

Xenon Two-photon Absorption Laser Induced Fluorescence: A Neutral Density Measurement and Absolute Calibration Tool

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Motivation

Laser induced fluorescence (LIF) of transitions from the ground state allow for direct and absolute measurement of the ground state density of a given atom or ion. Two-photon absorption LIF (TALIF) has advantages over traditional LIF in several ways: greater spatial resolution, intrinsically non-resonant emission and absorption, and TALIF removes the need to use vacuum ultraviolet light for high energy transitions. We have already identified an atomic hydrogen TALIF scheme, in which excitation occurs with two ~205nm photons, and emission is at ~656nm. In order to make absolute density measurements of hydrogen, one must calibrate the system with a measurement on a known density of gas. Since atomic hydrogen is not available under normal conditions, we need to absolutely calibrate our TALIF system by measuring a known density of atomic gas and then compare the two-photon absorption cross-sections of the atomic gas and hydrogen. We have identified a TALIF calibration scheme in xenon, in which excitation occurs with two ~209nm photons, and emission is at ~656nm. The common emission wavelength is advantageous eliminates chromatic effects on collection optics and photo detectors. We have noted significant absorption line broadening effects in early studies, of xenon gas. Xenon has several stable isotopes, so we performed the same measurements on a single isotope sample of xenon 132, in order to eliminate mass effect broadening. This single isotope measurement has resulted in no observed 656nm emission. This suggests the 656nm decay path for xenon 132 is not an allowed transition. All of the measurements presented are taken with confocal collection.

TALIF Laser System

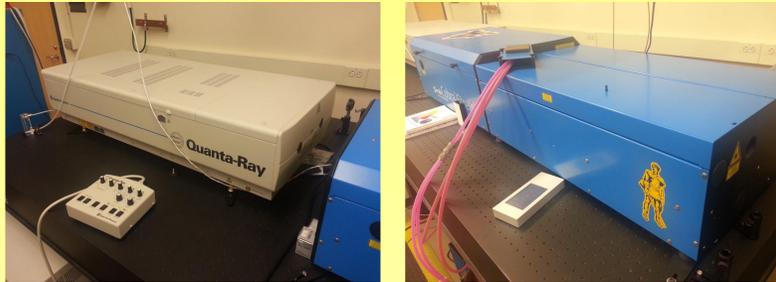
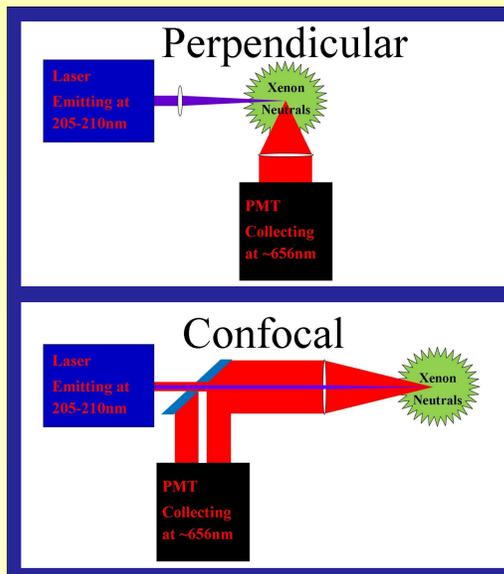


Figure 1. On the left is the pump laser which operates at 532 nm. On the right is the pulsed dye laser and frequency tripling stages that operate at spectral ranges of ~600-630 nm and ~200-210 nm, respectively.

Laser Stage	Energy/Pulse (mJ)	Pulse length (ns)	Spectral Width (1/cm)	Frequency (Hz)
Pump	~500	~9	< 1	20
Dye stage	~120	~12	.06-.08	20
Frequency tripling	~8	5-8	.12	20

Confocal and Perpendicular Collection Methods:



Perpendicular collection allows for some material advantages over confocal collection. Slightly more emitted light can be collected perpendicularly than confocally, because of the hollow center in our confocal set up. Perpendicular collection also allows for different sized collection and injection optics, which can be a materials cost advantage because special optics are required to manipulate the injected light.

