Hyperfine structure and isotope-shift investigations of atomic nitrogen by saturation spectroscopy

P. Cangiano, M. de Angelis, L. Gianfrani, G. Pesce, and A. Sasso

Dipartimento di Scienze Fisiche, Università "Federico II" di Napoli, Mostra d'Oltremare Pad. 20, 80125 Napoli, Italy

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In this work we report a Doppler-free investigation of atomic nitrogen. As a laser source we used a narrow-band semiconductor diode laser emitting in the near infrared. Atomic nitrogen was produced by dissociating N₂ molecules in a radio-frequency discharge sustained by helium as a buffer gas. Hyperfine structures and isotope shifts of the transitions $3p \, {}^{4}P_{1/2-5/2} - 3s \, {}^{4}P_{1/2-5/2}^{\circ}$ were investigated for the two stable isotopes ${}^{14}N$ and ${}^{15}N$ by using absorption-saturated laser spectroscopy.

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INTRODUCTION

Atomic nitrogen is of great relevance in many chemical and physical processes such as, for instance, atmospheric physics, combustion kinetics, plasma physics, and astrophysics. Nitrogen is also interesting because, being an element with low Z, a comparison of precise experimental data and theoretical calculations is still possible.

Nuclear effects in the spectrum of nitrogen, such as hyperfine interactions and isotope shifts, are particularly attractive. Indeed, nuclear angular momentum of the two stable isotopes ¹⁴N (99.63%) and ¹⁵N (0.37%) are both nonzero: ¹⁴N nuclei are bosons, I = 1, while ¹⁵N nuclei are fermions, $I = \frac{1}{2}$.

Despite these relevant features, little information is available on high-resolution spectroscopy of this element and, according to our knowledge, no Doppler-free investigations have been performed. Isotope-shift (IS) effects are among those less investigated, since the only experimental study by Holmes [1,2] dates back more than 40 years ago; interferometric techniques were used to measure isotope shifts of several transitions in the near infrared and in the ultraviolet.

The study of hyperfine structures (HFS) in nitrogen has been essentially devoted to the ground state $(1s^22s^2 2p^3)^4S_{3/2}$.

It is well known that for the ground state of the halffilled shell, both magnetic-dipole hyperfine constant Aand electric-quadrupole constant B would be zero [3]. In reality, A is approximately 10 MHz and B is about 1 Hz for the ${}^{4}S_{3/2}$ ground state of ${}^{14}N$, as was first measured by Heald and Beringer [4] and later, with greater accuracy, by Hirsch *et al* [5]. This result was explained by Dutta, Matsubara, and Pu [6] in terms of an unsymmetric interaction between the three valence electrons with the spin-up and spin-down core S electrons.

Experimental work on HFS in the ¹⁵N ground state was performed by Anderson, Pipkin, and Baiard, Jr. [7] who, because of the high accuracy of their experimental method, could investigate the ¹⁴N-¹⁵N hyperfine anomaly. Some attempts to investigate HFS of excited states were made in 1933 by Bacher [8], who explained the unresolved HFS patterns in terms of an unusually low magnetic moment. High-resolution studies of IS and HFS in the optical domain have been largely facilitated by the recent development of Doppler-free laser spectroscopic techniques [9]. Nevertheless, nitrogen has been elusive to sub-Doppler investigations because of the difficulty in producing free atoms. Moreover, optical transitions of this element arise from excited states, while the resonance lines fall in the vacuum ultraviolet, where tunable laser sources of high spectral purity are not available.

In this paper we show an efficient scheme to produce atomic nitrogen in excited states based on the dissociation of molecular nitrogen (present at trace level) in a radio-frequency discharge sustained by helium as a buffer gas. This has allowed us to perform a sub-Doppler investigation of nitrogen by using saturation spectroscopy.

In particular, we have measured ${}^{14}N{}^{15}N$ isotope shifts and investigated the hyperfine structures of the transitions $3p {}^{4}P_{1/2-5/2} - 3s {}^{4}P_{1/2-5/2}^{o}$ in the near infrared $(\lambda \approx 820 \text{ nm})$. From HFS splittings measurements we have estimated the magnetic-dipole (A) and the electricquadrupole (B) coupling constants.

