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DETECTOR CALIBRATION WITH LASERS - A REVIEW

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ABSTRACT

UV lasers have proven to be a simple but powerful and versatile tool for calibration and surveying of drift chambers and for precision measurements of electron transport parameters in gases. Much information has been added recently, in particular on the performance of various dopants with high 2-photon ionisation cross sections and a great variety of applications. Experimental results and theoretical ideas will be reviewed in detail. Applications of lasers to bubble chambers and Si-strip detectors will only be touched briefly.

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(Somewhat enlarged version)

1. INTRODUCTION

Tracking detectors in particle physics have reached such a scale and complexity that calibration and monitoring has become a formidable task. This is particularly true for multiwire proportional and drift chambers where long beam runs are required to evaluate a large set of chamber parameters; even then stability is difficult to monitor. Here, the advent of laser beams capable of imitating particle tracks, has brought a significant improvement. The principal advantages of UV lasers for the production of ionisation tracks in gases are:

- Cheap, transportable and flexible test beam set-up.
- Good resolution in space and time over several meters: no production of δ -rays, no multiple scattering.
- No curvature in magnetic fields.
- Ionisation density controllable with small fluctuations (Poisson instead of Landau statistics).
- Ideal tool for two-track studies, e.g. via simple beam reflection.

In the past few years, the field has expanded very rapidly. In the first attempt in 1979, ionisation from a N_2 laser was detected only after addition of Nickelocene [1(a)]. Shortly afterwards, signals were also detected in undoped chambers: 50 electrons/cm for about 100 μJ in 1 mm² with a focused N_2 laser and more than 400 electrons/cm for 1 μJ in 0.1 mm² at shorter wavelength (Nd Yag laser) [2]. The addition of 0.07 Torr of diethylaniline increased the ionisation with the N_2 laser by a factor of 10. First evidence for a 2-photon process is seen in the quadratic dependence of ionisation on photon flux.

In parallel, a special 2-stage N_2 laser had been developed to provide a low divergence (0.8 mrad) beam with high intensity, a technique later used in most N_2 lasers for detector calibration^(*). This laser produced high quality, long tracks: > 100 electrons/cm over more than one meter [3]. Single track resolution of < 50 μm was achieved with chamber pulseheight variation of only 9% fwhm.

In an attempt to produce ionisation tracks with visible light, 3-photon resonances were observed at 428.7 and 447.2 nm with 4-amino-azobenzene as additive [1(b)]. For the standard process, i.e. 2-photon ionisation, it was

(*) E.g. lasers from Multilasers, Geneva, Switzerland.

demonstrated that the quadratic dependence of ionisation on photon flux is turning into linear dependence at high flux, due to saturation effects [6]. Various groups started to construct and study the performance of special N_2 lasers in test chambers [7-13,24] apply them to large detectors in experiments [4,5], and compare ionisation at different photon energies with N_2 , Nd and excimer lasers [14,17,24]. Additives were studied systematically [15-19,21,22,24-27] and a detailed frequency scan was carried out on some of them [17-19]. Relatively clean systems were constructed to investigate the origin of ionisation in undoped chambers [24] and the consequences of gas cleaning [20,24]. Finally, phenol and toluene have been identified as trace impurities in some chambers [18,19].

Some of these results will be presented after a short discussion on the principles of ionisation in gases by UV lasers and a summary of laser characteristics. In the last section, application of lasers to bubble chambers and Si detectors will be discussed briefly.

2. GASEOUS DETECTORS

2.1 Basic Principles

UV-laser beams are used to produce ionisation tracks in the gas. Typical chamber gases have rather high ionisation energies: Ar: 15.7, CO_2 : 14.4, CH_4 : 13.1, C_2H_6 : 11.6 eV, much higher than photon energies of standard UV lasers: N_2 : 3.68 eV, Nd-Yag: 4.68 eV (frequency quadrupled).

As the cross section for 3-4 photon ionisation is extremely low, one has to search for substances with high cross section σ_i for 2-photon ionisation. Their ionisation potential has to be I.P. ≤ 9.4 eV and 7.4 eV in case of Nd-Yag and N_2 lasers, respectively. In addition, the single photon excitation level should be a real state or a virtual state with strong coupling to a real state.

