore, as shown in Fig. 3b, one can expect that at least the first 3 - 6 single-photon steps in the vibrational excitation are nearly resonant. Then, the density of states of a polyatomic molecule increases very rapidly and soon forms a quasi-continuum. In SF_6 for example, the density of states at 5000 cm^{-1} is already as high as $10^3/cm^{-1}$. Therefore, after the first 3 - 6 near-resonant steps, the radiation field can now resonantly excite the molecule all the way through the quasi-continuum to above the dissociation level. This then qualitatively explains the observed high probability of multiphoton dissociation. While isotopic selectivity comes in through the initial near-resonant steps over the discrete states, absorption in the quasi-continuum simply puts enough energy into the molecule for subsequent dissociation. Clearly, the above theory is not valid for diatomic molecules and perhaps also triatomic molecules. This is the reason why no multiphoton dissociation of such small molecules has ever been reported.

An experiment using a molecular beam to study collisionless multiphoton dissociation[45] has shown that each SF_6 absorbs around 35 photons from a CO_2 TEA laser pulse. The absorbed energy appears to be randomly

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distributed in all vibrational degrees of freedom before dissociation. The dissociation rate is finite and increases very rapidly with the excitation energy. It is around 10^7 sec^{-1} at the excitation level of 35 photons absorbed per molecule. The power or/and energy threshold and the power or/and energy dependence of multiphoton dissociation are not yet clearly understood. The quantitative aspect of multiphoton excitation and also other characteristic features of the dissociation process are yet to be investigated. A full understanding of the process will be essential for its future application to isotope separation or other induced chemical processes.

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V. Concluding Remarks

We have not discussed here a number of other important nonlinear optical effects. In particular, nonlinear optics in plasmas has developed into a major subfield in the study of laser interaction with plasmas and laser-induced fusion.[46] Nonlinear optics in optical waveguides is an important branch in the development of integrated optics. Applications of nonlinear optics to various disciplines have been growing rapidly in the recent years. We can anticipate that the field of nonlinear optics will certainly remain active for a number of years to come.

Acknowledgement

This work was done with support from the U. S. Energy Research and Development Administration.

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References

1. P. A. Franken, A. E. Hall, C. W. Peters, and G. Weinreich, Phys. Rev. Lett. 7, 118 (1961).

2. See, for example, the recent review article by Y. R. Shen, Rev. Mod. Phys. 48, 1 (1976) and references therein.

3. L. D. Landau and E. M. Lifshitz, "Electrodynamics in Continuous Media" (Pergamon Press, Oxford, 1960) p. 251.

4. N. Bloembergen, "Nonlinear Optics" (Benjamin Inc., New York, 1965).

5. P. N. Butcher, "Nonlinear Optical Phenomena" (Ohio State University Press, Columbus, Ohio, 1965) p. 43.

6. See, for example, K. G. Benbigh, Trans. Faraday Soc. 36, 936 (1940).

7. J. C. Phillips, "Covalent Bonding in Crystals, Molecules, and Polymers" (University of Chicago Press, Chicago, 1969); "Bonds and Bands in Semiconductors" (Academic Press, New York, 1973).

8. D. R. Penn, Phys, Rev. 128, 2093 (1962).

9. B. F. Levine, Phys. Rev. B7, 2600 (1973).

10. C. L. Tang and C. Flytzanis, Phys. Rev. B4, 2520 (1971); C. L. Tang, IEEE J. Quantum Electron. QE-9, 755 (1973); F. Scholl and C. L. Tang, Phys. Rev. B8, 4607 (1973).

11. C. Y. Fong and Y. R. Shen, Phys. Rev. B12, 2325 (1975).

12. Y. R. Shen, Progress in Quantum Electronics 4, 207 (1976) and references therein. See also the book on Nonlinear Infrared Generation, edited by Y. R. Shen (Springer-Verlag, Berlin, 1977).

13. J. Reintzes, R. C. Eckardt, C. Y. She, N. E. Karangelen, R. C. Elton, and R. A. Andrews, Phys. Rev. Lett. 37, 1540 (1976).

14. J. J. Wynne and P. P. Sorokin, in "Nonlinear Infrared Generation",

0 0 0 0 4 7 0 9 0 2 8

edited by Y. R. Shen (Springer-Verlag, Berlin, 1977).

