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H4.SMR/449-8

WINTER COLLEGE ON HIGH RESOLUTION SPECTROSCOPY

(8 January - 2 February 1990)

MULTIPHOTON IONISATION OF ATOMS

I Weak Field

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Multiphoton ionisation of atoms

Introduction
- Typical set up
- Interpretation
- Orders of magnitude

Weak field effect
- Perturbation theory at lowest non vanishing order
- Power laws. Saturation. Coherence effect
- Calculation of cross sections.
- Resonances. effective hamiltonian.
- Measurement of cross sections.

Strong field
- Multicharged ions
- Harmonic generation

Electron energy spectrum.
- Calculation of cross sections
- Examples
- Angular distribution.
- leaving the interaction volume
- Short pulses
- Non perturbative calculations

Multiphoton ionisation of atoms

Theoretical prediction of two-photon absorption
Goppert Mayer Ann Phys 9 273 (1931)
Multiphoton ionisation invoked to explain breakdown in gases
Meyerand + Haught PRL 11 401 (63) PRL 13 7 (64)
Phenomenological model Keldysh Sov Phys JETP 20 1307 (65)
Multiphoton ionisation of noble gases
Voronov + Delone Sov. Phys JETP 23 54 (66).
Agostini Barjot Bonnal Mainfray Manus IEEE QE 4 667 (68)

Laser pulses become stronger and shorter

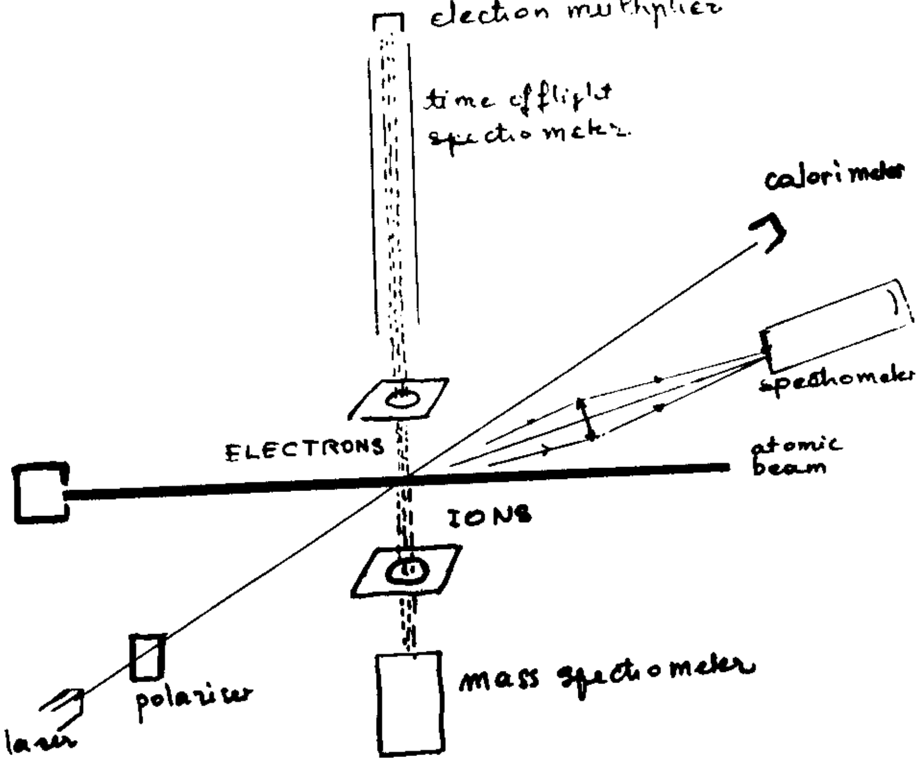
Improvement in fast electronics → pulse characteristics
in detection of electrons

Moderate fields – Yield of singly charged ions.

Strong fields – Multicharged ions.
– Harmonic generation

Short pulses – Electron yield.

Typical set up



laser pulse
frequency
bandwidth
time dependence
energy per pulse
polarisation
intensity distribution

electrons
total yield
energy spectrum
angular distribution

ions
yield
charge state distribution

emitted light
fluorescence
harmonics.

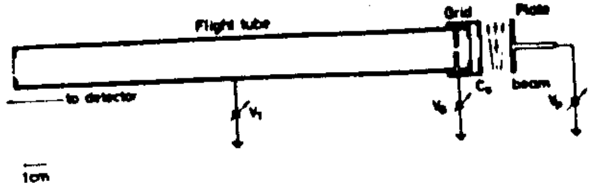


FIG. 5. Time-of-flight spectrometer.

L.Petite, F.Fabre, P.Agostini, M.Crance, M.Aymar Phys.Rev.A29 (1984) 2677

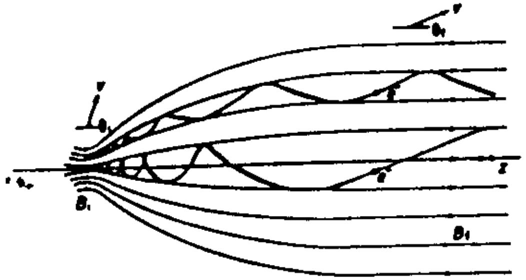
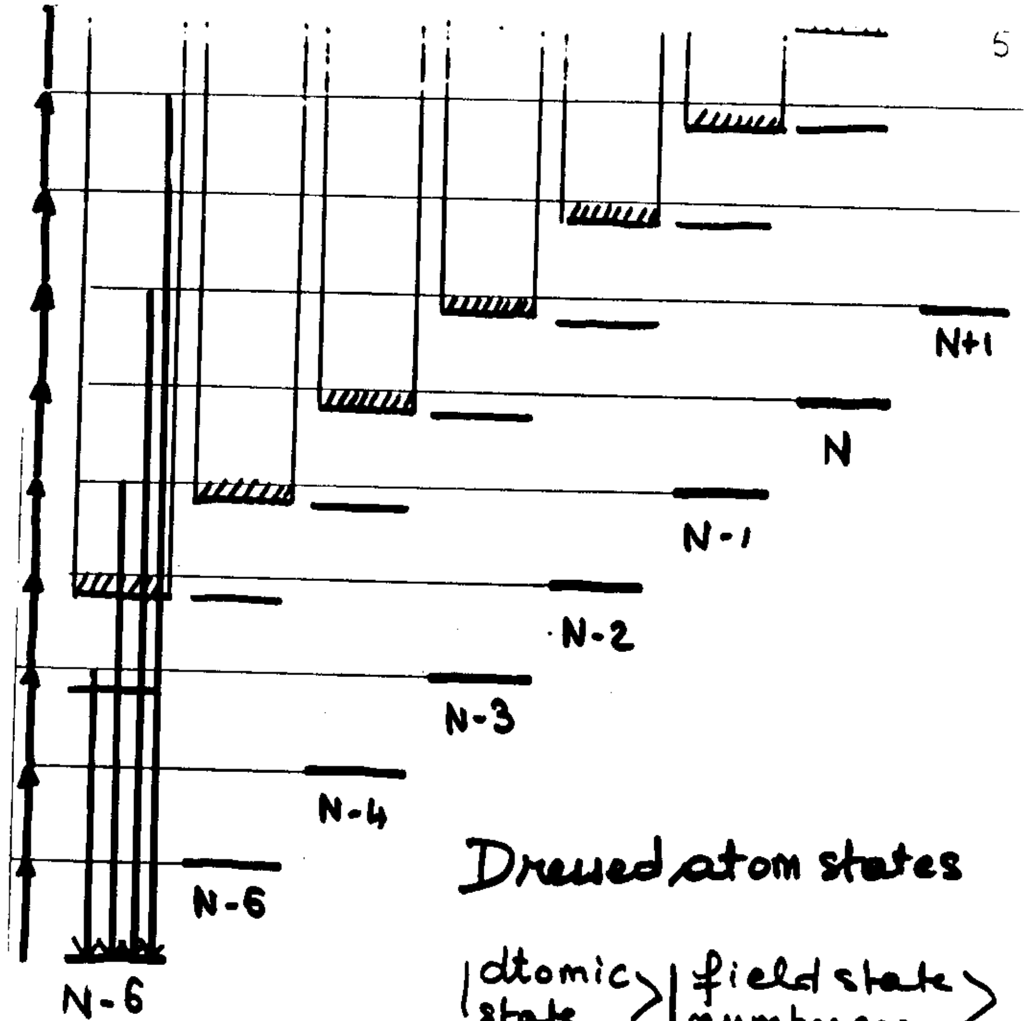


