basis of the one-electron model.[3] We suggest that further work is needed to calculate multi-electron excitation in the framework of the many-electron model.

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Raman-Induced Kerr Effect*

D. Heiman, R. W. Hellwarth, M. D. Levenson, and Graham Martin
Electronics Sciences Laboratory, University of Southern California, Los Angeles, California 90007
(Received 18 November 1975)

A new effect is demonstrated in which a laser pulse can be made to induce a Kerr effect only at Raman-shifted frequencies. This permitted the observation of a Raman spectrum with a single dye-laser pulse. The pulse power required is a small fraction of that required to produce stimulated instabilities. The spectral information emerges in a coherent beam, phase matching is not necessary, and spectra can be obtained at any scattering angle.

Spontaneous Raman scattering has been a productive tool for studying the excitations of the scattering medium. However, low signal levels have precluded the use of Raman scattering in short-lived or high-background media such as explosions, flames, shocks, and sparks. Nonlinear optical effects, such as the stimulated and inverse Raman effects, and coherent anti-Stokes Raman scattering, have been employed in attempts to increase the effective signal-to-noise ratio in Raman spectroscopy.[1-4] Each of these effects has disadvantages that have limited its usefulness. In this paper we demonstrate a new nonlinear optical effect which can produce a complete Raman spectrum in a coherent beam with high scattering efficiency, without phase-matching requirements, and without a concomitant background from nonresonant nonlinearities. We first describe this "Raman-induced Kerr effect" by several experimental examples, and then outline the theory, which suggests useful variations on our experiments.

In the Raman-induced Kerr effect, a strong polarized monochromatic pump beam at frequency ν induces complex, anisotropic changes in the refractive indices experienced by a weak probe

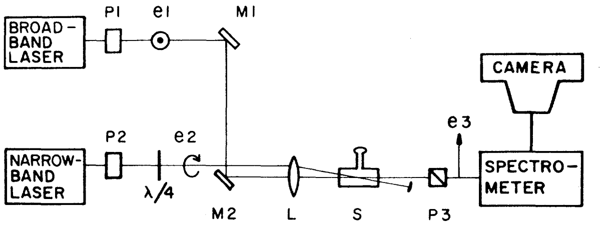

Fig. 1. Apparatus for observing Raman-induced Kerr effect. Glan polarizer $P1$ produces probe polarization $e1$. Glan polarizer $P2$ and mica $\lambda/4$ plate produce pump polarization $e2$. Probe and pump beams are made to overlap over a region ($\sim 8\times 0.1\times 0.1$ mm^3) in sample S by mirrors $M1$ and $M2$ and a 25-cm focal-length lens L. Glan analyzer $P3$ passes polarization $e3$ into spectrometer.

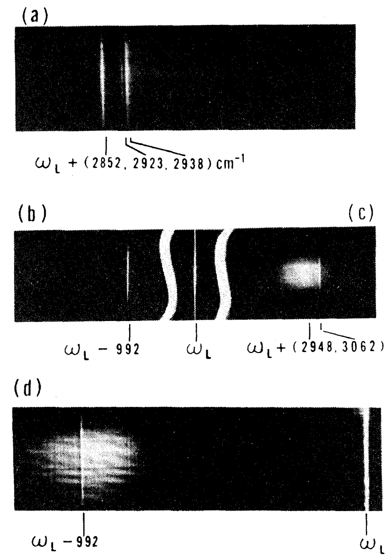

Fig. 2. Spectra taken with ASA-400 film, four pulses per exposure under polarization conditions described in text. Samples were (a) cyclohexane and (b)–(d) benzene. Asymmetric line shapes result from interference as described in the text. Structure appearing in background transmission in (c) and (d) is from spurious interfering reflections. The frequency of the narrow-band pump laser is marked ω_L.

beam at frequency ω. These changes exhibit resonances when $|\omega - \nu|$ is near the frequency of a Raman-active vibration in the scattering medium. Being anisotropic, these changes cause the exiting polarized probe beam to be partially transmitted through a polarizer that normally blocks it. The spectrum of the transmitted probe beam reflects the spontaneous Raman spectrum.