EXPERIMENTAL METHOD

The experimental setup for saturation spectroscopy used in our work is illustrated in Fig. 1. The laser source was an extended cavity semiconductor diode laser emitting at $\lambda = 826$ nm at room temperature. As a diode we used a commercial $Ga_x Al_{1-x}$ As-GaAs diode laser (Sharp Model LT015MD0). Typical output power was 30 mW with an injection current of 96 mA, while the laser bandwidth was found to be several tens of MHz. A narrower bandwidth of about 1 MHz was achieved by using a frequency stabilization scheme based on the optical feedback [10]. The diode was mounted in a pseudocavity consisting of a 1200-lines/mm diffraction grating mounted in Littrow configuration. The first-order diffracted beam was fed back in the diode cavity, whose output facet had a reduced reflectivity. A 30% reflectivity beam splitter was inserted between the laser and the grating in order to extract two laser beams used for the experiment. Coarse wavelength tuning was obtained by changing the diode temperature, while fine tuning was achieved by varying the injection current. Frequency scans of about 5

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FIG. 1. Experimental setup for saturation spectroscopy: PD photodiode; M, mirror; BS, beam splitter; L, lens; F, filter; FPI, Fabry-Pérot interferometer.

GHz could be realized by synchronously changing the injection current and the cavity length using a piezoelectric crystal on the grating. The laser wavelength was measured with a traveling Michelson interferometer with an accuracy of one part in 10^7 , while the frequency scan was calibrated using the markers from 1.5-GHz and 75-MHz free spectral range confocal Fabry-Pérot interferometers.

Doppler-free experimental arrangement was configured for saturated-absorption spectroscopy. The counter propagating pump and probe beams were weakly focused onto the discharge cell with 1-m focal length lenses and the power in the two beams was in the ratio 10:1. The intensity of the pump beam was mechanically modulated at 3 KHz, while the diode laser frequency was sweeped at frequencies of a few Hz.

Nitrogen atoms must be produced from the dissociation of a suitable molecule. This problem was first solved by Holmes [1], who dissociated N_2 atoms in a radiofrequency (rf) discharge. A different approach based on the photodissociation of N_2O was more recently employed by Svanberg's group in an experiment devoted to time resolved spectroscopy [11].

In recent papers [12] our group has demonstrated an efficient scheme for atomic oxygen production to perform high-resolution spectroscopy of this element. The method was based on the dissociation of O_2 in a rf discharge sustained by noble gases (technical details of the discharge cell are described elsewhere [13]). In spite

of the similarity between oxygen and nitrogen, the extension of O_2 apparatus to N_2 -noble-gas mixtures gave place to a quite low atomic nitrogen density. This motivated us to explore more efficient discharge configurations (among these, we also tested dc discharges, such as positive column and hollow cathode). The discharge used in this experiment consists of a resonant rf cavity. It is essentially the same as described by MacAlpine [14] and had already been applied by other groups [15,16] to produce atomic hydrogen beams. The scheme of our discharge is shown in Fig. 2 and here we give only a short description. A homemade rf oscillator was operating at 30 MHz with a maximum power of 100 W. The resonator consisted of a coil wounded in a helix within a copper shield, one end of the coil being connected to the shield. The helix was made of 14 turns ($\Phi=5$ cm) for a total length of 9 cm, while the shield diameter was 10 cm. The impedance matching was achieved by moving the contact along the helix until the reflected power (measured by inserting a power meter between the rf generator and the rf cavity) was minimized.

The discharge tube was in Pyrex; it was 42 cm long with an internal diameter of 6 mm and an external jacket to be cooled with water or liquid nitrogen (atomic nitrogen production improved by a factor of 2 when the discharge was cooled in liquid nitrogen). When the discharge was turned on, the plasma region extended over a distance of about 12 cm, while the electromagnetic noise around the apparatus was very low.

The discharge was operated in a sealed-off regime with a few mTorr of N_2 in the presence of several Torr of helium. We have used helium as a buffer gas because metastable noble-gas atoms are well known as efficient partners in molecular dissociating collisions, because of their high internal energy. A recent review concerning the basic mechanisms of such complex collisional processes is given in [17], while a phenomenological description of the case of O_2 -noble-gas collisions has been reported by one of the authors (A.S) in [18].

