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Observation of Population Inversion by Optical Adiabatic Rapid Passage

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I report the observation of population inversion by optical adiabatic rapid passage. These observations, on an NH_3 infrared transition with all the relevant parameters known, agree with theoretical expectations. The pressure dependence of T_1, obtained by this technique, makes possible the first estimate of the collision-induced rotational lifetime of NH_3 in an excited band, which is significantly longer than the rotational lifetime in the ground vibrational band.

The adiabatic rapid passage (ARP) technique has been widely used in magnetic resonance to achieve population inversion of spin systems.[1] It has been suggested that this technique can also be used in the optical regime, but the experimental results were inconclusive.[2] In this Letter, I report the first unambiquous observation of population inversion using optical ARP. The relevant physical parameters of the system are known, so that my observations can be compared with theoretical expectations. As in earlier NMR experiments, two passages were performed, with the first one inverting the system and the second monitoring the population difference at a later time. In my experiment, at the second passage, I observed a power emission signal, a clear sign that the system was inverted by the first passage. Monitoring the decay of this signal in time, I obtain T_1 measurements as a function of pressure in NH_3, thereby making possible the first estimate of the collision-induced transition rate between rotational levels in an excited vibrational band of NH_3.

The main requirements for ARP are (1) either the frequency of the two-level system or that of the laser must be swept through the linewidth of the system in a time short compared to T_1, and (2) the laser field must be strong enough to satisfy the adiabatic condition,

$$\kappa\mathcal{E} > (d/dt)|\Delta\omega/\kappa\mathcal{E}|, \quad \kappa \equiv \mu/\hbar,$$

where $\mathcal{E}$ is the laser field strength, μ is the dipole matrix element, and $\Delta\omega$ is the angular frequency offset between the laser and the two-level system. I choose to sweep the resonant frequency of the molecular system via the Stark effect and to leave the laser frequency unchanged. My method is similar to the very successful Stark switching technique of Brewer and Shoemaker,[3] except for the following: Their Stark pulses are switched on and off suddenly (nonadiabatically) for their laser intensity. My laser intensity is higher and the sweep rate is controlled so that the adiabatic condition above is satisfied.

I used the $R6$ line of the CO_2 laser at 10.35 μm, which is 2.98 GHz from the ν_2 band, $s \rightarrow a$ $Q(J=5, K=5)$ transition of NH_3. NH_3 is chosen because of its relatively large dipole matrix element ($\mu_{\nu_2} = 0.24$ D),[4] and because Stark spectroscopy has been performed on its ν_2 band.[5] A dc Stark field shifts the $M = \pm 5$ transitions to within 500 MHz of the laser frequency. All other M states can be neglected since they are more than 1 GHz away. The laser light is linearly polarized parallel to the Stark field, so the only relevant transitions are $(\nu_2, J, K, M) = (0, 5, 5, \pm 5) \rightarrow (1, 5, 5, \pm 5)$ with $\Delta M = 0$. The dipole transition element for this doubly degenerate system is uniquely given by $|KM/J(J+1)|\mu_{\nu_2} = 0.20$ D.

The Stark electrodes are 91.5 cm long and are separated by precision balls 2 mm in diameter. The dc voltage was 2650 V. The amplitude of the Stark pulse was 400 V, corresponding to a frequency shift of ~800 MHz. The sweep started ~500 MHz below the resonant frequency and the

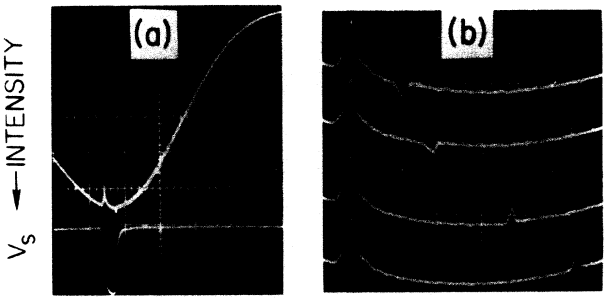

FIG. 1. (a) Dual-beam oscilloscope traces showing simultaneously the Stark pulse V_s (the lower beam) and the laser pulse (the upper beam) after passing through the Stark cell. The time scale is 1 μsec/div. The pressure of NH_3 in the cell was 10.4 mTorr. (b) Relevant part of the laser pulse at the expanded time scale of 0.2 μsec/div. Four separate shots at increasing Stark pulse durations are displayed.

