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WINTER COLLEGE ON LASERS, ATOMIC AND MOLECULAR PHYSICS

(21 January - 22 March 1985)

SPIN-POLARIZED HYDROGEN

(Lectures 2, 3 and 4)

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Kleppner - H↓

Lecture 2

Some historical background on H↓
(see Greytak & Kleppner for a more complete account)

C. Hecht - 1959
Short note in *Physica* predicting that H↓ is a gas at T=0. However, no experimental techniques were then possible, and the note was overlooked until after H↓ was created.

Etters, Dugan & Palmer
Miller, Nosanow & Parrish } 1975
Careful studies of equation of state of H↓.

Stwalley & Nosanow - 1976
Comment in Phys. Rev. Lett. pointing out potential interest in H↓, and suggesting that somebody do something.

Silvera & Walraven - 1980
First creation of H↓. Produced a few months later at MIT.

1985 - H↓ under study by at least 10 experimental groups, and many theorists.

Spin-Polarized Hydrogen

Experimental work

- Amsterdam   Silvera & Walraven
- M.I.T.
- U. of British Columbia !
- Cornell U.
- U. of Turku
- CERN
- SNCI - Grenoble
- Harvard U.
- Los Alamos
- Brookhaven

Work at MIT

T. J. Greytak
H. F. Hess
D. A. Bell
G. P. Kochanski
D. Kleppner

Primary goal of the field - B-E transition

$$n_0 = 2.621\left(\frac{2\pi M k T}{h^2}\right)^{3/2} \xrightarrow[T=0.3K]{} 8\times10^{19}\ \text{atoms cm}^{-3}$$

Problems

* System is metastable - atoms would much rather be molecules.
  (Note: energy per unit mass of H↓ approximately 100x greater than dynamite!)
* Normal H recombines very rapidly

  $$H + H + H \rightarrow H_2 + H + 4.8\,eV.$$

  3-body lifetime: $\dot{n} = -L' n^3$

  $$\tau_3 \sim \frac{-n}{\dot{n}} = \frac{1}{L' n^2} \xrightarrow[n=10^{19}\,cm^{-3}]{} 10^{-2}\,s.$$

  Recombination on surface even faster.

  $$H + H + \text{surface} \rightarrow H_2 + \text{surface} + 4.8\,eV$$

* High densities required - some way must be found to compress the gas.

<u>Basic approach</u> - use a "super Stern Gerlach" effect - work in regime where magnetic energy >> thermal energy.

EXPERIMENTAL STEPS

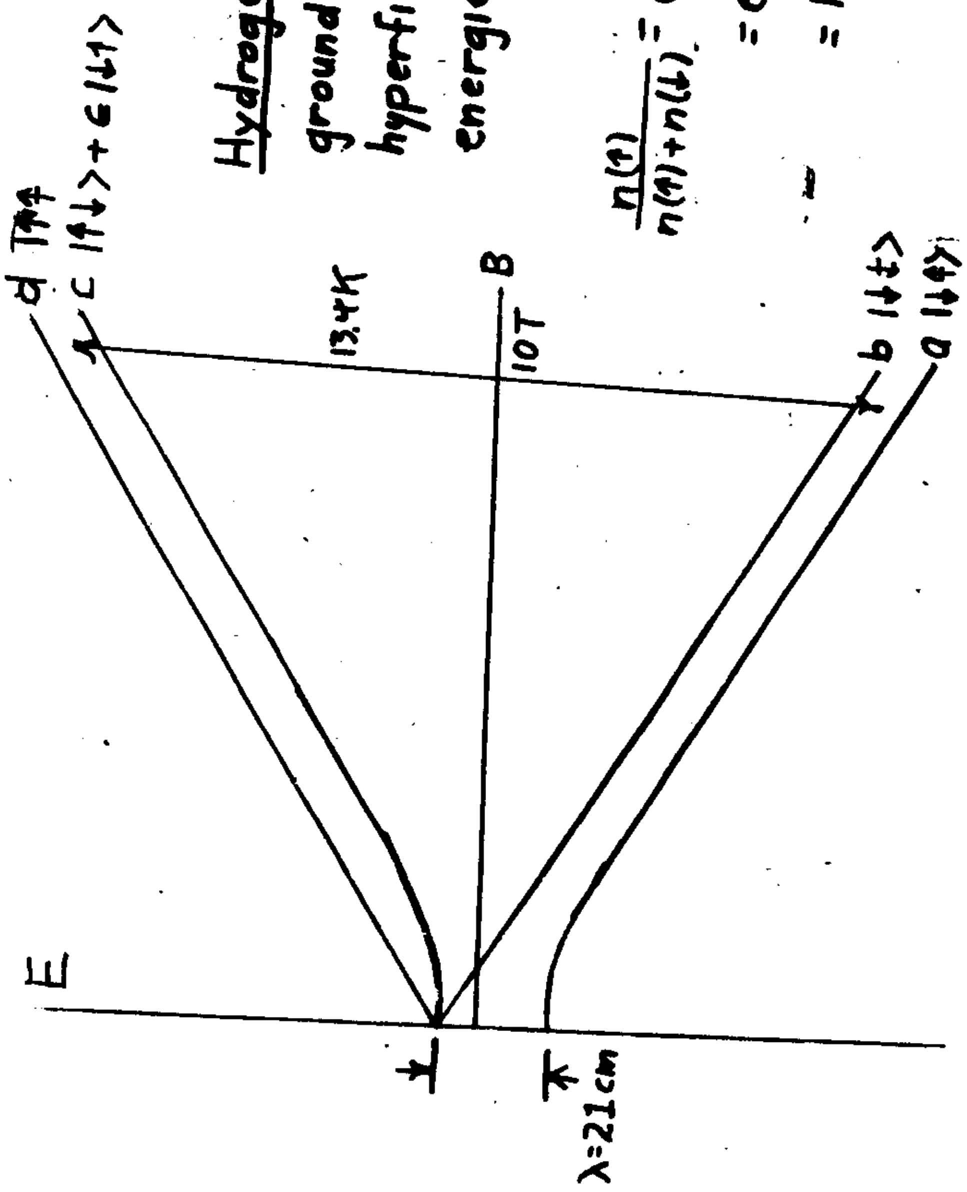
DISSOCIATE $H_2 \rightarrow H + H$

COOL $H \rightarrow H$

SPIN SELECT $H\uparrow$, ~~$H\downarrow$~~

STABILIZE $n\downarrow / n\uparrow \sim 10^{-19}$

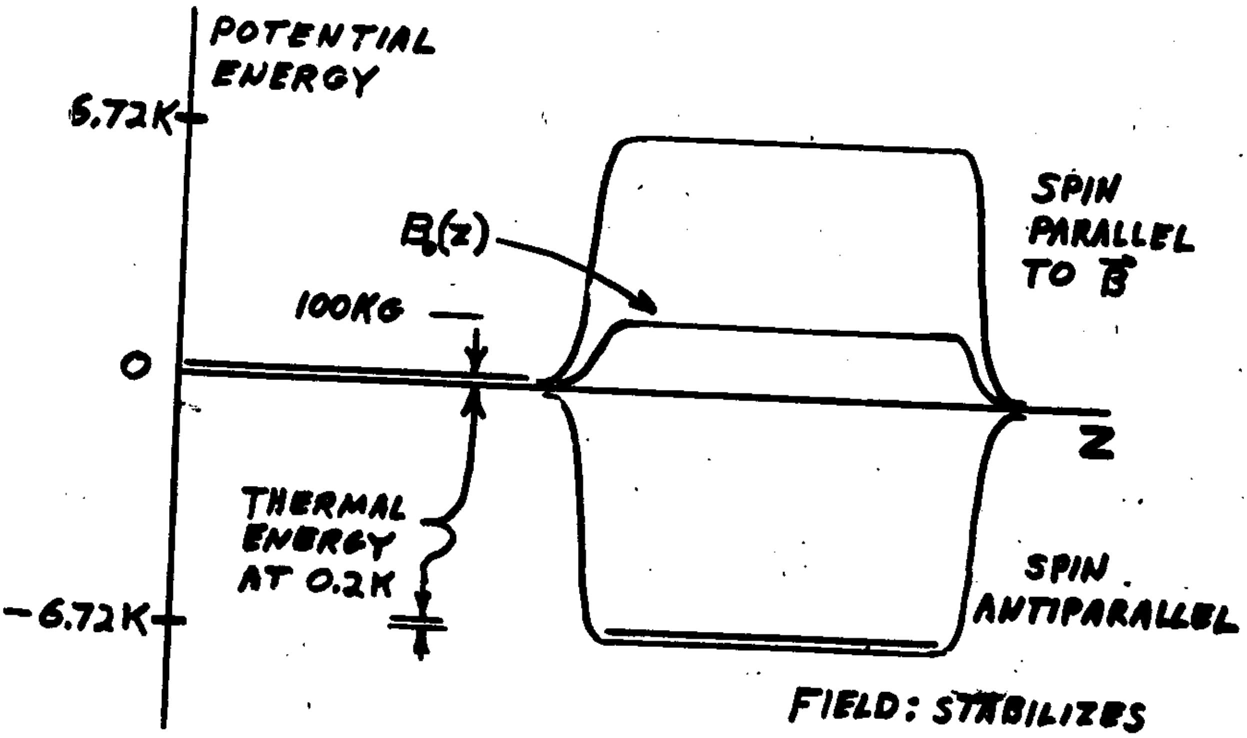
CONFINE ⫽ $H\uparrow$ ⫽



Hydrogen ground state hyperfine energies

$$\frac{n(\uparrow)}{n(\uparrow)+n(\downarrow)} = e^{-\frac{\Delta W}{kT}} = e^{-\frac{13.4K}{0.3K}} = 10^{-20}$$

TEMPERATURE	n^+/n^- AT 100 KG	$n_{B.E.}$	SEPARATI-
1.0 K	1.4×10^{-6}	$5.0\times10^{20}\ CM^{-3}$	13 Å
0.5	2.1×10^{-12}	1.8×10^{20}	18
0.3	3.6×10^{-20}	8.2×10^{19}	23
0.2	6.7×10^{-30}	4.4×10^{19}	28



FIELD: STABILIZES
SPIN SELECTS
CONFINES

Major experimental tools needed-

- High magnetic field - superconducting magnets became widely available in 1950's.
- Low temperature (≤0.3K) refrigerator - ^3He-^4He dilution refrigerators became available in 1960's & 70's.

New techniques which had to be developed

- Method for cooling H to 0.3K
- Inert surfaces to prevent H from sticking to the walls

Preliminary experiment:

- attempt to cool H in abundant quantities to liquid He regime (~5K)
- Method - Helium temperature hydrogen hyperfine resonance apparatus.

Wall coating: frozen H_2

Hyperfine resonance of H in H_2-lined bulb at T = 5 K.

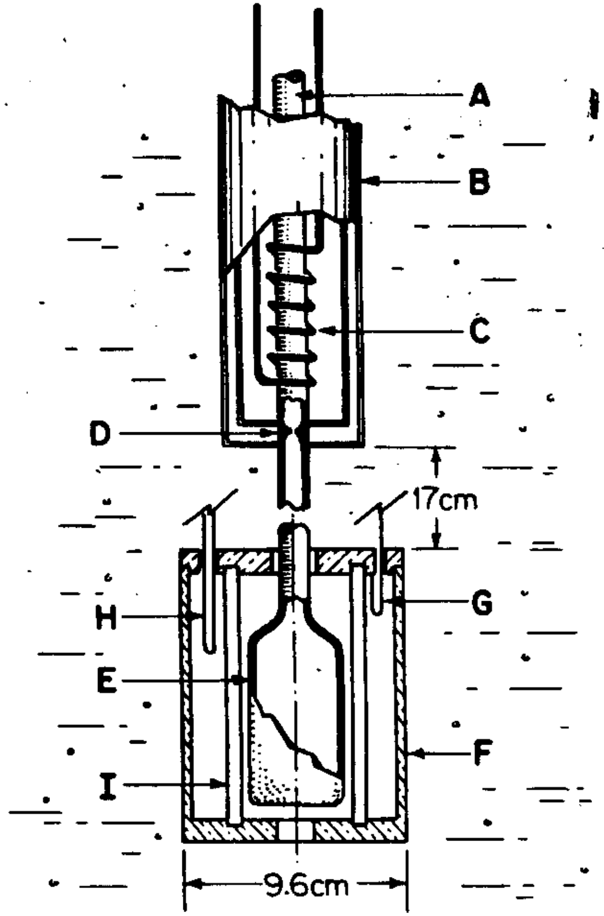


Fig. 6.1

Results:

a) H can be transported easily at 5K in H_2-lined apparatus. Hundreds of wall collisions possible.

b) Hyperfine frequency shifted by surface collisions.

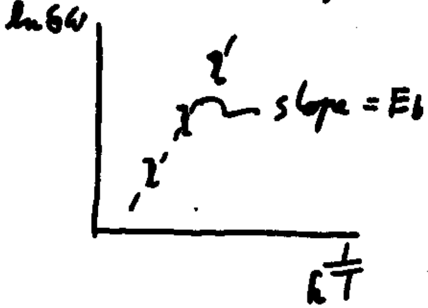
c) From b), adsorption energy of H on H_2 found to be ~ 35K. This precludes use of H_2 at low temperature, <1K.

Explanation: While atoms are on surface, hyperfine frequency is shifted by $\Delta\omega_s$.

Adsorption time on surface, τ_a, can be assumed to be short compared to mean time between surface collisions.

$$\omega(\text{observed}) \cong \omega_0 + \Delta\omega_s \frac{\tau_a}{\tau_c} = \omega_0 + \delta\omega.$$

$$\delta\omega = \text{"wall shift"} \sim \tau_a = \tau_0 e^{E_b/kT}$$ ← adsorption energy.



Implications:

at 5K, $\tau_a \sim 10^{-9}$ sec.

at 1K, $$\tau_a(1K) = \tau_a(5K) \times \frac{e^{35K/1K}}{e^{35K/5K}} = \tau_a(5K) \times e^{28}$$

Atom will simply stick to surface!

This is confirmed in practice. As apparatus is cooled below 5K, H rapidly disappears.

Next step:

Only remaining candidate for surface coating: ^4He (or ^3He)

Requires operating below 0.5K.

