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Two-Photon Self-Induced Transparency of Different-Frequency Optical Short Pulses in Potassium*

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Low-loss, coherent two-photon propagation and pulse breakup with peak amplification are observed resulting from two-photon resonant self-induced transparency of different-frequency optical pulses interacting with a three-level system in potassium vapor. Theoretical analysis yielding new area equations for two different-frequency pulses with computer solutions agrees well with the observed results.

Novel propagation effects of coherent short light pulses have been studied in connection with a resonant interaction with a two-level system such as in ruby¹ and gaseous SF₆²⁻⁴ and Rb.⁵ Self-induced transparency (SIT) due to a one-photon resonant transition was first investigated by McCall and Hahn.¹ Recently, coherent two-photon propagation in which twice the propagating frequency is resonant with a two-level system has also been analyzed,^{6,7} though the experimental study has not been reported yet. We wish to present here the first observation and theoretical analysis of a new type of coherent propagation of two different-frequency optical pulses causing a two-photon transition in a gaseous three-level system. The present model excludes two simultaneous transitions in double resonance.

The basic effect of coherent two-photon propagation can be analyzed in the limit of two different-frequency plane waves given as

$$E_i(z, t) = \epsilon_i(z, t) \cos[\omega_i t - k_i z + \varphi_i(z, t)] \quad (i = \lambda, \nu), \quad (1)$$

where the electric fields E_λ and E_ν induce the transitions between the energy levels 1-2 and 2-3 in a three-level system consisting of the ground state 1 and the two excited states 2 and 3, respectively. The frequencies ω_λ and ω_ν are assumed to be far off resonance from the eigenfre-

quencies Ω_λ and Ω_ν for each transition, although the sum of these frequencies $\omega_\lambda + \omega_\nu$ is equal to $\Omega_\lambda + \Omega_\nu$; i.e., resonant to the transition 1-3. Also, $\epsilon_i(z, t)$ and $\varphi_i(z, t)$ are considered slowly varying. A set of coupled equations of the time-dependent coefficients in the expansion of the wave function is obtained from the Schrödinger equation for the system. The solutions give the macroscopic induced polarizations, which act as sources in the self-consistent forms for the electric fields in accordance with Maxwell's equations. We can obtain from the reduced Maxwell's equations the area equations of different-frequency, two-photon propagation given by

$$\frac{d}{dz} \int_{-\infty}^{\infty} \epsilon_i^2 dt = -\frac{\beta_i}{k} [1 - \cos(k \int_{-\infty}^{\infty} \epsilon_\lambda \epsilon_\nu dt)], \quad (2)$$

where $\beta_i = 4\pi\Omega_i N_0 \mu_\lambda \mu_\nu / cn\hbar\Delta\omega$, $k = \mu_\lambda \mu_\nu / 2\hbar^2 \Delta\omega$, n is the refractive index of the medium, c is the light velocity in vacuum, $\Delta\omega = |\Omega_i - \omega_i|$ is the frequency deviation from the intermediate (virtual) state 2, μ_i is the matrix element for the electric dipole moment, and N_0 is the atomic density. Here it was assumed that the spectrum distribution function of the medium is uniform and the frequency shift from $\varphi_i(z, t)$ is negligible. The condition $T_2^* \ll \tau \ll T_2'$ is assumed for the actual pulse width τ . It is seen that Eq. (2) covers the single equation derived by Belenov and Poluektov⁶

for the special case of $\omega_\lambda = \omega_\nu$.

These equations indicate that if we define the area as

$$A = k \int_{-\infty}^{\infty} \epsilon_\lambda \epsilon_\nu dt, \quad (3)$$

a lossless propagation resulting in SIT exists for the condition $A = 2m\pi$. Accordingly, $A = 2\pi$ gives the threshold condition of different-frequency, two-photon SIT.

Equation (2) also represents an ordinary two-photon absorption law with the limit of small pulse area. It is noteworthy that the two-photon absorption length decreases with the rise of the input pulse area depending upon each intensity. On the moving frame, a Lorentzian shape solution is obtained. Then the pulse velocity is modified in the following manner:

$$V = \frac{c}{n + 2c\tau(\beta_\lambda\beta_\nu)^{1/2}}. \quad (4)$$

Computer solutions of ϵ_λ^2 and ϵ_ν^2 for the coherent two-photon propagation are illustrated in Fig. 1. The properties of the peak amplification and multiple breakup of both the pulses for the present case are analogous to those of one-photon SIT.⁵ It should be remarked that when varying the intensity ratio of the two input pulses keeping

