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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
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H4.SMR/449-9

WINTER COLLEGE ON HIGH RESOLUTION SPECTROSCOPY

(8 January - 2 February 1990)

MULTIPHOTON IONISATION OF ATOMS

II Strong Fields

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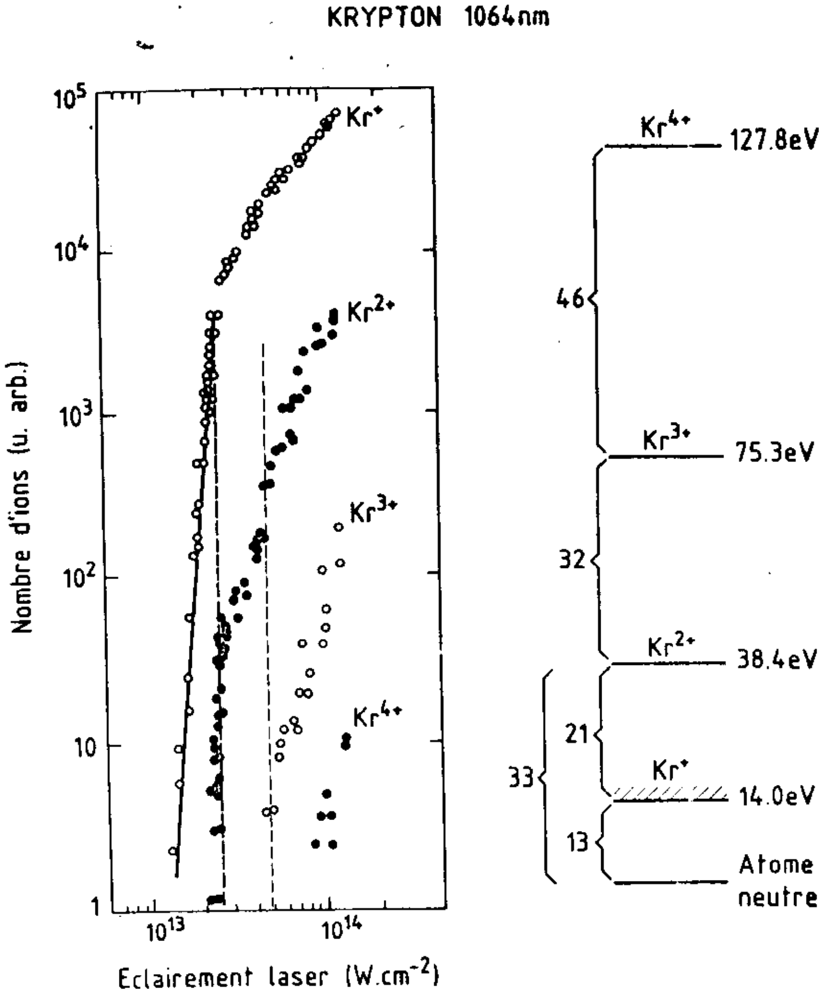
Formation of multicharged ions



(a) (b)

Figure 26- (a) Nombres d'ions Kr^+ à Kr^{4+} en fonction de l'éclairement laser à 1064 nm.
(b) Schéma des énergies d'ionisation du krypton.

Experiments

- alkaline earths, rare earths
  - Suran - Zopesochny 1975..... } dye laser. ~10^{10} Wcm^{-2} nsec.
  - Feldmann et al 1982..... }
  - Agostini - Petite 1985 ~ 10^{12} Wcm^{-2} psec
- noble gases
  - L'Huillier, Lompré, Mainfray, Manus 1982
    - 1.06μ. 0.53μ. → 10^{14} Wcm^{-2}. psec
  - Chin - Yergeau - Lavigne 1985
    - CO_2 laser → 10^{14} Wcm^{-2} n.sec.
  - Luk et al 1983 193nm → 10^{17} Wcm^{-2} psec.
  - Perry - Landen 1988
    - dye laser ~ 10^{14} Wcm^{-2} psec.
    - ~ 300nm → 10^{14} Wcm^{-2} psec.

Theoretical investigations.

- Statistical interpretations
  - Åberg et al 1983
  - Crance 1984
  - Geltman 1985
- Quantum calculations
  - screening of the field Wendin L'Huillier 1986
  - double ionisation Crance Aymar 1985
    Wendin L'Huillier 1987

Questions

Once single ionisation has been reached, it is easy to obtain multiple ionisation. Why?

Do electrons leave the atom one by one or several together?

Ions are left in ground state or in excited states?

How does the process vary with the laser frequency?

What parameters characterize the process?

How can various experiments be related?

→ simple features.
- time development
- coherence effects
- statistical interpretation

D. Feldmann, J. Krautwald, S.L. Chin, A. von Hellfeld, K.H. Welge
J.Phys.B15 (1982) 1663

9395cm⁻¹ →

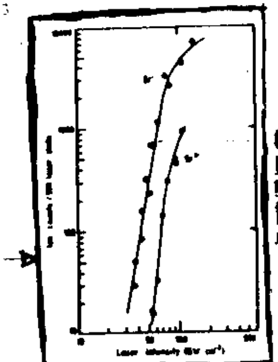


Figure 3. Log-log plots of the ion counts against laser intensity at λ = 10 642 Å.

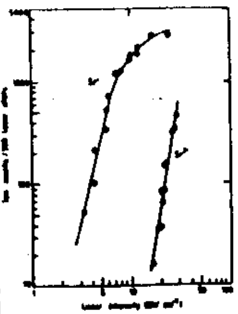


Figure 5. As figure 3, λ = 5321 Å.

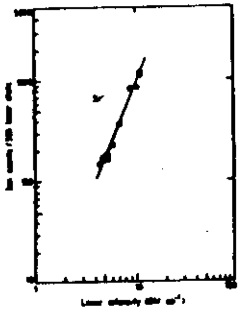


Figure 4. Log-log plots of the ion counts against laser intensity at λ = 3547.3 Å.

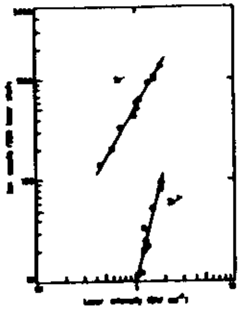


Figure 6. As figure 4, λ = 2660.5 Å.

