# Two-Photon Laser

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We have demonstrated a pulsed two-photon laser at the transition $3d\,^2D-2s\,^2S$ of atomic Li (frequency $\omega_{31}$) by (i) generation of a coherent light beam at $\omega_x$, stimulated by light pulses at $\omega_1=\omega_{31}-\omega_x$, and (ii) single-pass amplification of injected light at $\omega_3=\omega_{31}/2$.

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Atomic two-photon transitions[1] have been considered for the operation of optical high-power amplifiers[2,3] since the gain depends on the light intensity: No small-signal gain makes the excited levels prematurely deplete. Theoretical models of two-photon amplifiers have been worked out.[4,5]

Two-photon stimulated emission may alternatively include two quanta of unequal energy (nondegenerate case). Light injection on only one of the corresponding two frequencies is required for stimulation of the two-photon transition. An oscillator based on this principle has potential as a novel tunable coherent light source. Nondegenerate stimulated two-photon emission has been observed in the past from potassium vapor simultaneously irradiated by ruby-laser light and Stokes light generated in various organic liquids.[6] In that experiment, output and pump frequencies were tuned by changing the ruby temperature, which left the tuning range limited to $\sim 10\ \mathrm{cm}^{-1}$. Recently, a related incoherent source was demonstrated, where one of the quanta was stimulated by radiation from a high-power Nd-doped yttrium aluminum garnet laser whereas the second quantum in the two-photon emission was spontaneously emitted ("enhanced" spontaneous emission).[7] In this kind of source, inversion does not exist. On the other hand, population inversion on a resonance line has been recently accomplished by the application of a two-photon optical pumping scheme.[8]

We have observed, on the $2s\,^2S\rightarrow 3d\,^2D$ transition of atomic lithium vapor, two types of coherent two-photon emission: (ii) *nondegenerate* two-photon generation of coherent light at frequency $\omega_x$, stimulated by injected light pulses of frequency $\omega_1\neq\omega_x$, where $\omega_1+\omega_x=\omega_{31}=\omega(2s\text{-}3d)$, and (ii) *degenerate* single-pass amplification of injected light at frequency $\omega_3=\omega_{31}/2$.

Transient population inversion of the *S-D* resonant two-photon transition was achieved upon two-photon excitation close to or on resonance with the $4f\,^2F$ level. The large $3d$ population is caused by a resonant quadrupole Raman-Stokes process,[9] although some collisionally induced redistribution in the $4f$ level and subsequent fast fluorescent decay may contribute. The $3d$ population faces two possible decay paths: by Dicke superfluorescence[10] into the $2p$ level, and by nondegenerate two-photon emission at $\omega_x$ and $\omega_1$, stimulated by the pump pulse at $\omega_1$. Upon injection of a third light pulse of suitable frequency $\omega_3=\frac{1}{2}\omega(2s\text{-}3d)$, still another deexcitation is via degenerate two-photon emission, and two-photon gain occurs. (See Fig. 1.)

The lithium vapor is contained in an $Al_2O_3$ heat pipe and simultaneously irradiated by two 4-nsec-long light pulses from $N_2$-laser–pumped dye lasers at $\lambda_1=667.4$ nm (DCM) and at $\lambda_2=461.8$ nm (Coumarine 1). A third dye laser generates a probe light pulse at $\lambda_3=639.1$ nm. Laser emission at $\omega_x$, or the amplified probe pulses at $\omega_3$, are recorded by a monochromator, photomultiplier, boxcar integrator, and *XY* recorder.

We observe, at $\omega_x$, single-pass laser pulses

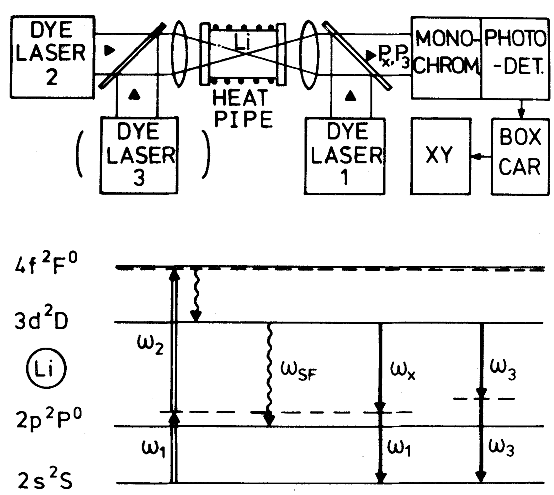


FIG. 1. Top: Experimental setup. Bottom: Li energy-level scheme, including excitation of $3d\,^2D$, and three modes of deexcitation: superfluorescence, and nondegenerate and degenerate stimulated two-photon emission.

of 30 W maximum power. They show minute fluctuations only, in contrast with superfluorescence at 610.3 nm from the competing decay $3d \rightarrow 2p$. The latter is eliminated by a suitable choice of peak power values $P_1$ and $P_2$ (high and low, respectively) and of the Li density.

