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WINTER COLLEGE ON HIGH RESOLUTION SPECTROSCOPY

(8 January - 2 February 1990)

MULTIPHOTON IONISATION OF ATOMS

IV Electron Energy Spectrum

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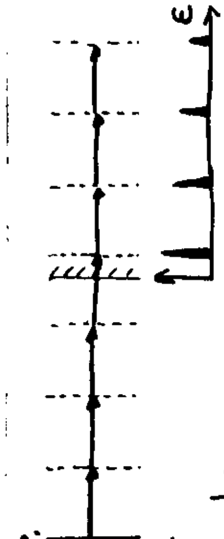


Electron energy spectrum.

$$\varepsilon = (n+k)\hbar\omega - |E_0|$$

↑ ionisation potential

first peak $\propto I^n$

second peak $\propto I^{n+1}$

⋮

Probability to reach

first peak : $\left[\sum \frac{V_{0\alpha} V_{\alpha i} V_{i\beta} V_{\beta k}}{(\omega-\omega_\alpha)(2\omega-\omega_i)(3\omega-\omega_\beta)}\right]^2$

second peak : $\left[\sum \frac{V_{0\alpha} V_{\alpha i} V_{i\beta} V_{\beta j} V_{jk}}{(\omega-\omega_\alpha)(2\omega-\omega_i)(3\omega-\omega_\beta)(4\omega-\omega_j)}\right]^2$

third peak $\left[\sum \frac{V_{0\alpha} V_{\alpha i} V_{i\beta} V_{\beta j} V_{j\gamma} V_{\gamma l}}{(\omega-\omega_\alpha)(2\omega-\omega_i)(3\omega-\omega_\beta)(4\omega-\omega_j)(5\omega-\omega_\gamma)}\right]^2$

$4\omega-\omega_j$ may vanish

→ replace $\int \frac{1}{4\omega-\omega_j}$ by $P.P.\int \frac{1}{4\omega-\omega_j} + i\,\delta(4\omega-\omega_j)$

Interpretation

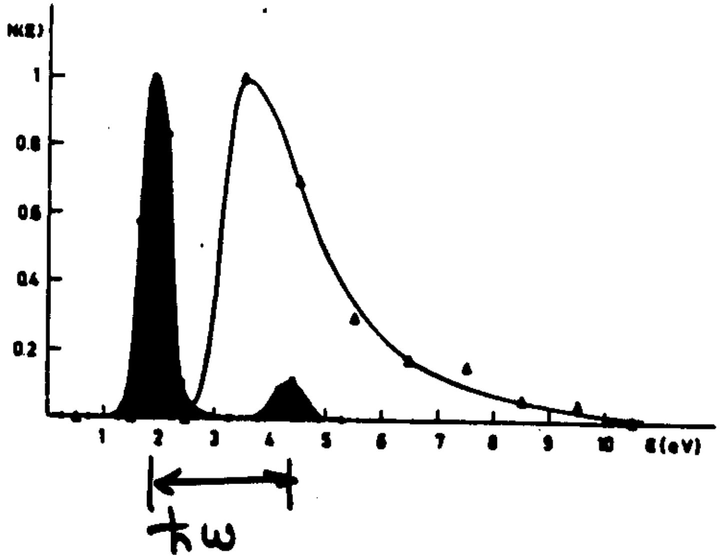
Probability amplitude to absorb 4+1 photons

$M_{05} + i\, M_{04} M_{45}$

M_{05} simultaneous absorption of 5 photons, no resonant step.

$M_{04} M_{45}$ simultaneous absorption of 4 photons.

Immediately followed by absorption of 1 photon.



532 nm

P. Agostini F. Fabre G. Mainfray G Petite N.K. Rahman Phys Rev Lett. 42 1127 (79).

Xenon
1.06 μ

Kruit et al 1981

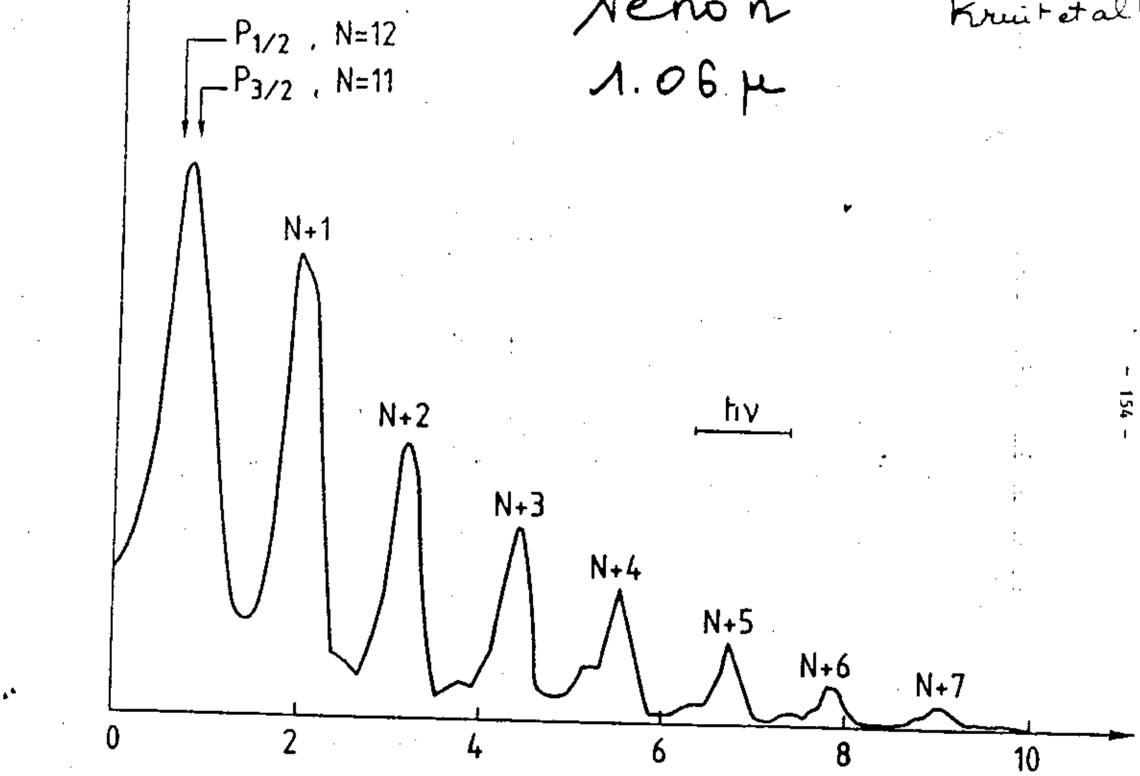
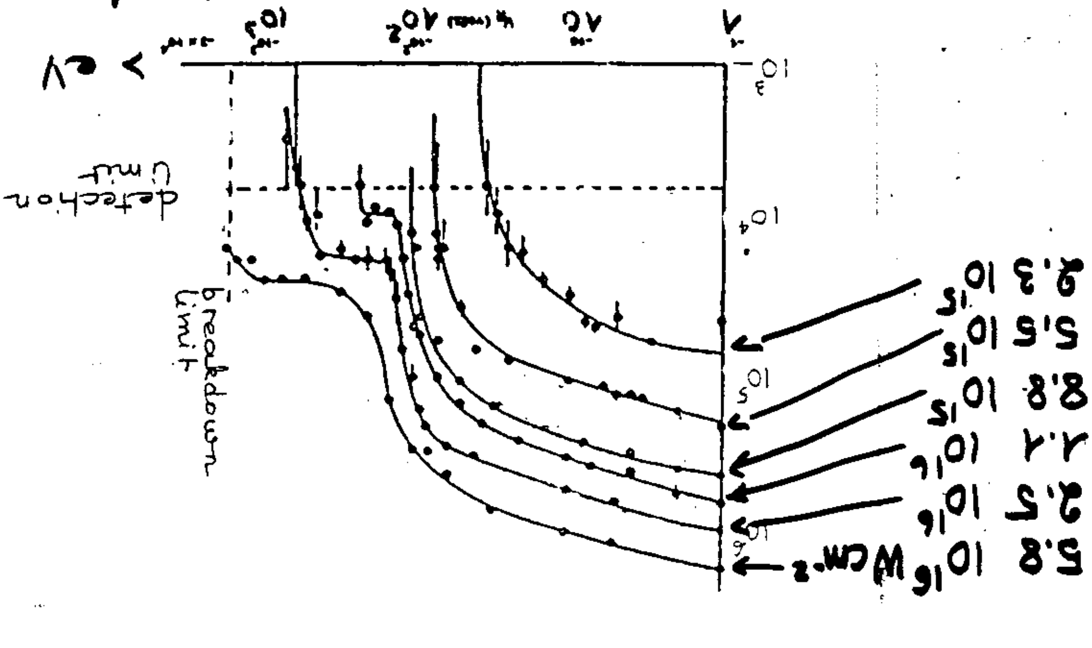