Figure 1. Schematic diagram showing the helical motion of an electron moving in a magnetic field that changes gradually from a strong field B_i to a weaker uniform field B_f.

P.Kruit, F.H.Read J.Phys.E16 (1983) 313



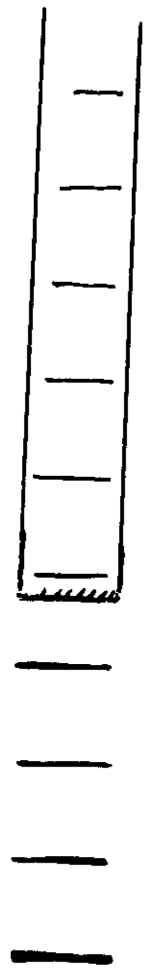
Dressed atom states

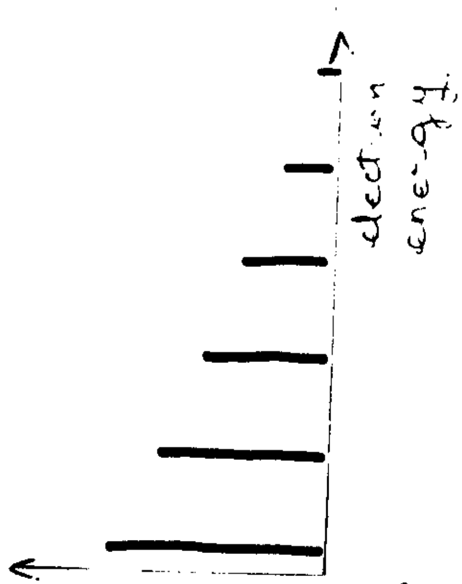
$|\text{atomic state}\rangle\,|\text{field state number rep.}\rangle$

$|g\rangle|N\rangle$
$|\varepsilon\rangle|N\rangle$

$|r\rangle|N\rangle$

$\varepsilon = E_g + 4\hbar\omega$





$\varepsilon_p = E_g + (n+p)\hbar\omega$

above threshold ionisation
or Excess Photon Ionisation
absorption of n+p photons.

What is a strong field?

ground – lowly excited states $\quad mW/cm^2$
→ saturation

bound – bound excited states $\quad \sim W cm^{-2}$

Resonant ionisation $\quad \sim 10^6\, W cm^{-2}$

Non resonant ionisation $\quad \sim 10^9\, W cm^{-2}$
in alkalis or alkaline earths

Non resonant ionisation $\quad \sim 10^{12}\, W cm^{-2}$
in noble gases

Excess Photon Ionisation $\quad \sim 10^{14}\, W cm^{-2}$
Multiple ionisation
Harmonic generation

→ STRONG SHORT PULSES

Perturbation theory at lowest non vanishing order.

Dipole approximation

time independent treatment → dressed atom pictures

$$H = H_{atom} + H_{field} + \vec{\mathcal{E}}.\vec{r}\,(a + a^+)$$

basis $|i\rangle \otimes |N\rangle$

time dependent treatment

$$H = H_{atom} + \vec{\mathcal{E}}.\vec{r}\cos\omega t$$

basis $|i\rangle$

$$|\Psi(t)\rangle = a_0 |0\rangle + \sum_i a_i |i\rangle + \sum_\alpha a_\alpha |\alpha\rangle$$

initial state $\quad$ even states $\quad$ odd states

$$i\hbar\frac{d}{dt}|\Psi(t)\rangle = H|\Psi(t)\rangle$$

$$i\dot{a}_0 = \sum_\alpha V_{0\alpha} a_\alpha \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$i\dot{a}_\alpha = \omega_\alpha a_\alpha + V_{0\alpha} a_0 \left(e^{i\omega t} + e^{-i\omega t}\right) + \sum_i V_{i\alpha} a_i \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$i\dot{a}_i = \omega_i a_i + \sum_\alpha V_{i\alpha} a_\alpha \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$E_0 = 0 \qquad \hbar V_{0\alpha} = \langle 0|\frac{\vec{\mathcal{E}}\vec{r}}{2}|\alpha\rangle$$

$$\hbar\omega_\alpha = E_\alpha \qquad \hbar V_{i\alpha} = \langle i|\frac{\vec{\mathcal{E}}\vec{r}}{2}|\alpha\rangle$$

$$\hbar\omega_i = E_i$$

→ double expansion according to
- perturbation order
- rotating / counter rotating terms

Perturbative treatment:

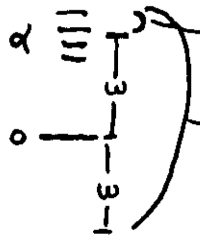
Order 0 $\rightarrow i\dot{a}_0 = 0 \rightarrow a_0 = 1$

