Investigation of laser temporal pulse duration on Rayleigh scattering

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Relative Rayleigh-scattering cross sections from nitrogen have been measured for various pulse durations and wavelengths of incident laser radiation. No pulse-duration dependence has been observed for laser pulses as short as 5 ns, and classical theory is found to be still valid over the pulse-width range (5 ns $\leq \Delta t \leq 110$ ns) of our observations.

I. INTRODUCTION

Rayleigh scattering has been generally accepted for decades as an *in situ* calibration to Thomson scattering in plasma diagnostics.[1,2] Its validity for probe laser pulse durations less than 50 ns was recently questioned by Skowronek and Alayli.[3] Their experimental evidence showed that the Rayleigh-scattering cross sections drastically deviated from the classical values, revealing strong laser pulse-duration dependence. The dependence was given by the equation

$$S_{\text{expt}}/S_{\text{theor}}=(\tau_1/\tau)\,(1-e^{-\Delta t/\tau})\,,$$

where τ is an experimentally determined time constant ($\tau=28$ ns in their experiment), S_{expt} and S_{theor} are the experimental and the theoretical Rayleigh-scattering cross sections, τ_1 is the classical dipole damping constant, and Δt is the laser temporal duration. Similar experiments had been carried out by Kilkenny and White,[4] using a Q-switched ruby laser at 694.3 nm, and by Selter and Kunze,[5] using a flashlamp pumped dye laser at 600 nm. Over the range of pulse durations used in Refs. 4 and 5 (≤ 30 ns), no such pulse width dependence was observed.

The purpose of this experiment was to carefully investigate the quantitative nature of the laser temporal pulse dependence of the Rayleigh scattering by making precise and careful relative cross-section measurements. We also performed supplemental experiments such as scattering radiation pattern and its wavelength dependence, plus a check of the linear dynamic range of the detection system and filling pressure to uniquely identify the measurement of Rayleigh scattering. To ensure minimal signal distortion over the entire measurement time scales, we put some emphasis on improving the high-frequency response of the detecting systems.

The experimental detail will be described in Sec. II. The results and comparison with other experiments will be given in the final section.

II. EXPERIMENTAL METHOD

A commerically made coaxial flashlamp pumped dye laser was used as the probe laser source. It typically delivered a 700 ns pulse, with an energy of 1 J. A 1200-line/mm grating, mounted as the end reflector of the cavity, was used for wavelength tuning. The cylindrical dye cell has an aperture of 11 mm and a length of 42 cm. Both the liquid inlet and outlet of the cell have been constructed in a way such that the dye flow is uniform along the cell axis.[6] This arrangement minimizes dye turbulence and temperature gradients, which smooths out the spatial profile of the laser beam. A beam divergence of 1.5 mrad and a tuning bandwidth of 0.13 nm was achieved. Two dyes were used in this experiment, Rhodamine 6G at concentration 1×10^{-4} M and LD 490 at concentration 2×10^{-4} M in 100% ethenol, providing lasing wavelength at 590 and 486 nm, respectively.

A Pockels cell consisting of a deuterated KDP crystal with two Glan-Thomson polarizers on each side provides temporal pulse slicing of the laser output. The cell is gated by a trigger with an extremely sharp 1 ns rise and fall time. The extinction ratio is generally better than 1000. Laser-pulse durations between 3 to 120 ns can be easily obtained.

Stray light constitutes a major problem in this experiment, particularly for the cylindrical scattering chamber with an azimuthal scattering plane. The surrounding wall acts like a concave mirror, tending to enhance the background light noise level at the chamber center where the scattering experiment is being carried out. Therefore the probe

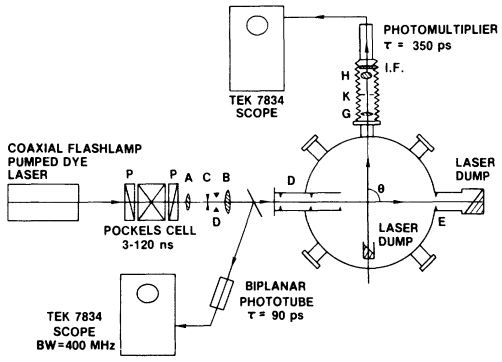

FIG. 1. Experimental setup with positive lenses *A*, *B*, *G*, and *H*, pinholes *C* and *K*, light baffles *D* and *E*, Glan-Thomson polarizes *P* and interference filter I.F. The test chamber has cylindrical symmetry, with the azimuthal plane chosen as the scattering plane. The laser polarization is perpendicular to the scattering plane.