Figure 44 - Spectre énergétique des photoélectrons provenant de l'ionisation du Xénon pour λ = 1,06 μm. (Kruit et al. / 27 /).

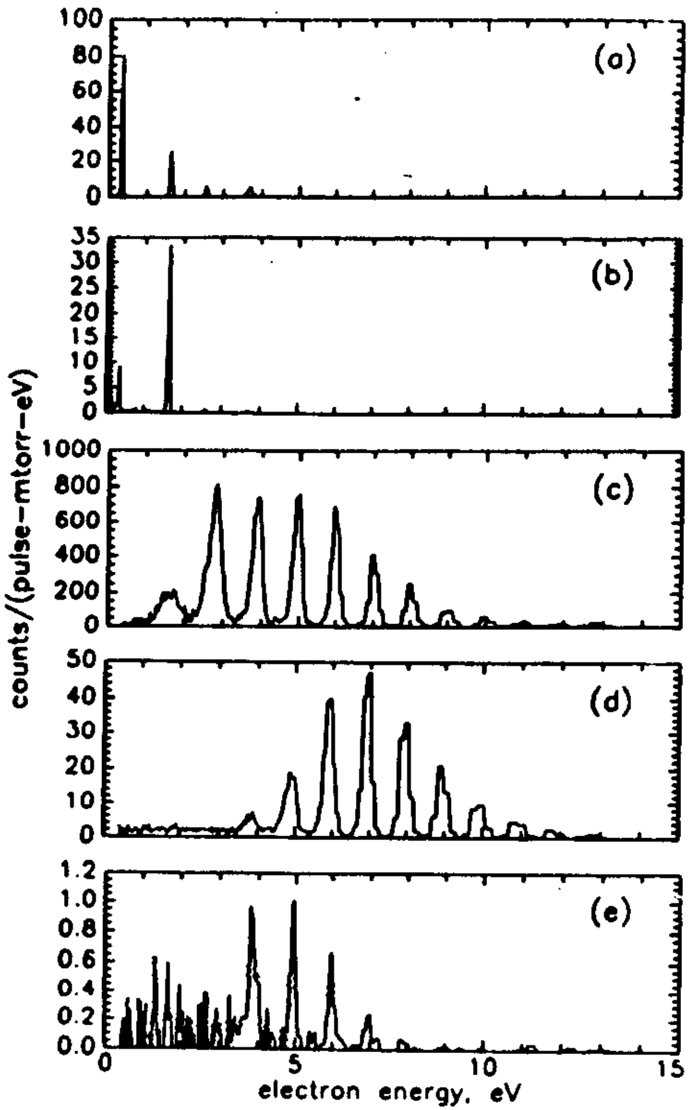
Boreham et al 1981

Retarding potential



58

59



P. H. Bucksbaum M. Bashkansky R.R. Freeman T.J. McIlrath
Phys Rev Lett. 56 2590 (86)

Angular distribution

Removing an electron of momentum ℓ by ~~absorption~~ of n photons provides an electron of momentum $\ell+n$ for circular polarization
$\ell+n$, or $\ell+n-2$ or $\ell+n-4$ ..... for linear polarisation

For each possible momentum in final state

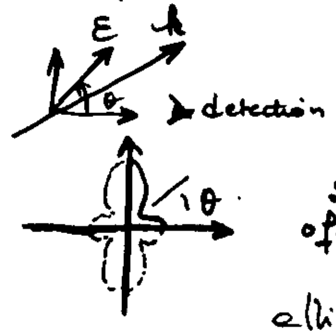
$$M_{o\ell} = \sum_{\substack{\text{interm.}\\ \text{state}}} \langle o | V G V G \ldots \, | \varepsilon \ell \rangle$$

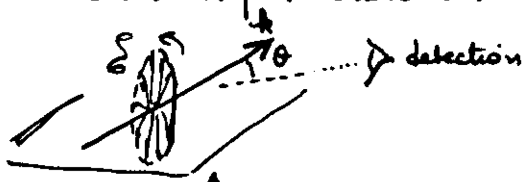
$|\varepsilon \ell\rangle$ stands for $R(r)\, Y_{\ell m}(\theta,\varphi)$ $\quad R(r) \to \sin(kr + \delta_\ell)$

Probability to detect an electron in direction θ, φ.

