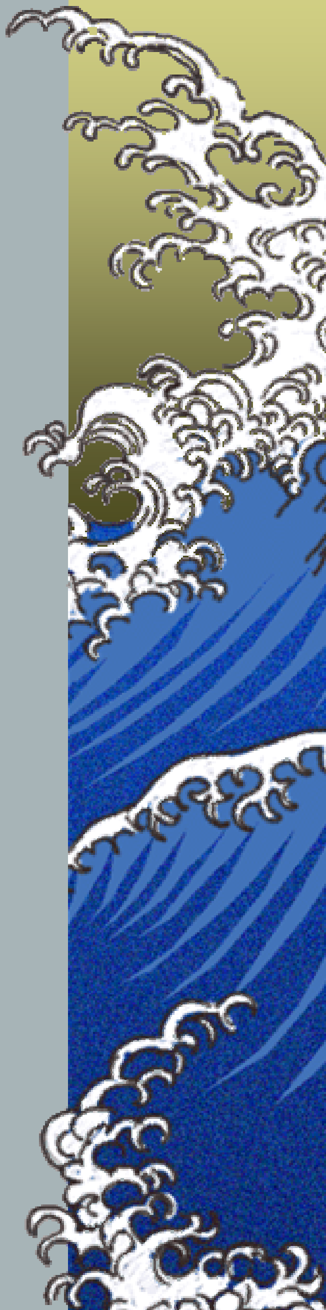
Institute for Physical Research, NAS of Armenia

Studies of Laser Induced Cesium and Rubidium Hydride Formation in Vapor Cells and Their Application for Isotope Separation



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- *Brief overview and background*
- *Basic concept*
- *Hydride formation in the cell volume*
- *Hydride formation on the cell window*
- *Experiments and results*
- *Model and calculations*
- *Hyperfine structure of D-lines of Cs isotopes*
- *Determination of abundance*
- *Conclusion*

Background

Industrially realized non-laser techniques:

Gas diffusion; Gas centrifugation

Industrially realized laser techniques:

Selective ionization

Published possible laser techniques:

Selective high-level excitation

The basic concept is:

Alkali atoms do not react with hydrogen molecules in their ground states.

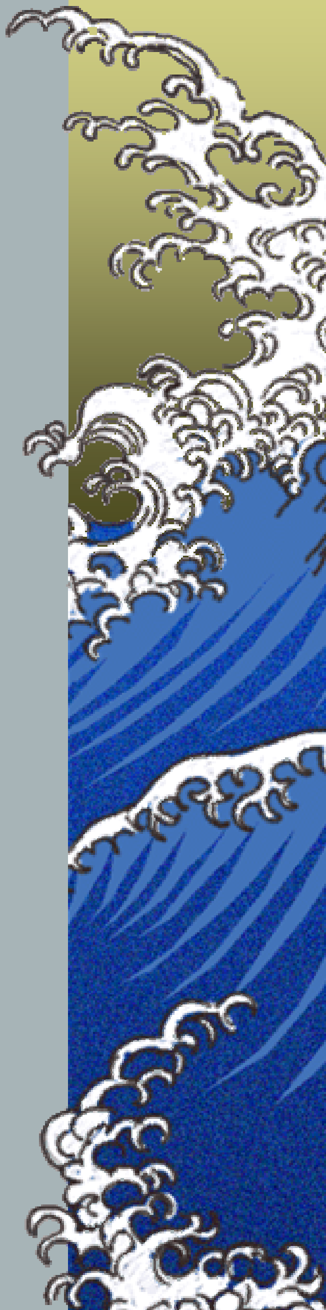
However, when excited, they do.

Excitation may be realized isotope- selectively by narrow-line laser radiation

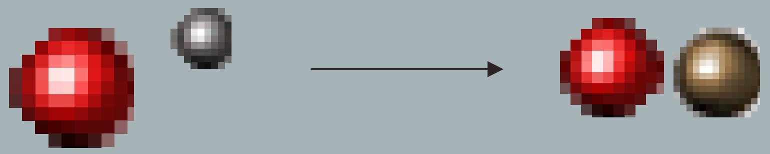
We use the lowest excited state of Cs and Rb

*As distinct from highly excited **Cs** atoms, the energy of those excited to the D-level is insufficient to form a hydride molecule in collision with a hydrogen molecule*

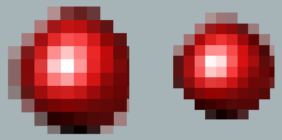
However, the reaction may proceed in two steps, as was obtained earlier in sodium vapor mixed with hydrogen