beam was reduced from 11 to 3 mm diameter by entrance optics shown in Fig. 1 to diminish this effect. A sharp-edged aperture *C* with diameter $=600$ μm, mounted at the focal spot of a positive lens *A* with focal length $=5$ cm was imaged into the chamber center by a second positive lens *B* with focal length $=20$ cm. The aperture *C* not only defines the illuminated area but also performs certain spatial filtration. Four light baffles, *D*, one in front of lens *B* and three behind the entrance windows, were used to eliminate additional stray light caused by scattering from each optical element. A laser dump, made of absorbing glass with optical density greater than 8, mounted at the Brewster angle, significantly reduced back-reflected light. A light baffle, *E*, is inserted between the scattering center and the laser dump to prevent any stray light coming back from the dump surface.

The observing optics consists of two achromatic lenses *G* and *H* and an aperture *K* which defines the observing area. On axis but opposite the chamber, within the solid angle (5×10^{-3} sr) of the observing lens, is mounted another laser dump to provide further reduction of the background light. Finally, an interference filter with a typical bandwidth of 6 nm was put in series with the observing optics. As a result, a 5 to 1 signal to background ratio at 90° scattering angle for 53.3 kPa (400 Torr) of dry nitrogen was obtained. This ratio reduces to 2 to 1 at a 45° scattering angle and hinders any small angle scattering measurements.

The Rayleigh-scattering signal was measured by a fast photomultiplier (Hamamatsu 1294), while the incident laser power was monitored by splitting off part of the beam before the chamber entrance window and was sensed by a high speed biplanar phototube (Hamamatsu 1328U). This latter signal was used for normalization on shot-to-shot basis. Two Tektronix 7834 storage oscilloscopes (bandwidth 400 MHz) were used to record both detectors. The overall time constants for the scattered and the incident laser monitor signals were measued by the use of Hamamatsu picosecond light pulser C1308, and both were found to be close to 600 ps, essentially limited by the oscilloscope bandwidth.

Rayleigh scattering was measured using dry nitrogen with filling pressure ranging from 40 to 140 kPa (300 to 1087 Torr). A particle filter was inserted at the gas inlet to reduce the amount of dust. In addition, a sufficiently long waiting period was allowed after each tank fill to permit any remaining dust to settle. Nonlinear effects in the scattered signal due to excessive gas filling pressure and laser intensity were carefully checked and avoided prior to each new set of measurements.

III. RESULTS AND DISCUSSIONS

Rayleigh scattering from molecular nitrogen at 53.3 kPa (400 Torr) with an incident laser wavelength of 590 nm was measured at a 90° scattering angle (defined as the angle between the incident and the observing axes), where the scattering plane were perpendicular to the laser polarization. Each signal was normalized by the synchronous laser monitor trace; generally, data was taken at the time of the peak Rayleigh-scattering signal. All data were than divided by the mean value at 100 ns pulse duration as a relative calibration. The results are shown in Fig. 2. A least-squares linear-curve fit, $y=p+q\tau$, gives a constant term $p=0.97\pm0.02$ and a slope $q=0.0003\pm0.0003$. It is evident that there is no pulse-width dependence on the relative Rayleigh-scattering cross section from 5 to 100 ns. The error bars indicated here are the standard deviation from a minimum of 5 shots at each laser-pulse duration.

Similar measurements were made at scattering angles of 70° and 135°. In contrast to Ref. 3, which conclude a $\sin(\theta/2)$ dependence of the time constant τ, we observed no pulse-duration dependence at all. A typical measurement at 70° and a fit to a constant cross section is illustrated in Fig. 3.

