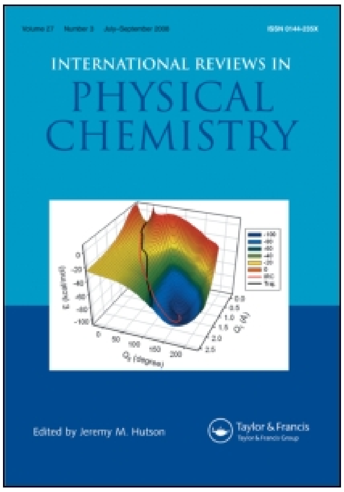

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Detection methods for atoms and radicals in the gas phase

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Detection methods for atoms and radicals in the gas phase

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This report lists atoms and free radicals in the gas phase which are of interest for environmental and flame chemistry and have been detected directly. The detection methods which have been used are discussed with respect to their range of application, specificity and sensitivity. In table 1, detection methods for the five atoms of group IV (C, Si, Ge, Sn, Pb) and about 60 radicals containing at least one atom of group IV are summarized

(CH, CD, CF, CCl, CBr, CN, CS, CSe, CH_2, CD_2, CHF, CDF, CHCl, CHBr, CF_2, CCl_2, CBr_2, CFCl, CFBr, CH_3, CD_3, CF_3, CH_2F, CH_2Cl, CH_2Br, CHF_2, $CHCl_2$, $CHBr_2$, HCO, FCO, CH_3O, CD_3O, CH_2OH, CH_3S, NCO, CH_4N, CH_3O_2, CF_3O_2;
C_2, C_2N, C_2H, C_2O, C_2HO, C_2H_3, C_2F_3, C_2H_5, C_2H_5O, C_2H_4OH, CH_3CO, CD_3CO, C_2H_3O, $C_2H_5O_2$, CH_3COO_2, C_2H_4N, C_2H_6N, C_3;
Si, SiF, SiF_2, SiO, SiC, Si_2;
Ge, GeC, GeO, GeF, GeF_2, $GeCl_2$, Sn, SnF, SnO, SnF_2, Pb, PbF, PbF_2, PbO, PbS).

In table 2 detection methods for about 25 other atoms and 60 radicals are listed:

(H, D, O, O_2, OH, OD, HO_2, DO_2, F, Cl, Br, I, FO, ClO, BrO, IO, FO_2, ClO_2, Li, Na, K, Rb, Cs, N, N_3, NH, ND, NF, NCl, NBr, NH_2, ND_2, NHD, NHF, NF_2, NCl_2, N_2H_3, NO, NO_2, NO_3, HNO, DNO, P, PH, PD, PF, PCl, PH_2, PD_2, PF_2, PO, As, AsO, AsS, Sb, Bi, S, S_2, SH, SD, SF, SF_2, SO, HSO, DSO, SN, Se, Te, Se_2, SeH, SeD, SeF, SeO, SeS, SeN, TeH, TeO, BH, BH_2, BO, BN, BO_2, Cd, Hg, UF_5).

The tables also cite some recent kinetic applications of the various methods.

1. Introduction

The detection of free radicals in the gas phase was first achieved by Paneth and Hofeditz in 1929 [1]. They showed that the reflecting surface produced by the thermal dissociation of Pb $(CH_3)_4$ or Zn $(CH_3)_2$ disappeared when the metal surfaces were exposed to the dissociation products from Pb $(CH_3)_4$ at some distance from the heated area; in the case of a Zn-film, Zn $(CH_3)_2$ was formed. In addition the authors were able to conclude from the experiments with their flow system that the free radicals produced had a short lifetime.

Since those early days, detection methods have improved substantially. Sensitive and specific detection methods were needed to study combustion processes and other reactive systems in detail. Most of the important elementary reactions in these systems involve radicals. Their production and depletion rates are high and their concentrations are small, so sensitive detection methods are required. Also products of elementary reactions do not have a Boltzmann distribution with respect to the surrounding temperature. The initial product state populations are not only of interest in producing inversion in chemical reactions but also for the detailed description of the reacting systems. Therefore detection methods are not only required for the atom or radical but also for its quantum state. For this reason, for some radicals, detection methods are required for various states.

Since there are so many methods known for detecting free radicals, a review seems worth while. There are several reviews containing information about detection methods for atom reactions [2–4] in general or for reactions of atoms in electronically excited states[5]. The kinetics of various atoms (H [6, 7], O [8, 9], F [10, 11], Cl [11], N [12] and others) and radicals (OH [13–16], HO_2 [17], CH_3 [18] and others) are reviewed in the literature; those papers contain detailed descriptions for detecting at least the atom or radical under consideration. Some systems in which atom and radical reactions are of significant importance (e.g. atmospheric chemistry [19], combustion and flame chemistry [20, 21] or gas phase kinetics in general [22–25]) are discussed in the cited reviews. Also there are excellent descriptions of the various detection methods (e.g. ESR [26], LMR [27], Mass spectrometry [28], LASER, LIF, LRA [29]). Clyne has described methods to detect radicals and to measure their concentrations in flow systems in several articles [30].

2. Classification of detection methods

A species can be detected quantitatively if a signal which is specific for that species and a function of its concentration is obtained. An electrical signal is nearly always preferred since its further treatment is easiest.

A general tabulation of small radicals together with the methods applied in the past to determine their concentrations in the gas phase should be useful at the moment since in the last few years remarkable progress has been made in experimental techniques, due to new and improved laser light sources and due to the development of better molecular beam sampling systems in mass spectrometry. In particular the experimental advances in laser methods which allow observation of the system without disturbance are still going on with respect to time and spatial resolution; the possibilities seem to be far from finished.

The various methods used can be arranged in a systematic way by dividing them into two groups: (i) only the quantum state of the radical is changed, and (ii) the chemical nature of the radical is changed, during the detection process. In the following we list the techniques with respect to this classification.

2.1. *Radical detection by changing its quantum state*

The radical can be supplied with the energy to change its quantum state by energy transfer or by absorption of photons. The first process is useful for detection if subsequent emission occurs. The emission (E) which can be observed with spectral resolution is often used in high-temperature systems (flames, shock tubes) and to observe reactions in which the radicals of interest appear in excited states (Chemiluminescence (ChL)). Connected to the second process, the absorption of light in a wide spectral range from the vacuum ultraviolet (VUV) to the microwave (MW) region, provides a whole variety of detection methods.

The absorption methods (A) depend on the weakening of the light beam passing through the absorbing probe, which is described for sufficiently dilute probes by the Lambert–Beer law:

$$I(\nu) = I_0(\nu)\exp(-\sigma_\nu l) \qquad (1)$$

$I(\nu)$, $I_0(\nu)$ = light intensity after the distance l and at $l=0$ respectively
l = absorption length
σ_ν = absorption coefficient.

The absorption coefficient is a feature of the radical and characteristic for the two quantum states involved in the transition. The values of σ_ν determine whether an atom or radical can be detected by absorption at a particular concentration, with a particular experimental arrangement. The sensitivity will be discussed later.

A change in light by absorption can be converted into an electrical signal without severe problems. The absorption can be produced either with white light or with a resonance line (RA). The resonance either already exists from the choice of the light source (resonance lamp or monochromator) or is obtained by applying a magnetic (Zeeman-spectroscopy) or electric (Stark-spectroscopy) field. A technique based on the tuning of the absorption line of the paramagnetic radical by the Zeeman effect is laser magnetic resonance (LMR); the anologue for the Stark effect is laser electric resonance (LER). Absorption between the components of one state produced by Zeeman splitting is observed in electron spin resonance (ESR). For strong absorbers single-pass absorptions may be sufficient, but for weakly absorbing species multipass absorption devices or intracavity absorption (ICA) may be required.

As in the case of energy transfer mentioned above, the absorption can be detected indirectly by emission (fluorescence or phosphorescence). The observed fluorescence intensity is given by:

$$I_F = \chi \phi I_{abs} \tag{2}$$

χ = geometric factor
ϕ = fluorescence quantum yield
I_{abs} = absorbed light

in the absence of radiation trapping. The obvious necessity for appearance emission is that $\phi > 0$; i.e. the excited state must have a finite lifetime. If the emission is obtained from the state initially populated, it is often called resonance fluorescence (RF), and laser-induced fluorescence (LIF) if laser light is used. Light absorption and thus emission ($I_{abs} > 0$) is only obtained if the transition is allowed. With high intensity laser beams, two-photon processes can be induced. This technique known as 'two-photon absorption laser induced fluorescence' (TALIF) has among others the advantage that scattered laser light has a frequency which differs from that of the emission. Since the fluorescence quantum yield as given in equation (2) is reduced by quenching the fluorescence, techniques are normally applied in low-pressure systems ($p = 1$–10 mbar).

Quenching problems can be avoided by using laser-induced saturation fluorescence (LISF). If the power density in the irradiated volume is sufficiently high (10^5–10^8 W/cm^2 cm^{-1}), the optical transition is saturated or nearly saturated. The fluorescence intensity becomes independent of the laser intensity and the quenching rate [52]. This method has been used to measure concentrations of radicals like CH and CN in flames at atmospheric pressure [53–55].

The quenching problem can also be circumvented by using scattering instead of fluorescence. The Raman effect and even the resonance-Raman effect are generally not sensitive enough to measure radicals and are thus used mainly for stable molecules. However intense laser light sources allow non-linear Raman spectroscopy to be used; a comparison of spontaneous Raman scattering (RS), inverse Raman scattering (IRS) and coherent-anti-Stokes Raman scattering (CARS) for flame studies is discussed in

[62]. The CARS method will be described in more detail here since in practice it seems to be, among the Raman methods, the most important technique for studying radical concentrations. A probe which is collinearly irradiated by two laser beams with frequencies ω_1 (pump radiation) and ω_2 (Stokes radiation, $\omega_1 > \omega_2$), and which has a Raman active frequency $\omega_v(\omega_v = \omega_1 - \omega_2)$ will emit a higher frequency

$$\omega_3(\omega_3 = \omega_1 + \omega_v = 2\omega_1 - \omega_2)$$

which results from a coherent interaction between the molecular vibration at ω_v and the radiation at ω_1 and ω_2. The new emission (CARS) can be observed collinear with the pump beams in the forward direction.

CARS has been used to analyse flames [56] and the method is particularly useful for analysing rotational and vibrational temperatures [57]. The only radical detected so far by resonance-CARS is C_2 ($X\,^1\Sigma_g^+$) [58, 59].

2.2. *Radical detection by changing its chemical nature*

A radical can be converted into another species for which detection is easier and/or more sensitive by dissociation (e.g. $HO_2 \rightarrow OH + O$) or by chemical reaction (e.g. $HO_2 + NO \rightarrow OH + NO_2$). In both examples OH is detected rather than the HO_2-radical. Techniques of this kind are normally not included in table 1 or 2. If an excited species is produced in such a chemical reaction the emission can be used to observe the radical.

Another technique which is connected with a change in the chemical nature of a radical is the measurement of the heat released to a catalytic probe by radical recombination. This method is often used for atoms.

By far the most important process which is covered by this heading is the formation of ions from radicals which are then detected in a mass spectrometer (MS) or with other devices. Ionization by electron impact can be used as a primary process in mass spectrometry. The ionization cross-section may be significantly different for various particles and some are not ionized without fragmentation. This fragmentation is a problem in mass spectrometry as discussed in detail later (§ 3.2). Specific mass spectrometric detection techniques for radicals with focusing fields using their paramagnetic or polar properties have been developed [64, 65]. Photoionization (PI) and multiphoton ionization (MPI) provide other ways to avoid the fragmentation problem (§ 3.2).

Some other techniques using ions, which are not necessarily connected to a mass spectrometer, are surface ionization (SI) in a Langmuir–Taylor detector, the laser-optogalvanic absorption (LOG) which is related to ion formation by laser radiation, and photoelectron spectroscopy (PES) which is often used to obtain information about the quantum states of the radical rather than about its concentration.

In addition to these techniques, there are indirect detection methods such as freezing the gas-phase radicals into low-temperature matrices. This report deals with direct methods only. In the following sections the qualities of the various procedures will be described.

3. Comparison of techniques

The comparison of radical detection techniques is not an easy task. The analytic method applied in a specific experiment will depend on the experimental arrangement used but nevertheless we are not going to deal with experimental methods used for kinetic studies. The most important criteria of the kinetic system studied is whether stationary (e.g. in flow systems, flames, photolytic or pyrolytic systems) or non-stationary (e.g. in flash photolysis, shock tubes) radical concentrations are to be observed. Several of the methods given below are less useful for the latter application. For a specific radical the options are summarized in tables 1 and 2.

The comparison is done with respect to three characteristics of a method; whether it is *universal, specific, sensitive.*

These characteristics seem to be of special importance and the following description will be divided to highlight these features. It is easily accepted that it is a great advantage if the detection system is universal. The method should be specific for the species and if possible for the quantum state. A high sensitivity is of special importance for kinetic applications as pointed out above. Very often reliable information about a kinetic system can be obtained only if it is possible to vary the concentrations over a wide range. Another problem, not yet mentioned, is that of determining absolute concentrations and since a calibration may be needed, another good quality of a detection method is that an absolute calibration can be made easily. One might argue that the characteristics given here are not independent of each other but nevertheless they give helpful guidelines in discussing and comparing the various methods.

3.1. *Universality*

In principle all particles are detectable by mass spectrometry. This method can be regarded as universal provided that it is possible to get the representative sample into the ion source. This method is still universal if photoionization is used instead of ionization by electron bombardment. The surface ionization (SI) is mainly used for alkali and alkaline earth atoms and is not universal at all.

Optical detection methods in general require a more specific interaction between photon and radical or atom than does photoionization. Two quantum states are needed which are connected by a photon; moreover, if emission is expected instead of absorption a sufficiently high fluorescence quantum yield is required. In particular, for absorption the sensitivity will be sufficient only if a narrow-band light source with suitable frequency is available but, as mentioned above, the absorption frequency of the radical can be shifted by the Stark or Zeeman effect. Since light sources are available for the whole spectral range including the microwave region one would expect to find one, or even several, appropriate light sources; nevertheless the experimental arrangements required for a specific problem and the sensitivity can show a great variety.

