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SMR 1302 - 8

WINTER SCHOOL ON LASER SPECTROSCOPY AND APPLICATIONS

19 February - 2 March 2001

COLD COLLISIONS AND CHEMICAL REACTION

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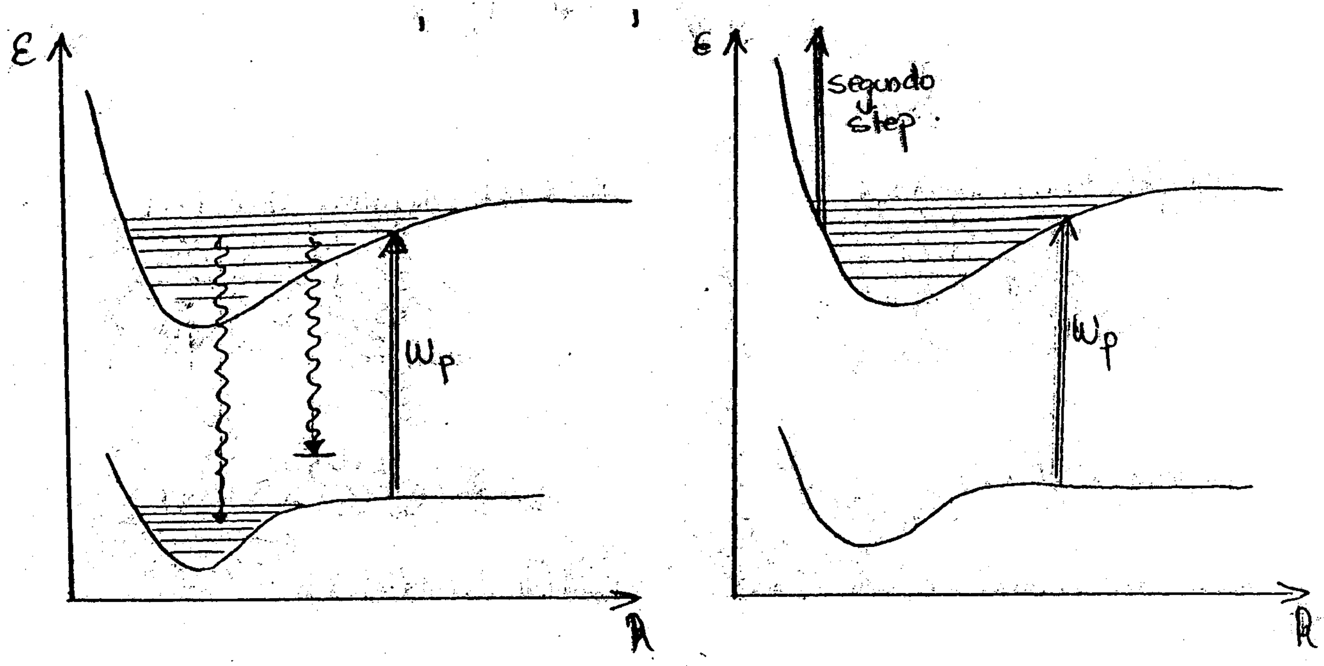
These are preliminary lecture notes, intended only for distribution to participants.

III - Cold Collisions and Chemical Reaction

* Laser can selectively excite bound states.
* Detection through trap loss or ionization
* Use of probe laser

— Photoassociation —



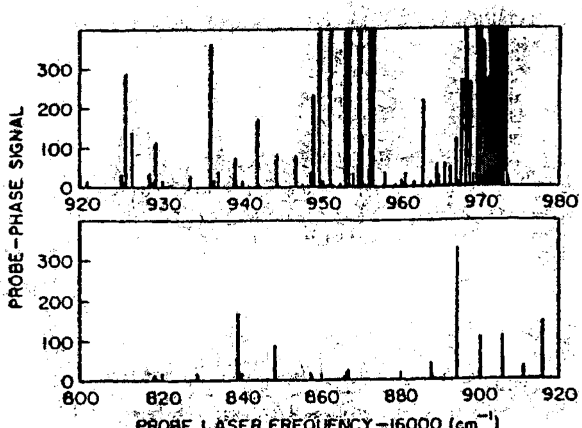


FIG. 57. Photoassociative ionization spectrum of Na collisions from the NIST dark-spot MOT. From Ratliff *et al.* (1994).

Ex: Na by Ionization (NIST)

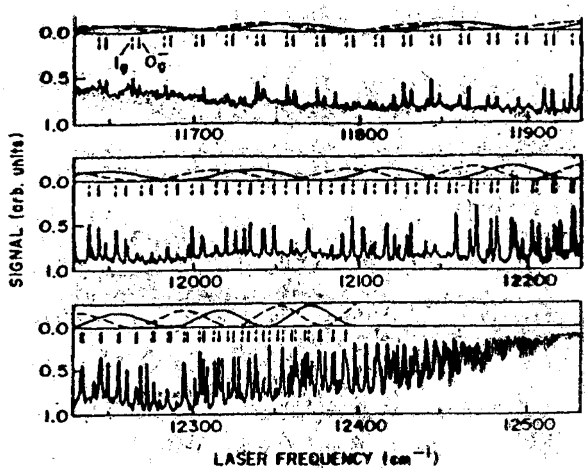


FIG. 61. Photoassociation FORT trap-loss fluorescence spectrum of Rb_2. From Miller et al. (1993b).

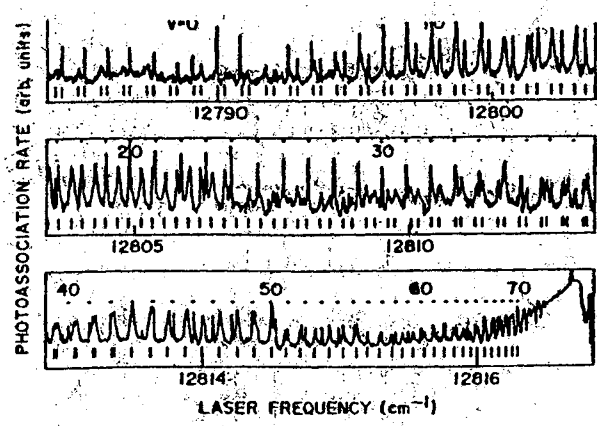


FIG. 62. Cline et al. (1994a, 1994b) separated the trapping and scanning functions with two different lasers: one laser at fixed frequency produced the FORT while a probe laser scanned through the resonances.

* Determination of potential terms

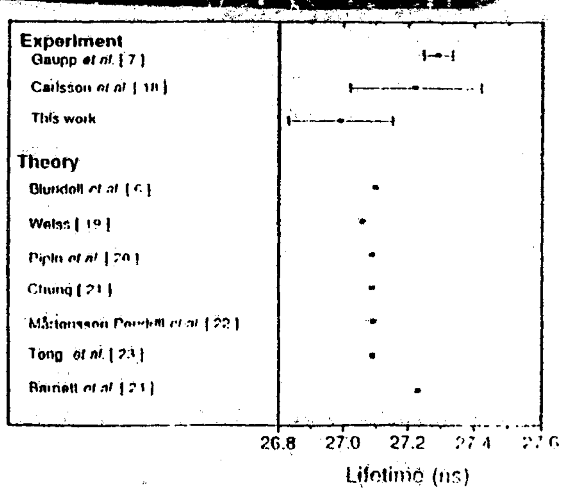
$$V \cong -\frac{C_3}{R^3} - \frac{C_6}{R^6} - \frac{C_8}{R^8}$$

C_3 is related to the excited state lifetime

$$C_3 = \frac{3\hbar}{2\tau}\left(\frac{\lambda}{2\pi}\right)^3$$

⟶ Determination of τ

Ex: Medida para Li



$\tau_{Li} = 26.99 \pm 0.16$ ns

Há medidas para Na, K, Rb, Cs

[Rev. Mod. Phys. 71, 1 (99)]

* Ultra-high resolution spectroscopy

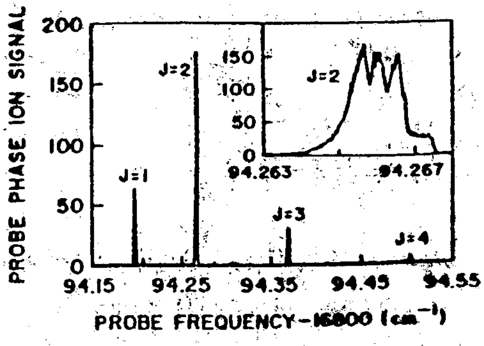


FIG. 59. High resolution photoassociative ionization spectrum of rotational levels associated with $v=48$ of the long-range 1_g state of Na_2. Note residual hyperfine splitting in the $J=2$ inset. From Ratliff *et al.* (1994).

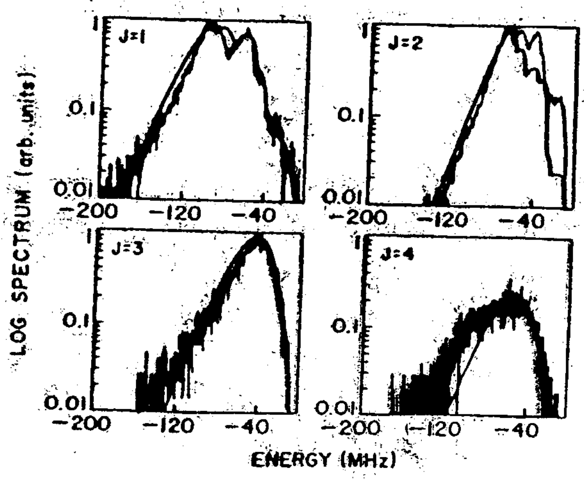
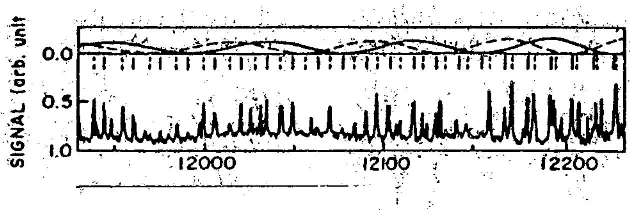


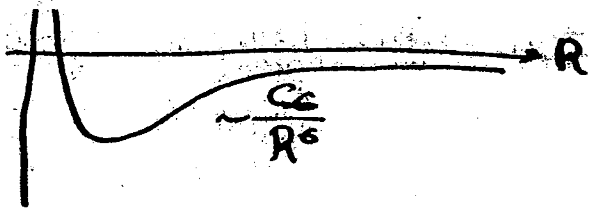
FIG. 60. High resolution line profile measurements and theory on rotational progression associated with $v=48$ of the long-range excited 1_g state of Na_2. From Napolitano *et al.* (1994).

