



OSCILLATOR STRENGTHS OF VIBRIONIC EXCITATIONS OF NITROGEN DETERMINED BY THE DIPOLE (γ, γ) METHOD

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Received 2015 December 16; accepted 2016 January 27; published 2016 March 8

ABSTRACT

The oscillator strengths of the valence-shell excitations of molecular nitrogen have significant applicational values in studies of the Earth's atmosphere and interstellar gases. In this work, the absolute oscillator strengths of the valence-shell excitations of molecular nitrogen in 12.3–13.4 eV were measured by the novel dipole (γ, γ) method, in which the high-resolution inelastic X-ray scattering is operated at a negligibly small momentum transfer and can simulate the photoabsorption process. Because the experimental technique used in the present work is distinctly different from those used previously, the present experimental results give an independent cross-check to previous experimental and theoretical data. The excellent coincidence of the present results with the dipole (e, e) and those that were extrapolated indicates that the present oscillator strengths can serve as benchmark data.

Key words: ISM: molecules – methods: laboratory: molecular – molecular data

1. INTRODUCTION

Nitrogen is widely distributed in the atmospheres of a host of planets and satellites, and the airglow emission in the extreme-ultraviolet and vacuum-ultraviolet spectral regions, which resulted from the photoexcitation or electron-impact excitation of nitrogen, was observed in the atmospheres of these planets and satellites such as Triton (Broadfoot et al. 1989) and Titan (Strobel & Shemansky 1982; Stevens et al. 1994; Ajello et al. 2007). On this account, the photoabsorption cross sections (or the equivalent quantities of oscillator strengths) of nitrogen play an important role in the photochemical processes of the Earth's upper atmosphere, and are used to interpret the observations of nitrogen-rich planets and satellites (Strobel & Shemansky 1982; Broadfoot et al. 1989; Stevens et al. 1994; Ajello et al. 2007). Meanwhile, the valence-shell spectrum of nitrogen coincides with the wavelength region covered by the Far Ultraviolet Spectroscopic Explorer, so the oscillator strengths of nitrogen have been used to determine the interstellar nitrogen abundance toward the moderately reddened star HD 124314 in the constellation of Centaurus (Knauth et al. 2004), and test the models of nitrogen chemistry in the interstellar medium (Petit et al. 2004). Therefore, the high-precision oscillator strengths of nitrogen and its isotopic variants are greatly needed.

With such importance and applicable values, the oscillator strengths for the vibronic excitations of nitrogen have been paid a lot of attention by both experimentalists and theorists. Experimentally, the different techniques have been used to determine the oscillator strengths for the vibronic excitations of nitrogen. Early photoabsorption measurements of the oscillator strengths of nitrogen were carried out by Lawrence et al. (1968), Carter (1972), and Gürtler et al. (1977) at a relatively low resolution of about 5 cm^{-1} (about 0.6 meV). In principle, the photoabsorption measurements provide a direct determination of the oscillator strengths. However, the insufficient instrumental resolution may result in the line-saturation effect

and complicate the analysis of strong absorption lines (Hudson 1971; Chan et al. 1991). Aiming to recede the line-saturation effect, it is a worthwhile effort to improve the instrumental resolution. Then, Stark et al. (1992, 2000, 2005, 2008) measured the oscillator strength of nitrogen with the photoabsorption method using a much better instrumental resolution of about 0.7 cm^{-1} (about 0.09 meV), which is a sevenfold improvement compared with Lawrence et al. (1968), Carter (1972), and Gürtler et al. (1977). However, it is difficult to eliminate the line-saturation effect unless the instrumental resolution is infinitesimal, especially for the discrete transitions with narrow natural linewidths and large cross sections.