Order 1 $\rightarrow i\dot{a}_\alpha = \omega_\alpha a_\alpha + V_{0\alpha} a_0 \left(e^{i\omega t} + e^{-i\omega t}\right)$

$$\rightarrow a_\alpha = V_{0\alpha} \left\{ \frac{e^{-i\omega_\alpha t} - e^{-i\omega t}}{\omega_\alpha - \omega} + \frac{e^{-i\omega_\alpha t} - e^{i\omega t}}{\omega_\alpha + \omega} \right\}$$

rotating term

counter rotating term



keep $a_\alpha = V_{0\alpha} \dfrac{e^{-i\omega_\alpha t} - e^{-i\omega t}}{\omega_\alpha - \omega}$

Order 2 $\rightarrow i\dot{a}_i = \omega_i a_i + \sum_\alpha V_{i\alpha} a_\alpha \left(e^{i\omega t} + e^{-i\omega t}\right)$

$$\rightarrow a_i = \sum_\alpha \frac{V_{0\alpha} V_{\alpha i}}{\omega_\alpha - \omega} \times \left\{ - \frac{e^{-i\omega_i t} - e^{-2i\omega t}}{\omega_i - 2\omega} - \frac{e^{-i\omega_i t} - 1}{\omega_i} + \frac{e^{-i\omega_i t} - e^{-i(\omega + \omega_\alpha)t}}{\omega_i - \omega - \omega_\alpha} + \frac{e^{-i\omega_i t} - e^{-i(\omega_\alpha - \omega)t}}{\omega_i + \omega - \omega_\alpha} \right\}$$

counter rotating

spurious term related to the sudden approximation

$$n_i = |a_i|^2 = \left\{ \sum_\alpha \frac{V_{0\alpha} V_{\alpha i}}{\omega_\alpha - \omega} \times \frac{2\left(1 - \cos(\omega_i - 2\omega)t\right)}{(\omega_i - 2\omega)^2} \right\}^2$$

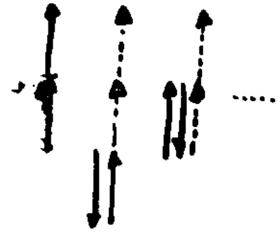
$\lim\limits_{t \to \infty} \dfrac{n_i}{t}$? 0 if $\omega_i \neq 2\omega$, ∞ if $\omega_i = 2\omega$ $\qquad \displaystyle\int_{-\infty}^{+\infty} \frac{2 - 2\cos\omega t}{\omega^2 t} d\omega = 2\pi\delta(\omega)$

Finally $\displaystyle\lim_{t \to \infty} \frac{n_i}{t} = \left[\sum_\alpha \frac{V_{0\alpha} V_{\alpha i}}{\omega_\alpha - \omega} \right]^2 2\pi\delta(\omega_i - 2\omega)$

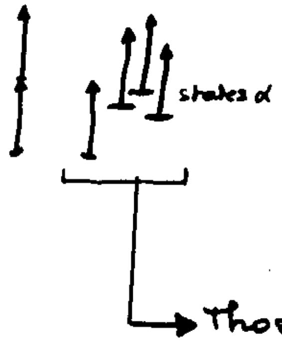
Perturbation theory at lowest non vanishing order.

The "spurious terms"

Counter rotating terms: they contribute at orders higher than the minimum one.

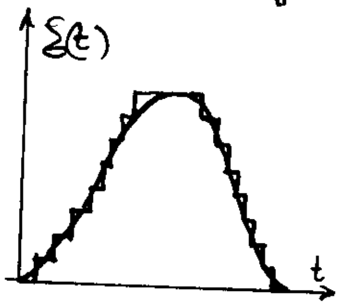


Terms related to the sudden approximation:



- We have assumed
  $\mathcal{E}(t) = 0$ for $t < 0$
  $\mathcal{E}(t)$ constant $\neq 0$ for $t > 0$
  This is the sudden approximation
- Let $\mathcal{E}(t) = (1 - e^{-\lambda t})\mathcal{E}$ for $t > 0$
  $\lambda \to 0$ is the adiabatic approximation

$\rightarrow$ Those terms disappear in the adiabatic approximation.



- Use of perturbative result: apply the result for each step.
- Using the adiabatic approximation is a way to preserve the continuity of the phase from a step to the next one.

Time independent perturbative treatment.

$$|\Psi(t)\rangle = a_0|0\rangle + \sum_i a_i|i\rangle + \sum_\alpha a_\alpha|\alpha\rangle$$

$$i\frac{d}{dt}|\Psi(t)\rangle = H|\Psi\rangle$$

$$i\dot{a}_0 = \sum_\alpha V_{0\alpha}\, a_\alpha \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$i\dot{a}_\alpha = \omega_\alpha a_\alpha + V_{0\alpha} a_0 \left(e^{i\omega t} + e^{-i\omega t}\right) + \sum_i V_{i\alpha} a_i \left(e^{i\omega t} + e^{-i\omega t}\right)$$

$$i\dot{a}_i = \omega_i a_i + V_{i\alpha} a_\alpha \left(e^{i\omega t} + e^{-i\omega t}\right)$$

Let $a_0 = b_0^n e^{in\omega t}$ $\quad a_i = b_i^n e^{in\omega t}$ $\quad a_\alpha = b_\alpha^n e^{in\omega t}$

$$\sum_n e^{in\omega t}\left(-i\dot{b}_0^n + n\omega b_0^n + \sum_\alpha V_{0\alpha}\left(b_\alpha^{n-1} + b_\alpha^{n+1}\right)\right) = 0$$

$$\sum_n e^{in\omega t}\left(-i\dot{b}_\alpha^n + (\omega_\alpha + n\omega) b_\alpha^n + V_{0\alpha}\left(b_0^{n-1} + b_0^{n+1}\right) + \sum_i V_{i\alpha}\left(b_i^{n-1} + b_i^{n+1}\right)\right) = 0$$

$$\sum_n e^{in\omega t}\left(-i\dot{b}_i^n + (\omega_i + n\omega) b_i^n + \sum_\alpha V_{i\alpha}\left(b_\alpha^{n-1} + b_\alpha^{n+1}\right)\right) = 0$$

each term of $\sum_n = 0$: Floquet représentation

Formally $\omega_\alpha + n\omega$ is the energy of the dressed state $|\alpha\rangle|n\rangle$

For strong intensity Floquet representation equivalent to dressed atom picture.