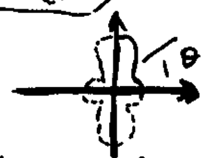
$$P(\theta,\varphi) = \left| \sum_{\ell} M_{o\ell}\, Y_{\ell m}(\theta,\varphi)\, e^{-i\delta_\ell} \right|^2$$

linear polarisation $\qquad$ circular polarisation





z axis of symmetry



elliptic polarization

conserved in π-rotation

Multiphoton ionisation by 1.06μ in elliptic polarisation

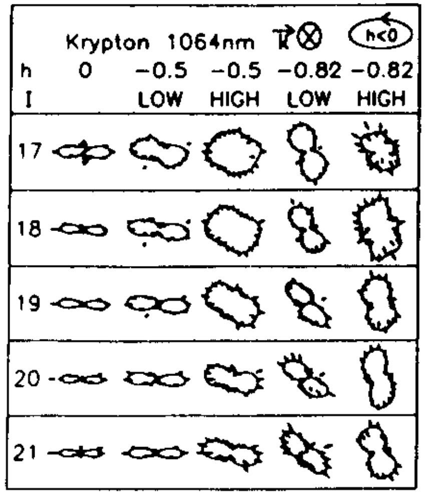


FIG. 4. Azimuthal distributions for various retardations and laser intensities, for krypton photoionized by 1064-nm light. Numbers to the left of each row designate photons absorbed to the $P_{3/2}$ ion final state for each ATI peak. First column: linear polarization, $I_{peak} = 2.5\times10^{13}$ W/cm². Second column: $h = -0.5$, $I_{peak} = 2\times10^{13}$ W/cm². Third column: $h = -0.5$, $I_{peak} = 4\times10^{13}$ W/cm². Fourth column: $h = -0.82$, $I_{peak} = 2\times10^{13}$ W/cm². Fifth column: $h = -0.82$, $I_{peak} = 4\times10^{13}$ W/cm².

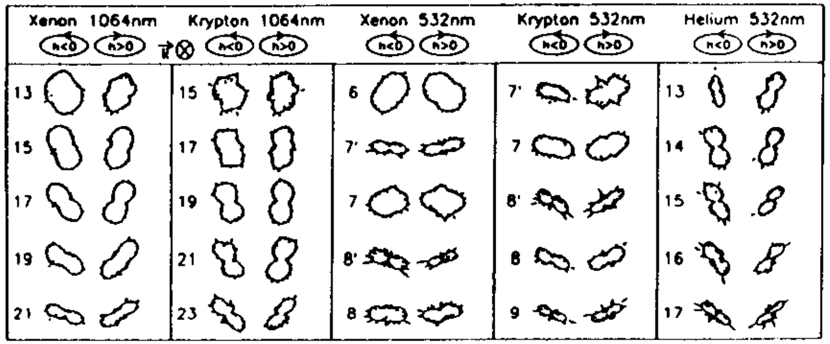


FIG. 3. Comparison between data obtained with positive-helicity light ($h = +0.82$), and data obtained under the same conditions with the helicity reversed ($h = -0.82$). The laser pulse width was 0.10 to 0.12 nsec. Xe 1064 nm: $I_{peak} = 4\times10^{13}$ W/cm²; $P_{1/2}$ and $P_{3/2}$ final states were not resolved, so numbers indicate photons absorbed for the $P_{3/2}$ final state. Kr 1064 nm: $I_{peak} = 4\times10^{13}$ W/cm²; $P_{3/2}$ final states only. Xe 532 nm: $I_{peak} = 1\times10^{13}$ W/cm²; primed numbers designate photons absorbed to the final $P_{1/2}$ state; unprimed numbers designate the $P_{3/2}$ state. Kr 532 nm: $I_{peak} = 1.5\times10^{13}$ W/cm; primes mean the same as for Xe 532 nm. Helium 532 nm: $I_{peak} = 1\times10^{14}$ W/cm².

Bashkansky Bucksbaum Schumacher PRL 60 2458 (88)

Multiphoton ionisation of hydrogen
Angular distribution for various peaks in electron energy spectrum

0,355 μm

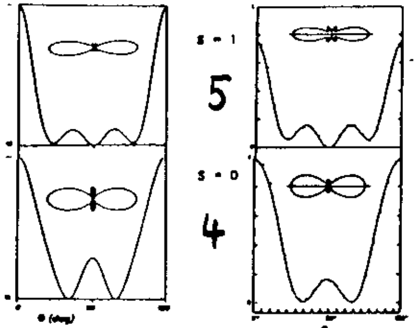


Figure 1. Photoelectron angular distributions at 355nm. S is the number of photons above threshold. Left column: calculated in this work. Right column: experiment [1,6] normalised to highest signal.

0.532 μm

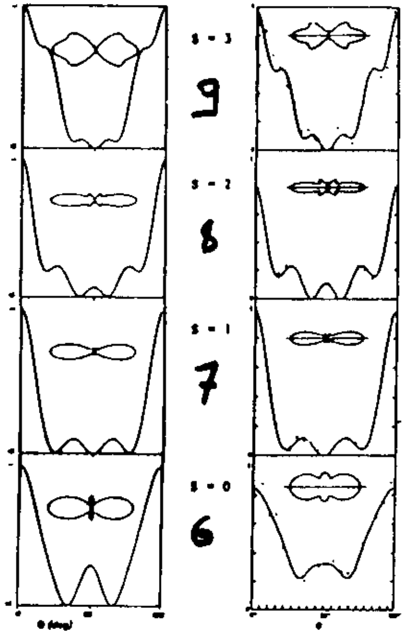


Figure 2. Photoelectron angular distributions at 532nm (notation as in Figure 1).

exp: Feldmann Wolff Wemhöner Welge Z f Phys (1987)
th : Kracke Marxer Broad Briggs Z f Physik (1988)

Electron energy spectrum

development in time

$$\dot{n}_0 = -\gamma n_0 \qquad \gamma = \sum_1^{\infty} P_k$$

$$\dot{n}_k = P_k n_0$$

$n_0(t)$ probability to remain neutral at time t

$n_k(t)$ probability to reach peak k before time t

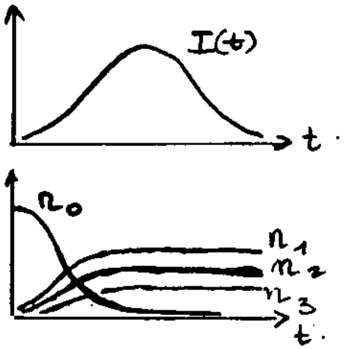
P_k probability per unit time to transit to peak k.