The low-amplitude noise exhibited by diode laser (one part in 10^6) allowed us to monitor atomic nitrogen by observing the absorption on the transition $3p \, {}^4P_{5/2}$ - $3s \, {}^4P_{3/2}^{o}$ at 824.4 nm; its density was optimized by varying the N₂ pressure, helium pressure, and rf power. The best conditions were achieved when the discharge was operated at 7



FIG. 2. Resonant radio-frequency cavity operating with He- N_2 mixtures to produce atomic nitrogen in excited states.

Torr of He and 0.1 Torr of N_2 , which produced an absorption of about 30%.

RESULTS AND DISCUSSION

In this work we have investigated the fine-structure transitions of the multiplet $3p \, {}^{4}P_{1/2-5/2} - 3s \, {}^{4}P_{1/2-5/2}^{o}$; a relevant energy-level diagram is shown in Fig. 3, where we report the wavelengths of the studied transitions and level positions [19].

A typical Doppler-limited recording is shown in Fig. 4(a) for the transition $3p \,{}^{4}P_{5/2}$ - $3s \,{}^{4}P_{3/2}^{\circ}$ of ${}^{14}N$ obtained with a N₂-He mixture ($p_{N_2} = 7$ Torr, $p_{He} = 0.1$ Torr) and a laser power of 2 mW. As shown in Fig. 4(a), the Doppler broadening ($\Delta v_D = 1.5$ GHz for a typical discharge temperature of 500 K) masks completely the HFS that, for the transition in question, gives place to six HFS components.

Doppler-free recording obtained with the saturation technique for the same transition ${}^{4}P_{5/2} - {}^{4}P_{3/2}^{o}$ is reported in Fig. 4(b). The small HFS splittings, combined with the relatively broad homogeneous width of the resonances, seriously limits the resolution of HFS components. Varying the laser power, the rf power, and the helium pressure, we have found that, in our experimental conditions, the main broadening was represented by collisions with buffer-gas atoms. In order to improve resolution, we have reduced the He pressure, reaching a good compromise between signal-to-noise ratio and line broadening at $p_{\rm He} = 1$ Torr, as shown in Fig. 4(c). A quantitative analysis of the homogeneous linewidth is not straightforward because the resonances are not completely resolved. Nevertheless, we have estimated the linewidth by fitting the whole HFS pattern to the superposition of a number of Lorentzian curves equal to the expected HFS components. In such a fit the amplitudes and the centers of the Lorentzians are, together with the homogeneous width, free parameters. From measurements of the spec-



FIG. 3. Fine-structure transitions of nitrogen investigated in this work.



FIG. 4. (a) Doppler-limited absorption recording of the transition $3p {}^{4}P_{5/2} - 3s {}^{4}P_{3/2}^{o}$ at 824 nm for 14 N obtained in a He-N₂ mixture ($p_{He} = 7$ Torr, $p_{N_2} = 0.1$ Torr, $P_L = 2$ mW, $P_{rf} = 80$ W); (b) absorption-saturated recording obtained under the same experimental condition as (a); and (c) absorption-saturated recording as in (b) but with $p_{He} = 1$ Torr.

trum as a function of helium pressure down to 0.3 Torr, we have estimated a natural width $\gamma = 67(4)$ MHz and a pressure broadening parameter of 123(5) MHz/Torr. The radiative width depends on the lifetime of the upper τ_u and lower τ_l levels involved in the investigated transition, i.e.,

$$\gamma_{\rm rad} = (1/2\pi)(1/\tau_{\mu} + 1/\tau_{1})$$
.

The lifetime of the level $3s {}^{4}P$ is $\tau_{l} = 2.5$ ns [20], while that of the upper level $3p {}^{4}P^{o}$, according to our knowledge, has not been measured. However, τ_{u} seems to be much larger than τ_{l} because the radiative width λ_{rad} calculated under the assumption $1/\tau_{u} \ll 1/\tau_{l}$ becomes 64 MHz, in good agreement with our estimation based on the measured profiles.