The universality is reduced if special absorption arrangements are needed like LRA or ICA. The Zeeman absorption techniques ESR, LMR have been used for many interesting radicals. In particular, the number of radicals and even atoms detected by LMR is still growing rapidly. As already mentioned the absorption-emission techniques require that the fluorescence quantum yield is high enough; this restriction narrows the number of radicals for which these techniques are available.

In contrast to these methods which can be called universal or semi-universal, techniques like chemiluminescence and the conversion of radicals to species, as mentioned above, will always be restricted to special chemical systems. The CARS-technique which until now has been used for only a very few radicals may soon become applicable for more radicals.

The question of how universal a detection method is, can be answered by looking at tables 1 and 2. In these tables methods for atoms and small radicals are summarized. From these tables (which do not claim to be complete) one can see that mass spectrometry (MS) appears to be nearly universal; this would be even more evident if the larger hydrocarbon radicals (C_n; $n > 2$) had been included. Looking at other methods, such as laser induced fluorescence (LIF), one has to keep in mind that for two radicals to be detected with LIF, two different lasers and thus two different experimental arrangements may have to be used. This means that the method may seem to be much more universal than a specific experimental arrangement really is.

3.2. *Specificity*

For any analytical method it is necessary that the signal obtained is clearly correlated with the observed species. This seems to be a problem in mass spectrometry, since electron bombardment is unselective and produces fragments of all the particles initially present in the sample. This problem is severe for radicals since the same ions may be obtained by electron impact from the precursor molecules in the sample. Reduction of the electron energy to avoid fragmentation of the precursor also reduces the sensitivity. An even more complicated situation is obtained where highly excited radicals are produced in a reactive system. A solution to these problems can be seen in the photoionization technique (specific photoionization by one, two, three or multiphoton ionization) [31–35]. This method is specific in two ways and is thus often called two-dimensional mass spectrometry. The focusing field technique is another attempt to avoid the fragmentation problem. Surface ionization in a kind of Langmuir–Taylor arrangement is unspecific. This is also true for the catalytic probe (CP) technique.

Compared to MS, high specificity can be obtained easily by optical methods.

Specific detection by optical methods (e.g. A, RA, LRA, RF, LIF, LISF, TALIF, LMR, ESR) is possible only if the spectroscopy of all particles present in the probe is known and adequate monochromatic light sources are available. The absorption or fluorescence is then not only specific for the absorbing particle but also for the population of its quantum states. Thus there is the possibility of detecting the product energy distribution in chemical reactions. Zare *et al.* [36] used laser induced fluorescence to probe the population of quantum states in BaO produced in the reaction $Ba + O_2 \rightarrow BaO + O$. This technique has been applied to various other molecules and radicals. The rotational and vibrational population of OH in the reaction $H + NO_2$ was studied using the well-known transition $A\,^2\Sigma^+ - X\,^2\Pi$ [37]. The population distribution was used not only for concentration measurements but also to determine temperatures [38]. Chemiluminescence observed with sufficiently high spectral resolution is also highly specific.

Measurements of emission intensities in the infrared region (IR-chemiluminescence) have been used to determine rotational and vibrational energy distributions in the product of chemical reactions [39, 40]. The application of Raman and resonance Raman scattering as well coherent-anti-Stokes Raman scattering (CARS) appear to be of increasing importance for measuring product state distributions.

3.3. *Sensitivity*

As mentioned above, the sensitivity is the most important feature of a detection method. In this section we will give some estimates and comparisons of the sensitivity of the various methods. There is no doubt that the sensitivity resulting for one method depends on the characteristics of the particular species and so the figures given in this section can only give a rough idea of the sensitivity of a method. Detailed data about the sensitivity for some atoms and radicals are included in the tables.

In the mass spectrometer the pressure in the volume where the detection is performed (ion source) is in the order of 10^{-5} to 10^{-6} mbar. The reaction system which is to be analysed by mass spectrometry is often at a pressure of 10 to 1000 mbar. The ratio of the number densities in the reaction and detection volume are in the order of 10^{-6} to 10^{-9}. Nevertheless, the mass spectrometer is a sensitive method since the ionization and the detection of the ions formed is very sensitive. For example, in 1 mbar of He, less than 10^{-6} mbar CH_4 can be observed quantitatively. For a CH_3-radical, the detection limit is about 10^{-5} mbar in typical laboratory conditions if the CH_3 is produced in an excess of CH_4 in the reaction $F + CH_4$. If a CH_3-radical source is used which contains only a minor CH_4 mole fraction, e.g. $CH_2N_2 + H$, the mass spectrometric detection limit is more than an order of magnitude higher. This high sensitivity is due to high efficiency for the detection of ions. The same argument explains the extremely high sensitivity obtained by Langmuir–Taylor detectors, which are therefore successfully used in molecular beam experiments. Photoionization is also a technique which shows high sensitivity for the same reason.

For the optogalvanic detection method the authors claim they obtain a substantially greater sensitivity for O-atoms in an atmospheric pressure flame than Raman or two-photon laser induced fluorescence techniques [66]. The detection sensitivity is substantially better than 10^{17} atoms/cm^3 given for the spontaneous Raman scattering in the literature cited [68].

For optical methods, and particularly for those based on absorption, numerous experimental arrangements are in use. As can be seen from equation (1), a narrow-band monochromatic light source is needed to obtain high sensitivity; moreover the absorption length can be increased by using multipass arrangements (White cell). An absorber placed inside a laser cavity (intra-cavity absorption) changes the quality factor of the resonator if the laser frequency and the absorbing frequency match. Thus the laser intensity is sensitive to the absorbing probe; the enhancement in sensitivity compared to a single absorption can be several orders of magnitude. The relative change of the laser output has its maximum near the lasing threshold (limited by spontaneous emission in the direction of the laser axis) whereas the absolute change of the laser intensity is higher above the threshold. If linearity between the signal and the radical concentration and a low-noise laser is required, it is preferable to measure significantly off the laser threshold. The intra-cavity arrangement is used in LMR (laser magnetic resonance). The resonance between the laser line (CO, CO_2 or FIR-laser) and the radical absorption line is obtained by applying a magnetic field [41] (Zeeman effect) which is modulated to improve the sensitivity (comparable to NMR-spectrometers).

The sensitivities obtained by LMR are remarkably high (e.g. $[OH] \geqslant 10^8$, $[HO_2] \geqslant 10^9$ radicals/cm^3 at 1 mbar inert gas pressure [42, 43]). The spatial resolution is limited by the laser cross-section (about 1 cm for CO_2-laser, 3 cm for FIR-laser). Laser electric resonance (LER), where the resonance is obtained by an electrical field, is similar to LMR.

The intra-cavity absorption in multimode dye lasers has been known for more than ten years [44] but only a very few radicals have been detected by this method [45]. The sensitivity obtained can be compared with results attained with other multipass absorption arrangements.

Intra-cavity absorption is also used in electron spin resonance spectrometers, where the change in quality of a microwave cavity due to absorption of a paramagnetic probe is observed. The sensitivity obtained is about three orders of magnitude smaller than for LMR-spectrometers. ($[OH] \geqslant 8 \times 10^9$ radical/cm^3 at 1 mbar inert gas pressure

[26]). The spatial resolution is determined by the active volume of the cavity (about 3 cm^3). To improve the spatial resolution, special probing systems have been used which will not be discussed in detail here.

Another sensitive absorption technique, of increasing importance for chemical kinetic studies, is the Fourier-transform infrared absorption technique [46–50]. This method, which has mainly been used for high-resolution spectroscopy, will be used to study radical reactions in detail.

Extremely high sensitivities are reached by absorption-emission techniques. The resonance fluorescence (RF) and laser-induced fluorescence (LIF) are methods with remarkably good spatial resolution. For example, for OH and NH_2-radicals concentrations of 10^6 to 10^8 radicals/cm^3 at a total pressure of 1–10 mbar can be measured quantitatively in a cross-section of several mm diameter. The rapid development in laser techniques has changed the detection limits drastically; some years ago the sensitivity for Na-atoms was 10^2 atoms/cm^3 [51]. This limit is now at least two orders of magnitude lower. Information about the dynamics of the electronic excited state are also obtained by LIF since one can determine the collision-free lifetime as well as the quenching rates for the excited states involved. In systems where the nature and concentrations of quenchers is not known (e.g. flames), quenching turns out to be a severe problem since absolute calibrations become very complicated.

Laser-induced saturation fluorescence (LISF) is a very sensitive method, in particular in systems at high pressures. For OH-radicals, for example, in a premixed laminar flame in the pressure range $20 \leqslant p/\text{mbar} \leqslant 1011$ a detection limit to $10^{13}-10^{12}$ radicals/cm^3 was obtained [67].

The two-photon laser-induced fluorescence technique (TALIF) applied to O-atoms at a pressure of 13 mbar resulted in a sensitivity which was claimed to be considerably more sensitive than 10^{15}–10^{16} atoms/cm^3 [69].

The scattering techniques which—like the above mentioned LISF-techniques—have the advantage of working at higher pressure are not very sensitive compared to LIF (at low pressure). The sensitivity for O-atoms obtained by spontaneous Raman scattering in a hydrogen/oxygen flame and by coherent-anti-Stokes–Raman scattering in the same system is given as $[O] \simeq 10^{17}$ atoms/cm^3 [70]. The CARS sensitivity is limited by the non-resonant background and was insufficient for detecting other short-lived radicals. However, gas-phase CARS spectra of OH, NH_2 and CH_3 have been observed recently [63]. The sensitivity with available laser light sources is given 10 p.p.b. CO in air at standard conditions. A detailed description of the CARS method is given in [60, 61].

In particular, this last section shows that it is complicated to compare techniques. Even if the discussion is restricted to one property at a time, there are still aspects such as the different experimental conditions under which the method can be used.

This compilation indicates that the sensitivity of the various methods can differ significantly. The aims for the applications can vary drastically so that other properties, like long time stability, reproducibility, reliability or the possibility of calibration, become more important for the choice of technique.

4. Prospect

The development of analytical techniques is certainly not at an end; on the contrary, improvements in this field are very rapid, particularly since the laser technique is developing rapidly. More highly sensitive methods will be developed with good spatial and extreme time resolution, which will lead to new kinetic experiments in real time.

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Appendix

Compilation of various atoms and radicals and its detection methods

Detection methods for atoms and radicals are summarized in tables 1 and 2. The abbreviations used are explained at the end of table 2.

Table 1. Atoms of the fourth group of the periodic table and radicals containing at least one atom of this group.

Atom, radical	Method	Explanations	References
C (^3P_J)	RA	($^3P_J^0$–^3P_J) $\lambda = 165{\cdot}7$ or $156{\cdot}1$ nm	[131, 132]
($^1D_2$), ($^1S_0$)	RA	($^1P_1^0$–$^1S_0$) $\lambda = 247{\cdot}9$ nm	[133]
	LIF	($^1P^0$–$^1S_0$) $\lambda_F = 247{\cdot}8$ nm	[134, 148]
	LMR	($^3P_{0,1}$) FIR (CO_2)	[261]
	MS	(electron impact ionization) (C^+)	[3, 247]
CH, CD ($X\,^2\Pi_{3/2}$)	A	($C\,^2\Pi$–$X\,^2\Pi$) 314 nm λ (0, 0) = 315·66 nm λ (1, 1) = 314·41 nm	[136]
	LIF	($A\,^2\Delta$–$X\,^2\Pi$) 0, 0 band = 431 nm dye/N_2-laser	[137, 139]
	LISF	($A\,^2\Delta$–$X\,^2\Pi$) laser intensity 10^5–10^6 W/cm^2 cm^{-1}	[140]
	E	($A\,^2\Delta$–$X\,^2\Pi$) 430 nm O_2/C_2H_2-flame	[135]
	LMR	$(J'-J'') = 1$, $J' = 7/2, 5/2, 3/2$ CH_3OH (180·7 μm)–CH_2 CHCl (567·9 μm) FIR/CO_2-laser	[138]
	MS	(electron impact ionization) (CH^+, CD^+)	[4]
CF ($X\,^2\Pi_{1/2,3/2}$)	A–UV	($A\,^2\Sigma^+$–$X\,^2\Pi$) λ (1, 0) = 224·3 nm	[152]
	A–FIR DL	$X\,^2\Pi\Omega = 3/2, 1/2$ $(J+1 \leftarrow J)$ $17/2 \leqslant J \leqslant 23/2$, $\lambda = 330$ μm	[180, 153]
	E	($A\,^2\Sigma^+$–$X\,^2\Pi_r$) λ (0, 0) = 233·1 nm	[179]
	LMR	($\Omega = 3/2$, $J = 9/2$–$11/2$, CH_2CHBr (635·4 μm) FIR/CO_2-laser	[151]
	ESR	($J = 3/2$ and $J = 5/2$, $\Omega = 3/2$ (X-band)	[5, 150]
	MS	(electron impact ionization) (CF^+)	[3, 4, 186]
CCl ($X\,^2\Pi_{1/2,3/2}$)	A–UV	($^2\Delta_{3/2}$–$^2\Pi_{1/2}$) (Q_1, 0, 0) $\lambda = 277{\cdot}7$ nm (strong absorption band)	[155]
	LIF	($A\,^2\Delta$–$X\,^2\Pi$) λ (0, 0) = 278 nm dye/N_2-laser (doubled)	[154]
	E	($^2\Delta$)($A\,^2\Delta$–$X\,^2\Pi$) $293 \geqslant \lambda/\mathrm{nm} \geqslant 271$	[181]
	MS	(electron impact ionization) (CCl^+)	[3, 4]
CBr ($X\,^2\Pi_{1/2,3/2}$)	A–UV	($^2\Delta_{3/2}$–$^2\Pi_{1/2}$) (Q_1, 0, 0) $\lambda = 301{\cdot}45$ nm (strong absorption band)	[149]
	MS	(electron impact ionization) (CBr^+)	[3, 4]
CN ($X\,^2\Sigma^+$)	A	($B\,^2\Sigma^+$–$X\,^2\Sigma^+$) (P, 0, 0) 388·34 nm ($0 \leqslant V'' \leqslant 7$)	[157, 158]
	LIF	($A\,^2\Pi$–$X\,^2\Sigma^+$) (0, 0), (0, 1) $570 \leqslant \lambda/\mathrm{nm} \leqslant 610$ dye/N_2-laser	[159, 160]
	LISF	($B\,^2\Sigma^+$–$X\,^2\Sigma^+$)	[331]
	E	($^2\Sigma^+$), C (^3P) + N (^4S) + M $\rightarrow$ CN ($B\,^2\Sigma^+$) + M	[156, 161]
	MS	(electron impact ionization) (CN^+)	[3, 4]

