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Spectrally resolved pressure dependence measurements of air fluorescence emission with AIRFLY

M. Ave [a], M. Bohacova [b], B. Buonomo [c], N. Busca [a], L. Cazon [a], S.D. Chemerisov [d], M.E. Conde [d], R.A. Crowell [d], P. Di Carlo [e], C. Di Giulio [f], M. Doubrava [g], A. Esposito [c], P. Facal [h], F.J. Franchini [d], J. Hörandel [i,1], M. Hrabovsky [b], M. Iarlori [e], T.E. Kasprzyk [d], B. Keilhauer [i], H. Klages [j], M. Kleifges [k], S. Kuhlmann [d], G. Mazzitelli [c], L. Nozka [b], A. Obermeier [i,*], M. Palatka [b], S. Petrera [e], P. Privitera [f], J. Ridky [b], V. Rizi [e], G. Rodriguez [f], F. Salamida [e], P. Schovanek [b], H. Spinka [d], E. Strazzeri [f], A. Ulrich [l], Z.M. Yusof [d], V. Vacek [g], P. Valente [m], V. Verzi [f], T. Waldenmaier [j], AIRFLY Collaboration

[a] Enrico Fermi Institute, University of Chicago, 5640 S. Ellis Ave., Chicago, IL 60637, USA
[b] Institute of Physics of the Academy of Sciences of the Czech Republic, Na Slovance 2, CZ-182 21 Praha 8, Czech Republic
[c] Laboratori Nazionali di Frascati dell'INFN, INFN, Sezione di Frascati, Via Enrico Fermi 40, Frascati, Rome 00044, Italy
[d] Argonne National Laboratory, Argonne, IL 60439, USA
[e] Dipartimento di Fisica dell'Università de l'Aquila and INFN, Via Vetoio, I-67010 Coppito, Aquila, Italy
[f] Dipartimento di Fisica dell'Università di Roma Tor Vergata and Sezione INFN, Via della Ricerca Scientifica, I-00133 Roma, Italy
[g] Czech Technical University, Technicka 4, 16607 Praha 6, Czech Republic
[h] Departamento de Física de Partículas, Campus Sur, Universidad, E-15782 Santiago de Compostela, Spain
[i] Institut für Experimentelle Kernphysik (IEKP), Universität Karlsruhe (TH), Postfach 6980, D-76128 Karlsruhe, Germany
[j] Forschungszentrum Karlsruhe, Institut für Kernphysik, Postfach 3640, D-76021 Karlsruhe, Germany
[k] Forschungszentrum Karlsruhe, Institut für Prozessdatenverarbeitung und Elektronik, Postfach 3640, D-76021 Karlsruhe, Germany
[l] Physik Department E12, Technische Universität Muenchen, James Franck Str. 1, D-85748 Garching, Germany
[m] Sezione INFN di Roma 1, Ple. A. Moro 2, I-00185 Roma, Italy

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ABSTRACT

The knowledge of the fluorescence emission as a function of atmospheric parameters is essential for the detection of extensive air showers with the fluorescence technique. In this paper, we summarize AIRFLY published measurements of the pressure dependence of the fluorescence yield. The spectral distribution of the fluorescent light between 280 and 429 nm has been measured with high resolution. Relative intensities of 34 spectral lines have been determined. The pressure dependence of 25 lines was measured in terms of quenching reference pressures p'_λ in air. This set of AIRFLY measurements yields the most comprehensive parametrization of the pressure dependence of the fluorescent spectrum.