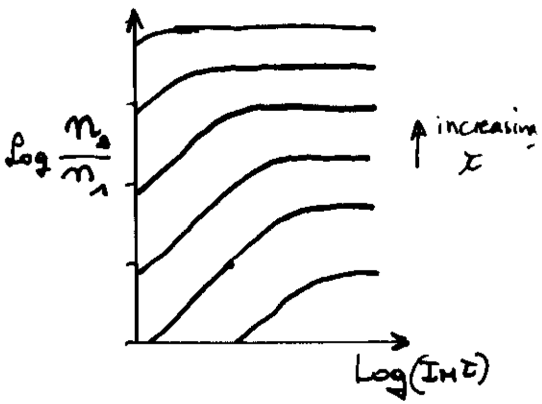
number of electrons in peak k $n_k(t=+\infty)$

$$n_k(t=+\infty) = \int_{-\infty}^{+\infty} P_k(I(t))\, n_0(t)\, dt$$

low intensity $\longrightarrow \alpha\, P_k(I_M)$

saturation intensity and above





Pulse duration determines saturation intensity I_S

For $I > I_S$ peaks remain in the ratio $n_1(I_S) \div n_2(I_S) \div n_3(I_S)$

The only way to increase higher order peaks is to decrease the pulse duration.

4 photon ionisation of Cesium

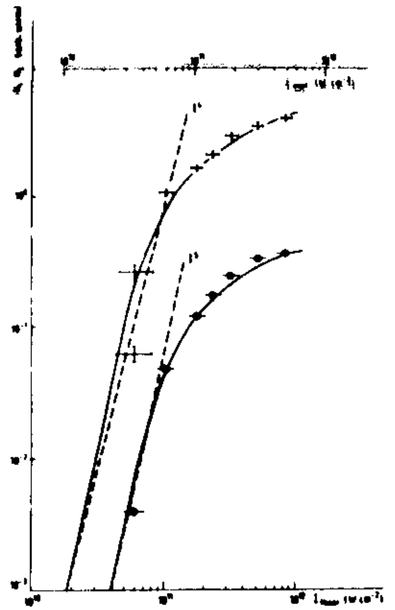


FIG. 11. $N_4(\theta=0)$ and $N_5(\theta=0)$ in arbitrary units as functions of intensity. Crosses and circles are experimental. Solid lines are calculated. Dashed straight lines of slope 4 and 5 are shown for comparison. Note the different experimental (I_{exp}) and theoretical (I_{theor}) intensity scales.

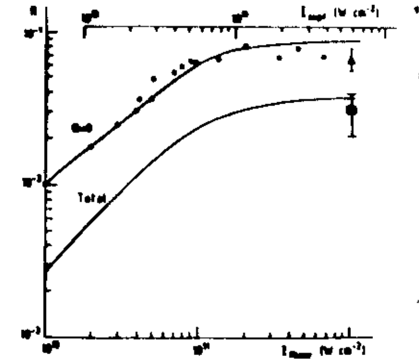


FIG. 12. Ratio N_5/N_4 as a function of intensity. Experimental values (dots) are normalized to the value at 6×10^{11} W cm^{-2} discussed in the text. Solid lines are theoretical. Note again the different intensity scales. Experimental point on the lower curve gives the experimental determination of the total signal ratio resulting from our angular-distribution measurements.

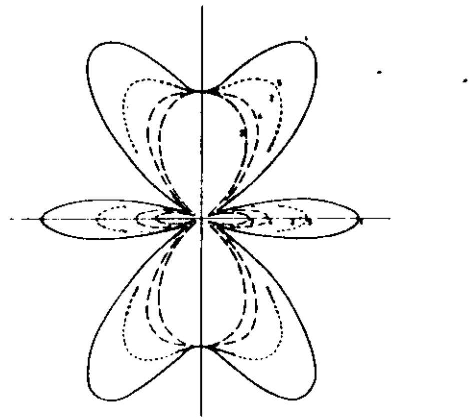


FIG. 2. Angular distributions for the four-photon ionization of cesium for different intensities (in W cm^{-2}): curve 1, 1×10^9; curve 2, 10^{10}; curve 3, 10^{11}; curve 4, 2×10^{11}; curve 5, 3×10^{11}.

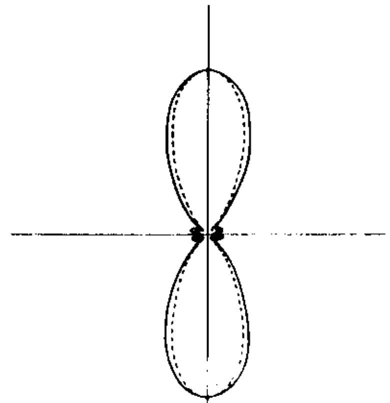
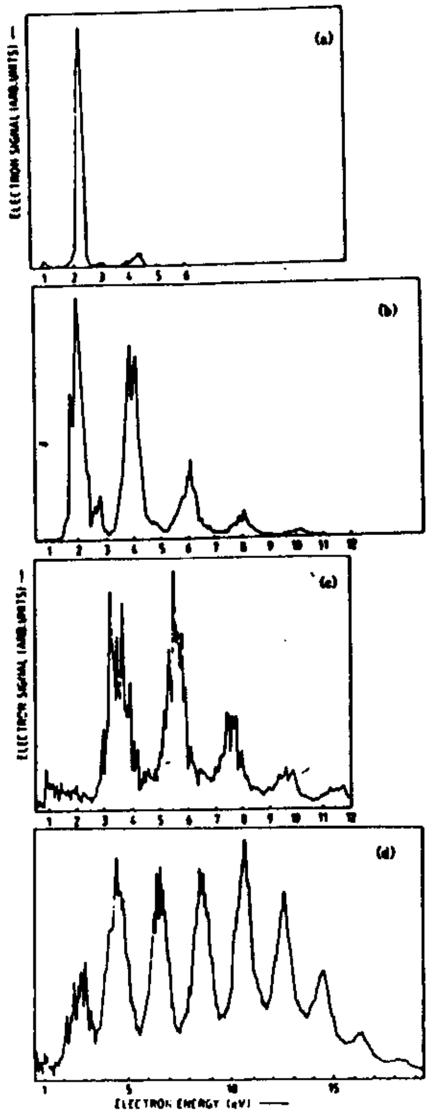


FIG. 3. Angular distributions for the five-photon ionization of cesium for two intensities: 10^9 W cm^{-2} (solid line), 10^{11} W cm^{-2} (dashed line).