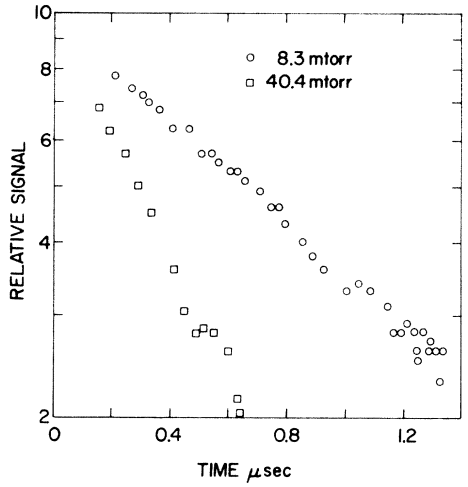

FIG. 2. Decay of the population inversion.

sweep rate was ~4 MHz/nsec. The laser pulse duration was ~5 μsec, with Stark pulse applied near the peak of the pulse. Typically, the intensity in the cell was 300 W/cm^2. The gas pressure was measured by a calibrated capacitive manometer (MKS Baratron, Type 210). The light, after passing through the Stark cell, was detected by a Ge:Cu photoconductor at 4.2°K and the output displayed on an oscilloscope, with an overall time constant of 10 nsec.

In Fig. 1(a), the laser pulse (upper beam) is shown together with the Stark pulse V_s (lower beam), both at 1 μsec/div sweep rate. At the leading edge of the Stark pulse, laser power was absorbed by the system, but at the trailing edge, there was an emission of light. In Fig. 1(b), the relevant part of the laser intensity pulse is shown at an expanded time scale of 0.2 μsec/div. Four shots, taken with increasing Stark pulse widths, show how the emission signal changed smoothly into absorption. For long Stark pulse widths, the strength of this absorption approached that at the leading edge. This agrees well with our theoretical expectations. The system was inverted by ARP at the leading edge, and power was absorbed by the system. Then another ARP was performed by the trailing edge, giving emission or reduced absorption depending on how much the inverted population has decayed. These results were not sensitive to the laser intensity as long as the adiabatic condition was satisfied.

The signal, $S(t)$, at the trailing edge of the Stark pulse is proportional to the population difference between the upper and lower state at t. To display the relaxation of this inversion towards equilibrium, I plot $S(t) - S(\infty)$ versus t, where $t = 0$ is the time of the first passage, and $S(\infty)$ is the absorption signal obtained when the system has returned to equilibrium at $t \gg T_1$. This is shown in Fig. 2 for two different NH_3 pressures. If we assume that the population difference decays exponentially we can obtain a relaxation time T_1 for each pressure. As shown in Fig. 3, the pressure dependence of T_1 can be expressed as $1/T_1 = 1/\tilde{T}_1 + 1/\tau_{IR} = 0.75 + (3.5 \times 10^{-2}$ mTorr$^{-1})p$ μsec for the $s \rightarrow a$ (0, 5, 5, ± 5) $\rightarrow$ (1, 5, 5, ± 5) transition of NH_3. The pressure-independent part $\tilde{T}_1$ is due to the molecular transit time across the laser beam, while the pressure-dependent part τ_{IR} is due to collision-induced transitions to and from neighboring rotational states[6] which are not affected by the pumping laser light. Both the vibrational relaxation time and the ra-

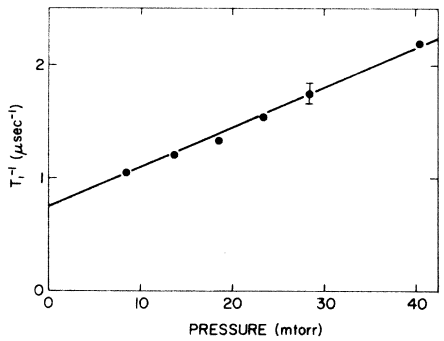

FIG. 3. The inverse of the relaxation time $1/T_1$ versus pressure.

diative lifetime are much longer than the rotational lifetime. Clearly, τ_{IR} depends on both τ_g and τ_e, the rotational lifetimes in the ground and excited vibrational bands.[7] While τ_e is not known, τ_g can be deduced from microwave pressure broadening measurements: $\tau_g = \frac{1}{2}\pi\Delta\nu$, where $\Delta\nu$ is the half width at half-maximum of the pressure broadened line.[8] For the $(J, K) = (5, 5)$ transition in the ground vibrational band, $\Delta\nu/p = 28$ kHz/mTorr or $\tau_g p = 5.68$ μsec mTorr. By comparison, the infrared (IR) value obtained in this experiment is $\tau_{IR} p = 28.5$ μsec mTorr, and $\tau_{IR} \approx 5\tau_g$.