1. Introduction

The fluorescence detection of ultra high energy cosmic rays is a well-established technique [1]. It allows the measurement of the longitudinal development of extensive air showers (EAS) in the atmosphere, thus providing a nearly calorimetric measurement of the energy as well as information on the mass of the primary particle. Charged particles, mainly $e^{\pm}$, of EAS excite the nitrogen molecules along their path in the atmosphere. Radiative de-excitation results in isotropic emission of fluorescence light in the 300–400 nm range. Fluorescence photons are attenuated along their path towards the fluorescence telescopes by wavelength-dependent scattering of the fluorescence light in the atmosphere. Also, the signal measured by fluorescence detectors is affected by the spectral dependence of the photomultipliers and broad-band filter used to detect the fluorescence light. Thus, the knowledge of the integrated fluorescence yield between 300 and 400 nm is not sufficient, and the relative intensities of air fluorescence bands must be known at every point along the EAS, that is, at different pressures and temperatures. Since the energy of EAS charged particles ranges from keV to GeV [2], fluorescence yield measurements over a wide range of energies should be performed.

The AIRFLY (AIR FLuorescence Yield) experiment is pursuing an extensive program of fluorescence yield measurements with electrons of keV to GeV energy at several accelerators. Preliminary results on the fluorescence yield dependence on temperature and humidity [3], as well as on the energy of the electron

* Corresponding author.
E-mail address: andreas.obermeier@ik.fzk.de (A. Obermeier).
[1] Now at Department of Astrophysics, Radboud University Nijmegen, Nijmegen, The Netherlands.

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which excites the nitrogen molecules [4] are reported elsewhere. The status of the measurement of the absolute yield of the 337 nm band is also presented at this Workshop [5]. In this paper, we summarize our published results [6] on the pressure dependence of the fluorescence spectrum.

Previous experiments [7,8] employed interference filters to isolate spectral lines, making use of sophisticated procedures to disentangle multiple lines within the filter range, or had a coarse spectral resolution [9]. AIRFLY has measured the air fluorescence spectrum in the range 280–430 nm with high resolution. Measurements with a grating spectrograph at the Chemistry electron Van de Graaff accelerator have been combined with precise photomultiplier (PMT) measurements of the most prominent spectral line at 337 nm at the Argonne Wakefield Accelerator. Both accelerators are based at the Argonne National Laboratory, near Chicago, USA. The gas was controlled in a cylindrical chamber with quartz windows that allowed the fluorescent light to be detected by the spectrograph or PMT. A detailed account of the experimental method is given in Ref. [6].

2. Fluorescence yield

Electrons passing through air excite molecular nitrogen directly as well as through secondary electrons produced along their path. This excitation is assumed to be proportional to the energy deposited in the gas. Excited nitrogen molecules then either radiate fluorescent light or relax due to quenching, e.g. intermolecular collisions. A more detailed description of the fluorescence process is given in Ref. [10].

Both the collisional quenching and the energy deposited by electrons are dependent on the gas density, and thus on the pressure p. The number of fluorescence photons emitted by a given volume of gas can be written [6] in its simplest form as

$$N_{\text{gas}} \propto E_{\text{dep}}^{\text{gas}} Y_{\text{gas}}(p) \propto p \cdot \frac{f_{\text{N}_2}}{1 + \dfrac{p}{p'_{\text{gas}}}} \tag{1}$$

where $Y_{\text{gas}}(p)$ is the fluorescence yield (measured in photon/MeV of energy deposited), f_{N_2} is the fraction of nitrogen molecules (100% for N_2 and 79% for air) and p'_{gas} is the collisional quenching reference pressure.

The parameter p' is defined as the pressure at which collisional quenching and fluorescent emission are equally likely. It can be expressed as

$$p'_{\text{N}_2} = \frac{\sqrt{\pi m_{\text{N}_2} kT}}{4\tau_0 \sigma_{\text{NN}}} \tag{2}$$

with the molecular mass of nitrogen m_{N_2}, the Boltzmann constant k, the thermodynamic temperature T, the lifetime of the excited state τ_0 and the nitrogen–nitrogen collisional cross section σ_{NN}. This model for fluorescence yield and quenching reference pressure is the most simple one, neglecting internal molecular processes and three body quenching processes. For the sake of simplicity, collisional cross sections are here considered constant.

