

SMR 1302 - 8

WINTER SCHOOL ON LASER SPECTROSCOPY AND APPLICATIONS

19 February - 2 March 2001

COLD COLLISIONS AND CHEMICAL REACTION

Vanderlei S. BAGNATO

**Univ. de Sao Paulo, IFSC - Center for Optical Science & Photonics
SP-13560-970 Sao Carlos
Brazil**

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 -

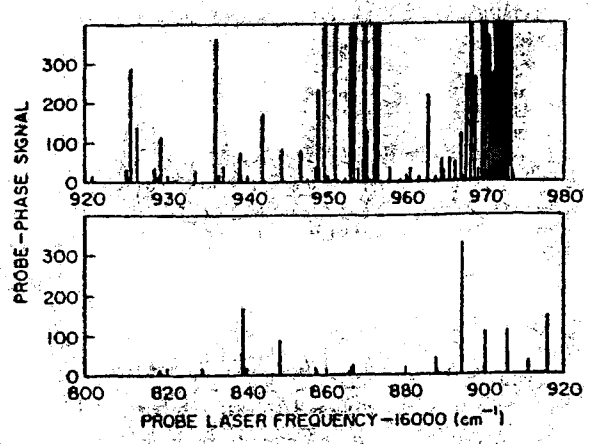
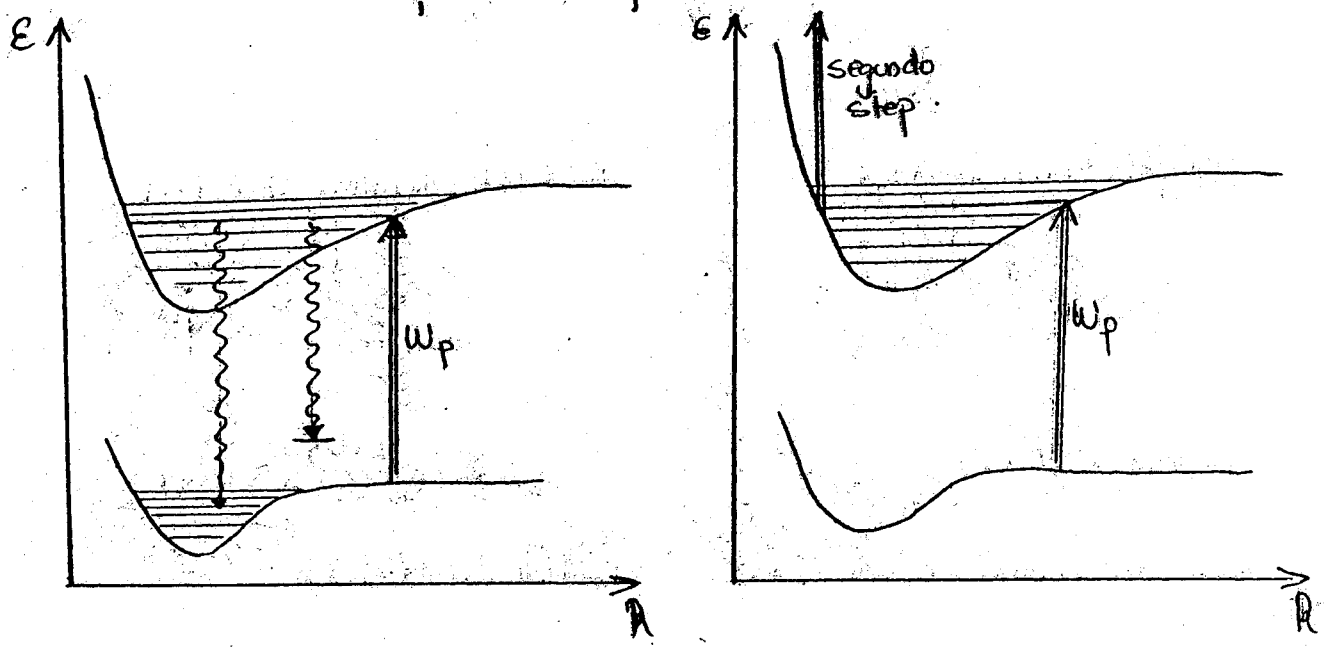


FIG. 57. Photoassociative ionization spectrum of Na collisions from the NIST dark-spot MOT. From Ratliff *et al.* (1994).

Ex: Na
by Ionization
(NIST)

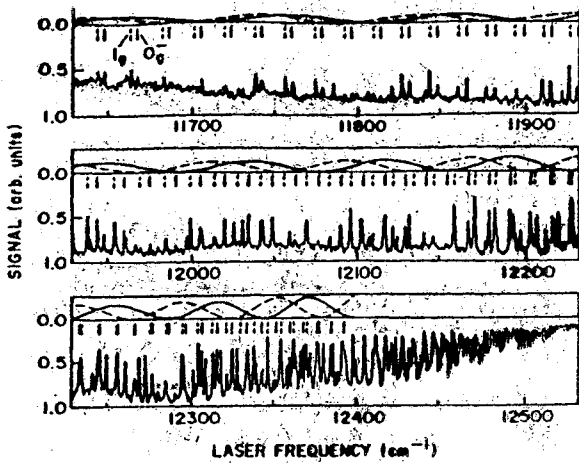


FIG. 61. Photoassociation FORT trap-loss fluorescence spectrum of Rb_2 . From Miller *et al.* (1993b).

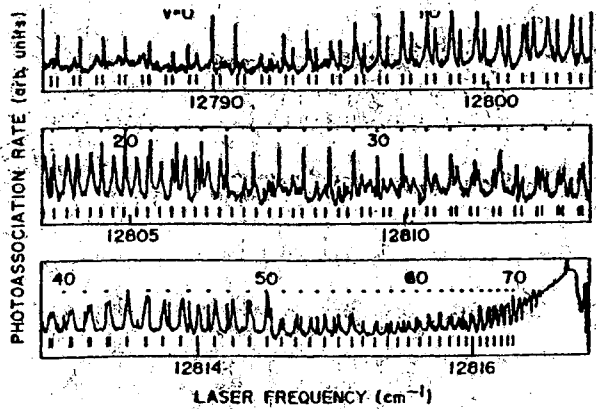


FIG. 62. Cline *et al.* (1994a, 1994b) separated the trapping and scanning functions with two different lasers: one laser at fixed frequency produced the FORT while a probe laser scanned through the resonances.

* Determination of potential terms

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

C_3 is related to the excited state life time

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

→ Determination of σ

Ex: Medida para Li

Experiment	Lifetime (ns)
Gaupp <i>et al.</i> [7]	27.1 ± 0.1
Caillon <i>et al.</i> [18]	27.0 ± 0.1
This work	27.0 ± 0.1
Theory	
Olundell <i>et al.</i> [6]	27.0
Wales [19]	27.0
Pipin <i>et al.</i> [20]	27.0
Citang [21]	27.0
Mansson-Pompl <i>et al.</i> [22]	27.0
Tong <i>et al.</i> [23]	27.0
Rinnell <i>et al.</i> [24]	27.0

$$\sigma_{Li} = 26.99 \pm 0.16 \text{ ns}$$

Ha medidas para Na, K, Rb, Cs
[Rev. Mod. Phys. 71, 1 (99)]

* Ultra-high resolution spectroscopy

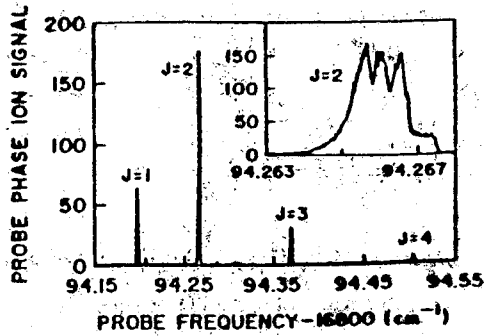


FIG. 59. High resolution photoassociative ionization spectrum of rotational levels associated with $v=48$ of the long-range 1_g state of Na_2 . Note residual hyperfine splitting in the $J=2$ inset. From Ratliff *et al.* (1994).

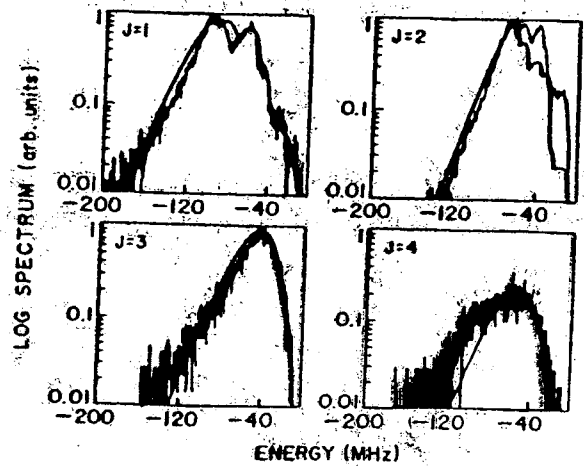
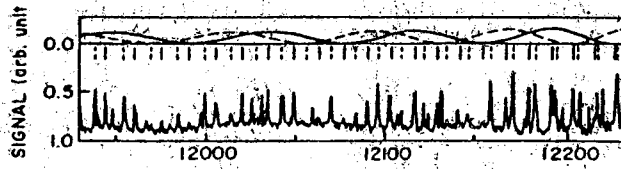


FIG. 60. High resolution line profile measurements and theory on rotational progression associated with $v=48$ of the long-range excited 1_g state of Na_2 . From Napolitano *et al.* (1994).

* Observation of Condon oscillations in the spectrum

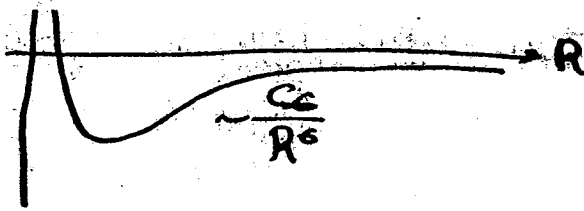


* Mapping the ground state wavefunction

→ Knowing zero positions determine the scattering-length.