The first step - quenching

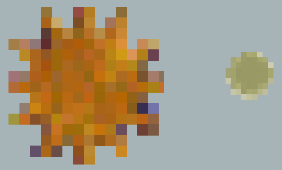


The second step – molecule formation



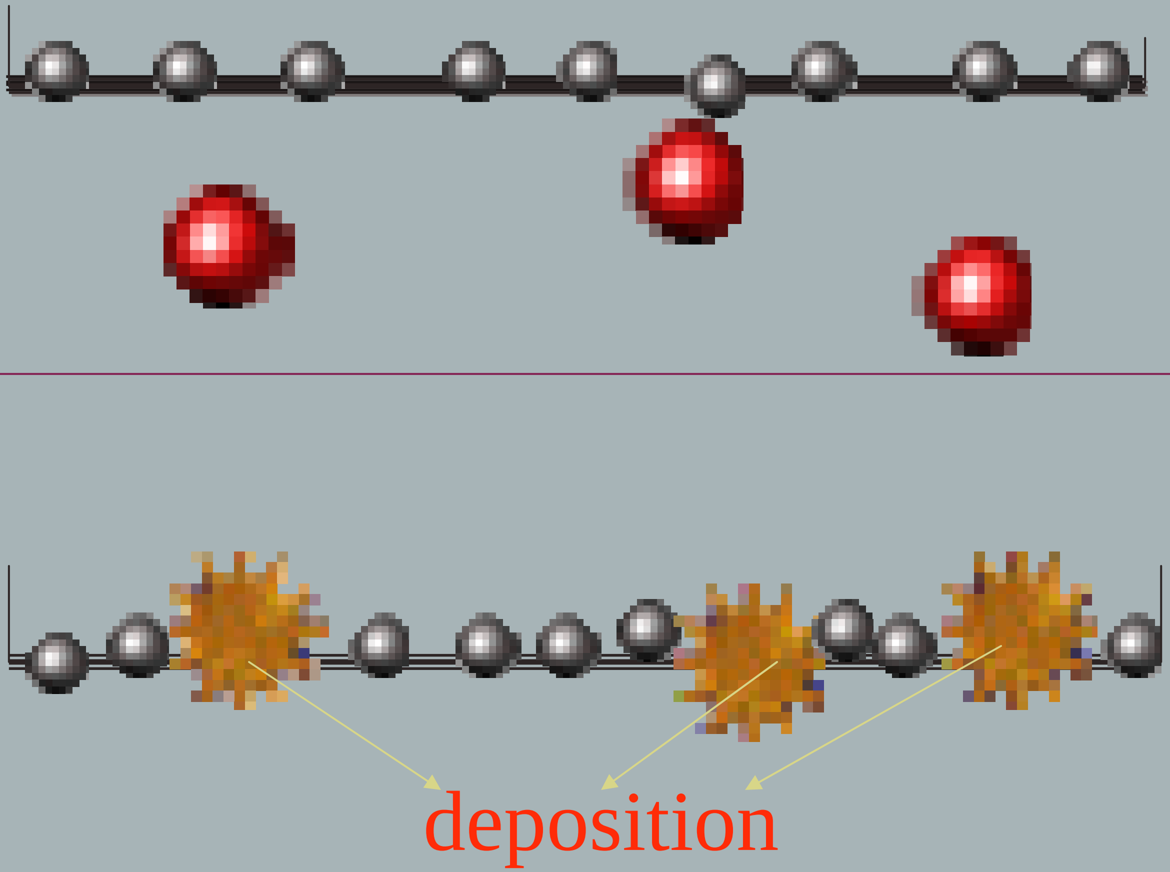
Excited and not cesium

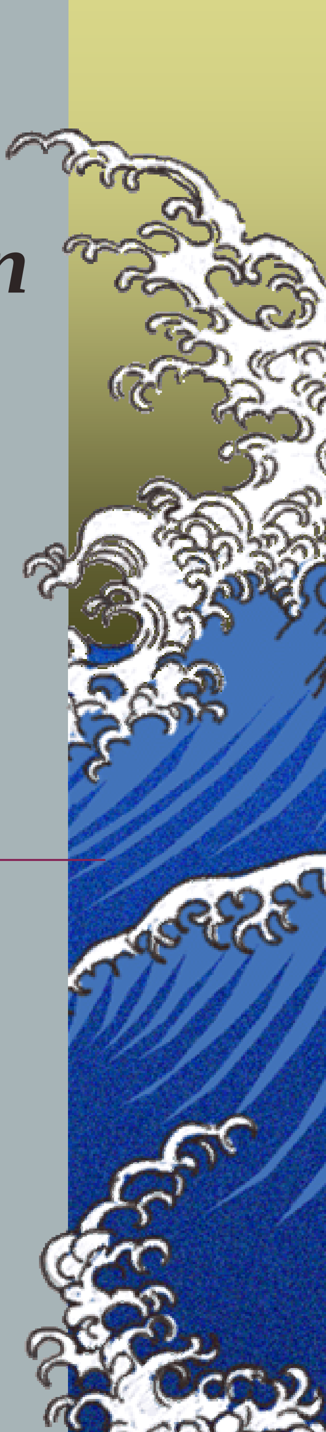
Excited and not hydrogen2

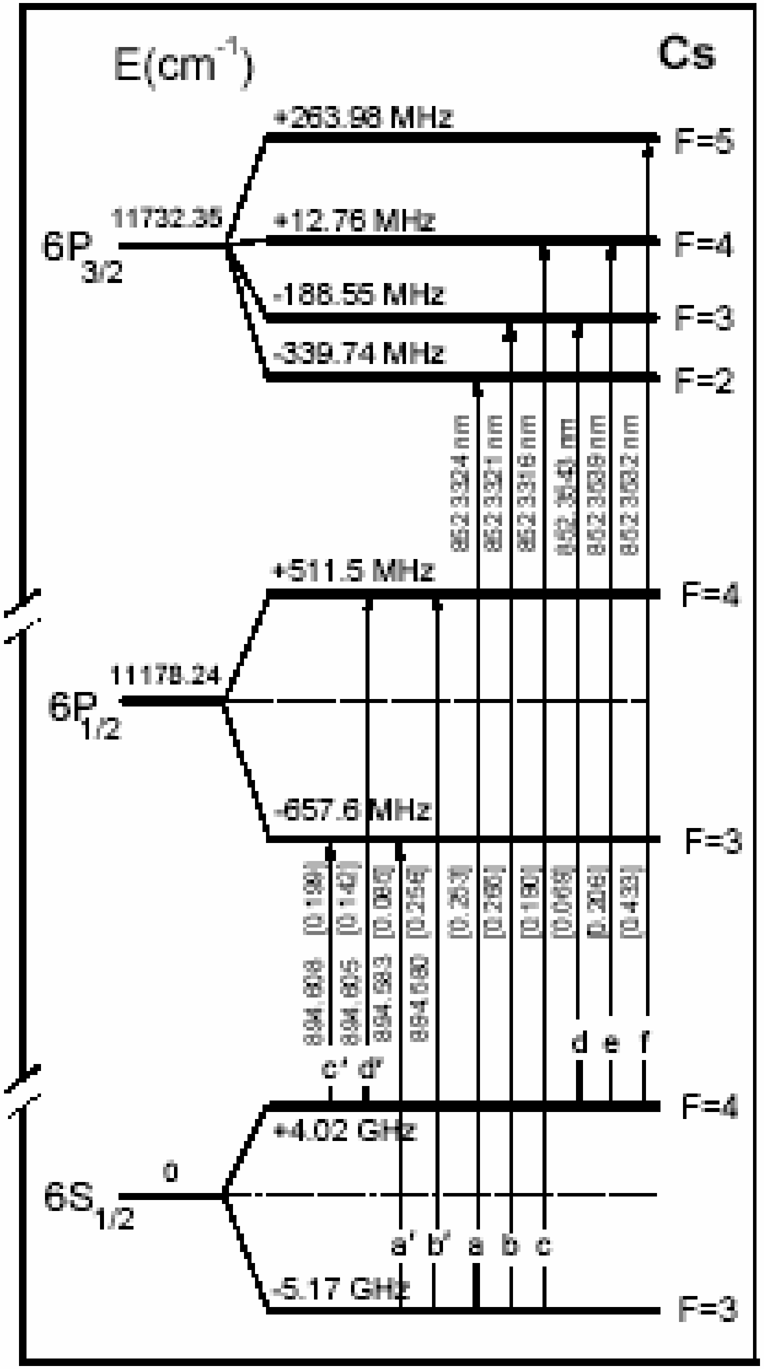


CsH and H

The reaction may also proceed on the cell wall, when an atom (rubidium in our experiments) being excited by laser radiation collides with the cell wall







In experiments with cesium vapor we exploited the group of hyperfine transitions F=4→F′=3;4;5 of D$_2$ line of Cs atom.

