IONIZATION OF MULTIWIRE PROPORTIONAL CHAMBER GAS
BY DOUBLE PHOTON ABSORPTION

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ABSTRACT

We have been able to produce substantial ionization in the gas of multiwire proportional chambers by means of pulsed lasers in the near ultraviolet (337 nm and 265 nm). The energy was modest (≤ 100 µJ/mm²). The mechanism involved appears to be double photon absorption by low ionization potential impurities in the gas. Applications, in particular to the calibration of large drift-chamber systems, are discussed.

(Submitted to Nuclear Instruments and Methods)
1. **INTRODUCTION**

The track reconstruction accuracy of large drift-chamber systems is usually limited by systematic effects such as the relative position of the sense wires, the knowledge of drift velocity and time origin, and the control of distortions due for instance to space-charge effects.

It is customary to evaluate these effects by fitting tracks which happen to go through the detector. However, the interpretation of the residue distributions is often difficult and is complicated by the fact that the track position and momentum (when working in a magnetic field) are not known independently.

It has been suggested\(^1\) to use laser beams or pulsed X-rays in order to ionize the chamber gas directly on a straight line. This could provide a set of fixed "fiducial marks", allowing easier calibration of large systems and potentially substantial improvement of accuracy.

This report deals with ionization by a laser beam. To be specific, let us consider a gas mixture of argon and ethane. The ionization potential (IP) of the components are 15.7 and 11.65 eV, respectively.

Direct photoionization can be attempted. However, it cannot be on the primary constituents of the gas. If the photoionization cross-section is \(\sigma_i\), the absorption length of the beam is

\[
\ell_{\text{abs}} = \frac{1}{\sigma_i \times N \times p}
\]

where \(N\) is the number of molecules per cm\(^3\) (\(\sim 2.7 \times 10^{19}\)) and \(p\) is the partial vapour pressure. For a typical cross-section of \(5 \times 10^{-17}\) cm\(^2\) and a required absorption length of 10 m, the vapour pressure should be smaller than \(7 \times 10^{-8}\) atm. It is unlikely that alkaline impurities can be used (Cs:IP = 3.89 eV; K:IP = 4.3 GeV) since they are chemically very active. Therefore one is limited to complex organic molecules such as diethylaniline (IP = 6.99 eV) or methylaniline (IP = 7.12 eV)\(^2\). The corresponding wavelength is 175 nm, which is unfortunately in the vacuum ultraviolet region where handling of the beam is difficult.
For this reason we were more interested in double photon absorption, which would allow us to work at a wavelength twice as large, i.e. in the close UV region (≈ 350 nm). Pulsed lasers in this region are commercially available (e.g. the N₂ laser at 337 nm, the Neodymium Yag laser tripled or quadrupled at 350 or 265 nm). The basic process is shown schematically in fig. 1a. A first photon excites the molecule to a usually virtual state with a cross-section \( \sigma_1 \) of the order of \( 10^{-16} \text{ - } 10^{-17} \text{ cm}^2 \). This virtual state lives for a time \( \tau \sim 10^{-16} \text{ s} \). If a second photon arrives during that time, it can ionize the virtual state with a cross-section \( \sigma_2 \), also between \( 10^{-17} \) and \( 10^{-16} \text{ cm}^2 \). The equivalent cross-section is then
\[
\sigma = \sigma_1 \sigma_2 \tau F = \sigma^{(2)} F,
\]
where \( F \) is the flux of photons. Indeed, typical electronic transition cross-sections \( \sigma^{(2)} \) are experimentally \(^1\) of the order of \( 10^{-46} \text{ - } 10^{-50} \text{ cm}^4 \text{ s}^{-1} \) as given by the foregoing naive computation. The number of electrons created per unit length is
\[
n_e = \frac{\sigma^{(2)} N P \eta^2}{T \eta} = \sigma^{(2)} N P F^2 T s,
\]
where \( n_e \) is the total number of photons, \( T \) the pulse width, and \( \eta \) the area. The ionization rate is, of course, quadratic with respect to the flux.

When the intermediate state exists (fig. 1b), cross-sections are much larger and one speaks of double-step processes. If the first transition is saturated, the ionization rate is expected to be linear with respect to the flux.

2. **EXPERIMENTAL SET-UPS**

We report results obtained in two different chambers exposed to two different lasers.

2.1 The Ecole polytechnique set-up

A small chamber has been built specially for testing gas ionization by UV light (fig. 2). Special care has been taken to prevent electrons extracted by the UV beam from windows and electrodes from reaching the sense wires. The pulse from a single wire was observed through a low-noise current amplifier. The chamber was
evacuated and filled with either of two gas mixtures: "magic gas" (argon 71.5% +
+ isobutane 24% + freon, CF₂Br 0.5% + methylal 4%) or argon + propane (50%, 50%).
Preliminary results have been reported elsewhere.