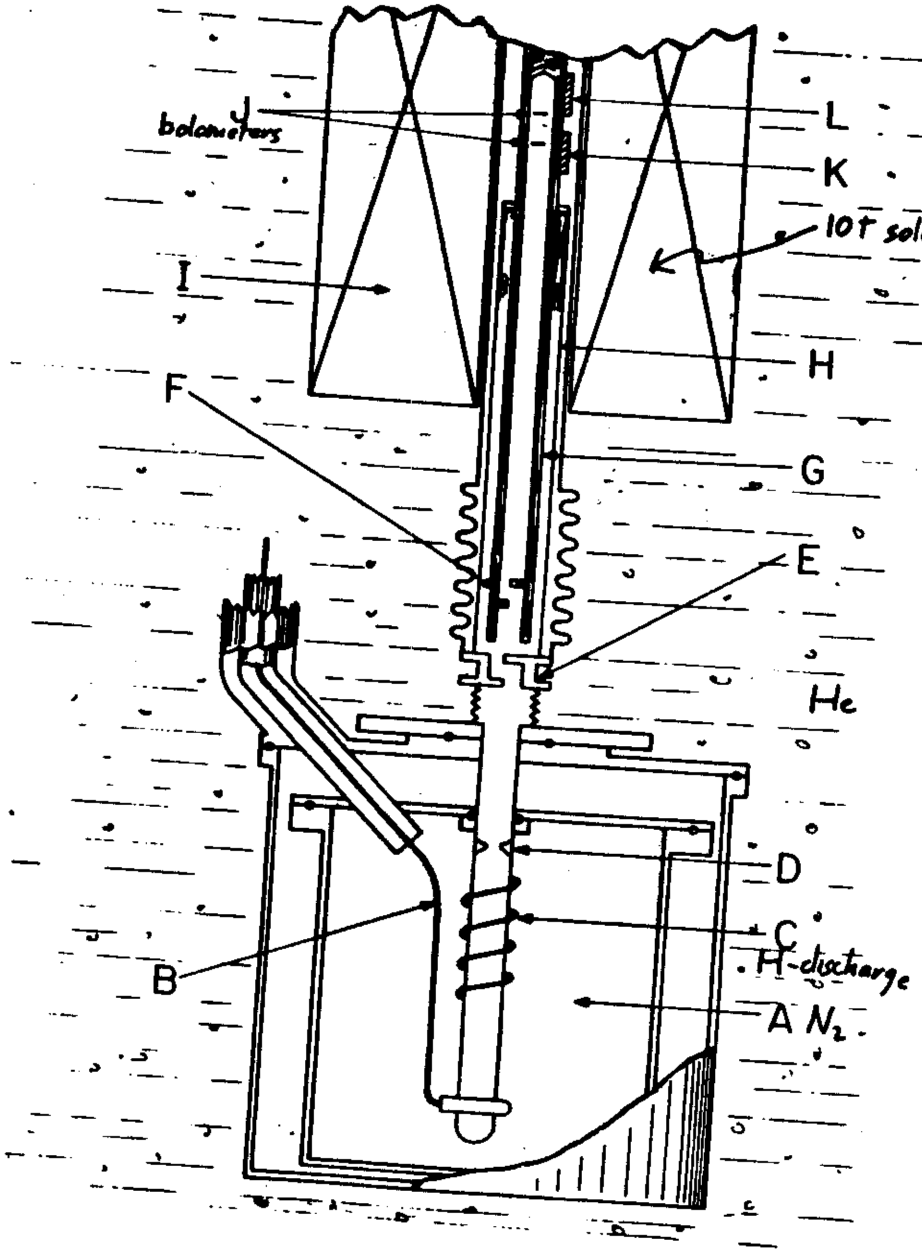
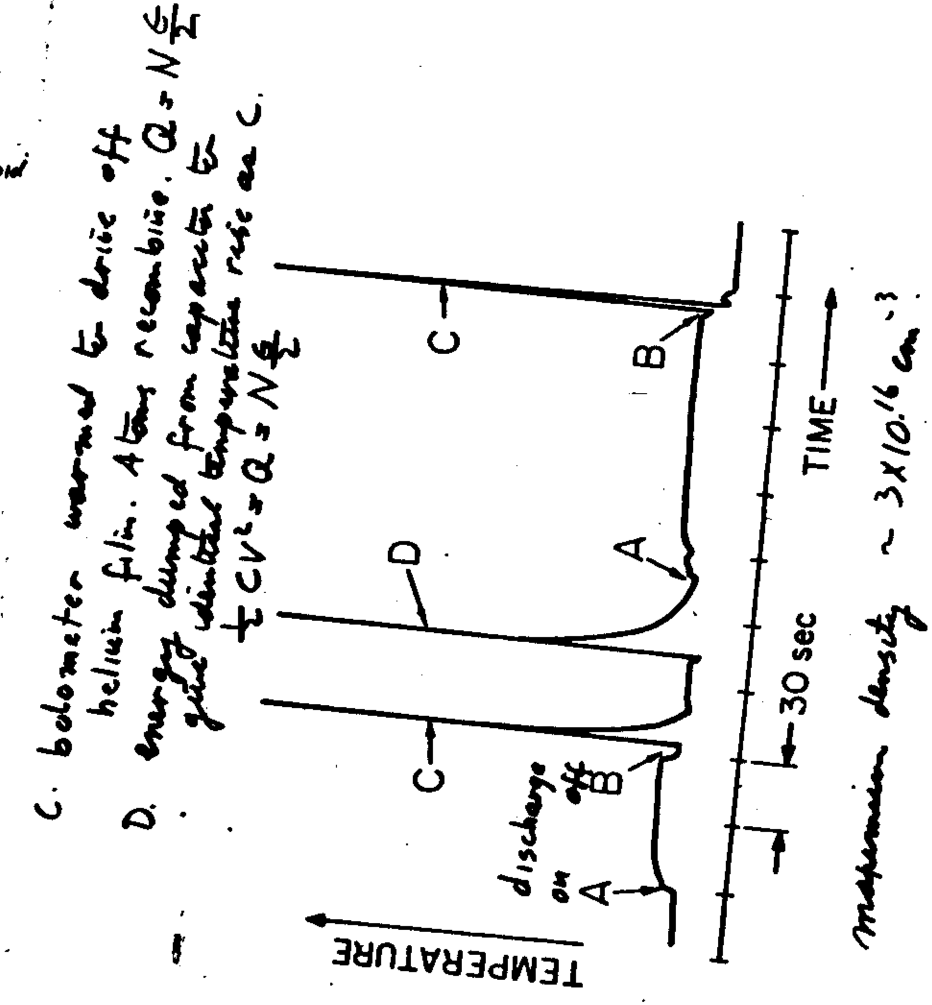

bolometers
J
L
K
10T solenoid
I
H
F
G
E
He
D
C
H-discharge
B
A N_2

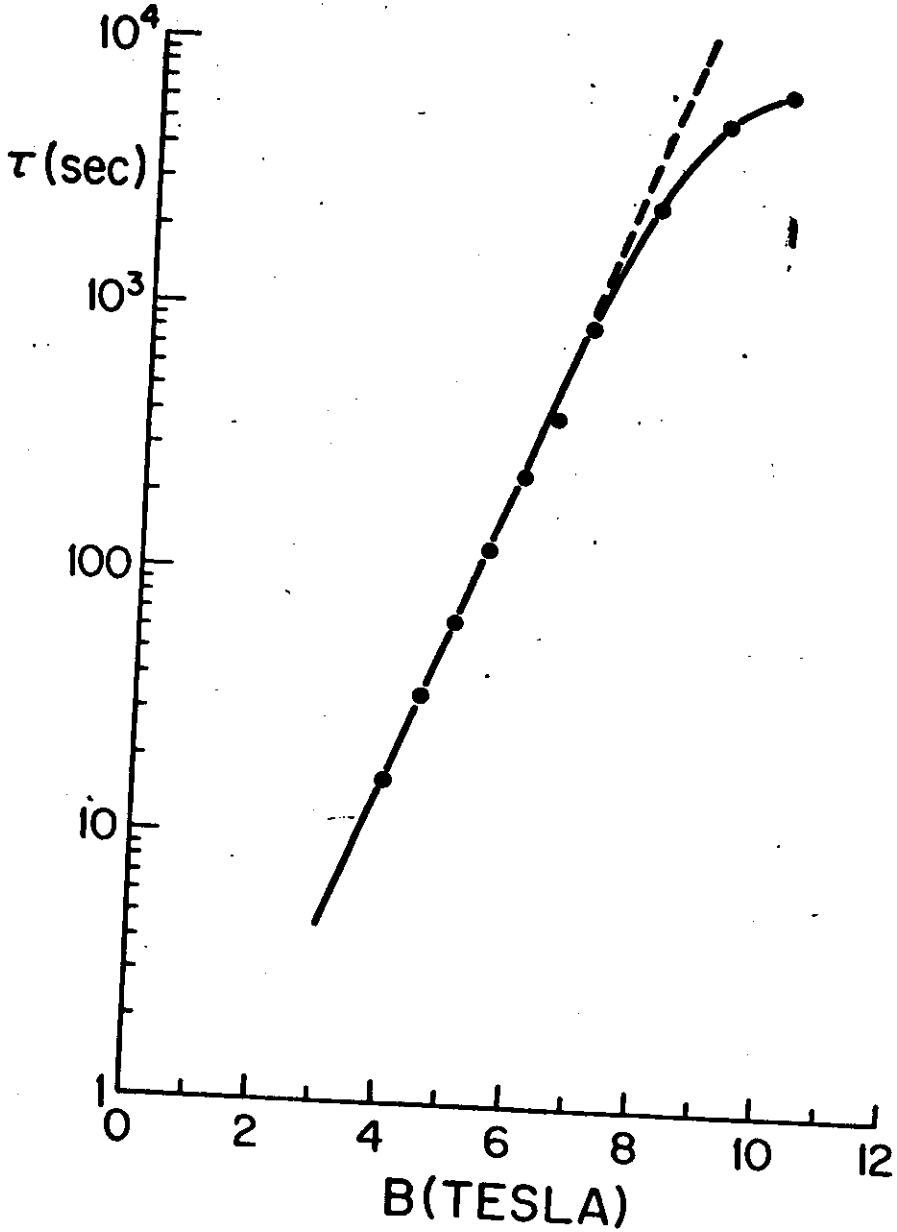
Fig. 7.2

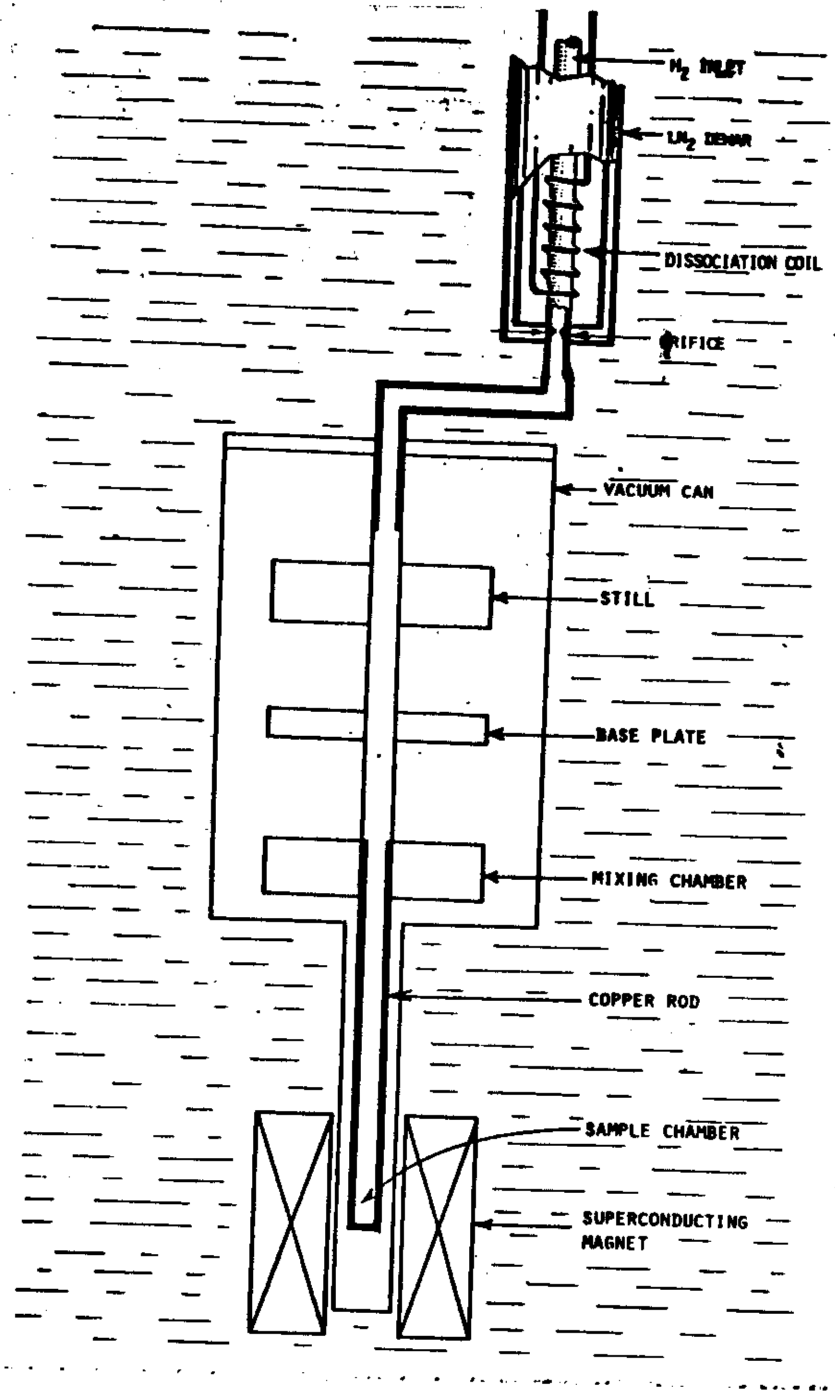

C. bolometer warmed to drive off helium film. Atoms recombine. $Q = N\frac{\epsilon}{2}$
D. energy dumped from capacitor to give identical temperature rise as C. $\frac{1}{2}CV^2 = Q = N\frac{\epsilon}{2}$
TEMPERATURE
TIME
30 sec
discharge on
discharge off
A
B
C
D
maximum density $\sim 3\times10^{16}\ cm^{-3}$

If magnetically confined, the confinement time is given by

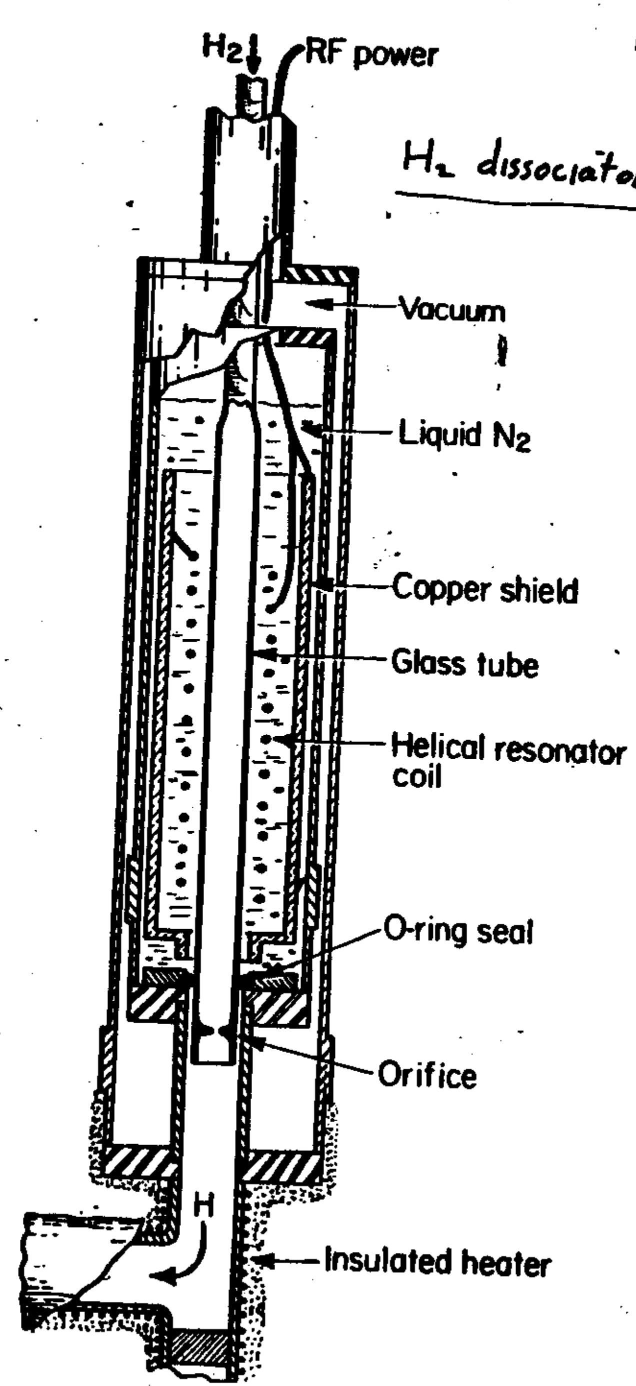
$$\tau = \tau_0 e^{\mu B/kT}$$

To check whether the H↑ is magnetically confined we then measure the decay time as a function of magnetic field.