Such substances do indeed exist. Composite molecules have a very large number of vibro-rotational states which at higher vibrational quantum numbers may overlap and form a quasi-continuum. Typical line-widths are 0.03 cm^{-1} . Large molecules may have 1000 transitions per cm^{-1} , i.e. 30 or more lines per unit line width [28(a)]. Obviously, this quasi-continuum may still have substantial structure. As an example, the 2600 Å system of benzene is shown in fig. 1 [28(b)].

Relevant processes are sketched in fig. 2, the most dominant ones being probably B and also D, if the singlet to triplet transition rate $S_1 \rightarrow T_1$ is high [22]. Typical cross sections are $\sigma_e \leq 10^{-16} - 10^{-20} \text{ cm}^2$ for excitation of S_1 or nearby levels S_1^* ; $\sigma_i \sim 10^{-18} \text{ cm}^2$ for single photon ionisation from S_0 near threshold (max. $\sigma_i \sim (2-6) \times 10^{-17} \text{ cm}^2$ far from threshold); $\sigma_i^* \sim \sigma_i$. For the lifetimes one expects: $\tau_e \sim 10^{-7} - 10^{-10} \text{ s}$; $\tau^* \sim 10^{-11} - 10^{-12} \text{ s}$; $\tau_T \geq 10^{-4} \text{ s}$ [22].

We set $N = \text{flux of photons/cm}^2\text{sec}$, n_o , $n_e(t)$, $n_i(t) = \text{density of molecules/cm}^3$ in the ground, excited and ionized state.

Then

$$\begin{aligned} dn_i/dt &= n_e \sigma_i^* N \quad (\text{assuming negligible recombination}), \\ dn_e/dt &= (n_o - n_e) \sigma_e N - n_e \sigma_i^* N - n_e/\tau = a - bn_e, \end{aligned}$$

with

$$a = n_o \sigma_e N, \quad b = (1/\tau + \sigma_e N + \sigma_i^* N).$$

Integration gives

$$\begin{aligned} n_e(t) &= \frac{a}{b} (1 - e^{-bt}) \quad \text{and} \\ n_i(T) &= \sigma_i^* N \frac{a}{b} (T + \frac{1}{b} (e^{-bT} - 1)) \end{aligned} \quad (1)$$

assuming $N = \text{const. during the pulse}$.

$$\rightarrow n_i(T) = n_o \sigma_e \sigma_i^* N^2 \frac{T}{b} [1 - \frac{1}{bT} (1 - e^{-bT})] \quad (2)$$

with $b = 1/\tau + (\sigma_e + \sigma_i^*)N$ and $T = \text{width of laser pulse}$.

We solve (2) for two limiting cases:

- $bT \ll 1$, i.e. low photon flux and long τ ($\gg T \sim 10^{-9} \text{ s}$):

$$n_i(T) = n_o \sigma_e \sigma_i^* N^2 T^2 / 2 \quad (3)$$

The ionisation density is proportional to $(NT)^2$, the square of the number of photons/cm² of the laser pulse. This is to be expected for a 2-photon process, when the lifetime of the intermediate state is longer than the laser pulse.

On the other hand, if

- $bT \gg 1$, i.e. $\tau \ll T$ and/or high flux N :

$$n_i(T) = n_o \sigma_e \sigma_i^* N^2 T / b. \quad (4)$$

We find a linear dependence on T . An experiment keeping $N = \text{const.}$ but varying T (e.g. with an optical shutter), could decide which condition is approached. This has unfortunately not been tried yet.

If $1/\tau \gg (\sigma_e + \sigma_i^*)N$, we find

$$n_i(T) = n_o \sigma_e \sigma_i^* N^2 T \tau, \quad (4a)$$

an ionisation cross section $(\tau/2T)$ times smaller than (3).

If the flux is high and $(\sigma_e + \sigma_i^*)N > 1/\tau$, a special situation arises. The excited state is saturated rapidly ($n_e \rightarrow a/b = n_o \sigma_e / (\sigma_e + \sigma_i^*)$) and

$$n_i(T) = n_o \sigma_e \sigma_i^* NT / (\sigma_e + \sigma_i^*) \quad (4b)$$

i.e. the ionisation becomes a linear function of $N \cdot T$! The transition from quadratic to linear dependence is defined by

$$(\sigma_e + \sigma_i^*)NT > 1. \quad (5)$$

For $(\sigma_e + \sigma_i^*) \sim 10^{-16} \text{ cm}^2$, $NTh\nu \geq 100 \text{ } \mu\text{J}/\text{mm}^2$ is required. This is close to thresholds observed in undoped chambers at 337 nm and 351 nm [6,14].