The electron-scattering technique operated at a negligibly small momentum transfer, i.e., the dipole (e, e) method, can be used to simulate the photoabsorption process of atoms and molecules, and is another commonly used experimental method to determine the oscillator strengths of atoms and molecules. Compared with the photoabsorption method, the dipole (e, e) method has the remarkable advantage that it is free from the line-saturation effect due to the nonresonant excitation character, though its typical energy resolution of tens of meV is lower than the photoabsorption method. In the early days, Geiger & Schröder (1969) reported the relative intensities of the vibronic excitations of nitrogen at an incident electron energy of 25 keV and a high-energy resolution of 10 meV. After correcting the scattering geometry, the relative oscillator strengths of the vibronic excitations were obtained from the relative intensities, but no absolute oscillator strength was derived. Then, Chan et al. (1993) measured the oscillator strengths of nitrogen at an incident electron energy of 3 keV and an energy resolution of 48 meV with the dipole (e, e) method. Using the same method, Zhang et al. (1997) also obtained the oscillator strengths of nitrogen at an impact energy of 1.5 keV and an energy resolution of 60 meV. In the same period, at an incident electron energy of 300 eV and the energy resolutions of 40–60 meV, Xu et al. (1995) obtained the oscillator strengths of nitrogen by extrapolating the generalized

oscillator strengths to the limit of $q \rightarrow 0$ based on the electron-scattering technique, where q is the momentum transfer. Based on the method used by Xu et al., Khakoo et al. (2008) also obtained the oscillator strengths of the $b^1\Pi_u$, $c^3\Pi_u$, $o^3\Pi_u$, $b^1\Sigma_u^+$, and $c^4^1\Sigma_u^+$ states at an incident electron energy of 100 eV, but the oscillator strengths of the vibronic states are not reported. In addition, Ajello et al. (1989) and James et al. (1990) also reported the oscillator strengths of nitrogen derived from the emission cross sections based on the electron-scattering technique.

In theory, Stahel et al. (1983) optimized a set of electronically coupled diabatic potential curves, then reproduced the anomalous absorption strength patterns within the vibrational progression. Then, Spelsberg & Meyer (2001) refined the analysis of Stahel et al. (1983) by combining ab initio potential curves with electronic couplings dependent on the internuclear distance R .

Despite considerable experimental and theoretical efforts, significant discrepancies remain in the previous oscillator strengths of vibronic excitations of nitrogen, e.g., for the strong transitions, the photoabsorption results (Stark et al. 1992, 2000, 2005, 2008) are systematically lower than the dipole (e, e) ones (Chan et al. 1993; Zhang et al. 1997). In this situation, it is very significant and important to investigate the oscillator strengths of nitrogen with a new experimental method, which can give an independent cross-check to the previous results. The oscillator strengths for the vibronic excitations of nitrogen were determined by the newly developed dipole (γ , γ) method (Kang et al. 2015; Xu et al. 2015) in this work.

2. EXPERIMENTAL APPARATUS AND PROCEDURES

The present experiment was carried out at the Taiwan Beamline BL12XU of SPring-8 at an incident photon energy of about 9889 eV and an energy resolution of about 70 meV. The experimental setup used in this work has been described in detail in our previous works (Xie et al. 2010; Zhu et al. 2011; Liu et al. 2014; Kang et al. 2015; Xu et al. 2015). Briefly, a Si(333) monochromator was used to achieve the high-energy resolution of 70 meV. Si(555) spherical analyzer with a 2 m radius of curvature was used to collect the scattered photons. In the measurement, the analyzer energy for the scattered photon was fixed, while the incident photon energy varied, from which the energy loss was determined. The energy loss spectrum of nitrogen measured at 2° by inelastic X-ray scattering (IXS) is shown in Figure 1 with the vibronic states assigned. For clarity, the vibronic states are represented by their term names and vibrational numbers, such as $b^1\Pi_u(v' = 0)$, written as $b(0)$ in this paper. In the present experiment, two different sample gases (helium and nitrogen) with pressures of 0.895 and 0.903 MPa were sealed in the same gas cell with Kapton windows, and then the gas cell was put on the experimental platform for the measurement. In addition, the actual transmissivities of two sample gases were measured, which were used to absolutize the experimental data obtained at the same experimental condition. All the spectra of helium and nitrogen were measured at room temperature.

