Doppler-Free Spectroscopy Measurements of Isotope Shifts and Hyperfine Components of Near-IR Xenon Lines

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Abstract. Xenon is currently used as propellant gas in electric thrusters, in which ejection of corresponding ions produces the desired thrust. As such a gas contains 9 stable isotopes, a non-intrusive determination of the velocity distribution function of atoms and ions in the thruster plasma plume, by means of absorption or fluorescence techniques, requires a precise knowledge of the line structure. We used Doppler-free Lamb-dip spectroscopy to determine isotopic shifts and hyperfine components of odd isotopes of several spectral lines of Xe atom and Xe^+ ion in the 825 – 835 nm range

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XENON AS A PROPELLANT GAS FOR ELECTRIC THRUSTERS

Xenon is currently used as propellant gas in electric thrusters like gridded ion engines and Hall effect thrusters [**1**]. In view of the performances of electric thrusters in terms of propellant mass savings and efficiency level, such devices are considered as a promising propulsion mean for space missions like platform station keeping and orbit transfer, space probe trajectory correction and long duration human interplanetary journeys [1,2].

A Hall Effect Thruster (HET) can be seen as a hollow annular ceramic channel confining a magnetized low pressure DC discharge generated between an external hollow cathode and an anode. Electrons emitted from the cathode are trapped around Larmor orbits and experience an azimuthal drift. The resulting low electron mobility induces a strong electric field that is concentrated in the vicinity of thruster exhaust. The propellant gas, which is fed through the anode, is efficiently ionized by trapped electrons in the downstream region. Created ions are then instantly accelerated along the thruster axis by the local electric field [3]. When operating near 1.3 kW, a HET ejects ions at 20 km/s and generates about 80 mN of thrust with efficiency in excess of 50%.

Profound understanding of physical phenomena governing the plasma dynamics inside a HET necessitates to accurately measure physical parameters such as the gas temperature, the magnetic field magnitude during thrusters' operation and especially the ion velocity distribution function. In order not to modify thruster plasma properties, these three quantities must be determined by means of laser-aided diagnostics providing information about Doppler broadening, Doppler shift and Zeeman splitting of xenon atomic and ionic lines. However, due to the existence of numerous Xe isotopes, some with non-zero nuclear spin, such measurements require a precise knowledge of the isotopic and hyperfine structure (HFS) of considered optical transitions.

DOPPLER-FREE SPECTROSCOPY

The hyperfine structure of a spectral line can be resolved when overcoming the limitation set by Doppler broadening. In this contribution, a non-linear absorption technique based on selective saturation of individual atomic transitions, the so-called Doppler free, or saturation, spectroscopy method, was applied with the use of a tunable single-mode diode laser [4,5]. A pump beam with wave vector +k is used to create a hole into the velocity distribution function (VDF) of atoms or ions in a resonant state. When tuning the laser, the hole is solely detected at

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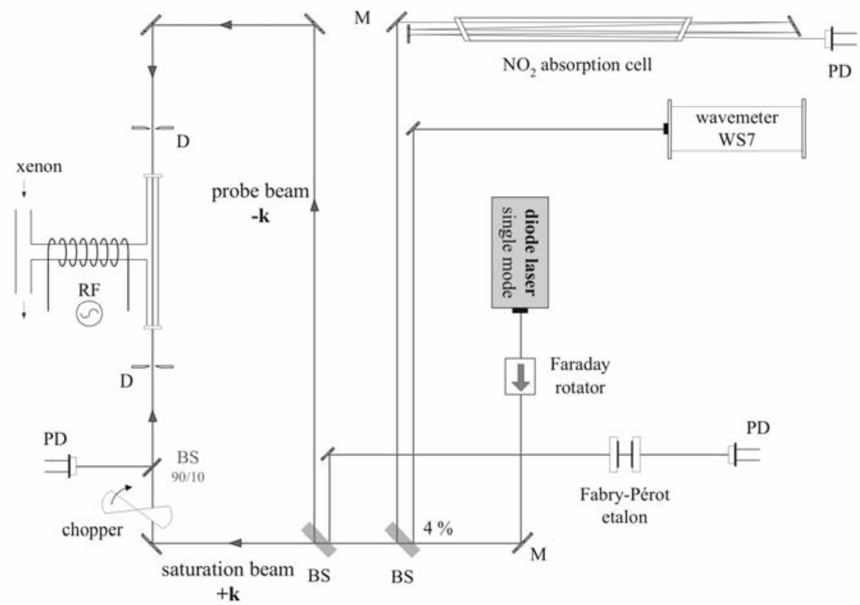