In Fig. 1 we show the experimental setup with which we observed the Raman-induced Kerr effect in several organic liquids by producing a photographic display of their Raman spectra. Polarized pump and probe beams (~ 30 kW for 8 nsec) were generated by nitrogen-laser–pumped dye lasers.[4] The linearly polarized probe beam had a continuous (~ 800 cm^{-1} wide) spectrum which enabled us to probe a large portion of the spectrum with a single pulse. The probe light exiting from the sample S was viewed through a "crossed" linear polarizer $P3$ aligned to block the probe when the pump beam was off. Figure 2 shows some typical spectra of transmitted probe beams tuned to different frequency ranges. In Figs. 2(a) and 2(b) the collinear pump beam was essentially circularly polarized (~ 99%), resulting in Stokes and anti-Stokes spectra with no observable background transmission. Theory predicts that transmission at a negative (Stokes) shift $(-w)$ is the same as at the corresponding positive (anti-Stokes) shift w; our observations were consistent with that prediction.

When the probe polarizer ($P3$ of Fig. 1) was slightly misaligned, a small fraction of the probe light was always transmitted. This interfered with the Raman-induced Kerr effect, as can be seen in Fig. 2(c). Figure 2(c) also shows that the "depolarized" (2948 cm^{-1}) and the "polarized" (3062 cm^{-1}) Raman lines of benzene, whose scattering cross sections differ by more than an order of magnitude, are visible on the same photograph.

When the pump-beam polarization was made slightly elliptical (~ 2% opposite circular), a stimulated background transmission occurred at all probe frequencies. This nonresonant transmision arose from the optical Kerr effect,[5,6] and interfered with the Raman-induced Kerr effect. The resulting constructive and destructive interference is illustrated in Fig. 2(d). Interference between neighboring Raman lines also can occur, as illustrated in Fig. 2(a). The interference phenomena give a more complex spectral profile than does ordinary Raman scattering.

The laser intensities used were less than those required to produce observable stimulated Raman scattering, as we verified by an unsuccessful search for stimulated lines with either the pump or the probe beam blocked. We also failed to find any pump-induced changes in the probe beam when the crossed polarizer $P3$ was removed. All the foregoing results were predicted from the following theory.

In the ideal Raman-induced Kerr effect, the probe field is weak enough so that the nonlinear electric polarization density is linear in its amplitude and quadratic in the pump field amplitude. This means that we may study the propagation of each frequency component of the probe field separately, superposing the results when, as in our experiments, a broad-band probe wave is used. The strong optical pump wave is monochromatic, so that we may write the ith spatial component of the total optical field in the medium as $\mathrm{Re}(F_i e^{-i\nu t} + E_i e^{-i\omega t})$, where F_i and E_i are the spatially varying pump and probe field amplitudes. The nonlinear electric polarization density at the probe frequency ω is written conventionally as $\mathrm{Re}[6c_{ijkl}(-\omega, \nu, -\nu, \omega)F_j F_k^* E_l e^{-i\omega t}]$.[6] Here as below, repeated space indices are assumed to be summed, and esu are used. The frequency arguments here are implied, if omitted.

To see the effect of this term on the probe field, we write E_i as $[e_i + s_i(\vec{r})]E(\vec{r})$, where e_i is the unperturbed probe polarization (normalized so that $e_i^* e_i = 1$) and $E(\vec{r})$ is the unperturbed probe amplitude in the absence of the nonlinear polarization density which causes the small correction $s_i(\vec{r})$ (of order F^2). Substituting these forms in Maxwell's equations, and solving iteratively, gives the following equation for the correction s_i at a distance L into the medium along a "stream line" in the vector field defined by $\mathrm{grad}E$[7]:

$$s_i(L) = (12\pi i\omega/nc)\int_0^L dx\, c_{ijkl} F_j F_k^* e_l, \qquad (1)$$

where the integral is taken along the stream line. The linear refractive index is n; the velocity of light is c.