* Observation of Condon oscillations in the spectrum



* Mapping the ground state wavefunction
→ Knowing zero positions determine the scattering-length. (a)

Two-body



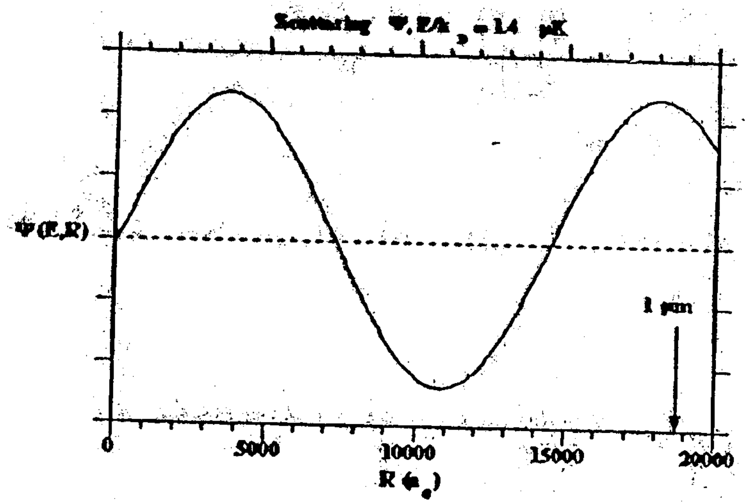
Near $E \to 0$

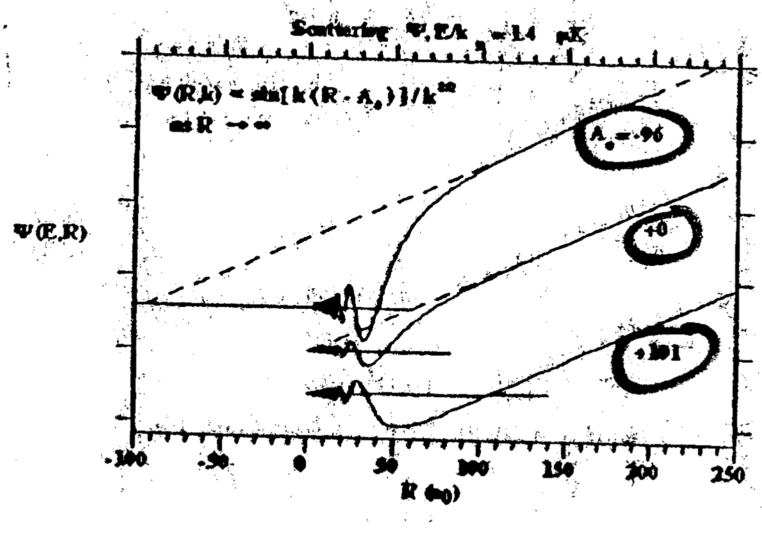
$$\lim_{k \to 0} k^{2\ell+1} \cot \eta_\ell = -\frac{1}{a_\ell}$$

S-wave

$$\sigma(E) = 8\pi a_0^2$$

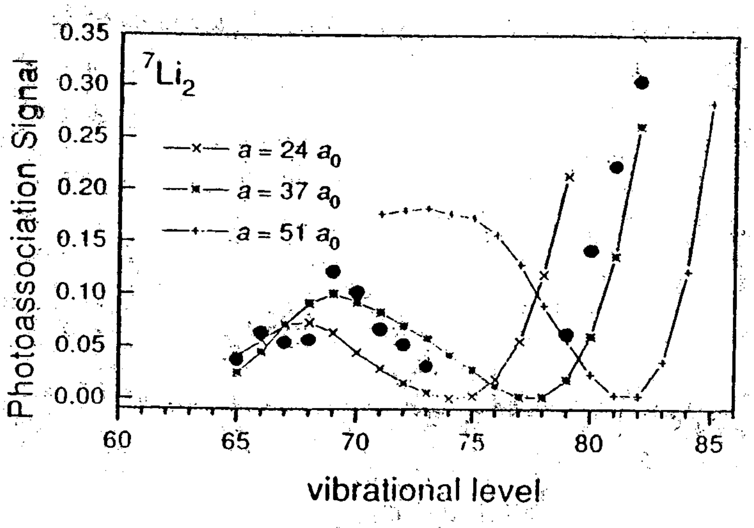
$$a_0 = -\lim_{k \to 0} \frac{\tan \delta_0}{k}$$



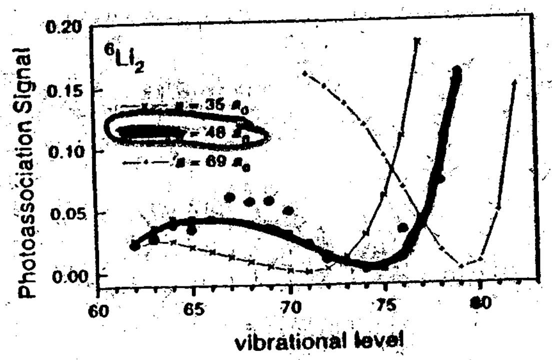


Why is a_0 important?

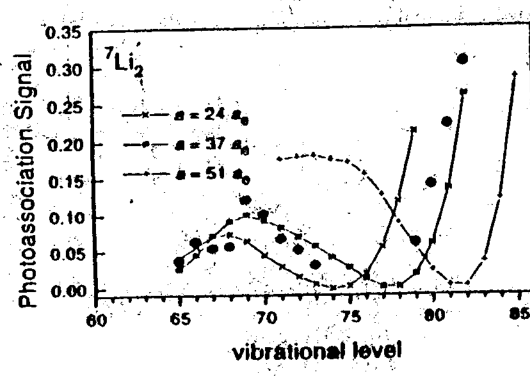
- Stability
- Evaporation
- thermodynamics



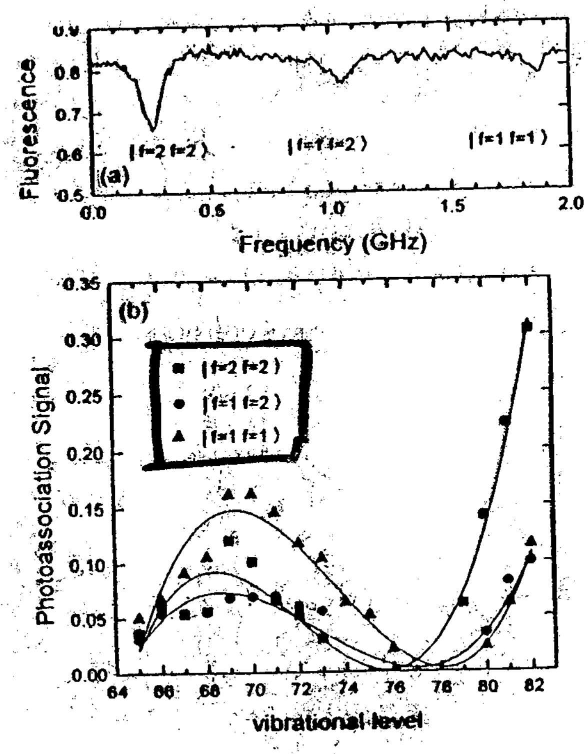
$^7Li_2$
a = 24 a_0
a = 37 a_0
a = 51 a_0
Photoassociation Signal
vibrational level
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
60
65
70
75
80
85



$^6Li_2$
a = 35 a_0
a = 48 a_0
a = 69 a_0
Photoassociation Signal
vibrational level



$^7Li_2$
a = 24 a_0
a = 37 a_0
a = 51 a_0
Photoassociation Signal
vibrational level



Fluorescence
(a)
|f=2 f=2⟩
|f=1 f=2⟩
|f=1 f=1⟩
Frequency (GHz)
(b)
|f=2 f=2⟩
|f=1 f=2⟩
|f=1 f=1⟩
Photoassociation Signal
vibrational level

TABLE III. Scattering Lengths (a_0)

	A_s	$A_t = A_{f_{max} m_{max}}$	A_{1-1}	$A_{3,-3}$	$A_{1,-1/2,2}$
$^6Li_2^a$	+45.5 ± 2.5	−2160 ± 250			
$^7Li_2^a$	+33 ± 2	−27.6 ± 0.5			
$^6Li^7Li^a$	−20 ± 10	+40.9 ± 0.2			
$^{23}Na_2^b$		+85 ± 3	+52 ± 5		
$^{39}K_2^c$	+132 ↔ +144	−1200 ↔ −60			
$^{41}K_2^c$	+80 ↔ +88	+25 ↔ +60			
$^{39}K_2^d$	+278 ± 14	+81.1 ± 2.4			
$^{40}K_2^d$	+153 ± 3	+1.7 ± 4.4			
$^{41}K_2^d$	+121 ± 2	+286 ± 36			
$^{85}Rb_2^e$		−500 ↔ −300			
$^{85}Rb_2^f$	+4500 ↔ +∞ −∞ ↔ −1200	−440 ± 140			
$^{87}Rb_2^e$		+85 ↔ 140			
$^{87}Rb_2^g$		+103 ± 5	+103 ± 5		+103 ± 5
$^{133}Cs_2^h$		|> 260|			
$^{133}Cs_2^i$				|46 ± 12|	

[a] Abraham, *et al.* (1997)
[b] Tiesinga, *et al.* (1996)
[c] Boesten, Vogels, *et al.* (1996)
[d] Côté, *et al.* (1997)
[e] Boesten, Tsai, *et al.* (1996)
[f] Tsai, *et al.* (1997)
[g] Julienne, *et al.* (1997)
[h] Arndt, *et al.* (1997)
[i] Monroe *et al.* (1993)

