Two-Photon Adiabatic Inversion

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This Letter reports the first experimental observation of population inversion of a two-photon transition in NH_3 using the adiabatic rapid-passage technique. The population inversion was demonstrated by the observation of stimulated two-photon gain and emission. By use of this technique, the two-photon population relaxation time T_1 was measured.

This Letter reports the first observation of population inversion of a two-photon transition by the technique of adiabatic rapid passage (ARP). The possibility of two-photon adiabatic inversion was initially pointed out by Grischkowsky and Loy[1] using a two-photon vector model.[2] In terms of this model, this process is seen to be the two-photon analog of adiabatic inversion in one-photon coherent optics and in magnetic resonances.[3] A number of two-photon coherent effects have been reported previously and dephasing time T_2 has been measured.[4-12] The experiment reported here is the first demonstration of two-photon population inversion, the signature of this inversion being the observation of two-photon gain and emission. Monitoring the decay of this two-photon emission allowed one to make the first measurement of the population lifetime T_1 in a two-photon transition in NH_3.

Consider a three-level system with excited state 1, intermediate state n, and ground state 2. The conditions for adiabatic inversion from 2 to 1 under two-photon excitation are as follows: (1) Either the two-photon transition frequency or the sum of the two input laser frequencies must be swept through resonance in a time short compared to T_1, and (2) the rate of the frequency sweep must be adiabatic, i.e.,

$$d\gamma_3/dt<(\kappa E_1 E_2)^2. \qquad (1)$$

In this expression γ_3 is the instantaneous difference between the sum of the laser frequencies and the two-photon transition frequency (including the optical Stark shifts); E_1, E_2 are the amplitudes of the laser fields; $\kappa = p_{1n}p_{n2}/2\hbar^2\Delta$ where p_{1n}, p_{n2} are, respectively, the dipole transition moments between the states 1 and 2 and the intermediate state n; and Δ is the offset between the laser frequency ω_1 and the transition frequency from 1 to n. Grischkowsky and Loy[1] suggested that by using the optical Stark effect to sweep through the two-photon transition while satisfying the adiabatic condition, one might obtain self-induced adiabatic rapid passage. Here, instead of the self-induced approach which depends critically on the laser pulse shape, the Stark-switching technique[13] using an external electric field was used. It was clearly shown in the two-photon free-precession experiments[8,9] that the Stark-switching technique, being electronically controlled, yielded very stable signals that allowed one to obtain accurate relaxation information.

The NH_3 two-photon transition, $(\nu_2, J, K, M) = (0^-, 5, 4, \pm 5) \rightarrow (2^-, 5, 4, \pm 5)$, has been previously used in the two-photon free-precession experiment.[9] This transition is 294 MHz away from the sum of the 10-μm CO_2 $P18$ and $P34$ lines. Here, the beam at $P18$ line was from a pulsed low-pressure CO_2 laser with intensity of 3 kW/cm^2 and duration of 150 μsec, and can be considered cw in this experiment. The other beam, at $P34$ line, was from a single-longitudinal-mode CO_2 TEA (transversely excited atmosphere) laser with peak intensity of 2.5 MW/cm^2 and duration of 1 μsec. These counter-propagating beams interested in a 40-cm-long Stark cell with 3.175-mm precision spacers. The polarizations of both lasers were parallel to the Stark field. The optical Stark effect, due to the strong beam at $P34$ line on the low-frequency side of the 2 to n transition, caused the two-photon transition to shift farther away from resonance. Application of an external Stark field, however, shifted the two-photon transition the opposite way, and could sweep through resonance. The Stark pulse duration was typically 200–300 nsec. The TEA laser pulse was monitored by a photon-drag detector. The two-photon emission signal was observed on the weak beam after it passed through the Stark cell using a Ge:Cu (4.2 K) photoconductor and a Tektronix 7904 oscilloscope. The gas pressure, in the range 0–200 mTorr, was monitored by a capacitive manometer.