With this explicit solution for the complex change in polarization anywhere on the exiting-probe-beam profile, we can calculate the result of any Kerr-type measurement on the probe beam. The results of such a calculation for the usual nonresonant "optical" Kerr effect are well known: In an isotropic medium a linearly polarized pump field produces a birefringence for the probe beam.[5,6] In our observations of the Raman-induced Kerr effect we could eliminate the nonresonant background arising from this usual optical Kerr effect by making the pump beam circularly polarized. To see how this possibility follows from (1), we must have an expression for the tensor properties and frequency dependence of c_{ijkl}. We will use the Born-Oppenheimer approximation (BOA) to obtain such an expression that is both useful for our experimental conditions and related to the Raman-scattering spectrum of the isotropic medium. (The BOA assumes that the optical fields produce no electronic transitions; the electrons follow the nuclear motions adiabatically.) In the BOA, c_{ijkl} may be expressed in terms of the real bulk electronic "hyperpolarizability" coefficient σ, plus two real, causal nuclear response functions $a(t)$ and $b(t)$.[7,8] In terms of the Fourier integral transforms A_w and B_w at angular frequency w of the latter,[8,9]

$$24c_{ijkl} \to \delta_{il}\delta_{jk}(\sigma + 2A_0 + B_w) + \delta_{ik}\delta_{jl}(\sigma + B_0 + B_w) + \delta_{ij}\delta_{kl}(\sigma + B_0 + 2A_w). \qquad (2)$$

Here, $A_0 \equiv A_w$ evaluated at $w = 0$, etc., and $w \equiv \omega - \nu$.

The fraction of any probe ray that will be transmitted by a polarizer which passes polarization f_i (normalized so that $f_i^* f_i = 1$) is $|f_i^* s_i|^2$. Therefore (1) and (2) give the fraction of an exiting probe beam at frequency $\nu + w$ that passes through a crossed linear polarizer on axis. For a circularly polarized pump beam, this fraction is[6]

$$|(2\pi^2\omega/n^2c^2)(2A_w - B_w)\int_0^L dz\, I(z)|^2, \qquad (3)$$

where $I(z)$ is the pump intensity at z along the probe beam axis. Nonresonant terms proportional to $\sigma + B_0$ have canceled out. To see, for example, that an isolated Lorentzian Raman resonance in the sample will cause a Lorentzian resonance in (3) of the same width and shift, we recall that[9]

$$\mathrm{Im}B_w = \frac{\pi c^4}{\hbar\nu\omega^3}\frac{d^2\sigma_{xy}}{d\Omega\, dw}(e^{\hbar\omega/kT} - 1). \qquad (4)$$

Here, $d^2\sigma_{xy}/d\Omega\, dw$ [$\mathrm{cm}^2\ \mathrm{cm}^{-3}\ \mathrm{sr}^{-1}\ (\mathrm{rad/sec})^{-1}$] is the differential Raman scattering cross section per unit volume to scatter photons from polarization x to polarization y (x, y, z is some Cartesian system) and from frequency ν to $\omega = \nu + w$. We take (4) to include a companion relation in which A_w and $\frac{1}{2}\sigma_{xx} - \sigma_{xy}$ replace B_w and σ_{xy}, respectively.[9] The real parts of A_w and B_w are obtained from their imaginary parts by the usual Kramers-Kronig relation. From this it is evident that an isolated Lorentzian Raman vibration of frequency w_R and half-width γ contributes a term proportional to $|w + i\gamma - w_R|^{-2}$ to the transmitted fraction (3). Furthermore, the spectrum of the Raman-induced Kerr effect can be predicted from (4) in (1) and (2) for any frequency dependence of the Raman cross section, and for arbitrary pump- and probe-beam polarizations and directions. For example, the interfering Kerr-effect background proportional to $\sigma+B_0$, that was observed in Fig. 1(d) when the pump beam was elliptically polarized, is easily calculated.