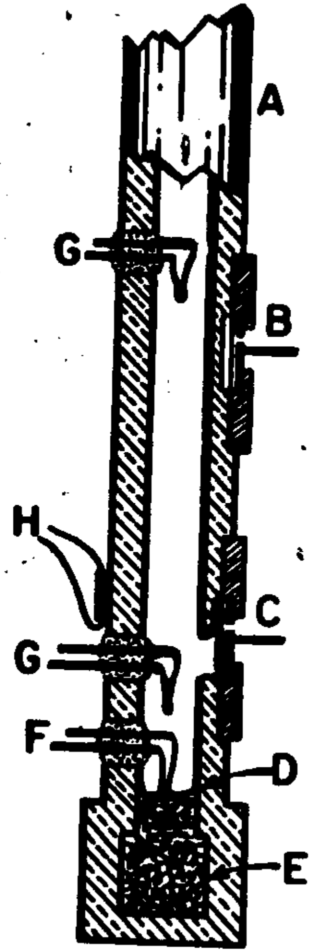




H_2 dissociator



A) Copper rod
B) Reference capacitor
C) Capacitive pressure transducer
D) Liquid ^{4}He pool
E) Sintered copper
F) Level sensor
G) Bolometer
H) Thermometer



A good way to find out what is happening to H↓:
- study how it decays

Types of decay

"single body", or first order

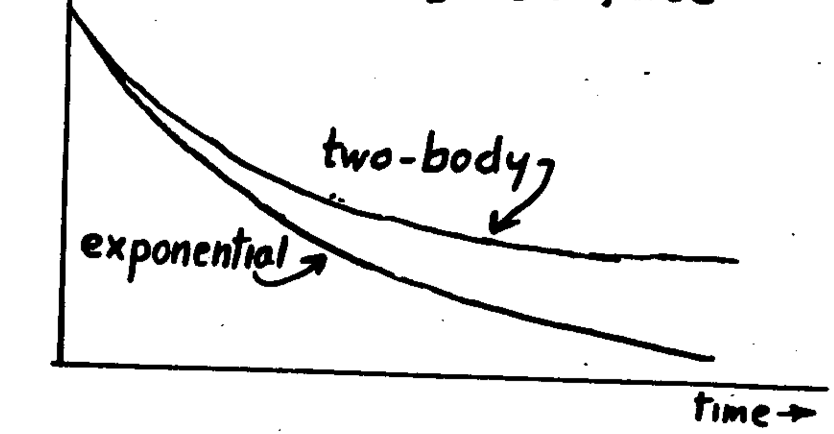
$\dot{n} = -\gamma n$, $\boxed{n = n_0 e^{-\gamma t}}$

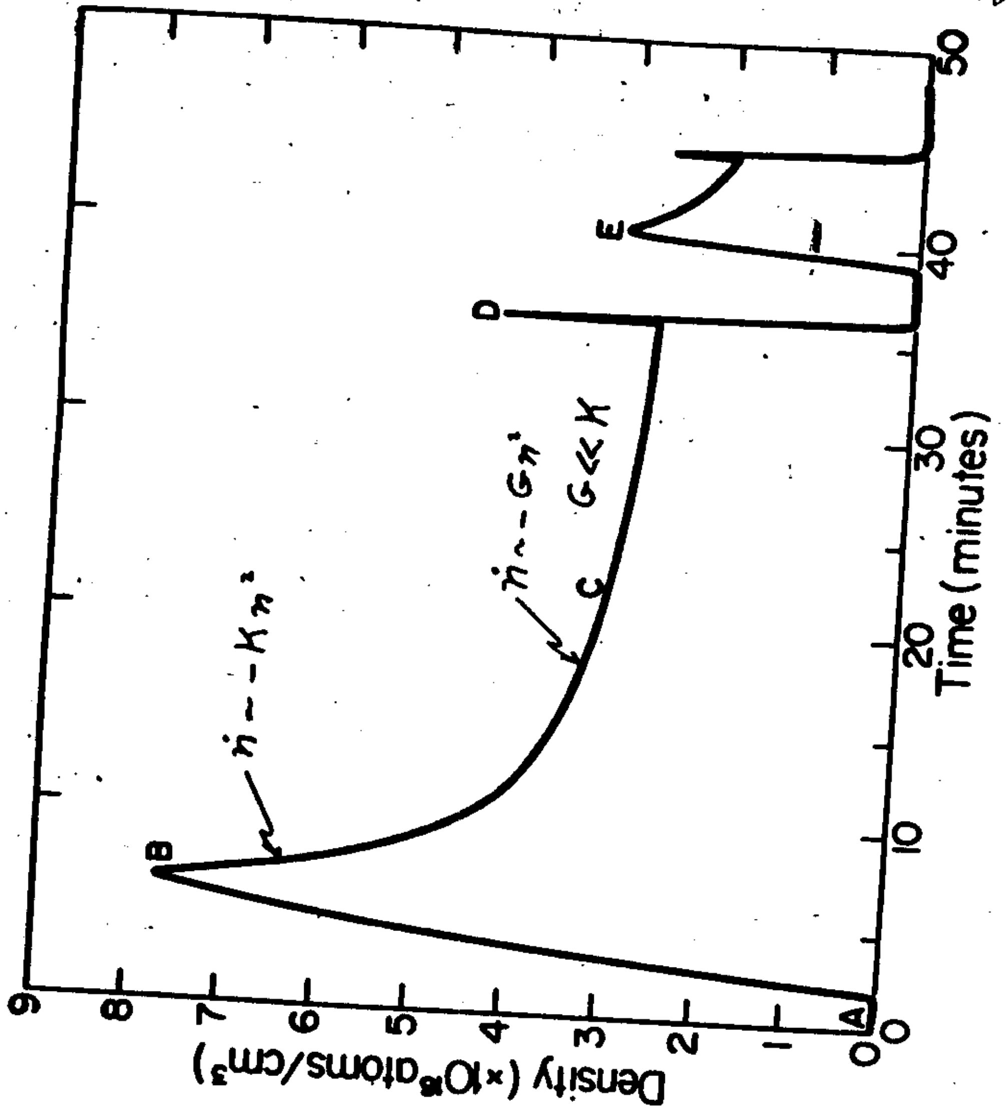
example: "evaporation from cell. $\tau = \gamma^{-1} = \tau_0 e^{\mu B/kT}$ ~ hours

"two body" or second order

$\dot{n} = -Kn^2$ $\boxed{\frac{1}{n} = \frac{1}{n_0} + Kt}$

example: H + H + surface → H_2 + surface





Explanation of M.I.T. results

Due to experimental and theoretical work of Hardy and Berlinsky - U.B.C.

H & B studied 0-field hyperfine resonance of H in He-lined cell, in range of 0.2 - 1.0 K.

Observations.

a) 3-body recombination rate, high T.

$$H\uparrow + H\downarrow + He \rightarrow H_2 + He + 4.8\,eV$$

$$\dot{n}_H = -(L_0 n_{He} n_H) n_H = -L_0 n_{He} n_H^2$$

b) 2-body recombination on surface, low T.

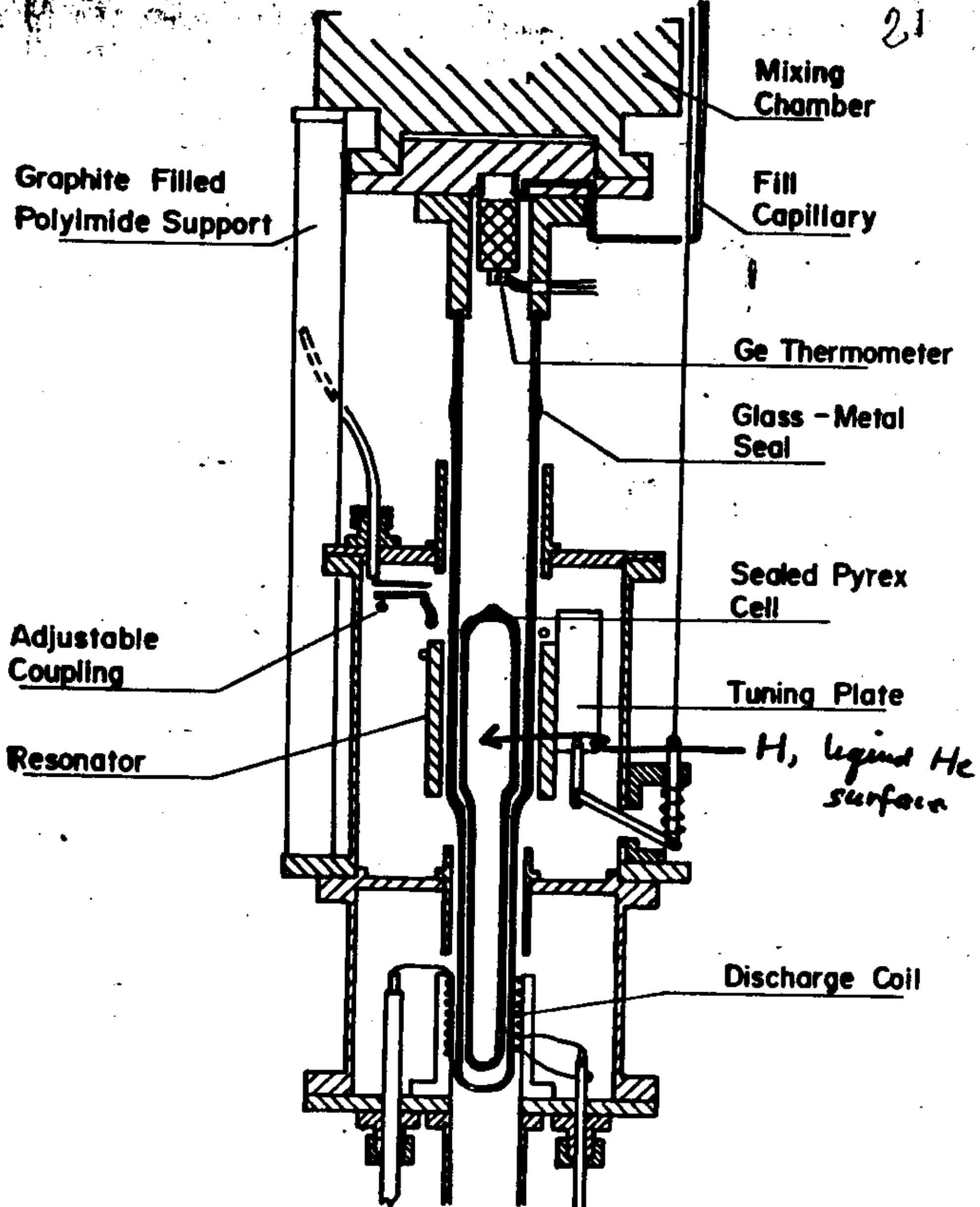
σ = surface density of H

$$\sigma = n \Lambda \exp\left[\frac{E_b}{kT}\right]$$ E_b = adsorption energy, H-He

surface recombination rate

$$\Gamma_s = l \bar{v} \sigma$$

scattering "cross length" (→ l); mean speed (→ $\bar{v}$)



Hyperfine resonance apparatus of Hardy & Berlinsky

Fig. 6.2

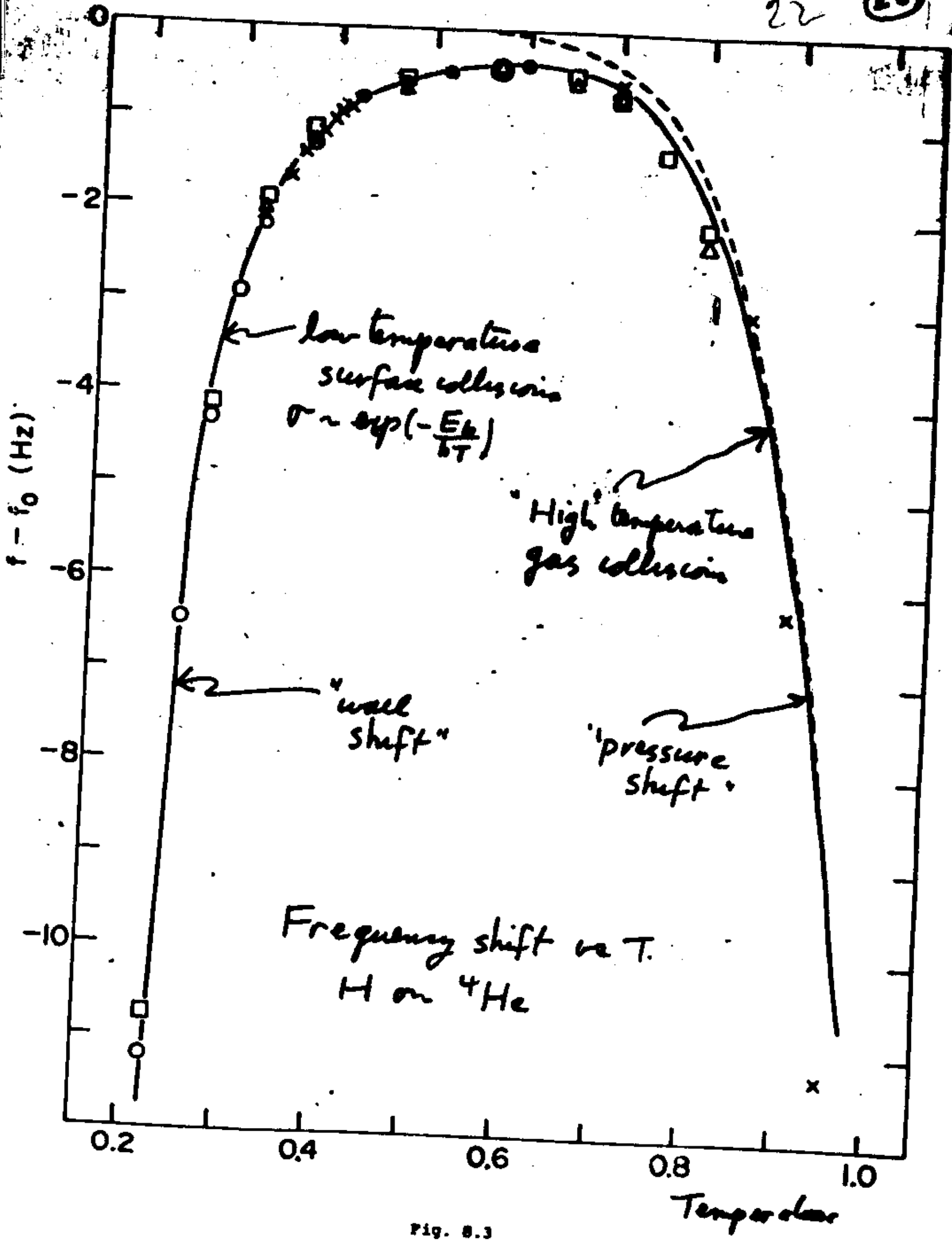


Fig. 8.3

explanation (continued)

surface recombination

$$\dot\sigma = -\Gamma_s \sigma = -\ell \bar v \sigma^2.$$

using $\dot N = A\dot\sigma$ (area), and $\sigma = n\Lambda e^{E_b/kT}$.

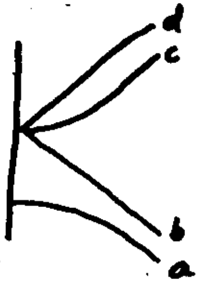
$$\dot N = -A\ell\bar v\Lambda^2 e^{2E_b/kT} n^2$$

$$\dot n = \frac{\dot N}{V} = -\left(\frac{A}{V}\right)\ell\bar v\Lambda^2 e^{2E_b/kT} n^2$$

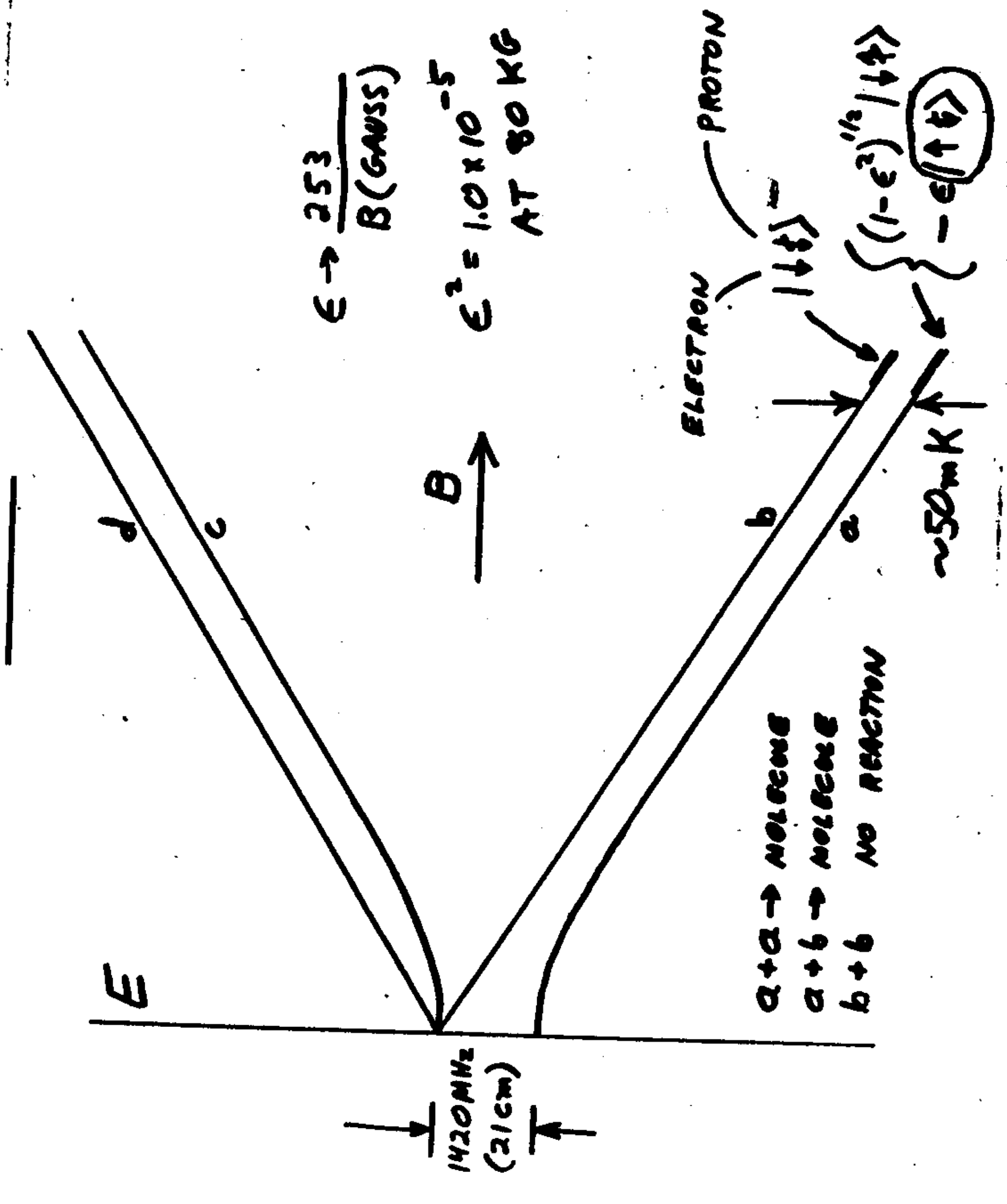
$$\dot n = -K_{eff}\, n^2$$

K_{eff} = 2-body decay rate $\sim e^{2E_b/kT}$

Implications for H↓ experiments



Due to hyperfine mixing, $|a\rangle$ is not a pure electron spin state. The spin impurity provides a channel for recombination.