9450 cm⁻¹

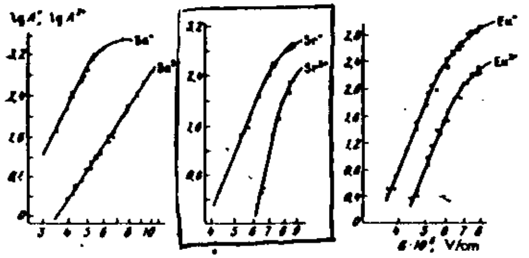
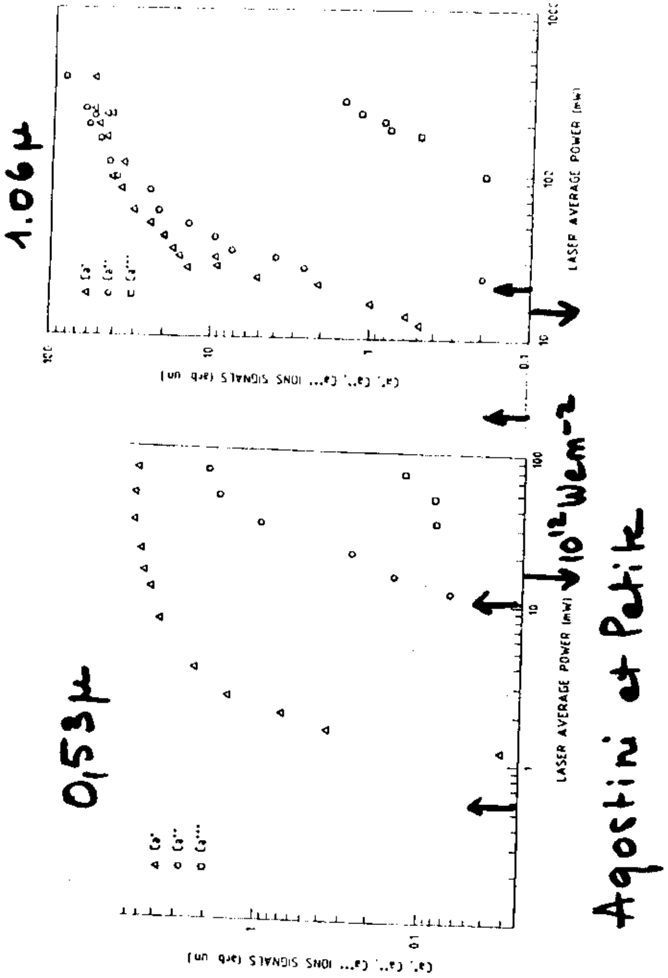
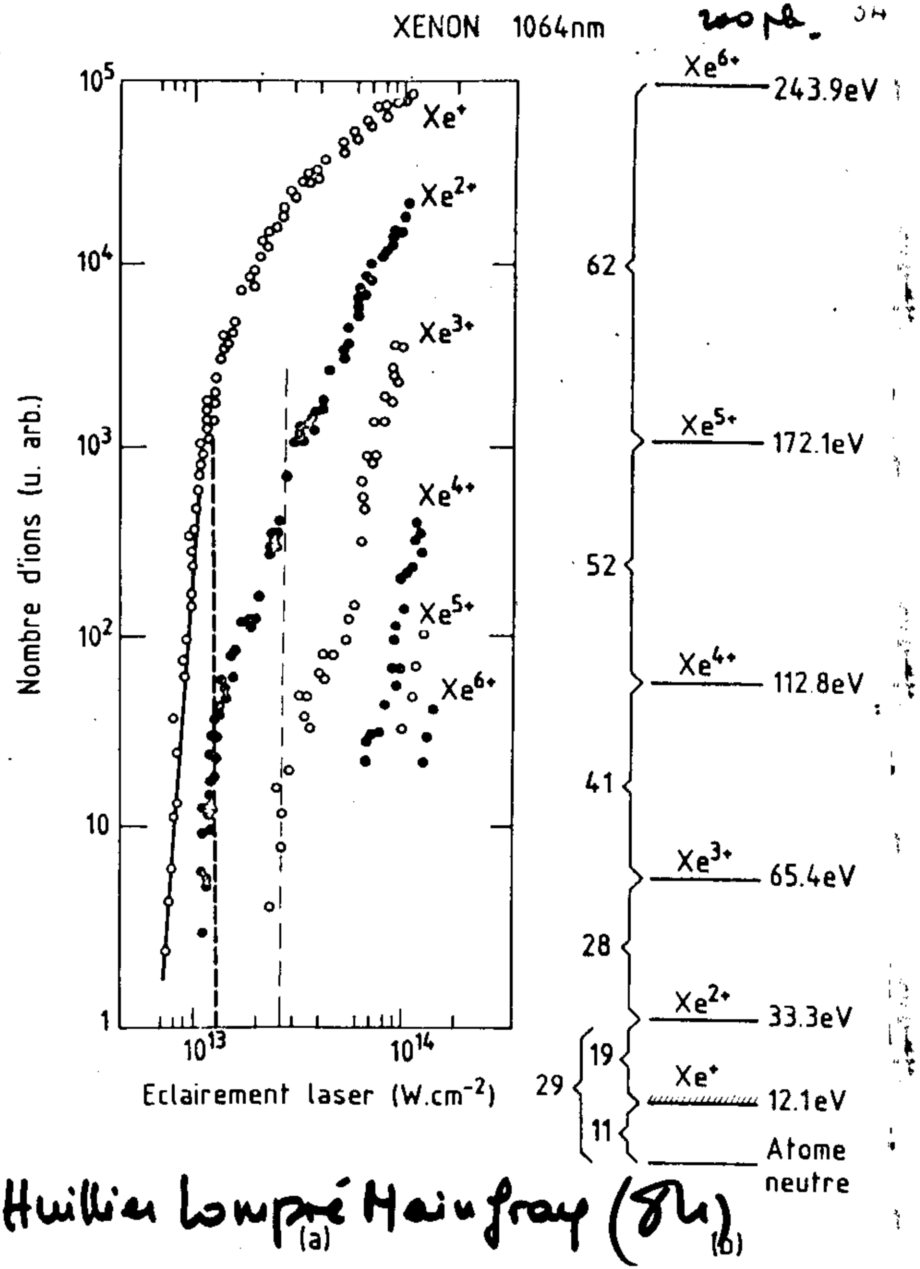


FIG. 2. Amplitudes of the ion signals from singly- and doubly-charged ions of barium, strontium, and europium vs the field strength 𝓔.