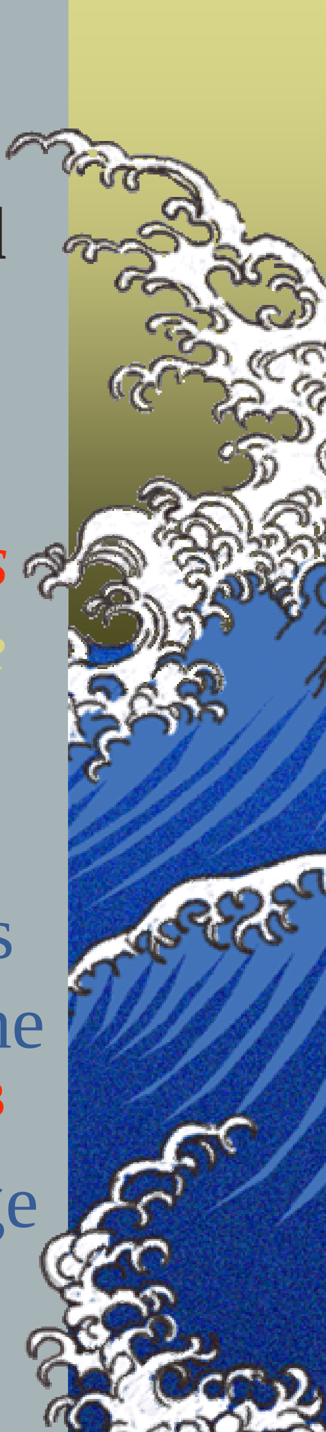
Scanning (fast or slow) the diode laser radiation through this group we measured the absorption in and fluorescence from the vapor versus the laser intensity

We used “pure” cesium cells and those filled with 20 Torr Hydrogen.

The fast and slow scan regimes determine the time of interaction of cesium atoms with laser radiation, which determines the probabilities of laser induced processes, such as saturation, optical pumping, etc.; this time is essentially different for cases of absence and presence of hydrogen.

We have the following relevant time scales: time of flight of atoms through the beam and time of scan, within which the radiation is in resonance with the transitions above; these times determine the time of interaction.

The physics is as follows



When the scan is fast, the time of interaction between Cs atoms and laser radiation equals nearly the time of scanning 12 µs, both with and without hydrogen