In our estimates, we have assumed $N = \text{const.}$ In practice, the laser beam may fluctuate substantially in time and over its cross section and correction terms have to be added.

To limit variations of the ionisation along the track, we may require a photon absorption length $l_{\text{abs}} > 10 \text{ m.}$ With $l_{\text{abs}} = (\sigma_e n_o)^{-1} > 10^3 \text{ cm}$ and $\sigma_e \sim 10^{-16} \text{ cm}^2$, the concentration of the additive has to be limited to $n_o < 10^{13} \text{ molecules/cm}^3$, i.e. to a partial pressure $< 0.3 \times 10^{-3} \text{ Torr.}$

It is interesting to look into the possibility of single photon ionisation. Assuming $\sigma_i \sim 10^{-16} \text{ cm}^2$, $n_o < 10^{13} \text{ molecules/cm}^3$ is required again. On the other hand, with a pulse of $1 \text{ } \mu\text{J}$ in a cross section $S = 1 \text{ mm}^2$ ($\text{NTS} \sim 10^{12}$), we find

$$n \text{ (ion pairs/cm track length)} = n_o \sigma_i \text{NTS} = n_o \times 10^{-4}.$$

If we require more than 100 ion pairs/cm, we need an additive with a concentration of $> 10^6 \text{ molecules/cm}^3$ only, i.e. $> 10^{-10} \text{ Torr}$! Alkaline additives, Cs (with I.P. = 3.89 eV and vapour pressure p_v of 10^{-6} Torr at 20°C) and K (with I.P. = 4.3 eV and $p_v = 10^{-8} \text{ Torr}$) could therefore in principle be used in connection with a Nd laser ($E_Y = 4.68 \text{ eV}$). These substances are however delicate as they are chemically very aggressive. Another combination would be an ArF laser (6.53 eV) ionizing TMAE (5.36 eV, $p_v = 0.27 \text{ Torr}$ at 20°C). None of these combinations has been tried so far.

2.2 Laser characteristics

A summary of lasers used so far in detector calibration is presented in table 1. The nitrogen laser is the most commonly used one, followed by the Nd-Yag. The nitrogen laser has many advantages: it is simple, compact and relatively cheap; the construction can easily be adapted to special requirements. The pulsewidth at 1 atm. can be as short as about 0.5 ns, the beam divergence close to diffraction limit: $0.2 \times 0.1 \text{ mrad}$ half-angle, for a cross section of $1 \times 2 \text{ mm}^2$. But the choice of additives which can be ionised at $\lambda = 337 \text{ nm}$ in a 2-photon process is much more restricted than that for a Nd laser with 2 frequency doublers ($\lambda = 266 \text{ nm}$). The excimers have a very short life span, unless cleaning is carried out continuously and carefully. On the other hand, a XeCl excimer coupled to dyes and a frequency doubler has provided very interesting information on the wavelength dependence of the ionisation [17-19, see below].

Laser beams may present a very irregular intensity profile in space, unless they are adjusted to a pure TME_{00} mode (Gaussian profile) or a clean multimode operation with an approximately flat profile. The time spectrum, too, may show strong fluctuations and may vary from pulse to pulse: fig. 3 shows an example for a particular N_2 laser. Also shown is the wavelength spectrum, containing several distinct lines within a band of about 0.1 nm.

2.3 Results on ionisation

2.3.1 Doped chambers

Many additives have been investigated. A summary of the most successful tests is presented in table 2. About 100 compounds were found with ionisation potential < 7.36 eV [29]. Most, however, are radicals, instable, rare compounds or contain halogens with strong electron attachment. Significant ionisation was also detected with a N_2 laser in 2, 2, 6, 6-dimethyl-piperidon-4-N-oxyl (7.36 eV), n, n'-tetramethyl-diaminodiphenyl (7.25 eV), Diphenylamine (7.14 eV), β -naphtylamine (7.25 eV; strongly cancerogene) and tritretbutyl-ferrocene (7.36) [22].