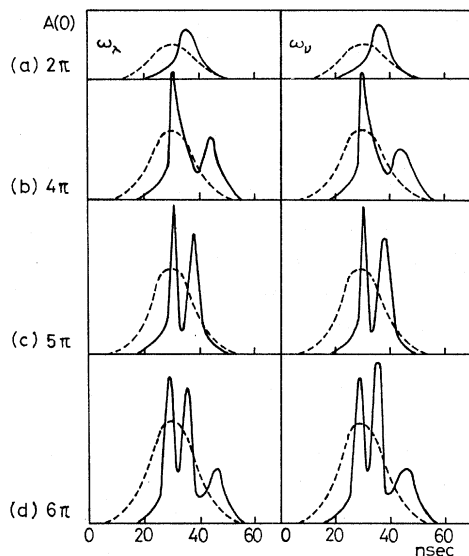


FIG. 1. Solid curves, computer-generated output pulses demonstrating the two-photon SIT of different-frequency optical beams ω_λ and ω_ν . Dashed curves, Gaussian input pulses with the pulse widths of $\tau = 20$ nsec at $z = 0$, respectively. The equal intensity $\epsilon_\lambda^2(0) = \epsilon_\nu^2(0)$ was assumed. The values of μ_i , N_0 , and $\Delta\omega$ as well as the propagation distance used in the calculation correspond to the experimental condition described in the text.

the area constant, we find a curious modulation effect with the low-intensity input pulse showing marked breakup and sharp peak amplification, in contrast to the high-intensity input pulse having a smooth variation with distance and time. This is understood by noting that the high-intensity pulse produces a deep modulation for the low-intensity pulse through the two-photon coupling.

In the experiments, we employed potassium vapor as a two-photon propagating medium in which $4S_{1/2}$, $4P_{3/2}$, and $6S_{1/2}$ levels (these corresponding to the levels 1, 2, and 3 in the analysis, respectively) comprise an inhomogeneously broadened three-level system. The experimental arrangement was similar to the previous works⁸ involved in four-wave parametric interactions in this vapor. The two coherent beams ω_ν and ω_λ , one at the Q-switched ruby-laser frequency and the other at ruby-generated stimulated Stokes frequency from nitrobenzene, are incident simultaneously upon potassium vapor. A sapphire etalon with a 0.5-mm spacer as the output mirror was used with a Q-switch solution to preserve a stabilized single transverse and longitudinal mode operation⁹ for both oscillations. The frequency tuning was performed by temperature control of the ruby-laser rod. For the double-quantum degenerate transition¹⁰ $4S_{1/2}$ - $6S_{1/2}$ in potassium characterized by $\mu_\lambda = 7.53 \times 10^{-18}$ esu cm, $\mu_\nu = 5.33 \times 10^{-18}$ esu cm, and $\Delta\omega = 10 \text{ cm}^{-1}$, a 2π square pulse with a width of $\tau = 20$ nsec at the threshold corresponds to a power density of 1.9 kW/cm². We used a maximum peak intensity of 0.1 MW/cm² for the ruby laser and stimulated Raman emission. Both the beams were made to be approximately uniform over a cross section of 3 mm diam through the vapor cell. The vapor cell is made of a 140-cm-long Pyrex tube and heated to $(150 \pm 0.5)^\circ\text{C}$ in an oven. The output radiation (through a 0.5-mm-diam aperture placed after the cell in order to isolate a nearly uniform plane-wave portion of Gaussian profile beam) was detected by a photomultiplier and was simultaneously displayed with the input pulse on an oscilloscope (with a rise time of 2 nsec) by the use of a matched delay circuit.

Since the inhomogeneous broadening time due to Doppler effect is ~ 0.2 nsec at 150°C and the natural lifetime of an upper state is ~ 70 nsec, the relation $T_2^* \ll \tau < T_2'$ held well in our experiment. The atomic density is about $10^{13}/\text{cm}^3$ at 150°C . We remark here that when the input power of both beams is increased to 10 MW/cm², intense violet emission corresponding to the $5P-4S$

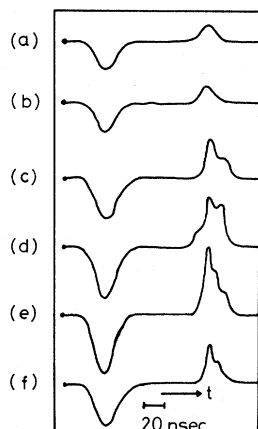


FIG. 2. Oscilloscope traces of the input ruby-laser pulse (first) and the output pulse (second) at the ruby-laser and stimulated Raman frequencies. In *a* and *b*, the second pulses show the ruby and Stokes output without potassium vapor, respectively. *c*–*e* represent ruby output pulses at different input intensities with the vapor present, while *f* shows the Stokes output pulse.

transition resulting from the resonant two-photon excitation was observed and its intensity showed the super-radiant behavior¹¹ depending on the square of the atomic density.¹²

The observed shapes of the incident and transmitted optical pulses at the ruby laser and stimulated Raman emission frequencies are shown in Fig. 2. The first pulse corresponds to the input ruby frequency and the second pulse to the output from the potassium cell for comparison. It was found experimentally that the laser pulse with high input energy yields breakup into two or more pulses corresponding to $2m\pi$ pulses and also peak amplification. The Stokes input pulse also exhibited a similar behavior as shown in Fig. 2(f). It is worthy to note that a time delay of several nanoseconds for the output pulses is also observed as the input energy is decreased. The observed delay time was found to be explained by the pulse velocity $V = c/4$ estimated from Eq. (5). These experimental results seem to be coincident with the computer solutions in Fig. 1 considering experimental smearing such as the time response, the uniformity of transverse profile of both beams, the fluctuation of laser oscillations, diffraction losses, and the level degeneracy.