The Rayleigh-scattering cross section has a

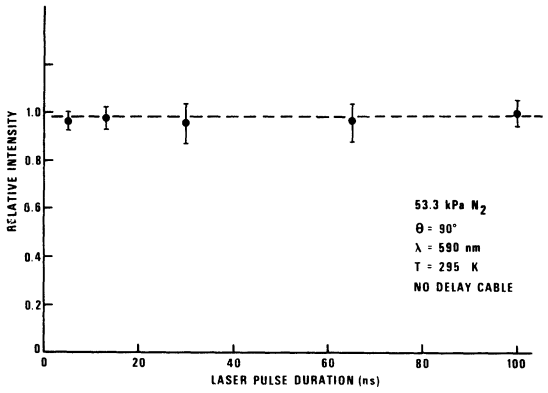

FIG. 2. Relative Rayleigh-scattering cross-section measurements for 53.3 kPa (400 Torr) dry nitrogen, at 90° scattering angle and laser wavelength 590 nm. The broken line is a least squares constant line fit.

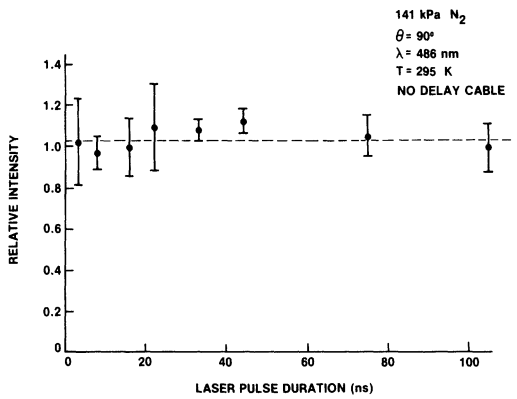

FIG. 4. Relative Rayleigh-scattering cross-section measurements on 141 kPa (1060 Torr) dry nitrogen, at 90° scattering angle and laser wavelength 486 nm. The broken line is a least squares constant line fit.

wavelength dependence λ^{-4}. This had been investigated by repeating the experiment with LD-490 dye at 486 nm. The measured ratio of the normalized signal between R6G and LD-490 was 0.43 ± 0.05, in excellent agreement with the λ^{-4} ratio 0.46. The conclusion of a pulse-duration independent cross section is also evident for 486 nm as shown in Fig. 4.

To further confirm the identity of Rayleigh scattering from other types of scattering such as Mie scattering by dust or oil contamination, etc., we measured the scattered radiation pattern in the same chamber azimuthal plane, but with the laser incidence normal to the observation plane (Z direction, in cylindrical geometry), and with 0° observing angle coinciding with the laser polarization.

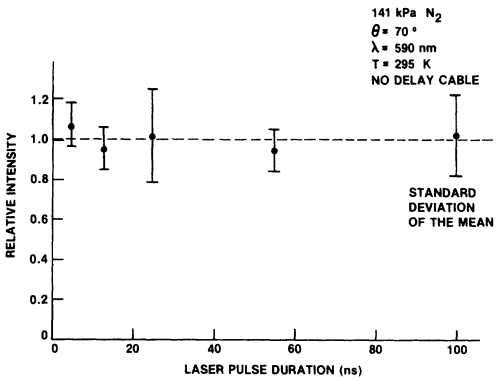

FIG. 3. Relative Rayleigh-scattering cross-section measurements for 141 kPa (1060 Torr) dry nitrogen, at 70° scattering angle and laser wavelength 590 nm. The broken line is a least squares constant line fit.