The program fit mentioned above, in addition to the homogeneous broadening analysis, has allowed us to assign the HFS pattern. A comparison of the experimental and the fit result of the transition $3p \ ^4P_{5/2} - 3s \ ^4P_{3/2}^o$ for the isotope ¹⁴N is shown in Fig. 5(a). The most prominent peak appearing at lower frequencies corresponds to the HF transition between the largest F values $(F'' = \frac{7}{2}$ and $F' = \frac{5}{2}$). All other HFS components are identified except for the weak $F'' = \frac{3}{2} - F' = \frac{5}{2}$ transition. From this analysis we can conclude that the upper-level hyperfine separations are inverted, while the lower-level ones are normal. Also, the agreement between the estimated intensities and the expected ones calculated from [21,22] is quite good, as shown in the lower part of Fig. 5.



FIG. 5. (a) Experimental (dashed line) and theoretical (dotted line) HFS pattern of the transition $3p \ ^4P_{5/2}$ - $3s \ ^4P_{3/2}^{\circ}$ for (a) ^{14}N and (b) ^{15}N (the numbers in brackets are the expected intensities).

Absorption-saturated spectra of the transition $3p {}^{4}P_{5/2} \cdot 3s {}^{4}P_{3/2}^{o}$ for the odd isotope ${}^{15}N$ obtained by using a 90% N₂ enriched sample are shown in Fig. 5(b). The nuclear spin of ${}^{15}N$ is $I = \frac{1}{2}$ and the angular momentum sum gives place to a simpler HFS pattern with respect to ${}^{14}N$. The identification of the positions of the three HFS components is done with the same program fit previously discussed. The result of the fit reveals a behavior of the HFS separations of ${}^{15}N$ that is opposite to that of the even isotope: upper-level separations are normal, while the lower-level ones are inverted. This result is easily explained because nuclear magnetic-dipole moments μ_I have opposite signs for the two isotopes: It is parallel to the nuclear spin I in ${}^{14}N$, $\mu_I = -0.2832\mu_N$ [21], with μ_N the nuclear magneton.

The values of the measured HFS splittings for the levels investigated in this work are reported in Table I (errors are one standard deviation obtained over a set of six recordings). We have extended Doppler-free investigations to other transitions of the multiplet, except for the transition ${}^{4}P_{1/2} {}^{-4}P_{1/2}^{o}$ for which the signal-to-noise ratio was very low.

We consider the transition ${}^{4}P_{5/2} - {}^{4}P_{5/2}^{o}$ at 821.8 nm, which has the lower level common with the previously investigated line at 824.4 nm. Doppler-free recordings are shown for ${}^{14}N$ and ${}^{15}N$ in Figs. 6(a) and 7(a), respectively. As one can see, the HFS patterns look very similar to the HFS patterns of the line at 824.4 nm, but only the strong-

 TABLE I. Experimental values of the hyperfine-structure

 splittings

Level	¹⁴ N		¹⁵ N	
	$\Delta_{F,F'}$	(MHz)	$\Delta_{F,F'}$	(MHz)
⁴ <i>P</i> _{3/2}	$\frac{3}{2} - \frac{5}{2}$ $\frac{1}{2} - \frac{3}{2}$		1-2	90±9
⁴ P _{5/2}	$\frac{5}{2} = \frac{7}{2}$ $\frac{3}{2} = \frac{5}{2}$	227±14 162±11	2-3	247±14
${}^{4}P_{1/2}^{o}$	22		0-1	154±10
⁴ <i>P</i> ^o _{3/2}	$\frac{3}{2} - \frac{5}{2}$ $\frac{1}{2} - \frac{3}{2}$	124±9 100±7	1-2	166±11
⁴ <i>P</i> ^o _{5/2}	$\frac{\frac{5}{2} - \frac{7}{2}}{\frac{3}{2} - \frac{5}{2}}$	23±20 29±20	3-2	158±19

est components are resolved at 821.8 nm. As a matter of fact, the experimental curves of Figs. 6(a) and 7(a) are well fitted with three and two Lorentzian curves, respectively, which does not allow a direct assignment of all the HFS components. Nevertheless, an estimation of splitting of the upper level can be done using the splittings from the lower level common with the previously studied transition ${}^{4}P_{5/2} {}^{4}P_{5/2}^{o}$. For sake of simplicity, we discuss this procedure for the case of 15 N which exhibits a simpler HFS pattern. The separation Δ_{a-c} between the two peaks of Fig. 7(a) corresponds to the sum of the splittings of the upper D_{3-2}^{o} and lower D_{2-3} level [for notation, see the level scheme of Fig. 7(a)]. Since the splitting D_{2-3} is known, the upper-level splitting is easily calculated.