Atom, radical	Method	Explanations	References
CS ($X\,^1\Sigma^+$)	A–UV	($A\,^1\Pi$–$X\,^1\Sigma^+$) (Q, 0, 0) $\lambda = 257{\cdot}675$ nm	[163]
	LIF	($A\,^1\Pi$–$X\,^1\Sigma^+$) (0, 0) $\lambda = 275{\cdot}5$ nm dye/N_2-laser (doubled)	[162, 165, 166]
	E	(UV) ($A\,^1\Pi$–$X\,^1\Sigma^+$) CS $A\,^1\Pi$ from discharge in CS_2	[164]
	MS	(electron impact ionization) (CS^+)	[3, 4]
CSe ($D\,^1\Pi$)	E	($D\,^1\Pi$–$X\,^1\Sigma^+$) $275 \leqslant \lambda/\mathrm{nm} \leqslant 305$	[332]
		CSe ($D\,^1\Pi$ from photodissociation of CSe_2)	[315]
CH_2, CD_2 ($\tilde{X}\,^3B_1$)	A–UV	($\tilde{B}$–$\tilde{X}$) $\lambda = 141{\cdot}5$ nm	[171]
	LMR	($\tilde{X}\,^3B_1$) CH_3OH (163·0 μm) FIR/CO_2-laser sensitivity:	[169, 170]
		$[CH_2(\tilde{X}\,^3B_1)] \geqslant 1{\cdot}7 \times 10^{-15}$ mol/cm^3	[396]
	MS	(electron impact ionization) (CH_2^+)	[4, 256]
CH_2, CD_2 ($\tilde{a}\,^1A_1$)	A–VIS–UV	($\tilde{b}\,^1B_1$–$\tilde{a}\,^1A_1$) $500 \leqslant \lambda/\mathrm{nm} \leqslant 900$	[168]
	LIF	($\tilde{b}\,^1B_1$–$\tilde{a}\,^1A_1$) $563 \leqslant \lambda/\mathrm{nm} \leqslant 568$ $544 \leqslant \lambda/\mathrm{nm} \leqslant 549$, dye/$N_2$-laser	[167, 172]
CHF, CDF ($\tilde{X}\,^1A_1$)	A–VIS	($\tilde{A}\,^1A''$–$\tilde{X}\,^1A'$) (0, 0, 0)–(0, 0, 0) $\lambda = 579$ nm	[175]
	LIF	($\tilde{A}\,^1A''$–$X\,^1A'$) $574 \leqslant \lambda/\mathrm{nm} \leqslant 582$, dye/$N_2$-laser	[173, 176, 177]
	ChL	$F + CH_4$, (CD_4) ($\tilde{A}\,^1A''$); $O + C_2HF_3$($\tilde{A}\,^1A''$)	[178]
CHCl, CHBr ($\tilde{X}\,^1A'$)	A–VIS	$\tilde{A}\,^1A''$–$\tilde{X}\,^1A'$ $820 \geqslant \lambda/\mathrm{nm} \geqslant 550$	[337]
CF_2 ($\tilde{X}\,^1A_1$)	A–UV	($^1B_1$–$^1A_1$), $300 \geqslant \lambda/\mathrm{nm} \geqslant 220$ at 1600 K, $\lambda = 249$ nm at 300 K	[152, 182, 189]
	A–IR–DL	(^RQ branch of $\nu_1\tilde{X}\,^1A_1$); multipass absorption (White-cell)	[185]
	RF	($\tilde{A}\,^1B_1(0,4,0)$–$\tilde{X}\,^1A_1(0,0,0)$) $\lambda = 253{\cdot}7$ nm Hg-lamp	[187]
	LIF	($\tilde{A}\,^1B_1$–$\tilde{X}\,^1A_1$) $\lambda\,(0) = 268{\cdot}74$ nm dye/N_2-laser (doubled)	[174]
	E	($^3B_1$) $O + C_2F_4$ ($\tilde{a}\,^3B_1$) $\lambda_E = 560$ nm	[188]
	MS	(electron impact ionization) (CF_2^+)	[3, 4, 186]
CCl_2, CBr_2 ($\tilde{X}\,^1A_1$)	LIF	(Cl) ($\tilde{A}\,^1B_1$–$\tilde{X}\,^1A_1$) $\lambda/\mathrm{nm} = 515$; 437, dye-laser	[154, 183]
	MS	(electron impact ionization) (CCl_2^+, CBr_2^+)	[3, 4]
CFCl ($\tilde{X}\,^1A'$)	LIF	($\tilde{A}\,^1A''$–$\tilde{X}\,^1A'$) $\lambda/\mathrm{nm} = 379{\cdot}4$; 384·8, dye-laser	[183, 190]
CFBr ($\tilde{X}\,^1A'$)	LIF	($\tilde{A}\,^1A''$–$\tilde{X}\,^1A'$) $400 \leqslant \lambda/\mathrm{nm} \leqslant 460$ dye-laser	[184]
CH_3, CD_3 ($\tilde{X}\,^2A_2''$)	A–UV	($\tilde{B}\,^2A_1'$–$\tilde{X}\,^2A_2''$) $\lambda = 216$ nm	[141, 142, 171, 392]
		$\varepsilon_{\mathrm{max}}$ (CD_3) $= 2{\cdot}0 \times 10^3$ l/mol cm $\varepsilon_{\mathrm{max}}$ (CH_3) $= 1{\cdot}8 \times 10^3$ l/mol cm at 1400 K	[143]
	E (IR)	ν_2 and ν_3 (antisym. stretching mode) at 3·1 μm, CH_3 from $Hg(CH_3)_2$ photolysis at 248, 193 nm	[146]
	MPI–MS	($\tilde{B}\,^2A_1'$–$\tilde{X}\,^2A_2''$) (0, 0) (266 nm Nd:YAG, 4. harm. + 215 nm)	[144]
	MS	(electron impact ionization) (CH_3^+, CD_3^+); photoionization Lyman α, Kr (MgF_2-window)	[3, 4, 145]
	CARS		[395]
CF_3 ($\tilde{X}\,^2A_1$)	A–UV	$200 \leqslant \lambda/\mathrm{nm} \leqslant 230$; $T = 1300$ K, $\lambda_{\mathrm{max}} = 218$ nm $\varepsilon_{218} = 57$ l/mol cm	[191]
	MPI–MS	$415 \leqslant \lambda/\mathrm{nm} \leqslant 490$ three-photon process	[192]
	MS	electron impact ionization, photoionization (CF_3^+)	[3, 4, 186, 259]
CH_2X (F, Cl, Br) ($\tilde{X}$)	LMR	CH_2F–N_2H_4 (301·3 μm), CH_2CHCl (567·9 μm) HCOOH (393·6 μm; 513 μm) FIR/CO_2-laser	[250]
	MS	(electron impact ionization) (CH_2F^+, CH_2Cl^+, CH_2Br^+)	[3, 4]

Atom, radical	Method	Explanations	References
CHX_2 (F, Cl, Br ($\tilde{X}$))	MS	(electron impact ionization) (CHF_2^+, $CHCl_2^+$, $CHBr_2^+$)	[3, 4]
HCO ($\tilde{X}\,^2A'$)	A–VIS–UV	($\tilde{A}\,^2A''$–$\tilde{X}\,^2A'$) $460 \leqslant \lambda/nm \leqslant 860$, ($\tilde{B}\,^2A'$–$\tilde{X}\,^2A'$) $210 \leqslant \lambda/nm \leqslant 260$ ε (230 nm) = 941 (l/mol cm)	[193, 198]
	A–IC	$\tilde{A}$–$\tilde{X}$(0, 9, 0)–(0, 0, 0) $\lambda = 613{\cdot}8$ nm (dye/flashl.-laser) (sensitivity not given)	[93, 203]
	A–LR	($\tilde{A}$–$\tilde{X}$) $\lambda = 614{\cdot}5$ nm, dye/N_2-laser	[204]
	LIF	$^2\tilde{A}'$(0, 0, 0)–$^2\tilde{A}''$(0, 9, 0) $638 \leqslant \lambda/nm \leqslant 664$, YAG dye-laser	[387]
	E	hydrocarbon flame $250 \leqslant \lambda/nm \leqslant 410$	[197, 340]
	LMR	MIR (ν_2, 5·3 μm CO laser) FIR (rot, 108 μm D_2O laser)	[194, 195]
	LER	(ν_3, 9·4 μm CO_2-laser, Stark field) ($10 \leqslant E/kV/cm \leqslant 60$)	[196]
	LOG	($\tilde{A}\,^2A''$–$\tilde{X}\,^2A'$) (0, 9, 0–0, 0, 0), 570–630 nm single-mode dye-laser	[341]
	ESR	(R-branch rotational states, X-band)	[5]
	MS	(electron impact ionization) (HCO^+), photoionization Lyman α (10·2 eV) MgF_2-window	[3, 4, (202), 258]
FCO ($\tilde{X}$)	A–IR DL	(ν_1 at 5 μm, with Zeeman modulation, multipass cell)	[201]
	E	$\lambda = 425$ nm FCO* from; O_3 + {*cis*, *trans* CHF=CHF, C_2HF_3, C_2ClF_3}	[199, 200]
CH_3O ($\tilde{X}\,^2E$)	A–UV	($\tilde{A}\,^2A_1$–$\tilde{X}\,^2E$) $280 \leqslant \lambda/nm \leqslant 320$ modulated Hg-sensitized dissociation of R–CO–OCH_3, R = H, CH_3	[207]
CD_3O ($\tilde{X}\,^2E$)	LIF	($\tilde{A}\,^2A_1$, $v_3' = 2$—$\tilde{X}\,^2E$, $v_3'' = 0$) $\lambda = 303{\cdot}9$ nm flashl. dye-laser (doubled)	[205, 210, 212, 213]
	E	($\tilde{A}\,^2A_1$–$\tilde{X}\,^2E$) $280 \leqslant \lambda/nm \leqslant 370$, Ar*, Kr* + CH_3OH	[206, 210, 211]
	LMR	(250–650 μm) FIR/CO_2-laser	[208]
	MS	(electron impact ionization) (CH_3O^+), (CD_3O^+)	[4, 209]
CH_2OH ($\tilde{X}$)	LMR	(117·7–657·2 μm) CH_2F_2 (a.o.) FIR/CO_2-laser	[215]
	MS	(electron impact ionization) (CH_2OD^+, CD_2OH^+, CD_2OD^+)	[209]
CH_3S ($\tilde{X}$)	MS	(electron impact ionization) (CH_3S^+)	[3, 4]
NCO ($\tilde{X}\,^2\Pi_i$)	A–UV	($\tilde{A}\,^2\Sigma^+$–$\tilde{X}\,^2\Pi$) $\lambda = 465$ nm ($\tilde{B}\,^2\Pi$–$\tilde{X}\,^2\Pi$) $\lambda = 440$ nm	[229]
	LIF	($\tilde{A}\,^2\Sigma^+$–$\tilde{X}\,^2\Pi$) $\lambda = 465{\cdot}8$ nm dye-laser	[227]
	LIF	(Intracavity excitation) cw-dye-laser/Ar^+ ion-laser	[228]
	LMR	($\tilde{X}\,^2\Pi$, ν_3 antisym. stretching mode) CO-laser	[230]
	ESR	($\tilde{X}\,^2\Pi$), (0, 0, 0), (0, 1, 0), (0, 2, 0) X-band	[4, 231]
CH_4N ($\tilde{X}$)	MS	photoionization (CH_4N^+)	[257]
CH_3O_2 ($\tilde{X}\,^2A''$)	A–UV	$220 \leqslant \lambda/nm \leqslant 280$, $\lambda_{max} = 235$ nm	[217, 220, 221, 342]
		ε (235 nm) = 870 l/mol cm	[218]
	A–IR	$\lambda = 1{\cdot}2$–$1{\cdot}4$ μm modulated Hg-sensitized production, phase sensitive detection	[343]
	MS	(electron impact ionization) ($CH_3O_2^+$)	[219]
CF_3O_2 ($\tilde{X}$)	MS	electron impact ionization, photoionization ($CF_3O_2^+$)	[224, 347]