The Raman-induced Kerr effect we have described is closely related to the stimulated and inverse Raman effects which involve different tensor elements of c_{ijkl} but the same frequency arguments $(\pm\omega, \pm\nu)$. In the latter effects one observes stimulated gain or loss in a probe wave,[2,6,8] whereas we have observed a stimulated optical anisotropy.[8] In all these effects, momentum and energy are conserved for any two frequencies ω and ν, and for any two relative beam angles. By contrast, the four beams (at three or more frequencies) involved in the coherent anti-Stokes Raman scattering effect must obey the familiar, and restrictive, "phase matching" conditions.[4,6]

In summary, we have found, as theory predicted,[8] that the Raman-induced Kerr effect (a) yields a complete Raman spectrum of a substance in times on the order of nanoseconds; (b) requires less laser power than is required for observable stimulated Raman scattering; (c) can provide the Raman spectrum in a coherent beam, which allows spatial filtering of background noise; (d) gives equivalent spectra at frequencies below (Stokes) and above (anti-Stokes) the pump frequency; (e) allows the broad-band background from the optical Kerr effect to be eliminated by proper choice of pump polarization; (f) allows interference between Raman lines and the optical Kerr effect, which can be used to calibrate one effect in terms of the other; and (g) requires no index matching, and thus can yield a spectrum with the probe beam at any angle to the pump beam. This latter capability allows the probing of spectra for a wide range of scattering wave vectors, and for a wide range of scattering frequencies at a fixed scattering direction. Media, like plasmas, in which phase matching is impossible can be studied with this effect. The observation of a Raman-induced optical Kerr effect does not depend on the the coherence of the probe beam. However, there are many advantages to using a spatially coherent probe beam. For example, one can then eliminate strong background signals due to fluorescence or elastic scattering by spatial filtering.

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Absolute Parametric Excitation by an Imperfect Pump or by Turbulence in an Inhomogeneous Plasma

G Laval, R. Pellat, and D. Pesme
Centre de Physique Théorique de l'Ecole Polytechnique, 91120 Palaiseau, France*
(Received 1 May 1975)

Finite-bandwidth effects are investigated by using the random phase approximation. The conditions of validity are checked by solving exactly a particular model. It is found that a finite coherence length for the pump wave can favor the existence of absolute instability.

Problems of laser fusion, plasma heating, and ionospheric modifications have drawn attention to the importance of plasma inhomogeneities for the excitation of parametric instabilities. The inhomogeneity introduces a wave number mismatch which depends on the coordinate x. In many cases this dependence can be approximately considered as linear. In such cases, it is now

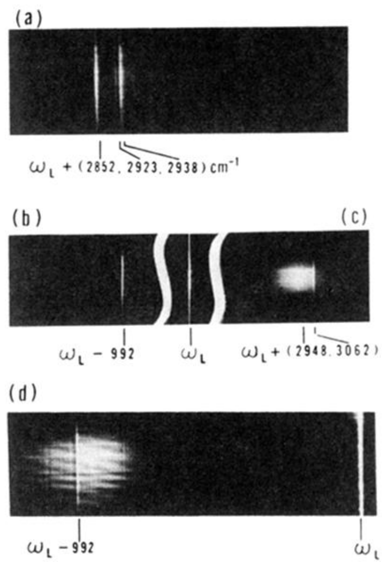

Fig. 2. Spectra taken with ASA-400 film, four pulses per exposure under polarization conditions described in text. Samples were (a) cyclohexane and (b)–(d) benzene. Asymmetric line shapes result from interference as described in the text. Structure appearing in background transmission in (c) and (d) is from spurious interfering reflections. The frequency of the narrow-band pump laser is marked ω_L.