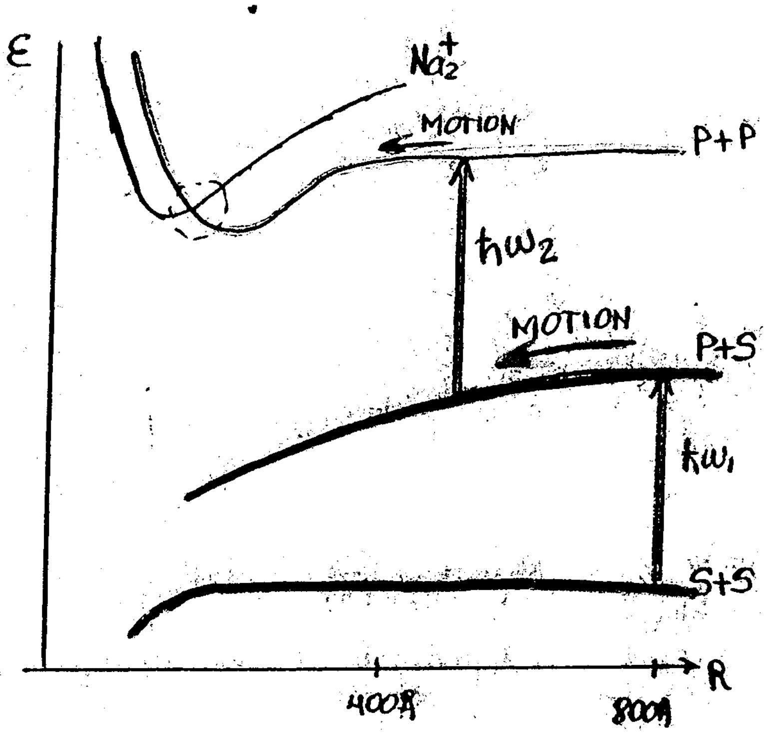
→ What states BEC is possible

→ Better evaporative cooling

→ corrections

Photo Associative Ionization

* Participation of double excited
* Final product is well known
* Nice prototype to study intermediate steps of the collision
* Mesoscopic system



$Na(3S) + Na(3S)$
$\rightarrow + \hbar\omega_1 \rightarrow Na \cdots Na^*$
$\rightarrow + \hbar\omega_2 \rightarrow Na^* \cdots Na^*$
$\rightarrow$ "Short range"
$\rightarrow Na_2^+ + e^-$

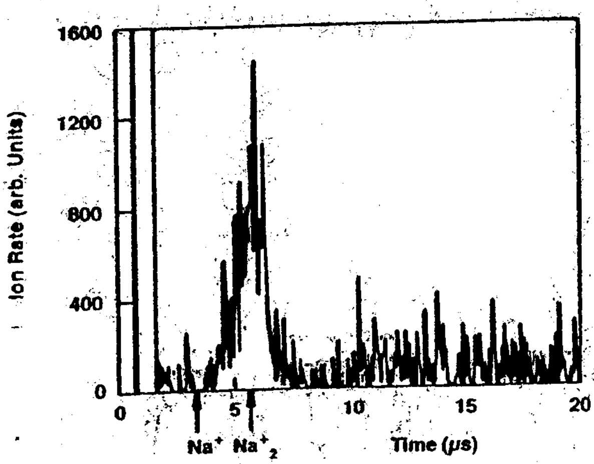
* It was the fisrt coed collision process investigated.

* Experimental set-up

MOT + Channeltron.

* Detection of Product

⟶ time of flight



Experiment done in Molasses

* Characterization of PAI

$$\frac{d[Na_2^+]}{dt} = K[Na]^2$$

* Determination of K allow to investigate the mechanisms

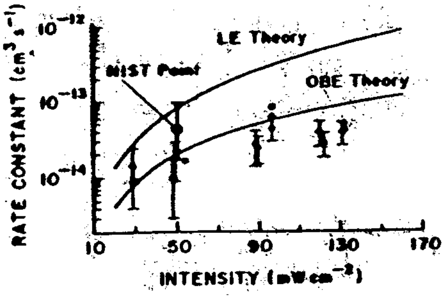


FIG. 53. Photoassociative ionization rate constant as a function of MOT light intensity. MOT laser detuned to the red of resonance about one natural linewidth. From Bagnato, Marcassa, Wang, *et al.* (1993).

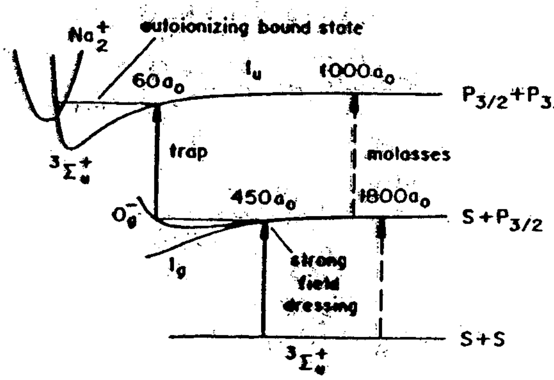


FIG. 52. Photoassociative ionization in Na collisions. From Heather and Julienne (1993).

Model for K_4 (Gallagher - 89)

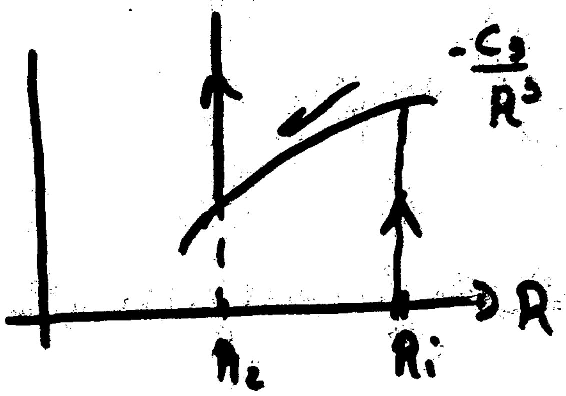
→ Local equilibrium theory

K_4 = Ionization rate = $\prod$(fators)

- fator (1) → excitation
- fator (2) → spontaneous emission while excited
- fator (3) → New excitation (phase space factor)
- fator (4) → oscillation
- fator (5) → New spontaneous emission
- fator (6) → Ionization

LOCAL EQUILIBRIUM THEORY

Simple rate calculation:



R_i → first excitation

R_2 → second absorption

ω_0 → atomic resonance

ω_1, ω_2 → two photons

$$\text{Rate of excitation } (R_i) = \underbrace{\frac{n^2}{2} 4\pi R_i^2 dR_i}_{\#\text{ pairs}} \left(\frac{I_1}{\hbar\omega_1}\right) \frac{\lambda_1^2}{2\pi} \frac{\Gamma^2}{\Gamma^2 + 4(\Delta_1 - V(R_i))^2}$$

~~probabi~~

$$\text{Probability of excitation } (R_2) = \left(\frac{I_2}{\hbar\omega_2}\right) \frac{\lambda_2^2}{2\pi} \frac{\Gamma^2}{\Gamma^2 + 4(\Delta_2 + V(R_2))^2} \underbrace{\frac{dR_2}{v(R_2, R_i)}}_{\text{phase-space factor}}$$

Decay going $R_i \to R_2$: factor $e^{-\Gamma t^*(R_i, R_2)}$

Possibility of oscillation: factor $= \dfrac{1}{1 - \eta_s e^{-2\Gamma t^*(R_i)}}$

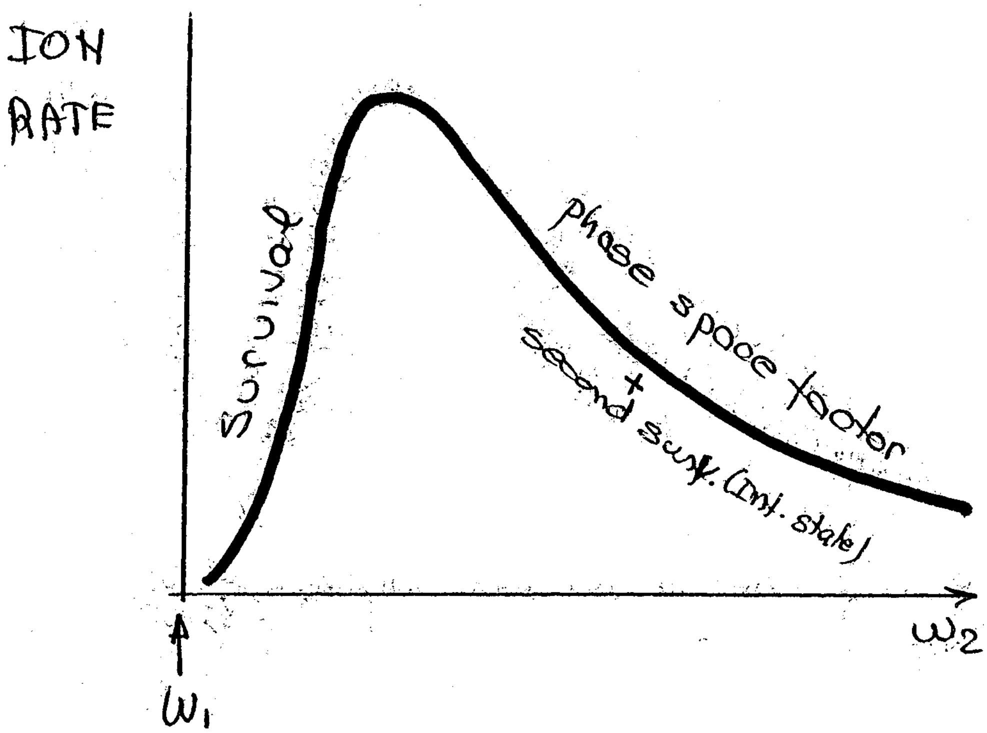
Decay going $R_2 \to$ short range $\quad e^{-\Gamma t^{**}(R_2, R_i)}$

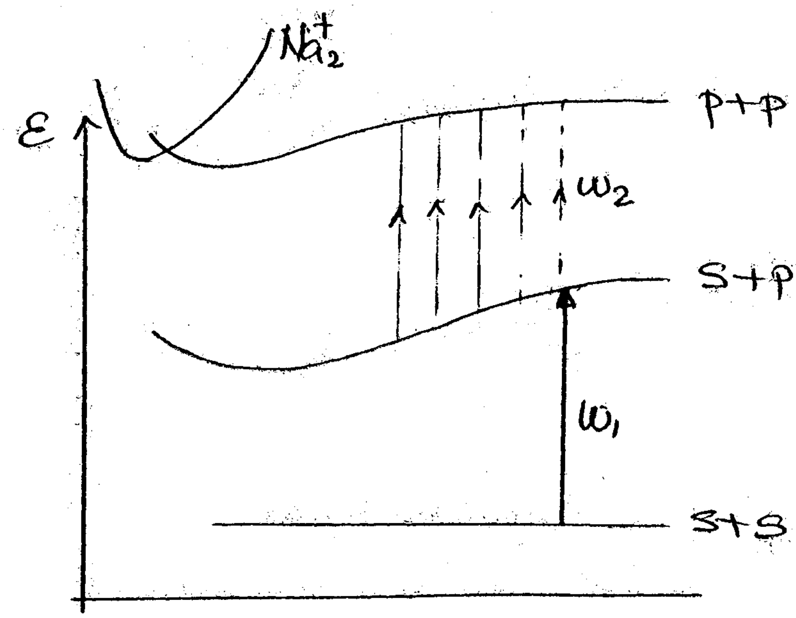
K_4 Total rate $= \int$ product.