The chamber was exposed to quadrupled Neodymium Yag lasers (λ = 265 nm).
The diameter of the beam was 0.36 mm and, depending on the laser used, the pulse
width was either 50 ps or 15 ns (FWHM).

2.2 The CERN set-up

Figure 3 gives a sketch of the chamber used at CERN. It is a projection
chamber built by P. Queru and collaborators. It has a drift space of 18 cm and a
wire length of 2.5 m. For this particular test, seven wires at the centre were
equipped with a wave-shape recorder sampling each pulse every 30 ns. The gas
used was argon + ethane (50%, 50%).

The UV beam was produced by a small N₂ laser, and was focused with a
ρ = 30 cm quartz lens to an area of about 1 mm² in the region of interest. The
energy was fixed and equal, within a factor of 2, to 0.1 mJ.

3. EVIDENCE FOR IONIZATION IN GAS

In both set-ups we have observed ionization in the gas without specifically
adding any impurities of low ionization potential.

In the case of the small chamber exposed to a 265 nm beam, more than 400 elec-
trons/cm were produced in magic gas with an energy per pulse of roughly 1 μJ. The
proof that the electrons are produced in the gas and not on the electrodes parallel
to the beam is given in fig. 4. The chamber was moved with respect to the beam,
and the variation of the drift time of the electrons was measured. A linear
dependence on the position is observed and the drift velocity inferred is realistic.

In the CERN chamber the electrons are clearly produced along the light-beam
in the gas, as can be seen in fig. 5a; this shows the pulses observed on seven
consecutive wires when the light-beam is parallel to the wire plane. The bin

*) From Quantel, 17 avenue de l'Atlantique, Z.I. 91400 Orsay (France).
**) Designed by H. Verweij and B. Walgren at CERN.
***) Produced by Sopra, 68 rue P. Joignieux, 92270 Bois-Colombes (France).
width is 30 ns. The particular case shown corresponds to a drift length of 10 cm, and the measured drift time is in excellent agreement with the expected value. Comparison with a $^{55}$Fe source shows that 50 electrons/cm were produced. If the light is shot perpendicularly to the wire plane, on a single wire, the typical pulse height shown in fig. 5b can be observed. The peak at a small time is a Jacobian peak due to the electrons produced at the shortest distance from the wire. The pulse then extends, with small fluctuations, to 4 µs, which corresponds to the full drift length (the fluctuations at 1 µs are due to laser noise). When the light hits an electrode which is made of Cu-Be wire, an enormous pulse is obtained, without apparent damage to the chamber.

There is therefore no doubt that the gas itself is ionized. The interpretation of this phenomenon, which occurs without specifically added impurities, is less clear. It is unlikely to be due to alkaline impurities (which with the N$_2$ laser can only be Cs). The absence of fluctuations in the pulse (fig. 5b) seems to rule out the explanation that light ionizes minute pieces of dust. Such a phenomenon is known to cause problems in the study of multiphoton-induced electric breakdown$^5$). The most likely explanation is the presence in the gas of heavy hydrocarbon impurities, such as pump oil, which have an ionization potential$^6$ as low as 7 eV. They are known to be a disturbing background for multiphoton ionization studies$^7$). They can be ionized by multiphoton or multistep processes, as discussed below.

4. EVIDENCE FOR DOUBLE PHOTON ABSORPTION AT 337 nm

After observing the above phenomenon in the control experiment, we proceeded in the CERN set-up to the planned experiment of adding diethylamine to the gas. This substance has a low ionization potential of 7 eV and is liquid at 20° with a vapour pressure of 0.14 Torr$^2$). By bubbling argon (which represents half of the gas) through a few millimetres of the liquid, the proportion of diethylamine has to be smaller than or equal to 0.07 Torr. This was done in the CERN set-up.

The pulse height generated by the laser beam was multiplied by approximately 10. We studied the flux dependence by interposing small sheets of mylar which
were previously calibrated with the same N₂ laser and a phototube. The observed dependence (fig. 6) is compatible with a quadratic behaviour: the fitted power is

\[ h = (1)^{1.7±0.2}. \]

This is direct evidence for double photon absorption. Moreover, we can compute an order of magnitude of the double photon cross-section \( \sigma^{(2)} \). Let us take in formula (1),

\[ \begin{align*}
  n_e &= 500 \text{ e/cm} \\
  p &= 0.01 \text{ Torr} = 10^{-5} \text{ atm (because of the inefficiency of our bubbler)} \\
  n_\gamma &= \frac{10^{-5}}{3.67 \times 1.6 \times 10^{-15}} = 1.7 \times 10^{-12} \text{ (since the photon energy is 3.67 eV)} \\
  T &= 3 \times 10^{-9} \text{ s} \\
  s &= 10^{-2} \text{ cm}^2.
\end{align*} \]

We then obtain

\[ \sigma^{(2)} = 2 \times 10^{-51} \text{ cm}^4 \text{ s}^{-1}, \]

which is of the correct order of magnitude and supports our claim to have observed a double photon process.