FIG. 6. Doppler-free recordings of the transitions (a) $3p \,{}^{4}P_{5/2}$ - $3s \,{}^{4}P_{5/2}^{o}$, (b) $3p \,{}^{4}P_{3/2}$ - $3s \,{}^{4}P_{1/2}^{o}$, and (c) $3p \,{}^{4}P_{1/2}$ - $3s \,{}^{4}P_{3/2}^{o}$ for ${}^{14}N$.



FIG. 7. Doppler-free recordings of the transitions (a) $3p \,{}^{4}P_{5/2}$ -3s ${}^{4}P_{5/2}^{o}$, (b) $3p \,{}^{4}P_{3/2}$ -3s ${}^{4}P_{1/2}^{o}$, and (c) $3p \,{}^{4}P_{1/2}$ -3s ${}^{4}P_{3/2}^{o}$ for ¹⁵N.

Similar considerations can be extended to the case of ¹⁴N. The calculated splittings are reported in Table I. As is evident, the splittings of the level ${}^{4}P_{5/2}^{o}$ are affected by large errors and, hence, they represent only an estimation of the order of magnitude of these quantities. For this reason, as will be discussed later, they are not used to estimate hyperfine coupling constants.

The fine-structure transition ${}^{4}P_{3/2} {}^{-4}P_{1/2}^{o}$ at 822.5 nm is another line of the investigated multiplet which exhibited a partially resolved HFS, as shown in Fig. 6(b) (${}^{14}N$) and in Fig. 7(b) (${}^{15}N$). Nevertheless, only for the odd isotope does the fit program allow us to assign the HFS spectra and, hence, to determine HFS splittings (see Table I).

For lower angular momentum of the lower level, the HFS became completely unresolved, as shown in Figs. 6(c) and 7(c) for the transition ${}^{4}P_{1/2} {}^{-4}P_{3/2}^{o}$ at 819.0 nm. In this case the HFS pattern appeared as a single broad peak [$\Delta \nu \approx 300$ MHz full width at half maximum (FWHM)].

As is known, in the case of Russell-Saunders coupling the HFS splitting decreases with decreasing J and increases again when the sequence of levels becomes inverted [22]. The data of Table I show a dependence of the splittings over the total angular momentum J. As a consequence, when the two fine-structure levels of the investigated transition exhibit splittings comparable with each other and with the homogeneous width of the resonances, the envelopes of the HFS components are completely unresolved.

From the splittings reported in Table I, the magnetic-

TABLE II. Magnetic-dipole (A) and electric-quadrupole (B) HFS constants for the levels investigated in this work.

Level	¹⁴ N		¹⁵ N	
	A (MHz)	B (MHz)	A (MHz)	B (MHz)
$^{4}P_{3/2}$			45±9	0
${}^{4}P_{5/2}$	61±5	-50 ± 3	-82 ± 5	0
${}^{4}P_{1/2}^{o}$			154±10	0
${}^{4}P_{3/2}^{o}$	-54 ± 4	-23 ± 3	83±5	0
⁴ P ^o _{5/2}			53±6	0

dipole (A) and the electric-quadrupole (B) coupling hyperfine constants can be calculated. Two hyperfine levels are separated by [22]

$$\Delta_{F,F-1} = A_{\alpha J}F$$

+3B_{\alpha J}F[F²+\frac{1}{2}-I(I+1)
-J(J+1)]/[2I(2I-1)J(2J-1)],

where J is the total angular momentum and α stands for all the other quantum numbers that completely specify the level in question (for ¹⁵N, B=0, with I < 1). It follows from this equation that, given the HFS separations of a given term, the constants A and B can be determined. The values for A and B measured in this work are reported in Table II.

Finally, we have performed isotope-shift measurements for all the transitions ${}^{4}P_{J} {}^{-4}P_{J'}^{o}$. For the transitions where HFS was resolved, IS was determined as the distance between the centers of gravity of the HFS pattern of the two isotopes.