$$K_4 = \frac{n^2}{2} I_1 I_2 \left[\frac{\lambda^2}{\Gamma \hbar\omega_0}\right]^2 \frac{4\pi}{9} \frac{C_3}{\hbar} \left(\frac{R_N}{R_2}\right)^{5/2} \times$$

$$\times \int_0^\infty d\Delta_i \int_0^\infty d\Delta \frac{(1 - \eta_s u)^{-1} e^{-\Gamma t^*} e^{-\Gamma t^{**}}}{\Delta_i^2 \Delta^{4/3} (\Delta - \Delta_i)^{1/2} [1 + 4(\Delta_1 - \Delta_i)^2] [1 + 4(\Delta_2 + \Delta)^2]}$$

Fix ω_1 and vary ω_2

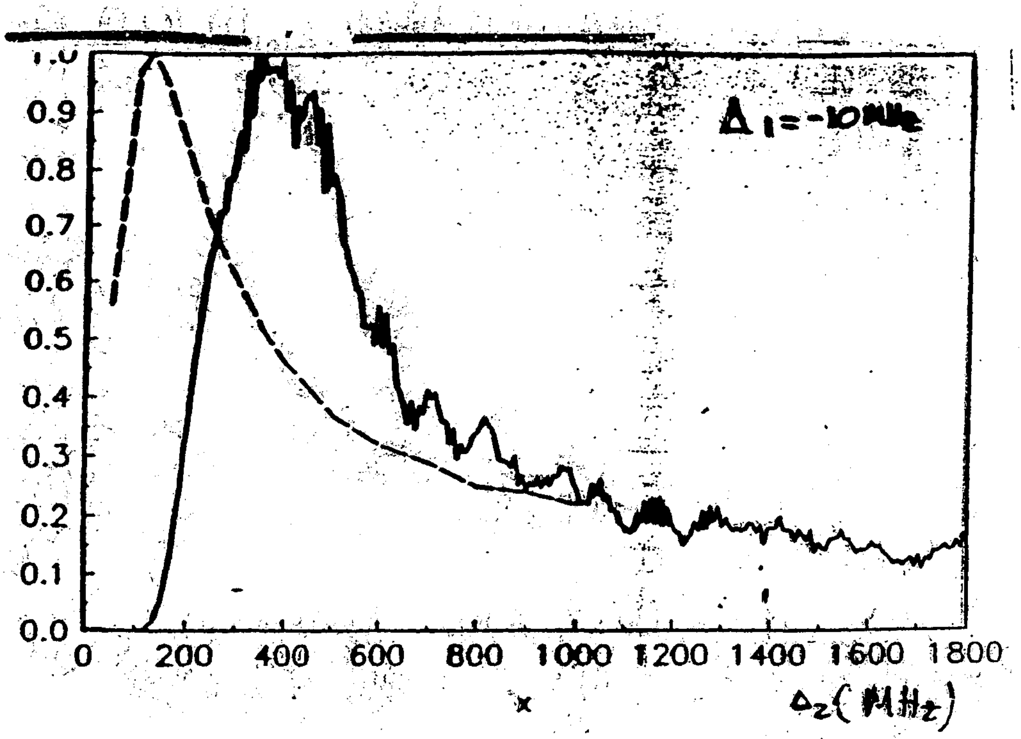




* ω_2 always bluer than ω_0

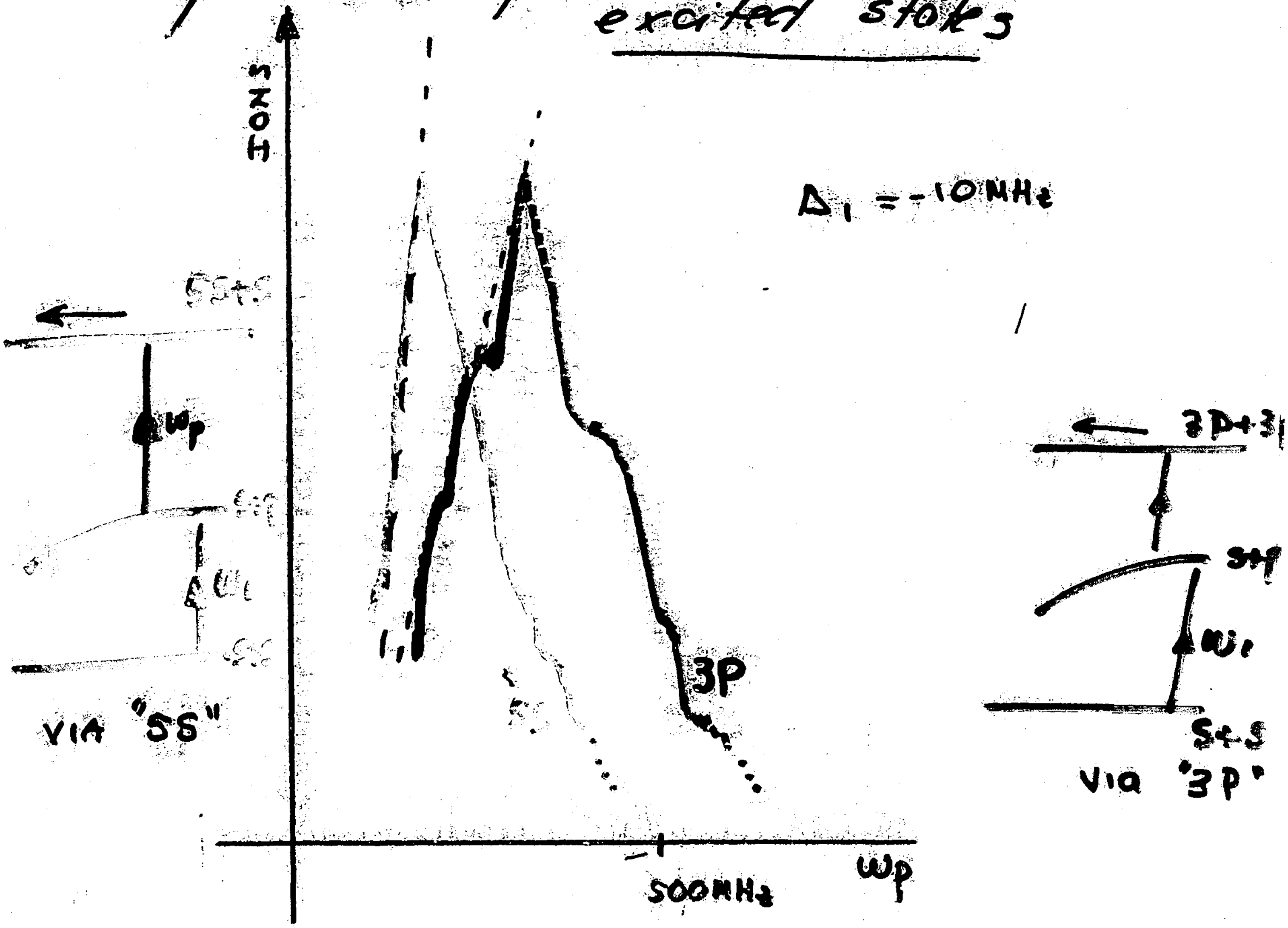
Experiment $\Delta_1 = -10\,MHz$ (MOT laser)

$\omega_2 \rightarrow$ probe laser



* Well explained behavior
* Shift ~ 300 MHz (probably due to excited state hyperfine structure)

Comparison of survival in two excited states



- Longer lived states
  - Maximum at lower frequency
  - Faster raise up

better survival

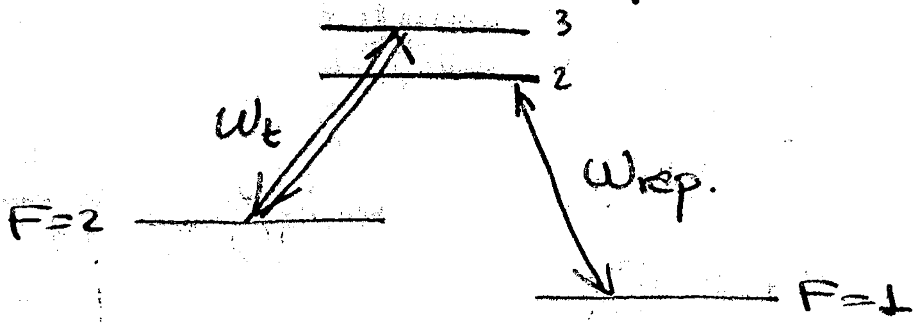
- Collisions can start with several possibilities

(F=1) + (F=1) ⟸ pure

(F=1) + (F=2) ⟵ mixture

(F=2) + (F=2) ⟸ pure

To prepar sample.



→ turn off ω_t (~ 100 μs)

⟶ sample in F=1

→ turn off ω_{rep} (~ 100 μs)

⟶ sample in F=2

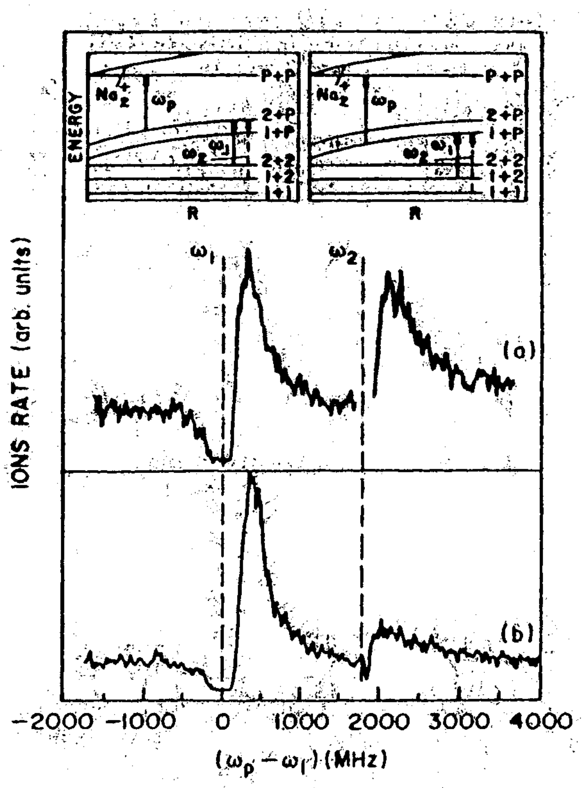


FIG. 55. Two-color photoassociative ionization. Inset shows MOT (ω_1) and repumper (ω_2) fixed frequencies and sweeping probe (ω_p) frequency. From Bagnato, Marcassa, Tsao *et al.* (1993).