FIGURE 1. Schematic view of the Doppler-free laser spectroscopy setup used to determine isotopic shifts and hyperfine components of near infrared xenon transitions in a low-pressure RF discharge. A narrow linewidth (10 MHz) laser beam is produced by a single mode tunable diode laser. The Fabry-Pérot etalon (FSR = 1.37 GHz) is used to linearize the frequency axis and to detect mode hops. The optical setup is displayed in saturated absorption spectroscopy configuration. Two diaphragms are employed to limit the dispersion in wave vectors and to permit an optimized overlapping of the two beams. The wavemeter and the NO_2 absorption cell are both used to determine the absolute wavelength of every components.

the line center by means of a weak probe beam that originates from the same laser, with a counterpropagating wave vector –k. The moving atoms perceive the two light beams as having different frequencies, due to the opposite Doppler shifts. However, at the line center, both pump and probe beams interact with the atom or ion group experiencing a null velocity along the laser beam axis. The Doppler-free spectroscopy optical setup is depicted in Fig. 1. The holes in the absorption spectrum, the so-called Lamb-dips, exhibit a linewidth much narrower than the Doppler width: see upper trace in Fig. 2a. Modulating the pump laser beam intensity and processing the probe laser beam signal with a lock-in amplifier allows to solely detect Lamb-dip with a highly improved accuracy, as can be seen in Fig 2a. A high accuracy (100 MHz) wavemeter together with the recorded absorption spectrum of the NO_2 molecule serve to obtain the exact wavelength of each isotope and hyperfine component, see Fig. 2a.

A low-pressure (≈ 1 Pa) inductively coupled RF plasma discharge operating at 100 MHz is used as a source of excited atomic and ionic states [6]. The low gas density and the low plasma density ensure pressure broadening and Stark broadening to be negligible in comparison with the line broadening caused by the Doppler effect. However, due to the small amount of ions produced in a metastable state, the absorption technique is no more suitable to probe the ionic VDF and one must turn towards a Laser-Induced Fluorescence technique [7]. In that precise case, the saturation beam (+k) is reflected back into the plasma cell by means of a mirror and it is superimposed to the incident beam. The counterpropagating laser beam is then used as a probe beam (-k). The fluorescence light is detected at 90° with respect to the laser beam axis. When the probe beam intensity is modulated and the fluorescence radiation is monitored at the modulation frequency, the observed lineshape exhibits Lamb-dips at line centers of each transition.

Xenon atom (Z=54) has nine stable isotopes with natural abundance accessible to high sensitivity laser spectroscopy techniques. In addition two of these nuclei have a non zero nuclear spin (^{129}Xe: I=1/2 and ^{131}Xe: I=3/2) which brings about a hyperfine splitting of the levels. Isotope shifts and hyperfine structure in xenon have already

TABLE 1. Optical transitions investigated in this work.

Atom	Transition		J	λ in air [Å]
Xe I	$6s'[1/2]^o_1 \rightarrow 6p'[3/2]_2$	$1s_2 \rightarrow 2p_3$	1-2	8346.823
Xe I	$6s[3/2]^o_1 \rightarrow 6p[1/2]_0$	$1s_4 \rightarrow 2p_5$	1-0	8280.116
Xe II	$5d\ ^4F_{7/2} \rightarrow 6p\ ^4D_{5/2}$		7/2-5/2	8347.227