In air, excited nitrogen molecules will also suffer collisions with oxygen molecules. The effective quenching reference pressure in air may then be written as [6]

$$\frac{1}{p'_{\text{air}}} = \frac{0.79}{p'_{\text{N}_2}} + \frac{0.21}{p'_{\text{O}_2}} \tag{3}$$

for an air mixture made of 79% nitrogen and 21% oxygen. The parameter p'_{O_2} has the same dependencies of Eq. (2), with σ_{NN} replaced by the nitrogen–oxygen collisional cross section σ_{NO}.

In this paper, we will not interpret the measurements in terms of collisional cross sections, but rather use the quenching reference pressure for each band as a phenomenological description of the data. On the other hand, Eq. (2) provides useful predictions that may be tested by our measurements. The quenching reference pressure p' is inversely proportional to the lifetime of the excited state, which does not depend on properties of the final state. Thus, band heads that share the initial state should have the same quenching reference pressure, assuming no contamination of lines by nearby transitions.

3. The fluorescence spectrum

The air fluorescence spectrum was measured at the Argonne Chemistry Van de Graaff facility. The accelerator provided a DC current of $\approx 10\,\mu$A of 3 MeV electrons. The fluorescent light was focused by a mirror into an optical fiber, and then recorded by the spectrograph. Lead shielding around the fluorescence chamber and the spectrograph reduced background. A very low background level in the measured fluorescence spectrum was determined in wavelength regions without spectral lines and subtracted.

Fig. 1 shows the highly resolved fluorescence spectrum as recorded by AIRFLY. Major band heads are labeled according to their electronic transition, initial, and final vibrational state. The area under the spectrum is normalized to unity. Relative intensities, I_λ, of all observed band heads in air at 800 hPa and 293 K, normalized to 2P(0,0), are given in Table 1. The spectrum

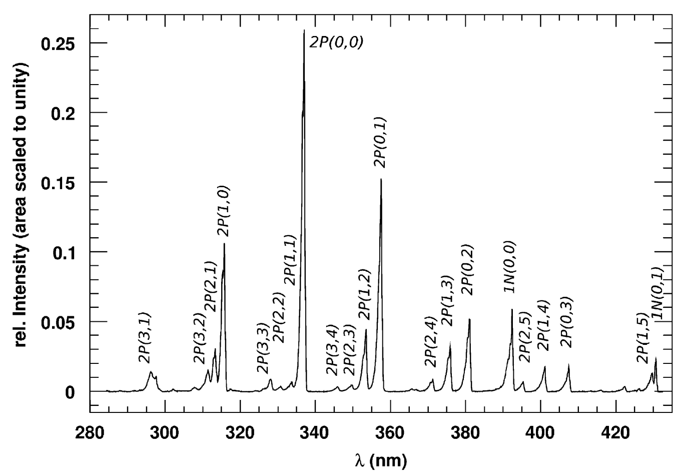

Fig. 1. Air fluorescence spectrum excited by 3 MeV electrons at 800 hPa. Labels indicate 21 major transitions.