(a)

Two-body



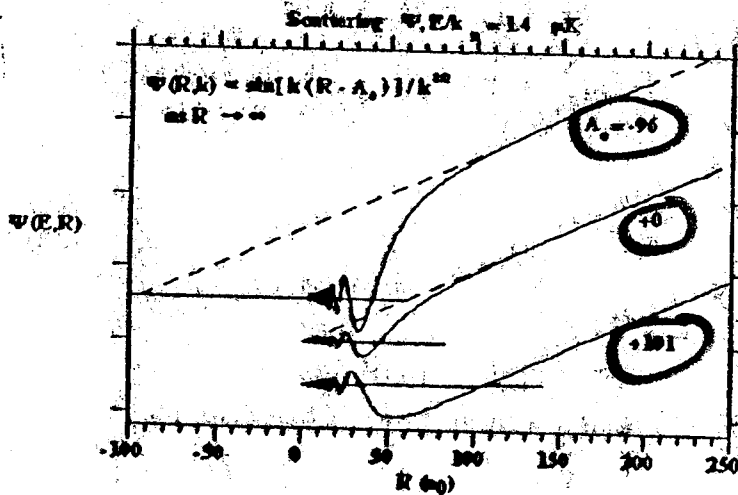
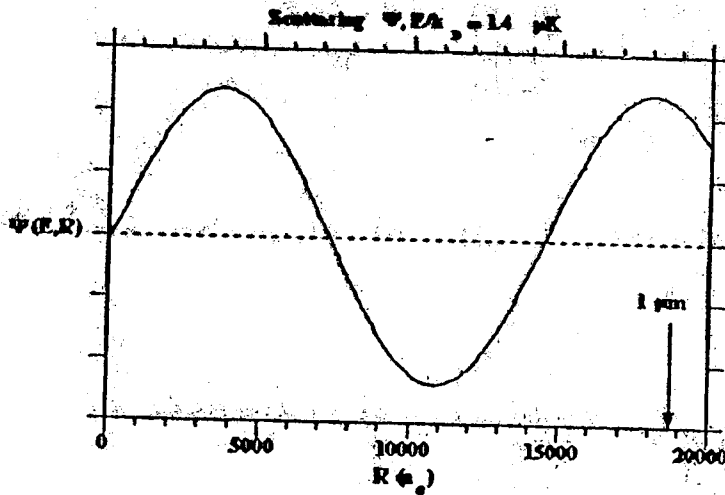
Near $E \rightarrow 0$

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

S-wave

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

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



Why is a_0 important?

{ Stability
Evaporation
thermodynamics

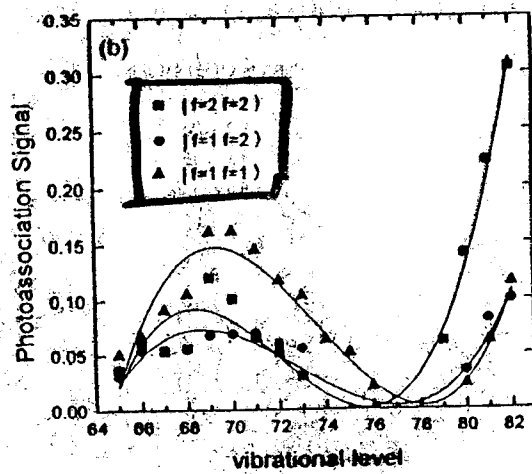
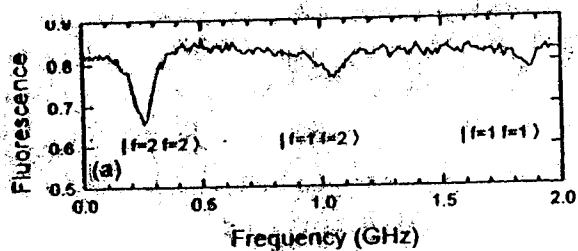
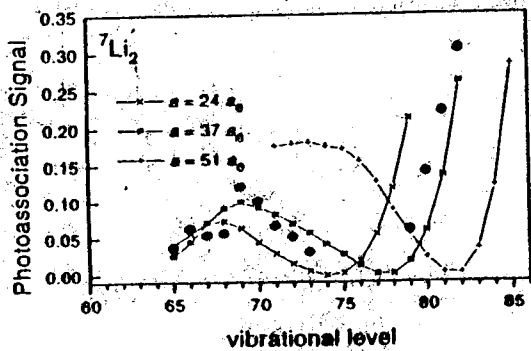
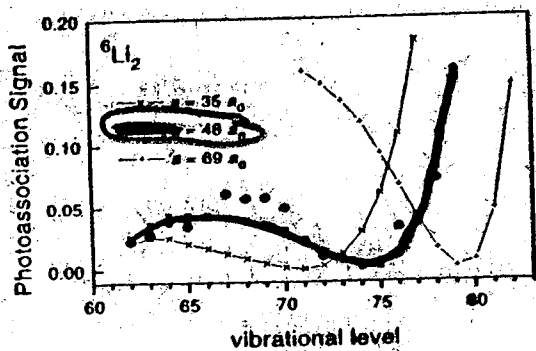
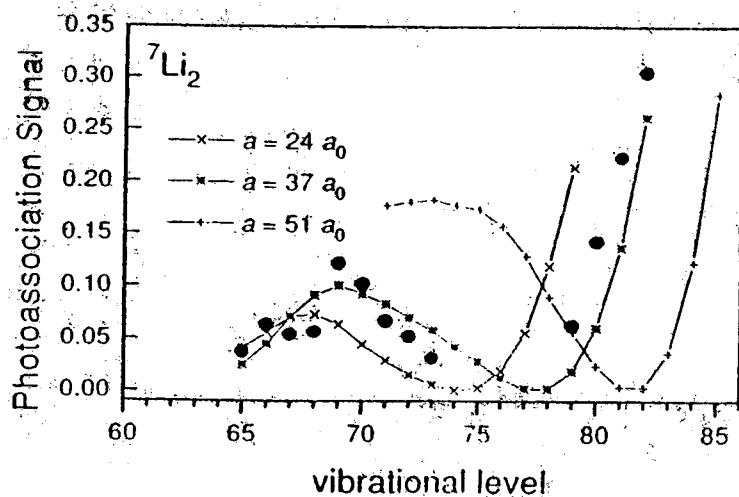


TABLE III. Scattering Lengths (a_0)

	A_s	$A_t = A_{f_{max} m_{max}}$	A_{1-1}	$A_{3,-3}$	$A_{1,-1/2,2}$
${}^6\text{Li}_2^a$	$+45.5 \pm 2.5$	-2160 ± 250			
${}^7\text{Li}_2^a$	$+33 \pm 2$	-27.6 ± 0.5			
${}^6\text{Li}^7\text{Li}^a$	-20 ± 10	$+40.9 \pm 0.2$			
${}^{23}\text{Na}_2^b$		$+85 \pm 3$		$+52 \pm 5$	
${}^{39}\text{K}_2^c$	$+132 \leftrightarrow +144$	$-1200 \leftrightarrow -60$			
${}^{41}\text{K}_2^c$	$+80 \leftrightarrow +88$	$+25 \leftrightarrow +60$			
${}^{39}\text{K}_2^d$	$+278 \pm 14$	$+81.1 \pm 2.4$			
${}^{40}\text{K}_2^d$	$+153 \pm 3$	$+1.7 \pm 4.4$			
${}^{41}\text{K}_2^d$	$+121 \pm 2$	$+286 \pm 36$			
${}^{85}\text{Rb}_2^e$		$-500 \leftrightarrow -300$			
${}^{85}\text{Rb}_2^f$	$+4500 \leftrightarrow +\infty$ $-\infty \leftrightarrow -1200$	-440 ± 140			
${}^{87}\text{Rb}_2^g$		$+85 \leftrightarrow 140$			
${}^{87}\text{Rb}_2^g$		$+103 \pm 5$	$+103 \pm 5$		$+103 \pm 5$
${}^{133}\text{Cs}_2^h$		> 260			
${}^{133}\text{Cs}_2^i$				$ 46 \pm 12 $	

^aAbraham, *et al.* (1997)

^bTiesinga, *et al.* (1996)

^cBoesten, Vogels, *et al.* (1996)

^dCôté, *et al.* (1997)

^eBoesten, Tsai, *et al.* (1996)

^fTsai, *et al.* (1997)

^gJulienne, *et al.* (1997)

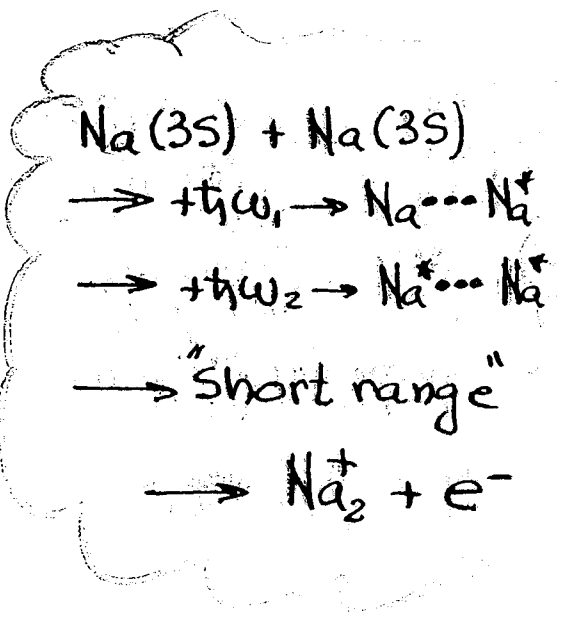
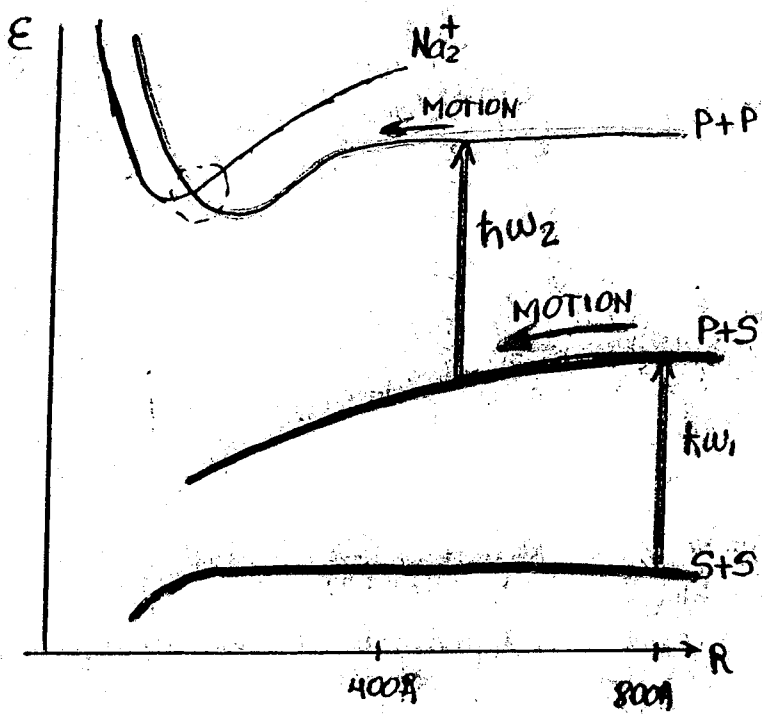
^hArndt, *et al.* (1997)