At low laser intensities (about 100 kW/cm^2 for the strong and weak beams, respectively) when the adiabatic condition (1) was not satisfied, application of the Stark pulse swept through the two-photon resonance in a nonadiabatic manner. As a result, free-precession signals appeared at the rising and trailing edges of the Stark pulse as observed in the previous free-precession experiment.[9] As the laser intensities increased, the excitation became more adiabatic and the free-precession signal decreased, since the two-photon polarization vector could no longer precess freely. There is an additional reason for the decrease of the precession signal at high intensities. The strong beam intensity was not uniform inside the Stark cell, being zero at the Stark plates and maximum at the midpoint between them. This field inhomogeneity, via the optical Stark effect, introduced an inhomogeneous broadening directly proportional to the strong beam intensity, and at high intensity substantially reduced the precession signal.

At sufficiently high laser intensities (about 2 MW/cm^2 and 3 kW/cm^2) the adiabatic condition was satisfied and two-photon inversion occurred. Figure 1 shows typical traces of the signal on the weak beam for two NH_3 pressures. The Stark

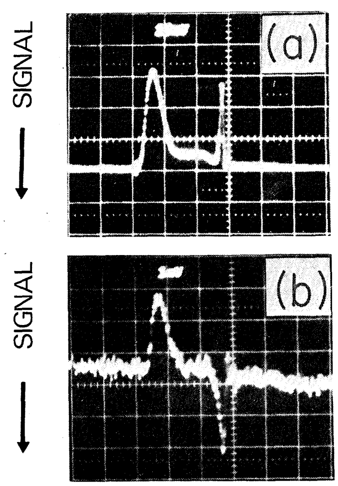

FIG. 1. Two-photon absorption signals observed on the weak beam after propagating through the Stark cell. The different absorption widths were due to the different rise and fall times of the Stark pulse. (a) The NH_3 pressure was 100 mTorr. In this case T_1 was shorter than the Stark pulse width of 210 ns and reduced absorption was observed when the system was probed at the trailing edge of the Stark pulse. Vertical scale was 10 mV/div and horizontal scale was 100 nsec/div. Detector signal was negative, with the full dc signal of the weak beam at -2.5 V. (b) Experimental conditions were the same as in (a) except the gas pressure was at 9 mTorr, when T_1 was long compared to the Stark pulse width. Two-photon emission was clearly observed at the trailing edge of the Stark pulse. Vertical scale was 1 mV/div and horizontal scale 100 nsec/div.

pulse for both cases was about 210 nsec in duration with field strength of 7650 V/cm. The shape of the Stark pulse was asymmetrical so that it swept through resonance more slowly at the rising edge than at the trailing edge. Calculation showed that the excitation was adiabatic on the rising edge, but nonadiabatic on the trailing edge. In Fig. 1(a), the gas pressure was 100 mTorr corresponding to a T_1 shorter than the Stark pulse width. One sees that while the absorption signal at the trailing edge was smaller than that at the rising edge, there was no two-photon emission signal, since the inverted population had substantially decayed during the Stark pulse. To observe emission, one must probe the system at a time after inversion short compared to T_1. This was achieved by performing the experiment with the same Stark pulse and laser intensities but at lower NH_3 pressure when the relaxation time T_1 is much longer. The result is shown in Fig. 1(b), where the NH_3 pressure was 9 mTorr. Here, in contrast to the trace in Fig. 1(a), when the system was probed at the trailing edge of the Stark pulse a two-photon emission signal was clearly seen, demonstrating the population inversion of this two-photon transition in NH_3. Further, this coherent emission signal on the weak beam occurred only in the presence of the strong beam, showing that this is definitely a stimulated two-photon emission and not one-photon emission to the intermediate state.