15. R. B. Miles and S. E. Harris, IEEE J. Quantum Electron. QE-9, 470 (1973).

16. N. Bloembergen, in "Laser Spectroscopy", edited by S. Haroche et al. (Springer-Verlag, Berlin, 1975) p. 31.

17. Articles in "Course on Nonlinear Spectroscopy", edited by N. Bloembergen (North-Holland Publishing Co., Amsterdam, 1976).

18. See, for example, R. L. Byer in "Quantum Electronics", edited by H. Rabin and C. L. Tang (Academic Press, New York, 1975) p. 588.

19. A Laubereau, L. Greiter, and W. Kaiser, Appl. Phys. Lett. 25, 87 (1974); T. Kushida, Y. Tanaka, M. Ojima, and Y. Nakazaki, Japan. J. Appl. Phys. 14, 1097 (1975).

20. M. Goeppert-Mayer, Ann. Physik 9, 273 (1931).

21. J. M. Worlock, in "Laser Handbook", edited by F. T. Arecchi and E. O. Schulz-Dubois (North-Holland Publishing Co., Amsterdam, 1972) p. 1323; H. Mahr, in "Quantum Electronics", edited by H. Rabin and C. L. Tang (Academic Press, New York, 1975) p. 286.

22. Y. R. Shen, in "Light Scattering in Solids", edited by M. Cardona (Springer-Verlag, Berlin, 1975) p. 275.

23. D. Heiman, R. W. Hellwarth, M. D. Levenson, and G. Martin, Phys. Rev. Lett. 36, 189 (1976); J. J. Song, G. L. Eesley, and M. D. Levenson, Appl. Phys. Lett. 29, 567 (1976).

24. R. Teets, R. Feinberg, T. W. Hansch, and A. L. Schawlon, Phys. Rev. Lett. 37, 683 (1976).

25. P. F. Liao and G. C. Bjorklund, Phys. Rev. Lett. 36, 584 (1976).

26. Y. R. Shen, Progress in Quantum Electronics 4, 1 (1975).

0 0 0 0 4 7 0 9 0 2 9

27. F. W. Dabby, T. K. Gustafson, J. R. Whinnery, Y. Kohanzadeh, and P. L. Kelley, Appl. Phys. Lett. 16, 362 (1970).

28. R. P. Feynman, F. L. Vernon, and R. W. Hellwarth, J. Appl. Phys. 28, 49 (1957).

29. L. Allen and J. H. Eberly, "Optical Resonance and Two-Level Atoms" (J. Wiley, New York, 1975).

30. I. Nebenzahl and A. Szöke, Appl. Phys. Lett. 25, 327 (1974).

31. J. C. Diels, Phys. Rev. A13, 1520 (1976).

32. C. LeCompte, G. Mainfray, C. Manus, and F. Sanchez, Phys. Rev. A11, 1109 (1975).

33. G. Baravian, J. Godart, and G. Sultan, Phys. Rev. A14, 761 (1976).

34. See, for example, C. S. Chang and P. Stehle, Phys. Rev. Lett. 30, 1283 (1973); P. Agostini and C. LeCompte, Phys. Rev. Lett. 36, 1131 (1976).

35. P. Esherick, J. A. Armstrong, R. W. Dreyfus, and J. J. Wynne, Phys. Rev. Lett. 36, 1296 (1976).

36. M. G. Littman, M. L. Zimmerman, T. W. Ducas, R. G. Freeman, and D. Kleppner, Phys. Rev. Lett. 36, 788 (1976).

37. S. A. Tuccio, J. W. Dubrin, O. G. Peterson, and B. B. Snavely, VIII International Quantum Electronics Conference, (San Francisco, 1974), postdeadline paper Q.14.

38. P. Lambropoulos, Phys. Rev. Lett. 30, 413 (1973).

39. S. A. Ramsden and W. E. R. Davis, Phys. Rev. Lett. 13, 227 (1974).

40. N. Kroll and K. M. Watson, Phys. Rev. A5, 1883 (1972).

41. See, for example, N. Bloembergen, IEEE J. Quantum Electronics QE-10, 375 (1974).

0 0 0 0 4 7 0 9 0 3 0

42. See, for example, articles on the subject in "Tunable Lasers and Applications", edited by A. Mooradian, T. Jaeger and P. Stokseth (Springer-Verlag, Berlin, 1976), and references therein.

43. R. V. Ambartzumian, Yu A. Gorokhov, V. S. Letokhov, and G. N. Makarov, JETP Lett. 22, 43 (1975).

44. M. J. Coggiola, P. A. Schulz, Y. T. Lee, and Y. R. Shen, Phys. Rev. Lett. 38, 17 (1977).

45. E. R. Grant, M. J. Coggiola, P. A. Schulz, Y. T. Lee, and Y. R. Shen (to be published).

46. See, for example, "Laser Interaction and Related Plasma Phenomena", edited by H. J. Schwarz and H. Hora, vols. I, II, III, and IV (1969, 1971, 1973, 1975).