Petite Fabre Agostini Crance Aymar PRA 29 2677 (84)

Multiphoton ionisation of Xenon
620 nm 100 fs



11 μJ

24 μJ

29 μJ

35 μJ

Muller, v.d. Heuvell, Agostini, Petite, Antonetti, Franco, Mysyrowicz
PRL 60 565 (88)

Multiphoton ionisation of Xenon 1.06μ.

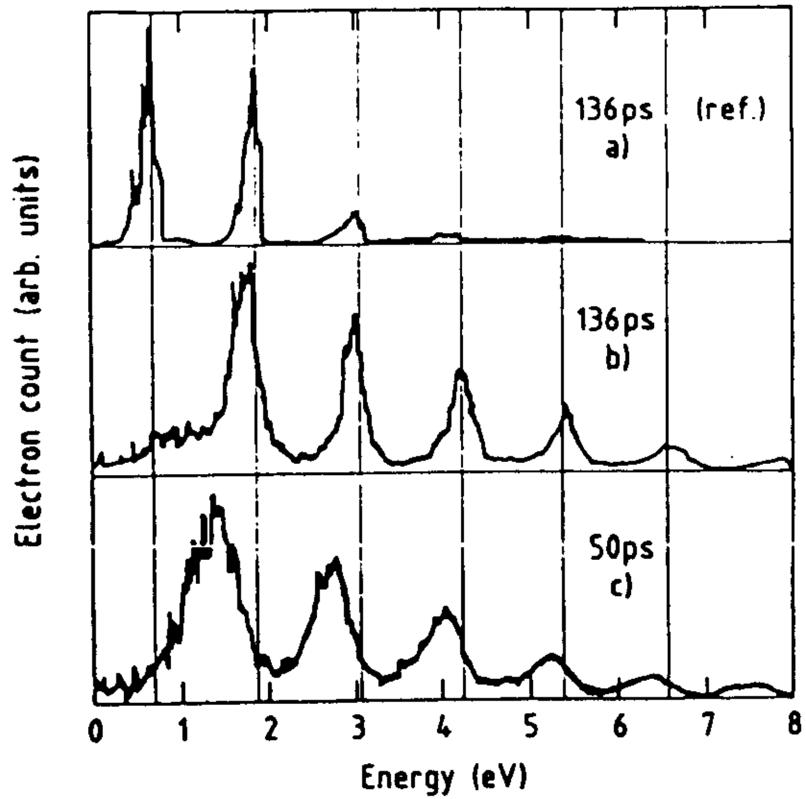


FIG. 2. Electron energy spectra for different laser intensities and pulse durations. (a) reference spectrum, $I = 2.2 \times 10^{12}$ W cm^{-2}; (b) and (c) $I = 7.5 \times 10^{12}$ W cm^{-2}.

Agostini Kupersztych Lompre Petite Yergeau
PRA 36 4111 (87)

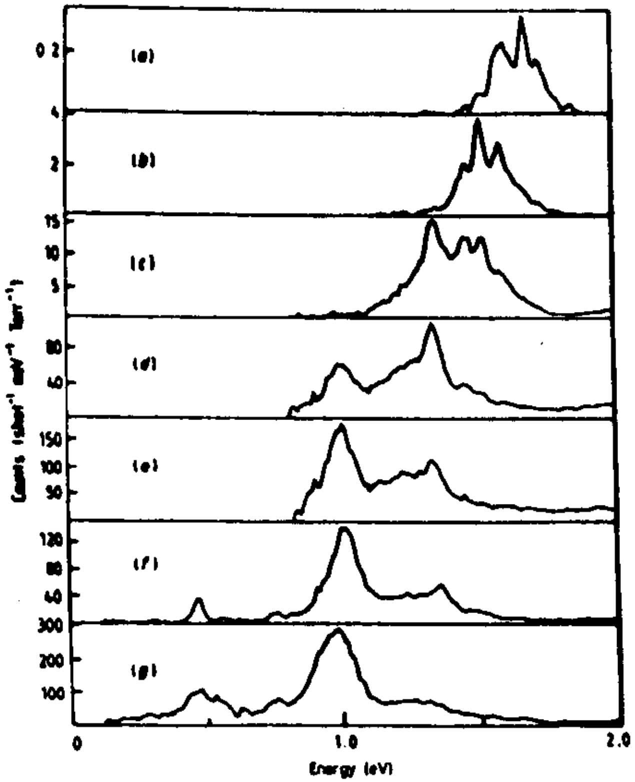
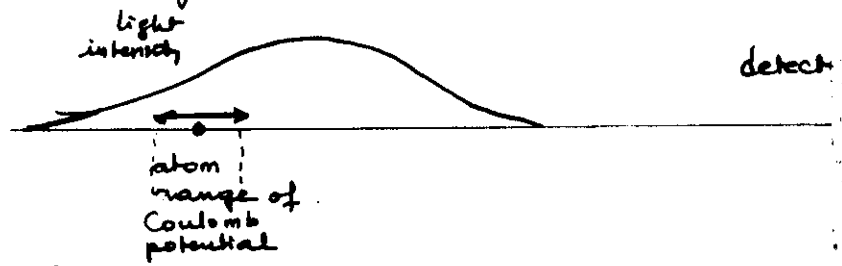


Figure 1. Selected electron energy spectra. The laser intensity increases from 1×10^{13} W cm^{-2} (*a*) to 3×10^{13} W cm^{-2} (*g*).