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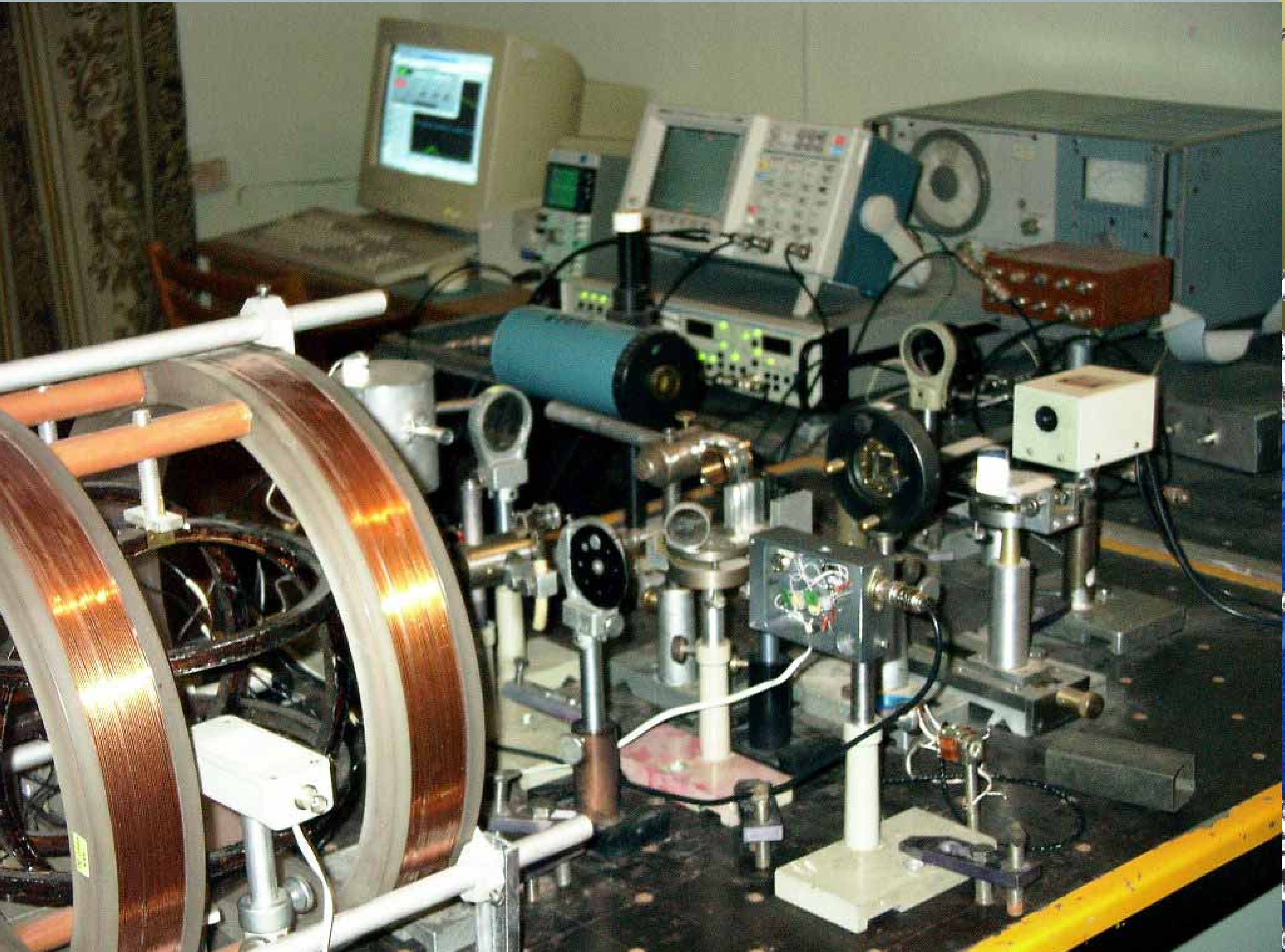
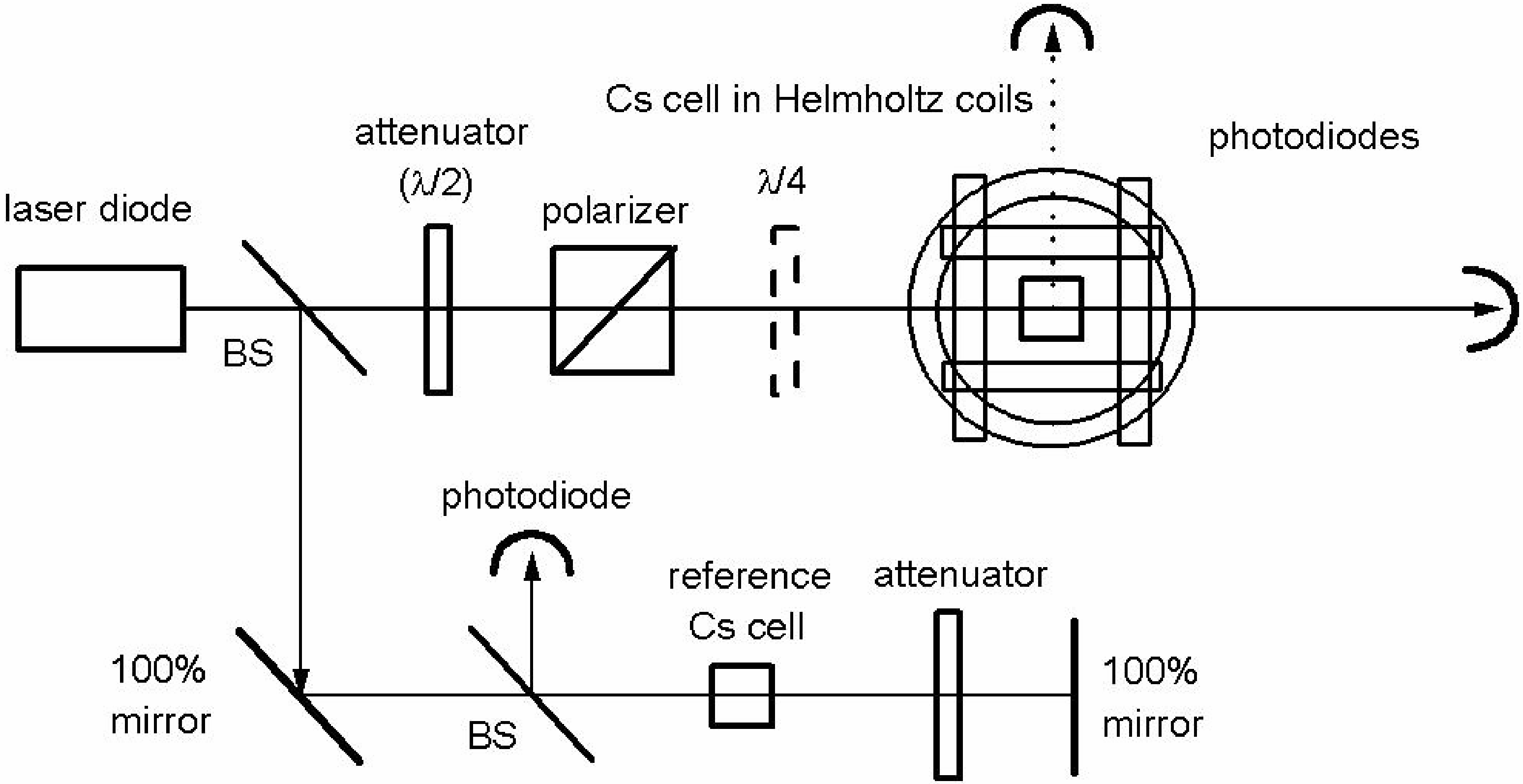
When the scan is slow, this time equals nearly the time of flight of atoms through the laser beam 15 µs in case of pure vapor and 30 ms in case of presence of hydrogen:

The cause: density of hydrogen in our experiments exceeds by 7 orders of magnitude that of cesium: the number density of cesium vapor was $2.7 \cdot 10^{10}$ cm^{-3} (room temperature); the intensity varied in the range $1.5 \cdot 10^{-4}$ to 40 mW/cm^2