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Figure Captions

Fig. 1. Schematic drawing showing the Stokes ($\omega_1 > \omega_2$) and anti-Stokes ($\omega_1 < \omega_2$) Raman transition between states $|i\rangle$ and $|f\rangle$.

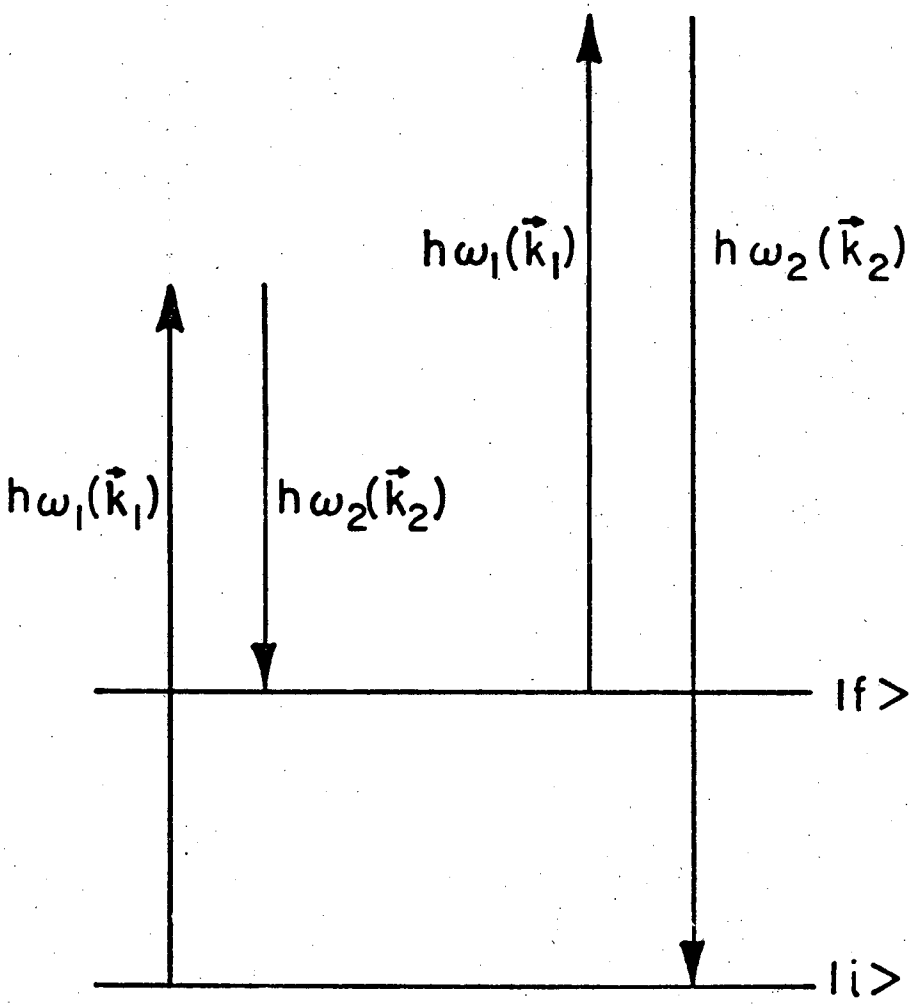
Fig. 2(a). Sketch showing the distortion of the wavefront and self-focusing of a laser beam in a nonlinear medium.

(b). Sketch showing transient self-focusing of a laser pulse in a Kerr liquid. Different parts (a, b, c, etc.) of the pulse focus and defocus along different ray paths. The pulse first gets deformed into a horn shape and then propagates on without much further change.

Fig. 3(a). Monochromatic laser radiation interaction with an anharmonic oscillator. The laser excitation quickly runs out of steps with the ladder of excited states.