Agostini et al J Phys B <u>22</u> (1989) 1971

Leaving the interaction volume



A free electron in a light beam has a quiver motion

$m\ddot{\gamma} = q\mathcal{E}\cos\omega t \quad v = \frac{q\mathcal{E}}{m\omega}\sin\omega t \quad \frac{1}{2}mv^2 = \frac{q^2\mathcal{E}^2}{2m\omega^2}\sin^2\omega t$

quiver energy $\frac{q^2\mathcal{E}^2}{4m\omega^2}$

The energy of an electron in a light beam is at least $\frac{q^2\mathcal{E}^2}{4m\omega^2}$

$\vec{f} = q(\vec{\mathcal{E}} + \vec{v} \wedge \vec{\mathcal{B}}) \rightarrow x$

Replace x in $\mathcal{E}$ and $\mathcal{B}$ $\rightarrow$ $\vec{f} = q(\vec{\mathcal{E}} + v \wedge \vec{\mathcal{B}}) \underbrace{- \frac{q^2}{4m\omega^2}\vec{\nabla}\mathcal{E}^2}_{\text{gradient force}}$

when $\mathcal{E}$ is time independent

$\rightarrow$ potential $\frac{q^2\mathcal{E}^2}{4m\omega^2}$

Right after ionisation $E = \frac{1}{2}mv_T^2 + \frac{q^2\mathcal{E}^2(r)}{4m\omega^2}$

The electron leaves the light beam with velocity v_T

the quiver energy decreases to zero $\quad \frac{q^2\mathcal{E}^2}{4m\omega^2}$ lost

The electron gains the work of the gradient force $\frac{q^2\mathcal{E}^2}{4m\omega^2}$ [illegible]

For a "long pulse"

Electron energy on the detector = energy right after ionisat[illegible]

LONG PULSE : $\underset{\text{duration}}{\tau} \gg \underset{\text{waist}}{R} / \underset{\text{translation velocity}}{v_T}$

(conversely for a short pulse, the quiver energy is

Electron energy on the detector =

energy right after ionisation − quiver energy −

Short strong pulse

Quiver energy can be interpreted as a "threshold shift"
A.C. Stark shift -
From perturbation theory at lowest order:
Ground state is a little shifted down δI
High Rydberg states are shifted as the threshold ΔI
An electron is emitted at time t, point $\vec{r} \to I(\vec{r}, t)$
Energy conservation

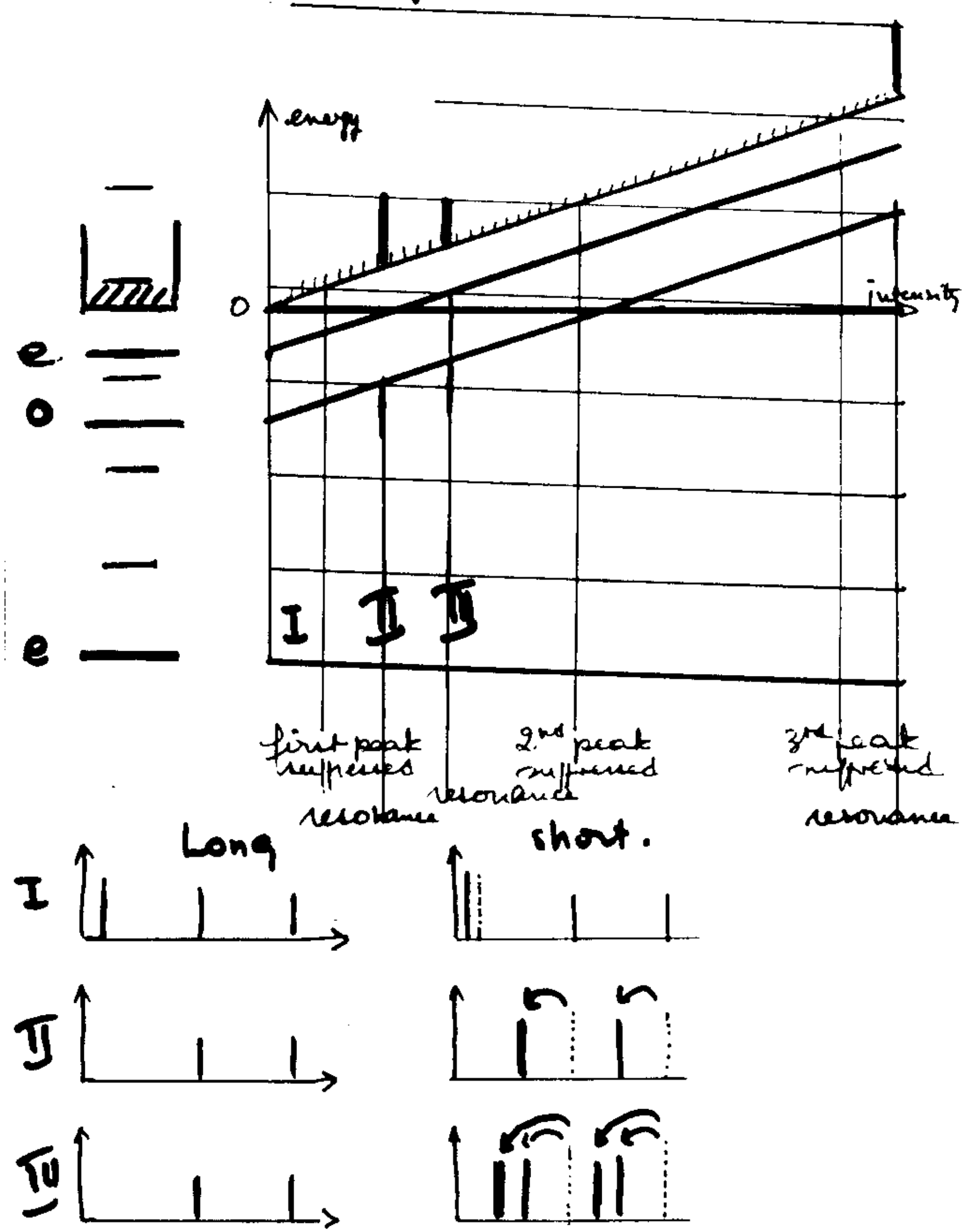
$$E_0 + \delta I + n\hbar\omega = \Delta I + E_T \qquad E_T = \frac{1}{2} m v_T^2$$

$\to$ E_T is a function of $I(\vec{r}, t)$

The electron energy spectrum reflects the ionisation probability as a function of intensity.

$\to$ sub peaks indicate transient resonances.