ⁱMonroe *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

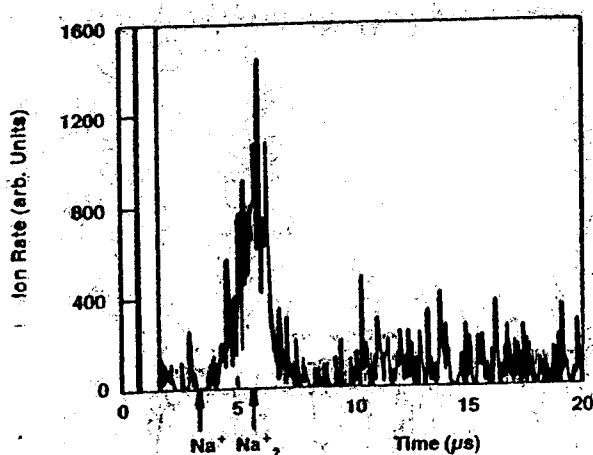


* It was the first coed collision process investigated.

* Experimental set-up

MOT + Channeltron.

* Detection of Product
 → time of flight



Experiment
 done in
 Molasses

* Characterization of PAI

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

* Determination of K allow to
 investigate the mechanisms

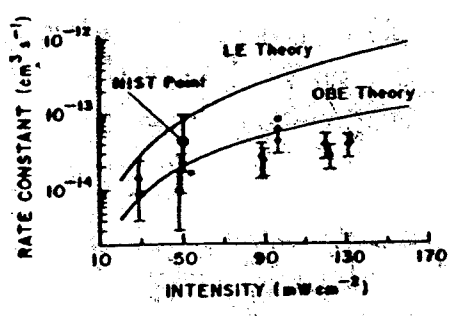


FIG. 53. Photoassociative ionization rate constant as a function of MOT light intensity. MOT laser detuned to the red of resonance about one natural linewidth. From Bagnato, Marcassa, Wang, et al. (1993).

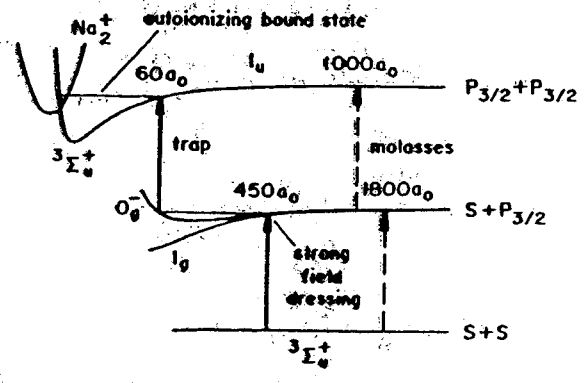


FIG. 52. Photoassociative ionization in Na collisions. From Heather and Julienne (1993).

Model for K_4 (Gallagher - 89)

→ Local equilibrium theory

$$K_4 = \text{Ionization rate} = \prod (\text{factors})$$

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

LOCAL EQUILIBRIUM THEORY

III-10

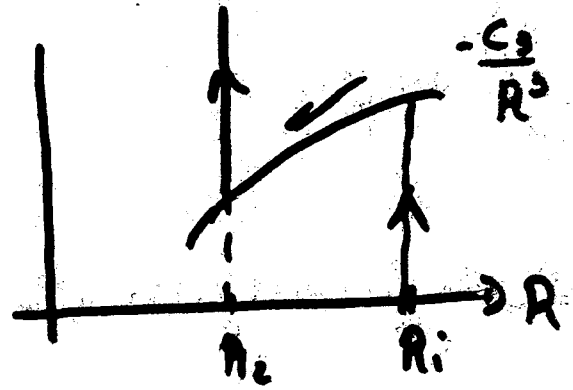
Simple rate calculation:

R_1 → first excitation

R_2 → second absorption

ω_0 → atomic resonance

ω_1, ω_2 → two photons



Rate of excitation (R_1) = $\underbrace{\frac{n^2 4\pi R_1^2 dR_1}{2}}_{\# \text{ pairs}} \left(\frac{I_1}{\hbar \omega_1} \right) \frac{\lambda_1^2}{2\pi} \frac{\uparrow^2}{\uparrow^2 + 4(\Delta_1 - \nu(R_1))^2}$

probabi:

Probability of excitation (R_2) = $\left(\frac{I_2}{\hbar \omega_2} \right) \frac{\lambda_2^2}{2\pi} \frac{\uparrow^2}{\uparrow^2 + 4(\Delta_2 + \nu(R_2))^2} \frac{dR_2}{\sigma(R_2, R_1)}$

Decay going $R_1 \rightarrow R_2$: factor $e^{-\uparrow t^*(R_1, R_2)}$ phase-sp factor.

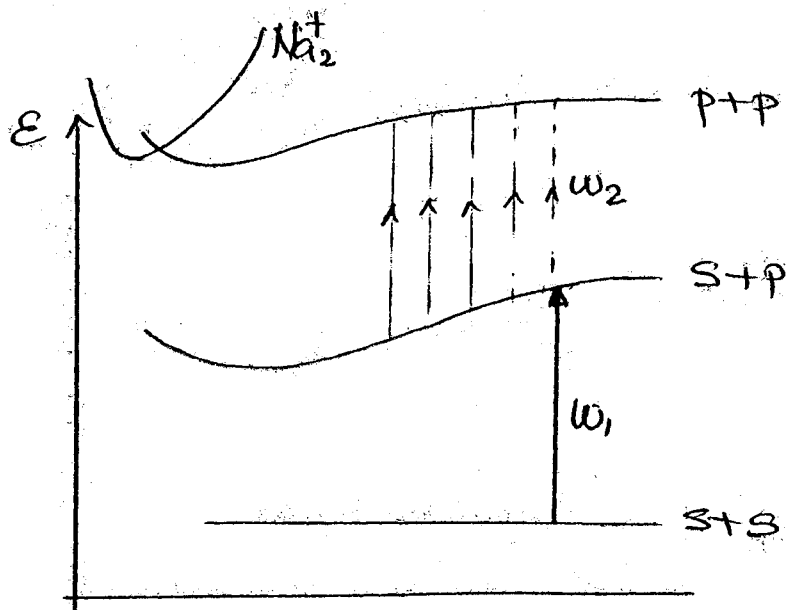
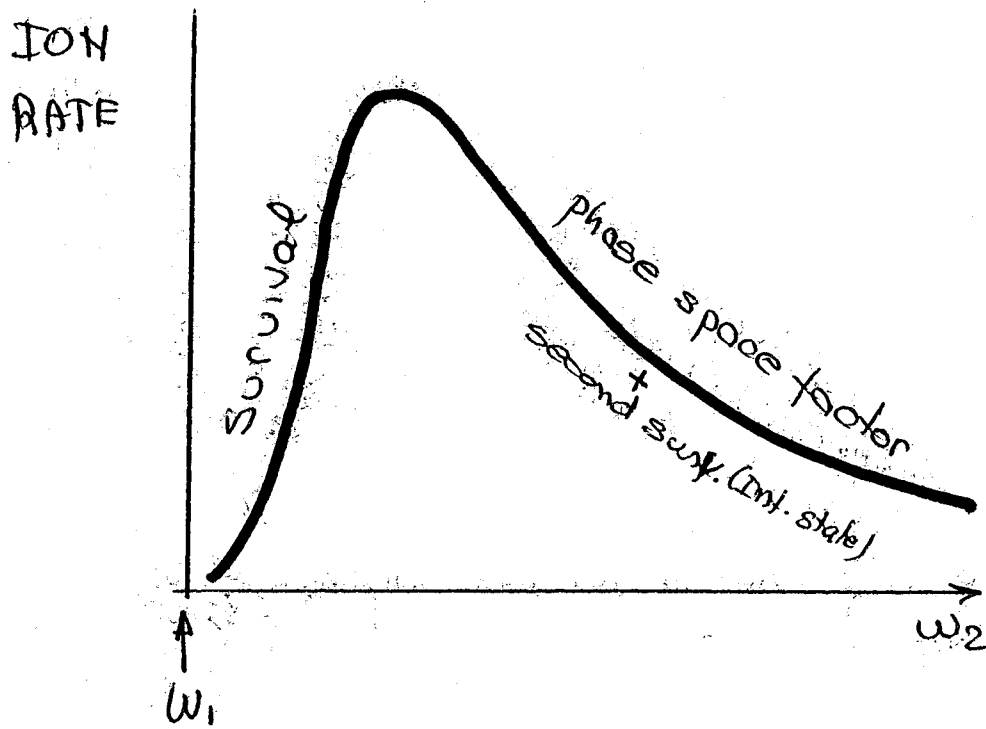
Possibility of oscillation : factor = $\frac{1}{1 - \gamma \int_0^\infty e^{-\tau t^*(R_1, R_2)} d\tau}$

Decay going $R_2 \rightarrow$ short range $e^{-\uparrow t^*(R_2, R_1)}$

K_4 Total rate = \int product.