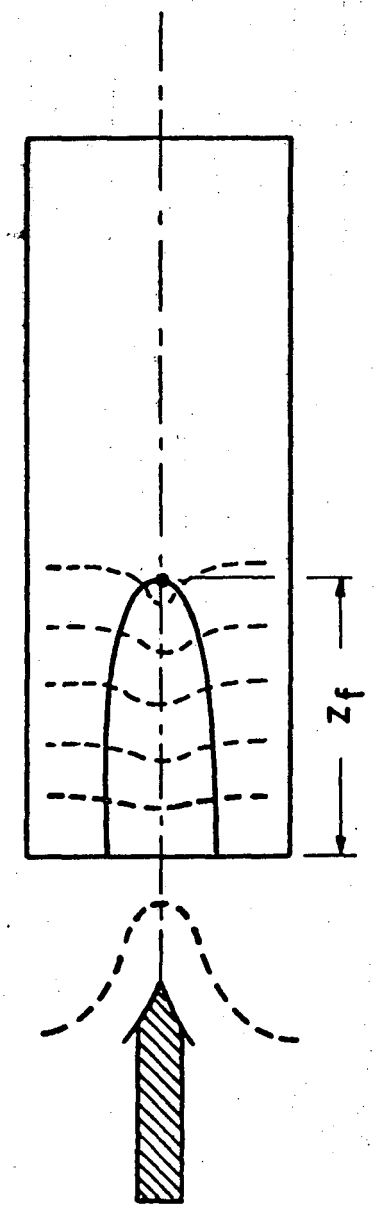
(b). Schematic diagram showing near-resonant transitions over the discrete rotation-vibrational states followed by successive resonant transitions in a quasi-continuum to beyond the dissociation level.