The results of these measurements are summarized in Table II where errors for HFS splittings represent the standard deviation over a set of six recordings. In the Table our results are also compared with the only experimental values obtained by Holmes [1]. The good agreement between the two determinations is remarkable although the investigations of [1] were Doppler limited. Holmes also investigated transitions involving doublet and quartet terms $(3p \, {}^2P_{1/2,3/2} - 3s \, {}^2P_{1/2,3/2})$ and $3p \, {}^4S_{3/2} - 3s \, {}^4P_{3/2,5/2})$. The IS turns out to be positive (${}^{15}N$ appeared at higher frequency) for doublet terms and negative for quartet terms.

It is well known [23] that the residual isotope shift (RIS) is the difference between the measured isotope shift and the normal mass shift (NMS). RIS is given by two contributions: the specific-mass shift (SMS) and the field shift (FS). The NMS is easily calculated as being about 950 MHz for the transitions investigated here. For light nuclei, such as nitrogen, the dominant contribution to RIS is the SMS, whereas the FS becomes not negligible for heavy nuclei (A > 60).

Since the IS for the transitions studied here is negative and the NMS is definitely positive, the SMS is also negative. The calculated values are reported in Table III.

It is apparent from the experimental results of Table III that the SMS is J dependent [24]. Indeed, the IS of the transition ${}^{4}P_{3/2} {}^{-4}P_{5/2}^{o}$ differs from the other transitions of the same multiplet by an amount larger than the

Transition	IS (This work) (MHz)	IS (Ref. [1]) (MHz)	NMS (MHz)	RIS (MHz)
${}^{4}P_{1/2} - {}^{4}P_{3/2}^{0}$	-1518±150		947	-2457
${}^{4}P_{3/2} - {}^{4}P_{1/2}^{0}$	-1610 ± 150	-1800 ± 180	945	-2555
${}^{4}P_{3/2} - {}^{4}P_{3/2}^{\circ}$	-1557±150		946	-2503
${}^{4}P_{3/2} - {}^{4}P_{5/2}^{\circ}$	-1235 ± 150		950	-2185
${}^{4}P_{5/2} - {}^{4}P_{3/2}^{0}$	-1901 ± 150	$-1788{\pm}18$	942	-2843
${}^{4}P_{5/2} - {}^{4}P_{5/2}^{o}$	-1788 ± 150	-1737±15	945	-2733

 TABLE III. Isotope-shift measurements. In the last column,

 Holmes' data are reported for comaprison.

probable errors. This effect has also been found in other experimental investigations [25,26] and it has been attributed by Keller [27] to relativistic corrections.

CONCLUSIONS

In this work we have demonstrated the possibility to perform high-resolution spectroscopic investigations of atomic nitrogen. This result was mainly obtained thanks to an efficient scheme for atomic nitrogen production based on dissociation of molecular nitrogen using a properly designed rf discharge. Doppler-free spectroscopy was performed by saturation spectroscopy using a frequency-stabilized semiconductor diode laser.

HFS and IS of the transitions ${}^{4}P_{J} {}^{4}P_{J'}^{o}$ were investigated for both ${}^{14}N$ and ${}^{15}N$ isotopes. The HFS was partially resolved and only for some of the transitions was the

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HFS pattern assigned with an appropriate fitting program. For these transitions the HFS splittings were measured and from such values HFS constants A and B were estimated. In order to improve the spectral resolution, we will explore other sub-Doppler techniques such as, for instance, polarization spectroscopy. The better sensitivity offered by this technique should allow us to operate the discharge at lower pressure in order to reduce the collisional broadening and, hence, to measure with greater accuracy HFS constants.

The isotope shift values reported in this work are mainly attributed to a large specific-mass shift, which is associated with correlations among electrons. It is also interesting to verify the J dependence of IS observed in this work by investigating other nitrogen transitions.

Our result should stimulate the theoretical analysis of the SMS in nitrogen. Indeed, such calculations require accurate knowledge of electronic wave functions which are based on Hartree-Fock methods. Very precise IS measurements represent a severe test for such complex quantum-mechanical calculations.

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FIG. 1. Experimental setup for saturation spectroscopy: PD photodiode; M, mirror; BS, beam splitter; L, lens; F, filter; FPI, Fabry-Pérot interferometer.