

Momentum Conservation in Raman-Induced Optical Kerr Effect

Tzer-Hsiang Huang, Chia-Chen Hsu, Tai-Huei Wei, and Jiunn-Yuan Lin

*Department of Physics, National Chung-Cheng University,
Ming-Hsiung, Chia-Yi, Taiwan 621, R.O.C.*

(Received November 7, 2001)

In the nonlinear interaction of femtosecond (fs) laser pulses with some simple molecular liquids we have observed and proved vibrational quantum interference (VQI) in the accompanying Raman processes. An analysis is given in this report of the associated momentum conservation. In addition to the propagation directions of the attending electromagnetic waves, the dispersion of the refractive indices around the vibrational resonances is invoked. The analysis leads to a prediction of active normal vibrations as well as the experimental condition for observing the VQI.

PACS. 42.50.Md – Optical transient phenomena: quantum beats, photon echo, free-induction decay, dephasings and revivals, optical nutation, and self-induced transparency.

PACS. 78.47.+p – Time-resolved optical spectroscopies and other ultrafast optical measurements in condensed matter.

I. Introduction

Nonlinear light-matter interaction can take place via very many processes and usually requires a careful analysis of momentum conservation in a macroscopic sense. As one of very many nonlinear processes, the optical Kerr effect (OKE) requires three incident laser fields (two pump and one probe) and one signal field whose polarization is rotated with respect to the probe field. The Raman-induced optical Kerr effect (RIKE) is based on the polarization rotation of the incident probe field due to the Raman resonances caused by the two pump fields. It was observed in the frequency domain as early as 1976 [1]. In a few years, it became one of the useful stimulated Raman techniques [2]. To observe the RIKE in a sample one needs a cw laser of fixed frequency nonlinearly interacting with another cw laser of tunable frequency. The latter laser is tuned to generate a Raman spectrum. Dynamic information attending the nonlinear process is deduced indirectly, through a Fourier transformation of the spectrum. To study slow dynamics one needs to record low frequency Raman spectra, which is not so easy experimentally.

After the invention of a femtosecond (fs) laser in 1981 [3], direct time-domain study of slow, as well as fast, molecular dynamics became possible [4, 5]. The electronic response of transparent molecular liquids to the incident fs laser field, as observed in the RIKE, is instantaneous, since no electronic resonance is involved. Next fastest in response is the molecular vibrations that are set in coherent motion by the fs laser pulses; the ensuing dephasing (or decoherence) are detected, in the form of quantum beats, by delaying the probe pulse. This coherent molecular vibration and the associated beats are characteristic of the time domain RIKE, as compared with

its frequency domain counterpart. Vibrations of some molecules have so far escaped detection in the time-domain RIKE, although the corresponding Raman transitions in the frequency domain are dipole allowed. Given a molecular vibration, whether the allowed Raman transition can be observed in the time-domain RIKE or not depends not only on energy conservation but also on momentum conservation. Although momentum conservation in the OKE is automatically fulfilled, we discover in this report that momentum conservation for vibrational resonances in the OKE is stringent. Namely, if the focusing angle of the laser beam is not large enough, momentum cannot be conserved for energetic vibrations, which, in turn, cannot be observed in the time domain RIKE, even if energy conservation is fulfilled.

The focusing effect of an incident laser beam on the third-order polarization was studied in 1975 by Bjorklund [6], who invoked the phase shift of $\pi/4$ radians that occurs as a beam traverses the region of focus in the sample and hence introduces a slip in phase between the driving field and the generated radiation. The generated radiations originating from different locations in the sample will interfere destructively unless this slip in phase is compensated by a wave vector mismatch of the proper sign. In our case the focusing effect arises from vibrational resonance in the Raman process caused by a femtosecond laser pulse.

II. Theoretical background

A powerful light field can induce in a molecular system the third order polarization [7]

$$P_i^{(3)}(t) = 2_{\pm} K \int_{i=1}^3 d\omega_1 \int_{i=1}^3 d\omega_2 \int_{i=1}^3 d\omega_3 r_{ijkl}^{(3)}(t; \omega_1; \omega_2; \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3); \quad (1)$$

where 2_{\pm} is electric permittivity in free space and K is the symmetry factor for the particular nonlinear process involved. $r_{ijkl}^{(3)}$ is the third order response function of the sample; the subscript i stands for the direction of polarization and $j, k,$ and l are the oscillation directions of incident fields, two supplied by the pump pulse and one by the probe pulse. Both the pump and probe pulses are derived from a single laser pulse that lasts for only about 20fs. The electric field of a fs laser pulse can be written as a superposition of quasi-monochromatic waves, each of which has some characteristic frequency ω_0 [7],