Ionisation densities are given as number of electrons produced in a track segment 1 cm long and 1 mm^2 in cross section. The values are often only extrapolated from measured numbers and have to be used with precaution; in particular when different groups are compared, as normalization may differ (e.g. measurement of intensity, variation of laser pulse in time and space).

More than 10^3 electrons/cm are produced with only $1\text{ }\mu\text{J}/\text{mm}^2$ and 10^{-3} Torr partial pressure in many substances at 266 nm and in TMPD and α -naphtylamine at 337 nm. Except for TMPD, the highest 2-photon ionisation cross sections at 337 nm are shown by substances with low vapour pressure: diacetyl-ferrocene, diethyl-diphenyl-molybdenium and α -naphtylamine. Some of the other substances listed, however, can provide adequate ionisation at higher partial pressure. The 2-photon ionisation cross sections estimated from these measurements at 337 nm range from 2×10^{-44} (TMPD) to $< 3 \times 10^{-50}\text{ cm}^4\text{ s}$ (ferrocene) [22].

The proportionality of the ionisation with $(\text{flux})^n \times \text{partial pressure}$ with $n \sim 2$, has been verified for most additives with typical values of flux $\leq 10\text{ }\mu\text{J}/\text{mm}^2$ and partial pressure ≤ 5 ppm (figs 4, 5 and ref. [2,14,16,21,22]). The coefficient n can deviate from 2 because of errors in the measurement of the average flux but also because of irregular space and time structure of the laser pulse.

A frequency scan obtained with a combination (XeCl-laser + dyes + frequency doubler) is presented in fig. 6 for several substances. The ionisation cross sections vary quite smoothly with frequency but rise by several orders of magnitude between 337 and 265 nm. Only weak resonance structures are visible [17,18].

Because of their high cross sections for ionisation by UV-lasers, some of the additives studied are clearly very attractive. Before actually applying them to large chambers, one has, however, to check various possible side effects. Most of the substances are toxic and chemically active (e.g. DEA attacks PVC; DPA is better [45]) and safety precautions have to be applied. Some tend to stick strongly to surfaces (e.g. phenol, DEA) and may be difficult to remove if so desired; in this respect, toluene, butylbenzene and xylene and mesitylene should be advantageous. TMAE attacks rubber and even some metals and forms conductive layers, once exposed to oxygen [46]. This can be disastrous for chambers with field cages. Little is known also on the influence of additives on chamber ageing under radiation. Work is just starting: in one test chamber, lifetime increased by a factor of 3 after addition of 100 ppm TMA and decreased by a factor of 9 after addition of 500 ppm naphthalene [47].

Large systems tend to introduce recirculating gas circuits to reduce costs and increase stability. The degree of absorption of the additives by the filters has to be measured. Only few results exist so far [20,24,26,36]. Molecular sieves (3 Å) absorb toluene chemically, but even when saturated with toluene, still absorb water [48].

For these reasons, work on undoped chambers is continuing, although higher laser powers are required.

2.3.2 Undoped chambers

Significant ionisation (> 100 electrons/cm for $\sim 100 \mu\text{J}/\text{mm}^2$ at 337 nm and $\sim 10 \mu\text{J}/\text{mm}^2$ at 266 nm) has also been observed in many chambers with various gas fillings without the use of additives. Again, ionisation was roughly proportional to $(\text{flux})^2$ and $(\text{flux})^1$ at low and high flux, respectively [e.g. 2, 3, 4 and many later reports]. The transition from $n \sim 2$ to $n \sim 1$ is shown in fig. 4(c) [6]. As an example of a long ionisation track, a photo from a streamer chamber is shown in fig. 9. A nitrogen laser with $20 \mu\text{J}/\text{mm}^2$ was used [5].