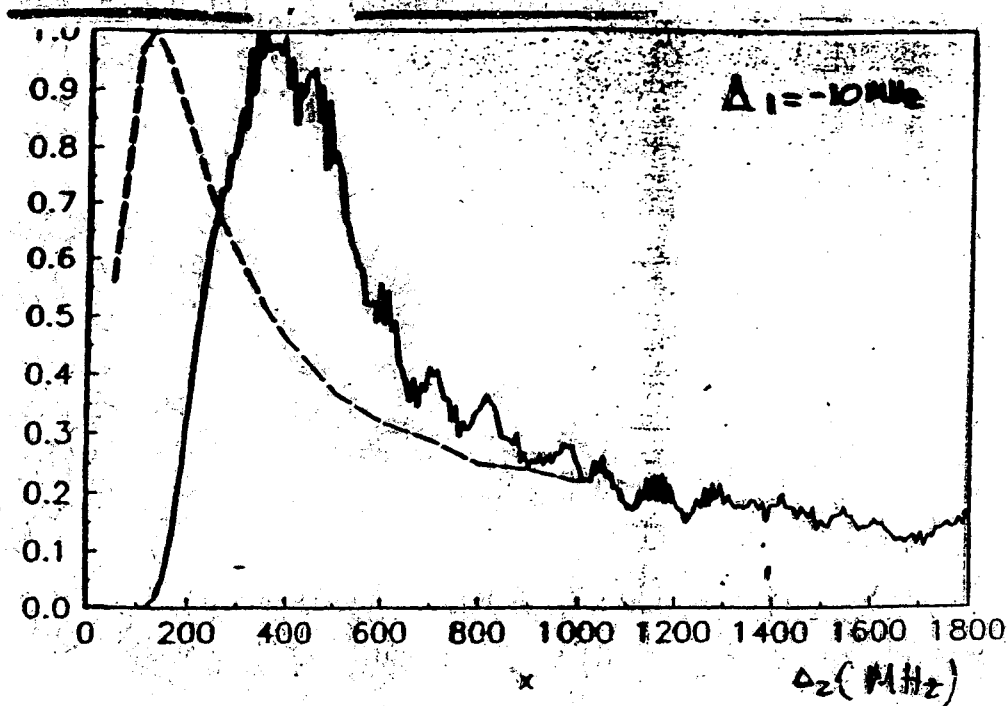
$$K_4 = \frac{n^2}{2} I_1 I_2 \left[\frac{\lambda^2}{\hbar \omega_0} \right]^2 \frac{4\pi}{9} \frac{C_3 \left(\frac{R_N}{R_2} \right)^{5/2}}{\hbar} \times \int_0^\infty d\Delta_i \int_0^\infty d\Delta \frac{(1 - \gamma \tau)^{-1} e^{-\uparrow \tau} e^{-\uparrow \tau \kappa}}{\Delta_i^2 \Delta^{3/2} (\Delta - \Delta_i)^{1/2} [1 + 4(\Delta_1 - \Delta_i)^2] [1 + 4(\Delta_2 + \Delta)^2]}$$

Fix ω_1 and vary ω_2



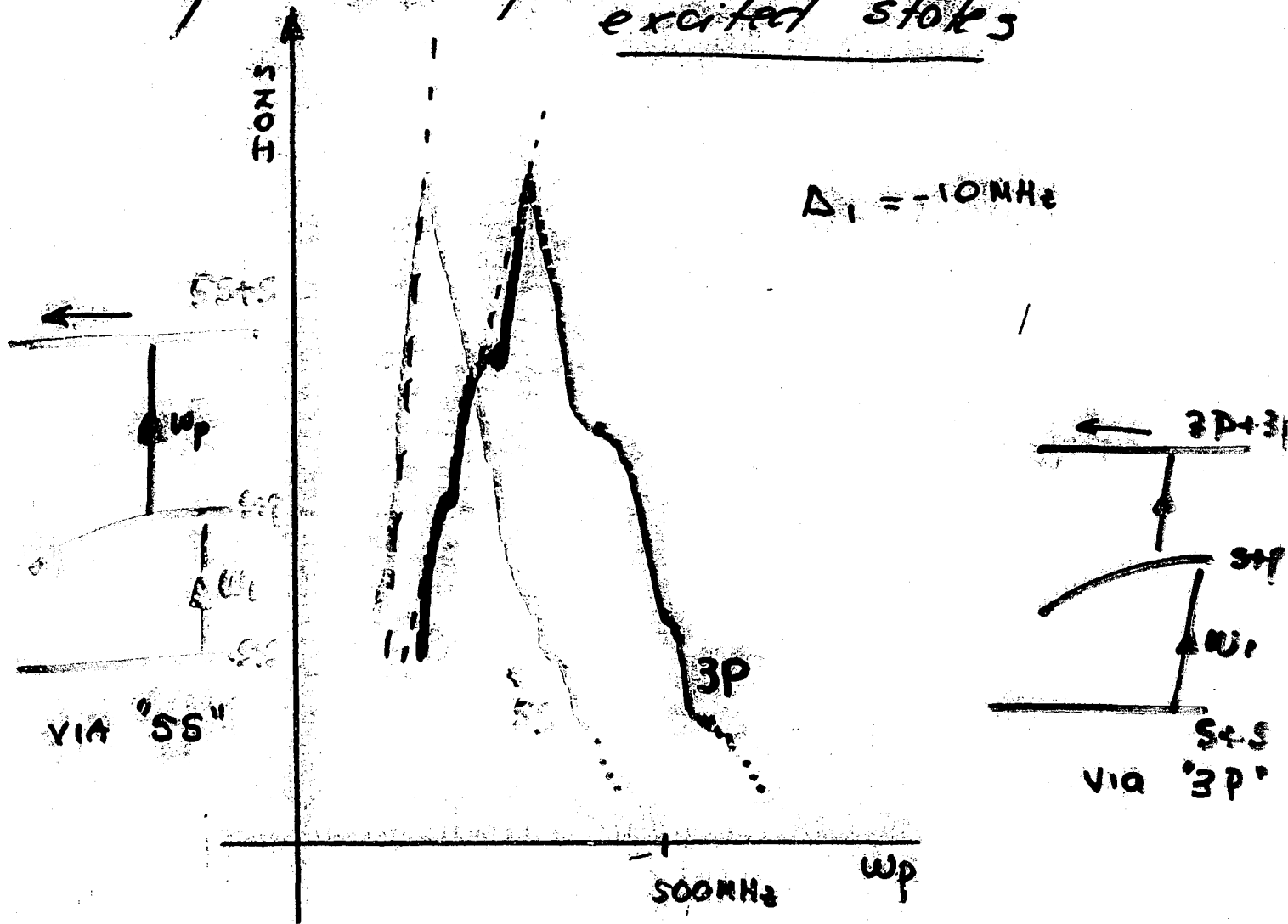
* ω_2 always slower than ω_0

Experiment $\Delta_1 = -10 \text{ MHz}$ (MOT laser)
 $\omega_2 \rightarrow$ probe laser



- * Well explained behavior
- * Shift $\sim 300 \text{ MHz}$ (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

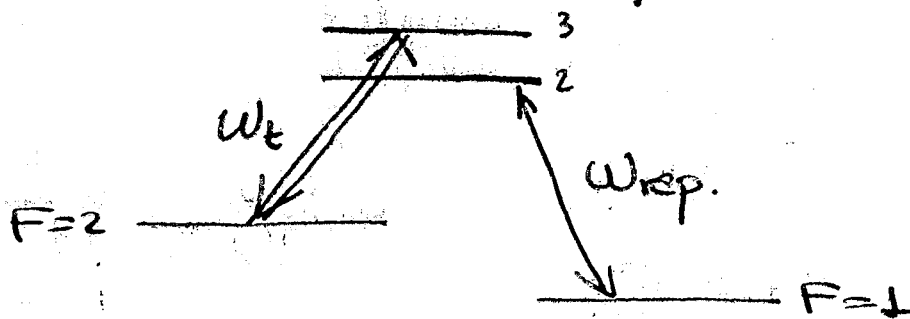
Collisions can start with several possibilities

$(F=1) + (F=1) \leftarrow$ pure

$(F=1) + (F=2) \leftarrow$ mixture

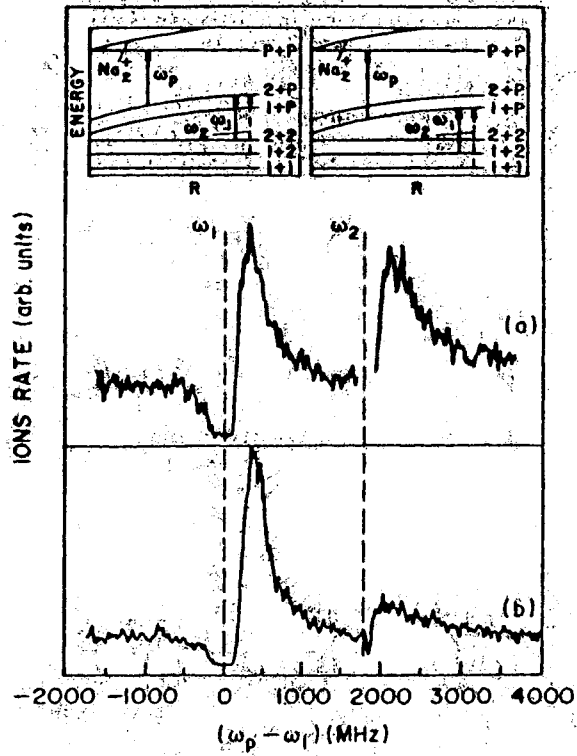
$(F=2) + (F=2) \leftarrow$ pure

To prepare sample.