$$E_m(\mathbf{r}; t) = \frac{1}{2} \sum_{\omega_0} \hat{\mathbf{e}}_m E_{\omega_0}(t) e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_0 t)} + C.C.; \quad (2)$$

where $\hat{\mathbf{e}}_m$ is the unit vector for the electric field and $C.C.$ means complex conjugate. $E_{\omega_0}(t)$ is the envelope function that varies much slower than the carrier wave $e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_0 t)}$. After some manipulation equation (1) can be rewritten as [8]

$$P_y^{(3)}(t) = 2_{\pm} K e^{i\omega_0 t} \int_{i=1}^3 \frac{1}{3} E_{\pm}(t) E_u(t) j^2 + E_{\pm}(t) \int_{i=1}^3 b(t; \omega) E_u(\omega) j^2 d\omega \\ + E_u(t) \int_{i=1}^3 a(t; \omega) + \frac{1}{2} b(t; \omega) E_{\pm}(\omega) E_u^*(\omega) d\omega; \quad (3)$$

In this equation the direction (y) of the third-order polarization is chosen to be perpendicular to the x -polarized probe field (labeled with the numerical subscript \pm). The pump field (labeled with the numerical subscript u) oscillates along the bisector of the positive x - and y -axes and hence has equal x - and y -components. The probe pulse arrives at the sample at time t when the y -component of the third order polarization, $P_y^{(3)}$, is to be measured, whereas the pump pulse arrives at a time ζ earlier (ζ is called the delay or delay time). The causality condition is satisfied: cause (pump) precedes the result $P_y^{(3)}$. The first two terms in equation (3) give rise to the ordinary Kerr effect: the effect of the pump intensity, $I_u(\zeta) = j \bar{E}_u(\zeta)j^2$, on the refractive index of the sample is interrogated by the probe field, $\hat{x} E_o(t=0)$. The first term arises from the electronic response ($\zeta = 0$). The corresponding response function χ is instantaneous (behaves like a \pm -function), as it does not involve any electronic resonance (absorption). The second term arises from the nuclear response. The corresponding response function $b(t; \zeta)$ has memory because nuclei are too heavy to respond instantaneously and also because some normal vibrations are Raman-excited (*vide infra*). It memorizes the aftereffect of the pump intensity. The third term gives rise to an effect called coherent coupling [9]. The pump field $\bar{E}_u(\zeta)$ interferes with the coherent probe field $\bar{E}_{\pm}(\zeta)$ to form a transient grating in the sample. Meanwhile, the second pump field, $E_u(t)$, scatters off the grating and propagates in the direction of the probe beam. The corresponding response functions $a(t; \zeta)$ and $b(t; \zeta)$ differ from the delta function. As such they have memory, due likely to the response of the molecular skeleton. If the pump and probe pulses do not overlap in time, their fields are unable to interfere and hence there is no coherent coupling to talk about. Although the transient grating might last for a time longer than the pulse duration because of the associated molecular response, the field experiencing the aftereffect is the second pump field. As a result, once the pump pulse is extinguished, coherent coupling should disappear.

The third order polarization given by equation (3) arises from the interaction of three laser fields with the sample, via various response functions. Henceforth, we focus on the second term, with the nuclear memory function $b(t; \zeta)$. The associated momentum conservation is analyzed in the following. Fig. 1 shows that the pump and probe beams are focused to cross in the sample cell. They are formed from a train of fs pulses derived from a single laser source. Being ultra short in duration ($\gg 16$ fs is the shortest), each of the pump and probe pulses consists of very many Fourier frequency components, $\omega_1; \omega_2; \text{ccc}; \omega_n$, and the corresponding propagation numbers, $k_1; k_2; \text{ccc}; k_n$, n being on the order of 10^4 . The cross sections of the pump and probe beams are $\gg 1$ mm in diameter. This cross-section corresponds, with a $f/6$ cm focusing lens, to a beam-focusing angle (μ) of about 1^\pm . The third-order polarization $P_y^{(3)}(t)$ generates the signal field, \bar{E}_{OKE} (left, Fig. 1), which is interfered with and hence enhanced by the local oscillator field, \bar{E}_{LO} (upper right, Fig. 1)—dubbed optical heterodyne detection (OHD).