This difference between τ_g and τ_{IR} implies that τ_e must be much longer than τ_g in NH_3. This is not surprising in view of the difference in inversion frequencies in the two bands and its effect on the collision-induced transition probability. In NH_3-NH_3 collisions, the dominant interaction is of the dipole-dipole type,[9] which has the form

$$V = [\vec{\mu}_1 \cdot \vec{\mu}_2 - 3(\vec{\mu}_1 \cdot \vec{r})(\vec{\mu}_2 \cdot \vec{r})/r^2]r^{-3},$$

where $\vec{\mu}_1$ and $\vec{\mu}_2$ are the interacting dipoles and r is the distance between them (see page 359 of Ref. 8). If one of the dipoles reverses direction during the collision, V will change sign, and the transition probability, which is proportional to the time-integrated V, will be reduced. For a collision between ground vibrational band NH_3 molecules, even though the dipole moments reverse direction every 20 psec, they can be considered static on the time scale of the collision duration, typically 0.1 to 1 psec. The resultant large transition probability gives a short τ_g. This is not true for a collision between an excited band molecule and a ground band molecule. (Since only 0.26% of the molecules are in the excited band in this experiment, collisions among excited molecules are rare.) The excited molecule, having a higher inversion frequency, reverses direction every 0.5 psec, which is of the same order as the collision duration. The time-integrated interaction with the ground molecule, which can be considered static, is therefore reduced. This is the main reason for $\tau_e > \tau_g$ in this experiment. Two other factors, of lesser importance, also contribute to a longer τ_e: (1) In this system, the collision-induced transitions are mainly between the inversion doublets. The larger inversion energy separation in the excited band makes transition less likely than in the ground band. (2) The static dipole moment in the excited band ($\mu_e = 1.25$ D) is smaller than that of ground band ($\mu_g = 1.47$ D).

As a further check, I measured the relaxation time τ_{IR} of the same transition in NH_3 due to collision with the nonpolar gas, N_2. Since N_2 has no permanent dipole moment, the N_2-NH_3 interaction should not be affected by the difference in inversion frequencies in the different bands of NH_3. Thus, even though we expect $\tau_e[NH_3\text{-}N_2]$ to be somewhat longer than $\tau_g[NH_3\text{-}N_2]$ because of the smaller moment and larger energy separation in the excited band, the difference between them should be much less than that in the pure NH_3 case. We therefore predict that τ_{IR} should be larger than τ_g, but only by a small factor. To my satisfaction, I obtained $\tau_{IR} p = 56.8$ μsec mTorr, about 1.4 times the ground-state value from microwave measurements, $\tau_g p = 41.8$ μsec mTorr.[10]

Recently, the IR pressure-broadened linewidth of NH_3 was measured by Lamb-dip spectroscopy.[11] The IR line-broadening parameter was found to be quite close to the microwave value for the ground vibrational band. The phase memory time τ_{IR_2} thus obtained is about $\frac{1}{5}$ of τ_{IR_1} measured in this experiment. This probably is due to the large energy separation between interacting states in the excited band, so that not all the collisions that change the phase of the molecules can induce transitions at the same time.

In closing, I note some advantages of ARP for population inversion. Since the inversion is not sensitive to pumping light intensity variation, population inversion can be obtained over a significant portion of the normally bell-shaped intensity profile of the pump beam. Also, complete inversion can be obtained over an entire inhomogeneously broadened line. Finally, at the completion of the passage, the pumping light is no longer on resonance and will not complicate the free decay of the inverted system. Besides T_1 measurements, this technique should be useful for other experiments requiring clean, inverted systems with known initial conditions.

I would like to thank N. S. Shiren for bringing my attention to this problem, and J. A. Armstrong, D. R. Grischkowsky, N. A. Kurnit, and J. J. Wynne for stimulating discussions. The skillful technical assistance of P. A. Roland is gratefully acknowledged.

[1]See, for example, A. Abragam, *The Principles of Nuclear Magnetism* (Oxford Univ. Press, Oxford, England, 1961).

[2]E. B. Treacy, Phys. Lett. 27A, 421 (1968); E. B. Treacy and A. J. DeMaria, Phys. Lett. 29A, 369 (1969).

The main question regarding this work is whether the frequency of their pump light swept *through* a transition of the system—a necessary requirement for adiabatic inversion. In NH_3, where the vibrational-rotational spectrum is known [see F. Shimizu, J. Chem. Phys. 52, 3572 (1970)], we found that this condition could not be satisfied under their experimental conditions. In the 10.3-μm region where their experiment was performed, there are numerous CO_2 lines ($R2$ through $R12$) and NH_3 transitions [$sQ(J,K)$ where $JK=1,2,\ldots,12$]. Neither the laser line nor the NH_3 transition was specified by the authors. However, among these lines, the minimum frequency separation between *any* CO_2 line and *any* NH_3 transition is that between the $R4$ CO_2 line and the $sQ(7,5)$ NH_3 transition, and is 690 MHz, which is more than ten times larger than the 60-MHz frequency sweep of their laser light.

[3]R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. 27, 631 (1971), and 28, 1430 (1972), and Phys. Rev. A 6, 2001 (1972).