60% F=1
40% F=2

30% F=1
70% F=2

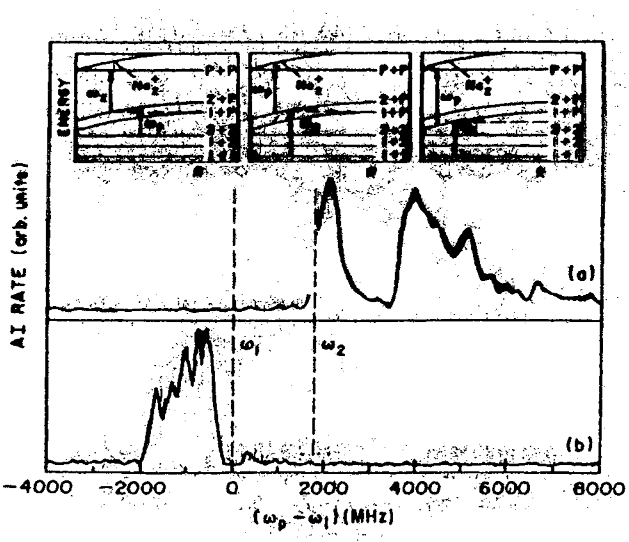


FIG. 56. Two-color photoassociative ionization. Insets above features describe excitation routes. Frequencies are labeled as in the previous figure. From Bagnato, Marcassa, Tsao, *et al.* (1993).

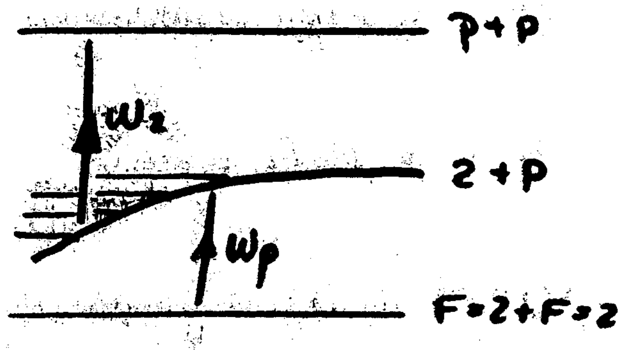
100% F=1

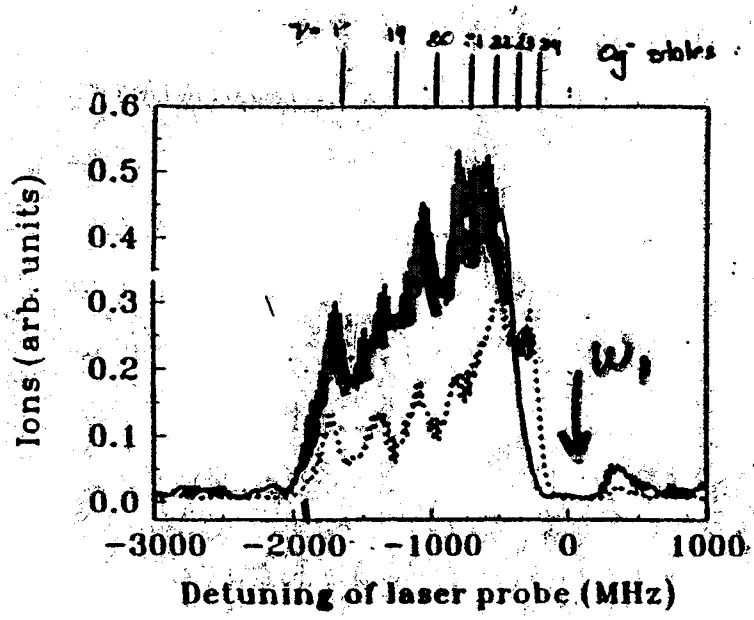
100% F=2

⇑

Free to bound

Gate B $\omega_2 + \omega_p$ (F=2+ F=2)
(Laser Phys. 4, 1062 (94))





Stwalley
(PRL - 41, 1164 (78))

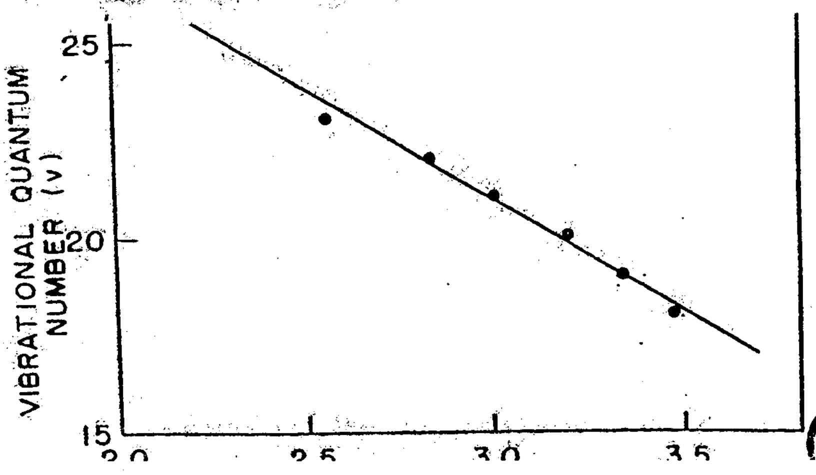
$\nu_0 - \nu = a_3 (E - D)^{1/6}$

0_g^- $\begin{cases} a_3 = 6.25\ MHz^{-1/6} \\ D \sim KHz \\ \nu_0 \sim 39.6 \end{cases}$

↑ Theory

Free to bound transition ⟹ vibrational structure of P+S long-range state)

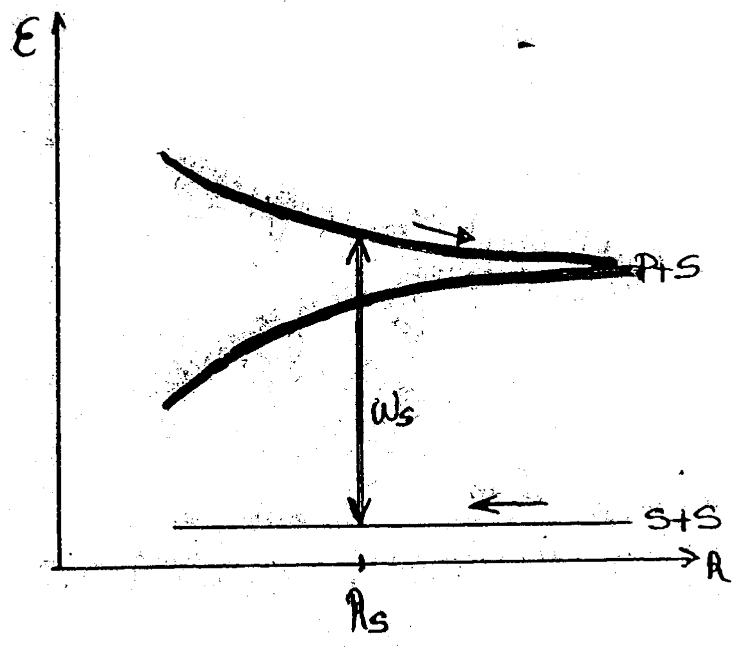
- very near threshold states



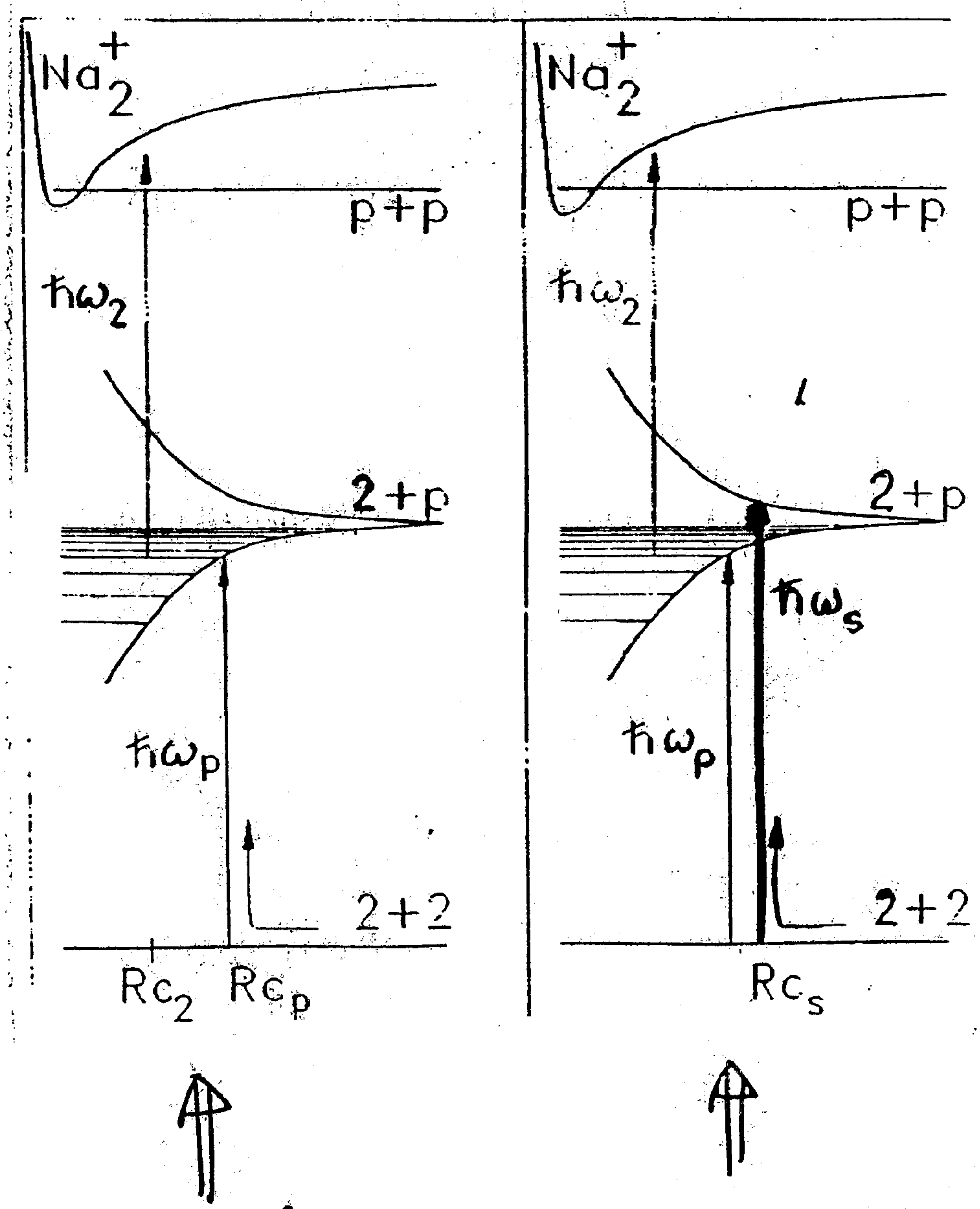
$a_3 = 6.54$
$\nu_D = 40.6$
D < 3 [illegible]

Optical Shielding

* Idea: To use repulsive states through photons to prevent atoms to get together.