Atom, radical	Method	Explanations	References
C_2 ($X\,^1\Sigma_g^+$)	A, E–VIS–IR	$A\,^1\Pi_u$–$X\,^1\Sigma_g^+$ (Phillips) $672 \leqslant \lambda/\mathrm{nm} \leqslant 1549$	[335]
	A, E–UV	$D\,^1\Sigma_u^+$–$X\,^1\Sigma_g^+$ (Mulliken) $231 \leqslant \lambda/\mathrm{nm} \leqslant 241{\cdot}5$	[335]
	A–UV	$F\,^1\Pi_u$–$X\,^1\Sigma_g^+$ $231 \leqslant \lambda/\mathrm{nm} \leqslant 134{\cdot}5$	[335]
	CARS	$\omega_1 = 532{\cdot}0$ nm Nd:YAG doub.; $\omega_2 = 631{\cdot}8$ nm OPA (420–730 nm) tunable and dye 547–654 nm respectively	[254, 255]
	LIF	($A\,^1\Pi_u$–$X\,^1\Sigma_g^+$, 0, 4) excitation at 690·9 nm emission (4, 1) at 790·8 nm, Ar^+-laser 514·53 nm	[242, 245, 246]
	MS	(electron impact ionization) (C_2^+)	[3, 247]
C_2 ($a\,^3\Pi_u$)	A–IR	$b\,^3\Sigma_g^-$–$a\,^3\Pi_u$ (Ballik-Ramsay) $1{\cdot}1 \leqslant \lambda/\mu\mathrm{m} \leqslant 2{\cdot}7$	[335]
	A, E–VIS–UV	$d\,^3\Pi_g$–$a\,^3\Pi_u$ (Swan) $340 \leqslant \lambda/\mathrm{nm} \leqslant 785$	[335]
	E–UV	$e\,^3\Pi_u$ $237 \leqslant \lambda/\mathrm{nm} \leqslant 328{\cdot}5$ (the three lowest states are mentioned)	[335]
	LIF	($d\,^3\Pi_g$–$a\,^3\Pi_u$) (0, 0) excitation at $\lambda = 516{\cdot}5$ nm emission at $\lambda = 559$ nm	[167, 242, 244]
C_2N ($\tilde{X}\,^2\Pi_r$)	A–VIS	($\tilde{A}\,^2\Delta_i$–$\tilde{X}\,^2\Pi_r$, $\tilde{B}\,^2\Sigma^-$–$\tilde{X}\,^2\Pi_r$, $\tilde{C}\,^2\Sigma^+$–$\tilde{X}\,^2\Pi_r$) $469 \leqslant \lambda/\mathrm{nm} \leqslant 472$	[249]
	LIF	($\tilde{A}\,^2\Delta(0,0,0)$–$\tilde{X}\,^2\Pi$ (0, 0, 0)) $471{\cdot}6 \leqslant \lambda/\mathrm{nm} = 468{\cdot}7$, dye-laser	[248]
	MS	(electron impact ionization) (C_2N^+)	[3]
C_2H ($\tilde{X}\,^2\Sigma^+$)	A–IR–CCL	$2{\cdot}38 \leqslant \lambda/\mu\mathrm{m} \leqslant 3{\cdot}3$; rot.-vib. $\tilde{X}$ and $\tilde{A}\,^2\Pi$–$\tilde{X}\,^2\Sigma$	[234]
	LMR	FIR/CO_2-laser	[260]
	MS	(electron impact ionization) (C_2H^+)	[4, 233]
C_2O ($\tilde{X}\,^3\Sigma^-$)	A–VIS	($\tilde{A}\,^3\Pi_i$–$\tilde{X}\,^3\Sigma^-$) $530 \leqslant \lambda/\mathrm{nm} \leqslant 900$	[251]
	LIF	($\tilde{A}\,^3\Pi_i$–$X\,^3\Sigma^-$), excitation at 689–588 nm flashl. dye-laser C_2O–from C_3O_2 photolysis (266 nm)	[262, 253]
C_2HO ($\tilde{X}$)	MS	(electron impact ionization-photoionization) (C_2HO^+)	[223]
C_2H_3 ($\tilde{X}$)	MS	(electron impact ionization) ($C_2H_3^+$)	[232]
C_2F_3 ($\tilde{X}$)	MS	(electron impact ionization) ($C_2F_3^+$)	[259]
C_2H_5 ($\tilde{X}\,^2A$)	A–UV	$234 \leqslant \lambda/\mathrm{nm} \leqslant 255$, $\lambda_{max} = 247$ nm $\varepsilon\,(247) = 4{\cdot}8 \times 10^2$ l/mol cm C_2H_5– from C_2H_5-N=N-C_2H_5 photolysis	[237]
	MS	electron impact ionization-photoionization ($C_2H_5^+$)	[235, 236, 257]
C_2H_5O ($\tilde{X}$)	LIF	($\tilde{A}$–$\tilde{X}$ (3–0)) CO-stretching mode $\lambda = 322{\cdot}8$ nm flashl./dye-laser (doubled)	[212, 214]
	E	($320 \leqslant \lambda_E/\mathrm{nm} \leqslant 440$) $C_2H_5O^*$ from C_2H_5ONO photolysis	[206, 210]
	MS	C_2D_5O, electron impact ionization, ($C_2H_5O^+$), ($C_2D_5O^+$)	[3, 222]
C_2H_4OH ($\tilde{X}$)	MS	(electron impact ionization) ($C_2H_4OH^+$), ($C_2H_4OD^+$)	[222]
CH_3CO ($\tilde{X}$)	A–UV	CD_3CO $200 \leqslant \lambda/\mathrm{nm} \leqslant 240$, $\lambda_{max} = 207$ nm $\varepsilon\,(207) = 1 \times 10^4$ l/mol cm CH_3CO $200 \leqslant \lambda/\mathrm{nm} \leqslant 240$, $\lambda_{max} = 215$ nm $\varepsilon\,(215) = 1 \times 10^4$ l/mol cm	[239]
CD_3CO ($\tilde{X}$)	MS	photoionization (CH_3CO^+)	[238]
C_2H_3O ($\tilde{X}$)	LIF	(0–0 band) $\lambda = 347{\cdot}2$ nm flashl./dye-laser	[216, 240]
	MS	photoionization ($C_2H_3O^+$)	[258]
$C_2H_5O_2$ ($\tilde{X}$)	A–UV	$220 \leqslant \lambda/\mathrm{nm} \leqslant 280$, $\lambda_{max} = 236$ nm $\varepsilon\,(236) = 1{\cdot}02 \times 10^3$ l/mol cm	[225, 226]

Atom, radical	Method	Explanations	References
	A–IR	$1 \leqslant \lambda/\mu\mathrm{m} \leqslant 1\cdot6$, electronic transition (0–0) $\lambda = 1\cdot317\,\mu\mathrm{m}$	[343]
CH_3COO_2 ($\tilde{X}$)	A–UV	$210 \leqslant \lambda/\mathrm{nm} \leqslant 270$, CH_3CO_3 from $CH_3CO + O_2$	[241]
C_2H_4N ($\tilde{X}$)	MS	photoionization ($C_2H_4N^+$)	[257]
C_2H_6N ($\tilde{X}$)	MS	photoionization ($C_2H_6N^+$)	[257]
C_3 ($\tilde{X}\,^1\Sigma_g^+$)	A–VIS–UV	($\tilde{A}\,^1\Pi_u$–$\tilde{X}\,^1\Sigma_g^+$) $340 \leqslant \lambda/\mathrm{nm} \leqslant 410$	[243]
	LIF	($A\,^1\Pi_u$–$X\,^1\Sigma_g^+$) (0, 0, 0)–(0, 0, 0) $\lambda = 405$ nm	[242]
	MS	(electron impact ionization) (C_3^+)	[3, 247]
Si ($^3P_{0,1,2}$)	RA	($4\,^3P_1$–$3\,^2P_0$) $\lambda = 251\cdot43$ nm ($4\,^3P_2$–$3\,^3P_1$) $\lambda = 250\cdot69$ nm ($4\,^3P_2$–$3\,^3P_2$) $\lambda = 251\cdot61$ nm	[310]
Si ($^1D_2$)	A–UV	$4s\ (^1P_1^0)$–$3p^2(^1D_2)$ $\lambda = 288\cdot16$ nm	[344]
Si ($^1S_0$)	RA–UV	$4s\ (^1P_1^0)$–$3p^2(^1S_0)$ $\lambda = 390\cdot53$ nm	[344]
SiF ($X\,^2\Pi$)	A–VIS–UV	($A\,^2\Sigma^+$–$X\,^2\Pi$) $\lambda = 436\cdot8$ nm ($B\,^2\Sigma^+$–$X\,^2\Pi$) $\lambda = 289$ nm	[336]
SiF_2 ($\tilde{X}\,^1A$)	PES	(photo electron spectroscopy)	[339]
	MS	(electron impact ionization) (SiF^+)	[3]
SiO ($X\,^1\Sigma^+$)	A	($A\,^1\Pi$–$X\,^1\Sigma^+$) $293 \geqslant \lambda/\mathrm{nm} \geqslant 210$	[336]
	MS	(electron impact ionization) (SiO^+)	[3]
SiC ($X\,^1\Sigma$)	MS	(electron impact ionization) (SiC^+)	[3]
Si_2 ($^3\Sigma_g^-$)	A–VIS–UV	($H\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$) $452\cdot6 \geqslant \lambda/\mathrm{nm} \geqslant 386\cdot3$	[333, 334, 335]
	MS	(electron impact ionization) (Si_2^+)	[3]
Ge ($^3\mathrm{P}_J$)	RA	$5s\ (^3P^0)$–$4p^2(^3P_2)$ $\lambda = 275\cdot5$ nm $5s\ (^3P^0)$–$4p^2(^3P_1)$ $\lambda = 259\cdot3$ nm $5s\ (^1P_1^0)$–$4p^2(^3P_0)$ $\lambda = 249\cdot8$ nm	[311, 345]
Ge ($^1S_0$)	RA	$4d\ (^1P_1^0) \leftarrow 4p^2(^1S_0)$ $\lambda = 274\cdot04$ nm	[316]
GeC ($X\,^1\Sigma$)	MS	(electron impact ionization) (GeC^+)	[3]
GeO ($X\,^1\Sigma^+$)	A	($D\,^1\Sigma^+$–$X\,^1\Sigma^+$) $225 \leqslant \lambda/\mathrm{nm} \leqslant 336$	[336]
	MS	(electron impact ionization) (GeO^+)	[3]
GeF ($X\,^2\Pi_{1/2}$)	A	($A\,^2\Sigma^+$–$X\,^2\Pi$) $390 \leqslant \lambda/\mathrm{nm} \leqslant 520$	[336]
	MS	(electron impact ionization) (GeF^+)	[3]
GeF_2 ($\tilde{X}\,^1A$)	PES	(UV-photoelectron spectroscopy)	[338]
	MS	(electron impact ionization) (GeF_2^+)	[3]
$GeCl_2$ ($\tilde{X}$)	MS	(electron impact ionization) ($GeCl_2^+$)	[3]
Sn (^3P_J)	RA	$6s\ (^3P_1^0)$–$5p^2(^3P_0)$ $\lambda = 286\cdot33$ nm	[312, 346]
SnF ($X\,^2\Pi_{1/2}$)	A	($A\,^2\Sigma^+$–$X\,^2\Pi_r$) $630 \geqslant \lambda/\mathrm{nm} \geqslant 460$	[336]
	MS	(electron impact ionization) (SnF^+)	[3]
SnO ($X\,^1\Sigma^+$)	A	($D\,^1\Pi$–$X\,^1\Sigma^+$) $449 \geqslant \lambda/\mathrm{nm} \geqslant 307$	[336]
	MS	(electron impact ionization) (SnO^+)	[3]
SnF_2 ($\tilde{X}$)	MS	(electron impact ionization) (SnF_2^+)	[3]
Pb (^3P_J)	RA	($7s\ (^3P_1^0)$–$6p^2\ (^3P_0)$) $\lambda = 283\cdot3$ nm	[313]
PbF ($X\,^2\Pi_{1/2,3/2}$)	A	($A(\Omega = 1/2)$–$X\,^2\Pi_{1/2,3/2}$) $\lambda\ (0, 0) = 444\cdot2$ nm $\lambda\ (0, 0) = 704\cdot3$ nm	[336]
	MS	(electron impact ionization) (PbF^+)	[3]
PbF_2, $PbCl_2$ ($\tilde{X}$)	MS	(electron impact ionization) (PbF_2^+), ($PbCl_2^+$)	[3]
PbO, PbS ($X\,^1\Sigma^+$)	A	($A\,O^+$–$X\,^1\Sigma^+$); PbO, $\lambda\ (0, 0) = 507\cdot0$ nm PbS, $\lambda\ (1, 2) = 549\cdot81$ nm	[336]
	MS	(electron impact ionization) (PbO^+), (PbS^+)	[3]

Table 2. Atoms and radicals without elements of the fourth group of the periodic table.

Atom, radical	Method	Explanations	References
H, D ($^2S_{1/2}$)	RA	(Lyman α (^2P–^2S) $\lambda = 121{\cdot}6$ nm sensitivity: $[\mathrm{H}] \geqslant 2 \times 10^{-14}$ mol/cm^3	[26, 27]
	RF	Lyman α (^2P–^2S) $\lambda = 121{\cdot}6$ nm sensitivity: $[\mathrm{H}] \geqslant 2 \times 10^{-15}$ mol/cm^3	[28, 29, 30]
	ChL	HNO*($^1A''$), (DNO)*	[31, 33]
	ESR	(X-band) $^2S_{1/2}$, H, $I = 1/2$, two lines D, $I = 1$, three lines sensitivity: $[\mathrm{H}] \geqslant 8 \times 10^{-13}$ mol/cm^3 $S/N = 1$	[5, 384]
	MS	electron impact-photoionization (H$^+$)	[3, 4]
O ($^3P_{2,1,0}$)	RA	$^3S_1$—$3P_J$ $\lambda/\mathrm{nm} = (130{\cdot}22;\ 130{\cdot}49;\ 130{\cdot}6)$	[34, 35]
	RF	($^3S_1$–$^3P_{2,1,0}$) triplet not resolved at $\lambda = 130$ nm sensitivity: $[\mathrm{O}(^3P)] \geqslant 6{\cdot}6 \times 10^{-14}$ mol/cm^3	[37, 38, 58]
	ChL	NO_2^* ($\mathrm{O} + \mathrm{NO} \rightarrow \mathrm{NO}_2^*$)	[32, 36]
	LMR	($^3P_0$–$^3P_1$) $\lambda = 147{\cdot}8$ μm; $145{\cdot}7$ μm (CH_3NH_2, CH_3OD/CO_2-laser)	[23, 24]
	ESR	(X-band) ($^3P_2$), four lines, ($^3P_1$), two lines $^3P_0$ (none) sensitivity: $[\mathrm{O}(^3P)] \geqslant 10^{-13}$ mol/cm^3	[5, 6, 7]
	MS	(electron impact ionization) (O$^+$)	[3, 4]
O_2 ($X\,^3\Sigma_g^-$)	A–UV	$A\,^3\Sigma_u^+$–$X\,^3\Sigma_g^-$ $243 \leqslant \lambda/\mathrm{nm} \leqslant 488$ $\lambda\ (0,0) = 285{\cdot}7$ nm	[1]
		$B\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$ (Schumann–Runge) $175 \leqslant \lambda/\mathrm{nm} \leqslant 535$, $130 \leqslant \lambda/\mathrm{nm} \leqslant 275$, $\lambda\ (0,0) = 202{\cdot}601$ nm (Two lowest transitions of the $^3\Sigma$-state are mentioned, more than 16 further systems are known, see e.g. 335)	[335]
	LMR	$X\,^3\Sigma_g^-$ ($N = 3$, $J = 4$, $M = 4$)–($N = 5$, $J = 5$, $M = 4$) 337 μm, HCN-laser $X(^3\Sigma_g^-)$ ($N = 13$, $J = 14$, M)–($N = 15$, $J = 14$, M') 119 μm and ($N = 21$, $J = 22$, M)–($N = 23$, $J = 22$, M') 78 μm, H_2O-laser	[2]
	ESR	($^3\Sigma_g^-$) X-band $v = 0, 1$ X-band (substance to calibrate the Q-factor for magnetic dipole transitions)	[5, 369]
	MS	electron impact- and photoionization (O_2^+)	[3]
O_2 ($a\,^1\Delta_g$)	E	O_2 ($^1\Delta_g$)–O_2 ($^3\Sigma_g^-$) + $h\nu$ ($\lambda = 1270$ nm)	[386]
	E	('dimol emission') vibrational states: $2O_2\ (^1\Delta_g) \rightarrow 2O_2\ (^3\Sigma) + h\nu$ ($\lambda = 634$ nm) $(0+0)$–$(0+0) + h\nu$ ($\lambda = 634$ nm) $(0+0)$–$(1+0) + h\nu$ ($\lambda = 703$ nm) $(1+0)$–$(0+0) + h\nu$ ($\lambda = 579$ nm)	[383, 385]
	E	emission $O_2(^1\Delta)$ from: $2O_2(^1\Delta_g) \rightarrow O_2(^1\Sigma_g^+)_{v'} + O_2(^3\Sigma_g^-)$ ($\lambda = 697$; $706{\cdot}1$)	[385]
	LMR	($a\,^1\Delta_g$) $v = 0$, $J = 1$, $J = 4, 5, 7, 8, 9$, $889{\cdot}1 \geqslant \lambda/\mu\mathrm{m} \geqslant 392{\cdot}1$ ($C_2H_2F_2$, CH_2F_2, C_2H_3Cl, CH_3OH) FIR/CO_2-laser	[366]
	ESR	('EPR') X-band magnetic dipole transitions sensitivity: $[O_2(^1\Delta_g)] \geqslant 5 \times 10^{-11}$ mol/cm^3	[5, 384]
	PI	(photoionization) Ar-lamp, LiF-window $\lambda = 106{\cdot}7$ nm absolute calibration with NO	[379, 380] [381]