These results can be explained by assuming ionizable impurities contained in the gas and/or outgassing from the chamber system and being practically omnipresent. This assumption has been solidified by many observations. Outgassing of chamber walls was demonstrated by heating the chamber: ionisation increased strongly [24,26]. Changing the pressure of the carrier gas hardly changed ionisation density in the laser beam, indicating again that the impurities were due to outgassing [23]. Gas

cleaning can reduce the ionisation drastically [24]: In a rather clean chamber (only consisting of stainless steel, glass-ceramics and viton sealing), filtering with a titanium getter pump (300°C) and activated carbon (-65°C) in a closed cycle reduces the ionisation by an order of magnitude in a few minutes. Even permanent cleaning of the system could be achieved: the ionisation signal dropped below the detection limit after 30 h of gas filtering limit when applying $10 \mu\text{J}/\text{mm}^2$ at 337 nm and after 100 h with $9 \mu\text{J}/\text{mm}^2$ at 266 nm.

An analysis of the gas which had circulated through the large OPAL prototype, showed small concentrations of benzene, toluene, xylene, naphtalene and some 25 more impurities [30]. In another set-up, ionisation was measured as function of wavelength in an undoped chamber and this spectrum compared to single photon absorption spectra of phenol and toluene [18,19]. Close correspondence was detected; these two substances could thus be identified as impurities in the undoped chamber (fig. 7). By comparison with well-controlled seedings, the background concentration of toluene was established to be at the few ppb level [19].

The close similarity between single photon and 2-photon ionisation spectra indicates that the second step ionisation cross section is rather independent of wavelength. A look at other single photon absorption spectra - easily obtainable with standard equipment - can, therefore, be useful to guess the magnitude of 2-photon ionisation cross sections. Some low resolution spectra are shown in fig. 8 [27].

2.4 Other results

Many groups (including all LEP collaborations) are using UV lasers to calibrate their drift chambers and measure particular gas parameters like diffusion coefficients. Only a few examples can be mentioned.

Tracking and alignment: Ionization tracks straight to $\sigma = 10 \mu\text{m}$ have been measured with a N_2 laser over 10 m length [13(b)]. The OPAL central detector group has transported the laser beam over 15 m and then measured $\sigma \sim 35 \mu\text{m}$ over 1.6 m in the prototype [31]. The ALEPH Collaboration has also tested a high precision long distance beam transport and splitting system [32]. For such long-distance measurements it is essential to operate the laser in the fundamental TME_{00} mode (Appendix).

The strong photo-electric effect on metallic surfaces can be used e.g. by hitting thin wires in a chamber with the laser beam, thus providing convenient reference points.

Diffusion measurements: High precision measurements on longitudinal and transverse diffusion of single electrons have been carried out with a N_2 laser by production of well localised single electrons in a metallic pin-hole [33].

Point scans: Focussed laser beams permit high precision point scans in chambers, e.g. near edges [34] or near sense and field wires [8,35].

2-track studies: A laser is an unique tool to study effects of a second near-by track and was used by several groups for this purpose [e.g. 3,10,36].

Transmission via mono-mode fibres: With fibres, light is easily transmitted to very inaccessible places. For good beam quality, only mono-mode transmission is acceptable. Unfortunately, transmission in the UV of significant power is not straightforward [37]. Most fibre research goes into I.R. transmissions which requires different material properties. UV mono-mode fibres would, therefore, require special developments. In addition, there are fundamental limitations: the diameter for a mono-mode fibre is proportional to the wavelength, the core diameter has to be of the order of $1\text{ }\mu\text{m}$ for UV. This causes input coupling losses, which is particularly important as the power density limitation for quartz fibres lies around 5 GW/cm^2 .

Nevertheless, with the use of efficient additives it might be possible to transmit the required intensity of $\sim 0.1\text{ }\mu\text{J/mm}^2$. This would certainly find interesting applications.

3. LIQUID AND SOLID TRACK DETECTORS

We shall only briefly summarize work related to these detectors.

3.1 Bubble chambers

Shortly after the first ionisation tracks in gases had been observed in a beam of a N_2 laser, the same laser was applied to various bubble chambers: with good success. Excellent tracks were observed first in the large BEBC filled with H_2 (3 m visible length with ≤ 25 bubbles/cm for $< 100\text{ }\mu\text{J}$, fig. 10) and then in small test chambers with argon and nitrogen fillings (< 8 and 20 bubbles/cm, respectively, with $\sim 10\text{ }\mu\text{J}$) [38]. To form bubbles, enough heat has to be deposited very fast to allow bubble growth to a critical radius in $\leq 1\text{ ns}$. Then, growth can continue due to the pressure difference between the vapour in the bubble and the surrounding liquid, as

the compressive force due to surface tension diminishes. The original heating is produced by δ -rays in the case of bubble formation by ionizing particles. In the case of bubble formation by laser beams, it is likely that heating of miniature dirt particles is responsible.