Table 1
Relative intensities of observed band heads in air at 800 hPa and 293 K

Trans.	λ (nm)	I_λ (%)	Trans.	λ (nm)	I_λ (%)
2P(3,1)	296.2	5.16 ± 0.29	GH(0,5)	366.1	1.13 ± 0.08
2P(2,0)	297.7	2.77 ± 0.13	2P(3,5)	367.2	0.54 ± 0.04
GH(6,2)	302.0	0.41 ± 0.06	2P(2,4)	371.1	4.97 ± 0.22
GH(5,2)	308.0	1.44 ± 0.10	2P(1,3)	375.6	17.87 ± 0.63
2P(3,2)	311.7	7.24 ± 0.27	2P(0,2)	380.5	27.2 ± 1.0
2P(2,1)	313.6	11.05 ± 0.41	2P(4,7)	385.8	0.50 ± 0.08
2P(1,0)	315.9	39.3 ± 1.4	GH(0,6)	387.7	1.17 ± 0.06
GH(6,3)	317.6	0.46 ± 0.06	1N(1,1)	388.5	0.83 ± 0.04
2P(4,4)	326.8	0.80 ± 0.08	1N(0,0)	391.4	28.0 ± 1.0
2P(3,3)	328.5	3.80 ± 0.14	2P(2,5)	394.3	3.36 ± 0.15
2P(2,2)	330.9	2.15 ± 0.12	2P(1,4)	399.8	8.38 ± 0.29
2P(1,1)	333.9	4.02 ± 0.18	2P(0,3)	405.0	8.07 ± 0.29
2P(0,0)	337.1	100	2P(3,7)	414.1	0.49 ± 0.07
GH(0,4)	346.3	1.74 ± 0.11	2P(2,6)	420.0	1.75 ± 0.10
2P(2,3)	350.0	2.79 ± 0.11	1N(1,2)	423.6	1.04 ± 0.11
2P(1,2)	353.7	21.35 ± 0.76	2P(1,5)	427.0	7.08 ± 0.28
2P(0,1)	357.7	67.4 ± 2.4	1N(0,1)	427.8	4.94 ± 0.19

All lines are normalized to the 2P(0,0)-transition at 337.1 nm.

was calibrated relative to an NIST-traceable quartz tungsten halogen lamp, with an uncertainty of 3%.

The good spectrograph resolution allowed the identification of 34 bands, belonging to three electronic transitions of molecular nitrogen; namely the "2P" (second positive) system of N_2, the "1N" (first negative) system of N_2^+ and the low intensity "GH" (Gaydon-Herman) system of N_2. Notice that the measured lines are widened and present a tail towards lower wavelengths, due to the rotational substructure and the experimental resolution. Some overlap between nearby lines is thus present. Since the accuracy of the spectral measurement is sufficient for the purpose of application to EAS detection, we did not attempt to disentangle overlapping bands. A comparison of the measured relative intensities to theoretical prediction based on ratios of Einstein coefficients yields very good agreement [6]. Furthermore, a similar measurement with 1% of argon added to the gas mixture revealed no difference. This means that argon has no effect on the fluorescence spectrum within AIRFLY's accuracy.

4. Pressure dependence of 2P(0,0)-transition

The intensity of the 2P(0,0)-transition was measured in nitrogen and air at 293 K in the pressure range from 2 to 1000 hPa. The fluorescence light was measured by a PMT, after passing through an interference filter. The filter isolates the 337 nm band head with a contamination of nearby spectral lines of about 1.7%. Because of their similar quenching reference pressures, their effect is negligible.

For each pressure scan, background measurements have been taken by closing a shutter in front of the PMT, and the background level was monitored by a second PMT during measurements.

Measurements were performed at the Argonne Wakefield Accelerator with 14 MeV beam energy, which was operated in pulsed mode with wide fluctuations in the bunch charge. The charge of each electron bunch was measured by an integrating current transformer (ICT). A typical measurement in nitrogen is shown in Fig. 2. The slope S_{FL} of the correlation of the PMT and ICT signals was used as a relative estimator of the fluorescence yield:

$$S_{FL,gas} \propto N_{gas} \propto \frac{p}{1 + p/p'}. \tag{4}$$

Notice that the fluorescence bands are mainly excited by secondary electrons produced in the gas by the beam electrons.