[4]D. C. McKean and P. N. Schatz, J. Chem. Phys. 24, 316 (1959); T. Shimizu, F. O. Shimizu, R. Turner, and T. Oka, J. Chem. Phys. 55, 2822 (1971). See also, R. G. Brewer and J. D. Swalen, J. Chem. Phys. 52, 2775 (1970) which gives a larger value of μ_{ν_2}.

[5]Shimizu, Ref. 2.

[6]T_1 of $C^{13}H_3F$ has recently been measured by the two-pulse nutation method using the Stark switching technique. J. Schmidt, P. R. Berman, and R. G. Brewer, Phys. Rev. Lett. 31, 1103 (1973). The values of the pressure-independent $\tilde{T}_1$ in their and my experiments, which should scale according to the pumping beam diameters and molecular weights, agree to within 25%.

[7]For example, if we assume that, after the population is inverted at $t=0$, the excited and ground state populations relax to equilibrium independently, $n_e=n_0\exp(-t/\tau_e)$ and $n_g=n_0[1-\exp(-t/\tau_g)]$, we would have $S(t)\sim n_e-n_g=n_0[\exp(-t/\tau_e)+\exp(-t/\tau_g)-1]$. In general, if τ_g and τ_e are different, the value of τ_{IR} should be between the two.

[8]C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955). Strictly speaking, the microwave pressure-broadening parameters give the phase memory time, τ_{g_2}, while we are interested in the population lifetime, τ_{g_1}. However, because of the small energy differences in microwave transitions, any phase-changing collision will also induce transition between states. Hence $\tau_{g_1}=\tau_{g_2}=\tau_g$. We note that τ_g of NH_3, $(J=8,K=7)$ line has also been directly measured by microwave transient nutation measurements. See J. H. S. Wang, J. M. Levy, S. G. Kukolich, and J. I. Steinfeld, Chem. Phys. 1, 141 (1973), and references therein.

[9]T. Oka, J. Chem. Phys. 48, 5919 (1968); T. Shimizu and T. Oka, Phys. Rev. A 2, 1177 (1970).

[10]Deduced from the pressure-broadening parameter of the $(J=3,K=3)$ line, whose value should be close to the (5,5) line in this experiment.

[11]A. T. Mattick, A. Sanchez, N. A. Kurnit, and A. Javan, Appl. Phys. Lett. 23, 675 (1973).

Decay of a Plasmon into Two Electromagnetic Waves

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In a plasma with a magnetic field, a plasmon can decay into two electromagnetic waves: a left-hand–polarized electromagnetic wave and right-hand–polarized whistler wave, both propagating in the direction of the magnetic field. Use of this process for a plasma laser is discussed.

Because an electromagnetic wave is cut off at a frequency below the plasma frequency, the de- of a plasmon into electromagnetic waves has been considered to be prohibited. I shall show here, however, that a plasma in a magnetic field is amenable to such a process.

In the presence of a magnetic field, electromagnetic waves are polarized. In particular, for the case of propagation parallel to the magnetic field, the left-hand polarized wave, whose dispersion relation is given by[1]

$$\epsilon_l(\omega,k)=1-\frac{\omega^2}{c^2k^2}+\frac{\omega_p^2\omega}{c^2k^2(\omega+i\nu+\omega_c)}=0, \qquad (1)$$

has a cutoff frequency

$$\omega_{cut}=\omega_p(1+\omega_c^2/4\omega_p^2)^{1/2}-\tfrac{1}{2}\omega_c, \qquad (2)$$

which is smaller than ω_p. Hence the wave propagates at a frequency below the plasma frequency. Here, ω_p, ω_c, and ν are electron plasma, cyclotron, and collision (angular) frequencies, respectively.

The decay of a plasmon (Langmuir wave at $k\sim 0$) to this wave will produce a low-frequency, right-hand–polarized electromagnetic beat wave at $\omega<\omega_c/2$. First I show that the beat wave satisfies the dispersion relation of the whistler wave,

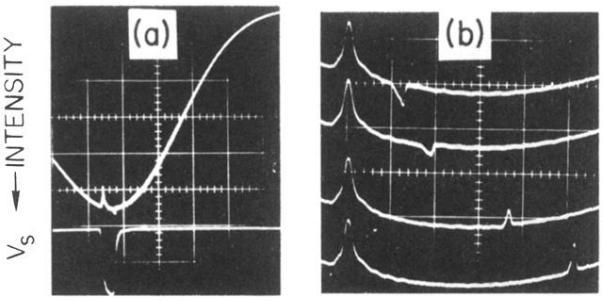

FIG. 1. (a) Dual-beam oscilloscope traces showing simultaneously the Stark pulse V_s (the lower beam) and the laser pulse (the upper beam) after passing through the Stark cell. The time scale is 1 μsec/div. The pressure of NH_3 in the cell was 10.4 mTorr. (b) Relevant part of the laser pulse at the expanded time scale of 0.2 μsec/div. Four separate shots at increasing Stark pulse durations are displayed.