→ turn off ω_t ($\sim 100 \mu\text{s}$)
 → sample in $F=1$

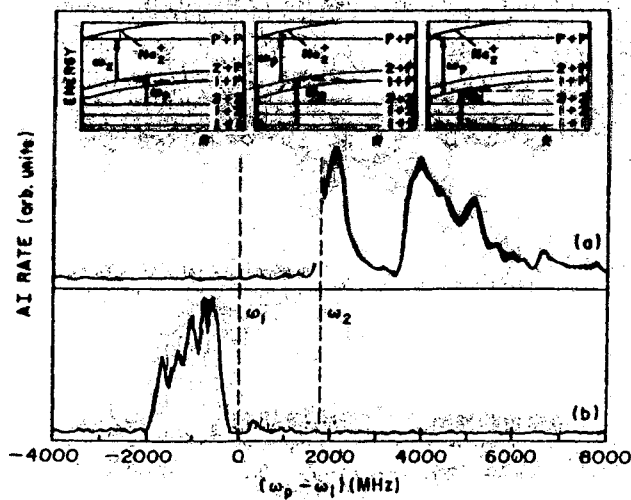
→ turn off ω_{rep} ($\sim 100 \mu\text{s}$)
 → sample in $F=2$



60% F=1
40% F=2

30% F=1
70% 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).



100% F=1

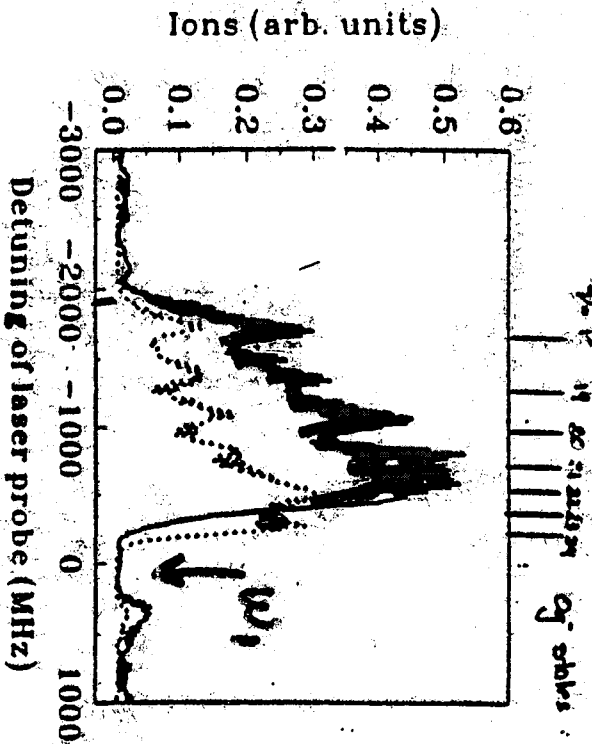
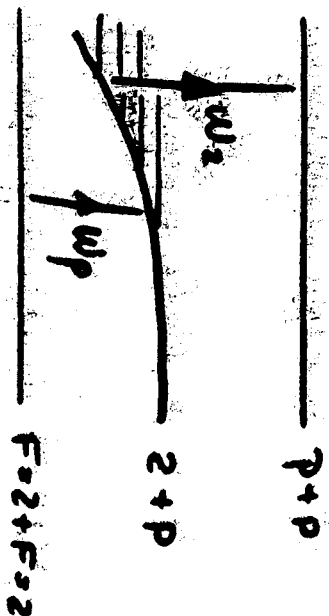
100% F=2



Free to bound

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).

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



Stuall ey
 (PRL - 41, 1164 (79))

$$\partial_0 \cdot \nu = a_3 (\epsilon \cdot D)^{1/2}$$

$$a_3 = 6.25 \text{ MHz}^{-1/2}$$

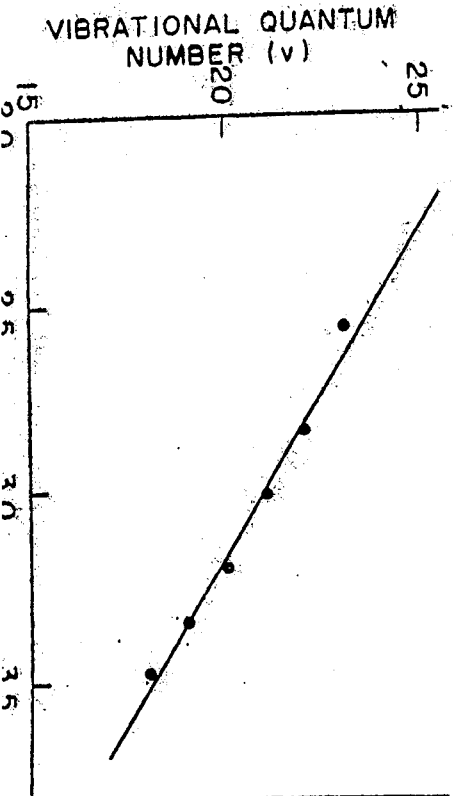
$$D \sim \text{kHz}$$

$$\partial_0 \sim 39.6$$

Theory

Free to bound transition \Rightarrow vibrational long-range state

- very near threshold states



$$a_3 = 6.59$$

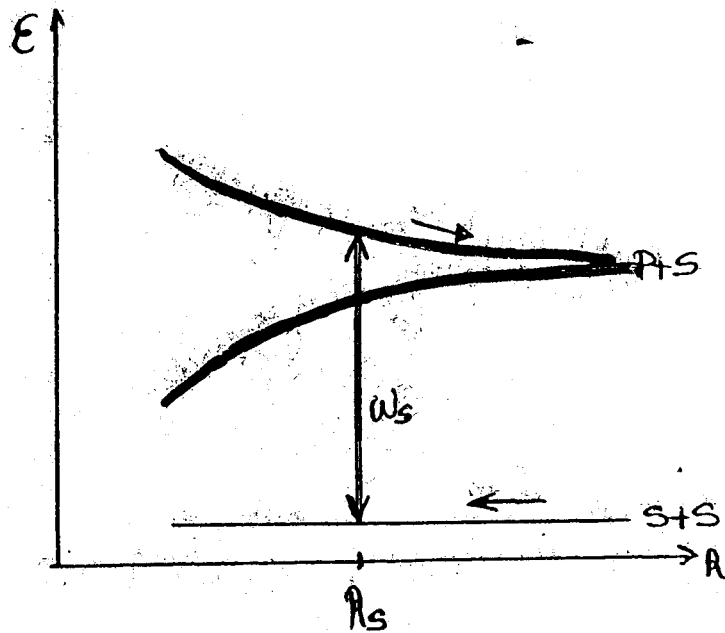
$$\partial_0 = 40.4$$

$$D < 3 \text{ kHz}$$

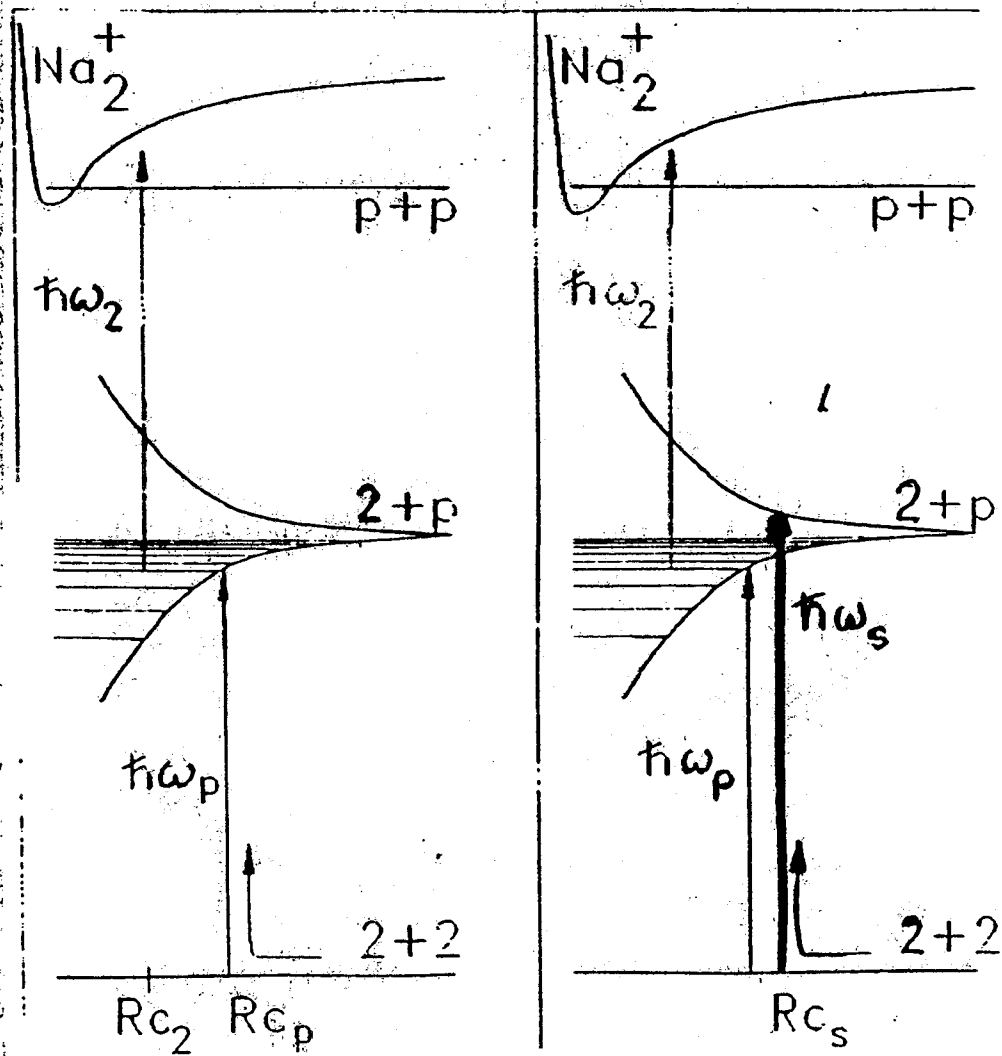
$$(\epsilon \cdot D)^{1/2}$$

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

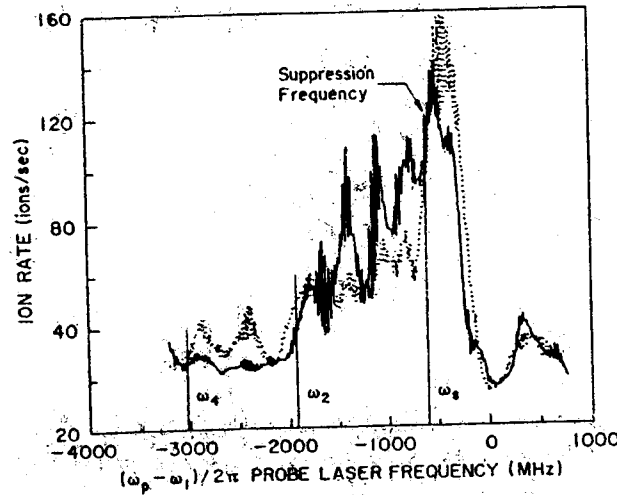


FIG. 77. Effect of optical suppression on the photoassociative ionization spectrum observed in a Na MOT. Solid curve is the photoassociative ionization spectrum without the suppression field ω_3 present. Dotted curve shows the photoassociative ionization spectrum ω_3 present. Note enhancement of signal to the right of ω_3 , 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_3 is present. From Marcassa *et al.* (1994).

