

ARTICLE

Isotope Shifts of Nitrogen around 800 nm

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The Doppler-limited absorption spectra of ^{14}N and ^{15}N atoms were measured around 800 nm using concentration modulation spectroscopy to study their isotope shifts. The nitrogen atoms were generated by discharging molecular nitrogen buffered with helium in a home-made discharge tube. The isotope shifts of four multiplets ($3s^4P_J \rightarrow 3p^4D_J^o$, $3s^4P_J \rightarrow 3p^4P_J^o$, $3s^2D_J \rightarrow 5p^2D_J^o$, and $3p^2P_J^o \rightarrow 5s^2P_J$) were measured and their J -dependent specific mass shifts were observed and discussed.

Key words: Doppler-limited absorption spectra, Isotope shifts, Specific mass shifts, J -dependence

I. INTRODUCTION

The isotope shifts (ISs) of atoms are needed in astrophysical studies such as the analysis of the quasar absorption spectra. By comparing the theoretically calculated and experimentally measured isotope shifts, it can be found that the change in the nuclear charge distributes from one isotope to another [1]. The study on the relative isotope abundance has many uses terrestrially, since various geological and biological processes tend to favor one isotope over another, it can help scientists to discover the chemical evolution of the universe. However, the IS parameters are limited and the IS characters are not well known for many atoms. The ISs of nitrogen atom are the topic of this study.

Holmes first observed a bunch of the isotope shifts of $3p^4P_J^o \rightarrow 3s^4P_J$, $3p^4S_J^o \rightarrow 3s^4P_J$, and $3p^2P_J \rightarrow 3s^2P_J^o$ transitions of ^{15}N - ^{14}N isotopic pair using a Fabry-Pérot interferometer [2]. In Holmes' experiment, no hyperfine structure in any component has been observed. After Holmes' work, several groups have reported the isotope shifts in some lines of nitrogen [3–7]. Cangiolo studied the hyperfine structures and isotope shifts of $3s^4P_J \rightarrow 3p^4P_J^o$ transitions using an external cavity diode laser and Doppler-free techniques [4]. Jennerich later reported the high-resolution saturation absorption spectra of $3s^4P_J \rightarrow 3p^4P_J^o$ and $3s^4P_J \rightarrow 3p^4D_J^o$ transitions [5]. Jönsson calculated the hyperfine structures of ^{14}N and ^{15}N using *ab initio* multiconfiguration Hartree-Fock method [6]. However, the hyperfine constants of $3s^4P_J \rightarrow 3p^4P_J^o$ and $3s^4P_J \rightarrow 3p^4D_J^o$ transitions from Jönsson's theoretical calculation are strongly inconsistent with these from Jennerich's experiment. Carette *et al.* studied the saturation spectra of the low

lying states of N I [7], their results are in agreement with Jönsson's theoretical ones.

A possible J -dependent specific mass shift of atomic nitrogen was found in some studies [2, 4–7]. However, the complete ISs of the involved multiplets were not measured. Here, we report our complete study on the ISs of $3s^4P_J \rightarrow 3p^4P_J^o$ and $3s^4P_J \rightarrow 3p^4D_J^o$ transitions, and some observed ISs on $3s^2D_J \rightarrow 5p^2D_J^o$ and $3p^2P_J^o \rightarrow 5s^2P_J$ transitions of atomic nitrogen. The J -dependent specific mass shifts of these multiplets were confirmed and discussed.

II. EXPERIMENTS

The experimental apparatus used in this work is illustrated in Fig.1, which is similar to that has been described previously [8, 9]. A single-mode Ti:Sapphire laser (Coherent Ring 899-29) pumped by a Nd:YVO₄ laser (Coherent Verdi-10, at 532 nm) was used as the excitation source. The laser beam passed through a home-made glow discharging (discharged at 23 kHz) absorption cell, which was made of a 60 cm long glass pipe with an internal diameter of 1.5 cm, then focused into a P-type layer/intrinsic layer/N-type layer (P-I-N) detector (Electro-Optics Technology, EOT-2030A). The output signal of the detector was demodulated using a lock-in amplifier at 2×23 kHz (concentration demodulation). Finally, the signal was acquired and processed by a computer to obtain the spectra of ^{14}N and ^{15}N . The nitrogen atoms were generated by discharging the mixture of trace $^{14}\text{N}_2$, $^{15}\text{N}_2$ buffered with helium (200 Pa). The absolute wavenumbers were determined by simultaneously recording the Doppler-limited absorption spectrum of I₂ [10]. The absolute wavenumbers were determined to an accuracy of ± 0.007 cm⁻¹.