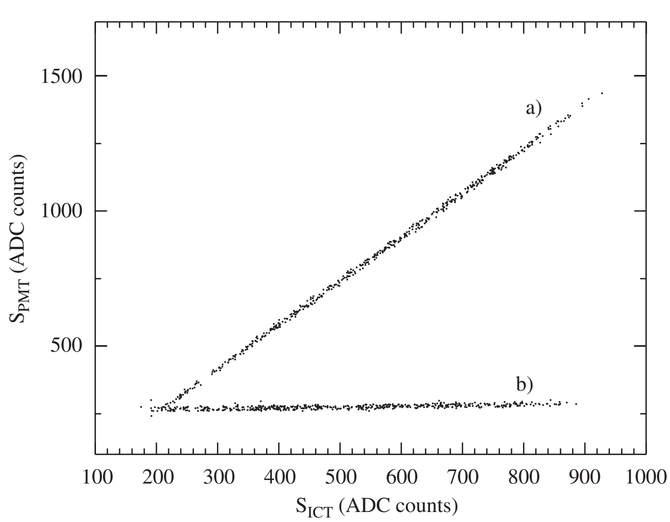

Fig. 2. Correlation of PMT and ICT signals for a fluorescence run (a) and a background run (b) in nitrogen at 180 hPa.

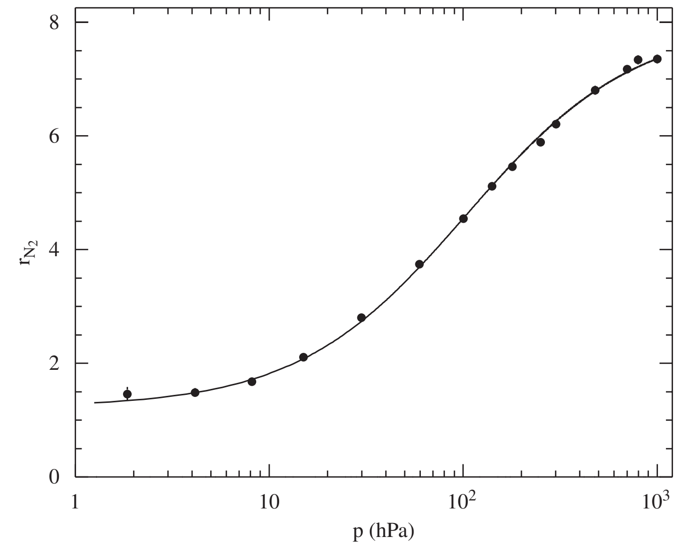

Fig. 3. Ratio of nitrogen to air signals as a function of pressure, together with the fit used to measure p'_{air} of the 337.1 nm band.

Decreasing the pressure, they eventually are able to leave the field of view of the PMT and part of the fluorescence emission is not detected. Neglecting the losses due to the secondary electrons escaping the field of view would cause an overestimation of the quenching reference pressure [11]. This bias is equally affecting the nitrogen and air measurements, so it can be eliminated by taking the ratio of the two measurements (cf. Eq. (3)):

$$r_{FL,N_2} = \frac{S_{FL,N_2}}{S_{FL,air}} = \frac{A}{0.79} \cdot \frac{1 + p[0.79/p'_{N_2} + 0.21/p'_{O_2}]}{1 + p/p'_{N_2}} \tag{5}$$

where A is a constant close to unity [6].

In Fig. 3, the measured r_{FL,N_2} is shown, together with a fit of Eq. (5) to the data. We obtained

$$p'_{N_2} = 103.7 \pm 5.0\,\text{hPa} \tag{6}$$

$$p'_{O_2} = 3.796 \pm 0.19\,\text{hPa} \tag{7}$$

from which the air quenching reference pressure is derived:

$$p'_{air} = 15.89 \pm 0.73\,\text{hPa}. \tag{8}$$

The quoted uncertainty includes statistical and systematic contributions. A direct fit of the pressure dependence in air using a correction estimated by Monte Carlo simulations of the effect of escaping secondary electrons yielded a compatible result within the uncertainty.

Also, including 1% of argon in the gas mixture had no detectable effect on the reference pressure.

5. Pressure dependence of other band heads in air

The pressure dependence of spectral bands other than the 2P(0,0) were performed by taking spectra at different air pressures with the same apparatus of Section 3.

The relative intensity of a spectral line at wavelength λ to 2P(0,0) is given by

$$I_\lambda = C \cdot \frac{1 + p/p'_{air}(337)}{1 + p/p'_{air}(\lambda)} \tag{9}$$

where C is the intensity ratio in the limit of $p \to 0$, and p'_λ is the quenching reference pressure of the line. The quenching reference pressure $p'_{air}(337)$, given in Section 4, is used here.