The output peak power at $\omega_x$ is shown in Fig. 2 versus stepwise tuning of $\omega_1$. Here, $\omega_2$ was accordingly tuned in order to preserve quasiresonance of the excitation with the $4f$ level. The central minimum is due to strong absorption of the pump beams at resonance with the intermediate $2p$ level. Laser emission at $\omega_x$ is still observable with detuning, from this level, as far as 500 $cm^{-1}$. Above 7 $cm^{-1}$, the tuning function of the output power agrees quite well with a $(\Delta\lambda_1)^{-4}$ dependence: The pumping rate and the stimulation by $P_1$ both contribute a factor $(\Delta\lambda_1)^{-2}$ due to the Lorentzian wings of the transition line shape.

We have assured that the coherent light emitted at $\omega_x$ (or $\omega_3$) had no correlations of phase or polarization with pump light $P_2$. In addition, the pump geometry does not provide phase matching. Thus, parametric $n$-wave processes are excluded as an origin of the emission.

Above generation threshold, the output power $P_x$ depends linearly on the power of the second pump step, $P_2$. The dependence on the first pump light just above threshold is, in contrast, approximately $P_1^2$, as suggested by $P_1$ acting both as a pump and as stimulating light. At higher $P_1$ values the output levels off because of saturation.

Additional light pulses at the probe frequency $\omega_3$ ($\lambda_3 = 639.15$ nm), which are injected into the optically pumped lithium vapor, become amplified. In Fig. 3 (top), the output $P_3(z)$ is plotted versus the input $P_3(0)$ under various pumping conditions. The data points of series 1 represent attenuation by two-photon absorption, when no pump light is present. Series 2 shows a reduction of this absorption, when only $P_1$ is on and some population appears in the $3d$ level from multiphoton excitation of high lying and ionized states with subsequent cascading. With *both* $P_1$ and $P_2$ on (series 3), the $3d$ level shows transient

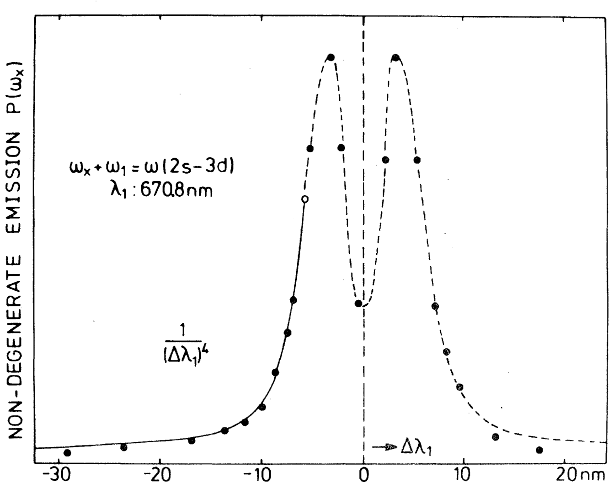


FIG. 2. Nondegenerate two-photon laser emission at $\omega_x$ vs detuning of pump laser 1 whose light simultaneously stimulates emission at $\omega_1 = \omega(2s\text{-}3d) - \omega_x$.