Atom, radical	Method	Explanations	References
	MS	electron impact- and photoionization (O_2^+)	[3]
	E	(calorimetric measurements: Co resistance-wire)	[382]
OH ($X\,^2\Pi_{3/2,1/2}$) ($0\leqslant v\leqslant 9$)	A–UV	$310\leqslant \lambda/\mathrm{nm}\leqslant 314$, $2700\leqslant T/\mathrm{K}\leqslant 6000$, Hg-high-pressure lamps	[42, 43]
	RA	($A\,^2\Sigma^+$–$X\,^2\Pi$) Q_1^4 $\lambda = 308{\cdot}3278$ nm, H_2O/Ar-discharge lamp $F = 1{\cdot}4\times 10^{-4}$	[45]
	LA	($A\,^2\Sigma^+(v=0)$–$X\,^2\Pi$ $(v=0)$ Q_1^1, $\lambda = 307{\cdot}844$ nm, Q_1^2, $\lambda = 307{\cdot}955$ nm, dye/Ar^+-laser (doubled), sensitivity: $[OH]\geqslant 2\times 10^{-15}$ mol/cm^3 $S/N = 1$ 1 m absorption length multipass (5 times)	[49]
	RF	($A\,^2\Sigma^+$–$X\,^2\Pi$ (0, 0)) $\lambda = 310$ nm, sensitivity: $[OH]\geqslant 1{\cdot}7\times 10^{-16}$ mol/cm^3 flow system $p = 3$ mbar, time 1 s	[40, 41, 46, 350]
	LIF	($A\,^2\Sigma^+$–$X\,^2\Pi$ (1, 0)) $\lambda_L = 282{\cdot}2$ nm, flashl./dye-laser (doubled) sensitivity: $[OH]\geqslant 2\times 10^{-17}$ mol/cm^3 ('low pressure')	[39, 44, 47]
	LMR	($^2\Pi_{3/2,1/2}$) $\lambda_L/\mu\mathrm{m} = 79$; 118·6; ($0\leqslant v\leqslant 9$) 108 μm H_2O, D_2O-laser ($\lambda/\mu\mathrm{m} = 78$; 84; 96; 118·8; 163) FIR/CO_2-laser sensitivity: $[OH]\geqslant 2\times 10^{-18}$ mol/cm^3 $S/N = 1$	[9, 10, 11, 12, 16]
	ESR	($^2\Pi_{3/2,1/2}$) $0\leqslant v\leqslant 4$, X-band sensitivity: $[OH]\geqslant 3{\cdot}3\times 10^{-14}$ mol/cm^3 $S/N = 1$	[5, 8, 13, 348]
	E	(IR) $OH^{\dagger}$ from: $H + O_3$, OH ($v\leqslant 9$) and $H + NO_2$, OH ($v\leqslant 4$)	[351]
	MS	electron impact-photoionization (OH^+)	[3, 4, 50]
	CARS		[395]
OD ($X\,^2\Pi_{3/2,1/2}$)	RA	($A\,^2\Sigma^+$–$X\,^2\Pi$) (0, 0) Q_1^5 $\lambda = 307{\cdot}6$ nm, D_2O/Ar discharge lamp	[48]
	LIF	($X\,^2\Pi$ ($0\leqslant v\leqslant 2$)) $A\,^2\Sigma^+$–$X\,^2\Pi$, dye/Nd : YAG (doubled)	[91]
	LMR	($X\,^2\Pi_{3/2}$) $0\leqslant v\leqslant 5$, $\lambda_L = 171$ μm D_2O-laser $\lambda/\mu\mathrm{m} = 118{\cdot}8$; 96·5; 215·4 ($CH_3OH$, CH_3OD) FIR/CO_2-laser	[12, 15]
	ESR	($^2\Pi_{3/2}$, $J = 3/2$) $0\leqslant v\leqslant 5$, X-band	[5, 14]
HO_2 ($\tilde{X}\,^2A''$)	A–UV	$220\leqslant \lambda/\mathrm{nm}\leqslant 280$, at $T = 1000$ K ε_{10} (230 nm) $= 6{\cdot}6\times 10^2$ l/mol cm $\lambda = 227{\cdot}5$ nm $\varepsilon_{10} = 4{\cdot}9\times 10^3$ l/mol cm at $T = 298$ K sensitivity: $[HO_2]\geqslant 8\times 10^{-13}$ mol/cm^3	[51, 53, 54]
	A–IR	($^2A'$–$^2A''$) $1{\cdot}1\leqslant \lambda/\mu\mathrm{m}\leqslant 1{\cdot}5$ ε (1·5 μm) = 10(l/mol cm)	[56]
	A–IR–DL	($^2A''$)ν_3 $\lambda = 9{\cdot}09$ μm	[57]
	ChL	($^2A'$ 0, 0, v_3')–($^2A''$ 0, 0, 0) from: $HCO + O_2(^1\Delta_g)\rightarrow HO_2(^2A', 0, 0, v_3') + CO$ $HO_2(^2A')$ after energy transfer $HO_2\,^2A'' + O_2(^1\Delta_g)\rightarrow HO_2(^2A', \nu_3) + O_2(^3\Sigma_g^-)$	[55]
	LMR	(HO_2 $\tilde{X}$, v_3) 9·1 μm CO_2-laser, FIR ($\lambda/\mu\mathrm{m} = 78{\cdot}4$; 79·09; 118·6) H_2O-laser ($\lambda/\mu\mathrm{m} = 72{\cdot}75$; 84·3; 107·72) D_2O-laser ($\lambda/\mu\mathrm{m} = 170{\cdot}1$; 392) CH_3OH/CO_2-laser sensitivity: $[HO_2]\geqslant 2\times 10^{-16}$ mol/cm^3	[19, 20, 22]
	ESR	($^2A''$) ($1_{1,0} - 1_{1,1}$) X-band (9 GHz) sensitivity: (very low)	[21]
	MS	(electron impact ionization) (HO_2^+)	[3, 4, 52]

Atom, radical	Method	Explanations	References
DO_2 ($\tilde{X}\,^2A''$)	A–UV	$\lambda = 227{\cdot}5$ nm $\varepsilon_{10} = 4{\cdot}1 \times 10^3$ l/mol cm at 298 K, sensitivity: $[DO_2] \geqslant 6{\cdot}7 \times 10^{-13}$ mol/cm^3	[54]
	A–IR	($^2A'$–$^2A''$) $1{\cdot}1 \leqslant \lambda/\mu m \leqslant 1{\cdot}5$ and $\lambda = 2{\cdot}05\,\mu m$	[56]
	ChL	($^2A'$, 0, 0, v_3')–($^2A''$, 0, 0, 0) from $DCO + O_2(^1\Delta_g) \rightarrow DO_2(^2A', 0, 0, v_3') + CO$	[55]
	LMR	($^2A''v_2$), 9·8 μm CO_2-laser FIR ($163 \leqslant \lambda/\mu m \leqslant 513$) ($CH_3OH$; N_2H_4; CH_3OD; a.o.)/CO_2-laser	[17, 18]
	ESR	($^2A''$) X-band 8·9; 9·3 GHz sensitivity: (low)	[17]
F ($^2P_{3/2,\,1/2}$)	RA–VUV	($3s\,^2P_J$–$2p^5\,^2P_J$) λ/nm = 95·48; 95·55; 95·85; 95·19 F_2/He-discharge lamp window: CHS collimated hole structures sensitivity: $[F(^2P_{3/2})] \geqslant (0{\cdot}2–2)\ 10^{-12}$ mol/cm^3	[60, 352] [353]
	RF	($3s^2\,^2P_J$–$2p^5\,^2P_J$) $95 \leqslant \lambda/nm \leqslant 98$, F_2/He-discharge lamp	[352]
	ChL	HF‡–IR	[59, 349, 354, 355]
		$F + NO \rightarrow NOF^*$ (yellow emission)	[61]
	ESR	($^2P_{3/2,\,1/2}$) X-band sensitivity: $[F(^2P_J)] \geqslant 5 \times 10^{-14}$ mol/cm^3	[5, 71]
	MS	(electron impact ionization) (F^+)	[6, 79]
Cl ($^2P_{3/2}$)	RA–UV	($^2P_{1/2}$) ($3p^4\ 4s\,^2P_{3/2}$–$3p^5\,^2P^0_{1/2}$) $\lambda = 136{\cdot}34$ nm $^2D_{3/2}$–$^2P_{3/2}$ $\lambda = 118{\cdot}875$ nm	[356] [353]
	A–IR–DL	($3p^5$, $^2P_{1/2}$–$^2P_{3/2}$) $\lambda = 11{\cdot}3\,\mu m$ IR-diode laser (tunable)	[70]
	RF–UV	(^4P–^2P) multiplet at $\lambda = 138{\cdot}0$ nm; sensitivity: $[Cl(^2P_{3/2})] \geqslant 8 \times 10^{-14}$ mol/cm^3	[58, 63, 65]
	TALIF	($4p^1(^2F_{7/2})$–$3p\,(^2P_{3/2})$) absorption, two photon process $\lambda = 210{\cdot}1$ nm, dye/YAG (doubled)/ArF-Exiplex-laser enhanced, Raman shifted H_2/D_2, emission ($4p^1(^2F_{7/2})$–$4s^1(^2D_{5/2})$) $\lambda = 904$ nm sensitivity: $[Cl(^2P_{3/2})] \geqslant 1{\cdot}7 \times 10^{-11}$ mol/cm^3	[66]
	ChL	$HCl(v = 1, 2)$ $\lambda = 3{\cdot}5\,\mu m$ from $Cl + H_2S$; NOCl titration: $Cl + NOCl \rightarrow O_2 + NO$, $NO + Cl \rightarrow ClNO^*$	[62, 64]
	LMR	($^2P_{1/2}$–$^2P_{3/2}$) $\lambda = 11{\cdot}33\,\mu m$ $^{13}C^{16}O_2$-laser	[25]
	ESR	($^2P_{3/2}$, $^2P_{1/2}$) (X-band) sensitivity: $[Cl(^2P_{3/2})] \geqslant 5 \times 10^{-14}$ mol/cm^3	[5]
	MS	(electron impact ionization) (Cl^+, Cl^{2+})	[6, 79]
Br ($^2P_{3/2}$)	RA–UV	Br $^2P_{3/2}$ ($5\,^2P_{3/2}$–$4\,^2P_{3/2}$) $\lambda = 148{\cdot}9$ nm; Br* ($5\,^2P_{1/2}$–$4\,^2P_{1/2}$) $\lambda = 153{\cdot}2$ nm	[67]
	RF	($5\,^4P_{3/2}$–$4\,^2P_{3/2}$) $\lambda = 157{\cdot}7$ nm, sensitivity: $[Br(^2P_{3/2})] \geqslant 2{\cdot}5 \times 10^{-14}$ mol/cm^3	[58, 69]
	ESR	($^2P_{3/2}$) Br^{79}, Br^{81} (X-band) sensitivity: $[Br(^2P_{3/2})] \geqslant 5 \times 10^{-14}$ mol/cm^3	[5]
	MS	(electron impact ionization) (20–30 and 70 eV) (Br^+, Br^{2+})	[6, 79]
I ($^2P_{3/2,\,1/2}$)	RA–UV	($6s\,^2P_{3/2}$–$5p\,^2P_{1/2}$) $\lambda = 206{\cdot}2$ nm, I_2-discharge lamp	[68]
	RF–UV	($6s(^4P_{5/2})$–$5p^2(^2P_{3/2})$) $\lambda = 183{\cdot}0$ nm sensitivity: $[I(^2P_{3/2})] \geqslant 1{\cdot}3 \times 10^{-14}$ mol/cm^3	[58]