Confocal collection has several operational advantages over perpendicular collection. The main advantage that confocal optics give is that they only require one access port, which is of great advantage in larger plasma devices where port access and space is limited. The collection and injection focal points are always overlapped, which maintains rough alignment at all times. The confocal optical setup can be put together as a single unit, which can be easily moved while maintaining rough alignment.

TALIF Fundamentals

Two-photon absorption is proportional to laser intensity squared

$$S \sim g * \sigma * n * I^2$$

where S is signal, g is absorption line shape, σ is the absorption cross section, n is the density of the state to be excited, and I is the intensity of incident light. We use a three state scheme so that the fluorescent light is at a distinctly different wavelength than the excitation wavelength.

TALIF of Multi-isotope Xe

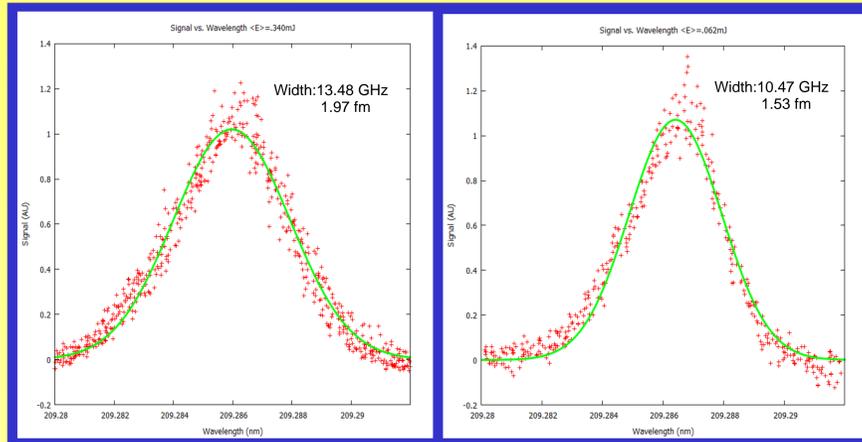


Figure 3: The intensity of collected light as a function of wavelength of injected light, where the collected light is normalized for injected laser energy.

We observe considerable broadening of the absorption spectra of xenon in these measurements. The likely causes of broadening are saturation broadening and hyperfine broadening. Xenon has four isotopes with over ten percent abundance and a total of nine stable isotopes. These measurements are all taken at room temperature (~0.25eV).

One contribution to isotopic broadening is just the mass shift of each isotope. For xenon, this is a very small because the mass of xenon is so large. The mass shifts are:

$$\Delta\lambda = \lambda_0 \frac{m_e \times (M_1 - M_2)}{M_1 \times M_2}$$

This expression gives a maximum shift of .05 fm from the farthest two isotopes and .0136 fm for the shift between the only two isotopes with non-zero nuclear spin. Neither of these two values are significant enough to account for the line width broadening observed in the above spectrum. The hyperfine shifts have not yet been calculated.

Xenon Saturation and Isotopic effects

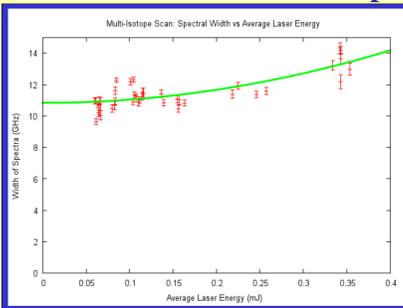


Figure 4: The spectral width as the average energy of the laser is decreased. The predicted spectral TALIF width at zero laser energy is 10.84 GHz. This is much higher than the Doppler width of a room temperature distribution.

Because projected width is not consistent with expectations for room temperature neutrals, there is clearly some other broadening term or isotopic splitting that is artificially enlarging the line width.

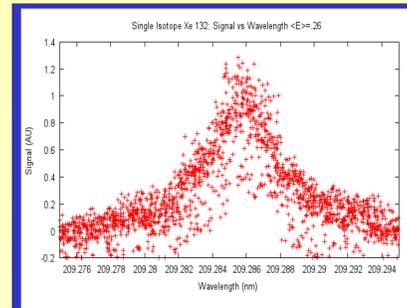


Figure 5: TALIF signal versus wavelength for several single isotope measurements. These data were obtained with a 1 Torr sample of isotopically pure Xe 132. The center of this distribution occurs at 209.2858 nm.