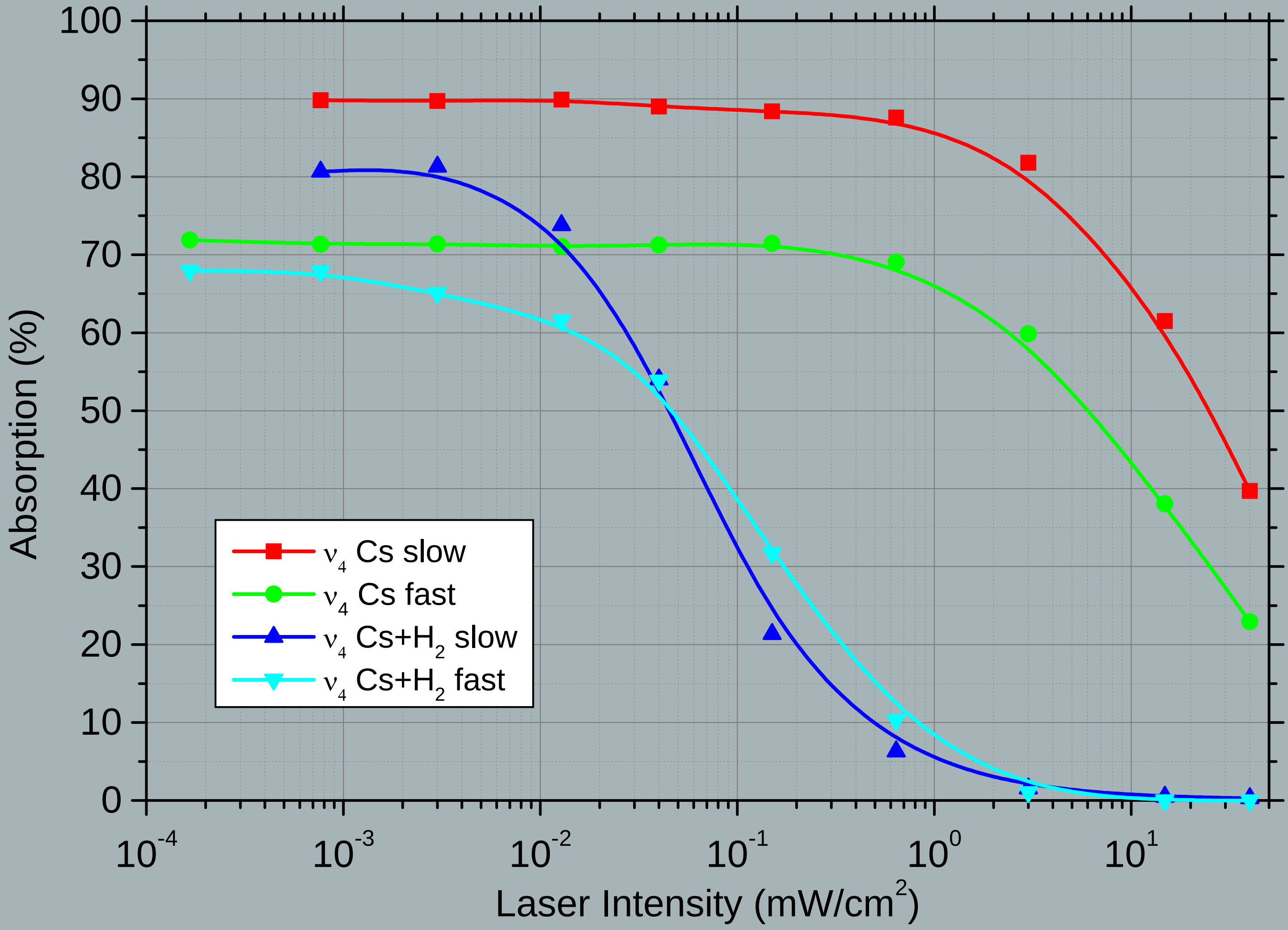
A fragment of experimental setup for cesium measurements



The diagram of the Cs setup



Results of absorption (transmission) measurements



What is observed

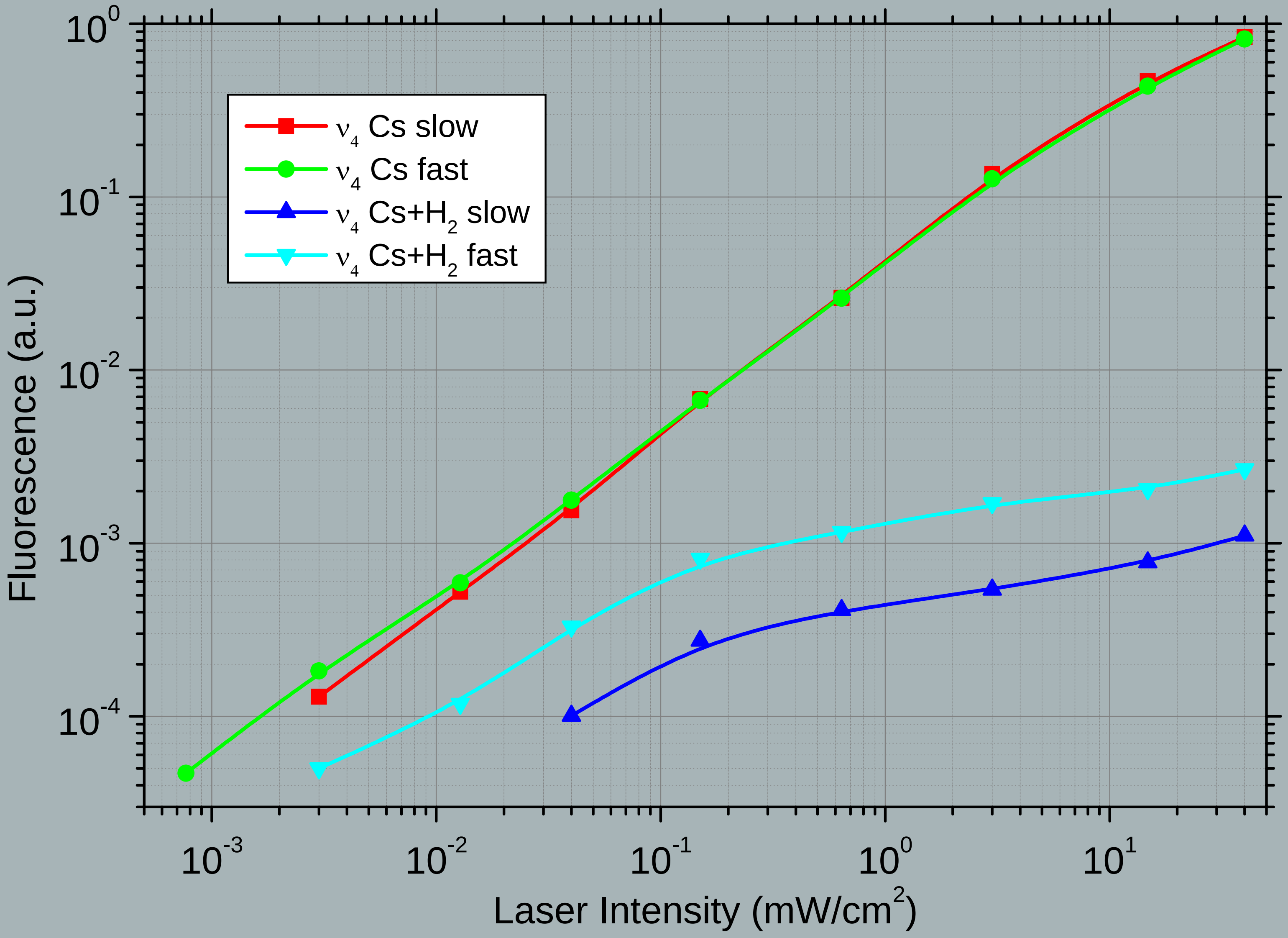
In the absence of hydrogen absorption remains unchanged going to saturation for intensities higher than 1 mW/cm^2

