Measurement of the $C\ ^3\Pi_u$ radiative lifetime of N_2 by laser-induced fluorescence

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The N_2 $C\ ^3\Pi_u$ lifetime was measured by laser-induced fluorescence (LIF) in a low-density plasma. To take account of radiation trapping efects, the lifetime was measured as a function of LIF signal amplitude, which is also a measure of the N_2 $B\ ^3\Pi_g$ density. To first order, radiation trapping is linearly proportional to B-state density. By extrapolating the data to zero density, values around 40.5±2 nsec were found for the C-state lifetime.

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The lifetime of N_2 $C\ ^3\Pi_u$ is important in the better understanding of nitrogen lasers as well as the use of excited nitrogen as a new light detection and ranging (LIDAR) target in the arctic and antarctic auroral zones. Measurements of the N_2 $C\ ^3\Pi_u$ lifetime over the last 30 years have ranged from 28–49 nsec [1–3]. These have included electron beam excitation measurements and recent laser-induced fluorescence (LIF) measurements at this laboratory [3,4]. Our new LIF measurements consider radiation trapping effects.

Radiation trapping occurs when the emitted fluorescence radiation is partially absorbed by the surrounding gas and reemitted, impeding its progress out of the gas. Radiation trapping, if it occurs, will increase the observed fluorescent decay time. To first order, for a long cylinder of radius A, the measured lifetime has been shown by Holt to be [5].

$$\tau_{\text{measured}}=\tau[1+(\alpha k_0 A\pi/2\sqrt{2})]\ , \tag{1}$$

where k_0 is the absorption coefficient at the line center,

$$k_0=\frac{\lambda^3 N}{8\pi\sqrt{\pi}u\tau}\frac{g_i}{g_f}\ . \tag{2}$$

For this experiment, N in Eq. (2) is the (assumed constant) density of N_2 $B\ ^3\Pi_g$, λ is the wavelength of the $C\ ^3\Pi_u$ to $B\ ^3\Pi_g$ transition (337.1 nm), g_i/g_f is the statistical weighting factor which is 1, u is the molecular thermal velocity, and τ is the radiative state lifetime. The α in Eq. (1) is a correction factor less than one which takes into account the branching ratios of various vibrational and rotational states.

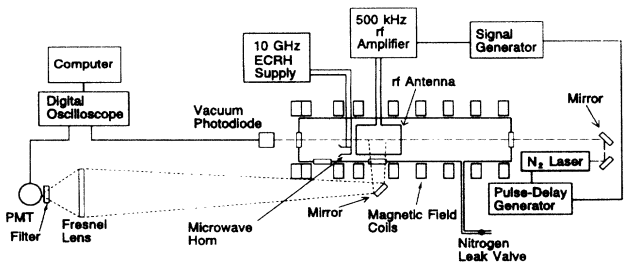

FIG. 1. LIF experimental setup for rf generated plasma case.

While the above expression was worked out for resonance trapping by atoms in the ground state, it is also relevant for molecules in an excited state when the lower level is populated, has a long lifetime, and the excitation efficiency is high [6]. This is the case for the nitrogen $B\ ^3\Pi_g$ to $C\ ^3\Pi_u$ transition in the present experiment. The B state has a lifetime of about 8 μsec.

The experiment was carried out with a neutral pressure below 1 mtorr. This is the low-pressure regime in which Eq. (1) is expected to be valid. Here low pressure means that the correction term in Eq. (1) is small, which also means that the radiation mean free path is greater than the chamber radius A. Emission lines are Doppler broadened. Collisional deexcitation of the C state is not important.

The effect of radiation trapping is linearly proportional to the N_2 $B\ ^3\Pi_g$ density. In the present experiment we measured the lifetime of the C state at different times in the afterglow of our nitrogen plasma. As the plasma decays and cools, the B-state density drops and the fluorescent decay time is seen to decrease slightly. We use the amplitude of the LIF signal as a relative measure of the $B\ ^3\Pi_g$ density at the time of the laser pulse.

The plasma was created by either a 10-GHz electron cyclotron resonance (ECR) source or by a rf discharge from an internal Nagoya type III antenna covering about one-quarter the length of the vacuum chamber. A

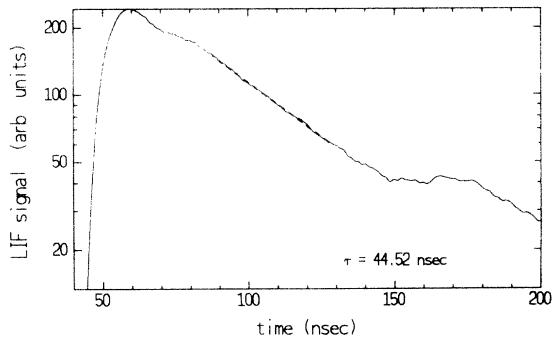

FIG. 2. Example of $N_2$357 nm LIF in the plasma afterglow. The dashed line shows the best fit slope over the fitted region.

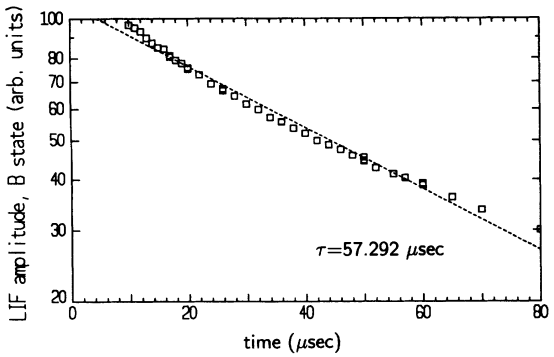

FIG. 3. N_2 B state as a function of time in the afterglow for the rf generated plasma. The B state decays much slower than its 8 μsec radiative lifetime.