Perturbation theory at lowest nonvanishing order

$$P = 2\pi \sum_k \left[\sum_{\substack{\alpha \\ i \\ \beta}} \frac{V_{0\alpha} V_{\alpha i} V_{i\beta} V_{\beta k}}{(\omega - \omega_\alpha)(2\omega - \omega_i)(3\omega - \omega_\beta)} \right]^2$$

$\omega_k = 4\omega$

summation over all possible final state.

summation over all intermediate states

$V \propto \mathcal{E}$ $\quad P \propto \mathcal{E}^{2n}$ $\quad P = \sigma^{(n)} I^n$

number of photons absorbed

generalised cross section

light intensity

$I = I_M f(t)\, g(\vec{r})$ $\quad \mathrm{Max}(f(t)) = 1$ $\quad \mathrm{Max}(g(r)) = 1$

$$\frac{d}{dt}|a_0|^2 = -\sigma^{(n)} I_M^n f^n g^n |a_0|^2$$

$$|a_0|^2 = \exp\left[-\sigma^{(n)} I_M^n g^n \int dt\, f^n(t)\right] = \exp -\left(\frac{g I_M}{I_s}\right)^n$$

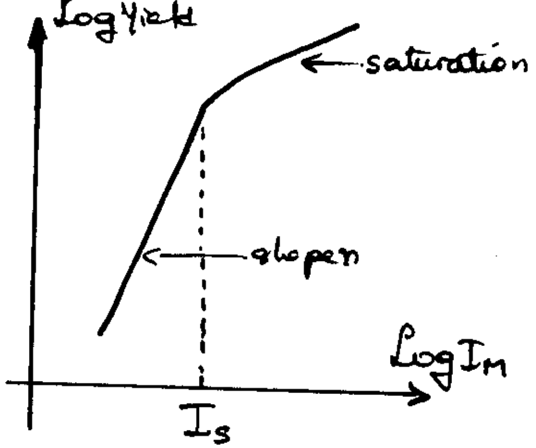
Saturation intensity $\sigma^{(n)} I_s^n \int dt\, f^n(t) = 1$

Total number of ions $= \int d^3\vec{r} \left[1 - \exp -\left(\frac{g I_M}{I_s}\right)^n\right] \times \rho$

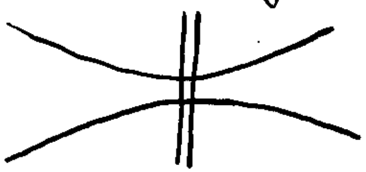
Weak field $\rightarrow \left(\frac{I_M}{I_s}\right)^n \int d^3 r\, \rho\, g^n$

Strong field $\rightarrow \rho \times$ volume where $I_M \gg I_s$

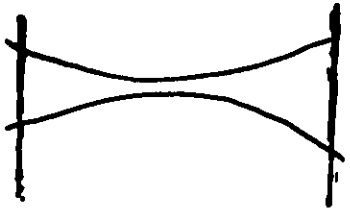
Ion yield



Saturation regime



$N_{ion} \propto \mathrm{Log}\, I$



spherical lens $N_{ion} \propto I^{3/2}$

cylindrical lens $N_{ion} \propto I^{2}$

Measurement of ionisation cross sections

Light intensity $I_M\, g(\vec{r})\, f(t)$ Max(g) = Max(f) =

Number of ions $N = \rho \int d^3\vec{r} \left[1 - \exp\left(-\sigma^{(n)} I_M^n g^n \int dt\, f^n(t)\right)\right]$

Weak field $N = \rho\, \sigma^{(n)} I_M^n \int d^3\vec{r}\, g^n(\vec{r}) \int f^n(t)\, dt$

Measure ρ, I_M, g, f

Strong field Measure $f(t) \rightarrow \mathcal{I}_n = \int dt\, f^n(t)$

Define I_s $\sigma^{(n)} I_s^n \mathcal{I}_n = 1$

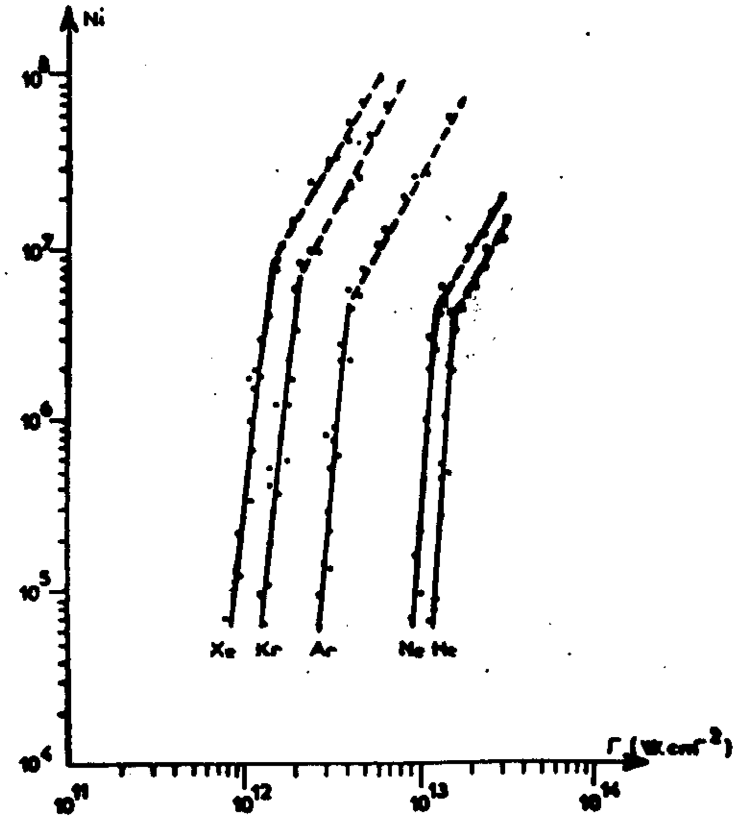
$\rightarrow N = \rho \int d^3\vec{r} \left[1 - \exp\left(-\left(\frac{I_M}{I_s}\right)^n g^n\right)\right]$

Measure $g(\vec{r})$ Calculate N

Fit Log N vs Log I with exp. points $\rightarrow I_s \rightarrow$

avoids measurement of ρ

absolute measurement of N.



Variation of number of multiphoton ions created asfunction of intensity Γ in focusing region; λ = 1.06 μ; dots represent experimental points.