5. POSSIBILITY OF SATURATION OF THE FIRST TRANSITION

5.1 Observation at 265 nm

This double photon behaviour is at variance with the dependence observed at 265 nm, without adding any impurity. Figure 7a shows an example of the observed dependence in the Ecole polytechnique set-up, using commercially available calibrated neutral filters. By varying the laser intensity in order to obtain the same pulse height with various filters, we checked their relative calibration and found that the highest attenuation filter was wrongly calibrated in our domain of wavelength and energy. The cross in fig. 7a refers to the intensity determined by our cross-calibration. Figure 7b combines the measurements done at various laser intensities and chamber amplifications. They have been rescaled according to known chamber amplification. The relative laser intensities were normalized.
by the requirement that, for a given number of collected electrons, light intensities should be the same. Normalization points are not included and the crosses refer to points dependent on our filter cross-calibration. Within our experimental errors, the number of created electrons is approximately linear over nearly two orders of magnitude.

With our available equipment, it is difficult to normalize the absolute flux given by the laser. A rough estimate, valid within a factor of 10, is that 1 μJ was producing 200 electrons per cm.

5.2 Interpretation

The observed linearity is at variance with a simple double photon absorption. It is extremely unlikely that molecules with ionization potential as low as 4.6 eV can survive in our gas. The most likely phenomenon is that the process at work is indeed double photon absorption but with a pumping mechanism that populates an intermediate level of relatively long lifetime. We show in the Appendix that our observation can be understood, in particular, in terms of double-step photon absorption, where the laser beam saturates a first transition to a normal excitation level which subsequent photons can ionize. Other pumping mechanisms can be imagined, such as creation of quasi-stable excited states in the avalanches, collision processes, etc.

6. CONCLUSIONS

We have observed direct ionization of the gas by UV light with two different chambers and three different gases. Only modest laser intensities are necessary. The data are compatible with an interpretation in terms of a double photon or double-step process. More detailed work with equipment more adequate than the one available to us is necessary in order to understand the linear dependence on the flux observed at 265 nm. However, the observed effects have interesting practical applications.

First, this phenomenon may provide a convenient way of calibrating large systems: straight tracks of known position can be drawn in the detector even in
the presence of a magnetic field. This task can also be performed by pulsed X-rays producing electrons by Compton effect\(^7\). However, the latter method produces isolated dots on a straight line and not a continuous track, and the chamber behaviour is different in the two cases. In particular, because of the intrinsic dispersion of electron arrival times due to electric field geometry, it is necessary to average over a large number of pulses in order to reach good accuracy. A laser beam can provide a better simulation of a straight track where the drift time is given by the arrival time of the first electron.

Obvious limitations of the laser beam are the need for transparent windows (quartz or some plastic material) and diffraction. Figures 8a and 8b give an evaluation of this effect for a 265 mm light-pulse going through a slit, 0.5 mm wide, for the two cases of linear and quadratic dependence of the pulse height on laser intensity. A typical 3 ns FWHM time spread has been added which increases the apparent dimension of the beam (the drift velocity taken was 1 mm/20 ns). A beam size of 0.5 mm FWHM, similar to the actual "width" that a track has because of diffusion and shape of electric field lines, is maintained over 1 m from the diaphragm. That is, by focusing in the centre of the system to be calibrated, about 2 m of "track" length can be used. A longer length needs intermediate re-focusing. It should be noted that the quadratic behaviour gives a less divergent beam but with a more rapid amplitude decrease. If a linear process is used, the ionization obtained is less sensitive to the optical quality of the beam, and good relative gain calibration of the different wires can be reached. However, a good control of the required impurity may be too difficult to reach for an absolute calibration of amplification. For this purpose it is possible to use X-ray radioactive sources or the Compton edge with a monochromatic pulsed X-ray beam\(^8\).

Secondly, this good simulation of a track by a UV laser beam makes it also potentially useful for understanding fundamental properties of proportional chamber systems and of the attached electronics. The "track" is fixed, has no Landau fluctuation (see fig. 5b), and can be varied in intensity. Such a method can bring
substantial simplification to the study of two-particle separation, cross-talk, electric field shaping, angular behaviour, dE/dx, space-charge problems, etc.

Acknowledgements


Discussions with G. Mainfray, P. Agostini and R. Papoular helped us in formulating the proposed physical interpretation of our findings.

We would like to thank particularly the people who lent us the necessary equipment for this test: the laboratory of ionized media physics from the Ecole polytechnique, Paris; E. Rossa and D. Brahy from CERN, who lent us a N₂ laser; and Messrs. Arie and Brassard from Quantel who made a Yag laser available to us. Mr. Brassard took part in some of the reported measurements (fig. 7).