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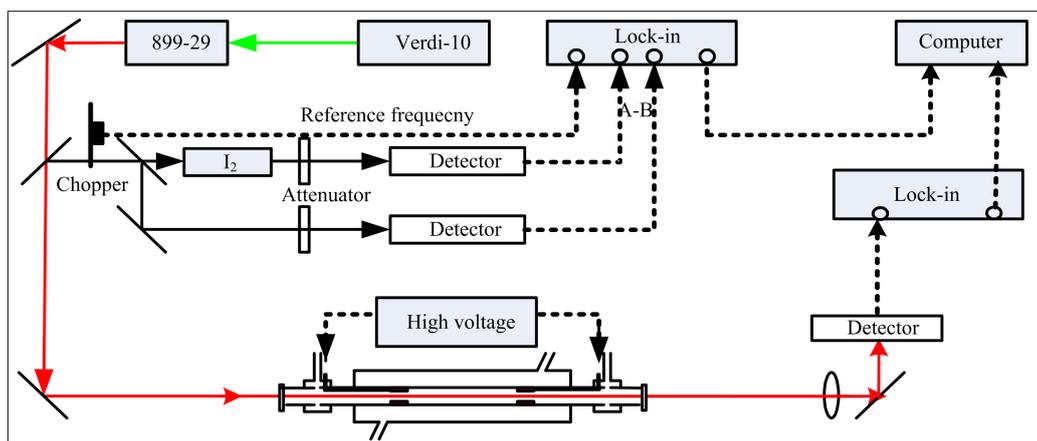


FIG. 1 Schematic of experimental setup.

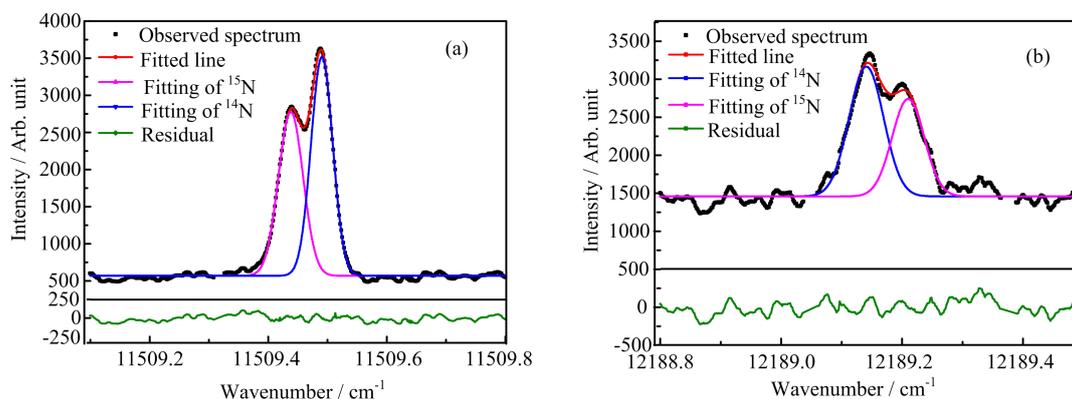


FIG. 2 Absorption spectra of (a) $3s^4P_{5/2} \rightarrow 3p^4D^o_{3/2}$ and (b) $3s^2D_{3/2} \rightarrow 5p^2D^o_{3/2}$ transitions of ^{14}N and ^{15}N . The black dot lines are our experimentally measured spectra, the red solid lines are the Gaussian fitting of the whole experimental spectra, pink and blue solid lines are the Gaussian fitting of the ^{15}N and ^{14}N spectra, the green solid lines are the residuals between the experimental data and the Gaussian fitting. In (a), the left peak is ^{15}N signal, right peak is ^{14}N , the value of IS ($(\nu(^{15}\text{N}) - \nu(^{14}\text{N}))$) is negative; In (b), the left peak is ^{14}N signal, right peak is ^{15}N , the value of IS is positive.