P. Agostini G. Barjot G. Mainfray C. Manus J. Thiebault.
IEEE J. Q.E. 6 782 (70)

Coherence effects

Space and time-dependent intensity

$$I = I_M f(t)\, g(\vec{r}) \quad \mathrm{Max} f(t) = \mathrm{Max}\, g(\vec{r}) = 1$$

Ion yield in perturbative regime

$$N = \rho\, \sigma^{(n)} \int d^3\vec{r}\, g^n(r) \int dt\, f^n(t)\, I_M^n$$

atomic density; cross section; depend on focusing; depend on coherence properties

Energy per pulse $I_M \int g(r)\, d^3r \int f(t)\, dt$

In a quantum description of the field

$$\int dt\, f^{(n)}(t)\, I_M^n = \langle N^n \rangle \qquad N \text{ number of photons}$$

Field state described by a density matrix $\sum_{M,N} P_{MN} |M\rangle\langle N|$

$$\langle N^n \rangle = \sum_N P_{NN} N^n$$

Two opposite cases:

Coherent (Glauber) state $P^G_{NN} = e^{-\langle N \rangle} \dfrac{\langle N \rangle^N}{N!}$

Chaotic (Poisson) state $P^P_{NN} = \dfrac{\langle N \rangle^N}{(1+\langle N \rangle)^{N+1}}$

Coherent state $\langle N^n \rangle = \langle N \rangle^n$

Chaotic state $\langle N^n \rangle = n!\, \langle N \rangle^n$

Mono mode laser $\rightarrow$ coherent state

Multi mode laser $\rightarrow$ chaotic state

Ionisation of Xenon   Nd:YAG laser

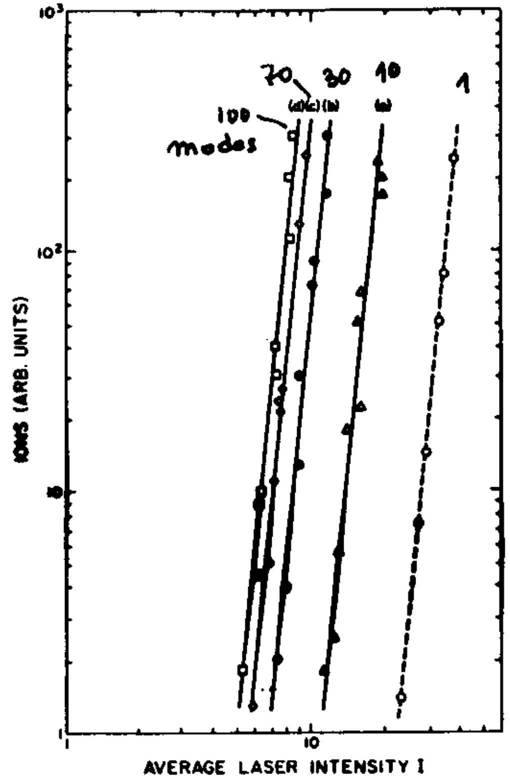


FIG. 3. Log-log plot of the variation of the number of ions in 11-photon ionization of Xe with a Nd-glass laser as a function of the average laser intensity I (arbitrary units) when the laser operates in (a) one spectral line (10 modes), (b) three lines (30 modes), (c) seven lines (70 modes), and (d) ten lines (100 modes). Dashed curve corresponds to a single-mode laser (Lecompte *et al.*, 1975).

Lecompte, Mainfray, Manus Sanchez
PRA 11 1009 (1975)

Calculation of multiphoton ionisation cross sections

weak field

$$P_o = \sum_{\substack{\mathcal{E} \\ \text{final states}}} M_{\mathcal{E}}^2 \qquad M_{\mathcal{E}} = \sum_{\substack{\alpha, j \\ \text{intermediate states}}} \frac{V_{o\alpha} V_{\alpha j} V_{j\mathcal{E}}}{(E_o+\hbar\omega-E_\alpha)(E_o+2\hbar\omega-E_j)}$$

$$\mathcal{E} = E_o + 3\hbar\omega$$

Approximate methods.

- Truncated summations.
- Babb and Gold method.
- Let us define an average energy $\langle E\rangle$ for excited states
- Replace E_α's and E_j's by $\langle E\rangle$

then $M_{\mathcal{E}} = \dfrac{\langle 0|V^3|\mathcal{E}\rangle}{(E_o+\hbar\omega-\langle E\rangle)(E_o+2\hbar\omega-\langle E\rangle)}$

- The latter method for highly excited states, while low lying are introduced explicitly -

Accurate methods

Green functions

$$M_{\mathcal{E}} = \langle 0|V\sum_\alpha \frac{|\alpha\rangle\langle\alpha|}{E_\alpha-E_o-\hbar\omega} V \sum_j \frac{|j\rangle\langle j|}{E_j-E_o-2\hbar\omega} V|\mathcal{E}\rangle$$

$$M_{\mathcal{E}} = \langle 0|V\, G(E_o+\hbar\omega)\, V\, G(E_o+2\hbar\omega)\, V|\mathcal{E}\rangle$$

Dalgarno method

$$M_{\mathcal{E}} = \langle \mathcal{E}|V \underbrace{\sum_j \frac{|j\rangle\langle j|}{E_j-E_o-2\hbar\omega} V \underbrace{\sum_\alpha \frac{|\alpha\rangle\langle\alpha|}{E_\alpha-E_o-\hbar\omega} V|0\rangle}_{|\rho_1\rangle}}_{|\rho_2\rangle}$$

$$(H-E_o-\hbar\omega)|\rho_1\rangle = V|0\rangle$$

$$(H-E_o-2\hbar\omega)\rho_2 = V|\rho_1\rangle$$

Solving step by step a hierarchy of inhomogeneous differential equations

Calculation of multiphoton ionisation crossections

any intensity

Expansion on finite basis

Slater type - Sturmian -

Tractable and accurate in the "single electron atom" approximation

alkalis and hydrogen.

noble gases and alkaline earths (MQDT).

Time dependent calculations.

Time dependent Hartree Fock.

Close coupling calculations.

splitts the space in 3-ranges

$0 < r < r_1$ core effects dominate

$r_1 < r < r_2$ electron-light interaction dominant

$r_2 < r$ Coulomb interaction dominant

Solving in each range - matching on the border -

Keldysh model.

"transition probability" between

atomic ground state and Volkov state

Multiphoton ionisation of hydrogen ground state

Perturbation at lowest non vanishing order

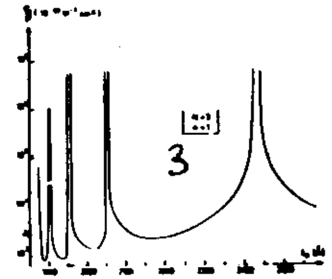


FIG. 2. Dispersion rate σ_n/I^n for the three-photon ionization of H in ground state versus the wavelength λ_p of the incident light.

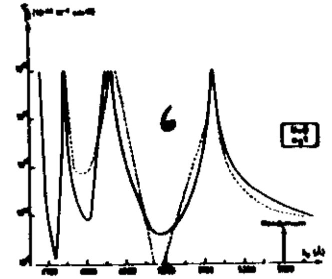


FIG. 5. Dispersion rate σ_n/I^n for six-photon ionization of H in ground state. The value λ_p = 5300 Å corresponds to the second-harmonic of the neodymium laser light. Presently calculated—solid lines; results obtained by Bebb and Gold (Ref. 12)—dashed lines.