[illegible] N.B. Delone, I.P. Zapesochnyi, V.V. Suran Sov. Phys. JETP 48 (1979) 447

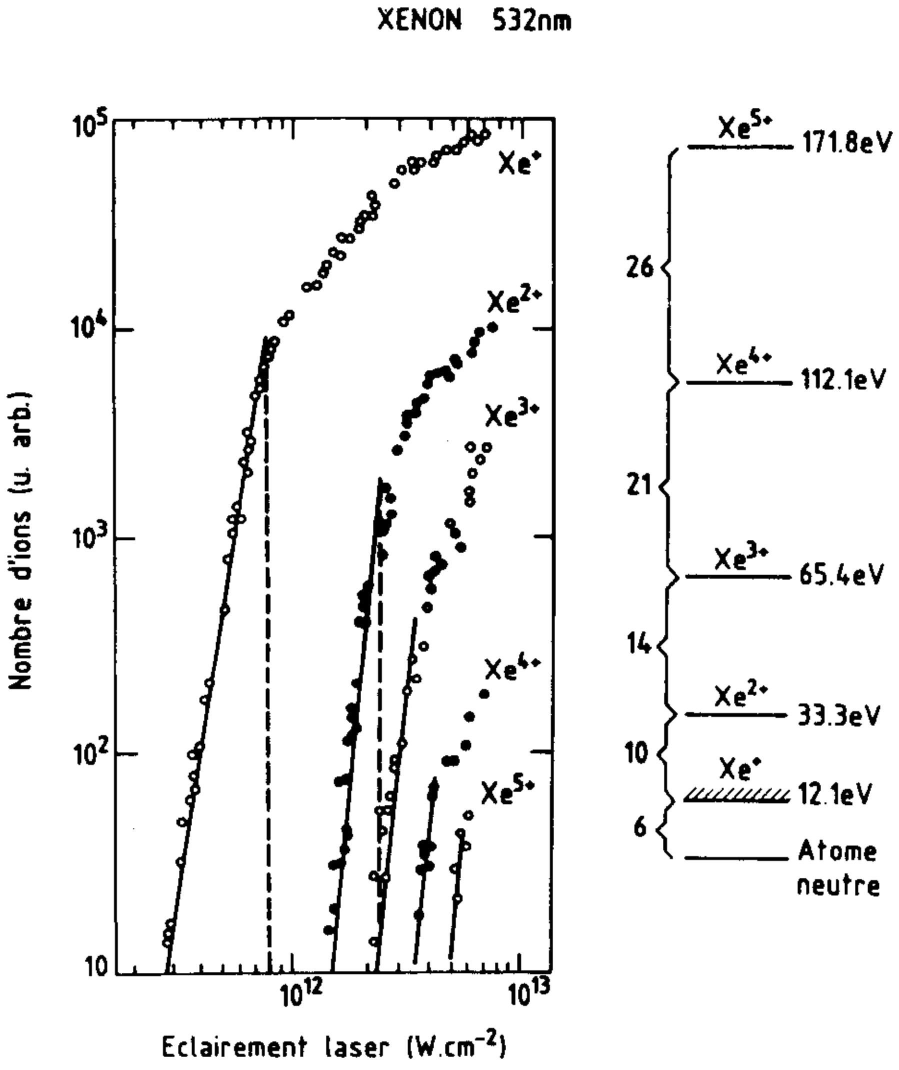


200 ps.



L'Huillier Lompré Mainfray (84)

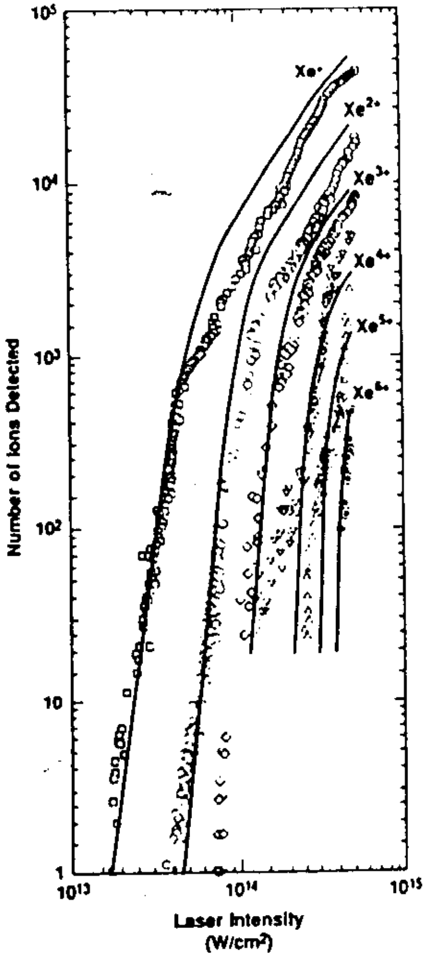
Figure 20- (a) Nombres d'ions Xe^{+} à Xe^{6+} en fonction de l'éclairement laser sur une échelle doublement logarithmique.
(b) Schéma des énergies d'ionisation du xénon.



(a) (b)

Figure 32- (a) Nombres d'ions Xe$^+$ à Xe^{5+} en fonction de l'éclairement laser (seuil de détection élevé d'un facteur 10).
(b) Schéma des énergies d'ionisation du xénon.

L'Huillier et al (1984)



Perry et al Phys Rev A 37 747 (88)
5860 Å ~1 ps

K.Sc er, H.Egger, T.S.Luk, H.Pummer, C.K.Rhodes JOSA B (1984) 1

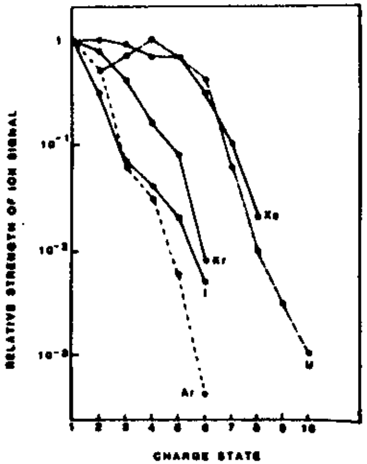


Fig. 4. Relative strength of ion signals for various atoms. No value is given for U^+ because of the interference with UF^+, UF_2^+, etc.

143nm

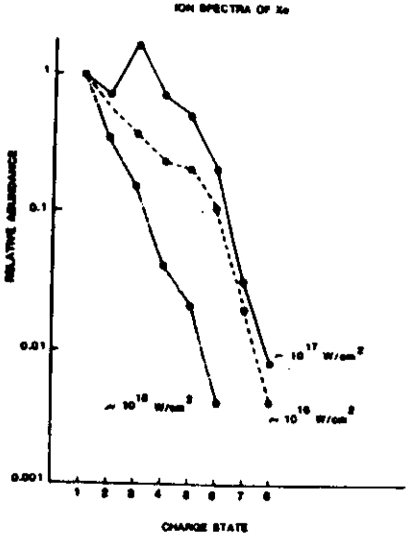
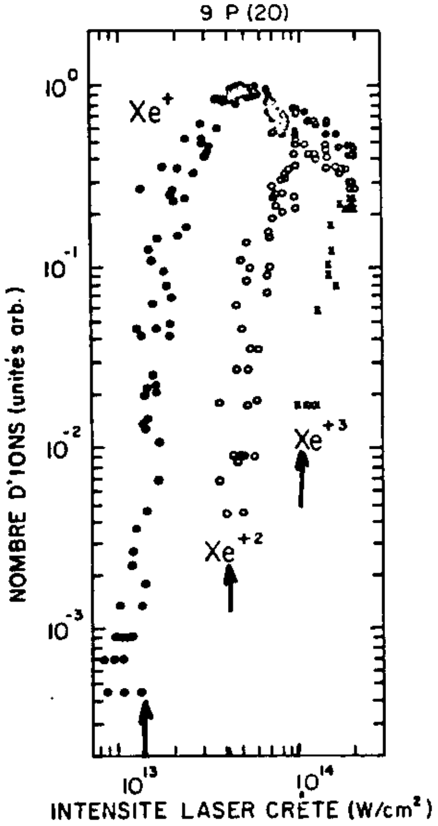


FIG. 5. Relative abundance of charge-state distributions observed in the ion spectra of xenon in the intensity range $10^{15}-10^{17}$ W/cm^2 at 193 nm.