Expect that

$$K(\text{high field}) \sim \epsilon^2 K(0\text{-field})$$

Temperature dependence of K:

$$K = \left(\frac{A}{V}\right) l \bar{v} \Lambda^2 e^{2E_b/kT}$$

$\bar{v} \sim \sqrt{T}$, $\Lambda^2 \sim \frac{1}{T}$

$$\sim \frac{1}{\sqrt{T}} e^{2E_b/kT}$$

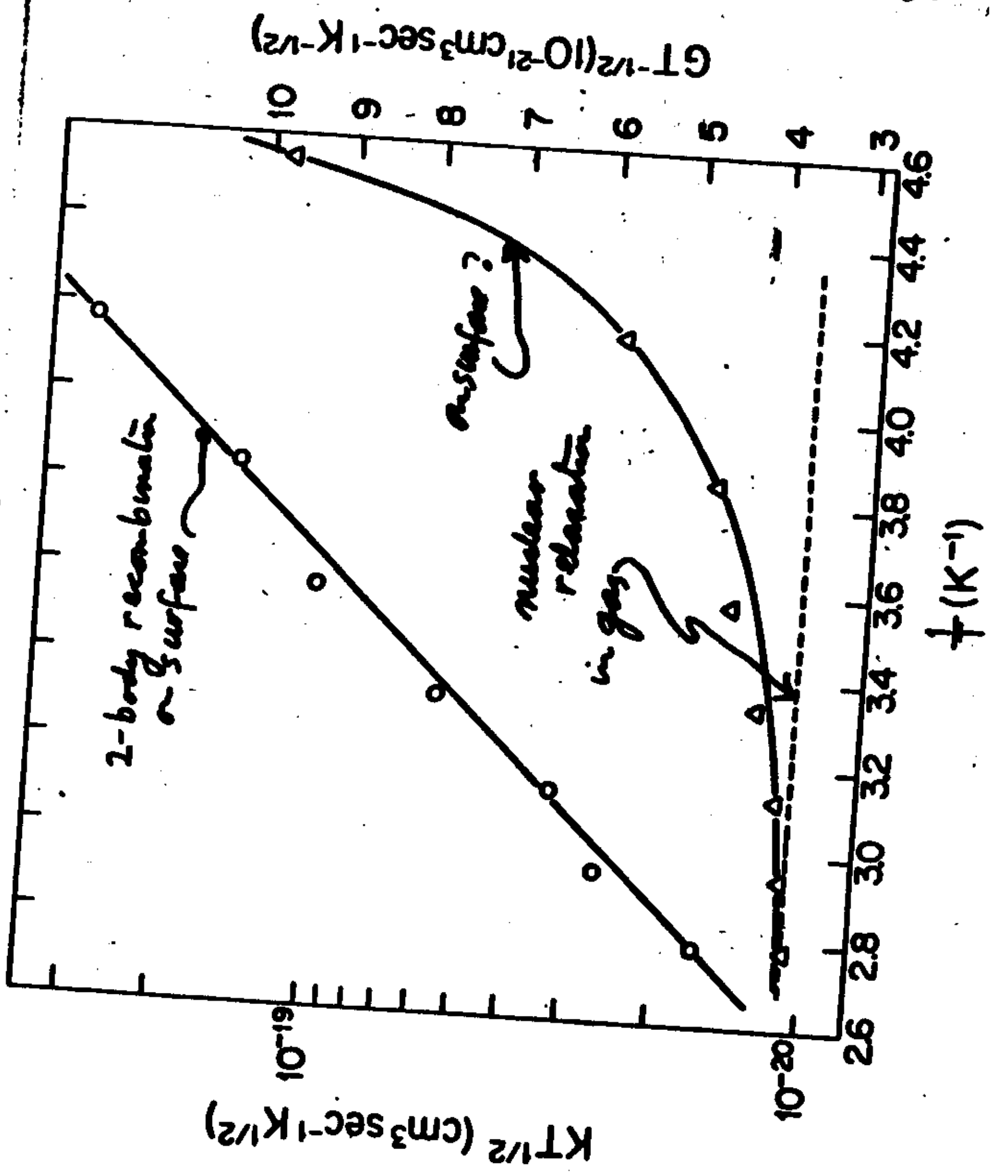
expect

$$\ln(K\sqrt{T}) = \frac{2E_b}{k} \times \left(\frac{1}{T}\right)$$

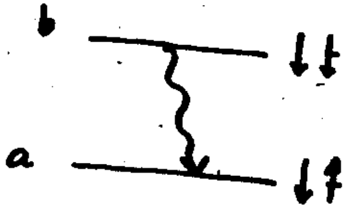
plot gives $E_b(H\text{-}^4He) \approx 1.0\,K$.

Further implications

When state |a> is completely recombined, a gas of |b> atoms will remain, stable against recombination. This is called doubly polarized H, $H\downarrow\Downarrow$ (↓ e, ⇓ proton).



Role of nuclear relaxation:



Berlinsky pointed out that state $|b\rangle$ will relax to $|a\rangle$ due to fluctuating magnetic fields during collisions.

Relaxation rate ∝ collision rate ∝ n.

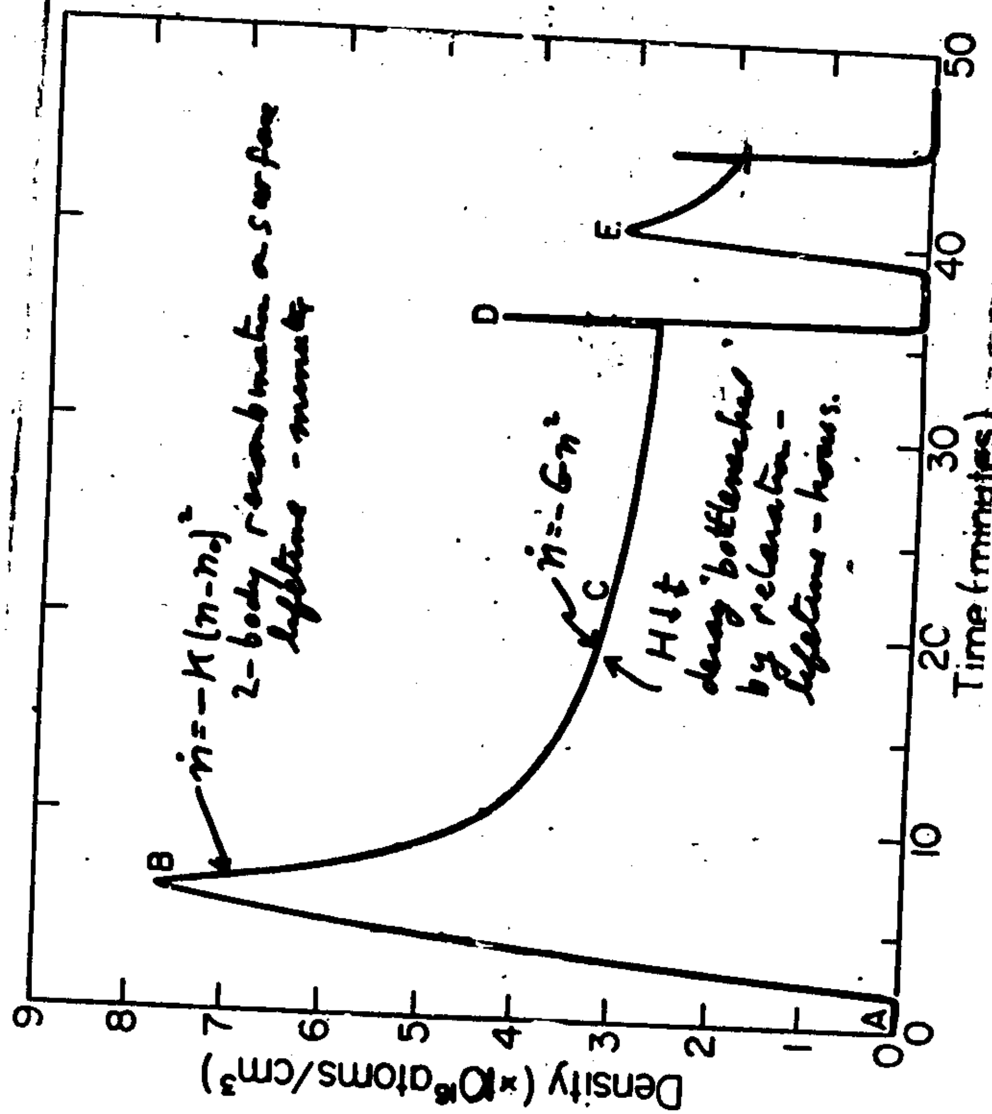
$$\dot{n}_b = -\Gamma n_b \ , \quad \Gamma = G n.$$

$$\dot{n}_b = -G n_b^2 \qquad (n_b \gg n_a)$$

If relaxation rate ≪ recombination rate, then recombination is "bottlenecked" by relaxation. In this regime

$$\dot{n} = -2 G n^2$$

2 atoms lost when 1 recombines.



Where to next?

- $H\downarrow$ can be created at moderate densities, $\sim 3 \times 10^{17}\ cm^{-3}$. By mechanically compressing the gas, higher density should be achievable. The relaxation rate will increase – at $8 \times 10^{19}\ cm^{-3}$, the lifetime is seconds. This is very long compared to the time required to measure the properties of $H\downarrow$.
- The above comments assume that no other processes become important at high density.

Spin-Polarised Hydrogen – lecture 3

A digression on laser-atomic physics

These lectures on $H\downarrow$ are offered essentially for general interest. The apparatus for $H\downarrow$ research is complicated and expensive. Low temperature expertise is essential. Thus, the research requires extensive resources.

The following proposal is for a research program in laser-atomic physics which can be started with a modest investment and which potentially provides many new research opportunities. It is based on the following thoughts:

* If you look through the NBS spectroscopy tables (Charlotte Moore) or other more recent compilations, you will find that large areas of data are simply missing. For instance, data on transitions to the higher angular momentum states of many atoms are often scarce.

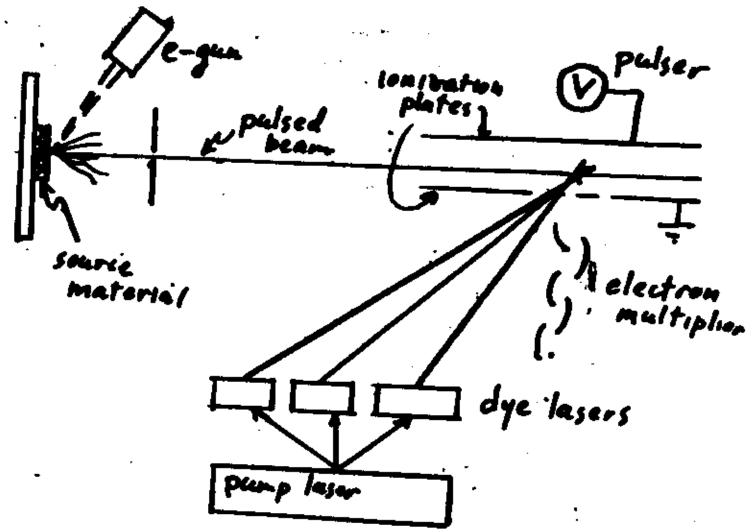
* With two or three pulsed dye lasers it is possible to attain access to many of these unstudied regions. (The atoms can be excited "stepwise" - i.e., from level to level.) This opens a large new area of atomic spectroscopy. More significantly, there is undoubtedly new and interesting atomic phenomena to be discovered.

* Spectra from these multiply excited states can be observed by fluorescence. An alternative method is to make the final excitation step terminate on a Rydberg (high-n) level. Rydberg states can be detected by field ionization, a simple method which is close to 100% efficient, and highly selective. With care, single atoms can be detected. The required field, which scales as $\frac{1}{n^4}$, is moderate - for $n=30$ it is about 300V/cm.

* A major problem in studying many atomic species, particularly refractory elements, is creation of a gaseous sample. An atomic beam is very well

suited to the proposed method. Since the lasers are pulsed, the beam can also be pulsed. Such beam can be created by bombarding a sample of target material with a pulsed electron beam. (alternative methods include laser heating, ion bombardment, etc.) Such a pulsed evaporation source is (more or less) universal, dissipates low average power, and places a very small load on the vacuum system.

* Some useful references:
  - Stebbings and Dunning, editors
    Rydberg States of Atoms & Molecules
    Cambridge University Press - 1983
  - Scientific American, May 1981 -
    Kleppner, Littman & Zimmerman
    (elementary discussion)



Equipment required -

- Pump laser - N_2 (alternative - Nd:YAG, excimer)
- tunable dye lasers - homemade
- e-multiplier or microchannel plate charged particle detector
- electron gun - from old cathode-ray tube?
- relatively straightforward electronics pulser for field ionizer, counters, timers, etc.
- simple vacuum system
  high vacuum not required, 10^{-5} torr probably okay.

Returning to H↓ –

To recapitulate "first generation" experiment:

2 states present in H↓

|b⟩ ———— ↓⇂

|a⟩ ———— ↓↿ − ε↑⇂

("spin impurity" marks the ε↑⇂ term)

Important processes

* 2-body recombination on surface
  at least 1 |a⟩ atom

* nuclear relaxation ∝ collision rate
  $\frac{1}{T_1} = -G(n_a + n_b)$

Let I_0 = flux/volume in each state.

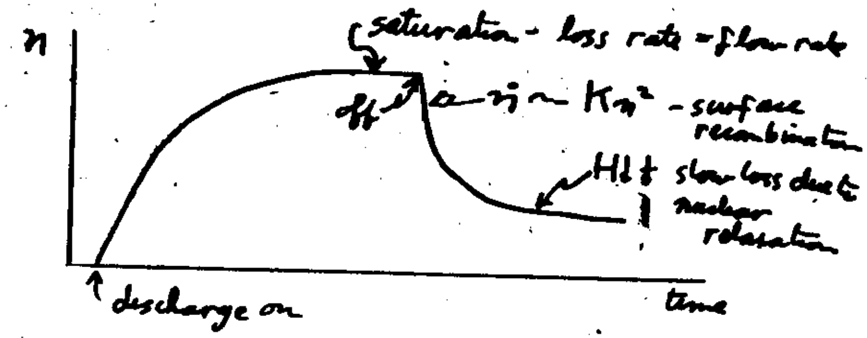
Rate equations

$$\dot{n}_b = I_0 - K n_a n_b - G(n_a+n_b)(n_b - n_a)$$

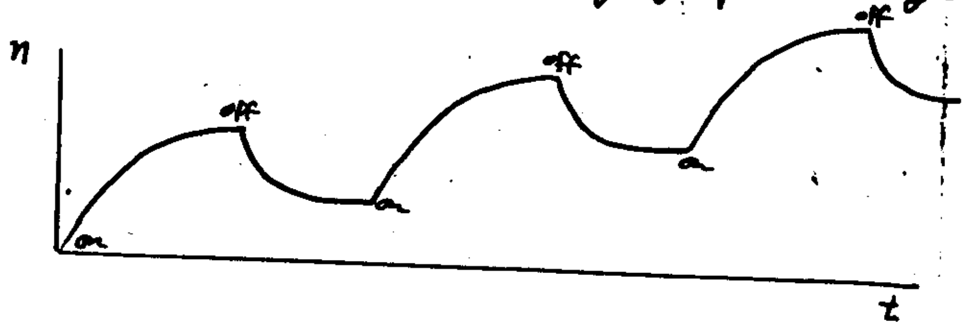
$$\dot{n}_a = I_0 - 2K n_b^2 + G(n_a+n_b)(n_b-n_a)$$