Of the enormous number of rays in the pump beam, let us consider the complex conjugate of the $(\omega_1; k_1)$ wave in ray 1 and the $(\omega_2; k_2)$ wave in ray 2, as indicated at the top of Fig. 2. The electric fields of these two waves are

$$\bar{E}_1^u(\mathbf{r}; t) = \hat{e}_y \bar{E}_1^u(t) e^{i(\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)} \quad (4)$$

and

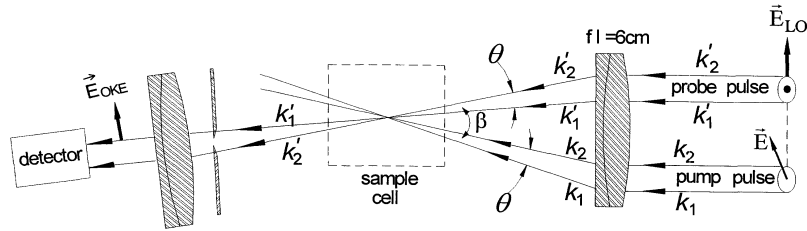


FIG. 1. Pump and probe pulses are focused at the sample cell. Both pulses have exactly identical content in wave propagation number k_i , though not in κ . \hat{x} stands for the \hat{x} -polarization of the probe field pointing out of the paper plane. \hat{y} , \hat{z} and \hat{y} (\hat{z} (pump)) bisect the \hat{x} and \hat{y} axes.

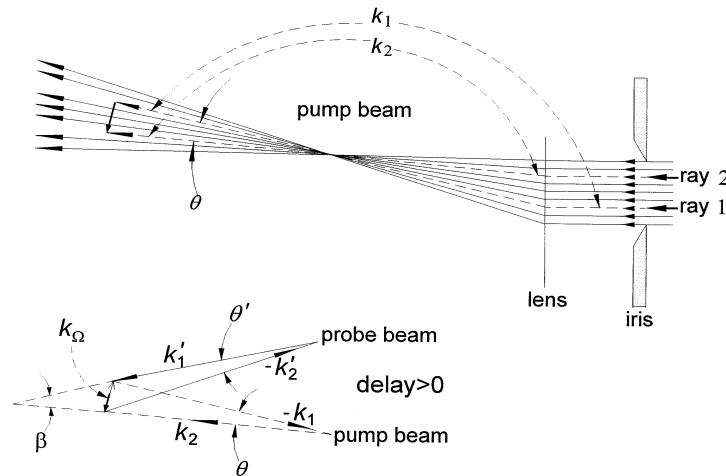


FIG. 2. Top: the pump pulse formed of many rays are focused towards the sample cell. Each of the rays consists of wave propagation numbers ($k_1; k_2; \dots; k_n$). Of the rays only ray 1 and ray 2 happen to form the angle μ that satisfies the phase matching shown at bottom: $k_2 - k_1 = k_0$.

$$E_2^u(\mathbf{r}; t) = \hat{e}_x E_2^u(t) e^{i(\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)}, \quad (5)$$

where we use the superscript u to denote the pump pulse, keeping in mind that the third incident field to cause $P_i^{(3)}(t)$ is provided by the probe pulse at a time τ fs later. During the course of interaction between the two pump fields and the sample, energy conservation follows $\omega_2 = \omega_1 + \omega_0$ (upper right, Fig. 3) and momentum conservation follows $\mathbf{k}_2 - \mathbf{k}_1 = \mathbf{k}_0$ (bottom, Fig. 2 and lower right, Fig. 3). As a result of energy conservation, individual molecules in the sample acquire energy $\hbar\omega_0$ whereas, through momentum conservation, the momentum $\hbar\mathbf{k}_0$ is imparted to the sample. The vibrational motions of neighboring molecules in the sample are such that the phonon wave (\mathbf{k}_0) sustains until the thermal motion of the molecules destroys it. For a certain molecular vibration (ω_0) driven by the pump fields (4) and (5) we have

$$\omega_0 = E_1 - E_0;$$

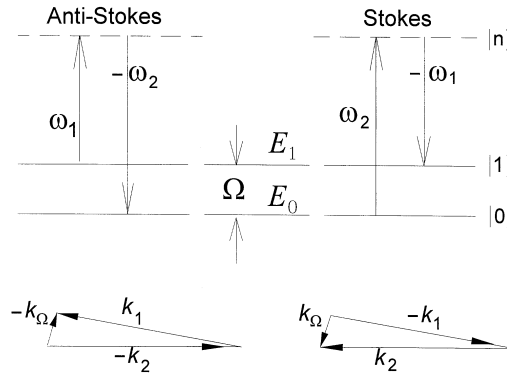


FIG. 3. Energy and momentum ($-k$) conservation for the femtosecond optical Kerr effect.

where E_0 and E_1 , respectively, are the energy of the ground vibrational state $|0\rangle$ and that of the first excited state $|1\rangle$. The ω_1 -vibration is excited to $|1\rangle$, as a result of pumping, in a process known as the Stokes Raman scattering. Henceforth, we focus on the vibrations of simple molecular liquids whose Raman transitions have been well characterized in the frequency domain.