There are a few notable differences between the isotopically pure spectra and the multi-isotope xenon gas. One is the much larger variance in the signal to wavelength measurements due to the poor signal-to-noise of the measurement.

Another important distinction is the peak of the distribution being at a slightly different wavelength. The multi-isotope scans all peaked at 209.2862 nm. The single isotope scan peaks at a wavelength just under 209.286 nm. This shift, of >.3pm from the mean of several isotopes, is significantly larger than the simple mass effect splitting.

Hydrogen and Xenon TALIF Schemes

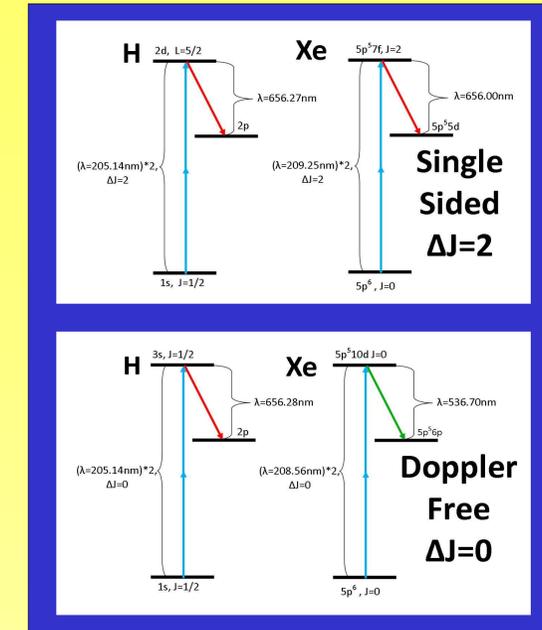


Figure 5. Grotrian diagrams for xenon and hydrogen in the single sided and Doppler free schemes.

The single sided TALIF scheme for hydrogen requires a $\Delta J = 2$ calibration scheme. Previous experiments in krypton have identified a TALIF scheme in krypton with nearly the same excitation wavelength as hydrogen, but with emission at 826 nm. The advantage of the single sided xenon scheme, is that the emission is nearly identical to that of the hydrogen scheme. This eliminates the issues of detection calibration across a wide spectral range for absolute calibration of hydrogen TALIF.

As yet, no Doppler-free calibration scheme, $\Delta J = 0$, has been developed for hydrogen wavelengths. Shown at right is a proposed scheme for Doppler-free excitation of ground state xenon. Emission, although not at 656 nm, is also in the visible and therefore the same well calibrated detector can be used for absolute calibration of Doppler-free TALIF in hydrogen.

TALIF Calibration

Absolute calibration of a hydrogen TALIF system is accomplished by comparing the TALIF signal attained from atomic hydrogen with that of the signal obtained from a noble gas at room temperature and a known density. The relationship between the unsaturated, intensity squared normalized signals is

$$\frac{S(P)}{S(C)} = \gamma \frac{\sigma(P) a(P) n(P)}{\sigma(C) a(C) n(C)}$$

P denotes the plasma species and C pertain the calibration species. Here S is signal normalized by laser energy squared, σ is the absorption cross section, a is the branching ratio of the observed decay path, and γ is the relative quantum efficiencies of detector at the observed wavelengths. Solving for $n(P)$

$$n(P) = \frac{S(P)\sigma(C)a(C)n(C)}{S(C)\sigma(P)a(P)\gamma}$$

Since the absorption cross section for the xenon transition is not known, we intend to measure the relative absorption cross section between the krypton and xenon TALIF schemes and then use the known krypton to hydrogen cross section relationship to determine the xenon to hydrogen relative cross section.

Summary

- **New single sided xenon TALIF scheme demonstrated.**
- **New xenon TALIF scheme emission line is nearly identical to hydrogen TALIF emission line.**
- **Doppler free measurements are needed to eliminate isotopic effects in xenon scheme.**
- **New Doppler free calibration scheme for hydrogen TALIF measurements proposed.**
- **Single isotope xenon shows dissimilar emission to regular gaseous xenon.**