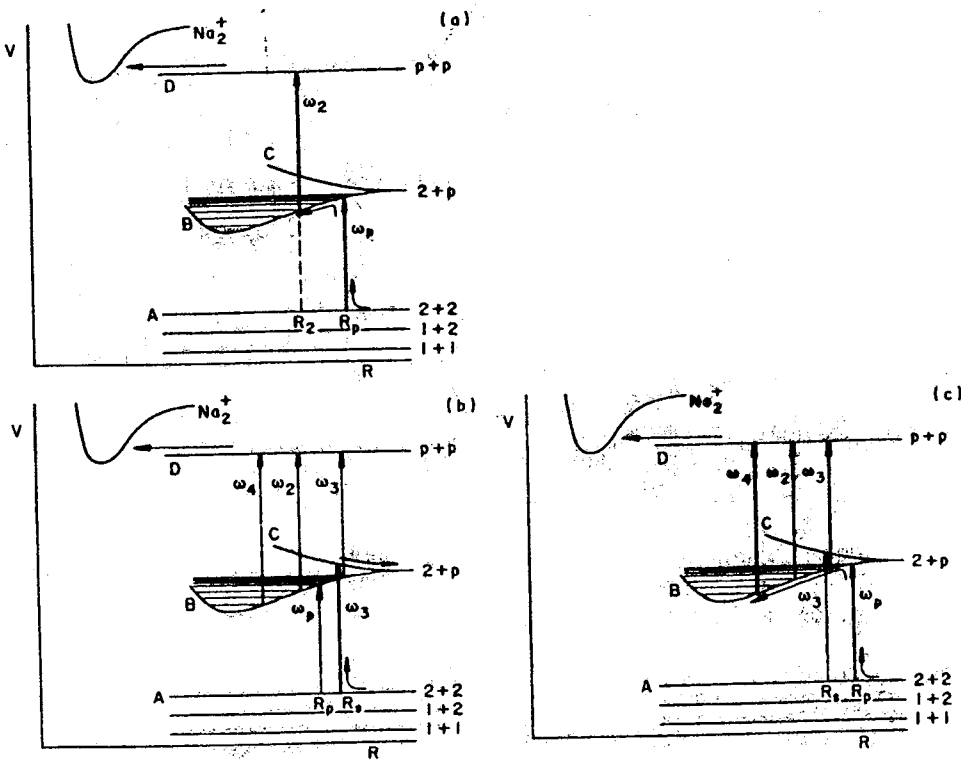
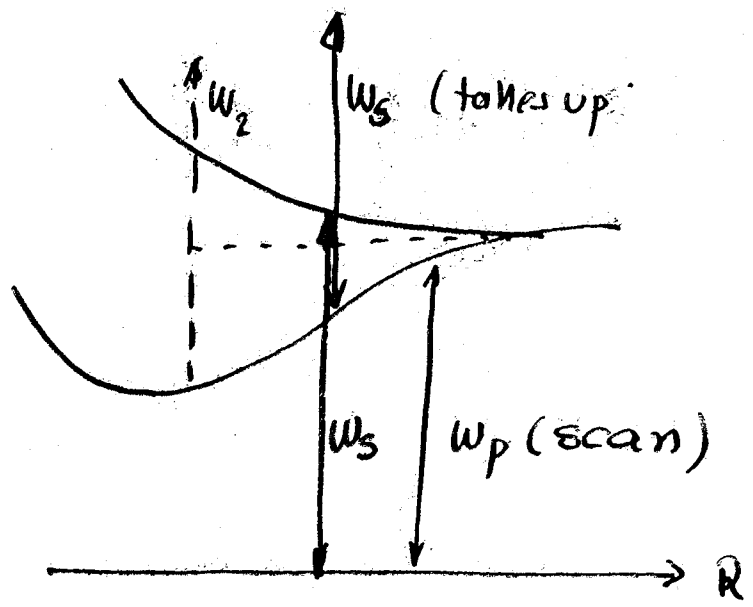
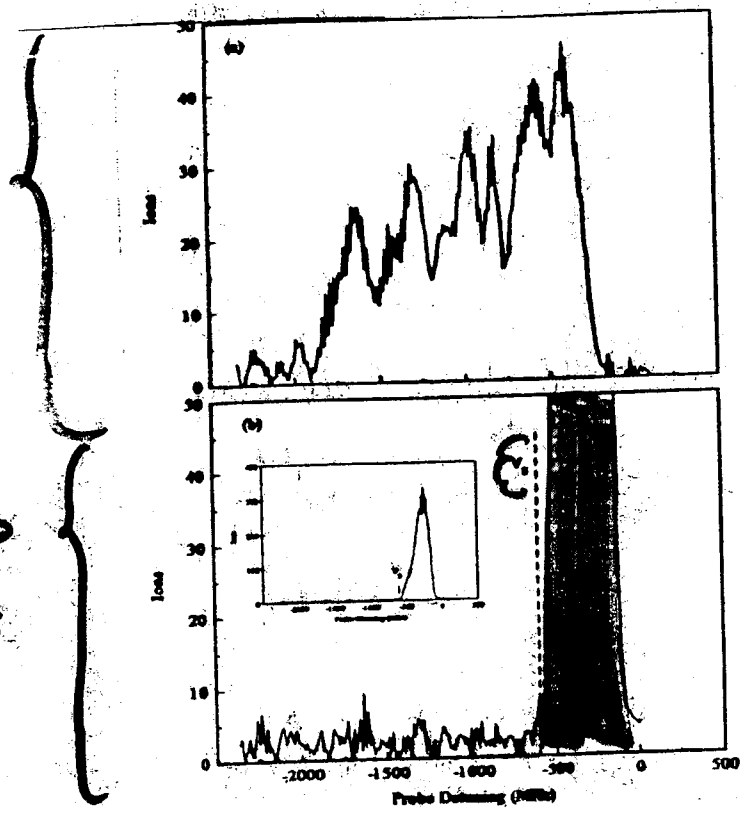


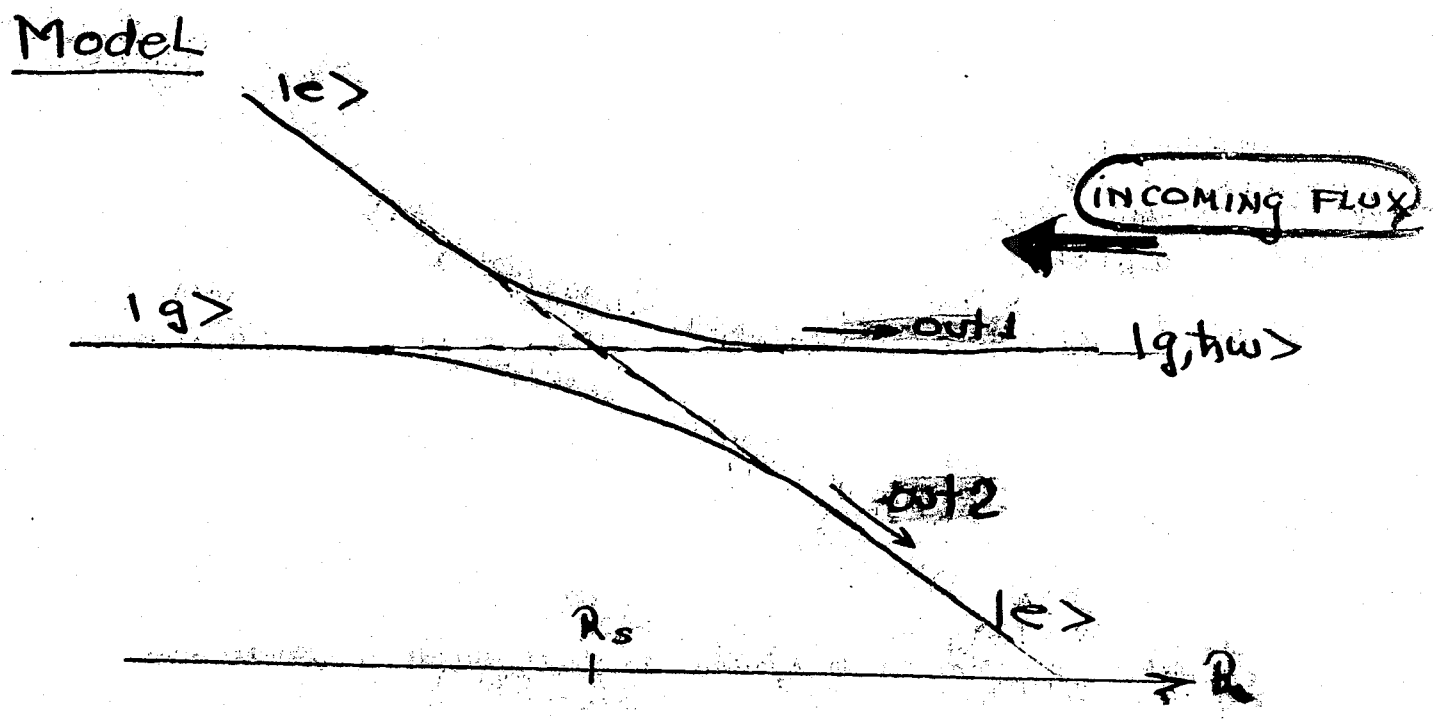
FIG. 76. Schematic of transitions showing photoassociative ionization and optical suppression of photoassociative ionization: (a) Two-step PAI process; (b) suppressor frequency ω_3 imposed on the collision, rerouting incoming flux to the repulsive excited curve; (c) with ω_p tuned to the right of ω_3 , 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.