These tests would not have been possible without the active support of the engineers and technicians who built the chambers and the electronics.
APPENDIX

INTERPRETATION OF THE LINEAR DEPENDENCE IN TERMS
OF DOUBLE-STEP ABSORPTION

The observed linearity can be understood in terms of double-step photon absorption. In the absence of a definite proof such as the UV absorption spectrum of our gas, we may present the following hypotheses and check that the orders of magnitude are compatible with our observations.

a) Amongst the impurities in the gas, there are complex hydrocarbon molecules (pump oil?) which happen to have an ionization potential lower than 9.3 eV and an absorption line close to 265 nm. Therefore it is the process of fig. 1b which is operative. This hypothesis is not unlikely since it is known that complex molecules have between 300 and 200 nm closely spaced "vibronic" excitation states, i.e. electronic excitation states which are multiplied by the various vibration modes of the molecules.

b) The first transition is saturated, i.e. a small fraction of the beam pumps up half of the impurity molecules and an equilibrium is reached soon after the beginning of the pulse between excitation and stimulated emission. This requires that two conditions be verified:

i) The inverse of the excitation cross-section should be much smaller than the number of photons per unit area. This can be the case since 1 μJ/0.1 mm² is equivalent to 1.3 × 10¹⁵ photons/cm², while typical excitation cross-sections \(^3\) are 10⁻¹¹ cm².

ii) The number of molecules should be small enough to absorb a negligible fraction of the beam. Assuming a 1% absorption over 1 m for \(n_γ/s = 1.3 \times 10^{15}\) photons/cm² requires a concentration \(p\) such that

\[
\frac{1}{2} p \times N < 10^{-4} n_γ/s
\]

\[
p < \frac{2 \times 10^{-8} \times 1.3 \times 10^{15}}{2.7 \times 10^{15}} \approx 10^{-8} \text{ atm}.
\]
c) This excited state lives long enough with respect to the duration of the laser pulse. Typical relaxation times\textsuperscript{3} are 10 to 50 ns, and the time between collisions at normal pressure and temperature is of the same order of magnitude.

d) Therefore, after a minute fraction of the beam has been fully absorbed, any incoming photon can ionize the pumped state directly, and the number of electrons per cm is

\[ n_e = \frac{1}{2} p N \sigma_i n_i \]

which, with \( \sigma_i \approx 5 \times 10^{-17} \text{ cm}^2 \), \( p = 10^{-8} \), \( n_i = 1.3 \times 10^{12} \), gives

\[ n_e = \frac{1}{2} \times 10^{-8} \times 2.7 \times 10^{18} \times 5 \times 10^{-17} \times 1.3 \times 10^{12} \approx 10^7 \text{ electrons/cm}. \]

This is much bigger than what we observed, and indicates that either \( p \) or \( \sigma_i \) can be decreased. This shows the extreme sensitivity of the process. In fact it is used for single atom detection\textsuperscript{10} and isotopic separation.
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Figure captions

Fig. 1 : a) Double photon absorption.
        b) Double-step absorption.

Fig. 2 : The Ecole polytechnique set-up.

Fig. 3 : The CERN projection chamber.

Fig. 4 : Time of arrival of electron versus position (arbitrary origins).

Fig. 5 : a) Pulse on six different wires, produced by a laser beam parallel to the wire plane.
        b) Pulse on one wire, produced by a laser beam approximately perpendicular to the wire plane. The small pulse produced on wire 3 may be due to electrons extracted from the 30 kV cathode (fig. 3).

Fig. 6 : Evidence for double photon absorption: pulse height versus laser intensity (log scales).

Fig. 7 : a) Linear plot of the number of electrons versus laser intensity (one set of measurements).
        b) Summary of all measurements on a log scale.

Fig. 8 : Effect of diffraction:
        a) Linear ionization;
        b) double photon absorption.

(Slot width = 0.5 mm, pulse width = 3 ns FWHM).
\begin{align*}
\text{ionization threshold} \\
\sigma_2 \\
\tau \sim 10^{-16} \text{s} \\
\sigma_1 \sim 10^{-16} - 10^{-17} \text{ cm}^2 \\
\end{align*}

\begin{align*}
\text{ionization threshold} \\
\sigma_2 \\
\tau \sim 10 - 50 \text{ ns} \\
\sigma_1 \sim 10^{-11} \text{ cm}^2 \\
\end{align*}

Fig. 1
Fig. 3
Fig. 4

Time of arrival (ns) vs. Position (mm)

26 ns/mm

Fig. 5

a) wire #1
wire #2
wire #3
wire #4
wire #5
wire #6

~35 ns per bin

b) wire #1
wire #2
wire #3
wire #4

~45 ns per bin
Fig. 7