The decay of the emission signal at the trailing edge of the Stark pulse is a direct measure of the relaxation of the population inversion. For example, the decay of this signal as the Stark pulse width increased would yield T_1. However, this method cannot be used in the present experimental setup since the maximum Stark pulse width cannot exceed 1 μsec, the duration of the TEA laser pulse. Instead, the Stark pulse width was kept constant, and the two-photon emission or absorption signal $S(p)$ at the trailing edge of the Stark pulse was measured as a function of gas pressure p. In Fig. 2, $S(p)-S(\infty)$ is plotted versus p, where $S(\infty)$ is the absorption signal at high pressure where the system has completely recovered from the adiabatic inversion at the time of probing. These data indicated that two decay constants are needed to describe the observed population relaxation. This can be understood by examining the relaxation mechanism, which in this case is due to collision-induced transitions to and from neighboring rotational states within the same vibrational band. In the $\nu_2=0$ ground vibrational

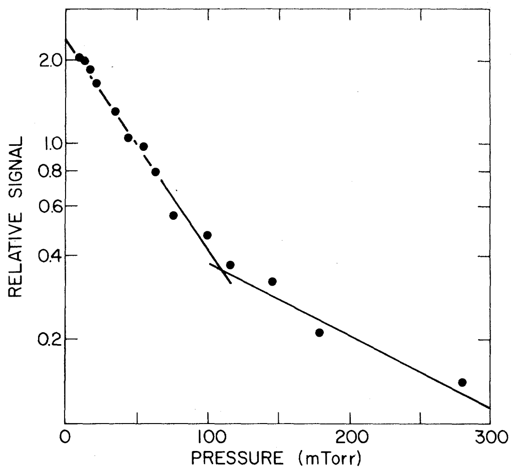

FIG. 2. Two-photon inversion signal, at fixed Stark pulse width, plotted as a function of NH_3 pressure p. The signal plotted was $S(p)-S(\infty)$ where $S(p)$ was the emission or absorption signal at pressure p and $S(\infty)$ was the absorption signal at high pressure when the system has completely recovered from the initial inversion.

band of NH_3, the dominant transition is that between the inversion doublets of the same J, K, and M, the transition rate being very high because of the small doublet spacing of 0.8 cm^{-1}. This doublet spacing increases substantially for the excited ν_2 bands (280 cm^{-1} for the $\nu_2=2$ band), and the collision-induced transition rate is expected to be much slower.[14] To model this relaxation, let the population relaxation times for the $\nu_2=0$ and 2 bands be T_{1l} and T_{1u}. Since both are inversely proportional to the gas pressure p, they can be written as $T_{1l}=k_l/p$ and $T_{1u}=k_u/p$. Assume that the system is inverted initially. The excited-state population will decay in time t as $\exp(-pt/k_u)$ while the ground state will be repopulated as $1-\exp(-pt/k_l)$. The relative signal plotted in Fig. 2 is then given by $S(p)-S(\infty) = A[\exp(-pt/k_u)+\exp(-pt/k_l)]$ where A is a proportionality constant. From the data in Fig. 2, with $t=210$ nsec, one can identify $k_l=11.5\pm1$ nsec Torr and $k_u=35.5\pm3$ nsec Torr for the $\nu_2=0$ and $\nu_2=2$ bands. It is instructive to compare the population relaxation times T_{1u} and T_{1l} with the phase memory time T_2 measured by the free-precession technique on this same two-photon transition. The phase memory time $pT_2=10.5\pm2$ nsec Torr is very close to the population relaxation time pT_{1l} for the $\nu_2=0$ band. This is because the phase coherence between 1 and 2 is destroyed whenever there is a collision-induced transition in either state, and thus is essentially determined by the shorter of the two population relaxation times. The much longer upper-state population relaxation time k_u therefore cannot be determined by measurement of T_2, or equivalently the linewidth of the two-photon transition, but is readily determined for the first time in the present experiment.