III. RESULTS AND DISCUSSION

Totally, we measured ISs for 20 transitions of 4 multiplets of nitrogen atoms. In the experiment, the volume ratio of ^{14}N , ^{15}N to helium mixture was properly prepared so that the spectra of two isotopes could be measured simultaneously with comparable intensity. Figure 2 shows partial of the absorption spectra that were fitted to extract the isotope shifts of nitrogen. The spectra of ^{14}N were individually measured to distinguish ^{14}N from the twin peaks of the IS spectra. Figure 2(a) shows the spectrum of $3s^4P_{5/2} \rightarrow 3p^4D^o_{3/2}$ transition, the peak at shorter frequency is ^{15}N , while the peak at longer frequency is due to the transition of ^{14}N . Figure 2(b) is the spectrum of $3s^2D_{3/2} \rightarrow 5p^2D^o_{3/2}$ transition, the peak at lower frequency is ^{14}N , the peak at higher frequency is due to the transition of ^{15}N . The exact line centers of absorption spectra were obtained by fitting the spectra with Gaussian functions and shared line widths. The residuals between the experimental

data and the Gaussian fitting are given in Fig.2. The full width at half maximum (FWHM) of these lines is around 1000 MHz, which is comparable to the Doppler broadening of this experimental system.

The isotope shift of nitrogen is the difference in the atomic spectra between ^{14}N and ^{15}N . It comes from two sources: the finite size of the nuclear charge distribution (the field shift, FS), and the finite mass of the nucleus (the mass shift, MS) [11, 12]. In first-order perturbation theory, the IS can be described as [13]:

$$\delta\nu_{\text{IS}}^{AA'} = \delta\nu_{\text{FS}}^{AA'} + \delta\nu_{\text{MS}}^{AA'} \quad (1)$$

The contribution from second source is traditionally divided into two parts: the normal mass shift (NMS) and the specific mass shift (SMS).

$$\delta\nu_{\text{MS}}^{AA'} = \delta\nu_{\text{NMS}}^{AA'} + \delta\nu_{\text{SMS}}^{AA'} \quad (2)$$

The NMS is calculated in a straightforward way from

TABLE I Summary of the isotope shifts of ^{14}N and ^{15}N around 800 nm. “Measured IS”= $v(^{15}\text{N})-v(^{14}\text{N})$, where $v(^{15}\text{N})$ and $v(^{14}\text{N})$ are the measured line-centers of the atom spectra. The NMSs are calculated from the experimentally measured ISs. The uncertainties for our experimental data are one standard deviation of the mean.

Transitions	Transitions		Measured IS/ MHz				NMS/MHz
	J''	J'	This work	Ref.[5]	Ref.[4]	Ref.[2, 3]	This work
$3s^4P_{J''} \rightarrow 3p^4P_{J'}^o$	5/2	5/2	-1761(10)	-1795.6(18)	-1788(150)	-1737(15) [2]	953
	5/2	3/2	-1805(10)	-1766.6(14)	-1901(150)	-1788(18) [2]	950
	3/2	5/2	-1590(15)		-1235(150)	-1500(300) [3]	957
	3/2	3/2	-1655(3)		-1557(150)	-1500(300) [3]	954
	3/2	1/2	-1609(14)	-1609.3(22)	-1610(150)	-1800(180) [2]	952
	1/2	3/2	-1549(27)		-1518(150)	-1500(300) [3]	956
	1/2	1/2	-1524 (14)			-1500(300) [3]	955
	$3s^4P_{J''} \rightarrow 3p^4D_{J'}^o$	5/2	7/2	-1847(10)	-1863.8(16)		
5/2		5/2	-1838(10)	-1853.07(84)		-1800(300)[3]	898
5/2		3/2	-1854(24)				895
3/2		5/2	-1660(10)				902
3/2		3/2	-1650(8)			-1800(300) [3]	899
3/2		1/2	-1669(27)				897
1/2		3/2	-1567(8)				901
1/2		1/2	-1565(6)	-1591.4(15)		-1800(300) [3]	900
$3s^2D_{J''} \rightarrow 5p^2D_{J'}^o$	5/2	5/2	1975(18)				959
	3/2	5/2	1876(21)				959
	3/2	3/2	2101(45)				955
$3s^2P_{J''} \rightarrow 3p^2P_{J'}^o$	3/2	1/2				2355(42) [2]	
	1/2	1/2				2253(18) [2]	
	3/2	3/2				2082(24) [2]	
	1/2	3/2				2034(27) [2]	
$3p^2P_{J''}^o \rightarrow 5s^2P_{J'}$	1/2	1/2	434(24)				961
	3/2	3/2	599(30)				963