Strong short pulse



Interpretation of transient resonances

$$E_g + \delta I + 6\hbar\omega = \Delta_q I + \varepsilon = E_r + \Delta_r I + \hbar\omega$$

$$\varepsilon = \frac{6\Delta_r - 5\Delta_q - \delta}{\Delta_r - \delta}\hbar\omega + \frac{E_r(\Delta_q - \delta) + E_g(\Delta_r - \Delta_q)}{\Delta_r - \delta}$$

Assign resonances by varying $\hbar\omega$

ok for large $n\ell$

A puzzle remains: missing peaks - extra peaks

Light shift in Hydrogen.

Complex dilatation method with a finite basis of complex functions.

Eigenstates: low energy : H-states.

high energy : wave packets of continuum + highly excited states.

→ interpretation

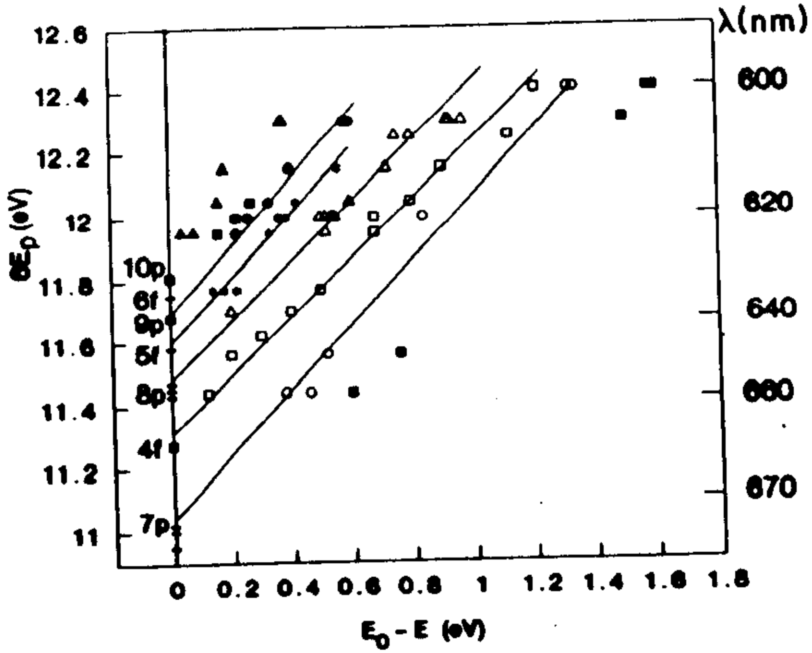
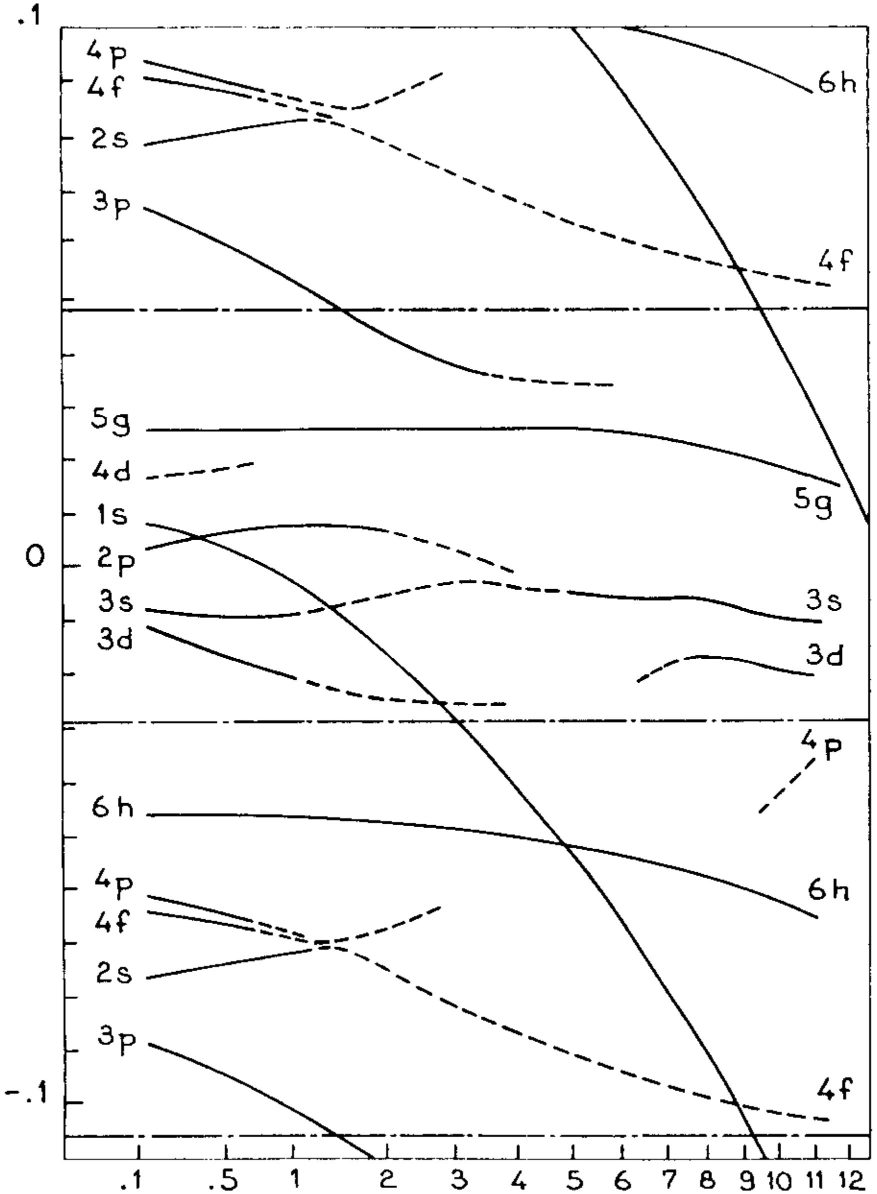


FIG. 2. Plot of $6E_P$ as a function of $E_0 - E$. For clarity, only a few lines have been drawn as guides to the eye. The filled squares below the line corresponding to the $7p$ state may be assigned to the $6p$ but do not extrapolate to the zero-field position of this state.