In bubble chambers, laser beams could be very useful, e.g. for the measurement of errors due to optical distortions and liquid movements, for the study of holographic recording techniques or of the unique properties of an Ar bubble chamber. However, the technique became available too late, at a time when bubble chambers were slowly being closed down.

3.2 Si-detectors

With the indication that a space resolution of $\sigma \geq 3 \mu\text{m}$ can be achieved, the interest in Si-detectors is rapidly growing. Clearly, such devices require powerful alignment and calibration tools. Again, the laser may help. This work is, however, still in its infant stage.

3.2.1 Alignment

Si-detectors are sensitive to light from 200–1100 nm. Thus, various lasers can be used, either to align the Si-strip detectors themselves, or taking the Si detectors as reference marks for the positioning of other detectors. Such applications are already quite common. Shorter wavelength can bring somewhat better precision. Positioning to $\sim 1 \mu\text{m}$ is possible.

3.2.2 Calibration

The applications mentioned in sect. 3.2.1 use ionisation in the surface layer. This process has also been applied to calibration of charge division techniques [39]: A junction was covered with strips spaced $20 \mu\text{m}$. Each fourth strip was connected to an amplifier A_i . A focussed light source ($\phi \sim 3 \mu\text{m}$) hit the surface between the strips A_i and A_{i+1} . The difference signal ($A_{i+1} - A_i$) was measured as function of the position of the light spot: an error of $\sim 3 \mu\text{m}$ was achieved. For such investigations, laser pulses are advantageous as adequate power is required in short pulses.

Another application has exciting prospects, making use of ionisation in depth, in a manner similar to gas ionisation by UV lasers [40]. In fact, Si becomes transparent when the photon energy approaches the energy of the gap between the conduction and the valence bands, i.e. $1.12 \text{ eV} \approx 1107 \text{ nm}$ (fig. 11).

For a wavelength around 1100 nm, the ionisation signal is still strong enough (>> noise) but the absorption is < 10% over a typical detector thickness of 230 μm . The ionisation density is thus almost constant over the full depth. An IR laser operating at this wavelength would, therefore, provide a powerful tool for 3-dimensional scanning of Si detectors, to measure precisely parameters as: homogeneity of the material, response as function of track position and angle, cross-talk and pulse smearing in a magnetic field.

4. OUTLOOK

In the past six years, lasers have become a powerful diagnostic tool for various tracking detectors, in particular for large drift chambers. Much knowledge has been gained on the ionisation process, efficiency of additives, construction of special lasers, etc. Nevertheless, there is ample space for further developments. We shall just mention a few:

- Development of smaller and cheaper lasers, in particular with $\lambda < 300 \text{ nm}$.
- More precise measurements and control of the time and space structure of the laser pulse.
- Further studies of the impurities: handling dangers; effects on chamber materials (chemical activity; stickiness, formation of conductive layers, etc.); influence on ageing of chambers under radiation; details of the ionisation process (e.g. studies with variable pulse width); identification of more of the impurities responsible for the ionization, in particular those active already at 337 nm.
- UV transmission via mono-mode fibres.

There is thus no risk for this field of research to loose its impetus over the coming years.

The author wishes to thank the experts of the work presented for the many interesting discussions and the rapid transmission of recent results. He appreciated in particular the efforts of the organizers of the workshops on calibration by UV lasers in Glasgow (1984) and in Dortmund (1985). Unfortunately, only a restricted sample of the large volume of interesting results could be mentioned in this short review.

APPENDIX

LASER BEAM OPTICS

For best performance as calibration tool, the laser should be adjusted to operate in the lowest order mode, in the TEM₀₀ [41]. The intensity profile then is and stays Gaussian as function of radius: $I = I_0 \exp(-2r^2/w^2)$, with 86.5% of I in $2r = 2w$, and the beam shows the lowest divergence. We consider only intensity profiles. The ionisation profile for a quadratic relationship will show larger amplitude variations but smaller beam diameter and divergence.