In the presence of hydrogen absorption drops essentially with the increase of intensity

Fast and slow scan cases do not practically differ in the absence of H_2, but exhibit significant difference in the presence of that

We explain this by the decrease of the number density of Cs atoms because of formation of cesium hydride molecules

Results of the fluorescence measurements



What is observed

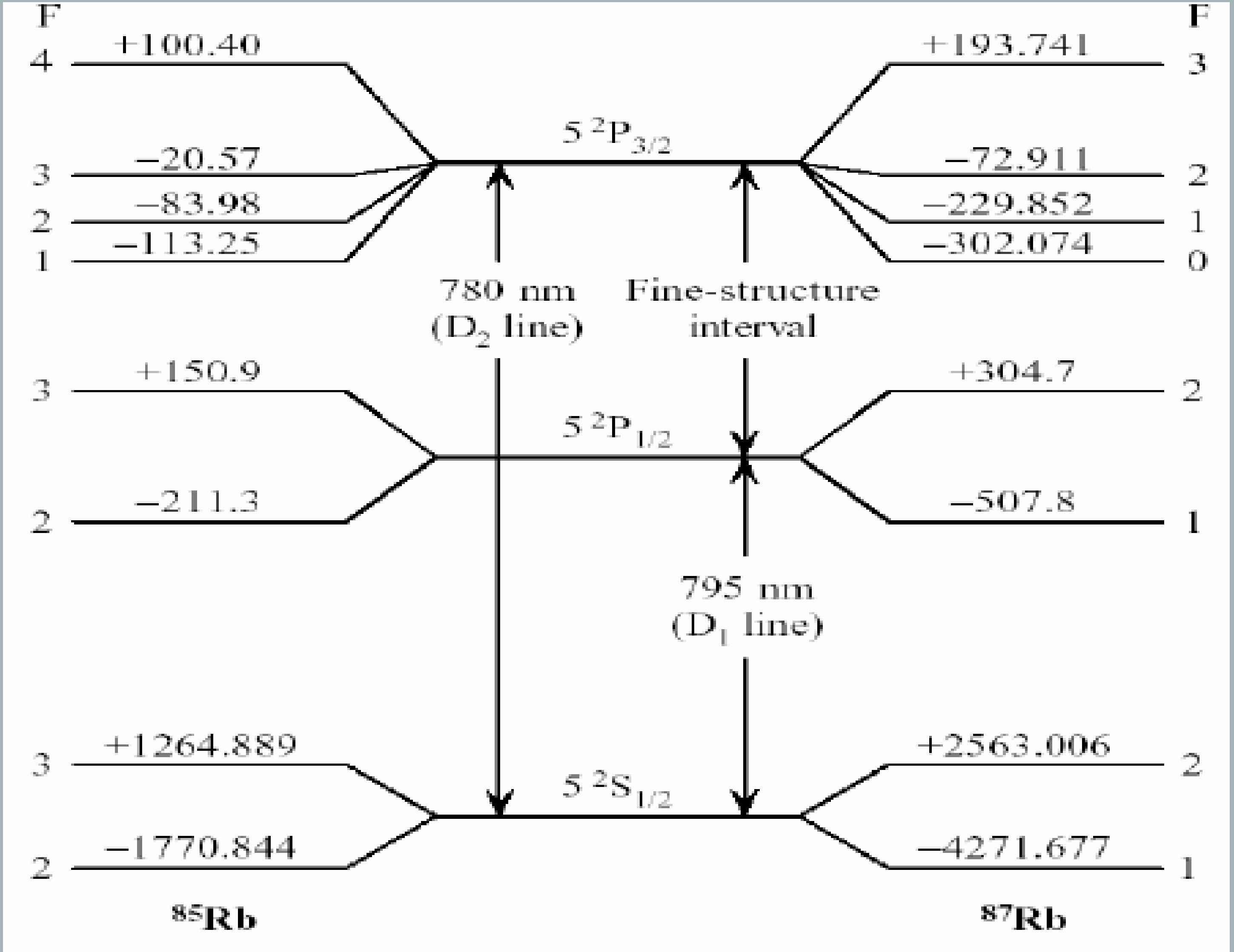
In the absence of hydrogen fluorescence rises up to highest intensities 40 mW/cm^2

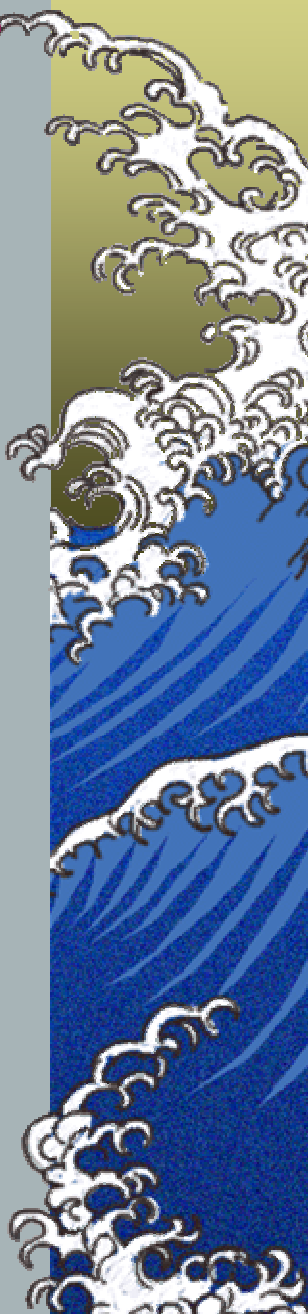
In the presence of hydrogen fluorescence drops by several orders of magnitude with the increase of intensity

Fast and slow scan cases do not practically differ in the absence of H_2, but exhibit significant difference in the presence of that

We explain this again by the decrease of the number density of excited Cs atoms because of formation of cesium hydride molecules and the optical pumping