these equations are simple, but tricky

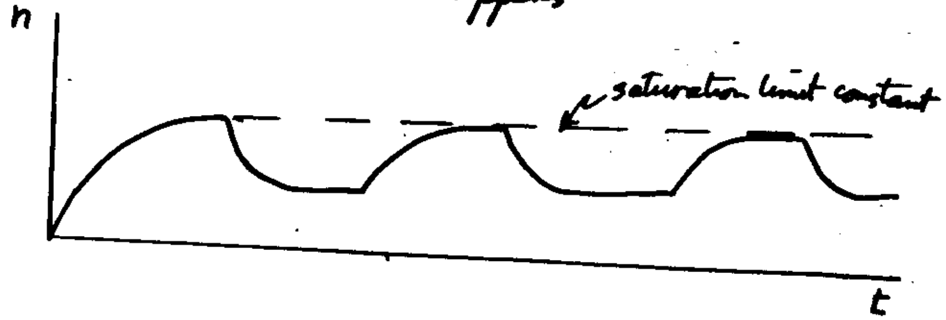
Experimental behavior



Q. Is it possible to increase density by repeated loading?



A. No! This is what happens

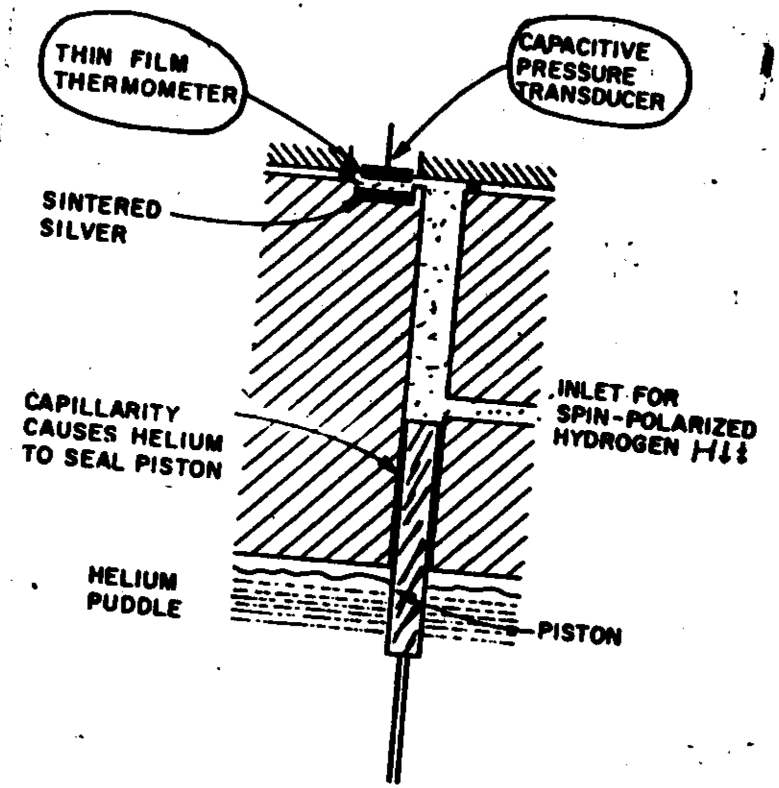


Compression Experiments

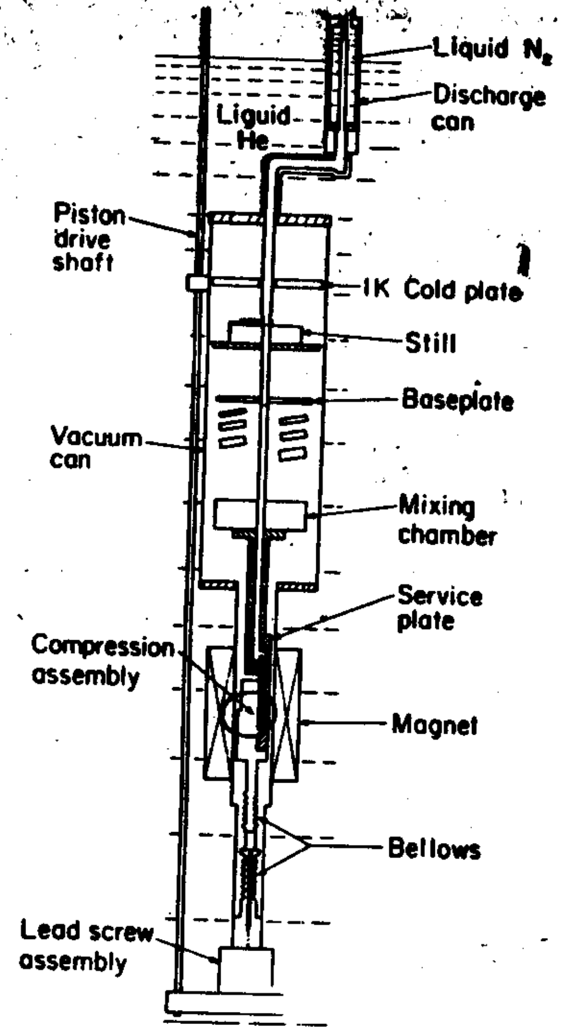
$H\downarrow\downarrow$ can be mechanically compressed to high density.

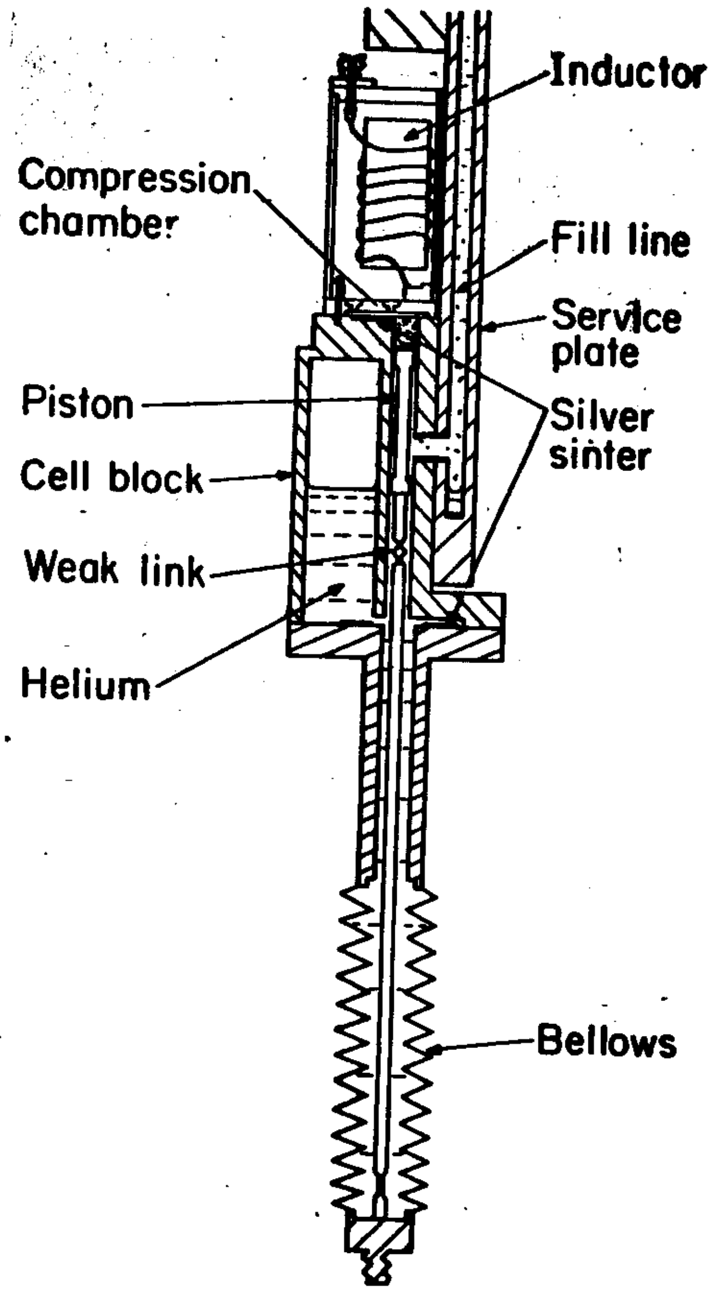
two approaches

- bubble – a bubble of $H\downarrow\downarrow$ is compressed in liquid helium – volume of bubble measured by change of capacitance
  used by Silvera & Walraven.
- piston – $H\downarrow\downarrow$ is compressed into a small sample cell by a piston in a cylinder.
  used by MIT group.
  (1st results in Phys. Rev. Lett, summer, 1983)

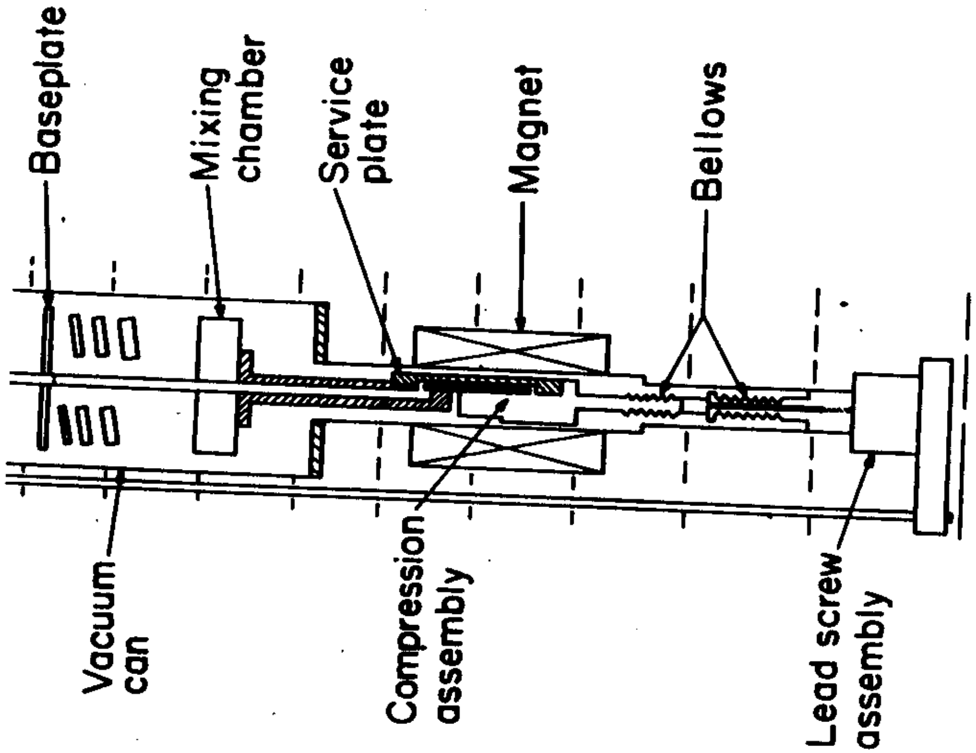


Compression Apparatus

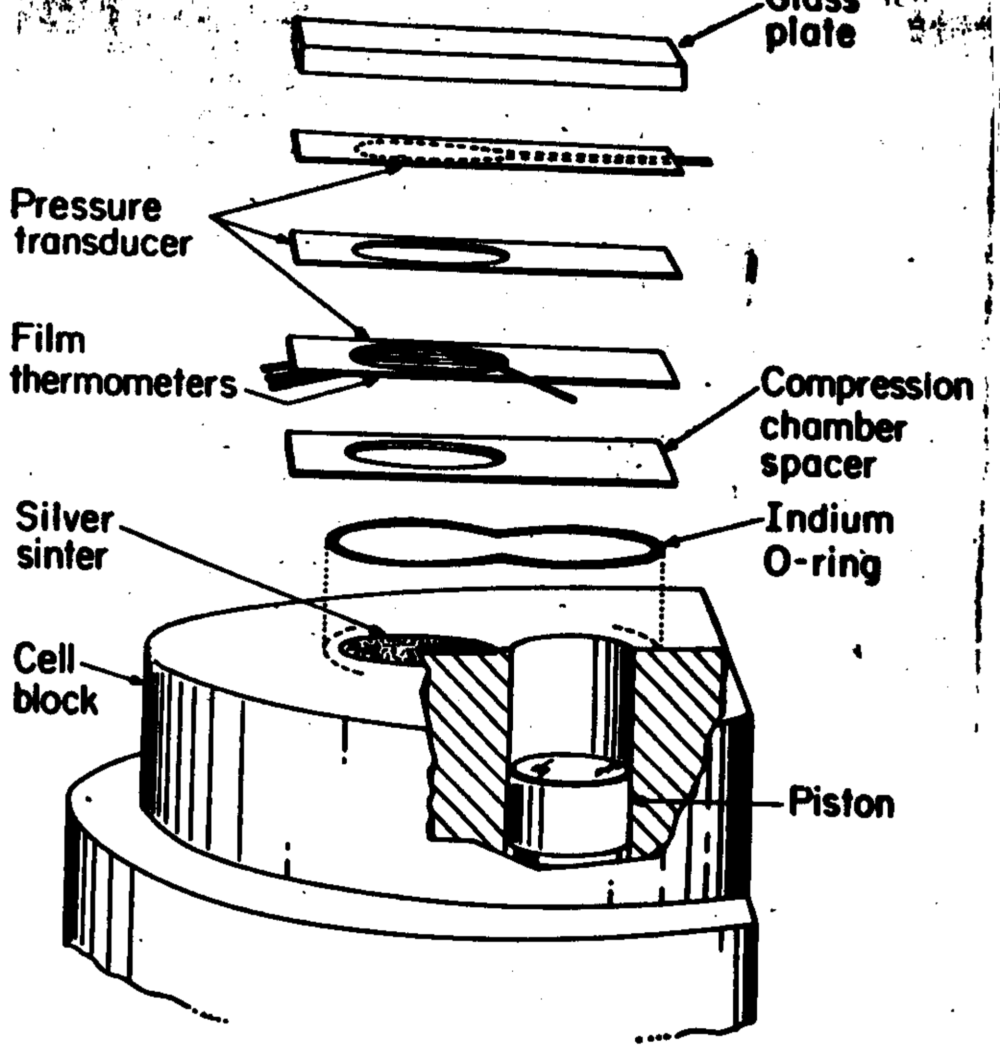

Liquid N_2
Discharge can
Liquid He
Piston drive shaft
1K Cold plate
Still
Baseplate
Vacuum can
Mixing chamber
Service plate
Compression assembly
Magnet
Bellows
Lead screw assembly


Inductor
Compression chamber
Fill line
Service plate
Piston
Silver sinter
Cell block
Weak link
Helium
Bellows

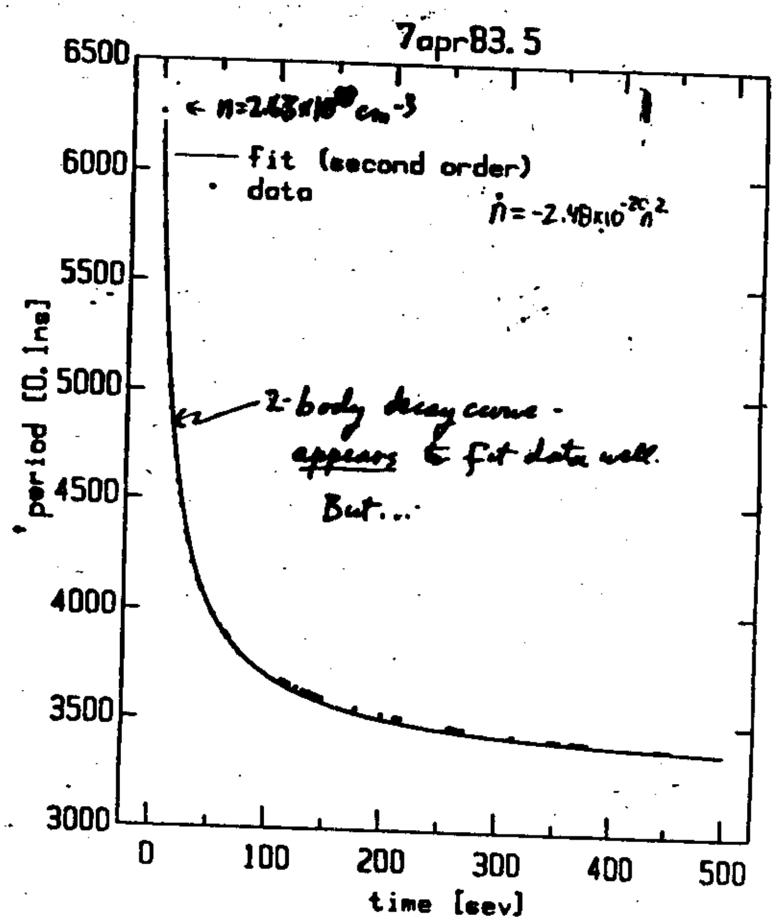
12 A



Baseplate
Mixing chamber
Service plate
Magnet
Bellows
Vacuum can
Compression assembly
Lead screw assembly



Glass plate
Pressure transducer
Film thermometers
Compression chamber spacer
Silver sinter
Indium O-ring
Cell block
Piston



first results of compression experiment
* (temperature not monitored)

Quantities measured are

Pressure vs time
and
Temperature vs time.