In order to determine the intensity of individual vibronic excitation, the least-squares fitting was used to fit the experimental spectra. In the fitting procedure, the energy positions of the vibronic excitations were fixed to the ones summarized by Khakoo et al. (2008) and the peak profile was described by a Gaussian function. Then the relative oscillator

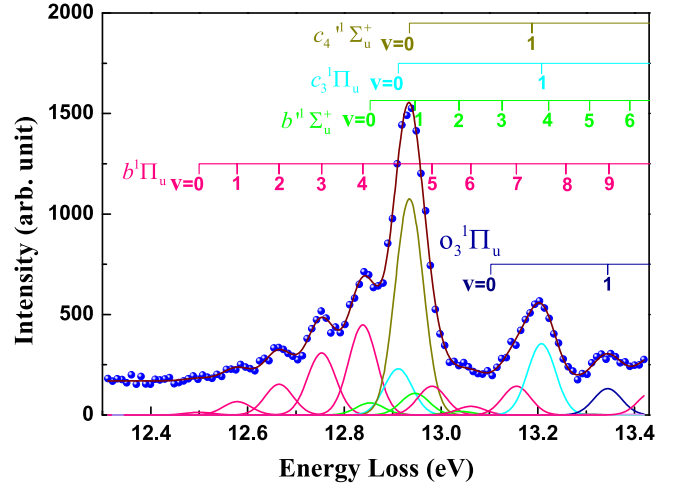


Figure 1. Energy loss spectrum of nitrogen at 2° measured by the IXS. Solid blue circles are the experimental data, and solid lines are the fitted curves.

strength was given as

$$f_0^r(\omega_n) = B_\gamma'(\omega_n) \frac{N(\omega_n)}{N_0} \frac{1}{D_0 \alpha} \frac{1}{l_{\text{eff}}} \frac{1}{n_0 P}. \quad (1)$$

$B_\gamma'(\omega_n)$ is the so-called Bethe-Born conversion factor at ω_n , which is determined accurately by simulating the actual arrangement of the light path in consideration of the rectilinear propagation of light (Xu et al. 2015). $N(\omega_n)$ and N_0 stand for the counts of the scattered photons and the intensity of incident photons, respectively. l_{eff} , n_0 , and P are the collision length, density of the target at 0.1 MPa, and pressure of the target in units of MPa, respectively. D_0 is a constant determined by the detection efficiencies of the ionization chamber and the detector of the scattered photon. The transmissivity α is determined by the sample species and its pressure, and it can be measured accurately with and without sample gas in the gas cell by an ionization chamber after the gas cell.

In order to obtain the absolute oscillator strengths of the transition of nitrogen at ω_n , a simple normalization method is used through

$$f_0(\omega_n) = \frac{B_\gamma'(\omega_n)}{B_\gamma'(2^1P)_{\text{He}}} \frac{[N(\omega_n)/N_0]}{[N(2^1P)/N_0]_{\text{He}}} \times \frac{\alpha_{\text{He}}}{\alpha} \frac{P_{\text{He}}}{P} f_0(2^1P)_{\text{He}}. \quad (2)$$

The 2^1P of helium, whose oscillator strength has been determined with a high accuracy both experimentally (Zhong et al. 1997) and theoretically (Grabowski & Chernoff 2011), was measured at the same scattering angle as nitrogen and used to normalize the results of nitrogen. The similar normalization method is used in our previous papers (Kang et al. 2015; Xu et al. 2015).