Atom, radical	Method	Explanations	References
	ESR	($^2P_{3/2}$) (X-band) sensitivity: $[\mathrm{I}(^2P_{3/2})] \geqslant 5\times10^{-14}$ mol/cm^3	[5]
	MS	(electron impact ionization) 70 eV (I^+)	[6, 79]
FO ($X\,^2\Pi$)	LMR	FO ($X\,^2\Pi_{3/2}$) $9{\cdot}76 \geqslant \lambda/\mu\mathrm{m} \geqslant 9{\cdot}59$ CO_2-laser	[358]
	MS	(electron impact ionization) 70 eV (FO^+)	[79, 80]
ClO ($X\,^2\Pi_{3/2,\,1/2}$)	A–UV	(A–X) (11–0) $\lambda = 277{\cdot}2$ nm, $\varepsilon = 4{\cdot}37\times10^3$ l/mol cm	[74]
	A–IR–DL	($^2\Pi_{3/2}$ and $^2\Pi_{1/2}$) $11{\cdot}77 \geqslant \lambda/\mu\mathrm{m} \geqslant 11{\cdot}44$ band strength (^{35}ClO R 13/2, $\Omega = 1/2$, $\omega = 853{\cdot}123$ cm^{-1} $\sigma = 12{\cdot}4$ cm^{-2} atm^{-1}.	[76, 359]
	MW	$^2\Pi_{3/2}$, $^2\Pi_{1/2}(v=0)$, $J = 9/2$–$7/2$ $164 \leqslant \omega/\mathrm{GHz} \leqslant 167$	[75]
	LMR	$^2\Pi_{3/2}(v = 2 \leftarrow 0)$ $\lambda/\mu\mathrm{m} = 6{\cdot}02$; 595; CO-laser $^{37}\mathrm{Cl}^{16}\mathrm{O}(X\,^2\Pi_{3/2},\ v=0)$, $J(23/2$–$21/2)$, $\lambda = 713{\cdot}1$ μm DCOOH/CO_2-laser sensitivity: $[\mathrm{ClO}\ (^2\Pi_{3/2})] \geqslant 5\times10^{-14}$ mol/cm^3	[72, 357]
	ESR	($^2\Pi_{3/2}$) X-band, electric dipole transitions	[5]
	MS	(electron impact ionization) 70 eV (ClO^+)	[73, 79]
BrO ($X\,^2\Pi$)	A–UV	($A\,^2\Pi$–$X\,^2\Pi$ ($v'' = 0$)) strong absorption $300 \leqslant \lambda/\mathrm{nm} \leqslant 355$ ε (338·3 nm) $= 4{\cdot}8\times10^3$ (l/mol cm) maximum at $\lambda = 338{\cdot}3$ nm (0–4)-band	[77, 81]
	ESR	($X\,^2\Pi_{3/2}$) X-band electric dipole transition Stark modulation	[5]
	MS	(electron impact ionization) 70 eV ($^{79,81}BrO^+$)	[78, 79]
IO ($X\,^2\Pi$)	A–VIS–UV	($A\,^2\Pi$–$X\,^2\Pi$) (3–0), (4–0), (2–0), (2–2), (1–1) $418 \leqslant \lambda/\mathrm{nm} \leqslant 470$, maximum (4–0), ε ($\Delta\lambda = 0{\cdot}5$ nm) $= 3\times10^3$ l/mol cm	[81]
	ESR	($X\,^2\Pi_{3/2}$) X-band, electric dipole transition	[5]
	MS	(electron impact ionization) 70 eV (IO^+)	[79]
FO_2 ($\tilde{X}$)	A–UV	$200 \leqslant \lambda/\mathrm{nm} \leqslant 240$, $\lambda_{\max} = 210$ nm, ε (210 nm) $= 10^4$ l/mol cm	[125, 361]
	LMR	FO_2 ($\tilde{X}$) $119 \leqslant \lambda/\mathrm{nm} \leqslant 742$; ($CH_3OH$, CD_3OD, CHOOH a.o.) FIR/CO_2-laser	[360]
	MS	(electron impact ionization) FO_2^+)	[3]
ClO_2 ($\tilde{X}\,^2B_1$)	A–UV	($\tilde{A}\,^2A_2$–$\tilde{X}\,^2B_1$) $\lambda = 322$ nm, 361·5 nm, ε (351·5 nm) $= 6{\cdot}86\times10^3$ (l/mol cm)	[74]
	LMR	($\tilde{X}\,^2B_1$) $v_1 = 1$–0 $\lambda = 10{\cdot}51$ μm P(12)-line CO_2-laser	[362]
	MS	(electron impact ionization) (ClO_2^+)	[3]
Li ($^2S_{1/2}$)	SI	Langmuir–Taylor detector, very sensitive for atoms of the first group: ion yield $n^+/n = c\exp\{-(J-\phi)/kT\}$ J = ionization energy, ϕ = work function	[325, 376, 377, 378]
Na, K ($^2S_{1/2}$)	LIF	($^2P_{1/2}$, $^2P_{3/2}$–$^2S_{1/2}$) $\lambda/\mathrm{nm} = 589$; 589·6, sensitivity: $[\mathrm{Na}] \geqslant 2\times10^{-24}$ mol/cm^3	[326]
	SI	Langmuir–Taylor detector, very sensitive molecular beam detector, sensitivity K > Na see above	[325, 376, 377, 378]
	ESR	($^2S_{1/2}$) X-band (H-atom analogous)	[5]
Rb, Cs ($^2S_{1/2}$)	SI	very sensitive molecular beam detector, Langmuir–Taylor detector, sensitivity Cs > Rb see above	[325, 376, 377]
	ESR	($^2S_{1/2}$) X-band, H-atom analogous	[5]

Atom, radical	Method	Explanations	References
N $(^4S_{3/2})$ $(^2D_{3/2,5/2})$ (^2P_J)	A–UV	$(^4S_{3/2}-^4P_{5/2,3/2,1/2})$ $(\lambda/\mathrm{nm}=119{\cdot}955;\ 120{\cdot}022;\ 120{\cdot}071)$ N_2/Ar-discharge lamp N $(2\,^2D)$ $2p^3$ (^2D)–$3s$ (^2P) $\lambda=149{\cdot}3$ nm	[84, 85, 88]
		N $(2\,^2P)$ $2p^3$ $(^2P_{3/2})$–$3s$ $(^2P_{3/2})$ $\lambda=174{\cdot}3$ nm, N_2/Ar-discharge lamp	[120]
	RF	$2p^2$ $3s$ $(^4P_{1/2,3/2,5/2})$–$2p^3$ $(^4S_{3/2})$, sensitivity: $\lambda=120$ nm $[N(^4S)]\geqslant(1-1{\cdot}7)\times10^{-4}$ mol/cm^3	[86, 340]
	TI	NO titration $N\ (^4S)+NO\rightarrow N_2+O\ (^3P)$ The appearance of a green-yellow emission indicates the end-point $NO+O\rightarrow NO_2+h\nu$	[87]
	ESR	$(^4S_{3/2})$ X-band, $(^2D_{5/2,3/2})$ X-band, sensitivity: $[N(^2D)]\geqslant5\times10^{-13}$ mol/cm^3 $S/N=1$	[5, 83]
	MS	(electron impact ionization, photoionization) (N^+)	[6, 79]
N_3 $(\tilde{X})$	MS	(electron impact ionization $E=20$ eV) (N_3^+)	[82]
NH, ND $(X\,^3\Sigma^-)$	RA–L	$(A\,^3\Pi-X\,^3\Sigma^-)\,0,0\,R_2\,(8)$ dye flashl.-laser (doubled)	[102]
	RF	$(A\,^3\Pi-X\,^3\Sigma^-)$ $\lambda=336$ nm	[89]
	LIF	$(A\,^3\Pi_i-X\,^3\Sigma^-)$ $(1,0)\lambda=305$ nm $(0,0)$ $\lambda=336$ nm dye flashl.-laser (doubled)	[90]
	LMR	$(^3\Sigma^-)$ $N=1\rightarrow0$ $\lambda/\mu\mathrm{m}=302;\ 315,$ CH_3OOH; CH_3NH_2/CO_2-laser	[103]
	MS	(electron impact ionization) (NH^+)	[3, 4, 328]
NH $(a\,^1\Delta)$ $(b\,^1\Sigma^+)$	LIF	$(c\,^1\Pi-a\,^1\Delta)$ $\lambda=324$ nm, dye/N_2-laser (doubled)	[363]
	ChL	NH $(^1\Delta)+HN_3$ $(^1A')$–NH_2 $(^2A_1)+N_3$ $(^2\Pi_g)$ NH_2 $(^2A_1)$–NH_2 $(^2B_1)+h\nu$ chemiluminescence	[329]
	E	NH $(b\,^1\Sigma^+)$ from VUV photolysis of NH_3 $(b\,^1\Sigma)$–$(X\,^3\Sigma)+h\nu$ $\lambda=470{\cdot}71$ nm	[330]
	MS	$(a\,^1\Delta)$ (electron impact ionization) (NH^+)	[328]
NF $(X\,^3\Sigma^-)$	A–VIS–IR	$(a\,^1\Delta-X\,^3\Sigma^-)$ $\lambda=874{\cdot}2$ nm $(b\,^1\Sigma^+-X\,^3\Sigma^-)$ $(0,0)$ $\lambda=528{\cdot}8$ nm	[336]
	E	NF $(b\,^1\Sigma^+)$–$(X\,^3\Sigma^-)$ $(0,0)$ $(0,1)$ $(1,1)$ NF $(b\,^1\Sigma^+)$ from energy transfer: NF $(a\,^1\Delta)+O_2$ $(a\,^1\Delta_g)\rightarrow$ NF $(b\,^1\Sigma^+)+O_2\,(^3\Sigma_g^-)$	[123]
	ESR	NF $(^3\Sigma^-)$, X-band	[5]
	MS	(electron impact ionization) (NF^+)	[4]
NF $(a\,^1\Delta)$ $(b\,^1\Sigma^+)$	E	NF $(a\,^1\Delta)$ from $H+NF_2$ $(a\,^1\Delta-X\,^3\Sigma)$ $(0,0)$ $(1,1)$ $(2,2)$ $870\leqslant\lambda/\mathrm{nm}\leqslant880$ NF $(b\,^1\Sigma^+-X\,^3\Sigma)$ $(0,0)$ $\lambda=530$ nm	[121, 122]
	LMR	$(a\,^1\Delta)$ $J=7\rightarrow8$, $\lambda=513\,\mu$m CHOOH/CO_2-laser	[108]
	ESR	NF $(a\,^1\Delta)$, X-band	[5]
NCl $(X\,^3\Sigma^-)$ $(b\,^1\Sigma^+)$	A–VIS	$(b\,^1\Sigma^+-X\,^3\Sigma^-)$ $(0,0)$ $\lambda=664{\cdot}67$ nm	[336]
	E	(NCl $b\,^1\Sigma^+$) $(0\leqslant v''\leqslant7)$ $(b\,^1\Sigma^+-X\,^3\Sigma^-)$ $\lambda\ (0,0)=664$ nm; NCl $(b\,^1\Sigma)$ from: $Cl+N_3\rightarrow NCl^*+N_2$	[262]
	MS	(electron impact ionization) (NCl^+)	[82]
NBr $(X\,^3\Sigma^-)$	A–VIS	$(b\,^1\Sigma^+-X\,^3\Sigma^-)$ $550\leqslant\lambda/\mathrm{nm}\leqslant650$	[336]
	E	$(A\,^1\Sigma^+-X\,^3\Sigma^-)$ $(0,0)$ $(1,0)$ $\lambda\ (0,0)=665$ nm NBr $(^1\Sigma)$ from: $Br+N_3\rightarrow NBr^*+N_2$	[262]