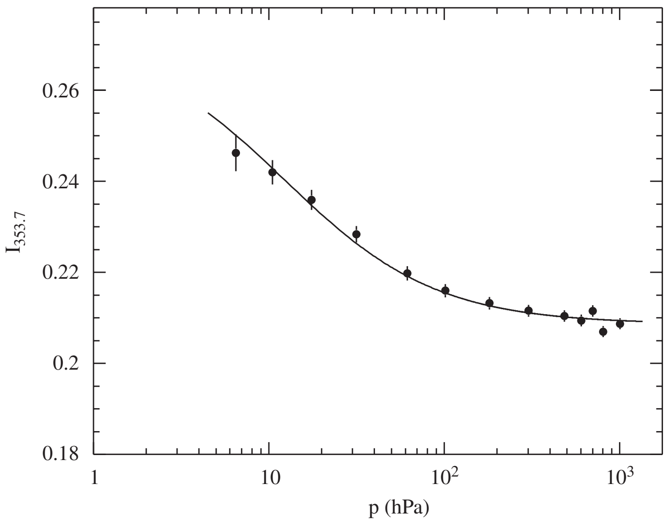

Fig. 4. Relative intensity $I_{353.7}$ of the 2P(1,2)-transition as a function of pressure. The full line represents the best fit to p'_{air} (353.7).

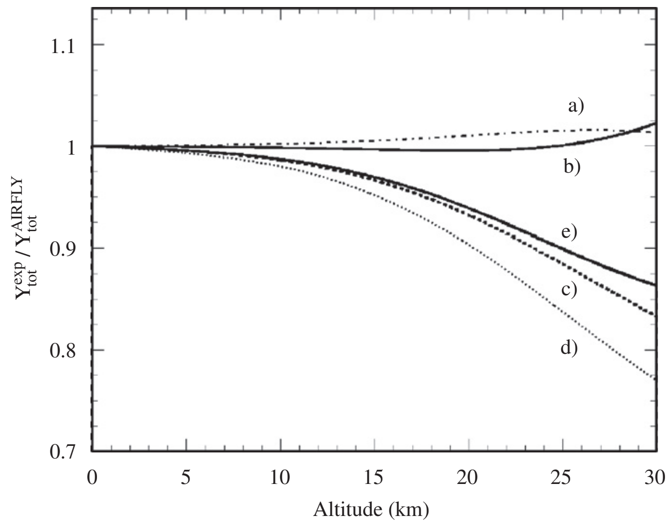

Fig. 5. Ratio of the total fluorescence yield in the range 300–400 nm as measured by other experiments to the one measured by AIRFLY as a function of altitude: (a) Belz et al., (b) Bunner, (c) Nagano et al., (d) Kakimoto et al., (e) Colin et al.