According to Equation (2), the experimental errors of the oscillator strengths of nitrogen, which are shown in the Table 1 and the corresponding figures, are attributed to the Bethe-Born conversion factors, the statistics of counts, the transmissivities and the pressures of both nitrogen and helium, as well as the oscillator strength of the 2^1P of helium. To be specific, the errors of $N(\omega_n)$ and $N(2^1P)$ were obtained from the fitting

Table 1
The Present and Previous Oscillator Strengths for the Vibronic Excitations of Nitrogen

Band	$b(0)$	$b(1)$	$b(2)$	$b(3)$	$b(4)$	$b(5) + b'(1)$ $+c_3(0) + c'_4(0)$	$b(6)$
Present dipole (γ, γ)	2.16(1.26)	10.8(1.3)	25.1(1.3)	51.2(2.3)	84.8(3.6)	261.5(10.5)	7.3(3.3)
Electron impact							
Chan et al. (1993)	2.54(0.25)	11.3(1.1)	27.2(2.7)	52.6(2.6)	86.1(4.3)	264.6(13.2)	5.0(0.5)
Zhang et al. (1997)	2.46(0.21)	11.9(0.7)	28.0(1.5)	54.1(2.7)	84.6(4.9)	280.97(16.69)	5.46(0.47)
Xu et al. (1995)	...	11.8(1.5)	27.8(3.6)	50.6(6.6)	78.2(10.2)
James et al. (1990)	1.4(0.3)	8.1(1.6)	18.2(3.6)	34.3(6.9)	55.0(11.0)	...	2.7(0.5)
Photoabsorption							
Lawrence et al. (1968)	20(10)	55(11)	180(41) ^a	...
Carter (1972)	35	58	47	102 ^a	...
Stark et al. (1992)	2.2(0.2)	9.6(1.0)	22(2)	51(5)	61(6)	201(29) ^a	...
Stark et al. (2000)	192(22) ^b	...
Stark et al. (2005)	2.4(0.2)	8.1(1.0)	21(2)	43(6)	68(7)	188.3(15.0)	4.0(0.4)
Calculation							
Stahel et al. (1983) ^c	2.56	11.63	29.38	54.94	75.38	242.74	4.86
Spelsberg & Meyer (2001) ^c	2.94	12.26	29.36	54.90	82.99	245.74	7.02
Band	$o_3(0) + b(7)$	$c_3(1) + b(8)$ $+c'_4(1) + b'(4)$	$b'(5) + o_3(1)$ $+b(9) + b'(6)$				
Present work	24.5(3.7)	61.0(3.5)	24.5(2.1)				
Electron impact							
Chan et al. (1993)	23.7(2.4)	65.5(3.3)	27.9(2.8)				
Zhang et al. (1997)	25.4(2.2)	62.6(4.4)	...				
James et al. (1990)	15.3(3.1)				
Photoabsorption							
Carter (1972)	19 ^d	58 ^e	24.75 ^f				
Stark et al. (2005)	17.1(2.0)	43.4(4.0)	...				
Stark et al. (2008)	19.8(2.9)				
Calculation							
Stahel et al. (1983) ^c	28.10	59.92	43.36				
Spelsberg & Meyer (2001) ^c	26.12	57.83	33.70				

Note. The listed data are amplified by a factor of 10^3 .

^a Not including the oscillator strengths of the weak transitions of $b'(1)$ and $b(5)$ (the oscillator strengths of $b'(1)$ and $b(5)$ are 0.45 and 2.8 reported by Stark et al. (2005), which are much less than the oscillator strength value 261.5 of $b'(5) + o_3(1) + b(9) + b'(6)$).

^b Same as footnote (a). The oscillator strength of $c_3(0)$ is taken from Stark et al. (1992) due to the fact that it is absent in Stark et al. (2000).

^c Normalized to the sum of the present oscillator strengths of all vibronic states listed in this table.

^d Not including the oscillator strength of the weak transition of $o_3(0)$ (the oscillator strength of $o_3(0)$ is 0.13, reported by Stark et al. 2005).

^e Not including the oscillator strengths of the weak transitions of $b(8)$ and $b'(4)$ (the oscillator strengths of $b'(4)$ and $b(8)$ are 1.8 and 0.44 reported by Stark et al. 2005).

^f Not including the oscillator strength of the weak transition of $b'(5)$ (the oscillator strength of $b'(5)$ is 1.1, reported by Stark et al. 2008).

procedure, which include the contributions of the statistics of counts. The measured accuracy of the transmissivities of helium and nitrogen are better than 0.1%. The pressures were measured by a commercial digital pressure manometer with a declared accuracy of better than 0.55%. The uncertainties of the Bethe-Born conversion factors are believed to be less than 0.2%. More details about the error analysis of the oscillator strengths obtained by the dipole (γ, γ) method can be referenced in Xu et al. (2015).