T.S.Luk, U.Johann, H.Egger, H.Pummer, C.K.Rhodes Phys.Rev.A32 (1985) 214



Chin Yergeau Lavigne

Figure 3.4 - Nombre d'ions de Xe versus Intensité crête en échelle doublement logarithmique avec le laser 1.1 ns à 9.55 µm.

Double ionisotion : direct vs stepwise proces

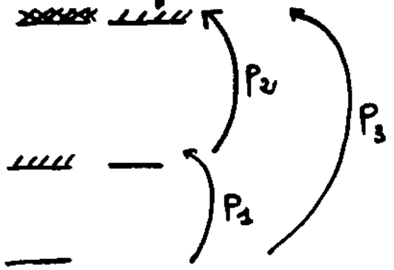
n_0 neutial
n_1 singly charged ions
n_2 doubly charged ions

$$\dot{n}_0 = -(\Gamma_{01}+\Gamma_{02})n_0$$
$$\dot{n}_1 = -\Gamma_{12}n_1 + \Gamma_{01}n_0$$
$$\dot{n}_2 = \underbrace{\Gamma_{02}n_0}_{\text{Direct}} + \underbrace{\Gamma_{12}n_1}_{\text{stepwise}}$$

Perturbative solution

$$n_0 = 1 \quad n_1 = \Gamma_{01}t \quad n_2 = \Gamma_{02}t + \Gamma_{01}\Gamma_{12}\frac{t^2}{2}$$

→ diferent time-development.



Coherence effects

$$\frac{\text{Yield in chaotic light}}{\text{Yield in coherent light}} = n!$$

Direct process : p_3 photons simultaneously absorbed

Stepwise process: p_1 photons simultaneously absorbed incoherently followed by p_2 photons simultaneously absorbed

chaotic versus coherent enhancement =

$p_3!$ for direct process.
$p_1!\,p_2!$ for stepwise process.

Example Xenon 1.06μ. $p_1 = 11$ $p_2 = 19$ $p_3 = 29$
$p_1!\,p_2! = 4.8\ 10^{24}$ $p_3! = 8.8\ 10^{30}$

Statistical interpretation

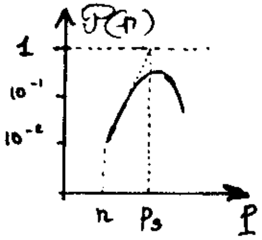
Noble gases. outer shell $ms^2\,mp^6$
comparable orbital size and extioction potential.

↳ $S = \pi\langle r\rangle^2$ ↳ E ionisation potential

$\frac{E}{\hbar\omega} \sim n$ minimum number of absorptions required

Assumption: an election is ejected if it absorbs at least n photons in a time $\leqslant T = \frac{\hbar}{E}$

Intensity I → $\frac{IST}{\hbar\omega} = p$ photons



8 elections share p photons
→ ionisation probability $\mathcal{P}(n)$
→ saturation p_s

$$I_s(T) = \frac{p_s\hbar\omega}{ST} = (\sigma^{(n)}T)^{\frac{1}{n}} \longrightarrow \sigma^{(n)}$$

gives an estimate of
- ionisation probability
- direct vs stepwise processes.
- in ground state vs excited states -

Saturation intensities

excitation 0.53μ   0.586μ

	Xe	Ne	Xe	Kr	Ar
exp.	$6.8\ 10^{11}$	$8.6\ 10^{13}$	4.10^{13}	5.10^{13}	$6.7\ 10^{13}$
th.	$6.8\ 10^{12}$	$2.2\ 10^{14}$	10^{13}	$2\ 10^{13}$	$4.4\ 10^{13}$

excitation 1.06μ

	Xe	Kr	Ar	Ne
exp.	10^{13}	$2.1\ 10^{13}$	$2.55\ 10^{13}$	$3.5\ 10^{14}$
th.	$1.24\ 10^{13}$	$2.15\ 10^{13}$	$4.67\ 10^{13}$	$3\ 10^{14}$

excitation 10μ. $\times 10^{13}$

ionisation from	Xe	Xe^+	Xe^{++}	Kr	Kr^+	Ar	Ar^+	Ne	He
exp	1.2	4.8	12	1.9	8.8	2.4	12	6.5	10
th	1.3	4.1	9.7	2.5	7.7	4.5	13.6	19.7	34.5

Saturation intensity

$\sigma^{(n)} I^n$ = probability per unit time

$\sigma^{(n)} I^n \tau$ = probability per pulse

$\sigma^{(n)} I_s^n \tau = 1$ defines saturation intensity

$$I_s = (\sigma^{(n)}\tau)^{-\frac{1}{n}}$$

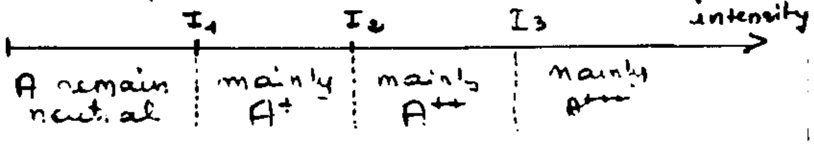
allows one to relate experiments with ≠ pulse durat