Starting with a waist, or a place where the wave front is flat (inside the laser or in a focal plane), of radius w_0 , the radius increases with distance z as:

$$w(z) = w_0[1 + (\Theta \cdot z/w_0)^2]^{1/2} \quad (A1)$$

where the divergence Θ (half-angle) for large z is given by

$$\Theta = \lambda/(\pi \cdot w_0), \quad (A2)$$

i.e. $\Theta \sim 0.1$ mrad for $\lambda = 337$ nm and $w_0 = 1$ mm.

Optical elements in the beam line should always have a radius $> 1.5 w$.

The divergence can be reduced by a beam expander, which increases the beam radius: $\Theta_2/\Theta_1 = w_1/w_2$.

Minimum variation of $w(z)$ over distance $2z_R$ can be achieved by focussing at $z = z_R = w_0/\Theta$. One obtains:

$$w(0) = w(2z_R) = w(z_R) \sqrt{2} \quad (A3)$$

A radius variation from 1 to 1.4 mm is thus possible over $2z_R = 20$ m!

The focal spot radius of a perfectly collimated laser beam ($r = w$) focussed by an aberration free lens with focal length f is:

$$s = \lambda \cdot f/(\pi \cdot w),$$

i.e. $s = 2 \mu\text{m}$ for $f = 10$ mm, $\lambda = 337$ nm and $w = 1$ mm.

For a special focussing singlet lens, s has to be multiplied by approximately 4/3 [42].

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TABLE I Laser characteristics. Typical performance of devices used in this work.

Laser	λ (nm)	h (eV)	Δt (ns)	Power (mJ)	Comments
N_2	337	3.68	0.5	0.250	Simple, compact, cheap; custom design Short Δt !, $\Delta PH \sim 10\%$
Nd-Yag	1060 530 354 266	1.17 2.34 3.51 4.68	4.5 [26,27] 6 [24] 25 [14]	40 20 4	Mostly clumpy, expensive, $\Delta PH \geq 30\%$, but high power and short λ ! Frequency quadrupling e.g. with DCDA + KDP
XeF [7,12] KrF [7] ArF XeCl + dyes + f doubling	351 248 193 (308) 262- 330	3.53 5.0 6.53 4.73- 3.76	15 13 11 6	~ 100 [14] $\gg 5$ [14] $\gg 2$ [14] > 0.01 [17-19]	"Excimers": short lifetime, Continuous cleaning necessary Spectroscopy carried out with $\Delta\lambda \sim 0.01$ nm! Dyes exist with emission from 420-780 nm; e.g. DCM, Rhodamin 6G, Coumarin 153 Frequency doubling e.g. with KDPB + R6G [17]

TABLE 2 Properties of additives with low ionization potential. Values for ionization density I.D. extrapolated assuming $I.D. \propto I^2 \cdot p$, a laser pulse of $1 \mu J$ and 1 mm^2 cross section and a partial pressure of additive $p = 10^{-3}$ Torr unless otherwise stated.