the wavelength of the optical transition of ^{14}N [14].

$$\Delta\delta_{\text{NMS}}^{AA'} = \frac{m_e}{m_p} \frac{A' - A}{AA'} v_i \approx \frac{v_{\text{expt}}}{1822.888} \frac{A' - A}{AA'} \quad (3)$$

where, m_e is the electron mass, m_p is the proton mass, v_{expt} is the experiment wave number of ^{14}N in cm^{-1} , A' is the mass number of ^{15}N , A is the mass number of ^{14}N . The residual isotope shift (RIS) will be written as:

$$\delta v_{\text{RIS}}^{AA'} = \delta v_{\text{IS}}^{AA'} - \delta v_{\text{NMS}}^{AA'} = \delta v_{\text{SMS}}^{AA'} + \delta v_{\text{FS}}^{AA'} \quad (4)$$

The isotope shift values extracted out from experimental spectra are listed in Table I, along with those from literatures. The sign assigned to the isotope follows the rule that a shift to higher frequencies in going ^{14}N to ^{15}N is positive. In Table I the signs of the ISs of $3s^2D_J \rightarrow 5p^2D_J^o$ and $3p^2P_J^o \rightarrow 5s^2P_J$ are positive which have the same sign as the reported ISs of $3s^2P_{3/2} \rightarrow 3p^2P_{1/2}^o$ in Holmes' measurement [2], the ISs of $3s^4P_J \rightarrow 3p^4D_J^o$ and $3s^4P_J \rightarrow 3p^4P_J^o$ are negative which are the same as previous investigation [6–8]. The NMS

values were calculated using Eq.(3), so the uncertainties will not be given here.

The measured isotope shifts of $3s^4P_J \rightarrow 3p^4D_J^o$ transitions are listed in Table I and compared with the results measured using saturated absorption spectroscopy [5], and with those measured with Doppler limited spectroscopy [2, 3]. Our results show good agreement with Jennerich's measured ones [5]. We measured the ISs of all eight transitions, and also gave the systematic IS and NMS values.

The measured isotope shifts of $3s^4P_J \rightarrow 3p^4P_J^o$ transitions are also listed in Table I and compared with the results from Jennerich *et al.* [5], Cangiano *et al.* [4], and Holmes [2, 3]. Our results are close to Jennerich's results [5]. Our results are also close to Cangiano's results [4] except a large difference in $3s^4P_{3/2} \rightarrow 3p^4P_{5/2}^o$ transition.

Holmes reported the IS of the $3s^2P_J \rightarrow 3p^2P_J^o$ transitions [2]. However, we could not obtain high quality spectra to get the IS of this multiplet. Jennerich *et al.* [5] and Cangiano *et al.* [4] did not report these IS values. Here, we cited Holmes' results in Table I for further analysis. We also measured some ISs of $3s^2D_J \rightarrow 5p^2D_J^o$

and $3p^2P_J^o \rightarrow 5s^2P_J$, these values were newly reported as we know. Unfortunately, not all the fine structure transitions were observed.