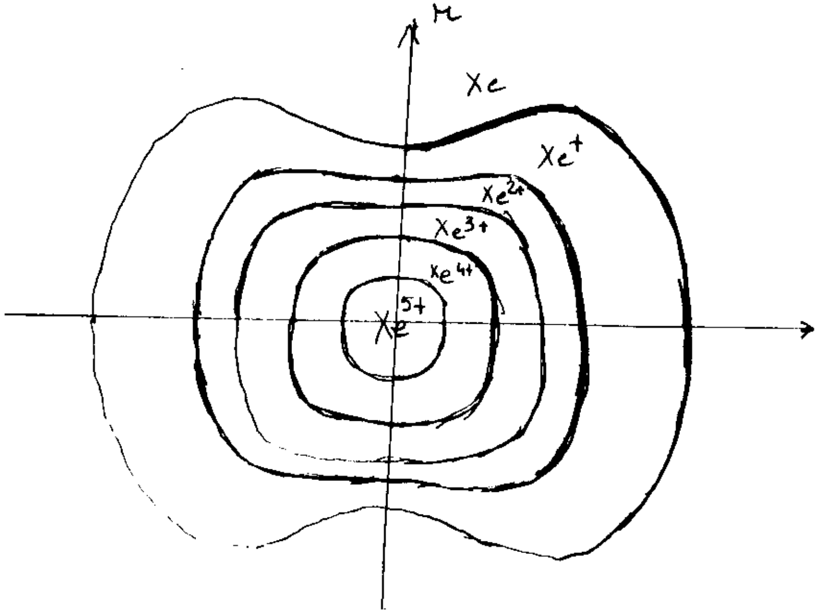
$A \longrightarrow A^+ + e^-$ saturation intensity I_1

$A^+ \rightarrow A^{++} + e^-$ — I_2

$A^{++} \rightarrow A^{+++} + e^-$ — I_3

$$I_1 < I_2 < I_3 \ \ldots\ldots$$





Application to a gaussian beam

$I(r) = I_M \, e^{-r^2}$

Volume where $I > I_0 \propto \mathrm{Log}\,\frac{I_M}{I_0}$

$A \rightarrow A^+$ saturation I_1

$A^+ \rightarrow A^{++}$ — I_2

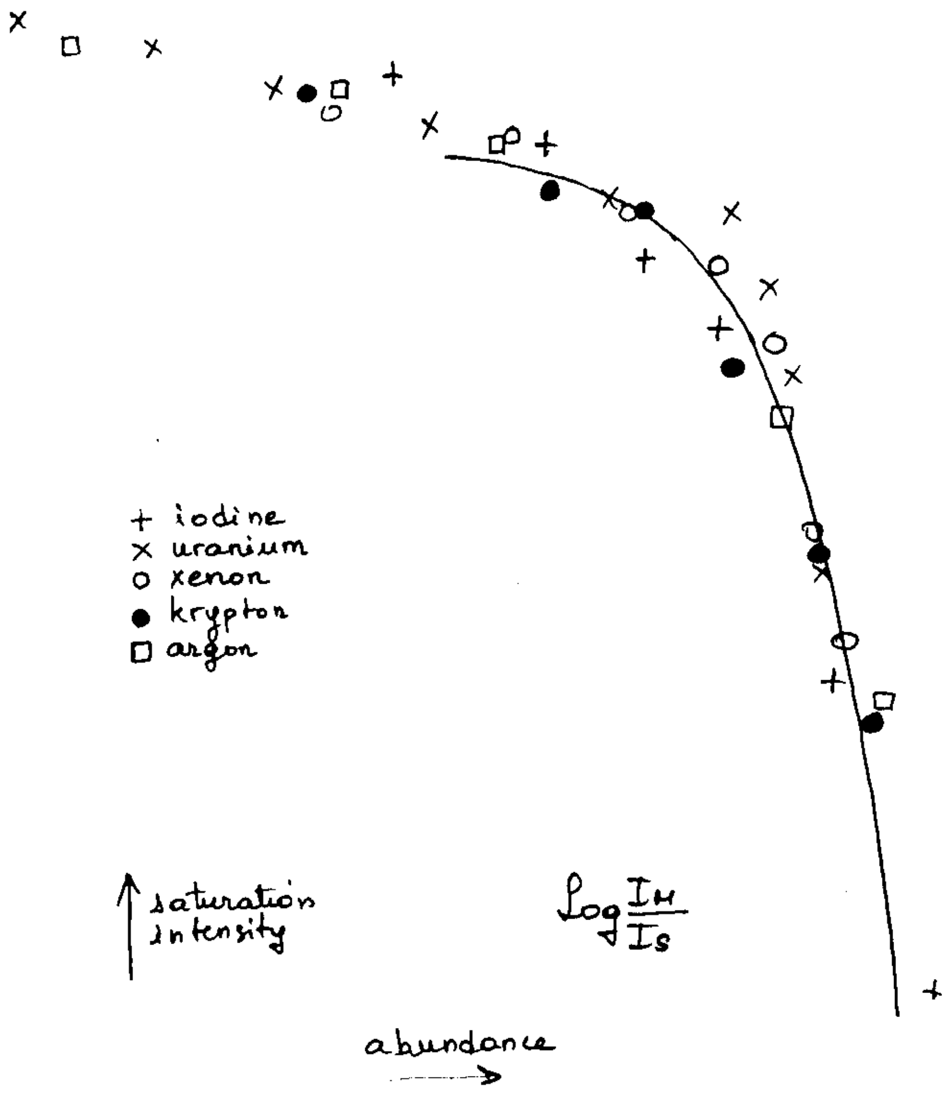
....

number of ions $A^+ A^{2+} \ldots\ldots \propto \mathrm{Log}\,\frac{I_M}{I_1}$

number of ions $A^{2+} A^{3+} \ldots\ldots \propto \mathrm{Log}\,\frac{I_M}{I_2}$

cumulated abundances should fit $\mathrm{Log}\,\frac{I_M}{I_S}$

Theoretical saturation intensities
versus
experimental abundances (from Bayer et al JOSA B 1 (1984) 1)



Ratio A^{++}/A^{+} at saturation intensity of $A \rightarrow A^{+}$

$A + 6\hbar\omega \rightarrow A^{+} + e^{-}$ $A^{+} + 10\hbar\omega \rightarrow A^{++} + e^{-}$

the process needs 16 photons: negligible

$A + 15\hbar\omega \rightarrow A^{++} + 2e^{-}$ direct

$A + 10\hbar\omega \rightarrow A^{+*} + e^{-}$; $A^{+*} + 5\hbar\omega \rightarrow A^{++} + e^{-}$

stepwise

Two possible processes involving 15 photons

		Xe	Kr	A	Ne
exp.		0.015	0.007	.006	.0015
th.	direct	10^{-6}	7.10^{-8}	$3\,10^{-9}$	$1.4\,10^{-15}$
	stepwise	.026	.005	.01	$4\ 10^{-8}$

= Resonance $Ne^{+} + 23\hbar\omega$ detuned by $174\ cm^{-1}$

$$:(2s^2 2p^5)\,{}^2P_{1/2} \rightarrow (2s\,2p^6)\,{}^2S_{1/2}$$

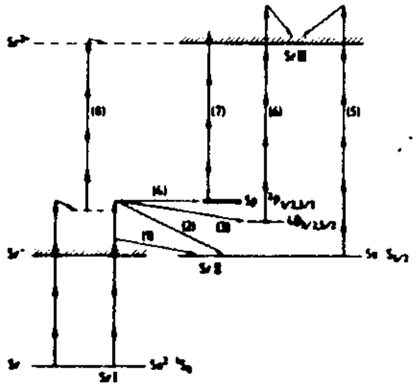


FIG. 1. Energy-level diagram of Sr I and Sr II showing the processes discussed in the text. The process labeled (8) is an example of "direct" or "nonresonant" double ionization.