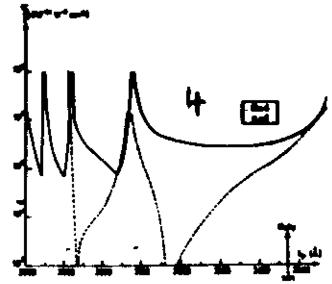


FIG. 3. Dispersion rate σ_n/I^3 for four-photon ionization of H in ground state. The value λ_p = 3471 Å corresponds to the second harmonic of the ruby laser light. Presently calculated—solid lines; results obtained by Bebb and Gold (Ref. 12)—dashed lines.

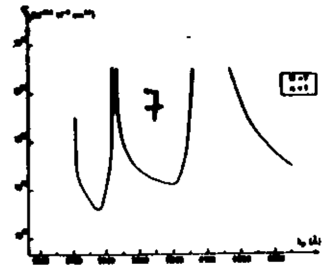


FIG. 6. Dispersion rate σ_n/I^n for seven-photon ionization of H in ground state versus the wavelength λ_p of the incident light.

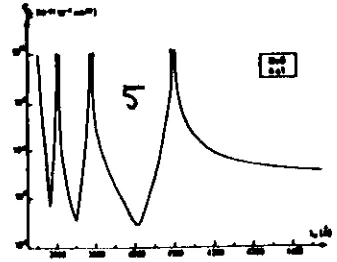


FIG. 4. Dispersion rate σ_n/I^n for five-photon ionization of H in ground state versus the wavelength λ_p of the incident light.

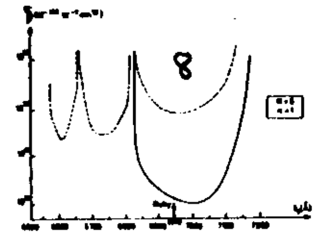
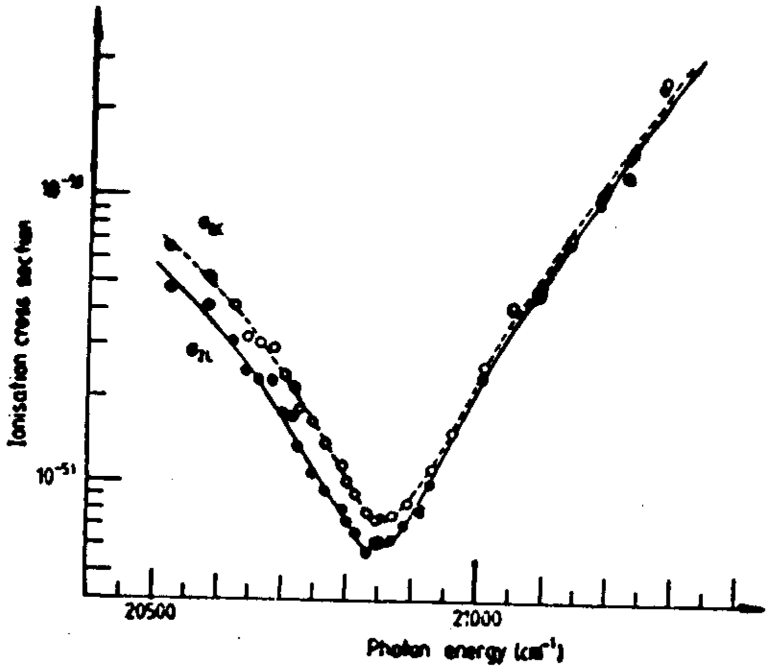


FIG. 7. Dispersion rate σ_n/I^n for eight-photon ionization of H in ground state. The value λ_p = 6943 Å corresponds to the wavelength of the ruby laser light. Presently calculated—solid lines; results obtained by Bebb and Gold (Ref. 12)—dashed lines.

Gontier Trahin (1968)



Two-photon ionisation cross sections of Cs atoms

D. Normand J. Morellec
J Phys B 14 L401 (81)

2-photon ionisation of Cs ground state

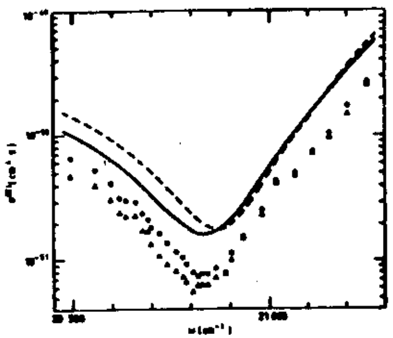


Figure 2. Two-photon ionisation cross sections of the Cs ground state as a function of the photon energy ω for linearly polarised light: ——, this work; Δ, experimental data (Normand and Morellec 1981). For circularly polarised light: - - - -, this work; ○, experimental data (Normand and Morellec 1981).

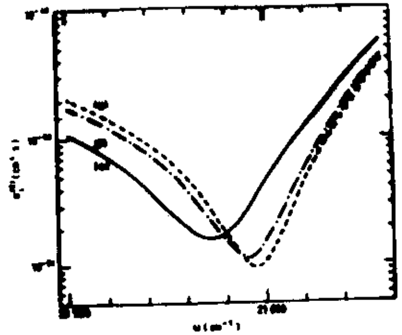


Figure 3. Two-photon ionisation cross section of the Cs ground state by linearly polarised light as a function of the photon energy ω calculated with three levels of approximation: - - - -, (γ), truncated summation limited to [illegible] levels with [illegible]; —·—, (β), truncated summation including all the bound intermediate levels; ——, (α), infinite summation.

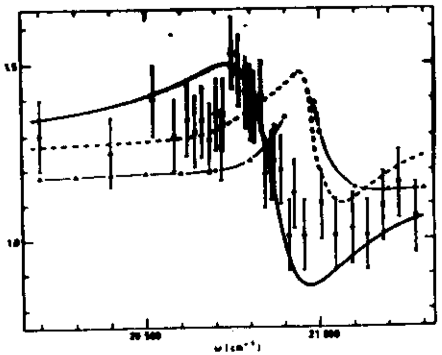


Figure 1. The ratio $R=\sigma_C^{(2)}/\sigma_L^{(2)}$ of the two-photon ionisation cross sections of the Cs ground state for circularly and linearly polarised light as a function of the photon energy ω. ⌀, experimental data (Normand and Morellec 1981). Theoretical predictions: ——, this work; - - - -, Geltman (1980); -Δ-Δ, Teague et al (1976).