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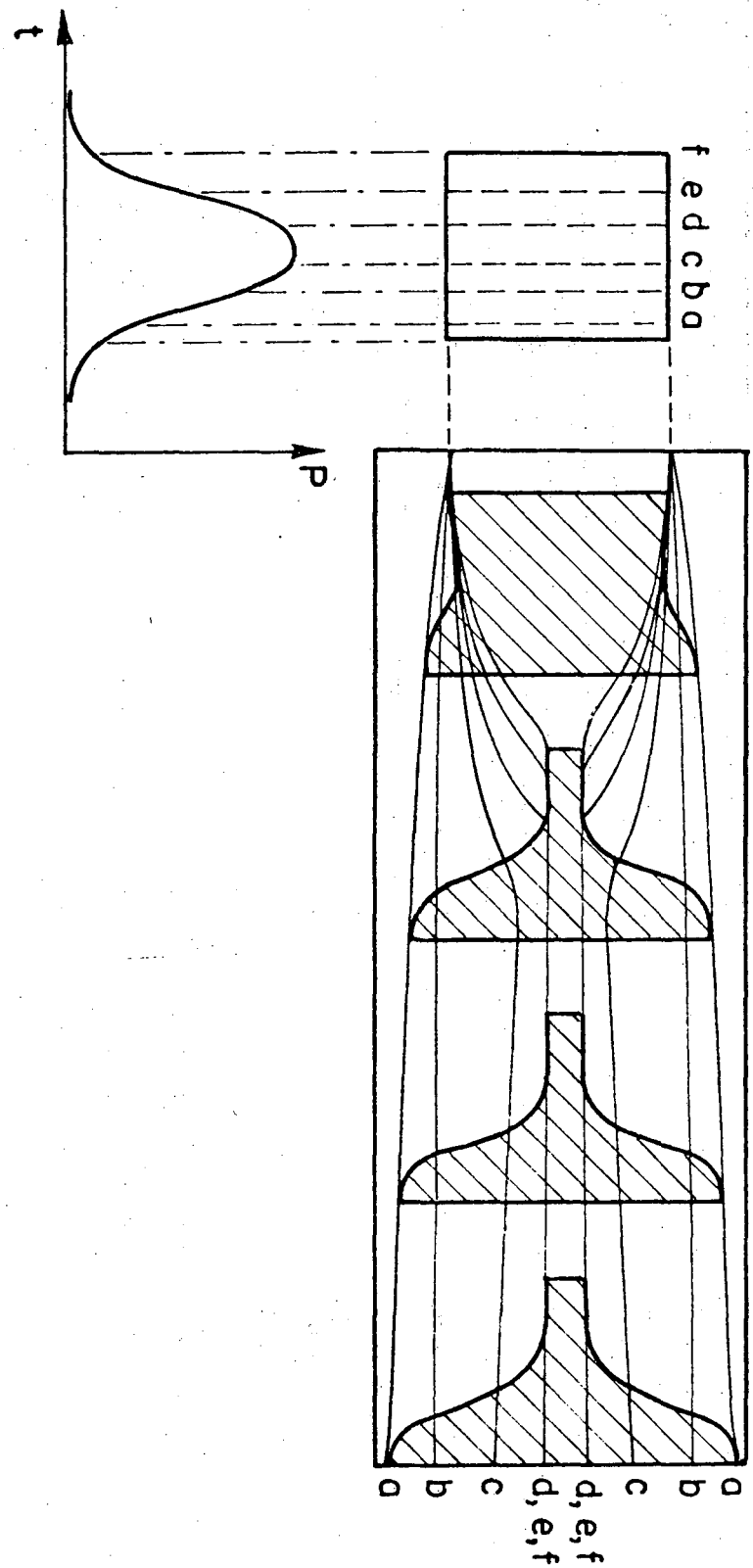
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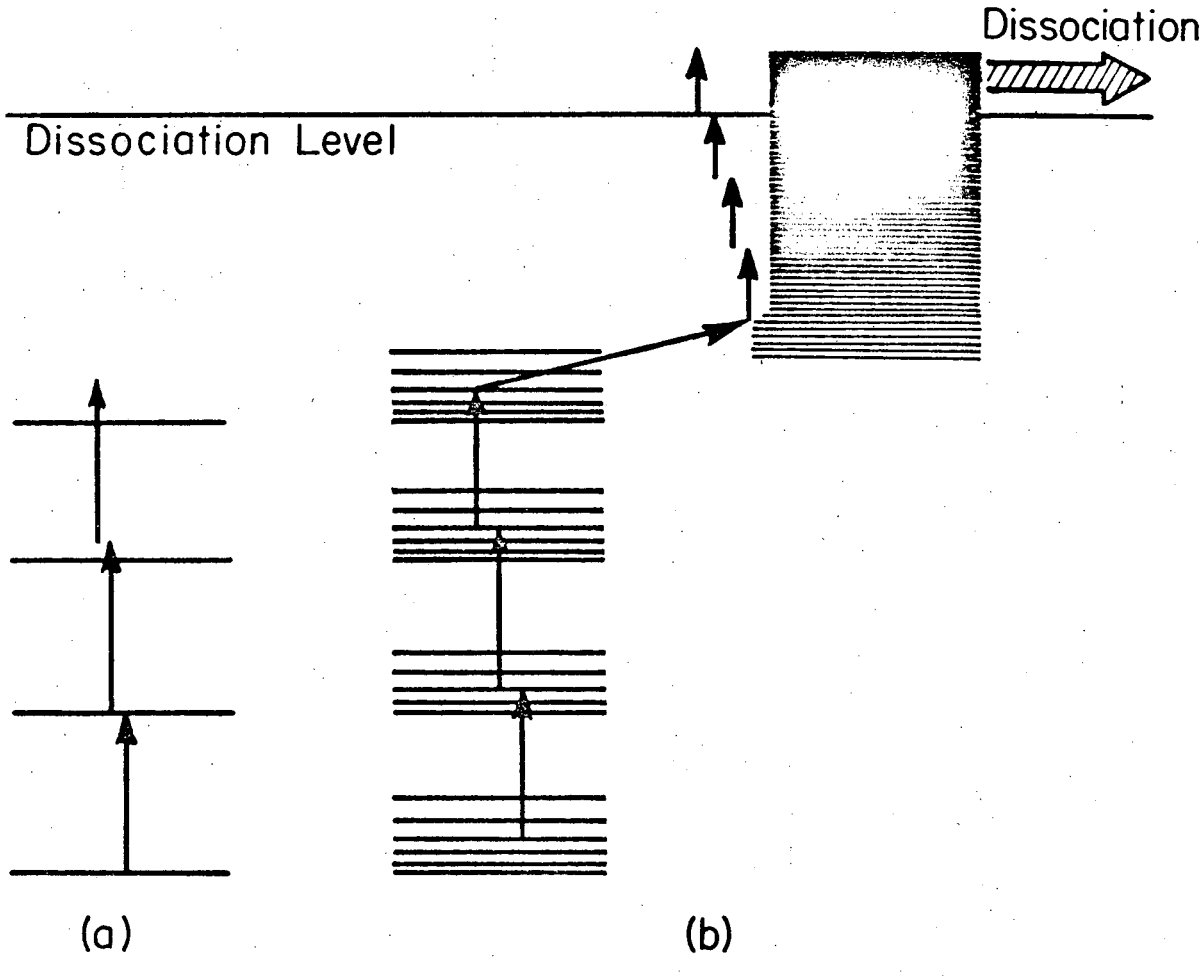
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This report was done with support from the United States Energy Research and Development Administration. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the United States Energy Research and Development Administration.

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