Multiphoton ionisation of Strontium

~ 570 nm

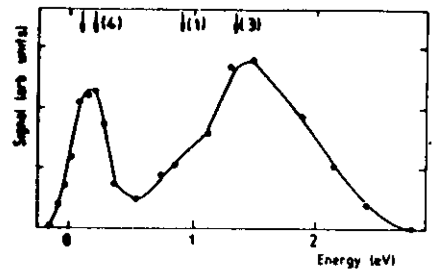


FIG. 2. Typical electron-energy spectrum at 100 GW cm⁻². The numbers inside parentheses refer to the processes illustrated in Fig. 1.

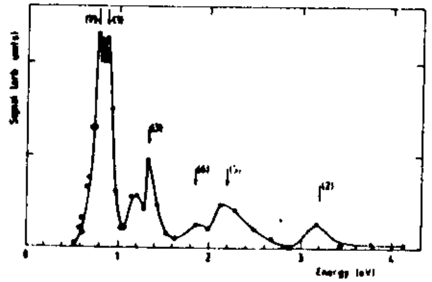


FIG. 4. Typical energy spectrum at 2.8 TW cm⁻² showing the 2.2-eV component (arrow) corresponding to the second ionization. The numbers in parentheses refer to the processes illustrated in Fig. 1.

Agostini Petite PRA 32 3800 (1985)

Harmonic generation

For an atom

$$|\psi(t)\rangle = a_0|0\rangle + \mathcal{E}\sum a_\alpha^{(1)}|\alpha\rangle + \dots + \mathcal{E}^{2p}\sum a_i^{(2p)}|i\rangle$$

$$\dots + \mathcal{E}^{2p+1}\sum a_\alpha^{(2p+1)}|\alpha\rangle$$

opposite parity

same parity as $|0\rangle$

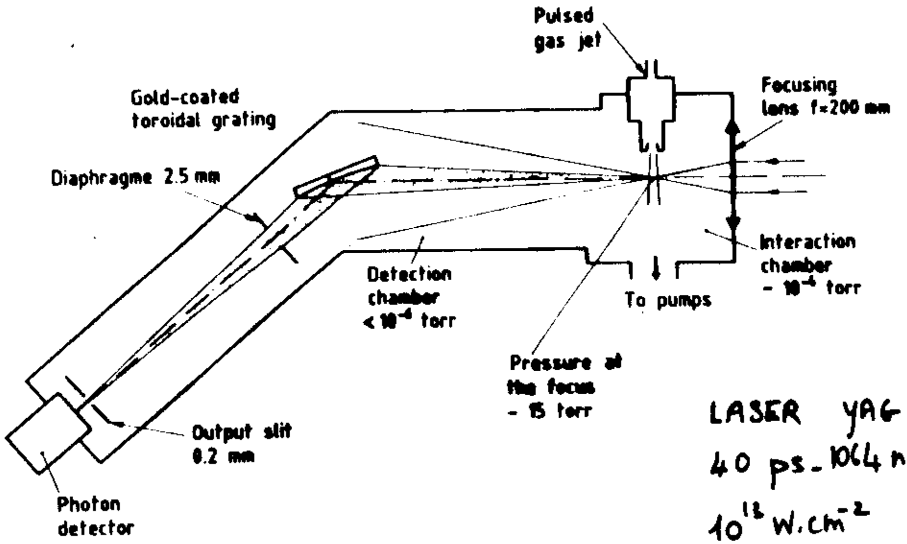
polarisation $\langle\psi(t)|z|\psi(t)\rangle$

polarisation at frequencies $(2p+1)\omega$
with leading term $a_0^* a_\alpha^{(2p+1)} \propto \mathcal{E}^{2p+1}$

Starting point : Observation of multicharged ions
What states are reached ? → look at fluorescence
Need to increase the pressure -
Does the process involve atoms or ions ?

Open question : phase matching conditions.