Traveling along a given ray in the pump beam (top, Fig. 2) are numerous wave pairs, for which the energy and momentum conservations are not satisfied and are hence ignored in this report. Of the one that satisfies the conservation requirements, the complex conjugate can also cause light-matter interaction whose energy and momentum conservations are shown in the left half of Fig. 3. The associated process is termed the anti-Stokes Raman scattering. Population in $|1\rangle$ at room temperature under thermal equilibrium is negligibly small since $(E_1 - E_0) \gg kT$ for most of the normal vibrations of interest to us. We thus neglect the anti-Stokes Raman scattering in the pumping process. As a result of pumping the $|1\rangle$ population is no longer negligibly small and hence the probe pulse can cause Stokes and anti-Stokes Raman scatterings with equal probability, leading to vibrational quantum interference [10].

The energy $\hbar\Omega$ needed for energy conservation in the pumping process corresponds to the vibrational energy and falls in the infrared region of the spectrum. The corresponding wavelength, $\lambda_\Omega = 2\pi/\Omega$, ranges from $\gg 1 \mu\text{m}$ to $\gg 100 \mu\text{m}$ (1/10 of the light path through the sample cell), and extends over 10^3 to 10^5 molecules. The momentum ($\hbar k_\Omega$) induced in each of these molecules must be correlated (i.e., have a definite phase relationship) in order to sustain the wave vector k_Ω . That the $(\Omega; k_\Omega)$ wave is established in the sample by the pumping process is manifested by the data presented in Fig. 4. First of all, if signals from individual molecules were uncorrelated as in spontaneous Raman scattering, the signal strength would have been many orders of magnitude weaker than that shown in the figure and at the same time non-directional, contrary to our observation. Thus, the pumping process is stimulated Raman, rather than spontaneous Raman. Further support for the $(\Omega; k_\Omega)$ wave lies in the long lasting oscillation shown in Fig. 4, which originates from the quantum interference of the ω_1 vibration that carries a momentum of $\hbar k_1$. [10]. By analogy to the optical field expressions given by equations (4) and (5), we write the electric field of the excitation wave $(\Omega; k_\Omega)$ as

$$\vec{E}(\vec{r}; t) = \frac{1}{2} \sum_{\vec{k}_\Omega} \vec{e} E_\Omega(\vec{k}_\Omega; t) e^{i(\vec{k}_\Omega \cdot \vec{r} - \Omega t)} + \text{C.C.} \quad (6)$$

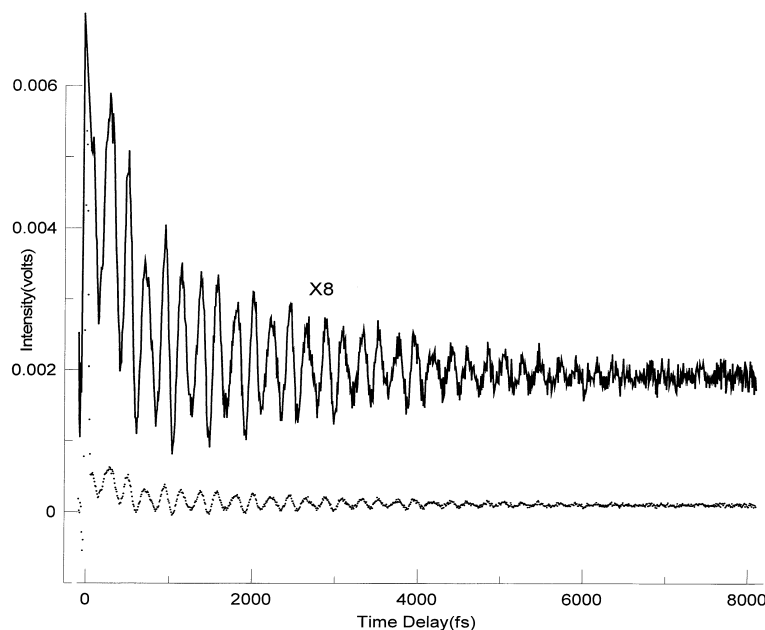


FIG. 4. OHD Kerr signal of CHBr_3 liquid observed at room temperature. Delay time is the time the probe pulse arrives at the sample minus the arrival time of the pump pulse.