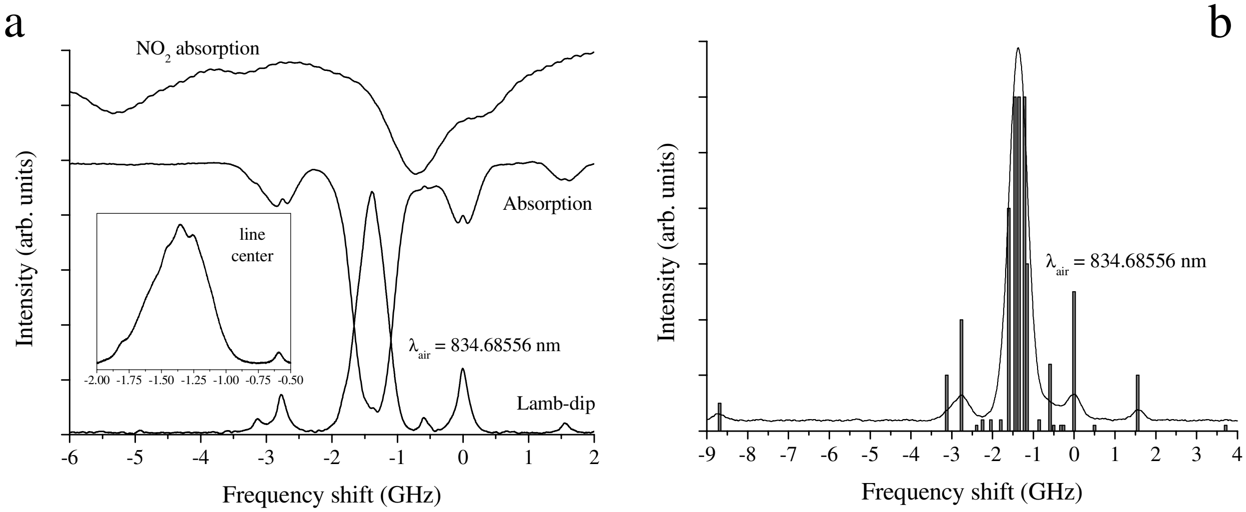

FIGURE 2. a) Part of the isotopic and hyperfine structure of the Xe atom $1s_2 \rightarrow 2p_3$ transition at 834.68 nm. Upper curve: Probe beam signal with Lamb-dips. Lower curve and insert: Lock-in outcome when modulating the pump beam. The NO_2 absorption spectrum is also shown. b) Complete experimental structure superimposed to a LIF spectrum: 21 components were observed. The bar height is here to guide the eye.

been measured for several transitions by means of high resolution Doppler-free laser spectroscopy [5]. In this work we investigate the structure of 3 strong near-infrared transitions in xenon gas, one of them being related to Xe^+ ion. The list of studied transition is shown in Table 1.

834.68 NM AND 828.01 NM XE I LINE STRUCTURE

The measured structure of the $1s_2 \rightarrow 2p_3$ transition of Xe atom at 834.68 nm is shown in Fig. 2b. The two odd mass isotope ^{129}Xe (I=1/2) and ^{131}Xe (I=3/2) are at the origin of 11 hyperfine components. To calculate the hyperfine structure, one must consider the total angular momentum $\mathbf{F} = \mathbf{J} + \mathbf{I}$ where **J** is the electronic angular momentum and **I** the nuclear spin. The selection rules for allowed dipole-dipole transitions are: $\Delta F = 0, \pm 1$ ($\Delta F = 0$ is forbidden if $F = 0$). Hence, ^{129}Xe yields 3 transitions and ^{131}Xe gives rise to 8 transitions. The overall hyperfine structure of the line can be described by means of 6 HFS constants. The 11 hyperfine transitions add up to the 5 mass shifted lines of the most abundant even isotopes.

The Lamb-dip spectrum shown in Fig. 2b exhibits 21 peaks whereas 16 peaks are expected in view of theoretical outcome. The isotopic transitions are likely to be concentrated in a narrow frequency domain. They are at the origin of the large and broad peak, however, saturated absorption spectroscopy permits to resolve them as can be seen in Fig. 2a. Isolated lines certainly belong to the HFS of the transition. Extra lines that increase the complexity of the measured spectrum can have two distinct origins. If two atomic transitions at center frequencies ν_1 and ν_2 with a common lower or upper level overlap within their Doppler width (in the RF plasma $T \approx 300$ K and $\Delta\nu_D \approx 390$ MHz) so-called cross-over signals appear in the Lamb-dip spectrum [4]. The frequency of the cross-over signal is just at the center between the two transitions: $\nu = (\nu_1+\nu_2)/2$. In Fig. 2b, the Lamb-dip at -2.04 GHz may well be a cross-over signal. “Ghost” lines, which are in fact measurement artifacts, may originate from the Lamb-dip detection accuracy. Noise fluctuations can easily generates small peaks that can by error be assigned to low intensity Lamb-dips. In order to diminish the probability of measuring ghost lines it is important to accumulate numerous saturated spectra in order to perform statistical checks.

The accuracy of the component frequency position is taken to be the narrowest linewidth that can be achieved for a single line under saturation conditions. When the pump beam power is low enough to avoid strong power broadening, one can achieve FWHM ≈ 50 MHz for each lamb-dip.