Additive	Ionis. pot. (eV)	Vapour pressure (Torr) 20°C (0°C)	Melting/boiling point (°C)	Ionization (e^-/cm) for $1 \mu J$ in 1 mm^2 and 10^{-3} Torr additive
Benzene C_6H_6	9.24	77	55/80	266: 5×10^3 [26,27]
Toluene C_7H_8	8.82	22	-95/110	266: 5×10^4 [26,27] 0.5×10^4 [19]
Cumene C_9H_{12}	8.69	3.8	-96/152	266: 11×10^4 [26,27]
n-Butylbenzene $C_{10}H_{14}$	8.69	1	-88/183	266: 5×10^4 [26,27]
m-Xylene C_8H_{10}	8.56	6	-48/139	266: $\leq 4 \times 10^4$ [26,27]
TMA (CH_3) ₃ N	8.5 [21] 7.8 [26]		-117/2.9	266: 60-130 [15,17] 80 [24]
1, 3, 5 Mesithylene C_9H_{12}	8.40	1.5	-45/165	266: 1.8×10^4 [26,27]
Phenole C_6H_5OH	8.3 8.51 [17]		41/	266: > 250 [25]
Naphtalene $C_{10}H_8$	8.12 [17]		81/	266: 560 [25]
TEA (CH_3CH_2) ₃ N	7.5 [26]	50 (12)	-114.7/893	266: 200 [17]
N_2 laser α -Naphtylamine ↓	7.3	2.7×10^{-4}	50/301	337: 800 at 2.7×10^{-4} Torr [21,22]
DMA $C_6H_5N(CH_3)_2$	7.14 [26]	0.25 (0.065)	2.5/194	266: 10^4 [17] 337: 2000 (40 x bgr) at 0.25 Torr with $14 \mu J/\text{mm}^2$ [16]
DPA $C_6H_5N(C_3H_7)_2$	7.1	1.2×10^{-2}	/245	337: 90 [21,22]
Diacetyl-ferrocene	7.08	1.2×10^{-6}	127/	337: 10 at 1.2×10^{-6} Torr [21,22]
DEA $C_6H_5N(C_2H_5)_2$	6.99 6.76 [11]	0.14 (0.021)	-39/216	337: ~ 10 x bgr. at ~ 0.07 Torr [2] ~ 0.03 Torr [8] ~ 0.045 Torr [13]
Diethyl-ferrocene	6.6	5.4×10^{-2}	/130 (5 Torr)	337: 7 [14,15]
Nickelocene $Ni(C_5H_5)_2$	6.5	1.1×10^{-3}		337: >> bgr. [1,7]
TMPD	6.18 [22]	2.3×10^{-3}	51/260	266: 6×10^4 [24] 337: 7×10^4 [21,22]
TMAE $C_{10}H_{24}N_4$	5.36	0.27 (0.06)	-10/59	337: > 300 for 10^{-3} Torr [16]
Diethyl-diphenyl-molybdenum	5.5	5.4×10^{-6}	/150 (1 Torr)	337: 120 at 5.4×10^{-6} Torr [21,22]

DEA: N,N-Diethylaniline; DMA: N,N-Dimethylaniline; DPA: N,N'-dipropylaniline; TMA: Trimethylamine; TFA: Triethylamine; TMAF: Tetrakis (Dimethylamine) Ethylene; TMPD: N,N,N',N' tetramethylphenyldiamine

Fig. 1 2600 Å system in benzene. (a) Low resolution single photon absorption spectrum, (b) the B_0^0 (6_1^0) band under high resolution (scale $\times 200$) [28(b)].

Fig. 2 Schematic energy level diagram of complex molecule, indicating possible channels for 2-photon ionisation (detailed discussion in [22]).

Fig. 3 N_2 -laser at 1 atm. (a) Intensity I as function of wavelength [43]. (b) Time structure of pulse [44].

Fig. 4 Dependence of ionisation density (electrons/cm) on energy density of laser pulse in doped and undoped chambers [(a) 26, (b) 24]. The approximately quadratic dependence at low flux becomes linear at high energy density (c) [6].

Fig. 5 Linear dependence of ionisation density on concentration of additive [(a) 26, (b) 24(a)].

Fig. 6 Variation of ionisation density I.D. (electrons/cm) with wavelength for some additives (upper lines). The background response of the counters is indicated by the lower points [17,18].

UV-ABSORPTION

Fig. 8 Single photon absorption spectra for some additives [27]: (a) m-xylene, (b) 1,3,5-mesitylene, (c) toluene, (d) i-propylbenzene, (e) N-butylbenzene, (f) benzene.

Fig. 7 Identification of toluene as trace impurity. (a) Two-photon ionisation spectrum in undoped chamber ("background"). (b) Single photon absorption of toluene. (c) Two-photon ionisation spectrum of chamber seeded with toluene [19].

Fig. 9 Ionisation track (2 m long) in EMC streamer chamber produced by a N_2 -laser beam ($20 \mu J/mm^2$). No additive used [5].

Fig. 10 Laser induced bubble track in the Big European Bubble chamber. N_2 -laser beam enters from the top (visible length = 3.5 m). Also visible are muon tracks with smaller bubble density and calibration wires [38(a)].

Fig. 11 Light absorption in Si. (a) Absorption coefficient as function of photon energy [39]. (b) Absorption in a $230 \mu\text{m}$ thick layer as function of wavelength [40].