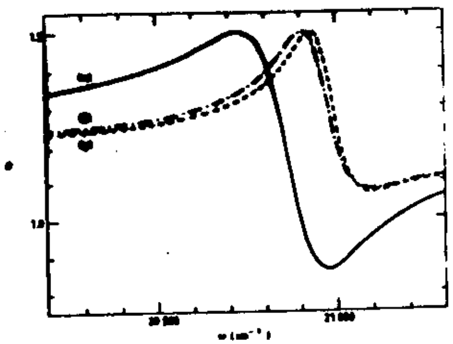


Figure 4. The ratio $R=\sigma_C^{(2)}/\sigma_L^{(2)}$ of the two-photon ionisation cross sections of the Cs ground state for circularly and linearly polarised light as a function of the photon energy ω calculated with three levels of approximation: - - - -, (γ); —·—, (β) and ——, (α).

Normand-Morellec (1981)
Geltman 1980
Aymar Crance 1982

Resonant multiphoton ionisation

$$\underline{P} = 2\pi \sum_k \left| \sum_{\alpha, i, \beta} \frac{V_{0\alpha} V_{\alpha i} V_{i\beta} V_{\beta k}}{(\omega - \omega_\alpha)(2\omega - \omega_i)(3\omega - \omega_\beta)} \right|^2$$

energy denominators may be small

Basic assumption of the previous treatment

$$|\langle\psi|0\rangle| \gg |\langle\psi|\alpha\rangle|, \; |\langle\psi|i\rangle|$$

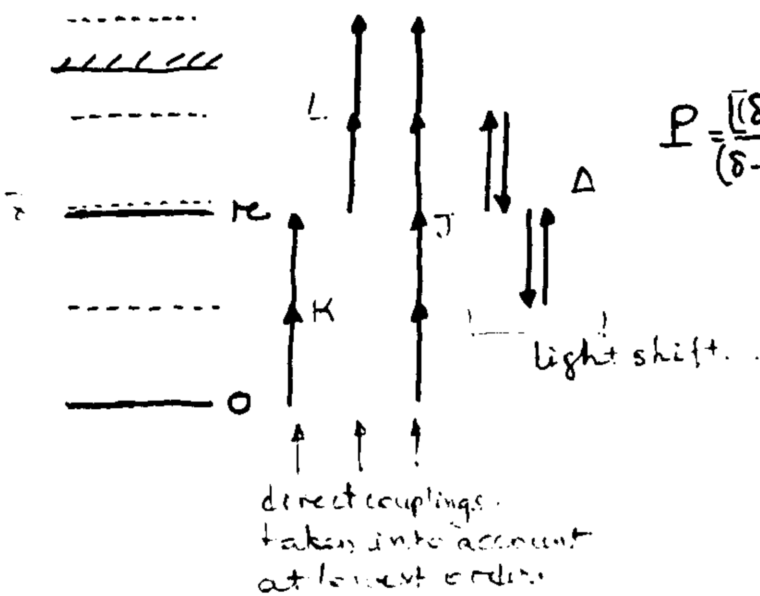
A resonance with $|r\rangle$ means $\omega_r = \omega \times$ integer

$|\langle r|\psi\rangle|$ large.

→ Look for the evolution of

$$(|0\rangle\langle 0| + |r\rangle\langle r|)|\psi\rangle = P|\psi\rangle$$

→ effective operator



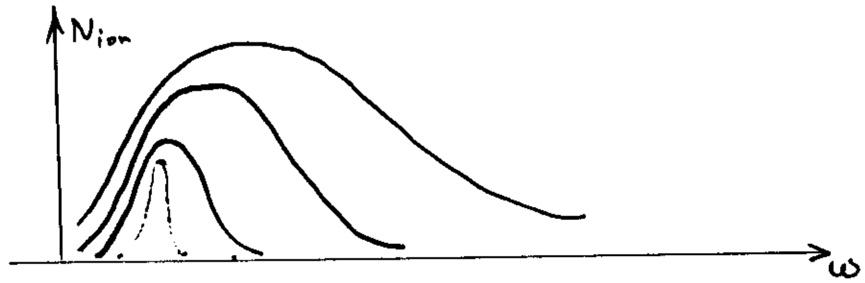
$$\underline{P} = \frac{[(\delta - \Delta)J + KL]^2}{(\delta - \Delta)^2 + L^4} + J'$$

Resonant multiphoton ionisation

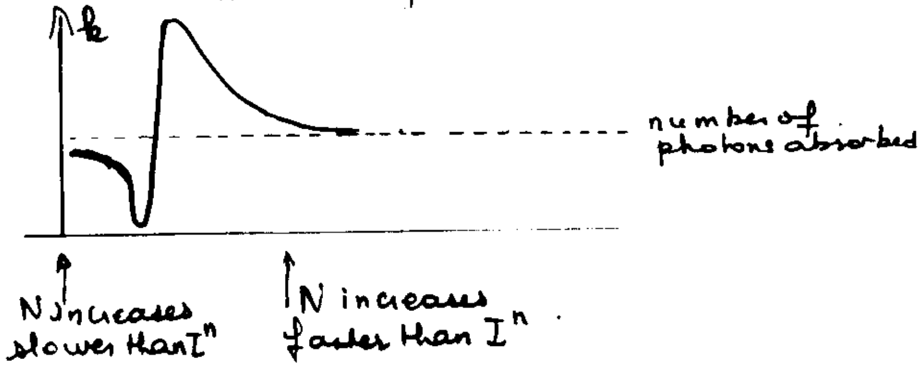
$$\underline{P} = J_0 + \frac{[(\delta - \Delta)J + KL]^2}{(\delta - \Delta)^2 + L^4}$$

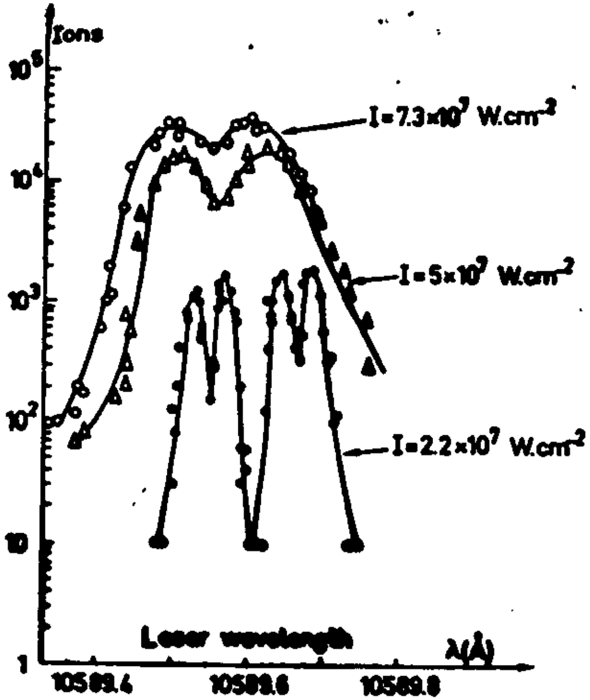
$\propto I$ $\quad I^{2P}$

When I increases, resonances shift and broaden.