Use PAI as a prototype to investigate optical Shielding



Normal PAI

Shielding

• Primeira demonstração

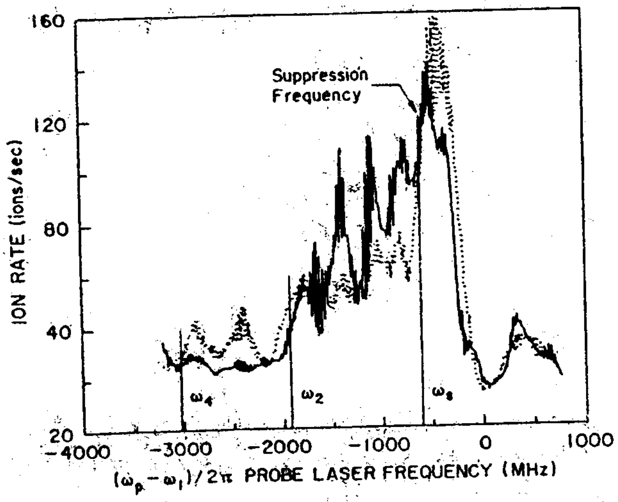


FIG. 77. Effect of optical suppression on the photoassociative ionization spectrum observed in a Na MOT. Solid curve is the photoassociative ionization spectrum without the suppression field ω_s present. Dotted curve shows the photoassociative ionization spectrum ω_s present. Note enhancement of signal to the right of ω_s, suppression to the left. Note also the cutoff of the photoassociative ionization signal to the left of ω_2 without the suppression field and the extension of this cutoff to ω_4 when ω_s is present. From Marcassa *et al.* (1994).

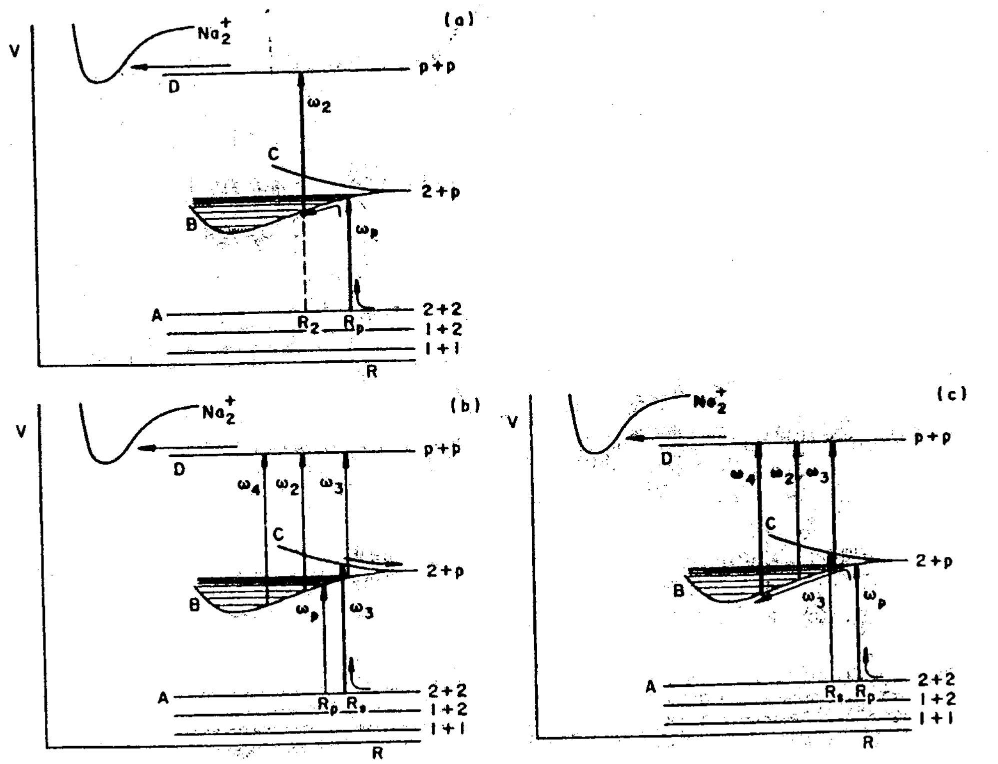
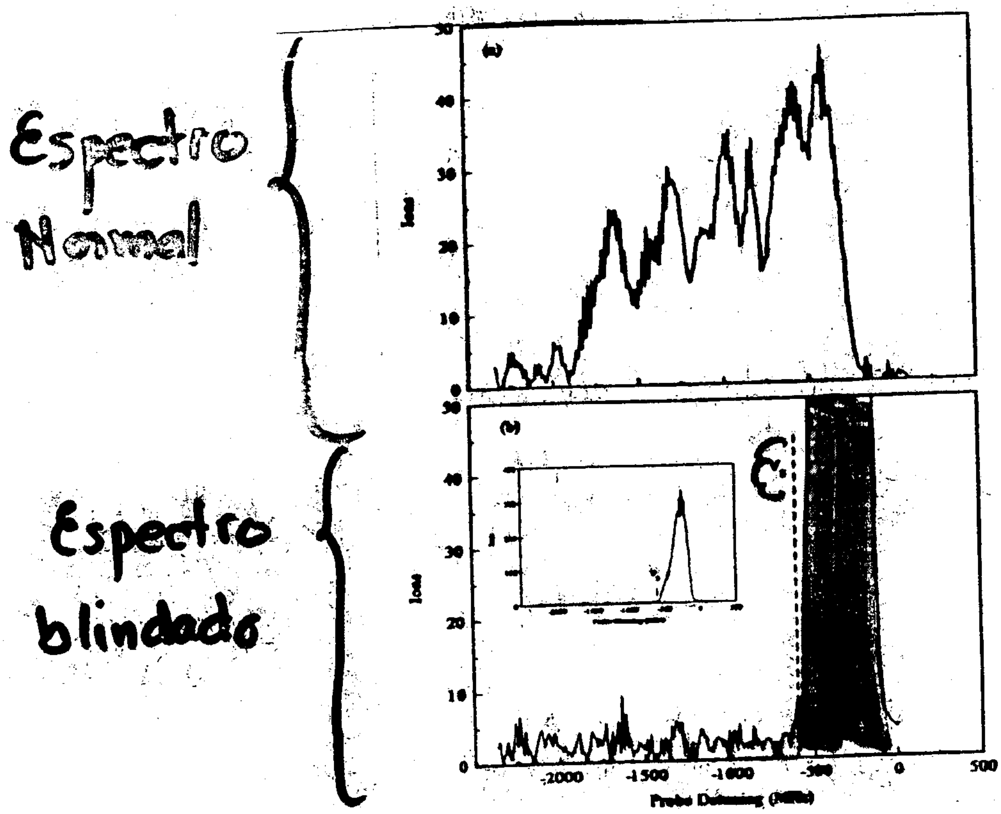
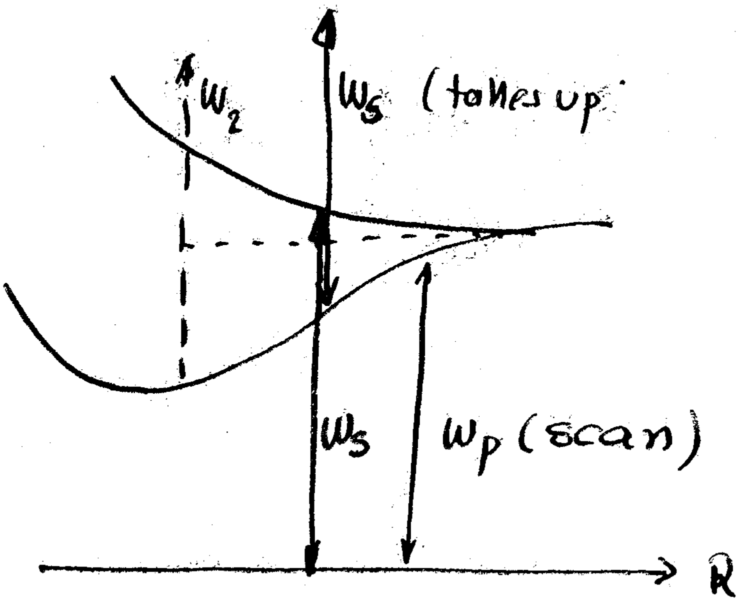


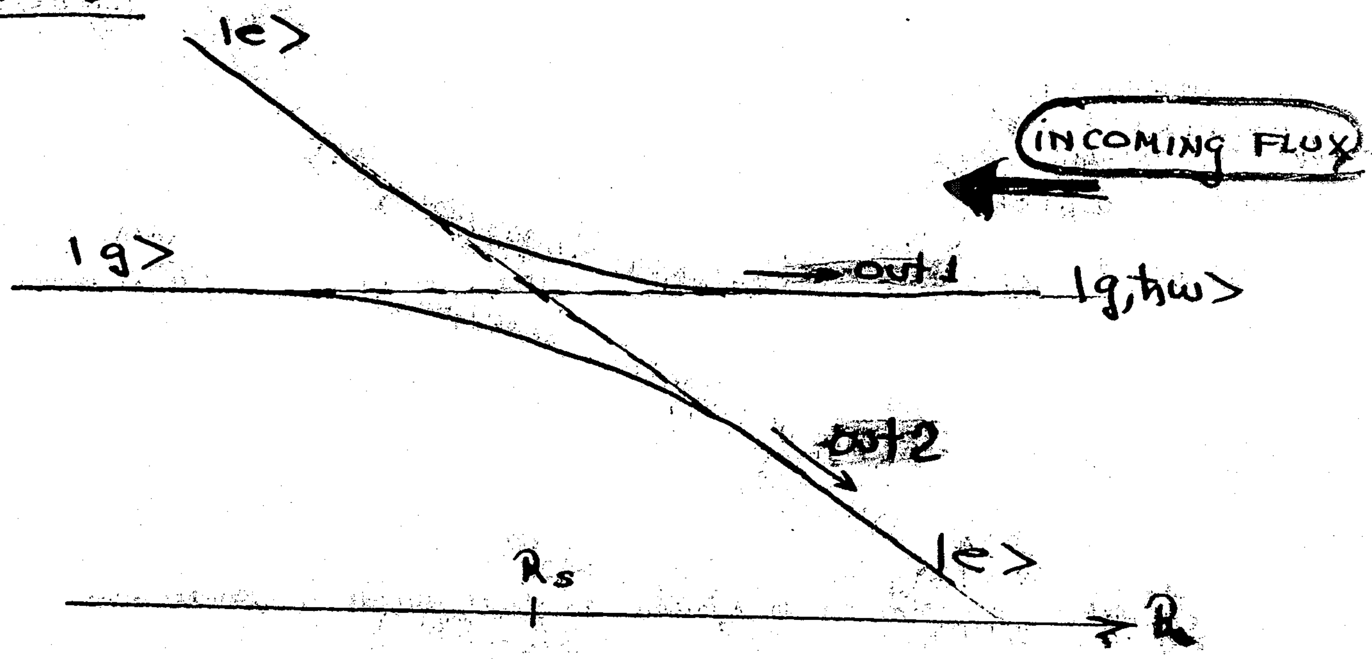
FIG. 76. Schematic of transitions showing photoassociative ionization and optical suppression of photoassociative ionization: (a) Two-step PAI process; (b) suppressor frequency ω_3 imposed on the collision, rerouting incoming flux to the repulsive excited curve; (c) with ω_p tuned to the right of ω_3, photoassociative ionization takes place with enhanced probability due to addition of ω_3 and ω_4 to ω_2. From Marcassa *et al.* (1994).