3. RESULTS AND DISCUSSIONS

In the measured energy region of 12.3–13.4 eV of molecular nitrogen, the most prominent transitions correspond to the valence-shell excitations of $b^1\Pi_u$, $b'^1\Sigma_u^+$, $c'^1\Sigma_u^+$, $c^1\Pi_u$, and $o^1\Pi_u$. Table 1, as well as Figures 2 and 3, summarize the vibrationally resolved oscillator strengths for the electronic transitions from the ground state to each vibronic state of $b(0 - 4, 6)$, $b(5) + b'(1) + c_3(0) + c'_4(0)$, $o_3(0) + b(7)$, $c_3(1) + b(8) + c'_4(1) + b'(4)$ and $b'(5) + o_3(1) + b(9) + b'(6)$, including

the present dipole (γ, γ) ones and the previously published results by the electron-scattering measurements (James et al. 1990; Chan et al. 1993; Xu et al. 1995; Zhang et al. 1997; see Figures 2(a) and 3(a)), photoabsorption measurements (Lawrence et al. 1968; Carter 1972; Stark et al. 1992, 2000, 2005, 2008; see Figures 2(b) and 3(b)), and calculations (Stahel et al. 1983; Spelsberg & Meyer 2001; see Figures 2(c) and 3(c)). It should be noted that, $b'(0, 2, 3)$ are very weak (Stark et al. 2005) and unresolvable with nearby vibronic states in the present work, so the contributions of $b'(0)$, $b'(2)$ and $b'(3)$ are included in $b(4)$, $b(6)$, and $b(7)$, respectively. For the overlapping transitions, such as $b(5)$, $b'(1)$, $c_3(0)$, and $c'_4(0)$, their energy intervals are much less than the present finite energy resolution, so the sum of their intensities is used to determine the total oscillator strengths of these states.

As shown in Figures 2(a) and 3(a), within the mutual experimental uncertainties, the present oscillator strengths match the dipole (e, e) results of Chan et al. (1993), Zhang et al. (1997), and the extrapolated ones of Xu et al. (1995) excellently. As for the oscillator strengths derived from the

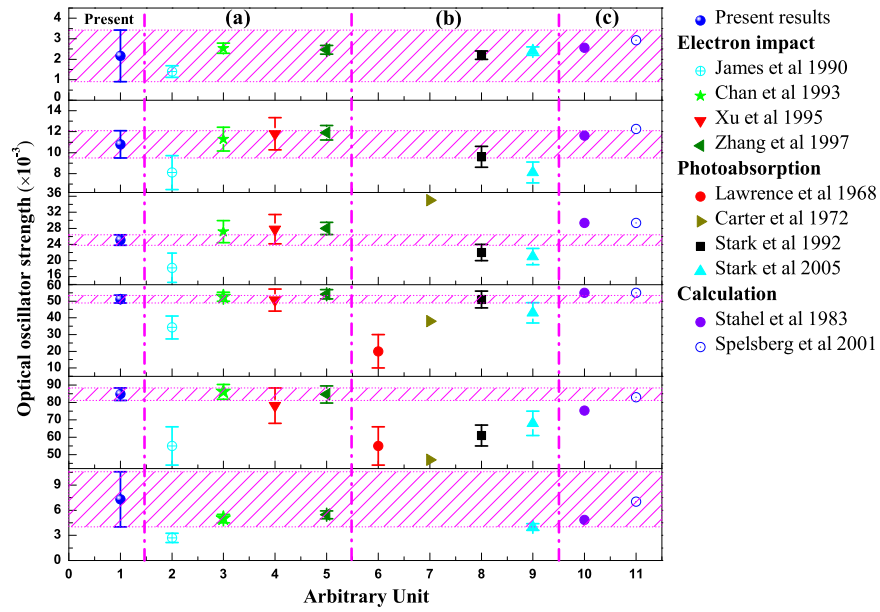


Figure 2. Present and previous oscillator strengths for $b(0 - 4, 6)$ from top to bottom.