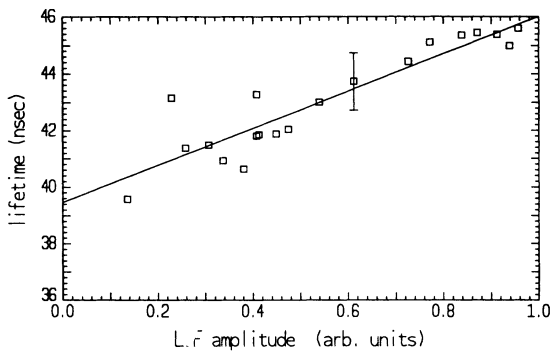

FIG. 5. N_2 $C\,^3\Pi_u$ lifetime as a function of B-state density in the afterglow of an ECR generated plasma. The base pressure was 5×10^{-4} torr.

schematic of the experiment is shown in Fig. 1. The plasma was pulsed on for 1 msec at a repetition rate of 10 Hz. Measurements were made in the quiet afterglow plasma after the exciter sources were off and the plasma optical emission had decayed.

The 337 nm laser radiation, used to excite the nitrogen B- to C-state transition, was produced by a Molectron UV-14 transverse discharge pulsed nitrogen laser. The laser pulse width was 15 nsec, the energy per pulse was 3 mJ, and the beam cross section was about 1 cm by 3 cm. The laser beam was directed down the axis of the chamber. The fluorescence was observed via a side port perpendicular to the beam path. The LIF light was relayed by mirrors and lenses to a large plastic Fresnel lens 2 m away from the vacuum chamber which focused the light through a narrow band interference filter onto an RCA 1P28 photomultiplier tube (PMT). The PMT was kept 12 feet away from the chamber to avoid any effects due to the 4-kG magnetic dipole field used with the ECR source. The magnetic field at the PMT was less than 10 G when the ECR source was used. The PMT was also surrounded by a magnetic shield.

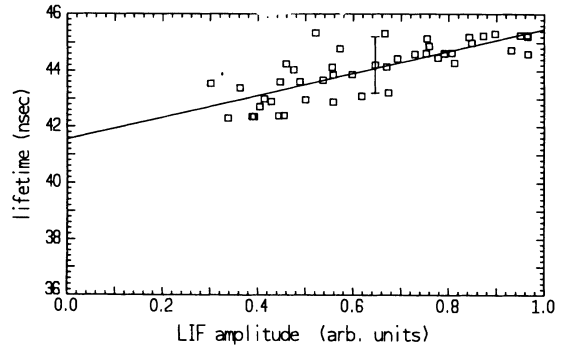

FIG. 4. N_2 $C\,^3\Pi_u$ lifetime as a function of B-state density in the afterglow of a rf generated plasma. The base pressure was 8×10^{-4} torr.

The interference filter was centered at 356.5 nm with a bandwidth of 8.8 nm. It passed the 357 nm (0–1) vibrational transition while blocking any direct laser light scattering as well as other plasma emission.

The data were acquired with a sampling rate of 2 GHz on a LeCroy 7200 digital oscilloscope. Each fluorescent decay curve was the average of 300 or more shots. The time constant was determined from about 80 points of the LIF decay curve starting 25 nsec after the laser pulse was over. We chose the fitting region to be away from any transient behavior near the laser pulse and away from any late-time nonexponential behavior.

A typical average fluorescent decay is shown in Fig. 2. The decay of the N_2 $B\,^3\Pi_g$ density for the rf drive case is shown in Fig. 3. The decay in Fig. 3 is much longer than the 8 μsec radiative lifetime and is due to plasma processes and the decaying rf drive field which create more B states from the 2-sec radiative lifetime $A\,^3\Sigma_u^+$ state. The plasma or antenna creation of the $C\,^3\Pi_u$ state was monitored by watching the plasma emission at 357 nm. The lifetime measurements are made at least a few μsec into the afterglow plasma when the emission at 357 nm has decayed away to less than 1% of the LIF signal. As we move further into the afterglow there are no detectable emissions at 357 apart from the LIF. Any baseline emissions are subtracted off before the lifetime calculations are made.

Figures 4 and 5 show the data and least-squares linear fit for the rf and ECR generated plasmas. The data show the expected linear dependence of B-state density. The ECR data do not show any reduction of the radiation trapping which might be expected from Zeeman splitting of the levels. The ECR source creates hotter electrons and presumably more B state than the rf drive. Increased B state could offset any effect from the Zeeman splitting. Projecting the slopes back to zero density gives a lifetime of 41.6 and 39.5 nsec for the rf and ECR plasmas. The scatter in the data as well as other measurements we have made suggest an error of ±2 nsec.

In summary, we have observed a dependence of the N_2 $C\ ^3\Pi_u$ lifetime measured by LIF on the $B\ ^3\Pi_g$ density which we attribute to radiation trapping. By extrapolating the data to zero B-state density, we find the radiative lifetime of the C state. Our average value for the N_2 $C\ ^3\Pi_u$ lifetime is about 40.5 ± 2 nsec.

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