In our work, the measured residual isotope shifts (RIS=IS–NMS) vary from -2781.7 MHz for $3s^4P_{5/2} \rightarrow 3p^4D_{7/2}^o$ to -2487.3 MHz for $3s^4P_{1/2} \rightarrow 3p^4D_{1/2}^o$ and from -2856.6 MHz for $3s^4P_{5/2} \rightarrow 3p^4P_{5/2}^o$ to -2549.6 MHz for $3s^4P_{1/2} \rightarrow 3p^4P_{1/2}^o$. From Eq.(4), the RIS comes from two components, the SMS and the FS. The FS depends on the variation of the electron density inside the nuclear charge distribution. As a general rule, for light atoms ($Z < 30$) the mass shift is the dominant contribution while for heavy atoms ($Z > 60$) the field shift plays the main role. The *ab initio* calculation of Jönsson *et al.* [6] gives the estimated value of FS around 0.2 MHz for these considered transitions. Thus for nitrogen, the FS can be neglected in the present work and the dominant part in RIS is SMS.

The schematic energy level diagram of the $2p^23s^4P_J \rightarrow 2p^23p^4P_J^o$ and $2p^23s^4P_J \rightarrow 2p^23p^4D_J^o$ transitions is shown in Fig.3. The transitions were assigned base on the data in NIST's database. The transition frequencies are extracted from our experimental data. From Fig.3, we can give the difference between two energy levels. For instance, the difference between the observed transitions in lines $3s^4P_{5/2} \rightarrow 3p^4D_{7/2}^o$ and $3s^4P_{5/2} \rightarrow 3p^4D_{5/2}^o$ gives the difference between the energy levels of $3p^4D_{7/2}^o$ and $3p^4D_{5/2}^o$.

A. $3s^4P_J \rightarrow 3p^4L_J^o$ (L=P or D) transitions

Table II listed the experimentally measured specific mass shifts of $3s^4P_J \rightarrow 3p^4P_J^o$ and $3s^4P_J \rightarrow 3p^4D_J^o$ transitions. The field shifts are neglected. In Table II, the lines of a multiplet do not exhibit the same isotope shifts and the shifts in the energy levels depend on the J -value. That is due to that the involved transitions of nitrogen atom are not a case of pure Russell-Saunders coupling, the shifts are J -dependence.

Holmes [2] first listed some J -dependence for the $3s^4P_J^o$ terms of nitrogen, for example the ISs measured difference relative to $3s^4P_{5/2} \rightarrow 3p^4P_{5/2}^o$ and $3s^4P_{5/2} \rightarrow 3p^4P_{3/2}^o$ is 51(33) MHz. Cangiano *et al.* [4] reported some J -dependence for the SMS of $3p^4P_J^o$ term, they obtained 110(300) and 318(300) MHz for the SMS differences $5/2-3/2$ measured relative to $3s^4P_{5/2}$ and $3s^4P_{3/2}$, respectively. We got SMS differences of 41(20) and 62(18) MHz for these two levels respectively. Carette *et al.* and Jennerich *et al.* gave differences of 1.0(35) and $-32.0(32)$ MHz with respect to the level $3s^4P_{5/2}$. We obtained $-48(17)$ and $-26(41)$ MHz for the SMS differences $3/2-1/2$ measured relative to $3s^4P_{3/2}$ and $3s^4P_{1/2}$, respectively. Only Cangiano *et al.* found a difference of $-52(300)$ MHz for the level of $3s^4P_{3/2}$.

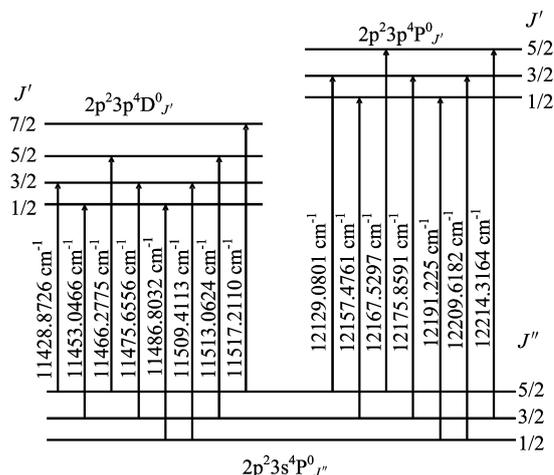


FIG. 3 Schematic energy level diagram of the $2p^23s^4P_J$, $2p^23p^4P_J^o$ and $2p^23p^4D_J^o$ states, and the observed transitions between these states around 800 nm.