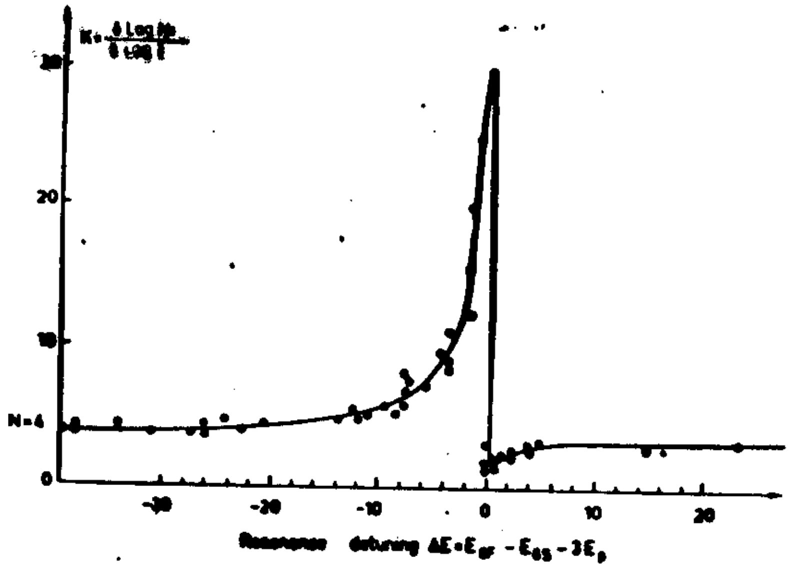
This is reflected by the effective order of non linearity $\frac{d \log N}{d \log I}$ = slope of the curve $\log N$ vs $\log I$





Resonance profiles in the four-photon ionization of Cs for three values of laser intensity I. Hyperfine structure of the ground state and the fine structure of the 6F resonant state are well resolved for the lowest intensity value.

G. Petite J. Morelle & D. Normand
J. Phys. 40 115 (79).

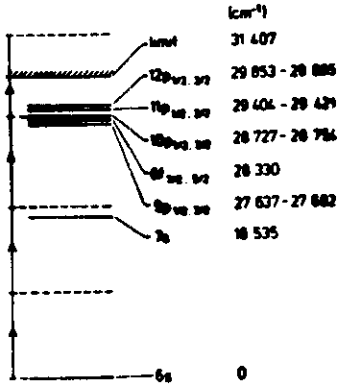


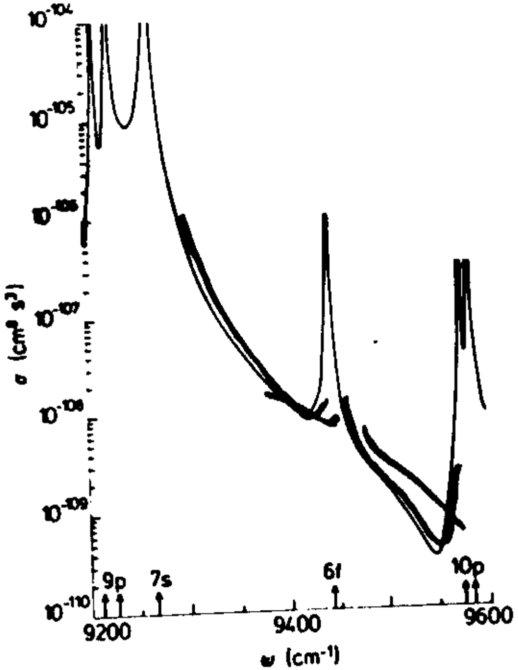
Variation of the effective order of nonlinearity K as a function of the resonance detuning ΔE in the four-photon ionization of Cs.

J. Morelle D. Normand
G. Petite Phys. Rev. A 14 300 (76)

ordre effectif de non linéarité

4 photon ionisation of Cezium.

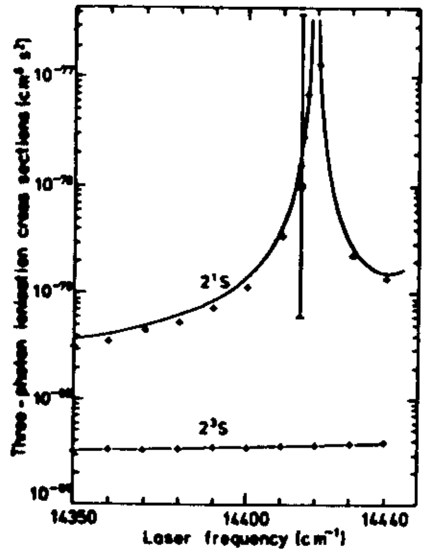


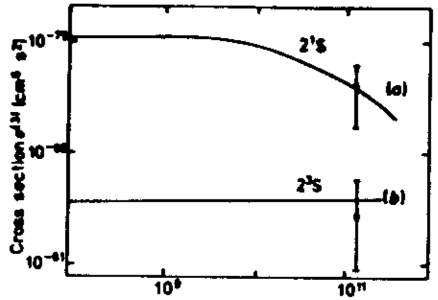


10 GWcm⁻²
100 GWcm⁻²

Figure 2. Four-photon ionisation cross sections for various light inte
results (up to 0·1 GW cm², ———; 10 GW cm⁻², — — — —; 100 GW

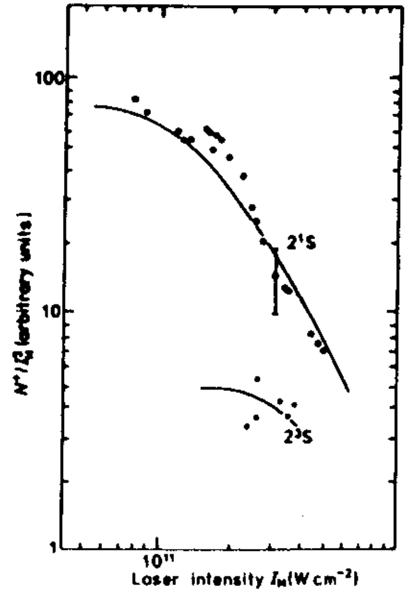
Crance Aymar JPhys B13 4129 (1980).





ω = 14398,5 cm⁻¹

3-photon ionisation of metastable Helium off resonant by 40.5 cm⁻¹



Lompré Mainfray Mathieu Watel Aymar Crance JPhys B13 1799 (1980)