The RIKE is a four-wave mixing process, in which the sample receives, in the pump process, a momentum of $\sim \mathbf{k}_-$ from the radiation field and returns the exact amount back to the radiation field in the probe process at a certain delay $\tau \in 0$. The associated phase matching is shown in the lower portion of Fig. 2, where the wave vector \mathbf{k}_2^0 arises from the third-order polarization $\mathbf{P}_i^{(3)}(t)$. (Henceforth, the primed wave vectors pertain to the probe process.) Once the ω_2 -vibration is excited to $|j\rangle$ and the $(-\mathbf{k}_-)$ wave is established, the probe process can occur so long as the excitation wave $(-\mathbf{k}_-)$ is sustained. This material excitation wave cannot propagate as fast as the non-resonant waves (4) and (5) whose frequencies do not agree with any of the (electronic) transitions of the sample molecules.

III. Results and discussion

From the phase matching condition $\mathbf{k}_- = \mathbf{k}_2 + \mathbf{k}_1$ for the pumping process shown at the bottom of Fig. 2 (also lower right, Fig. 3), we have

$$k_-^2 = k_1^2 + k_2^2 + 2k_1k_2 \cos \mu \quad (7)$$

Since $\omega = ck$ and $\omega_- = ck_-$, equation (7) can be converted to

$$\frac{\omega_-}{c} = \frac{\omega_1}{c} + \frac{\omega_2}{c} + 2 \frac{\omega_1}{c} \frac{\omega_2}{c} \cos \mu; \quad (8)$$

$$(\omega_-)^2 = (\omega_1)^2 + (\omega_2)^2 + 2(\omega_1)(\omega_2) \cos \mu;$$

or, with energy conservation $\theta_2 = \theta_1 + \theta$,

$$\begin{aligned} (n_- \cos \theta)^2 &= (n_1 \cos \theta_1)^2 + [n_2 (\theta_1 + \theta)]^2 - 2(n_1 \cos \theta_1)n_2 (\theta_1 + \theta) \cos \mu; \\ (n_-^2 - n_1^2) \cos^2 \theta &= n_1^2 (n_1^2 - n_2^2) \cos^2 \theta_1 + 2n_1 n_2 \cos \mu + n_2^2 + 2\theta_1 - (n_2^2 - n_1 n_2 \cos \mu); \end{aligned} \quad (9)$$

The refractive indices n_1 and n_2 are approximately equal, since the corresponding wavelengths λ_1 and λ_2 are so close to each other, both being very far from resonance for the transparent samples we used. Taking $n_1 \cong n_2$ equation (9) becomes

$$\mu \frac{n_-^2}{n_1^2} \cos^2 \theta - 2\theta_1 \cos \mu \cong 2\theta_1^2 (1 - \cos \mu) + 2\theta_1 - (1 - \cos \mu); \quad (10)$$

which we rewrite as

$$\mu \frac{n_-^2}{n_1^2} \cos^2 \theta - 2\theta_1 \cos \mu \cong 2\theta_1 (\theta_1 + \theta) (1 - \cos \mu); \quad (11)$$

or, in view of $\theta_2 = \theta_1 + \theta$, as

$$\mu \frac{n_-^2}{n_1^2} \cos^2 \theta - 2\theta_1 \cos \mu \cong \theta_2 (1 - \cos \mu); \quad (12)$$

The refractive index n_- is associated with the wavelength $\lambda_- = 2\pi/k_-$ that is in resonance with the normal mode ω_- . It is thus true that $n_- > n_1$ and $n_-^2 > n_1^2$.

Wavelengths λ_1 and λ_2 are usually so close that we can take $\theta = \theta_1 \cong \theta_2$. With this approximation equation (12) becomes

$$\mu \frac{n_-^2}{n_1^2} \cos^2 \theta - 2\theta \cos \mu \cong 2\theta (1 - \cos \mu); \quad (13)$$

where $\theta = 7.327 \times 10^{14}$ Hz is taken from the carrier wavelength (820 nm) of the titanium-sapphire laser we used. For the small molecules of interest to us the energies (ω) of most of the fundamental vibrations are known and hence the factor $(\omega - \omega_-)^2$ in equation (13) is also known. Thus, given a normal mode (with a known ω), we have one equation, (13), and two unknowns, $X = n_-/n_1$ and μ . This equation can be solved by plotting

$$Y(\mu) = 1 - \cos \mu \quad (14)$$

and

$$Y^0(x) = \frac{1}{2} (x^2 - 1) \frac{\omega - \omega_-}{\omega_-}; \quad (15)$$

and by finding a common functional value for both equations. This is illustrated in the following for two liquid samples.