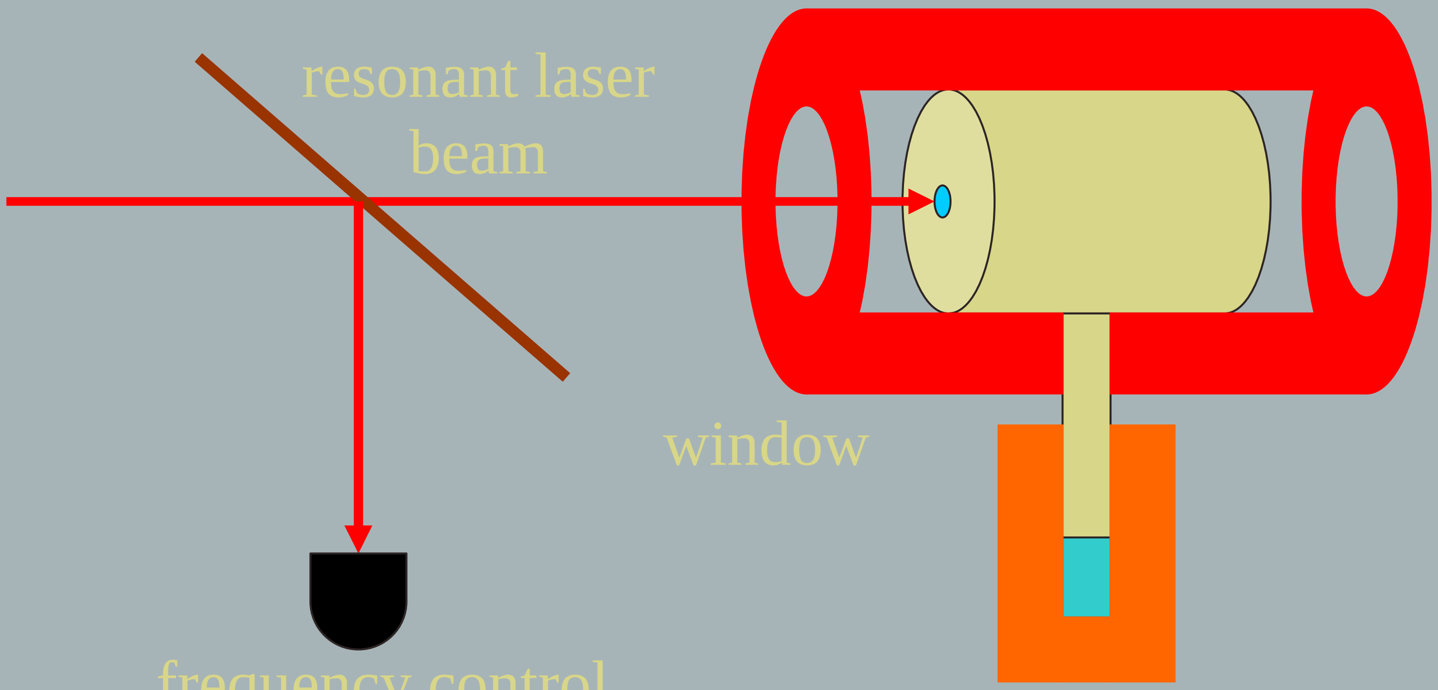
In experiments with rubidium vapor we exploited hyperfine transitions of the D_1 and/or D_2 lines of the Rb atom. Here is the relevant energy level diagram





A fragment of experimental setup for rubidium measurements

Simplified diagram of the Rb setup



Rb cell in the oven

resonant laser beam

window

frequency control

side arm

Three types of rubidium vapor cells were used.

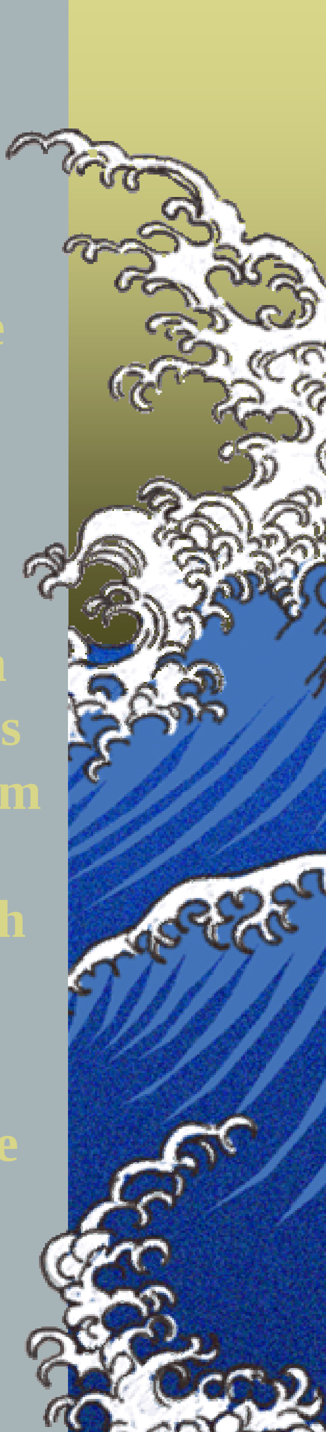
The first type were the cells purified up to 10^{-6} T vacuum and containing only rubidium vapor.

The second type were the rubidium vapor cells purified in the same way as the first one, but filled with hydrogen.

The third type cells were purified to the 10^{-3} T vacuum without hydrogen.

The cells of all three types were exposed to the diode laser radiation from 30 min to 2.5 hours. The radiation in different experiments was either tuned to a specific D_2 hyperfine transition of a specific rubidium isotope or scanned through the entire spectrum of all hyperfine transitions of D_2 line (in this case the deposition if any contained both isotopes).

The windows of the cells of all types were made of either glass or sapphire. Two or more cells of each type were used. The temperature of the cells varied in the range 190-210°C in the side arm and 220-240°C in the cell body. This temperature range corresponds to the number density of rubidium atoms on the order of 10^{14} cm^{-3}.



What is observed

The first type cells:

Strong blue emission in the direction opposite to that of laser radiation. This emission was observed as a spot inside the red ring.

No deposition on both cell windows and side surface.

The second type cells:

No backward blue scattering.

A deposition on both the sapphire windows and the glass side surface (cylindrical) when irradiated sufficiently long.