Atom, radical	Method	Explanations	References
NH_2 ($\tilde{X}\,^2B_1$)	RA–L	($\tilde{A}\,^2A_1$–$\tilde{X}\,^2B_1$) (0, 9, 0–0, 0, 0) $^PQ_{1,7}$ $\lambda=597{\cdot}375$ nm, dye flashl.-laser $f_i=2{\cdot}04\times10^{-4}$ NH_2/O_2-flame atm. pressure $\varepsilon_{max}=8{\cdot}40\times10^3$ l/mol cm, 4 bar NH_3 c.w. dye-laser	[95, 102]
	A–IC	$600\leqslant\lambda/nm\leqslant610$ dye flashl.-laser	[93]
	LIF	($\tilde{A}\,^2A_1$–$\tilde{X}\,^2B_1$) (0, 9, 0)–(0, 0, 0) $\lambda=597{\cdot}8$ nm c.w. dye/Ar^+ ion-laser; sensitivity: [NH_2 ($\tilde{X}\,^2B_1$)] $\geqslant10^{-16}$ mol/cm^3	[92, 94]
	LMR	($\tilde{X}\,^2B_1$)ν_2 $0\leqslant N\leqslant7$, $0\leqslant K_a\leqslant5$ CO-laser	[98, 100]
		$^2B_1$ (2_{20}–3_{31}) $\lambda=78$ μm, (1_{11}–2_{20}) $\lambda=118{\cdot}6$ μm H_2O laser 7_{35}–7_{44} ($\lambda=108$ μm D_2O-laser)	[99]
	MS	(electron impact ionization) (NH_2^+)	[3, 4]
ND_2 ($\tilde{X}\,^2B_1$)	LMR	ND_2 $^2B_1\nu_2$ $9\leqslant\lambda/\mu m\leqslant10$ CO_2-laser	[97]
NHD ($\tilde{X}\,^2A''$)		NHD ($\tilde{X}\,^2A''$) (0, 0, 0) $3_{13}\leftarrow2_{02}$ and $4_{13}\leftarrow3_{22}$ $\lambda/\mu m=211{\cdot}263$; $374{\cdot}086$ FIR/CO_2-laser	[98]
NHF ($\tilde{X}\,^2A''$)	A	$\tilde{A}\,^2A'$ (0, v_2', 0)–$\tilde{X}\,^2A''$ (0, 0, 0) $0\leqslant v_2'\leqslant3$ $400\leqslant\lambda/nm\leqslant500$	[389, 390]
	E	$\tilde{A}\,^2A'$ (0, 0, 0)–$\tilde{X}\,^2A''$ (0, v_2'', 0) $0\leqslant v_2''\leqslant3$ $450\leqslant\lambda/nm\leqslant650$ NHF ($\tilde{A}\,^2A'$) in N_2H_4-F_2-flames	[391]
NF_2 ($\tilde{X}\,^2B_1$)	A–UV	($\tilde{A}$–$\tilde{X}$) $235\leqslant\lambda/nm\leqslant280$	[147]
	A–IR–DL	($\tilde{X}\,^2B_1$) ν_1; ν_3 $\lambda/\mu m=9{\cdot}091$; $10{\cdot}75$	[101]
	LMR	NF_2 ($\tilde{X}$) ν_1, $\lambda=9{\cdot}4$ μm CO_2-laser	[96]
	ESR	X-band magnetic dipole transition (weak)	[5]
	MS	(electron impact-, photoionization) (NF_2^+)	[3, 79]
NCl_2 ($\tilde{X}\,^2B_1$)	MS	(electron impact ionization) (NCl_2^+)	[82]
N_2H_3 (X)	MS	(electron impact ionization) ($N_2H_3^+$)	[4]
NO ($X\,^2\Pi_{1/2,3/2}$)	A, E	$B\,^2\Pi$–$X\,^2\Pi$ (β-bands) $650\leqslant\lambda/nm\geqslant200$	[336]
		$A\,^2\Sigma^+$–$X\,^2\Pi$ (γ-bands) $340\geqslant\lambda/nm\geqslant195$ $C\,^2\Pi$–$X\,^2\Pi$ (δ-bands) $210\geqslant\lambda/nm\geqslant184$ $D\,^2\Sigma^+$–$X\,^2\Pi$ (ε-bands) $190\geqslant\lambda/nm\geqslant170$ $B'\,^2\Delta$–$X\,^2\Pi$ (β'-bands) $200\geqslant\lambda/nm\geqslant140$	[364]
	A–LR	$5{\cdot}17\leqslant\lambda/\mu m\leqslant5{\cdot}57$ CO-laser (White cell)	[265]
	LIF	($A\,^2\Sigma^+$, v')–($X\,^2\Pi$, v'') (v', v'') = (0, 0), (0, 1), (0, 2) $222\leqslant\lambda/nm\leqslant300$ dye/N_2-laser (doubled)	[263, 269]
	LMR	($^2\Pi_{1/2}$, $J=25/2$)–($^2\Pi_{3/2}$, $J=23/2$) $\lambda=78$; 79; 119 μm H_2O-discharge-laser	[264]
	ESR	$^2\Pi_{3/2}$, $J=3/2$ X-band magnetic and electric dipole transitions (substance to titrate the filling factor for electric dipole transitions)	[5, 266]
	MS	(electron impact ionization, photoionization) resonant photoionization (NO^+)	[3]
	MPI	enhanced by resonance two-photonionization $270\leqslant\lambda/nm\leqslant317$ dye/N_2-laser (doubled), sensitivity: [NO] $\geqslant$ 1 p.p.m. atm. flame	[270]
NO_2 ($\tilde{X}\,^2A_1$)	A UV–VIS–IR	$\tilde{A}$($^2B_2$)–$\tilde{X}\,^2A_1$ $1000\geqslant\lambda/nm\geqslant320$ (transitions to the lowest state only are mentioned)	[275]
	LIF	$\tilde{A}\,^2B_2$–$\tilde{X}\,^2A_1$ $250\leqslant\lambda/nm\leqslant900$ lasers used: dye (tunable),	[74, 276]
		He-Cd (441·6 nm), Ar^+ (514·5 nm) Kr^+ Nd : YAG (532 nm)	[274]

Atom, radical	Method	Explanations	References
	LMR	$\tilde{X}\,^2A_1$, $\nu_2(v_2=1\text{–}0)\ ^rR_6(\mathrm{N})\text{–}^rR_{11}(\mathrm{N})$ CO_2-laser $\tilde{X}\,^2A_1$, $\nu_3\,^QQ_4$ (4), $\lambda=6{\cdot}202\ \mu$m–CO-laser	[277, 278]
	ESR	$(^2A_1)$ X-band magnetic dipole transitions (very broad and weak lines)	[5]
	MS	(electron impact- and photoionization) (NO_2^+)	[3]
NO_3 ($\tilde{X}$)	A–VIS	$A\text{–}X$ $\nu_1(0\text{–}0)$ $\lambda=662$ nm $\nu_1(1\text{–}0)$ $\lambda=627$ nm ε (662 nm) $=1\times10^4$ l/mol cm	[393, 394]
HNO ($\tilde{X}\,^1A'$)	A–VIS	$(^1A''\text{–}^1A')$ $650\leqslant\lambda/\mathrm{nm}\leqslant770$	[280]
DNO ($\tilde{X}\,^1A'$)	A–IC	$(^1A''\text{–}^1A')$ $(1,0,0)\text{–}(0,0,0)$ $K'=4\text{–}K''=3$, $\varepsilon=20$ l/mol cm flashl. rhodamine B-dye-laser	[279]
	LIF	$\tilde{A}\,^1A''$ $(0,1,0)\text{–}\tilde{X}\,^1A'$ $(0,0,0)$ $\lambda=740$ nm $(0,0,1)\text{–}(0,0,0)$ $\lambda=710$ nm $(0,1,1)\text{–}(0,0,0)$ $\lambda=640$ nm Oxazin 720, Rhodamine 640 dye flashl.-laser	[213]
	E	(HNO $(^1A'')$) from: $O(^3P)/O_2+NO+C_2H_2$, C_2H_4, C_3H_6, CH_2O, CH_3CHO DNO $(^1A'')$ from: O $(^3P)/O_2+NO+C_2D_4$	[281]
	MS	(electron impact ionization) (DNO^+)	[3]
P $(^4S_{3/2})$	RA	$3p^24s(^4P_{5/2,3/2,1/2})\text{–}3p^3(^4S_{3/2})$ $\lambda/\mathrm{nm}=177{\cdot}5$; 178·29; 178·77, measured at: $\lambda=177{\cdot}5$ nm	[116]
	RF	$3p^24s$ $(^4P_{1/2,3/2,5/2})\text{–}3p^3(^4S_{3/2})$ $\lambda/\mathrm{nm}=178{\cdot}77$; 178·29 and 177·50	[112]
	ESR	$(^4S_{3/2})$ X-band (analogous ^{15}N)	[5]
	MS	(electron impact ionization) (P^+) (P^{2+})	[3]
PH ($\tilde{X}\,^3\Sigma^-$) ($a\,^1\Delta$)	A–UV	$A\,^3\Pi\text{–}X\,^3\Sigma$ $318\leqslant\lambda/\mathrm{nm}\leqslant360$ λ (0, 0) nm $=342{\cdot}68$ (P_1)	[336]
	LMR	$(^3\Sigma^-)$ $J=4\text{–}5$, $\lambda=118{\cdot}6\ \mu$m H_2O-discharge-laser	[109, 111]
	LMR	($a\,^1\Delta$), $J=4\text{–}5$, $\lambda=118{\cdot}6\ \mu$m H_2O-discharge-laser	[109, 111]
PD ($X\,^3\Sigma^-$)	LMR	($X\,^3\Sigma^-$) $v=1\text{–}0$ (N', $N''=3$) $\lambda/\mu\mathrm{m}=5{\cdot}95\text{–}6{\cdot}15$ CO-laser	[110]
PF ($X\,^3\Sigma^-$)	A–UV	($B\,^3\Pi\text{–}X\,^3\Sigma^-$) $300\leqslant\lambda/\mathrm{nm}\leqslant400$ ($b\,^1\Sigma^+\text{–}X\,^3\Sigma^-$) $744{\cdot}4\leqslant\lambda/\mathrm{nm}\leqslant748{\cdot}3$ λ (0, 0) $=748{\cdot}308$ nm	[336]
	E	($b\,^1\Sigma^+\text{–}X\,^3\Sigma^-$) λ (0, 0) $=748$ nm $NF(^1\Sigma^+)$ from: $Ar(^3P_{0,2})+NF(^3\Sigma)$	[124]
PCl ($X\,^3\Sigma$)	A–UV	$B\text{–}X$, $230\leqslant\lambda/\mathrm{nm}\leqslant248{\cdot}8$ λ (0, 0) $=241{\cdot}9$ nm $C\text{–}X$, $224{\cdot}3\leqslant\lambda/\mathrm{nm}\leqslant233{\cdot}6$ λ (0, 0) $=230{\cdot}7$ nm PCl from PCl_3-photolysis	[273]
PH_2 ($\tilde{X}\,^2B_1$) PD_2 ($\tilde{X}\,^2B_1$)	A–VIS	PH_2 $\tilde{A}\,^2A_1$ $(0,v_2',0)\text{–}\tilde{X}\,^2B_1\text{–}(0,0,0)$ $1\leqslant v_2'\leqslant8$ $(0,0,0)\text{–}(0,0,0)$ $\lambda=547{\cdot}1$ nm	[365]
	A–VIS	$PD_2(\tilde{A}\,^2A_1\text{–}\tilde{X}\,^2B_1)$ $(v_2'\text{–}0)$ $1\leqslant v_2'\leqslant10$	[130]
	LIF	$(\tilde{A}\,^2A_1\text{–}\tilde{X}\,^2B_1)$ $(0,v_2',0)\text{–}(0,0,0)$, $0\leqslant v_2'\leqslant5$, $436\leqslant\lambda/\mathrm{nm}\leqslant551$, dye/$N_2$-laser	[104, 107]
	LMR	$(\tilde{X}\,^2B_1)$ (ν_2) $9{\cdot}0\leqslant\lambda/\mu\mathrm{m}\leqslant9{\cdot}6$ CO_2-laser $(\tilde{X}\,^2B_1)$ $(0,0,0)$ $(6_{43}\text{–}5_{32})$ rot.-transition $\lambda=118{\cdot}6\ \mu$m, H_2O-laser	[105, 106]
	MS	(electron impact ionization) (PH_2^+)	[3]

Atom, radical	Method	Explanations	References
PF_2 ($\tilde{X}\,^2B_1$)			
PO ($X\,^2\Pi_{1/2,\,3/2}$)	LIF	($B\,^2\Sigma^+$–$X\,^2\Pi_{1/2}$) (0,0), $\lambda = 325$ nm, cresylviolet-dye/Nd-YAG laser (doubled)	[271]
	MPI	laser induced two photon ionization with as resonant intermediate state $B(^2\Sigma^+)$ $302 \leqslant \lambda/\mathrm{nm} \leqslant 334$, sensitivity: $x_{PO} \geqslant 10^{-8}$ C_2H_2-air-flame	[272]
As ($^4S_{3/2}$)	RA	As $4p^3(^4S_{3/2})$–$5s$ ($^4P_{5/2}$)–$4p^3(^4S_{3/2})$ $\lambda = 189{\cdot}0$ nm	[117]
	RF	$4p^2$ $5s$ ($^4P_{5/2,\,3/2,\,1/2}$)–$4p^3(^4S_{3/2})$, $\lambda/\mathrm{nm} = 189{\cdot}04$, $193{\cdot}76$ and $197{\cdot}23$	[113]
	ESR	($^4S_{3/2}$) (X-band)	[5]
AsO ($X\,^2\Pi_{3/2}$)	E	$A\,^2\Sigma^+$–$X\,^2\Pi$ ($0 \leqslant v \leqslant 5$), $295 \leqslant \lambda/\mathrm{nm} \leqslant 345$ ($G\,^2\Pi$–$X\,^2\Pi$, $H\,^2\Pi$–$X\,^2\Pi$, $D\,^2\Sigma^-$–$X\,^2\Pi$, $C\,^2\Delta$–$X\,^2\Pi$)	[268]
	LMR	$X\,^2\Pi_{3/2}(v = 1 \leftarrow 0)$, $\lambda = 10{\cdot}45$ μm, CO_2, 10 P (14) laser-line	[267]
AsS ($X\,^2\Pi$)	MS	(electron impact ionization) (As S^+)	[388]
Sb, Bi ($^4S_{3/2}$)	RA	Sb(6($^4P_{1/2}$)–5($^4S_{3/2}$)) $\lambda = 231{\cdot}3$ nm Bi(7($^4P_{1/2}$)–6($^4S_{3/2}$)) $\lambda = 306{\cdot}77$ nm	[118, 119]
	RF	Sb(6($^4P_{1/2}$)–5($^4S_{3/2}$)) $\lambda = 231{\cdot}3$ nm Bi(7($^4P_{1/2}$)–6($^4S_{3/2}$)) $\lambda = 306{\cdot}77$ nm	[114, 115]
	ESR	(^{121}Sb, ($^4S_{3/2}$) $I = 5/2$) (^{123}Sb, ($^4S_{3/2}$) $I = 7/2$) (X-band)	[5]
S ($^3P_2$), ($^1D_2$)	A–UV	($S\,^3P_2$) ($4s(^3S_1)$–$3p^4(3P_2)$) $\lambda = 180{\cdot}73$ nm (strong absorption) S ($^1D_2$) (from OCS 200–260 nm) ($3p^34s$ ($^1D_2$)–$3p^4$ ($^1D_2$)) $\lambda = 166{\cdot}67$ nm (H_2S/He discharge-lamps) sensitivity: [S (3 $^1D_2$)] $\geqslant 3 \times 10^{-14}$ mol/cm^3 $S/N = 1$	[282, 283, 284]
	RF	(^3P_J) ($4s$ $^3S_1$–$3p^4$ ^3P_J) $\lambda = 181$ nm (non-resolved triplet)	[285]
	ESR	($^3P_{1,2}$) X-band (analogous O($^3P_{1,2}$))	[5]
	MS	(electron impact ionization) (S^+)	[3]
S_2 ($^3\Sigma_g^-$)	A–UV–VIS	$B\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$ $240 \leqslant \lambda/\mathrm{nm} \leqslant 711$, $\lambda_{0,0} = 315{\cdot}5$ nm $C\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$ $165 \leqslant \lambda/\mathrm{nm} \leqslant 187$, $\lambda_{0,0} = 179{\cdot}6$ nm $D\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$ $165 \leqslant \lambda/\mathrm{nm} \leqslant 175$ $\lambda_{0,0} = 170$ nm	[335]
	LIF	($B\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$) ($v' = 3$, $N' = 24$, $J' = 25$)–($v'' = 3$, $N'' = 25$, $J'' = 26$) $\lambda = 325$ nm c.w.-He-Cd-laser ($B\,^3\Sigma_u^-$–$X\,^3\Sigma_g^-$) $\lambda = 337$ nm N_2-laser	[287]
	ESR	$^{32}S_2$, ^{34}S (^{32}S natural abundance) ($X\,^3\Sigma_g^-$) X-band (9 GHz) magnetic dipole transitions	[286]
	MS	photoionization (S_2^+)	[3]
SH, SD ($X\,^2\Pi_i$)	A–UV	SH, SD, ($A\,^2\Sigma^+$–$X\,^2\Pi_i$) (0,0) (1,0) $245 \leqslant \lambda/\mathrm{nm} \leqslant 330$ SH $^2\Pi_{1/2}$ (0,0) band head λ (R_2) $= 232{\cdot}66$ nm, λ (Q_2) $= 327{\cdot}93$ nm	[289]
	LIF	($^2\Sigma^+$–$^2\Pi_{3/2,\,1/2}$), $323{\cdot}6 \leqslant \lambda/\mathrm{nm} \leqslant 325{\cdot}7$ flashl. dye-laser (doubled) $320 \leqslant \lambda/\mathrm{nm} \leqslant 334$	[127, 128]
	E	$A\,^2\Sigma^+$ from ($SOCl_2$, $CHCl_3$, $CDCl_3$) K-vapour–low pressure diffusion flame $A\,^2\Sigma^+$–$X\,^2\Pi$ SH (0,0) and SD (0,0) (0,1) respectively.	[290]