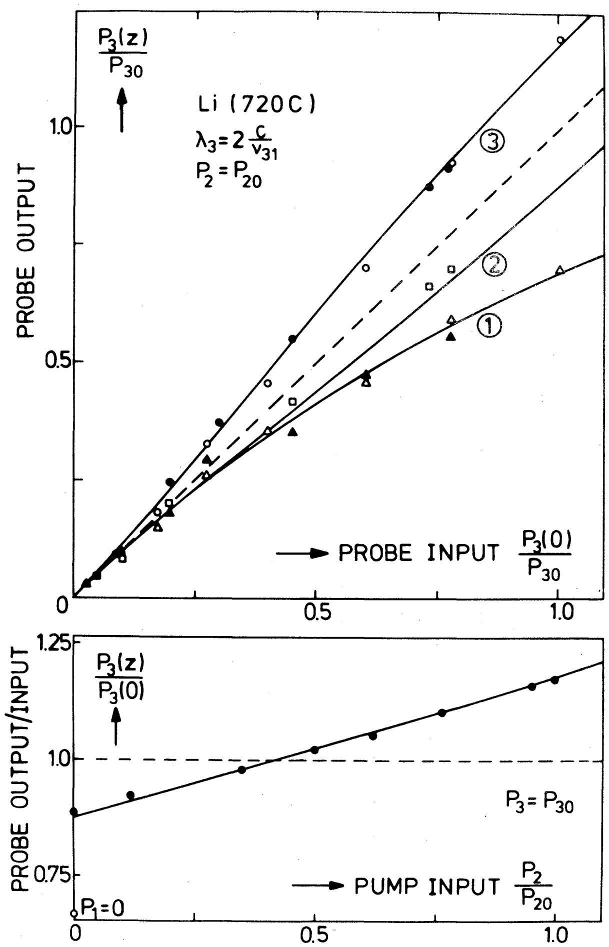


FIG. 3. Top: Output power $P_3(z)$ vs input power $P_3(0)$ of injected probe light pulses at $\omega_3$. Series 1, no pump light; two-photon absorption. Series 2, pump light pulse 1 only; reduced two-photon absorption. Series 3, both pump light pulses 1 and 2 present; two-photon gain. Solid and open data points are independent sets of measurements. Full lines give calculated output power, including saturation for curves 2 and 3. Bottom: Probe output-input ratio vs power $P_2$ of pump light 2 shows buildup of two-photon probe gain.

inversion against the $2s$ ground state, and two-photon gain at $\omega_3$ shows up. This gain increases with the probe light intensity, since for degenerate two-photon emission

$$dI_3/dz = B_2 \Delta N I_3^2 \qquad (1)$$

holds, where $\Delta N$ is the inversion density, and $B_2 I_3^2$ is the two-photon transition rate per atom. At the maximum achievable peak power of the injection pulse, $P_{30} = 3$ kW, the gain in the active zone approximately 2 cm long is $dI_3/I_3 \simeq 20\%$. The full lines represent fits based on the two-photon analog of the Lambert-Beer law,

$$I(z)/I(0) = [1 \pm I(0)/\hat{I}]^{-1}, \qquad (2)$$

where $\hat{I}^{-1} = \Delta N \sigma z$, and $\sigma$ is the emission cross section. However, saturation has been included in the calculation of curves 2 and 3. In contrast, curve 1 is a direct fit of Eq. (2), since saturation is overcome by powerful $3d$-$2p$ superfluorescence following two-photon absorption. This superfluorescence is efficiently quenched upon $P_1$ pumping which generates appreciable $2p$ population.

The bottom portion of Fig. 3 gives the dependence of the probe gain on the power of the second pump step, $P_2$. It displays the continuous transition from two-photon absorption to transparency to gain.

In brief, we have demonstrated nondegenerate two-photon light generation and degenerate two-photon amplification in Li vapor. After the (single-photon) laser and maser, the parametric oscillator, and the Raman sources, the two-photon laser is the fourth principally different quantum-optical device. If electric excitation becomes feasible, this novel type of laser will turn out to be particularly useful as a tunable source and as a high-power optical amplifier.

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# Energy-Transport, Group, and Signal Velocities near Resonance in Spatially Dispersive Media

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Theoretical results are reported for energy propagation in bounded spatially dispersive (exciton-polariton) media. A remarkable slowing down of the energy-transport velocity $V_E$ is predicted near resonance. For GaAs the decrement is by a factor of $10^4$, with $V_E \sim 4\times10^3$ m/sec at resonance. Some comparison is made between energy-transport, group, and signal velocities, and available data.

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In this Letter we report new theoretical results for the dispersion of the velocity of energy transport, $V_E$, in a nonlocal or spatially dispersive exciton-polariton medium. Loudon[1] studied energy propagation in an absorbing local dielectric; Bishop and Maradudin[2] extended the analysis to include spatial dispersion, and we follow the latter authors. We find a decrement by a factor of $10^4$ in $V_E$ at resonance.

We consider a simplified, idealized situation. The surface of a semi-infinite isotropic spatially dispersive medium lies in the $x$-$y$ plane, with the medium occupying the half-space $z \geq 0$. A steady-state monochromatic plane electromag-