P. Agostini P. Breger A. L'Huillier H.G. Muller G Petite
PRL 63 2208 (89)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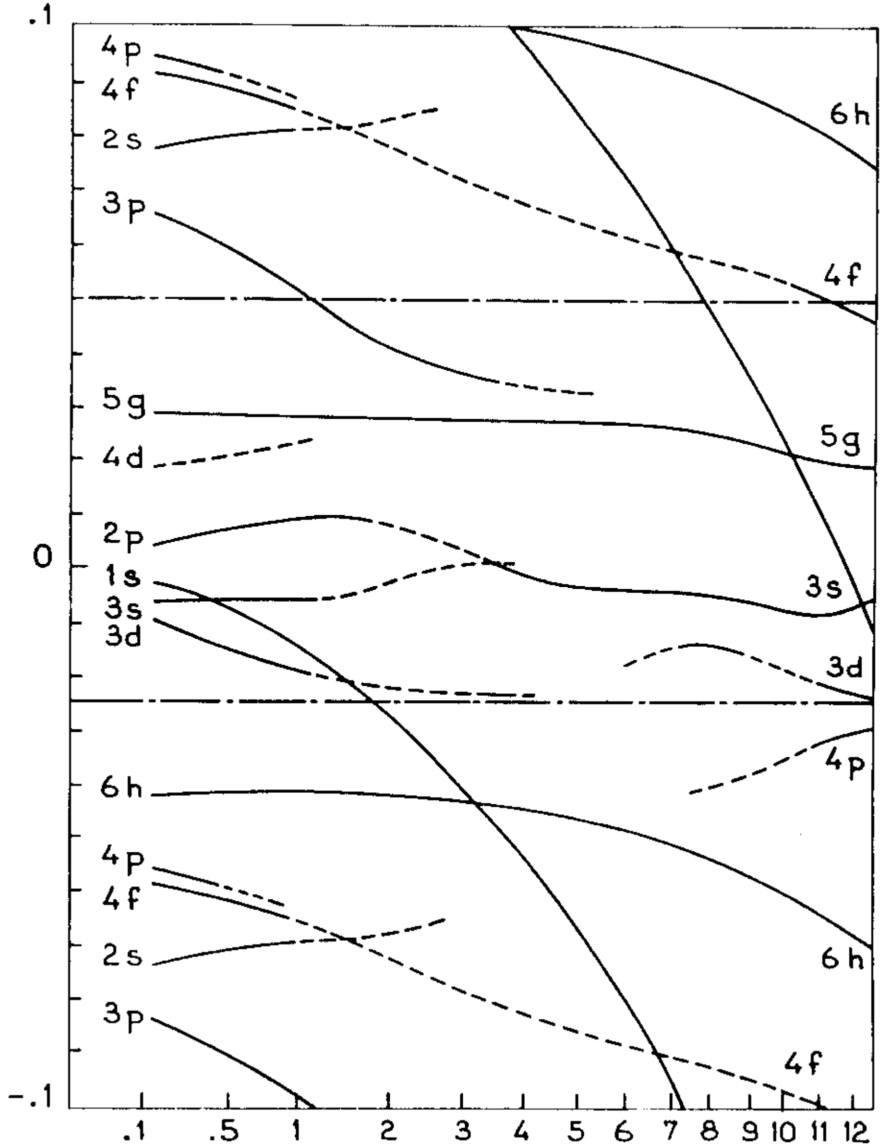
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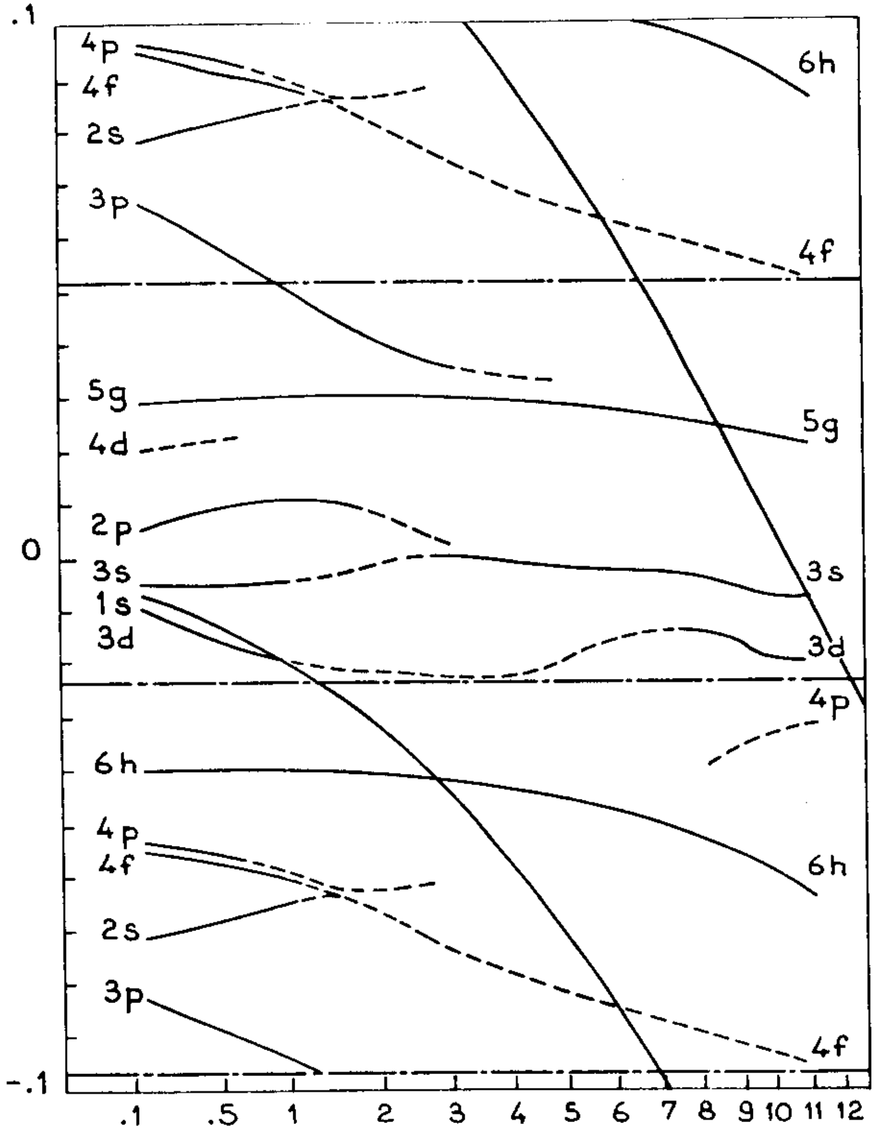

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Fig 1c