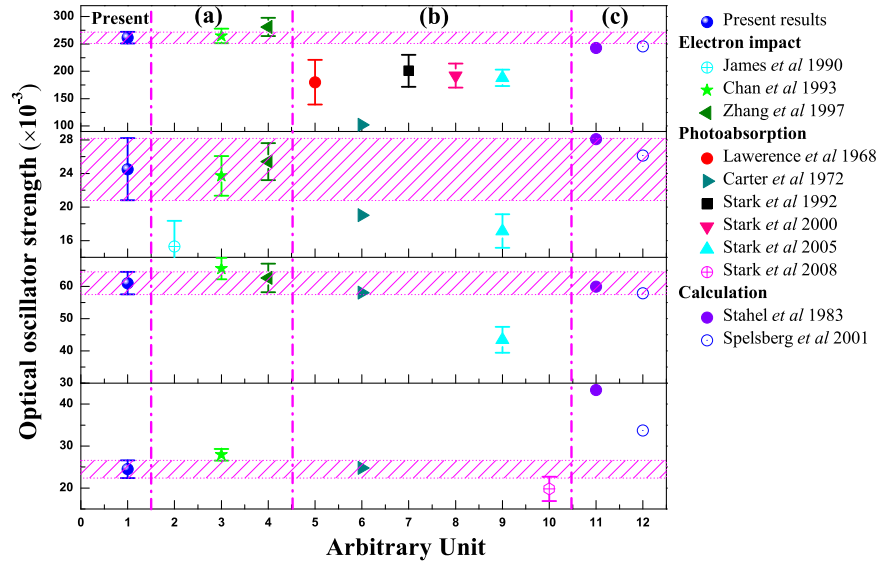


Figure 3. Present and previous oscillator strengths for $b(5) + b'(1) + c_3(0) + c_4'(0)$, $o_3(0) + b(7)$, $c_3(1) + b(8) + c_4'(1) + b'(4)$ and $b'(5) + o_3(1) + b(9) + b'(6)$ from top to bottom.

emission cross sections, the results of James et al. (1990) are about 30% lower than the present ones. It should be noted that the data of James et al. (1990) were normalized to the oscillator strength of $b(4)$ reported by the photoabsorption measurement of Lawrence et al. (1968). However, the oscillator strength of $b(4)$ of Lawrence et al. (1968) is about 35% lower than the present result. If the present oscillator strength of $b(4)$ is used as the reference standard to rescale the data of James et al. (1990), their oscillator strengths (James et al. 1990) are also consistent with the present ones within the mutual experimental uncertainties.

It can be seen from Table 1, as well as Figures 2(b) and 3(b), that, although for the weak transitions of $b(0)$ and $b(6)$ the present dipole (γ, γ) oscillator strengths are in agreement with the photoabsorption ones (Lawrence et al. 1968; Carter 1972; Stark et al. 1992, 2000, 2005, 2008), the apparent discrepancies, i.e., the photoabsorption oscillator strengths are much

lower than the present dipole (γ, γ) and the electron-impact measurements (Chan et al. 1993; Xu et al. 1995; Zhang et al. 1997), are observed for the strong transitions such as $b(4)$ and $b(5) + b'(1) + c_3(0) + c_4'(0)$. This is the typical feature of the line-saturation effect, which is more heavy for the stronger transitions. Furthermore, for the not so early photoabsorption measurements, there is not a tendency for the accuracy of the oscillator strengths to be improved with the ameliorating energy resolution. For example, the oscillator strengths of $b(4)$ and $b(5) + b'(1) + c_3(0) + c_4'(0)$ measured at the energy resolutions of 5 cm^{-1} (Lawrence et al. 1968) and 0.7 cm^{-1} (Stark et al. 1992, 2000, 2005, 2008) are in agreement with each other while they are systematically lower than the present dipole (γ, γ) and dipole (e, e) ones (Chan et al. 1993; Zhang et al. 1997). Therefore, it seems that the line-saturation effect for the strong transitions cannot be neglected even at an energy resolution of 0.7 cm^{-1} in the photoabsorption method.