The whole structure of the $1s_4 \rightarrow 2p_5$ transition of Xe atom at 828.01 nm is composed of 5 HFS that add up to the 5 isotopic lines. The Lamb-dip spectrum of the Xe I $1s_4 \rightarrow 2p_5$ transition is shown in Fig. 3a: 13 lines have been experimentally identified. The line structure is compact since the lineshape solely spreads over 2 GHz. As can be seen in Fig. 3a, the strongest lines have been calibrated in wavelength.

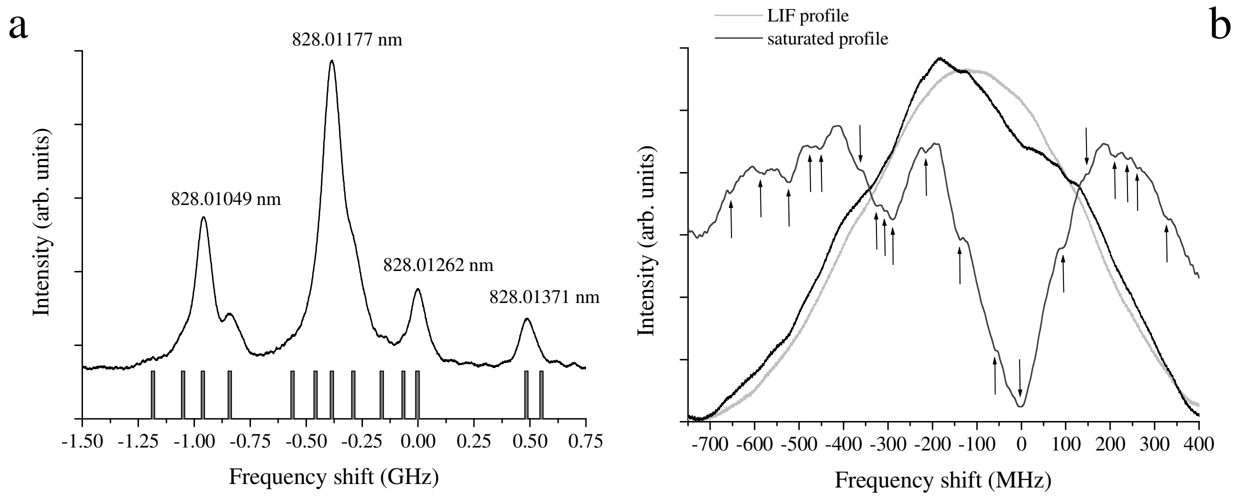

FIGURE 3. a) Isotopic and hyperfine structure of the Xe atom $1s_4 \rightarrow 2p_5$ transition at 828.01 nm: Lamb-dip spectrum and experimental structure. Numbers indicate λ_{air}. b) Structure of the Xe^+ $5d\ ^4F_{7/2} \rightarrow 6p\ ^4D_{5/2}$ transition at 834.72 nm: Fluorescence profile and saturated profile with Lamb-dips. Also shown is the difference between the two profiles, which is used to better determine dip position (arrows) especially in the wings.

834.72 NM XE II LINE STRUCTURE

The measured complex structure of the $5d\ ^4F_{7/2} \rightarrow 6p\ ^4D_{5/2}$ transition of Xe^+ ion at 834.72 nm is shown in Fig. 3b. The fluorescence light is monitored at 541.915 nm. The ^{129}Xe isotope yields 4 transitions and ^{131}Xe gives rise to 10 transitions. The 14 hyperfine components add up to the 5 even isotope lines. Holes in the fluorescence profile, which correspond to Lamb-dips, are clearly visible in Fig. 3b. The whole Xe^+ line stretches over 2 GHz. In order to improve the accuracy when determining the Lamb-dip positions, it is better to calculate the difference between the unsaturated fluorescence profile and the saturated profile. This method is better suited to than the ratio calculation on a noise generation viewpoint, especially in the wings of the profile. The lineshape difference is shown in Fig. 3b: 19 components are identified. Analysis of measured Xe^+ ion saturated profiles is still in progress.

All measured profiles must now be fully analyzed in order to determine isotopic shifts of the concerned transitions as well as the hyperfine constants of the lower and upper states of the transition [5]. Krypton is a good candidate to be employed as a propellant gas for electric propulsion. Therefore, HFS and IS of near infrared Kr and Kr^+ lines will be investigated in the near future.

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