In the case of the $3p^4D_J^o$ term, Jennerich [5], Carette [7], and this work obtained $-14.73(100)$, $1.77(102)$, and $-13(20)$ MHz for $7/2-5/2$ SMS differences of $3s^4P_{5/2}$, respectively. We also gave 13(34) and $-13(18)$ MHz SMS differences for the $5/2-3/2$ related to $3s^4P_{5/2}$ and $3s^4P_{3/2}$, 17(35) and $-3(14)$ MHz for the $3/2-1/2$ with respect to $3s^4P_{3/2}$ and $3s^4P_{1/2}$, respectively.

The experimental results of Cangiano *et al.* [4] showed a large J -dependence of the $3p^4D_J^o$ and $3p^4P_J^o$ states. Carette *et al.* [7] argued that the results from Cangiano *et al.* [4] are affected by a wrong assignment of the spectral lines. Our measured concentration modulation absorption spectra and Jennerich's saturated absorption spectra gave the SMS differences which are in the range 3–62 MHz. Carette's [7] saturation spectra gave a small J -dependence of these states. The results of Carette, Jennerich and present work exhibit small J - and L -dependence of the SMS for the $3p^4L^o$ (L=P and D) upper states. However, the limited relativistic *ab initio* calculations estimate a J -dependence of maximum 1 MHz [7] for $3p^4L^o$, which is an order smaller than our and Jennerich's results. Thus, more accurate experimental and theoretical results need to be proceeded by different methods.

For the lower $3s^4P_J$ term, Cangiano *et al.* gave the values of $-340(300)$ and $-548(300)$ MHz for the SMS differences between $3s^4P_{5/2}$ and $3s^4P_{3/2}$ sharing the same $3p^4P_{3/2}^o$ and $3p^4P_{5/2}^o$, respectively. Our work gave the values of $-146(13)$ and $-167(25)$ MHz for the SMS difference related to $3s^4P_{5/2-3/2}$, and $-104(30)$ and $-82(28)$ MHz for the SMS differences related to $3s^4P_{3/2-1/2}$.

The SMS difference between the different J -values of the lower levels are larger than those between the different J -values of the upper levels in $3s^4P_J \rightarrow 3p^4L_J^o$ transitions. This result is the same as that of Carette's [7].

TABLE II Specific mass shifts (in MHz) of the transitions of $3s^4P_{J''} \rightarrow 3p^4L_{J'}$. The field shifts are neglected.

Transitions			SMS/MHz			
	J''	J'	This work	Ref.[7]	Ref.[5]	Ref.[4]
$3s^4P_{J''} \rightarrow 3p^4P_{J'}$	5/2	5/2	-2714(10)	-2745.4(18)	-2745.4(18)	-2733(150)
	5/2	3/2	-2755(10)	-2746.4(17)	-2713.4(14)	-2843(150)
	3/2	5/2	-2547(15)			-2185(150)
	3/2	3/2	-2609(3)			-2503(150)
	3/2	1/2	-2561(14)	-2579.4(68)	-2558.3(22)	-2555(150)
	1/2	3/2	-2505(27)			-2457(150)
	1/2	1/2	-2479(14)			
$3s^4P_{J''} \rightarrow 3p^4D_{J'}$	5/2	7/2	-2749(10)	-2746.4(18)	-2762.9(16)	
	5/2	5/2	-2736(10)	-2748.17(84)	-2748.17(84)	
	5/2	3/2	-2749(24)			
	3/2	5/2	-2562(10)			
	3/2	3/2	-2549(8)			
	3/2	1/2	-2566(27)			
	1/2	3/2	-2468(8)			
	1/2	1/2	-2465(6)	-2488.1(15)	-2488.1(15)	

B. $3s^2D_J \rightarrow 5p^2D_J^o$ and $3s^2P_J \rightarrow 3p^2P_J^o$ transitions

Table III listed the specific mass shifts of $3s^2D_J \rightarrow 5p^2D_J^o$ and $3s^2P_J \rightarrow 3p^2P_J^o$ of ^{14}N and ^{15}N . The values deduced from our data and that deduced from Holmes' analysis are shown.