Table 2
Collisional quenching reference pressures in dry air at 293 K

Band	λ (nm)	$p'_{\mathrm{air}}(\lambda)$ (hPa)
2P(0,0)	337.1	15.89 ± 0.73
2P(0,1)	357.7	$15.39 \pm 0.25 \pm 0.72$
2P(0,2)	380.5	$16.51 \pm 0.48 \pm 0.72$
2P(0,3)	405.0	$17.8 \pm 1.5 \pm 0.8$
2P(1,0)	315.9	$11.88 \pm 0.31 \pm 0.62$
2P(1,1)	333.9	$15.5 \pm 1.5 \pm 0.7$
2P(1,2)	353.7	$12.70 \pm 0.34 \pm 0.64$
2P(1,3)	375.6	$12.82 \pm 0.45 \pm 0.62$
2P(1,4)	399.8	$13.6 \pm 1.1 \pm 0.6$
2P(1,5)	427.0	$6.38 \pm 0.68 \pm 0.43$
2P(2,0)	297.7	$17.3 \pm 4.0 \pm 0.8$
2P(2,1)	313.6	$12.27 \pm 0.78 \pm 0.64$
2P(2,2)	330.9	$16.9 \pm 3.5 \pm 0.76$
2P(2,3)	350.0	$15.2 \pm 3.7 \pm 0.7$
2P(2,4)	371.1	$14.8 \pm 1.9 \pm 0.7$
2P(2,5)	394.3	$13.7 \pm 3.3 \pm 0.7$
2P(2,6)	420.0	$13.8 \pm 4.0 \pm 0.7$
2P(3,1)	296.2	$18.5 \pm 5.0 \pm 0.8$
2P(3,2)	311.7	$18.7 \pm 3.8 \pm 0.8$
2P(3,3)	328.5	$20.7 \pm 2.6 \pm 0.8$
1N(0,0)	391.4	$2.94 \pm 0.58 \pm 0.31$
1N(0,1)	427.8	$2.89 \pm 0.64 \pm 0.30$
1N(1,1)	388.5	$3.9 \pm 1.7 \pm 0.3$
GH(0,4)	346.3	$21 \pm 10 \pm 1$
GH(0,6)	387.7	$7.6 \pm 1.6 \pm 0.5$

Quoted uncertainties are statistical and the propagated uncertainty of $p'_{\mathrm{air}}(337)$, respectively.

The pressure dependence of the spectrum lines was fitted with Eq. (9), and good agreement between model and data was found. As an example, the measured $I_{353.7}$ for the 2P(1,2)-transition is shown in Fig. 4. The deviation from a constant line reflects the difference in the quenching reference pressures.

Notice that the calibration of the spectral response of the spectrograph only affects the fitted value of the parameter C. This reduces systematic uncertainties, although the uncertainty of p'_{337} needs to be propagated. The measured quenching reference pressures are summarized in Table 2. Due to limited counting statistics, the reference pressures of nine weak lines could not be measured. The measured reference pressure of spectrum lines that share the same initial state and whose contamination by other lines is small are similar. This confirms the expectation of Section 2.

6. Discussion

The set of AIRFLY measurements summarized in this paper yields the most comprehensive parametrization of the pressure dependence of the fluorescent spectrum. A detailed discussion on the application of AIRFLY data to the fluorescence detection technique of EAS is found in Ref. [6].

The AIRFLY spectrum is significantly more precise compared to prior experiments, which were limited by coarser spectrograph resolution [9] or relied on optical filters [7,8]. For comparison with previous measurements, the total fluorescence yield between 300 and 400 nm can be used, because this was also measured by Refs. [12,13] with broad-band optical filters. The ratio of the total fluorescence yield in the range 300–400 nm as measured by other experiments to the one measured by AIRFLY is shown as a function of altitude in Fig. 5. All yields were normalized to have the same value at ground level. The total yields of Bunner [9] and Belz et al. [12] are in very good agreement with AIRFLY, but Kakimoto et al. [7], Nagano et al. [8], and Colin et al. [13] present significant deviations, which reflect the systematically higher values of p' measured by these experiments.

7. Conclusions

A high-resolution measurement of the fluorescence spectrum of molecular nitrogen excited by 3 MeV electrons has been made in dry air. The intensities of 34 clearly identified spectral lines have been reported and were found to be in good agreement with theoretical predictions using Einstein coefficients.

The pressure dependence of the fluorescence spectrum in dry air has been measured from a few hPa to atmospheric pressure. Particular care was taken to avoid the bias introduced by secondary electrons escaping the field of view, by using ratios of measured intensities as a function of pressure. Quenching reference pressures of spectral lines with the same initial molecular state are found to be the same within uncertainties.

The effect of argon has been found to be negligible.

The high-resolution spectra recorded by the spectrograph allowed many more closely spaced bands to be separated than in previous experiments. In the ratio of fluorescence intensities, which was extensively used, many systematic uncertainties cancel. Thanks to these improvements in the experimental method, the details and precision of the AIRFLY measurements surpass that of previous experiments.

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