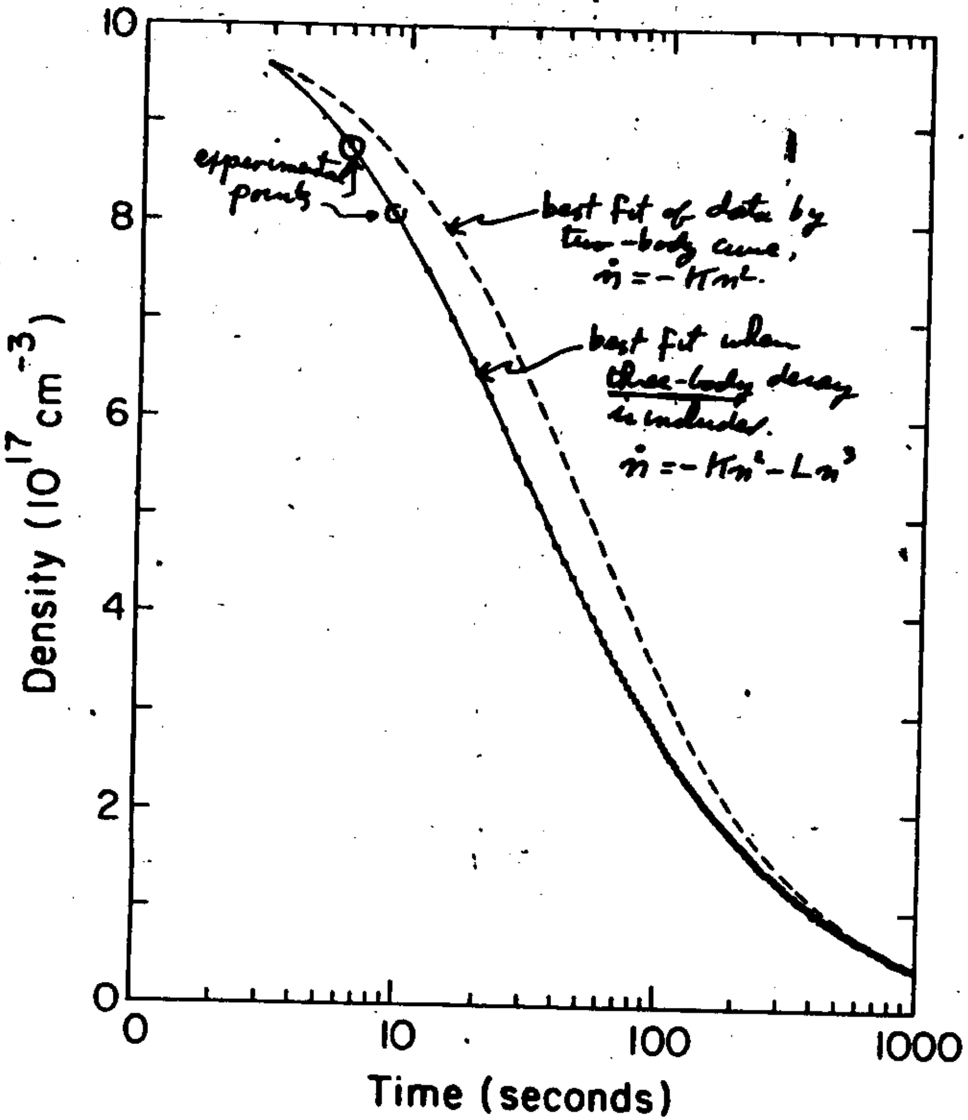
Quantity of interest is density vs time.
In region of operation, ideal gas law holds

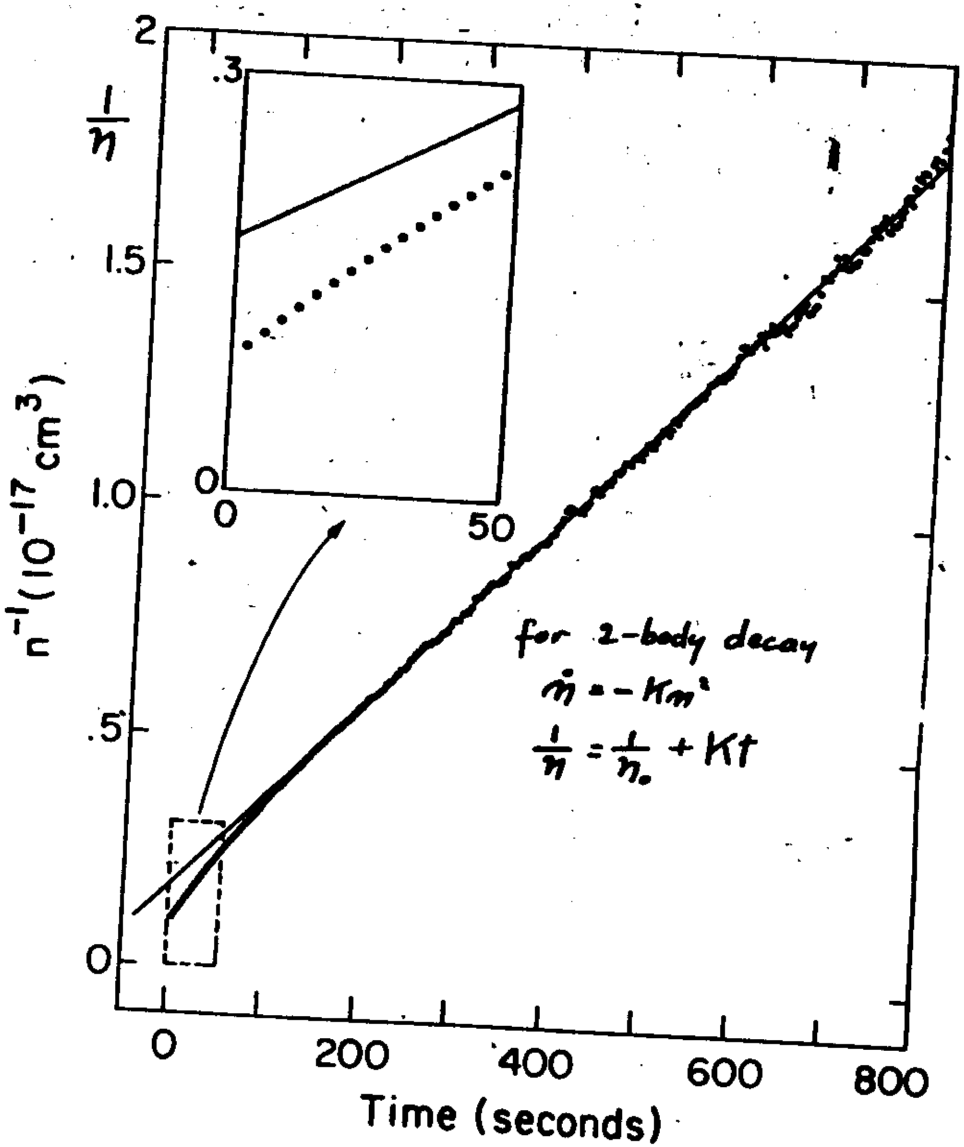
$$PV = NkT.$$

$$n = \frac{N}{V} = \frac{P(t)}{kT(t)}.$$

In practice, it is found that the temperature can increase significantly at high density, due to flow of heat of recombination. Typically, temperature is observed to rise from 300 to 350 mK.

When the data is reduced by formula above, the results look like...

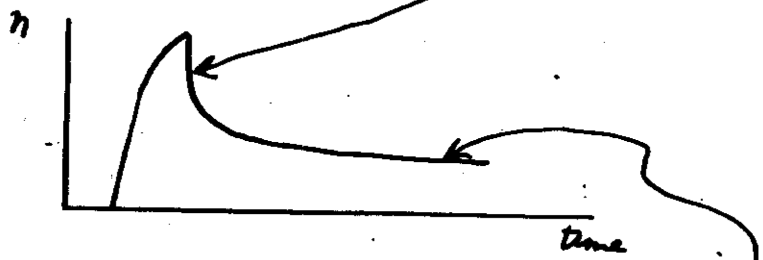

experimental points
best fit of data by two-body curve, $\dot{n} = -Kn^2$
best fit when three-body decay is included. $\dot{n} = -Kn^2 - Ln^3$
Density (10^{17} cm^{-3})
Time (seconds)
10
8
6
4
2
0
0
10
100
1000


$\frac{1}{n}$
n^{-1} (10^{-17} cm^3)
for 2-body decay
$\dot{n} = -Kn^2$
$\frac{1}{n} = \frac{1}{n_o} + Kt$
Time (seconds)
2
1.5
1.0
.5
0
.3
0
0
50
0
200
400
600
800

How to tell what is going on.

It is easy to make errors in interpreting density decay data.

* Much of the important "action" is over in the first few moments.



* At long times decay rate is very small - easy to confuse 2-body and 1-body processes ($\dot{n} = -2Gn^2$; $\dot{n} = -\gamma n$).
(Nuclear relaxation due to paramagnetic impurities in surfaces gives rise to 1-body decay.)

* Parameters may be changing with temperature - surface density very sensitive.

** Curve fit can be non-unique -

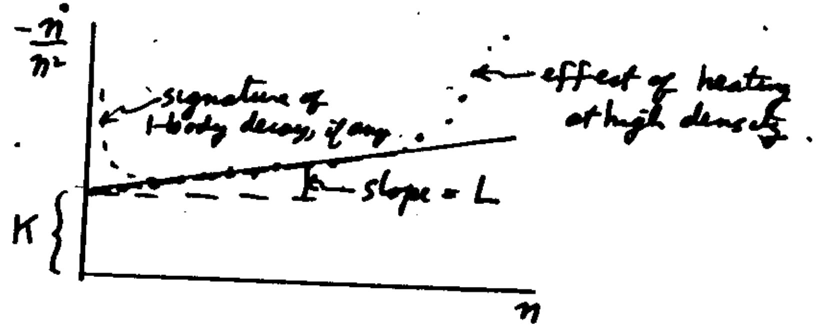
Assume system evolves according to

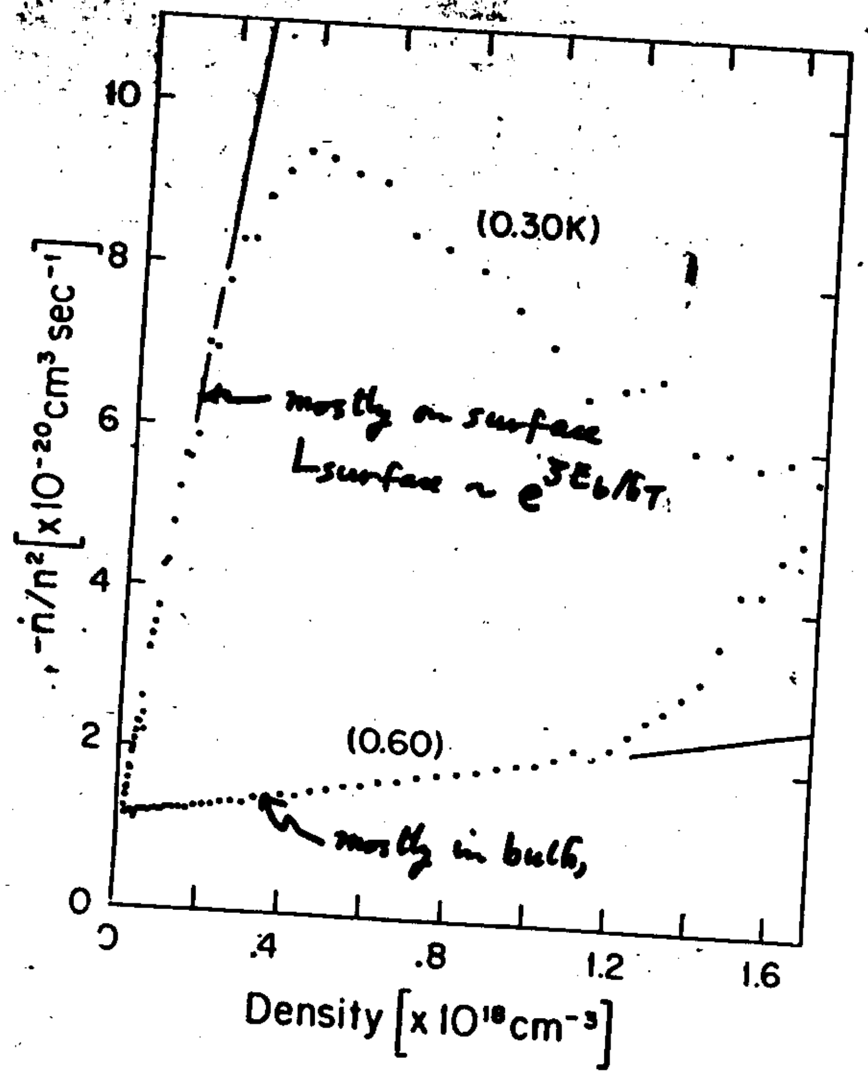
$\dot{n} = -Kn^2 - Ln^3 - \gamma n$.

1). evaluate $\dot{n}(t)$ from $n(t)$, numerically

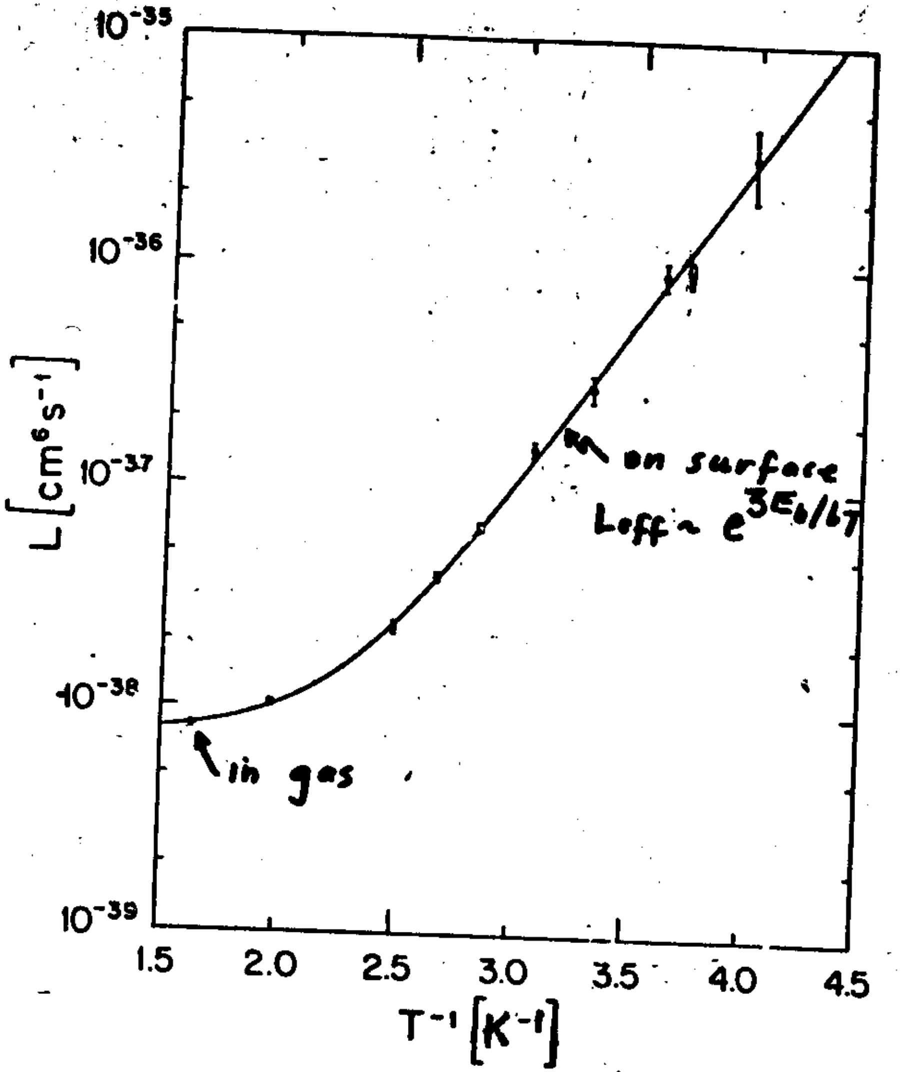
2). plot $-\frac{\dot{n}}{n^2}$ vs n.

$$-\frac{\dot{n}}{n^2} = K + Ln + \frac{\gamma}{n}$$





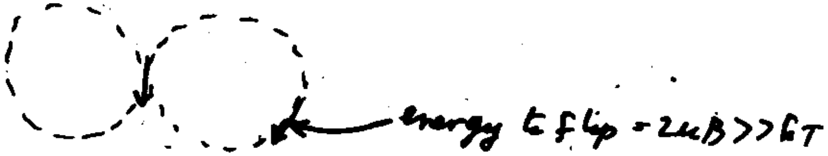
10
8
6
4
2
0
$-\dot{n}/n^2$ [x 10^{-20} cm^3 sec^{-1}]
(0.30K)
(0.60)
mostly on surface
$L_{surface} \sim e^{3E_b/kT}$
mostly in bulk,
0
.4
.8
1.2
1.6
Density [x 10^{18} cm^{-3}]



10^{-35}
10^{-36}
10^{-37}
10^{-38}
10^{-39}
L [cm^6 s^{-1}]
on surface
$L_{eff} \sim e^{3E_b/kT}$
in gas
1.5
2.0
2.5
3.0
3.5
4.0
4.5
T^{-1} [K^{-1}]

Explanation of three-body recombination by Kagan, Vartanyantz and Shlyapnikov

Dipole mixing → electron spin flip.
forbidden in 2-body collision.



electron spin relaxation energetically forbidden by barrier of 2μB ≈ 10K.