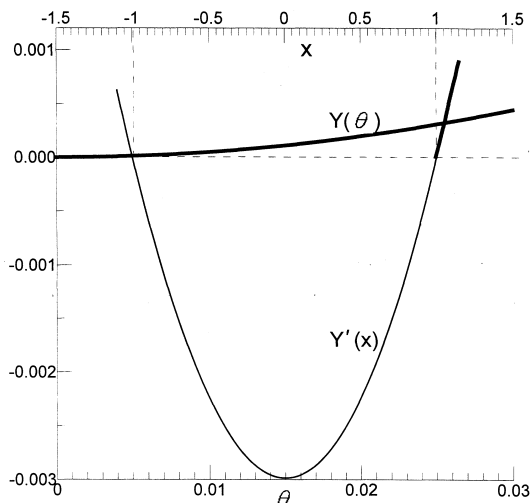


FIG. 5. Plots of $Y(\mu) = 1 - \cos \mu$ and $Y^0(X) = (X^2 - 1)/355$ for liquid CS_2 , where μ is in radian and $X = n_2/n_1$ and $\omega = 656 \text{ cm}^{-1}$.

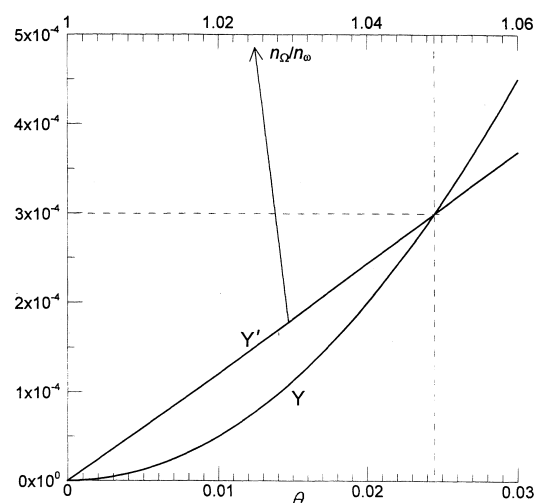


FIG. 6. A blown-up plot of the bold-faced line segments shown in Fig. 5.

We first consider liquid CS_2 . The fundamental energy of the symmetric stretch vibration of the CS_2 molecule in liquid has an angular frequency of $\omega = 2\pi \times 656 \text{ cm}^{-1} = 4\pi \times 10^{13} \text{ Hz}$, corresponding to the fundamental energy of $\omega = 656 \text{ cm}^{-1}$. Replacing this ω into equation (15) we have

$$Y^0(X) = (X^2 - 1)/355 \quad (16)$$

which, along with equation (14), is plotted in Fig. 5 using a single Y-scale. It is noticed that, for $1 < n_2/n_1 < 1$, $Y^0(X)$ is negative whereas $Y(\mu)$ is always positive since $\mu < 90^\circ$. Therefore, equation (13) cannot have any solution in this region. Solutions are possible for $n_2/n_1 > 1$ and for $n_2/n_1 < -1$. The latter is to be dropped because refractive indices cannot be negative. For $n_2/n_1 > 1$ the solution is found graphically from Fig. 6. It occurs at $Y = 0.0003$, where $\mu = 0.0244$ radians (or 1.4°) and $n_2/n_1 = 1.0488$.

We next consider the liquid formed of CBrCl_3 molecules, whose CCl_3 degenerate deformation has a fundamental energy of $\omega_6 = 193 \text{ cm}^{-1}$ and an angular frequency of $\omega = 2\pi \times 5.79 \times 10^{12} \text{ Hz}$. Equation (15) becomes

$$Y^0(X) = (X^2 - 1)/4000; \quad (17)$$

which, along with equation (14), is plotted in Fig. 7. From the plots in the upper half of the figure it is noticed that solutions can be found in a manner similar to the case of CS_2 . The plots of $Y(X)$, for $n_2/n_1 > 1$, and $Y(\mu)$ are blown up in the lower half of Fig. 7. The solution is found to occur at $Y = 8.1 \times 10^{-6}$, where $\mu = 0.00402$ radians (or 0.23°) and $n_2/n_1 = 1.0162$.