The normalized angular intensity distribution is illustrated in Fig. 5 in polar coordinates. The solid line indicates a least squares fit to $0.032 + 1.06\sin^2\theta$, from which a depolarization factor, $0.032/1.06=0.030$, is derived and found in reasonable agreement with the value of 0.032 obtained by Watson *et al.*[7] The observed fit can thus be interpreted as due to the presence of a weak depolarized component superimposing on the normal polarized dipole radiation, a typical Rayleigh-scattering characteristic. This measurement along with the

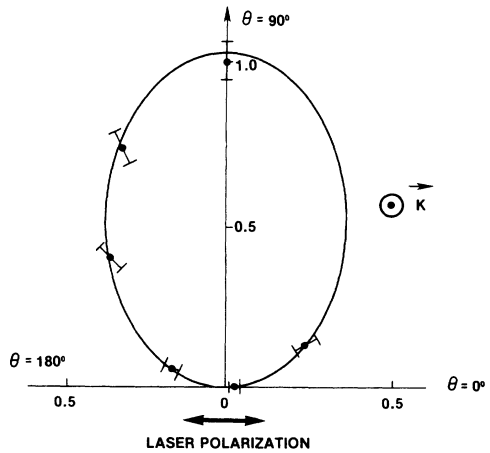

FIG. 5. Normalized scattered radiation in polar coordinates representation. θ is the azimuthal angle in the cylindrical chamber; and $\vec{K}$, being along the Z axis, is the laser propagation vector. Solid curve is a least squares fit to $0.032 + 1.06\sin^2\theta$. Laser polarized in $\theta=0°$ direction.

λ^{-4} dependence provides a definitive identification of Rayleigh scattering.

Since our results indicated no pulse-width dependence on the Rayleigh-scattering cross sections, in agreement with Refs. 4 and 5, we investigated possible causes of the $1-e^{-\Delta t/\tau}$ dependence of Ref. 3. To this end a mathematical simulation in which a Gaussian pulse was convoluted with the transfer function of an electronic integrator was performed on a computer. The output peak value was divided by the input Gaussian peak to simulate the way data is usually anaylzed. It was found that a time constant, τ, of 20 ns will reproduce the $1-e^{-\Delta t/\tau}$ dependence. The question then lies in what kind of integrator in the detecting system will produce a 20 ns integration time. We did a series of brief investigations with RG-58 coaxial cable, terminated with 50Ω at both ends, to provide a time delay between two experimentally matched phototubes both sensing the laser pulse simultaneously to simulate the recording of two signals on one oscilloscope. According to Ref. 8 this is the method used in Ref. 3 for recording the monitor and the scattered signals, with a 100 ns delay cable adding on the latter to separate the two signals in a single scope trace. The ratio of the two peaks as a function of laser-pulse duration is illustrated in Fig. 6. The results of Ref.3 are plotted on the same figure for comparison. It is clearly seen that a 100 ft delay cable will possess an effective time constant of 7 ns, whereas a 300 ft one will give 15 ns. The decrease of the delayed pulse amplitude is believed to be due to cable dispersion effects. Because of the use of the two recording oscilloscopes to avoid the delay cable problem, as mentioned in Sec. II, the effective time constant in our experiment is found to be at most 600 ps, essentially limited by the scope bandwidth. Consequently, no noticeable distortion was detected for laser-pulse widths between 5 and 110 ns. However, if we added the RG-58 delay cable to the photomultiplier channel, we obtained the same curves as in Fig. 6. It is therefore believed that the cable associated time delay may be the major reason for the pulse-width dependence of Ref. 3.

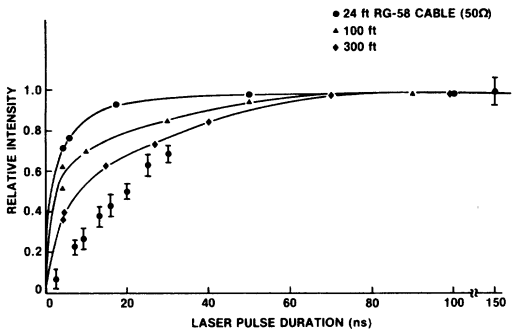

FIG. 6. Variations of signal attenuation due to RG-58 delay cables. Reference 3 results, shown as the group of circles with error bars, is presented for comparison.

In conclusion, we have measured the relative Rayleigh-scattering cross sections from molecular nitrogen for laser duration ranging from 5 to 110 ns, and found no temporal pulse-duration dependence at all. The classical Rayleigh-scattering formula has been investigated for its λ^{-4} dependence, its dipole angular radiation distribution and its depolarization factor, and was found to be valid over the pulse-duration range of our experiment.

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