It has long been recognized that an inverted two-photon transition could lead to a two-photon laser.[15] Because of the various technical difficulties, the two-photon laser has yet to be experimentally realized. In fact, while the related stimulated anti-Stokes emission from an inverted Raman transition has been observed,[16] to my knowledge stimulated emission or gain from a two-photon transition has not yet been reported. The experiment reported here clearly demonstrates two-photon stimulated emission and gain from the inverted two-photon transition in NH_3. Unfortunately the two-photon gain as seen by the weak beam was relatively small, about 0.2% in in 40 cm. This is due to the low gas pressure used in the experiment to avoid excessively fast population relaxation in the $\nu_2=0$ band. The low gas pressure also means low energy storage in the excited medium. Thus the NH_3 system excited by two-photon adiabatic inversion does not appear to be promising for achieving two-photon laser oscillation. Nonetheless, the two-photon adiabatic rapid-passage method should be a useful and clean excitation technique to obtain two-photon inversion in the continuing search for the two-photon laser.

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Ion-Ring Igniter for Inertial Fusion

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This paper outlines a scheme employing magnetically compressed ion rings for the transport of energy to a deuterium-tritium pellet.

Several candidates, e.g., lasers,[1] relativistic electrons beams,[2] intense beams of light ions in the 10-MeV range,[3] and 25–100-GeV heavy-ion beams,[4] have been suggested for the high-powered energy source or "ignitor" required for pellet fusion. In this Letter I present a scheme that employs ion rings for energy compression and transport to the pellet.

The basic idea is to inject a pulse of ions into a magnetic mirror, trap these ions in the form of an ion ring, and magnetically compress the ion ring to increase its energy and reduce its dimensions. The compressed ion ring is accelerated axially to impact a D-T pellet. Figure 1 shows a schematic of the proposed system. A pulse of ions from a magnetically insulated annular ring diode[5] is injected through a cusp-shaped magnetic field. By virtue of the conservation of the canonical angular momentum the ions begin to rotate and give rise to a θ current. Previous experimental work[6] has demonstrated that (i) intense ion beams are charge neutralized by electron flow along field lines, (ii) such electrons are created at nearby boundary surfaces, (iii) charge-neutralized intense ion beams propagate across the field in ballistic cyclotron orbits, and (iv) magnetic neutralization of the circulating ion current by azimuthally drifting electrons does not take place because the radial electric field required for this drift is shorted out by electron flow along field lines. The rotating beam is expected to be trapped between mirrors M_1 and M_2 because of axial momentum loss by dissipation induced in surrounding structures.[7-9]

Magnetic compression of the trapped ring is achieved in two stages. The first stage utilizes superconducting coils that generate a spatially increasing field from, say, 10–20 kG to approximately 100 kG determined by the state of the art. However, pulsed coils will be required to trans-

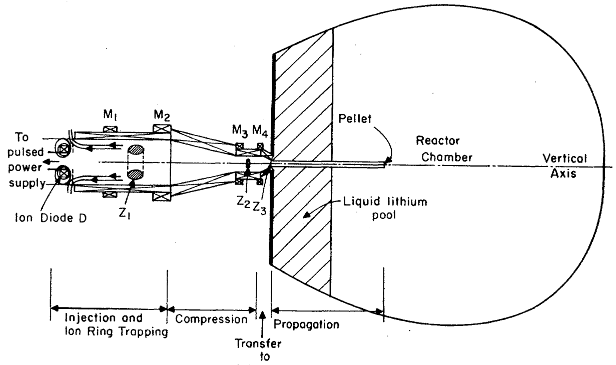
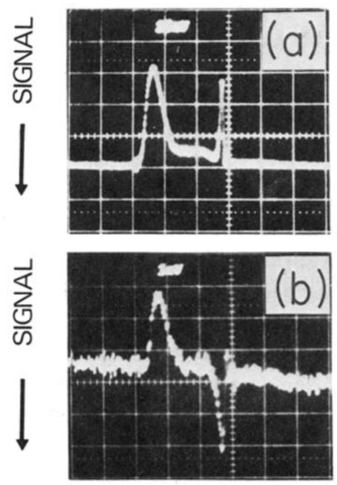


FIG. 1. Schematic of ion-ring–pellet fusion reactor.



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