From the two examples given above we learn that the more energetic the molecular vibrations are, the larger n_2/n_1 and μ are required to be. For example, with the 656 cm^{-1} vibration of the CS_2 molecule, as compared with the 193 cm^{-1} vibration of CBrCl_3 , the μ (1.4°) and n_2/n_1

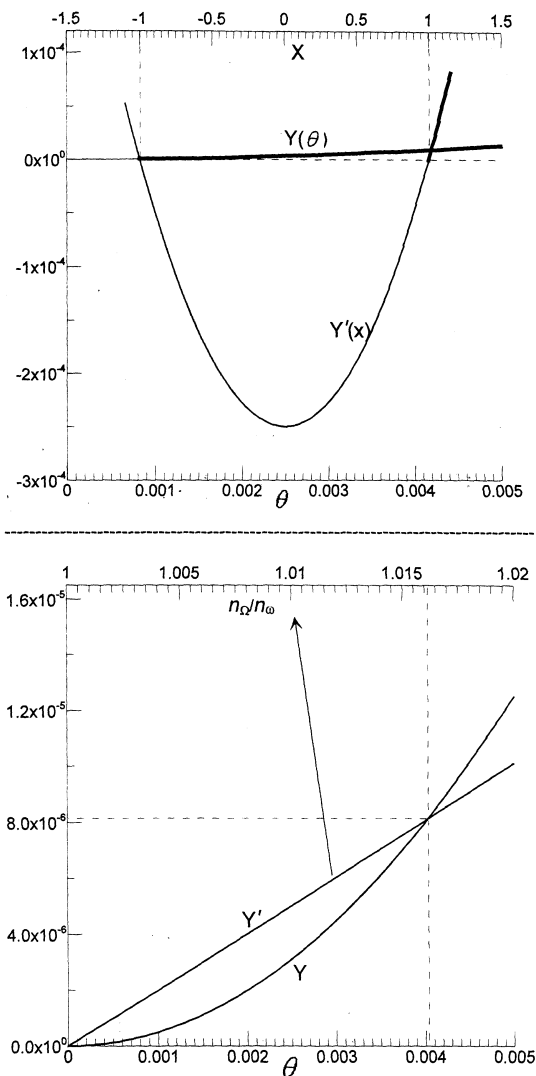


FIG. 7. Plots of $Y(\mu) = 1 - \cos \mu$ and $Y'(X) = (X^2 - 1)/4000$ for CBrCl_3 , where μ is in radian. Plots in the lower half is a blown-up of the bold-faced line segments in the upper half.

(1.0488) that have to be satisfied are larger than those of CBrCl_3 . This refractive index requirement is easy to satisfy, in view of dispersion of the refractive index at resonance. To satisfy $\mu = 1.4^\pm$ (and larger) we have to either expand the laser beam before focusing on the sample or use a focusing lens with a focal length shorter than the one we used, keeping in mind that the titanium:sapphire laser we used is in the TEM_{00} mode and has a very small cross section. This is likely the reason why the 656 cm^{-1} vibration of CS_2 has eluded detection so far. Similarly, other higher-energy vibrations will require a larger μ angle.

Given a liquid sample, usually more than one normal vibrations of the molecule are involved in the OHD RIKE signals such as the ones shown in Fig. 4, where the beats do not appear

harmonically due to the interference of more than one frequency. By decreasing the μ angle the higher frequency vibrations will be suppressed and as a result the oscillatory signal will appear more harmonic. When the μ angle is small enough only the least energetic vibration is active. Then, we should see harmonic beats in the OHD RIKE signals. As we decrease the μ angle experimentally, $n_{\omega} = n_1$ should decrease toward unity, as required by equation (13). The latter condition is automatically fulfilled because of the characteristic dispersion of n_{ω} at the resonance frequency given by $\omega = E_1 - E_0$.

If $n_{\omega} = n_1$, then $\mu = 0$, as required by equation (13). In this case, the pump/probe beams are collinear, and equation (7) becomes

$$k_{\omega}^2 = k_1^2 + k_2^2 + 2k_1k_2 \quad (18)$$

or

$$k_{\omega} = |k_2 + k_1| \quad (19)$$

The corresponding momentum conservation is depicted in the left half of the c diagram in Fig. 8, where k_1 , k_2 and k_{ω} are collinear. Meanwhile, k_{ω} must be collinear with the wave vectors k_1^0 and k_2^0 of the probing process, see the right half of the c diagram in Fig. 8. Consequently, the pumping and probing wave vectors as well as the material excitation wave vector are all collinear. Notice that the signal wave k_2^0 travels in the same direction as the phonon wave k_{ω} does. It is then interesting to see if the detector shown in Fig. 1 also detects the k_{ω} wave in addition to the signal wave k_2^0 . This is an experimental situation that we have yet to try; it was excluded by the apparatus setup we used. Note that $n_{\omega} = n_1$ can be easily satisfied because of the dispersion in n_{ω} around a resonance.