Espectro Normal

Espectro blindado





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

$$P = 1 - \exp\left\{-\frac{\hbar^2 J^2}{D(R) \cdot V(R)}\right\}$$

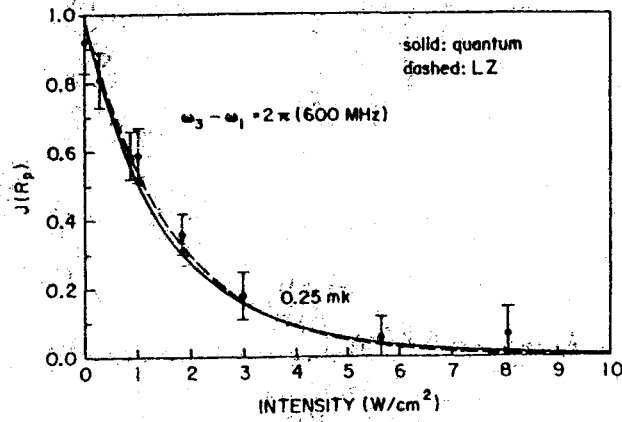
- If excited at $R_s \rightarrow$ revert motion
- If stay $|g\rangle$ with $1-P$ new possibility on the return

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

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

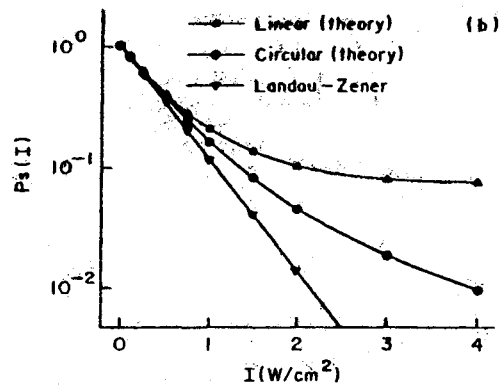
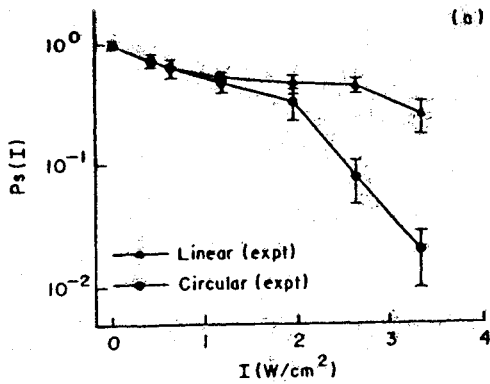
High intensity $\begin{cases} P(\text{out 1}) \rightarrow 1 \\ P(\text{out 2}) \rightarrow 0 \end{cases}$

* Shielding as a function of intensity



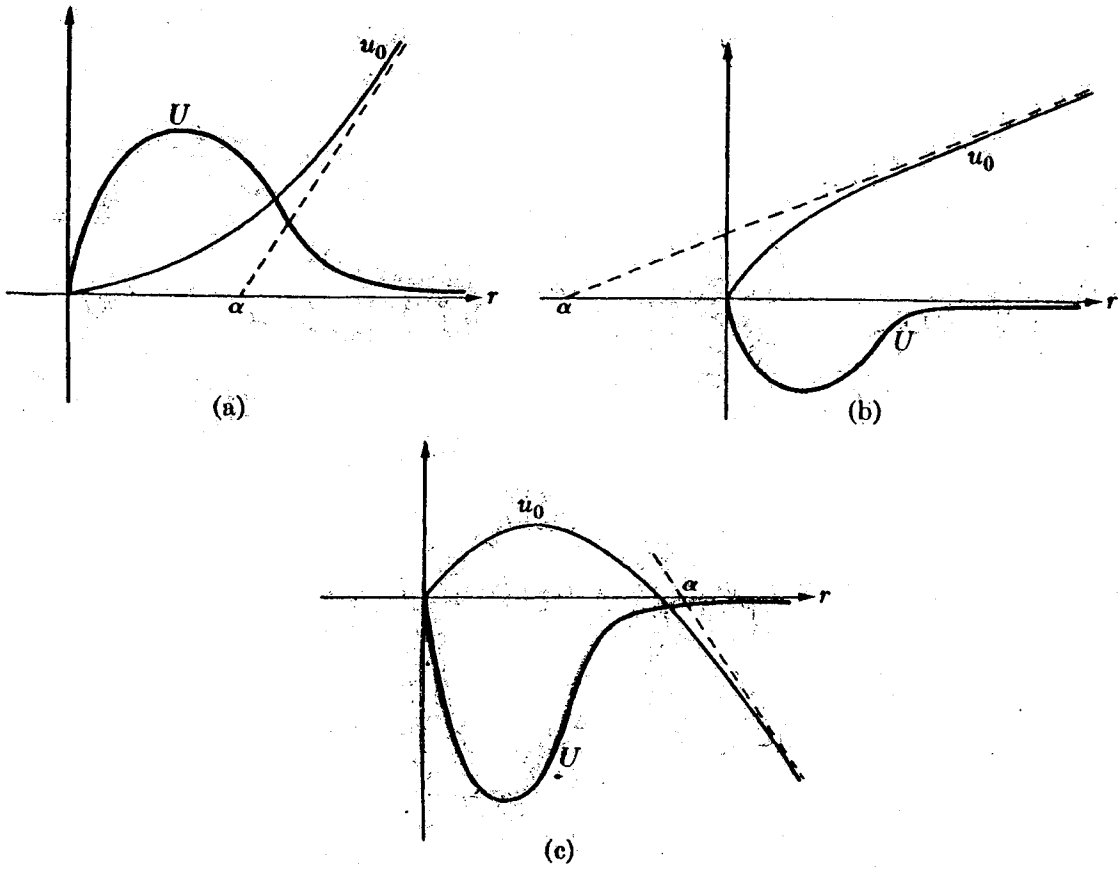
→ Experiment $\int \rightarrow \neq 0$
 for $I \rightarrow \infty$

* Polarization dependence:



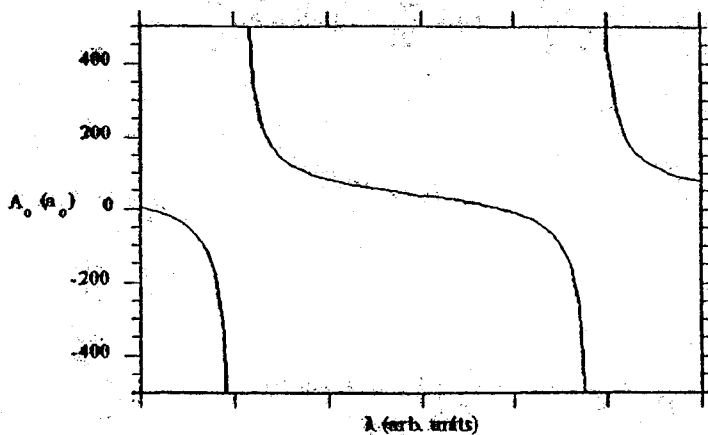
Variation of a with fields

→ Small variation of the potential can change a .

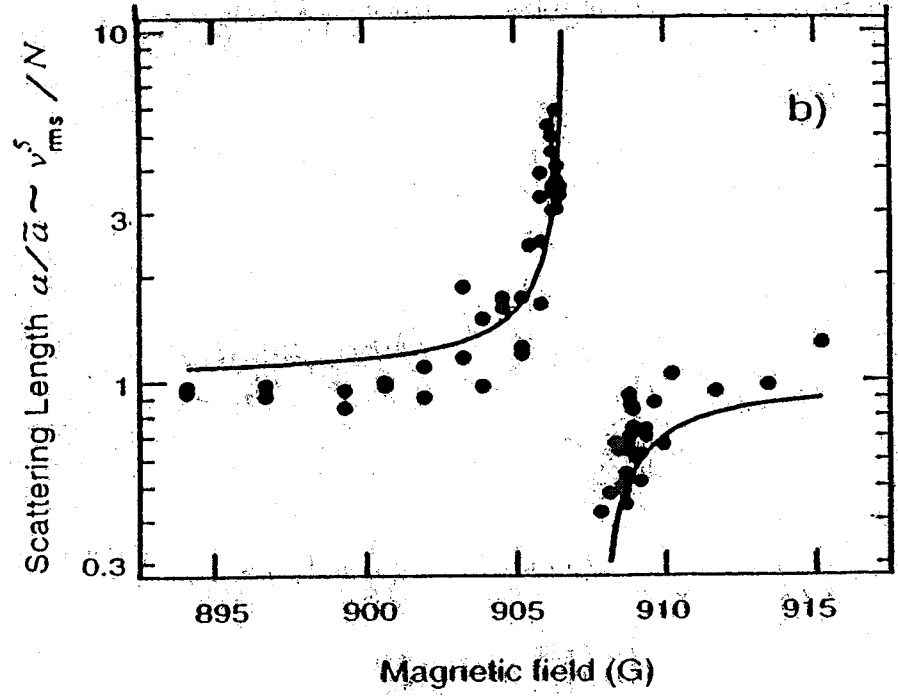


Using B-field (change in hyperfine levels)