BUT – in 3-body collision..

spin flips and the atoms <u>simultaneously</u> recombine – no energy barrier due to availability of recombination energy.

3-body recombination can occur in gas or on surface – but 3 H-atoms are needed in both cases.

Current status of H↓.

- Highest density under controlled conditions – $n = 4.5 \times 10^{18}$ atoms cm^{-3}! (n_{BE} (0.3K) $= 8 \times 10^{19}$ cm^{-3})
- Limit to density – 3-body dipole-induced recombination
- Where to from here?

  $n_{BE} \propto T^{3/2}$: go to low T.

  Limitation – at low T the atoms will stick to the walls. Can the walls be eliminated? Is a pure magnetic trap possible?

Summary - What has been accomplished

* Recombination processes studied and understood, 2- and 3-body, in gas and on surface.

* Nuclear and electronic relaxation rates

* Spin exchange at low temperature

* Thermal conductivity and diffusion constant, + other transport phenomena.

* Hyperfine shifts

* Adsorption energies on ^3He and ^4He

* New technology created.

There have been large investments in time and resources by several groups. The B-E transition has not yet been observed, and it is an open question as to whether it ever will be.

Neverthless, the payoff has been considerable

Uses for Spin-Polarized Hydrogen

Condensed Matter Physics

- Quest for Bose-Einstein transition
- Study of a new spin-polarized quantum fluid (Lhuillier, Laloë)
  - transport properties
  - two-dimensional phase
  - spin-waves (Lévy et al, Cornell)

Metrology

- Hydrogen maser atomic clock, T≈1K.
  - many advantages - stability, power, etc.
  - ultimate accuracy, >1 in 10^{16} ?

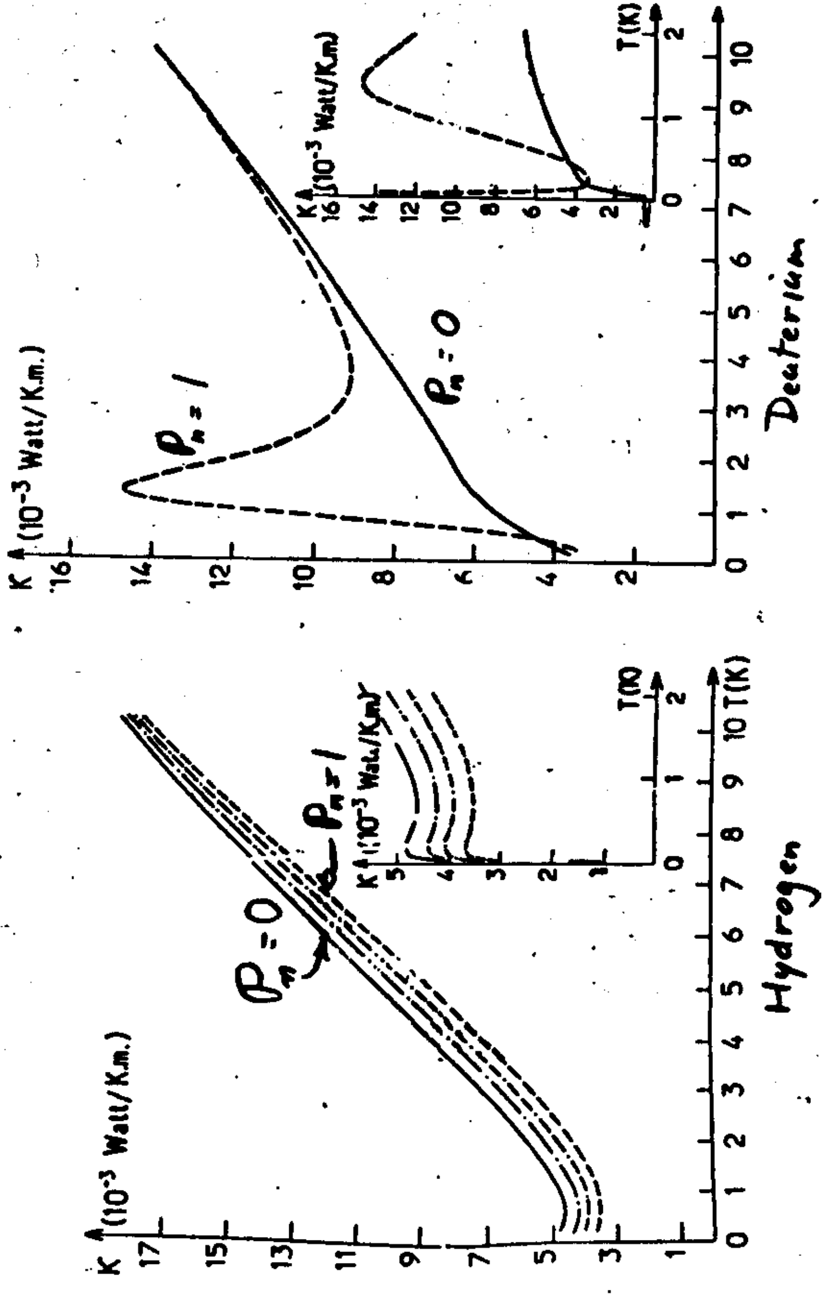
Atomic Physics

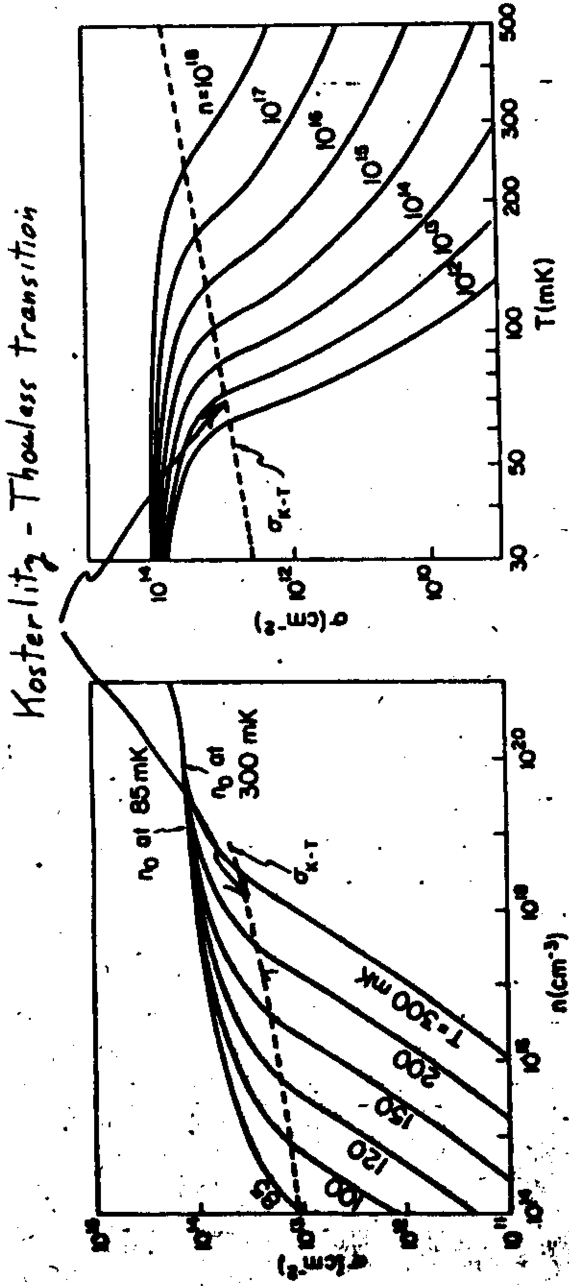
- Dense H target
  - charge exchange, spin exchange
- Cold H beam for "super-spectroscopy"
  - low 2nd-order Doppler: $\sim 10^{-15}$
  - long interaction time, "fountain"?
  - permits observation of 1s-2s two-photon transition $\tau(2s) = 1/7$ sec, $Q > 10^{15}$

Particle Physics, Nuclear Physics

- Intense beams of polarized protons
  - polarized proton source
  - polarized proton jet

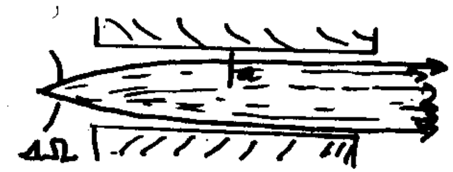
Thermal Conductivity calculated by C. Lhuillier





Cold H atomic beam

- Conventional beam with state-selecting focusing hexapole magnet



$$\Delta\Omega = (\sim 2)\,\frac{\mu B_0}{kT}$$

figure of merit: $\frac{\mu B}{kT} \simeq 0.6\,\frac{B(\text{tesla})}{T(K)}$

$= 0.6\,\frac{1\text{ tesla}}{300K} = 2\times 10^{-3}$ conventional

$= 0.6 \times \frac{10T}{0.3K} = 20$ low temp

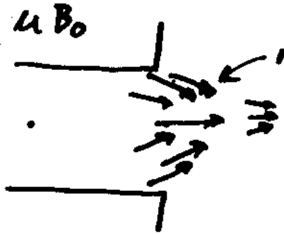
Should be possible to design very efficient atomic beam - useful for H collision studies, spectroscopy, H maser, polarized proton sources, etc.

Beam optics for cold H

New type of optics based on magnetic deflection possible in regime $\mu B >> kT$. In general, the ideal field seekers are focused.

- Simple coil behaves like a lens.

-- A solenoid-ejection source is self focusing - 2 reasons: a) acceleration focusing
b) radial gradients



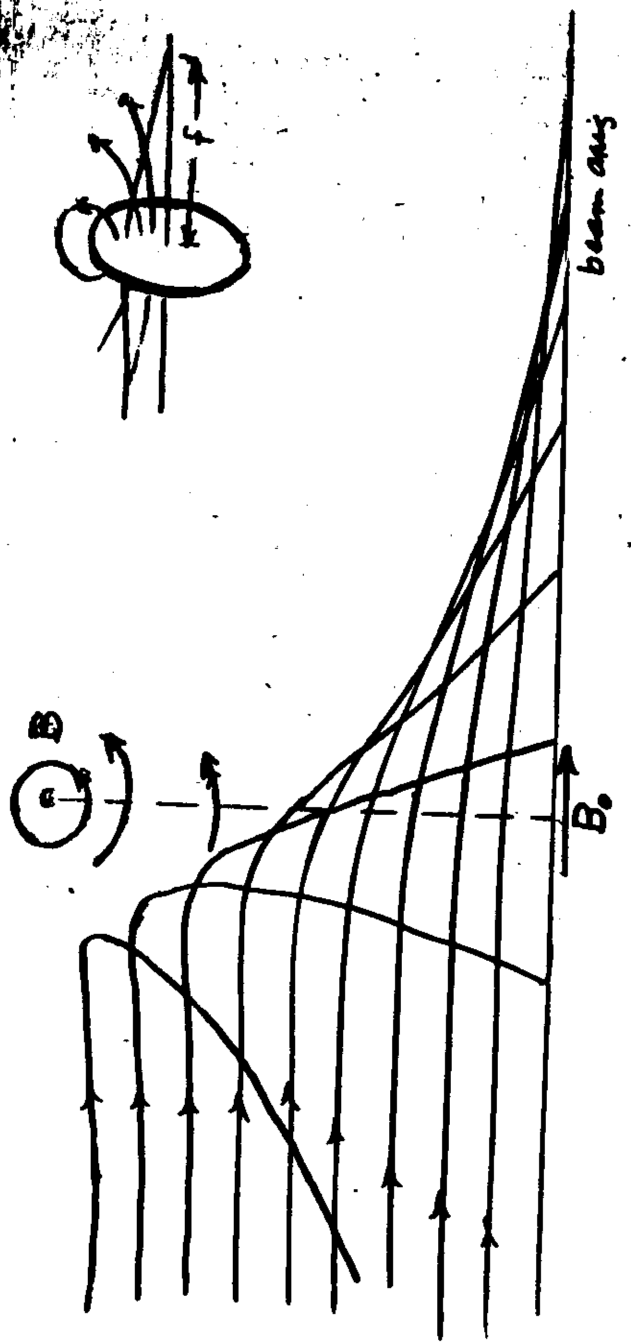
source,
$v_t \approx \frac{2kT}{M}$

$\Omega \approx \frac{kT}{\mu_0 B}$

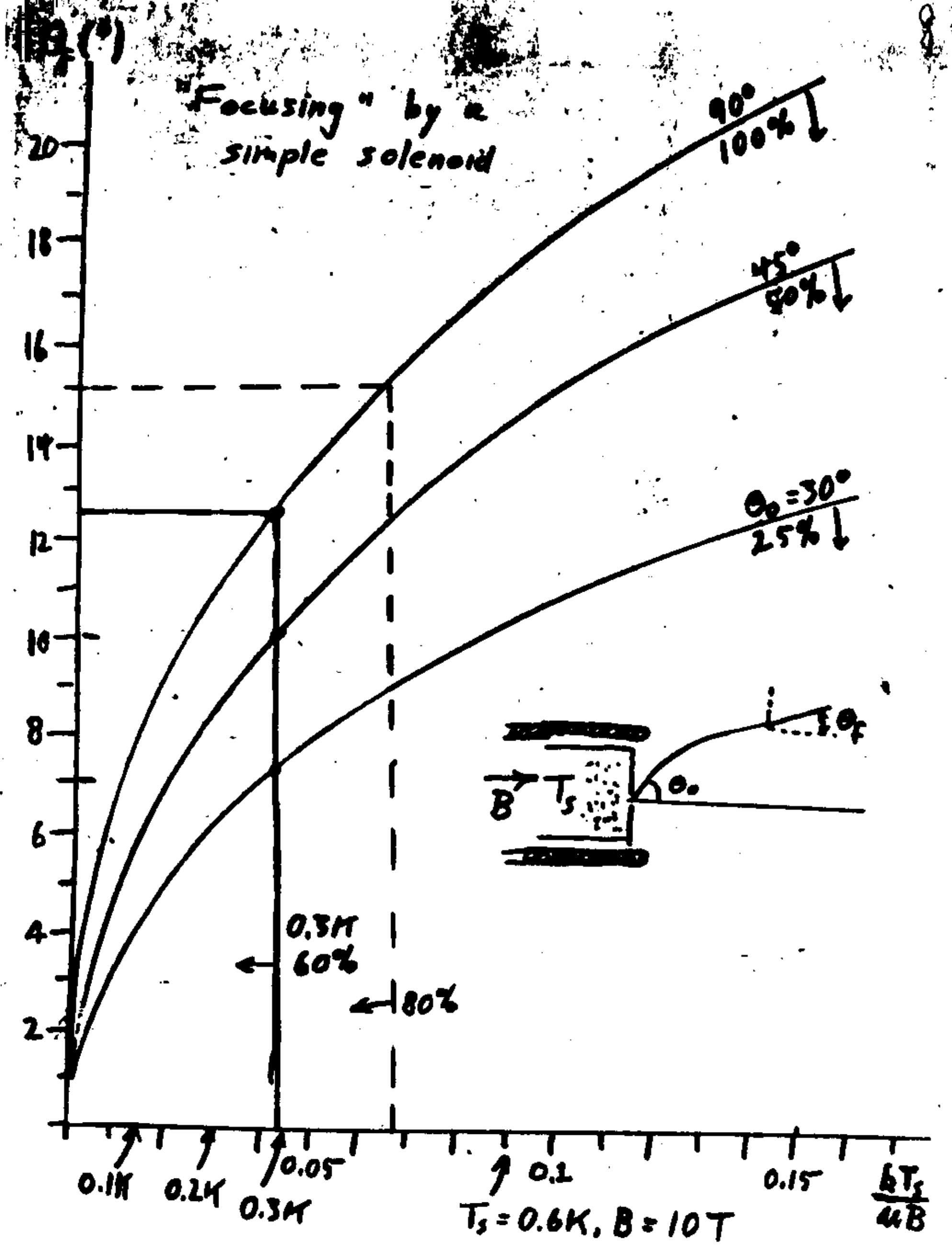
$\frac{1}{2} M V^2 = \mu_0 B$

$V = \sqrt{\frac{2\mu_0 B}{M}} >> v_t$

"acceleration focusing"



focusing by current loop
$kT = 1.1 B_0$

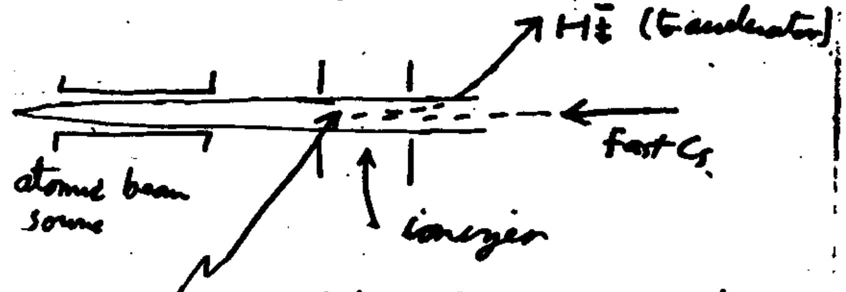


Application to particle physics

Polarized proton source.