In theory, Stahel et al. (1983) and Spelsberg & Meyer (2001) calculated the relative oscillator strengths of the vibronic states of nitrogen. It is clear that the present results are in agreement with the ones of Spelsberg & Meyer (2001) except for those of $b(2)$ and $b'(5) + o_3(1) + b(9) + b'(6)$ from Figures 2(c) and 3(c). It should be noted that the theoretical calculations only reproduce the experimental relative intensity distribution since they were normalized to the sum of the present s of all vibronic states.

4. SUMMARY AND CONCLUSION

In the present work, the accurate oscillator strengths of the vibronic excitations of $b(0 - 4, 6)$, $b(5) + b'(1) + c_3(0) + c_4'(0)$, $o_3(0) + b(7)$, $c_3(1) + b(8) + c_4'(1) + b'(4)$ and $b'(5) + o_3(1) + b(9) + b'(6)$ of nitrogen have been determined by the novel dipole (γ, γ) method based on the high-resolution X-ray scattering technique. The cross-check of the oscillator strengths measured by the present dipole (γ, γ) method, the dipole (e, e) method (Chan et al. 1993; Zhang et al. 1997), and the extrapolating method (Xu et al. 1995) based on the electron impact give us confidence that the present oscillator strengths can serve as benchmark data because the experimental techniques, experimental conditions, and normalization procedures used are distinctly different. For example, the target pressure in the present dipole (γ, γ) method are about 1 MPa, while it is about 0.01 Pa in the dipole (e, e) method (Chan et al. 1993; Zhang et al. 1997) and the extrapolating method (Xu et al. 1995). This work uses the oscillator strength of the 2^1P of helium to absolutize the oscillator strengths of nitrogen, while the dipole (e, e) method uses the TRK sum rule to normalize their data. The good agreement of the present results and those of the dipole (e, e) and the extrapolating methods (Chan et al. 1993; Xu et al. 1995; Zhang et al. 1997) excludes the possibility of any systematic error. Therefore, it is reasonable to think that the oscillator strengths of some strong transitions of nitrogen determined by the photoabsorption method are influenced by the line-saturation effect.

It is worth mentioning that every experimental technique has its own merits and demerits. Although for the strong transitions the photoabsorption method is influenced by the line-saturation effect, it has the highest energy resolution to determine the oscillator strengths of the rotational states. Although the present dipole (γ, γ) method, the dipole (e, e) method, and the extrapolating method with the moderate energy resolution of tens of meV can only measure the oscillator strengths of the vibronic states at the present stage, they are free from the line-saturation effect. The dipole (e, e) method has the advantage of a large cross section, which can achieve a good signal-to-noise ratio and a good statistical count, while the rapid variation of its Bethe-Born conversion factor may bring some uncertainties, as pointed out by Xu et al. (2015). The accurate Bethe-Born

conversion factor of the dipole (γ, γ) method can avoid this problem, and the only limitation of the dipole (γ, γ) method is its low cross section, which results in the poor statistical counts for the weak transitions such as the $b(0)$ and $b(6)$ in this work. However, the dipole (γ, γ) method is free from any systematic error, to the best of our knowledge. Thus it is possible to achieve a higher precision for the oscillator strengths with the dramatic progress of the third generation synchrotron radiation and the free electron laser.

This work was supported by the National Natural Science Foundation of China (Nos. U1332204, 11274291, and 11504361) and the Fundamental Research Funds for the Central Universities (WK2030040067). The experiment was carried out in a beam time approved by the Japan Synchrotron Radiation Research Institute (Proposal Nos. 2013B4250 and 2014B4251) and the National Synchrotron Radiation Research Center, Taiwan, Republic of China (Proposal Nos. 2013-3-057-1 and 2014-3-035-1).

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