EXPERIMENTAL SET.UP

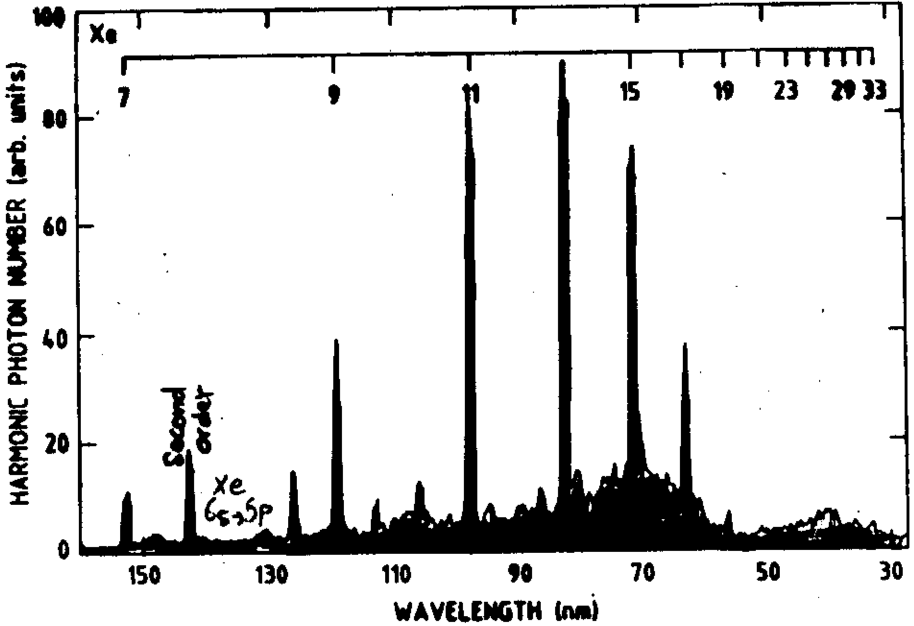


LASER YAG
40 ps - 1064 nm
10^{13} W.cm^{-2}

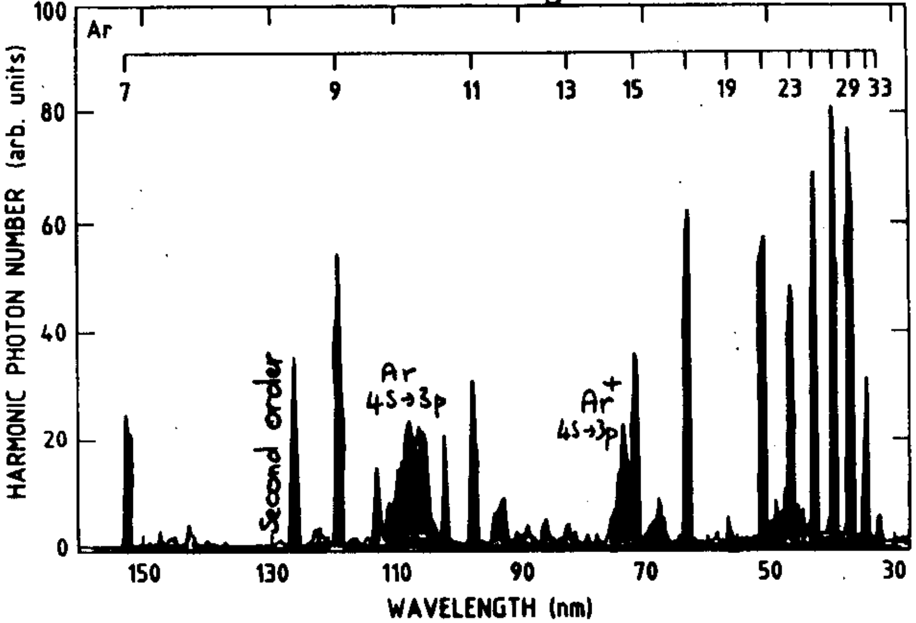
10 nm $< \lambda <$ 400 nm

L'Huillier et al 1988

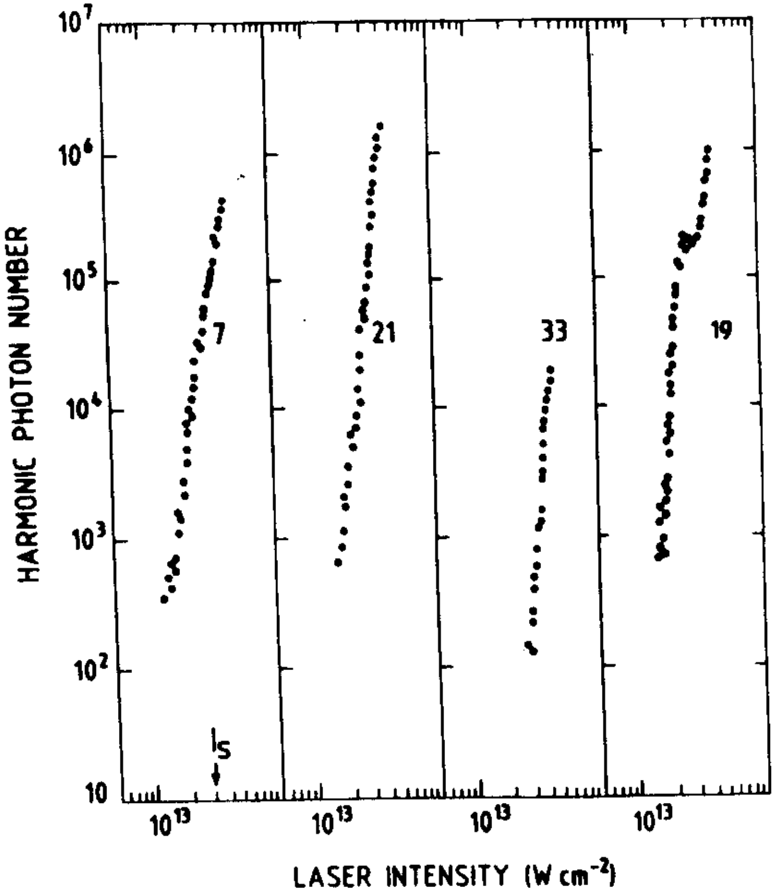
PHOTON SPECTRUM Xenon
5×10^{13} W.cm^{-2} 15 Torr



PHOTON SPECTRUM 5×10^{13} W.cm^{-2}
Argon 15 Torr.