In Fig. 3, the energy transmission of the ruby-laser pulse is plotted against the input energy for the two different pulse durations, $\tau = 15$ and 19 nsec. The dependence of observed transmission on the pulse duration could be explained by the fact that a narrower ruby laser pulse usually

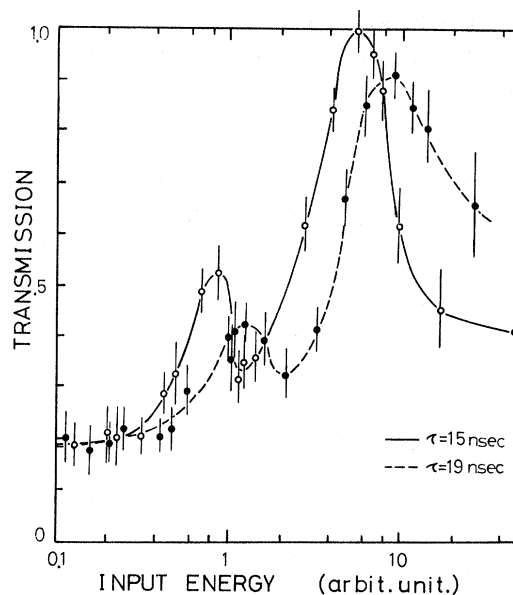


FIG. 3. Energy transmission ratio versus input energy for two different pulse widths at ruby-laser frequency.

generated higher a Stokes pulse due to the nonlinear process.¹³ The first small peak in the figure seems, from the observed pulse shapes, to correspond to 2π pulse area and the second to 4π pulse area, although the periodicity is not well defined because of experimental averaging, natural lifetime, deviation from the resonance condition, and so on. Unexpectedly, the third peak of 6π pulse area does not appear in the plots of transmission. In addition to this, the dips in transmission are very deep. These behaviors may be understood from a deviation from SIT due to the unfavorable energy transfer to a single-photon transition between $6S_{1/2}$ and $5P_{3/2}$,¹² besides coherent nonlinear processes⁸ in the high-input-power region. Rigorously analyzing the anomalous transmission is left as a future problem.

In conclusion, the data presented in this Letter are believed to be compatible with the concept of resonant two-photon SIT, and potassium vapor offers the first evidence of this phenomenon in the inhomogeneously broadened three-level system. Two-photon SIT should give a useful technique for controlling pulse compression at one frequency by the increase and decrease of another input-pulse intensity. Further studies of two-photon pulse compression and also double-resonance SIT should be a valuable future contribution to the field of pulsed, multiphoton, coherent,

nonlinear interactions with matter.

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Particle Loss in the Levitated Spherator FM-1*

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The plasma confinement times were studied in the levitated spherator FM-1 as a function of the electron density and electron temperature. It was found that below approximately 1 eV, the plasma confinement time is proportional to T_e^m/n_e ($\frac{1}{2} \leq m \leq \frac{3}{4}$) with an absolute confinement time 4-6 times below the classical confinement time. Above 1 eV the confinement time decreases as T_e^{-1} with an absolute confinement of 300 times the so-called Bohm diffusion time.

Recent experiments¹⁻⁴ suggest that over a limited range of plasma parameters the plasma confinement time increases with increasing electron temperature and decreases with increasing density. This result is consistent with the scaling of classical diffusion, but the absolute value may differ by a factor of 1-10 (pseudoclassical diffusion).⁵ In the LSP experiments,¹ the maximum plasma confinement time was about 200 msec, which was 260 times the so-called Bohm time.

The important question both from the basic plasma-physics point of view and for fusion application is whether there is any deviation from this scaling law as the electron temperature increases. Experiments to answer this question are reported here.

The experiments were carried out in the second-generation levitated spherator called FM-1.⁶ The previous levitated ring experiments performed in the device called LSP have been reported elsewhere¹ and a detailed report is now in prepara-

tion. The spherator has a single levitated current-carrying ring which produces the poloidal magnetic field, and with the aid of external conductors provides an azimuthally symmetric, toroidal confinement configuration with strong magnetic shear.

Table I lists the parameters at which LSP was operated along with the parameters at which FM-1 is presently operated. The maximum design ring current (I_p) for FM-1 is 375 kAt; however, the data presented here were obtained at 150 kAt. FM-1 has not as yet been operated with Ohmic heating, a fact which the lower density and electron temperature reflect.

Electron-cyclotron resonant heating is used to form the plasma in FM-1. Nonresonant microwave heating (above resonant frequency) is used to control the electron temperature in the afterglow. The nonresonant microwaves produce resistive heating of very low efficiency. The $T_e^{-3/2}$ dependence of the heating efficiency due to the