In the ordinary OKE experiment with fs laser pulses, no material excitation need be invoked since the OKE response is usually probed without any delay. In that case only electronic motion

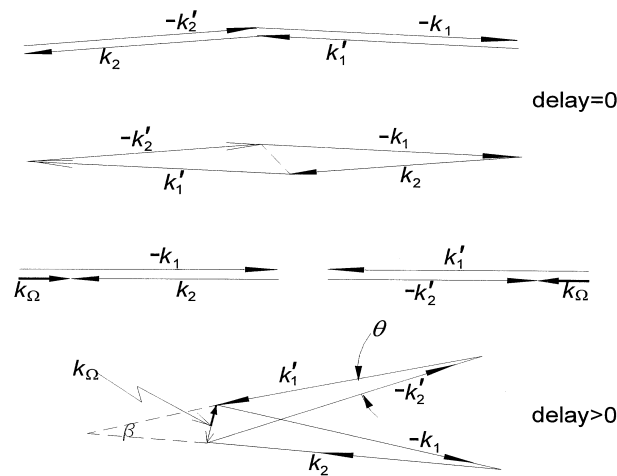


FIG. 8. Wave vector conservations for pump-probe femtosecond optical Kerr experiment. The primed k_i vectors belong to the probing process.

can be detected since the nuclear motion is too slow to respond to fs pulses. Once the pump pulse extinguishes, the OKE signal should disappear; whatever signals remaining after delay zero must arise from a nuclear response, e.g., the vibrational resonances. The strong peak at delay zero in Fig. 4 arises from the electronic response. The associated momentum conservation is shown in the a and b diagrams of Fig. 8, where the momentum $\sim k_{\perp}$ that we have to invoke for momentum conservation need not have memory. Hence, the associated n_{\perp} cannot possibly be equal to n_{\parallel} because the field oscillating frequencies ω and ω' are so much different. As such, the μ angle will differ from zero and hence the momentum conservation shown in c, Fig. 8 is not appropriate for ordinary OKE. The long lasting oscillation shown in Fig. 4, which decays and lasts for almost 8 picoseconds, cannot be attributed to the ordinary OKE. Rather, it arises from the RIKE with the momentum conservation that we have discussed so far. For easy comparison with other processes that can occur in the four wave mixing in question we reproduce in d, Fig. 8 the momentum conservation of the RIKE.

IV. Conclusion

Going through the momentum conservation analysis for the coherent transient optical Kerr experiment, we have come to a clear understanding as to why the 656 cm^{-1} vibration of CS_2 has escaped detection so far. It is because the focusing angle of the excitation laser is not large enough. The analysis applies to another similar experiment where two separate pump beams rather than one (as in our case) are used to excite the coherent vibrational wave. Our preliminary experimental data agree with the results of the momentum conservation analysis presented in this report. We are gathering more data for a stronger support.

References

- [1] D. Heiman, R. W. Hellwarth, M. D. Levenson and G. Martin, Phys. Rev. Lett. **36**, 189 (1976).
- [2] G. L. Eesley, *Coherent Raman spectroscopy*, (Pergamon, Oxford, 1981), Chap 3.
- [3] R. L. Fork, B. I. Greene and C. V. Shank, Appl. Phys. Lett. **38**, 671 (1981).
- [4] Sanford Ruhman, Alan G. Joly, and Keith A. Nelson, J. Chem. Phys., Letters to the Editor, **86**, 6563 (1987) and IEEE J. Quantum Electron., **QE-24**, 460 (1988).
- [5] Dale McMorro, William T. Lotshaw, and Geraldine A. Kenney-Wallace, Chem. Phys. Lett. **145**, 309 (1988).
- [6] Gary C. Bjorklund, IEEE J. Quantum Electronics, **QE-11**, 287 (1975).
- [7] Paul. N. Butcher and David Cotter, *The Elements of Nonlinear Optics*, (Cambridge University, Cambridge, 1990).
- [8] J.-L. Oudar, IEEE J. Quantum Electron. **QE-19**, 713 (1983).
- [9] T.-H. Huang *et al.*, Chin. J. Phys. **36**, 606 (1998).
- [10] Tzer-Hsiang Huang and Chin-Hsien Wang, Molecular Physics, **86**, 1249 (1995).