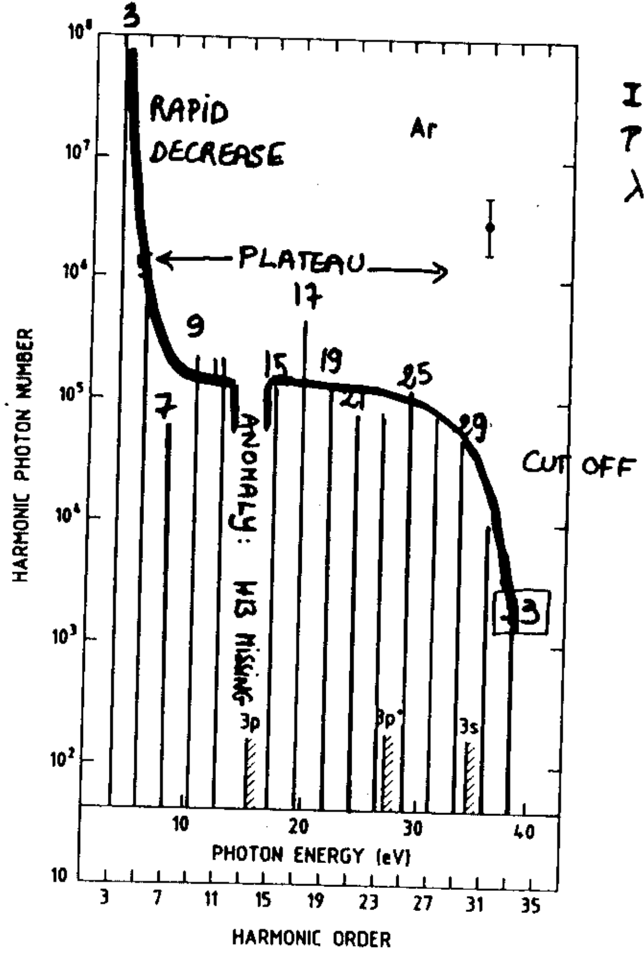


ODD HARMONICS
FLUORESCENCE (RECOMBINATION)
CONTINUOUS BACKGROUND



L'Huillier et al 1988

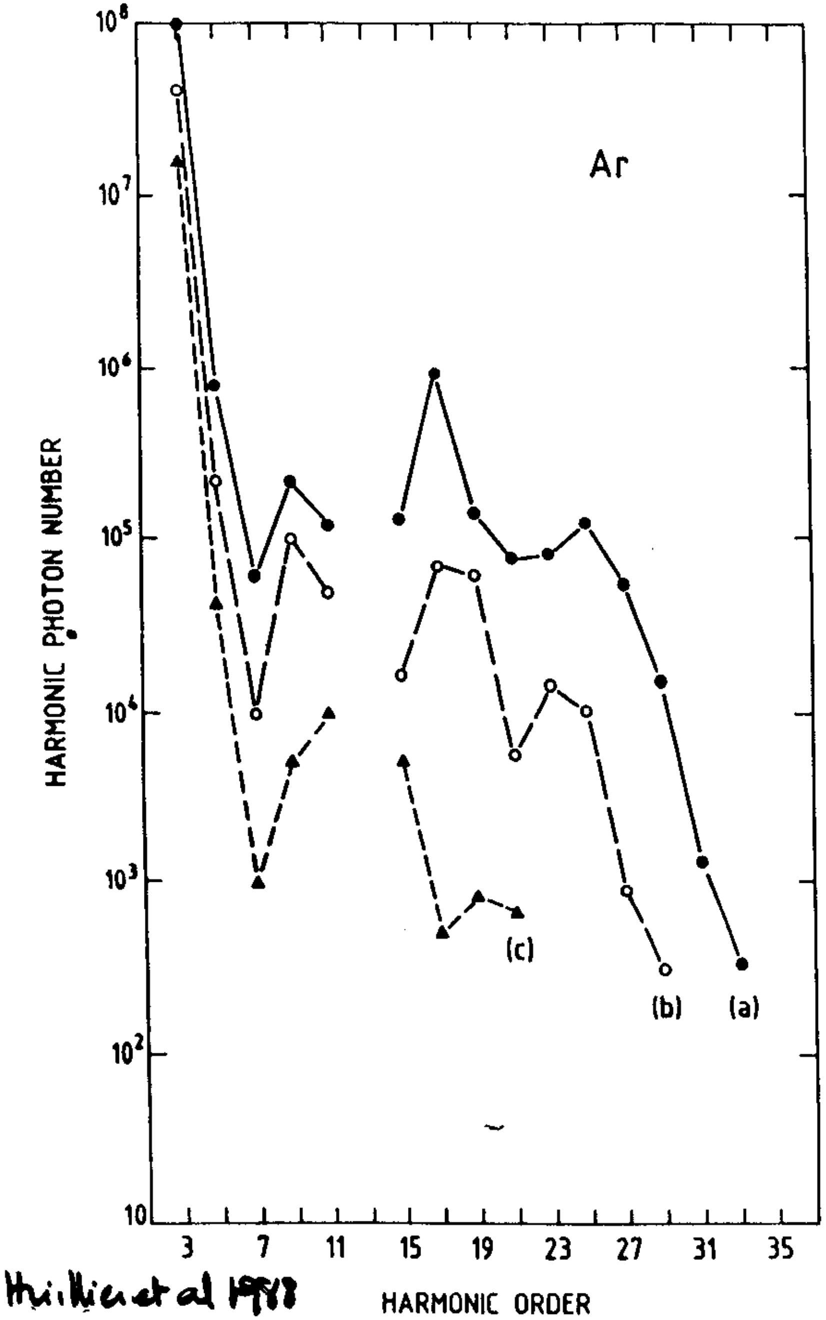
HARMONIC DISTRIBUTION



$I = 3 \times 10^{13}$ W.cm^{-2}

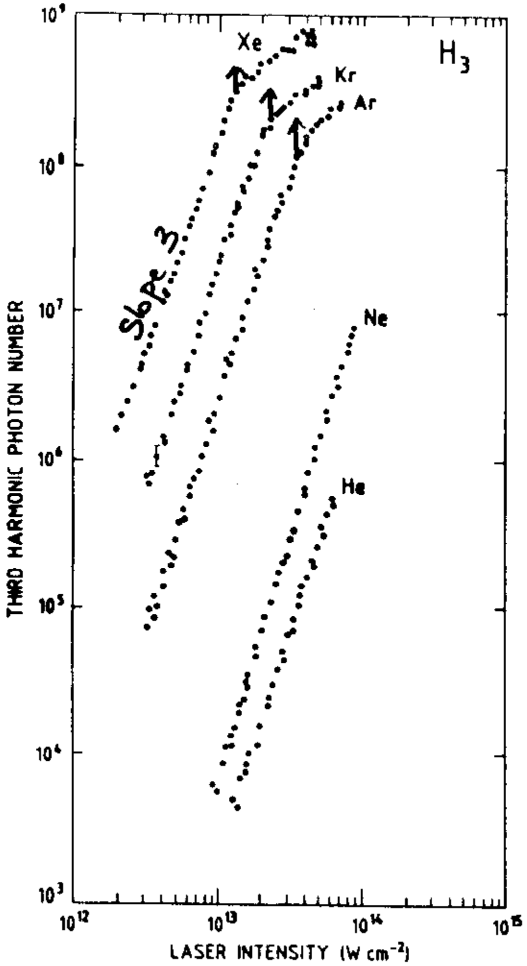
$P = 15$ Torr

$\lambda = 1064$ nm



L'Huillier et al 1988

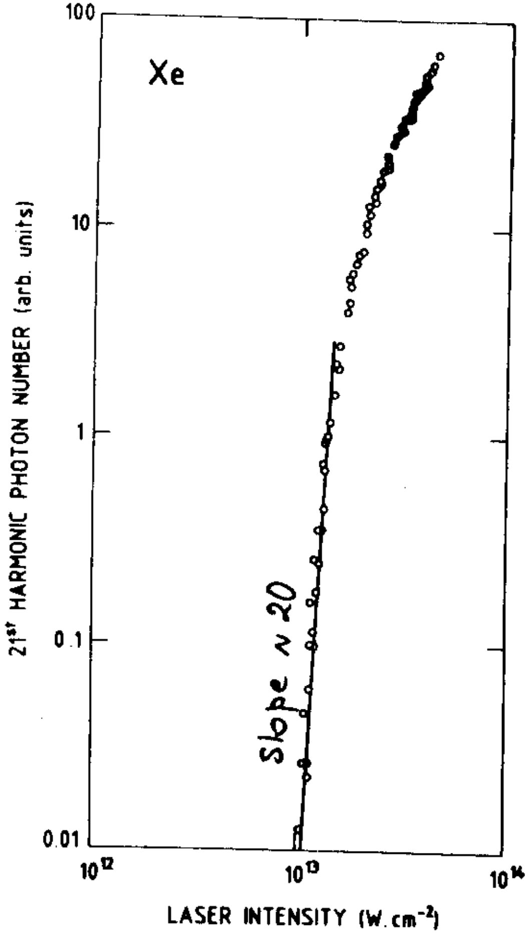
THIRD. HARMONIC GENERATION.



SATURATION WHEN THE MEDIUM IS IONIZED

L'Huillier et al 1989

HIGH-ORDER HARMONIC GENERATION



SATURATION
DUE TO
IONIZATION

L'Huillier et al 1989