Shielding - Enhancement.





Model



* Incoming flux start in ground state
* R_s anti-crossing
* Probability to stay |e> at R_s (Landau-Zener)

$$\boxed{P = 1 - \exp\left\{-\frac{\xi \Omega^2}{D(R) \cdot v(R)}\right\}}$$

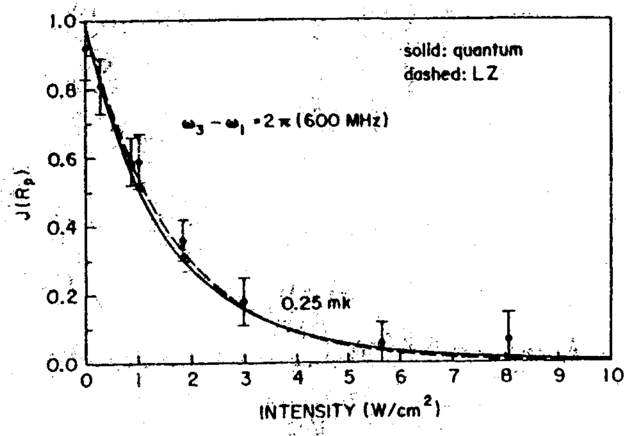
* If excited at R_s ⟶ revert motion
* If stay |g> with 1-P new possibility on the return

$$P(\text{out}1) = p^2 + (1-p)(1-p) = 1 - 2p + 2p^2$$

$$P(\text{out}2) = p(1-p) + (1-p)p = 2p(1-p)$$

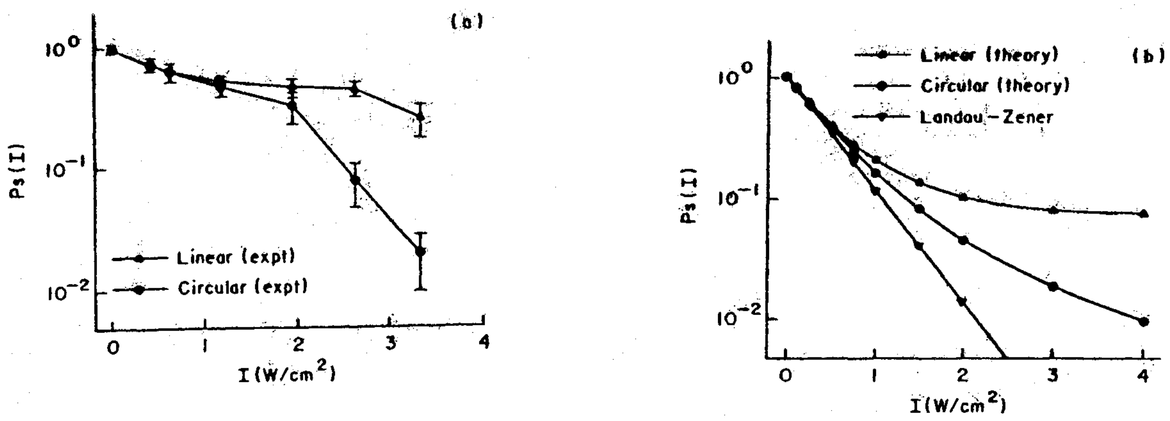
High intensity $\begin{cases} P(\text{out}1) \longrightarrow 1 \\ P(\text{out}2) \longrightarrow 0 \end{cases}$

* Shielding as a function of intensity



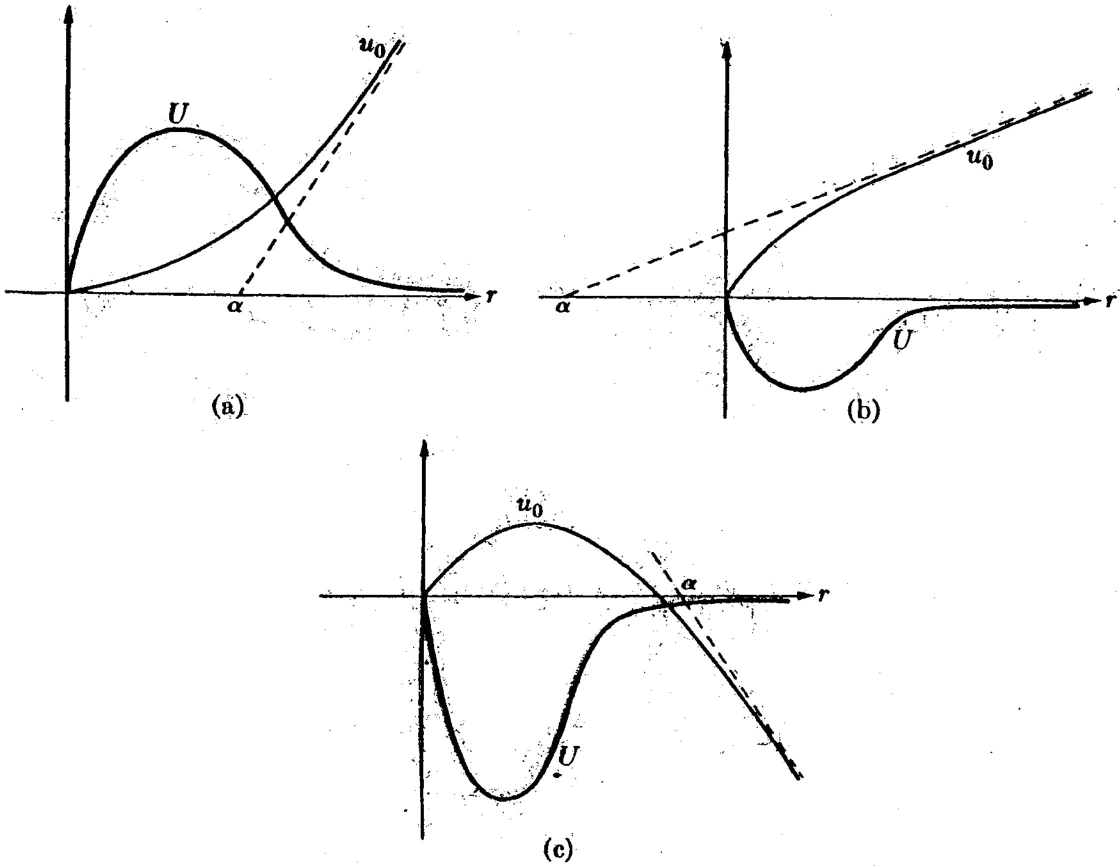
→ Experiment { $J \to \neq 0$ for $I \to \infty$

* Polarization dependence:



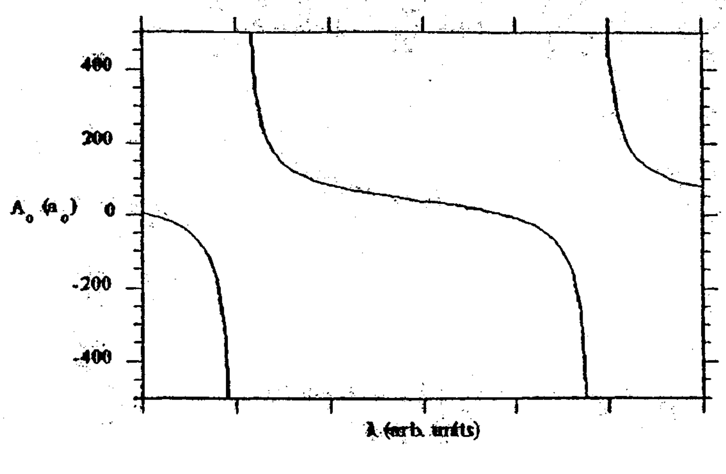
Variation of a with fields

→ Small variation of the potential can change a.