Atom, radical	Method	Explanations	References
	LMR	(SD, $X\,^2\Pi_{3/2}$, $v=1\leftarrow 0$) $\lambda/\mu\mathrm{m}=5{\cdot}303$; $5{\cdot}373$ CO-laser sensitivity: $[\mathrm{SD}]\geqslant 3\times 10^{-15}$ mol/cm^3 ^{32}SH, ^{34}SH $^2\Pi$, $J=3/2\rightarrow 5/2$, ^{32}SD $^2\Pi$ $J=5/2\rightarrow 7/2$ $\lambda/\mu\mathrm{m}=216{\cdot}295$ (CH_3OD)/CO_2-FIR-laser	[129, 291]
	ESR	^{32,33}SH (SD) $^2\Pi_{3/2}$ (X-band) electric dipole transitions (OH analogous)	[5, 367, 368]
	MS	electron impact ionization (SH^+)	[3, 4]
SF ($X\,^2\Pi_{3/2}$)	ESR	$^2\Pi_{3/2}$ $J=3/2$ (X-band) electric dipole transitions	[5]
	MS	electron impact ionization (SF^+)	[3, 79]
SF_2 ($\tilde{X}$)	MS	electron impact ionization (SF_2^+)	[3, 79]
SO ($X\,^3\Sigma^-$)	A–UV	$B\,^3\Sigma^- - X\,^3\Sigma^-$ $190\leqslant\lambda/\mathrm{nm}\leqslant 457$	[336]
($a\,^1\Delta$)		$A\,^3\Pi - X\,^3\Sigma^-$ $240\leqslant\lambda/\mathrm{nm}\leqslant 263$	[295]
	LIF	($A\,^3\Pi - X\,^3\Sigma$) $0\leqslant v'\leqslant 6$, $246\leqslant\lambda/\mathrm{nm}\leqslant 262$ Dye (doubled)/Nd-YAG (tripled)	[297]
	ChL	SO ($^1\Delta_g$, $^1\Sigma_g^+$) SO ($^1\Delta - {}^3\Sigma^-$) λ $(0,0)=1{\cdot}705\,\mu\mathrm{m}$ SO ($^1\Delta$) from energy transfer with O_2 ($^1\Delta_g$).	[298]
	LMR	($X\,^3\Sigma^- v=1\leftarrow 0$) $N=-1$, $\Delta J=0$ $\lambda=9{\cdot}09\,\mu\mathrm{m}$ CO_2-laser	[300]
	ESR	($^3\Sigma^-$) X-band, electric dipole transitions ($0\leqslant v\leqslant 6$) ^{34}SO, ^{33}SO ^{32,33}SO ($^1\Delta$) X-band electric dipole transitions	[5, 299]
	MS	(electron impact- and photoionization) (SO^+)	[3, 294, 296]
HSO ($\tilde{X}\,^2A''$)	LIF	($\tilde{A}\,^2A' - \tilde{X}\,^2A''$) (0, 0, 3)–(0, 0, 0)	[305, 306]
DSO ($\tilde{X}\,^2A''$)		(0, 0, 4)–(0, 0, 0) $570\leqslant\lambda/\mathrm{nm}\leqslant 620$ rhodamine 6G, kiton-red S dye/Ar^+ and flashl. pump respectively	
	ChL	($\tilde{A}\,^2A'$) from: SH (SD) $+ O_3 \rightarrow$ HSO (DSO) ($\tilde{A}\,^2A'$) $+ O_2$ $570\leqslant\lambda/\mathrm{nm}\leqslant 700$	[304, 305]
SN ($X\,^2\Pi$)	A–UV	$A - X\,^2\Pi$ at $\lambda=250$ nm	[317]
		$C\,^2\Sigma - X\,^2\Pi$ at $\lambda=230$ nm (strong absorption)	[314]
Se, Te (^3P_J)	RA	Se ($4\,^3P_0$) $5\,^3S_1 - 4\,^3P_0$ $\lambda=206{\cdot}3$ nm	[373]
		Te ($5\,^3P_1$) ($5\,^3P_0$) $210\leqslant\lambda/\mathrm{nm}\leqslant 240$	[372]
	ESR	Se ($^3P_1$, $^3P_2$) X-band (O ^3P_J) analogues ^{77}Se $I=1/2$ Te ($^3P_2$) observed	[5]
	MS	Knudsen cell electron impact ionization (Se^+)	[370]
Se_2 ($X\,^3\Sigma^-$)	A	$B\,^3\Sigma_u^- - X\,^3\Sigma_g^-$ $325\leqslant\lambda/\mathrm{nm}\leqslant 670$	[335]
	LIF	$B\,^3\Sigma_u^- - X\,^3\Sigma_g^-$ $\lambda/\mathrm{nm}=514{\cdot}5$; $351{\cdot}1$ Ar-laser; 413·1 nm Kr-laser (and dye-laser)	[288]
	MS	Knudsen cell electron impact- and photoionization (Se_2^+)	[3, 370]
SeH, SeD ($^2\Pi_{3/2}$)	A–UV	SeH, SeD, $300\leqslant\lambda/\mathrm{nm}\leqslant 325$	[293]
	LMR	^{76,77,78,80,82}SeH ($^2\Pi_{3/2}$) $J=5/2-7/2$ $\lambda=194\,\mu\mathrm{m}$ CD_3OD/CO_2-laser ($^2\Pi_{1/2} - {}^2\Pi_{3/2}$) $J=3/2-3/2$, $M_J=3/2-3/2$ $\lambda=5{\cdot}62\,\mu\mathrm{m}$ CO-laser ^{74,76,77,78,80,82}SeD $X\,^2\Pi_{3/2}$ $J=3/2-5/2$ $\lambda=513\,\mu\mathrm{m}$ $CHOOH/CO_2$-laser	[292]
	ESR	SeH (SeD) $^2\Pi_{3/2}$ (X-band)	[5]
SeF ($X\,^2\Pi_{3/2}$)	ESR	($^2\Pi_{3/2}$) (X-band) analogous SF $^2\Pi_{3/2}$	[5]
SeO ($X\,^3\Sigma^-$)	A–UV	($A\,^3\Sigma^- - X\,^3\Sigma^-$) $292\leqslant\lambda/\mathrm{nm}\leqslant 510$	[302, 336]

Atom, radical	Method	Explanations	References
$(a\,^1\Delta)$	LMR	^{76,77,78,80,82}SeO $(X\,^3\Sigma^-)$ $(v=1\leftarrow 0)$ $\lambda = 10\,\mu$m $P(6)$–$P(12)$ (^{13}C$\,^{16}$O$_2$-laser line)	[301]
	ESR	($^3\Sigma$) (X-band) analogous SO ($^3\Sigma$)	[5]
SeS $(X\,^3\Sigma^-)$	LIF	(^{78,80}Se^{32}S) (B–X) $(0 \leqslant v \leqslant 4)$ $(1 \leqslant v' \leqslant 14)$ $350{\cdot}9 \leqslant \lambda/\mathrm{nm} \leqslant 476{\cdot}6$ Ar$^+$, Kr^{2+}-laser	[303]
	MS	Knudsen cell electron impact ionization (SeS$^+$)	[371]
SeN $(^2\Pi_{3/2})$	LMR	$(^2\Pi_{3/2})$ $v=1\leftarrow 0$ CO$_2$-laser	[374]
TeH $(X\,^2\Pi_{3/2})$	ESR	$^2\Pi_{3/2}$, $J=3/2$ X-band	[5]
TeO $(X\,^3\Sigma)$	MS	(electron impact ionization) (TeO$^+$)	[3]
BH $(X\,^2\Sigma^+)$	A–UV–VIS E	$(A\,^1\Pi$–$X\,^1\Sigma^+)$ $394{\cdot}5 \leqslant \lambda/\mathrm{nm} \leqslant 460$ $\lambda\ (0,0) = 433{\cdot}3$ nm $(B\,^1\Sigma^+$–$X\,^1\Sigma^+)$ $182{\cdot}6 \leqslant \lambda/\mathrm{nm} \leqslant 192{\cdot}6$ $\lambda\ (0,0) = 190{\cdot}99$ nm (further transitions at shorter wavelengths)	[336]
	LIF	$(A\,^1\Pi,\ v'=0,\ J'=1,4,7)$ $430 \leqslant \lambda/\mathrm{nm} \leqslant 437$ dye/N$_2$-laser	[307]
	MS	(electron impact ionization) (BH$^+$)	[3]
BH$_2$ $(\tilde{X}\,^2\Pi_u)$	MS	(electron impact ionization) (BH$_2^+$)	[3]
BO $(X\,^2\Sigma^+)$	E	$(A\,^2\Pi$–$X\,^2\Sigma^+)$ $310 \leqslant \lambda/\mathrm{nm} \leqslant 850$ $\lambda\ (0,0) = 425{\cdot}0$ nm $(B\,^2\Sigma^+$–$X\,^2\Sigma^+)$ $210 \leqslant \lambda/\mathrm{nm} \leqslant 370$, $\lambda\ (0,0) = 233{\cdot}1$ nm $(C\,^2\Pi$–$X\,^2\Sigma^+)$ $163 \leqslant \lambda/\mathrm{nm} \leqslant 204$, $\lambda\ (0,0) = 181{\cdot}5$ nm	[336]
	LIF	$(A\,^2\Pi$–$X\,^2\Sigma^+)$ $11 \geqslant v' \geqslant 0$ $277 \leqslant \lambda/\mathrm{nm} \leqslant 425$ dye/YAG-laser	[308, 309]
	MS	(electron impact ionization) (BO$^+$)	[3]
BN $(X\,^3\Pi)$	E	$(A\,^3\Pi$–$X\,^3\Pi)$ $340 \leqslant \lambda/\mathrm{nm} \leqslant 385$ $\lambda\ (0,0) = 359{\cdot}9$ nm	[336]
BO$_2$ $(\tilde{X}\,^2\Pi_g)$	LIF	$(\tilde{A}\,^2\Pi_u\ (0,0,2)$–$\tilde{X}\,^2\Pi_g)$ $\lambda = 434$ nm dye/YAG-laser; c.w.-dye-laser, ion-laser	[308, 375]
Cd (^3P_J)	LIF	Cd $(^1P_1$–$^3P_J)$ $\lambda = 228{\cdot}8$ nm dye/N$_2$-laser (doubled)	[322]
	MS	(electron impact ionization) (Cd$^+$)	[3]
Hg $(6(^1S_0))$	RA	$6(^3P_1)$–$6(^1S_0)$ $\lambda = 253{\cdot}7$ nm	[318, 321]
$(6(^3P_{0,1,2}))$	LIF	$(^3P_0$–$7(^3S_1))$ $\lambda_L = 404{\cdot}7$ nm, $\lambda_F = 546{\cdot}1$ nm	[320]
	LMR	6s 6p $(^3P_1$–$^3P_0)$ $\lambda = 5{\cdot}658\,\mu$m CO-laser	[319]
	MS	(electron impact- and photoionization) (Hg$^+$, Hg$^{2+\cdots 6+}$)	[3]
UF$_5$ $(\tilde{X})$	A–UV–VIS	(UF$_5$) $350 \leqslant \lambda/\mathrm{nm} \leqslant 700$ ε (400 nm) = 600 l/mol cm	[126]
	AJR	diode laser 16–17 μm	[323]
	LIF	(UF$_5$) 17 emission lines $682{\cdot}7 \leqslant \lambda/\mathrm{nm} \leqslant 877$ after KrF-laser photolysis	[324]

Abbreviation used in tables 1 and 2

A	absorption
ICA	intracavity absorption
RA	resonance absorption
LRA	laser resonance absorption
RF	resonance fluorescence

LIF	laser induced fluorescence
LISF	laser induced saturation fluorescence
TALIF	two photon-absorption laser induced fluorescence
LMR	laser magnetic resonance
LER	laser electric resonance
LOG	laser-optogalvanic absorption
ESR	electron spin resonance
DL	diode laser
MW	microwave absorption
E	emission
ChL	chemiluminescence
MS	mass spectrometer
MPI	multiphoton ionization
PES	photoelectron spectroscopy
SI	surface ionization (Langmuir–Taylor detector)
CCL	colour centre laser
CARS	coherent anti-Stokes Raman scattering

Indirect detection methods (e.g. probes taken from the gas phase and frozen in a low-temperature matrix (e.g. (327)) are not included in tables 1 and 2.

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