Feedback - Resonances

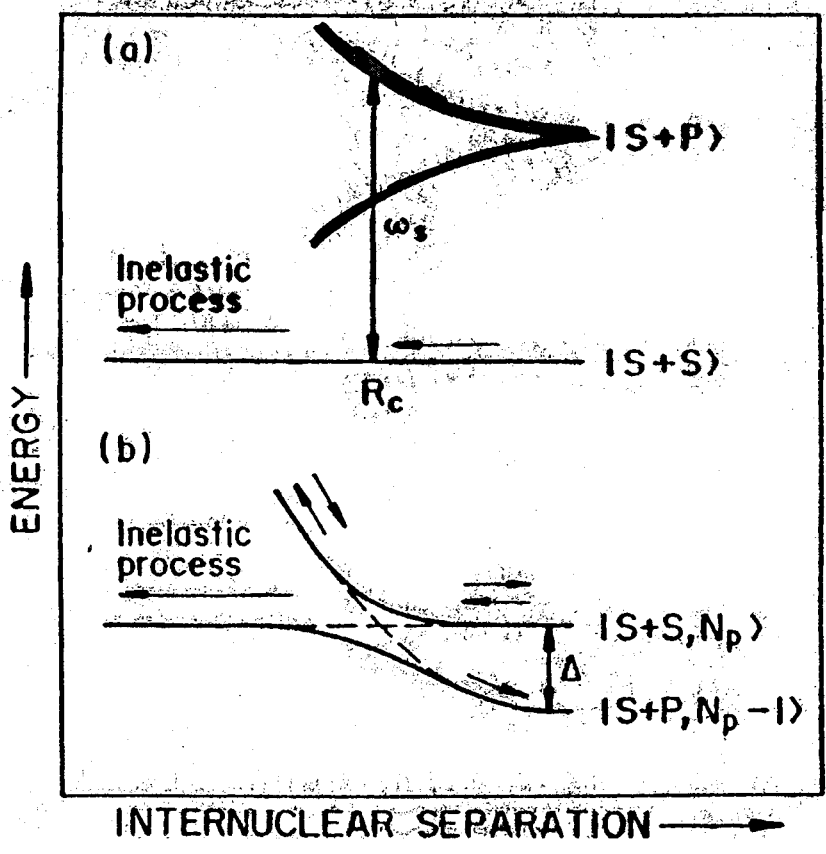


First measurement in Na.



* Using Photons

First we thought in processes like optical shielding



Phys. Rev. Lett. 73, 1911 (1994)
 Phys. Rev. A 52, 1913 (1995)
 Phys. Rev. Lett. 76, 2003 (1996)
 Phys. Rev. A 55, 4407 (1997)
 Science Spect. 7, 50 (1986)

"in" in IS+S

$P(\text{out}(IS+S)) \rightarrow \downarrow$
 High Intensity

Knowing q_0

$$H(p, r) = \frac{p^2}{2M} + U(r) + \int M C(r)$$

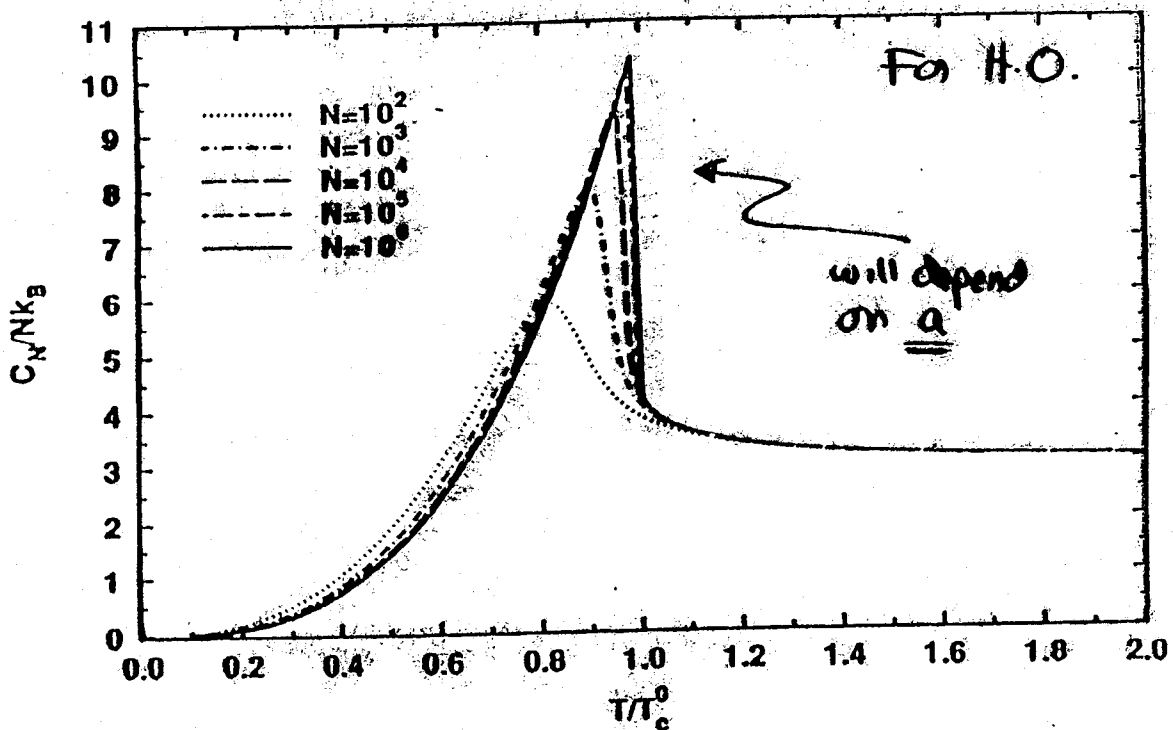
$$V(\vec{r}_1 - \vec{r}_2) = V_0 \delta(\vec{r}_1 - \vec{r}_2) \rightarrow V_0 = \frac{4\pi\hbar^2 a}{M}$$

$$\frac{\text{Energy}}{\text{particle}} = \frac{1}{2} V_0 M C(r) \rightarrow \boxed{f = \frac{2\pi\hbar^2 a}{M}}$$

* Determine new T_c ($\neq T_c^0$)

* Determine new $\frac{N_0}{N}$ [Phys. Rev. A 35 (1987)
Phys. Rev. A 44 (1991)]

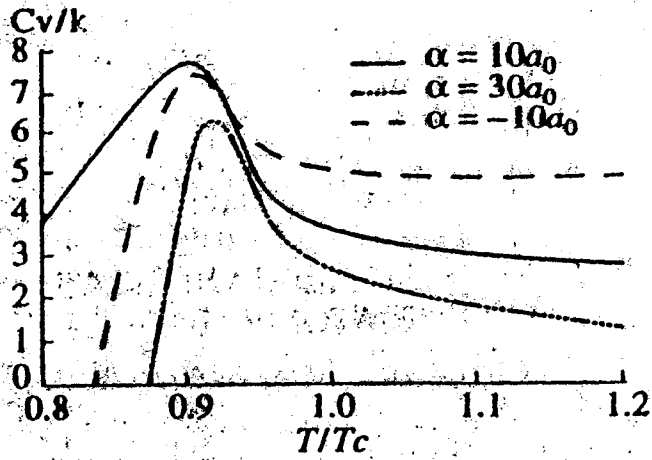
* Heat Capacity: $C = \left(\frac{\partial E}{\partial T} \right)_N$



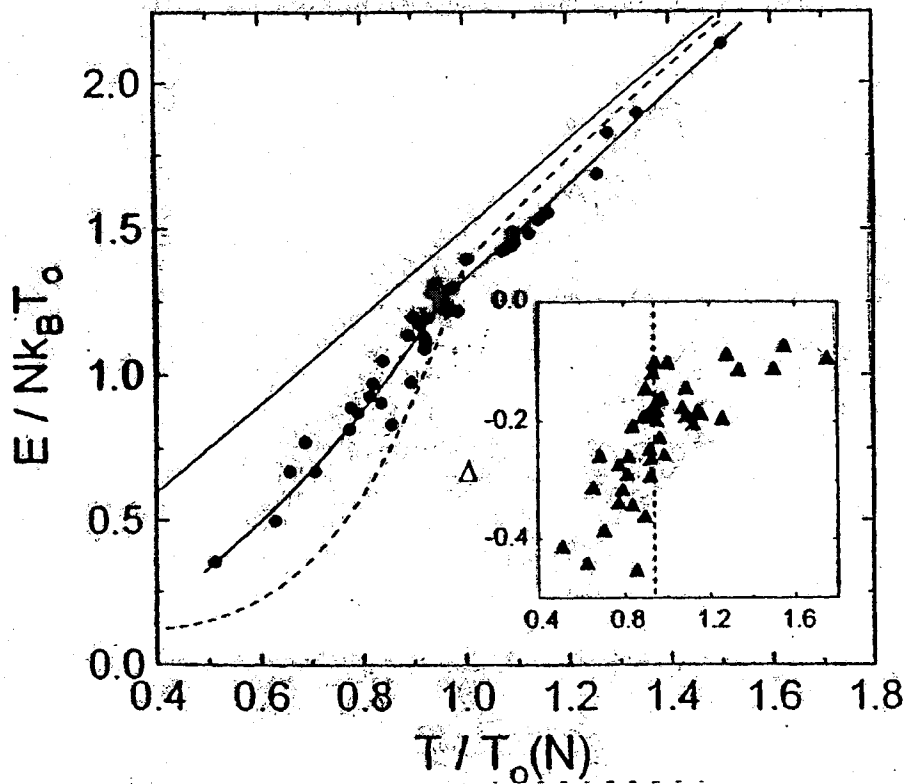
[Phys. Rev. A 55, 3954 (1997)]

neutron interaction

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



Lack of data for heat capacity.



Conell & Wieman

em colisões frias

- Marcossa et al - Phys. Rev. A, R4563 (1993)
- Santos et al - Phys. Rev. A, S2, R4340 (1995)
- Bagnato et al - Los. Phys. 4, 1062 (1994)
- Bagnato et al - Phys. Rev. Lett. 70, 3225 (1993)
- Bagnato et al - Phys. Rev. A, R2523 (1993)
- Napolitano et al - Phys. Rev. Lett 73, 1352 (1994)
- Marcossa et al - Phys. Rev. Lett 73, 1911 (1994)
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