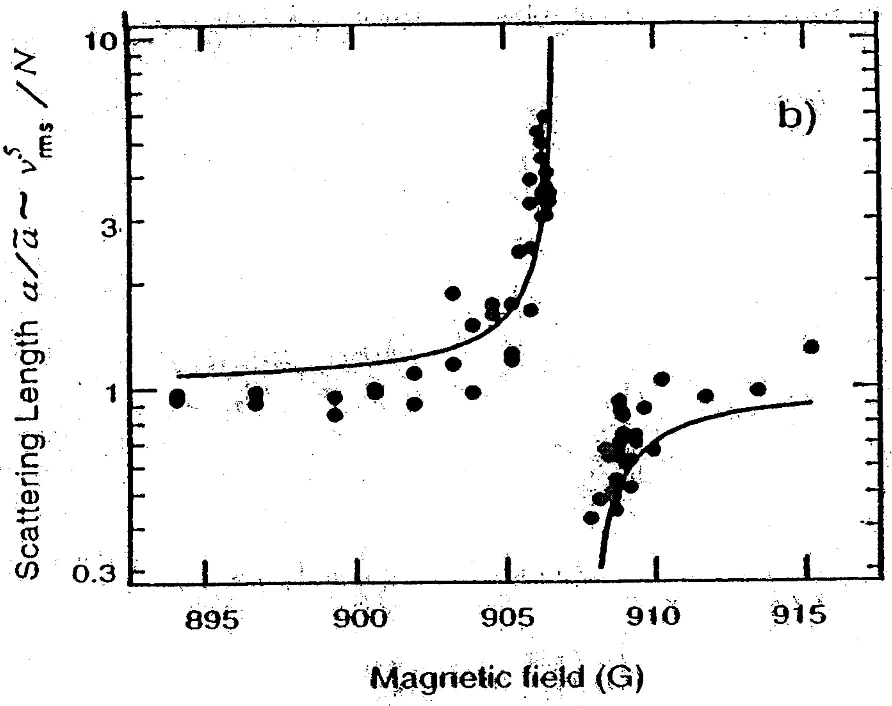


Using B-field (change in hyperfine levels)

Feshback - Resonances

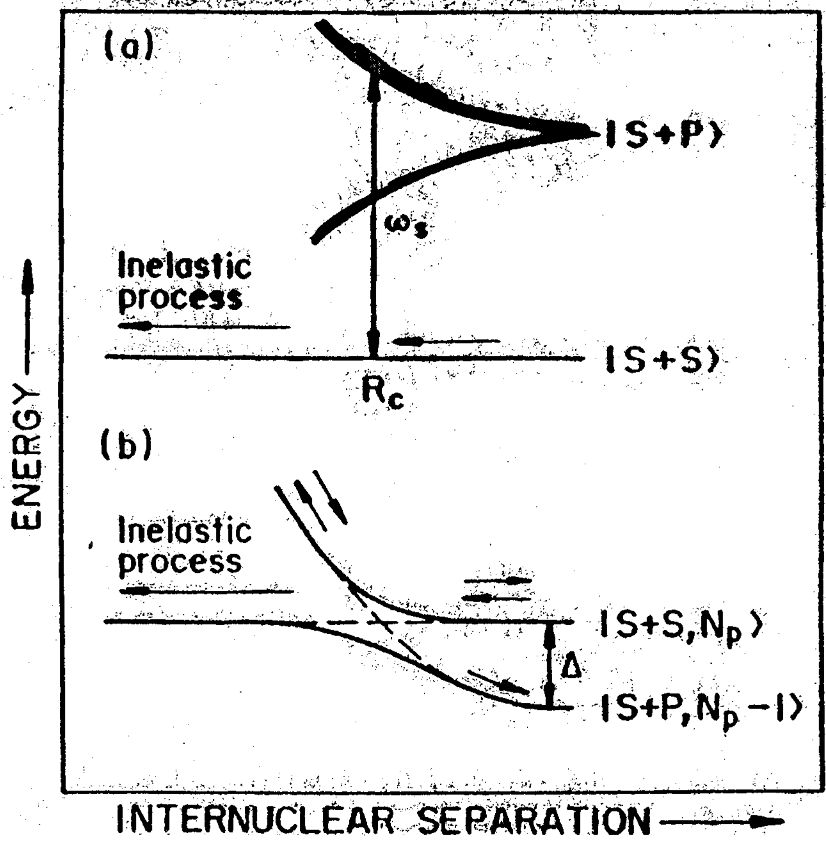


First measurement in Na.



* Using Photons

First we thought in processes like optical shielding



- Phys. Rev. Lett 73, 1911 (1994)
- Phys. Rev. A 52, R913 (1995)
- Phys. Rev. Lett. 76, 2033 (1996)
- Phys. Rev. A 55, 4407 (1997)
- Science Spect. 7, 50 (1996)

"in" in |S+S⟩

P(out |S+S⟩) → ↓

High Intensity

Knowing a_0

$$H(p,r) = \frac{p^2}{2M} + U(\vec{r}) + g\, n(\vec{r})$$

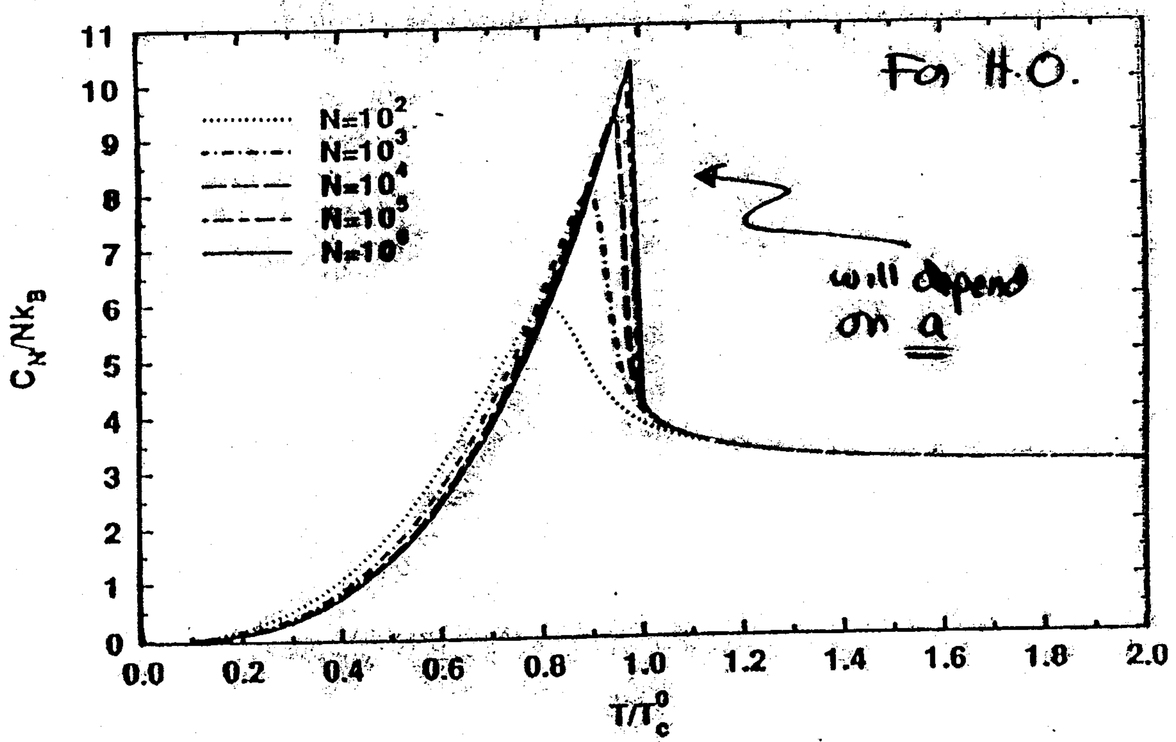
$$V(\vec{r}_1 - \vec{r}_2) = V_0\, \delta(\vec{r}_1 - \vec{r}_2) \longrightarrow V_0 = \frac{4\pi\hbar^2 a}{M}$$

$$\frac{\text{Energy}}{\text{particle}} = \frac{1}{2} V_0\, n(r) \longrightarrow g = \frac{2\pi\hbar^2 a}{M}$$

- Determine new T_c ($\neq T_c^0$)
- Determine new $\frac{N_0}{N}$

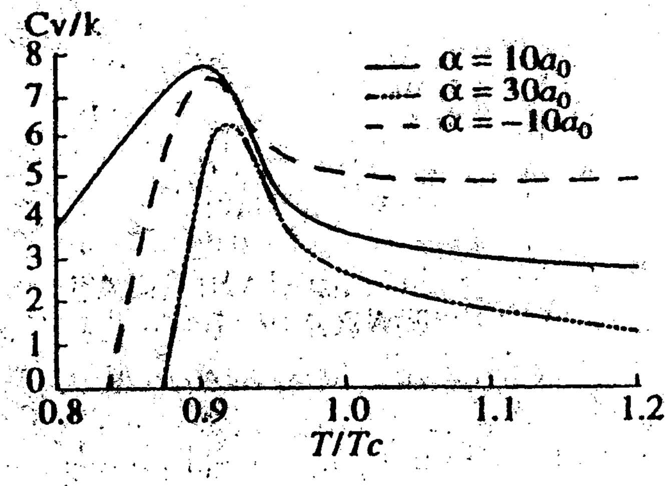
[Phys. Rev. A 35 (1987)
Phys. Rev. A 44 (1991)]

- Heat Capacity: $C = \left.\frac{\partial E}{\partial T}\right)_N$

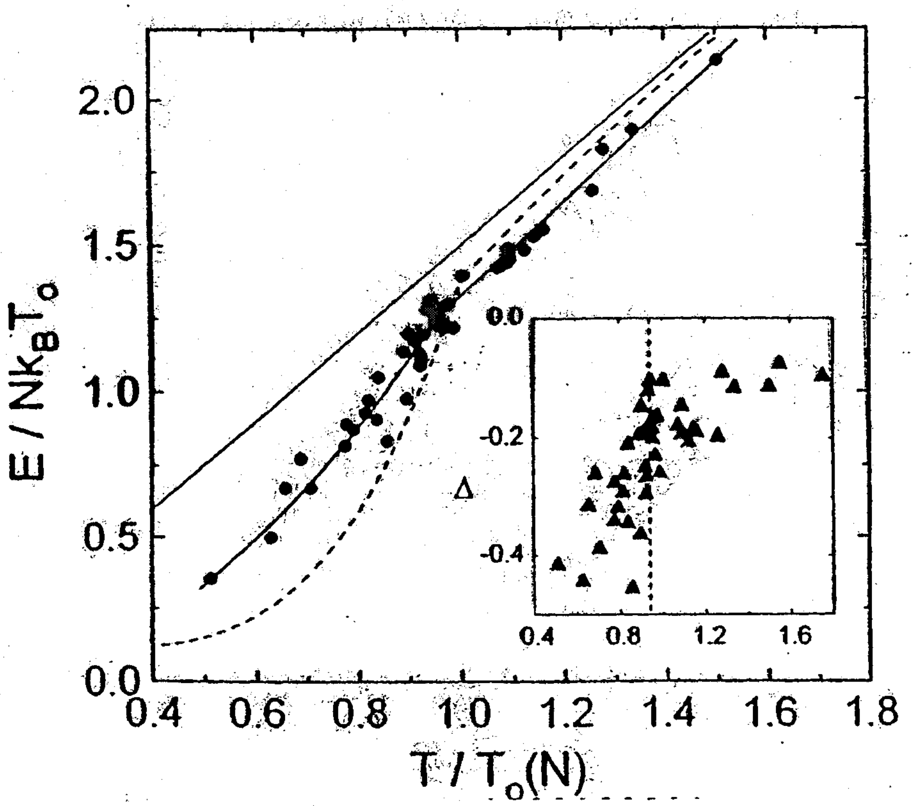


[Phys. Rev. A 55, 3954 (1997)]

[Cos. Phys. 7, 40 (1997)]



Lack of data for heat capacity.



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em colisões frias

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