Polarized proton "jet" (gaseous atomic beam target for use inside of accelerator)

Conventional polarized proton H^- source.



$H\downarrow + Cs \rightarrow H\downarrow^- + Cs^+$

density of $H\downarrow$
$10^{11} \sim 10^{12}\ cm^{-3}$?

density of spin polarized H: $H\downarrow\uparrow \sim 10^{17}\ cm^{-3}$?

Big improvements should be possible.

Elements of a Polarized Proton Jet

1) **Source of H atoms**

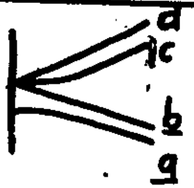
conventional discharge

2) **Storage cell for a, b states**

^4He - coated

$T \sim 0.5\,K$

$B \sim 9$ tesla

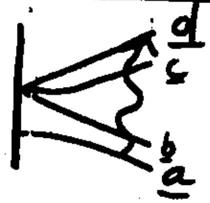


3) **Microwave Ejection (Niinikoski)**

$\underline{a} \rightarrow d$

$\nu \sim 240$ GHz

($\lambda = 1.2$ mm)



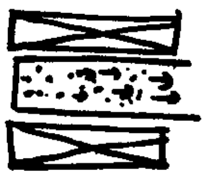
4) **Transport stage**

processes to consider

- diffusion
- recombination
  surface, gas
- nuclear relaxation
- electron relaxation
- spin-exchange

$\underline{d} + \underline{b} \rightarrow \underline{a} + \underline{c}$

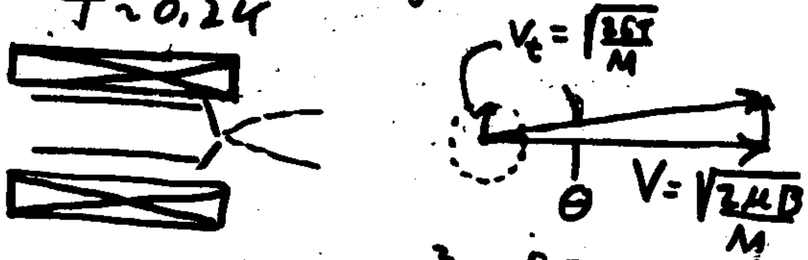
nuclear polarization lost but state $\underline{b}$, as well as $\underline{a}$, is expelled.



5. **Ejection stage**

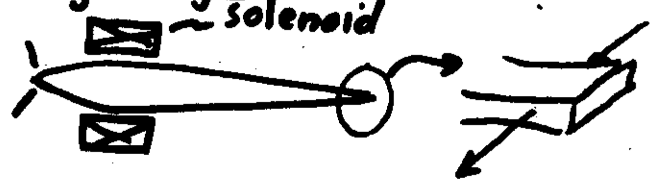
- atoms cooled for efficient focusing

$T \sim 0.2\,K$



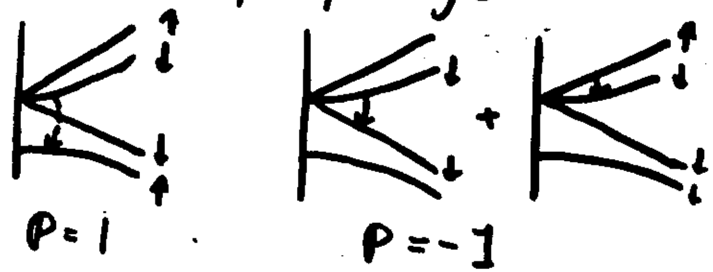
$$\theta^2 \sim \frac{kT}{\mu B}$$

6). **Focusing stage**



7) **e-p polarization transfer**

- adiabatic rapid passage



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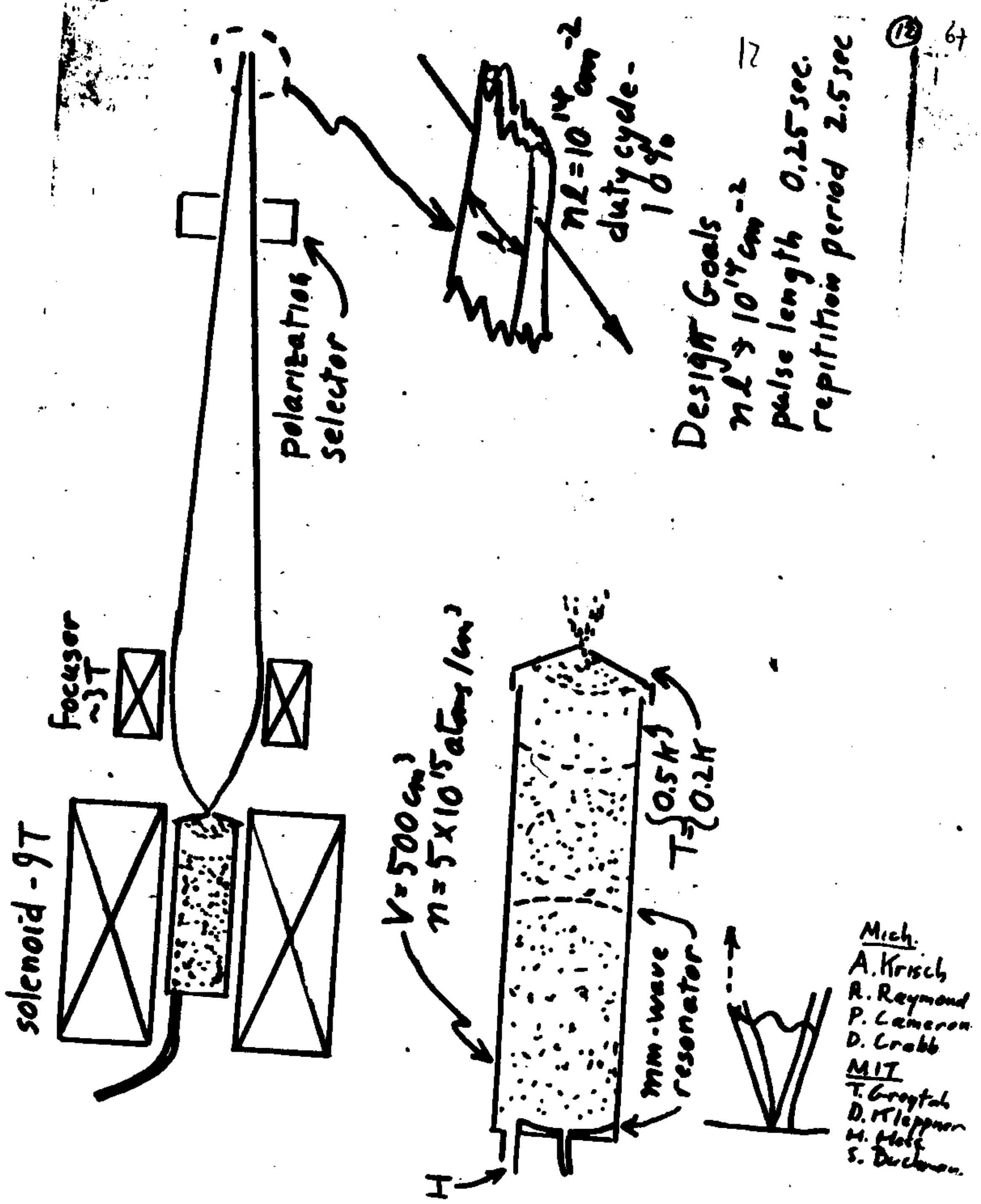

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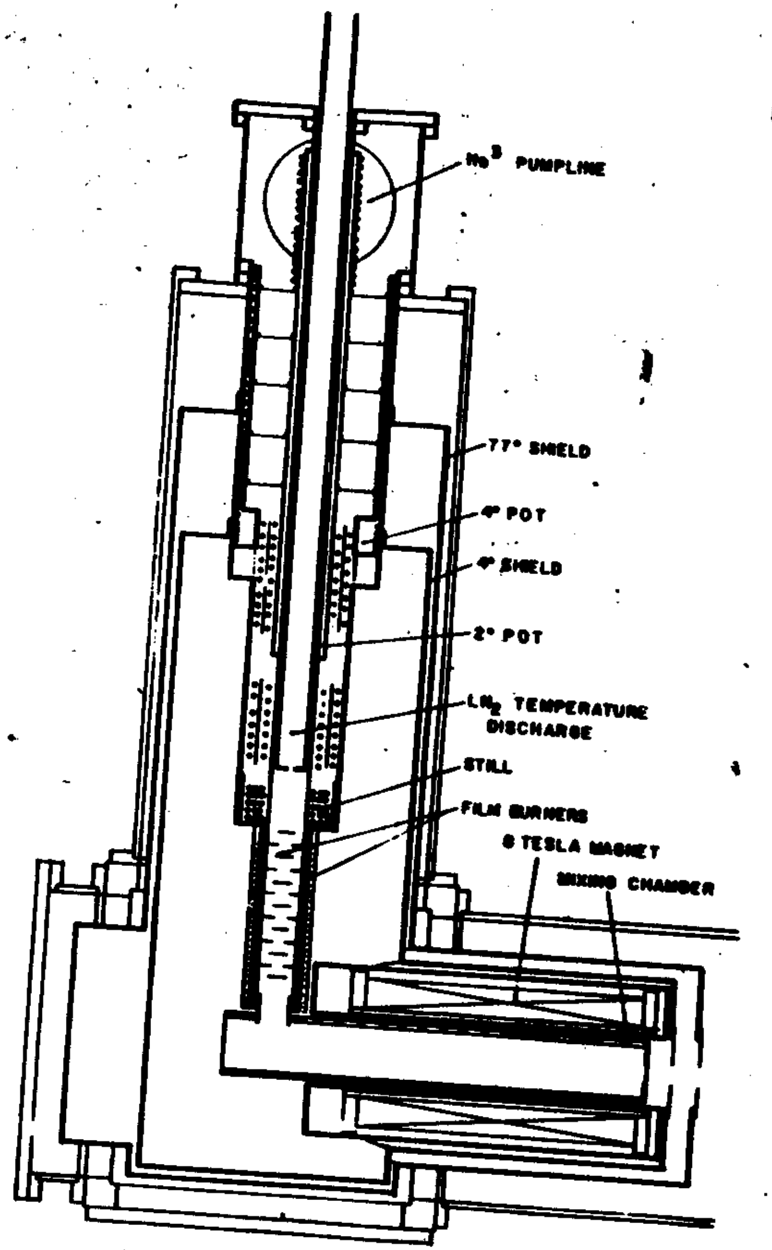
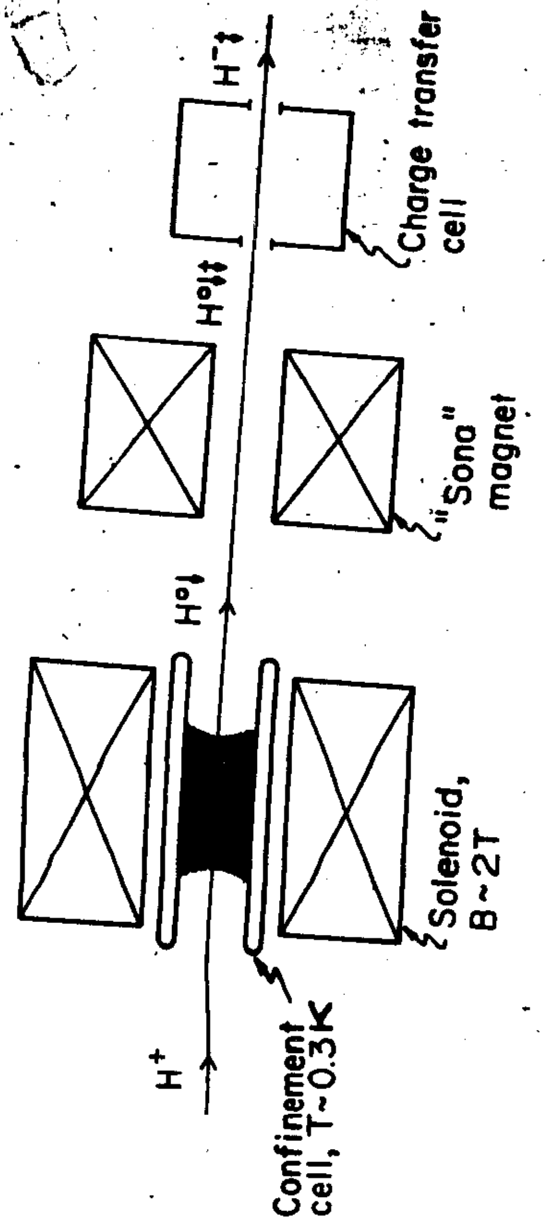
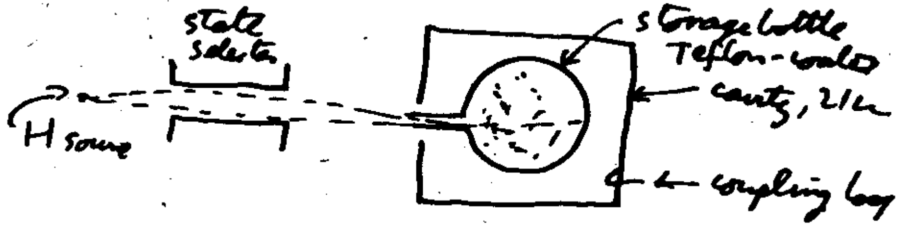


MICHIGAN-MIT-BROOKHAVEN
POLARIZED COLD ATOMIC HYDROGEN JET



Cold H Maser (Hardy & Berkeley)



conventional maser

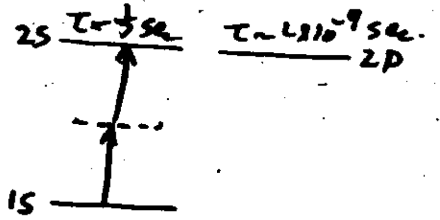
Advantages of cold (0.3K) maser

- He instead of Teflon on walls - more reproducible.
- Spin-exchange cross section much smaller - provides much more power.
- Larger beam flux available.
- kT noise suppressed.
- Many sources of drift diminished - cavity drift, magnetic fields, etc.

Present state of the art
$\frac{\delta f}{f} \sim 3 \times 10^{-13}$ absolute
$\sim 10^{-15}$ relative (hours-days)

Projected improvement - $< 10^{-16}$

Application of cold H to H spectroscopy



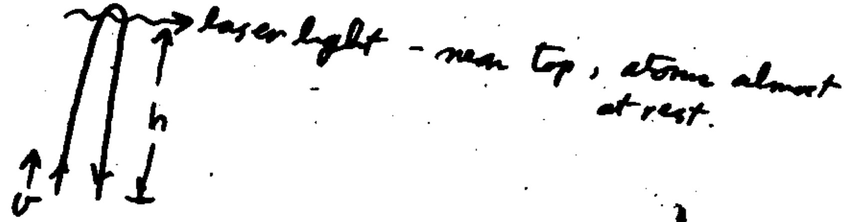
1S — 2S (2 photon) transition attractive

- Very high Q — $Q = \omega\tau \sim 10^{16}$
- Useful spectroscopy of H
- Potential optical frequency "standard"

Requirements

- Lasers stable to 1 in 10^{16} — not available, but progress appears steady
- Some method for observing H for 1/7 sec. Natural approach — cold H

H "fountain"



$v^2 = 2gh$. take $h = 1$ m

$v = 5\times10^{2}$ cm/s.

$T = \sqrt{\frac{2Mgh}{k}} \sim \underline{10^{-2}}$ K.

There are various possibilities for cooling H to 10 mK. One is to thermalize by a few collisions on a cold He surface. In equilibrium, the atoms would be completely adsorbed — but it takes many collisions for this to occur.

END