The measured SMS difference between the upper levels $5p^2D_{5/2}^o$ and $5p^2D_{3/2}^o$ is $-229(66)$ MHz, and the one between lower levels $3s^2D_{5/2}$ and $3s^2D_{3/2}$ is $99(39)$ MHz in present work. In Holmes' experiment, he measured the values of $-276(66)$ and $-222(45)$ MHz for the SMS difference between the upper levels $3p^2P_{3/2}^o$ and $3p^2P_{1/2}^o$ sharing the same $3s^2P_{3/2}$ and $3s^2P_{1/2}$, respectively. He also gave the value of $54(51)$ and $108(60)$ MHz for the SMS difference between the lower levels $3s^2P_{3/2}$ and $3s^2P_{1/2}$ sharing the same $3p^2P_{3/2}^o$ and $3p^2P_{1/2}^o$. Holmes' and our results show that SMS difference between the different J -values of the lower levels and those between the different J -values of the upper levels of the $3s^2D_J \rightarrow 5p^2D_J^o$ and $3s^2P_J \rightarrow 3p^2P_J^o$ transitions are small compared with that of $3s^4P_J \rightarrow 3p^4L_J^o$ transitions.

Cangiano *et al.* pointed out the J -dependence of the RIS values have been attributed by Keller *et al.* [15] to relativistic contributions and electron correlations affecting the SMS. The J -dependence of IS results from the higher-order IS effect, *i.e.*, the so-called crossed-second-order (CSO) effect or the far configuration-mixing effect [16]. The CSO effect between the magnetic interaction and IS operators leads to the J -dependence of IS in a term of the pure configuration. In Jennerich's report, he argued that identification of these effects has generally been limited by the availability of experimental data. SMS measurements reveal the nuclear shell very distinctly, the structure at magic

TABLE III Specific mass shifts of the transitions of $3s^2D_{J''} \rightarrow 5p^2D_{J'}^o$ and $3s^2P_{J''} \rightarrow 3p^2P_{J'}^o$. The field shifts are neglected.

Transitions			SMS/MHz	
	J''	J'	This work	Ref.[2]
$3s^2D_{J''} \rightarrow 5p^2D_{J'}^o$	3/2	3/2	1146(45)	
	3/2	5/2	917(21)	
	5/2	5/2	1016(18)	
$3s^2P_{J''} \rightarrow 3p^2P_{J'}^o$	3/2	1/2		1449(42)
	3/2	3/2		1173(24)
	1/2	1/2		1341(18)
	1/2	3/2		1119(27)

numbers can cause striking anomalies in the behavior of the IS of nitrogen.

IV. CONCLUSION

We systematically measured the isotope shifts spectra of ^{14}N and ^{15}N around 800 nm by using concentration modulation absorption spectroscopy. By analyzing these experimental data, we obtained the ISs between the two stable isotopes of atomic nitrogen. Our experimental results are close to other groups' results using saturated absorption spectroscopy method. The SMS value of $3s^4P_J \rightarrow 3p^4D_J^o$ transitions coming from the $3s^4P_{5/2}$ level is around 200 MHz larger than these coming from the $3s^4P_{3/2}$ level, and around 300 MHz larger than these coming from the $3s^4P_{1/2}$ level. For $3s^4P_J \rightarrow 3p^4P_J^o$ transitions, we can see the similar result. It may be concluded that the J value of the lower

level $3s^4P$ has a dominant contribution to the SMS values in both the $3s^4P_J \rightarrow 3p^4P_J^o$ and the $3s^4P_J \rightarrow 3p^4D_J^o$ transitions. The larger J value in $3s^4P$ level is, the larger IS value of the transition is. Similar phenomenon can be seen in Jennerich and Carette's results. For $3s^2D_J \rightarrow 5p^2D_J^o$ transitions, the J value of the upper level $5p^2D_J^o$ has dominant role in the SMS values. The larger the J value is, the smaller the IS value is. Holmes' experimental data show the similar result for the $3s^2P_J \rightarrow 3p^2P_J^o$ transitions. These results may be useful for comparing with highly red-shifted quasar absorption lines of nitrogen atom in order to obtain the fine structure constants, and can be used as a comparable experimental data for performing theoretical calculations on the anomalous isotope shift behavior in N atoms.

V. ACKNOWLEDGMENTS

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