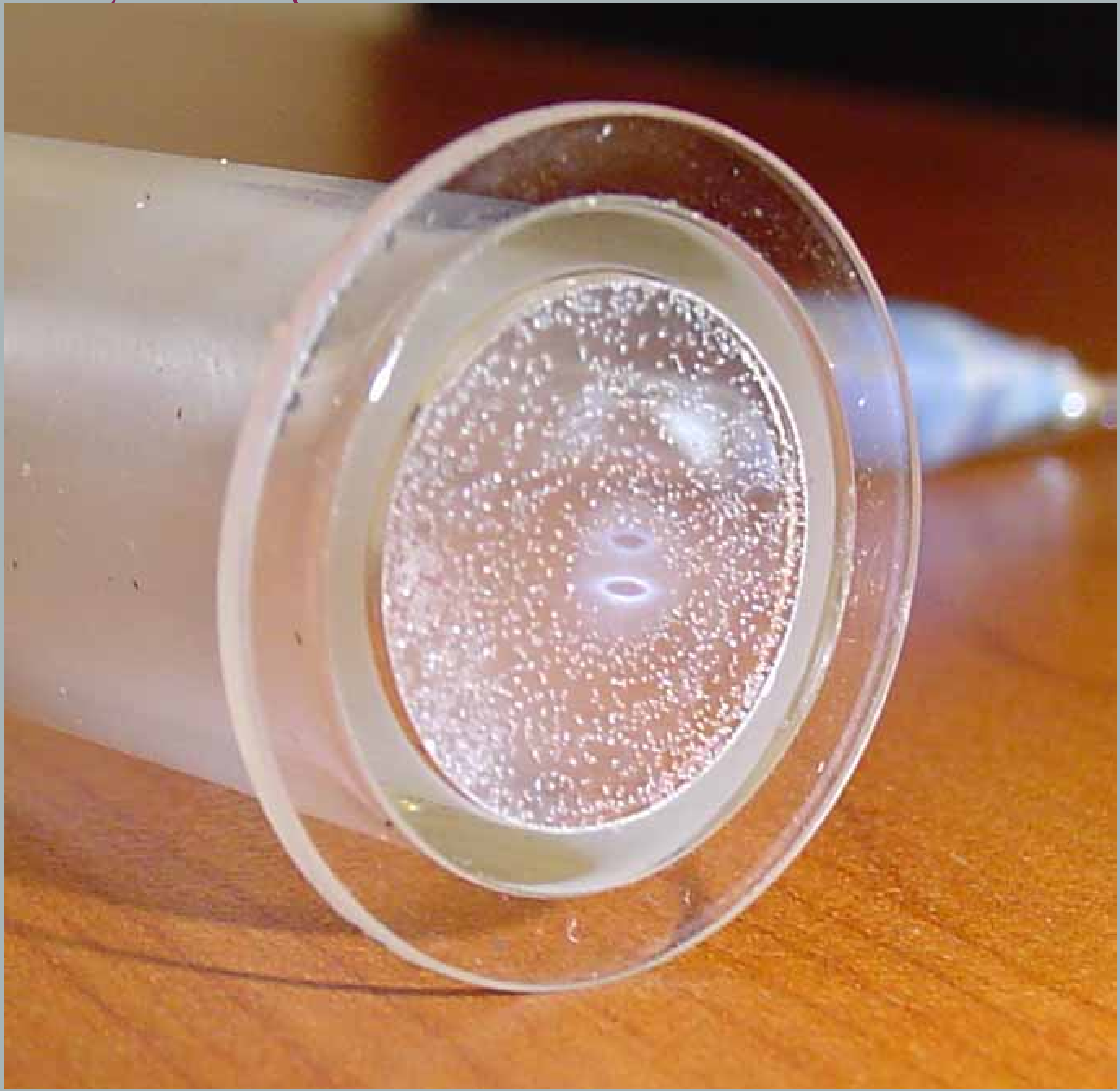
The third type cells:

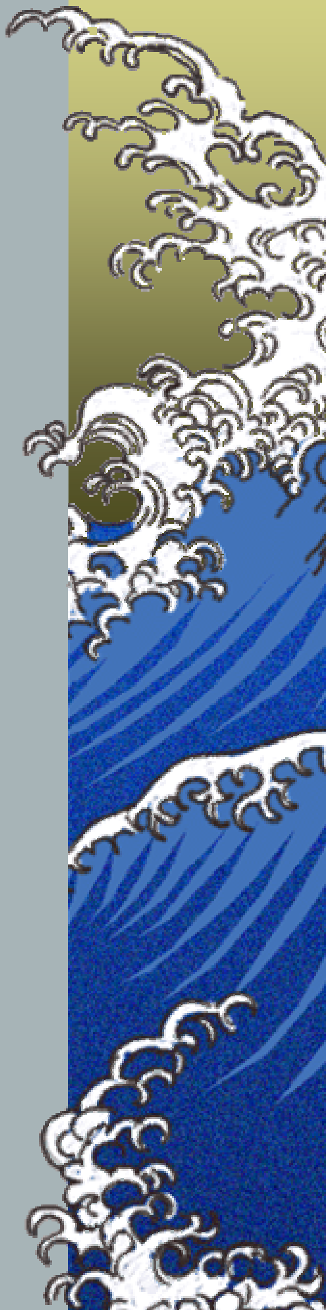
Bright blue scattering.

A deposition.

Example:

10^{-3} vacuum, no hydrogen, 240°C, half-hour irradiation, $F_g=1 \rightarrow F_e=0; 1; 2$ transitions of the ^{87}Rb.

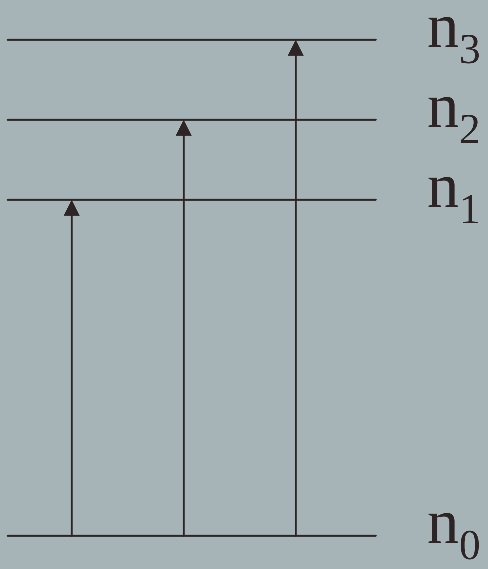




Analysis allow us to conclude that the deposited spot consists of mainly the rubidium hydride (along with other hydrogen containing compounds) which is formed in reactions between the laser excited rubidium atoms and the hydrogen (or water) molecules adsorbed on the inner surface of the cell.

The blue backward scattering is caused by the energy pooling processes and is suppressed by the presence of hydrogen in the cell volume.

Model and calculations



w_i,

γ_i, γ_i',

λ_a, λ_m,

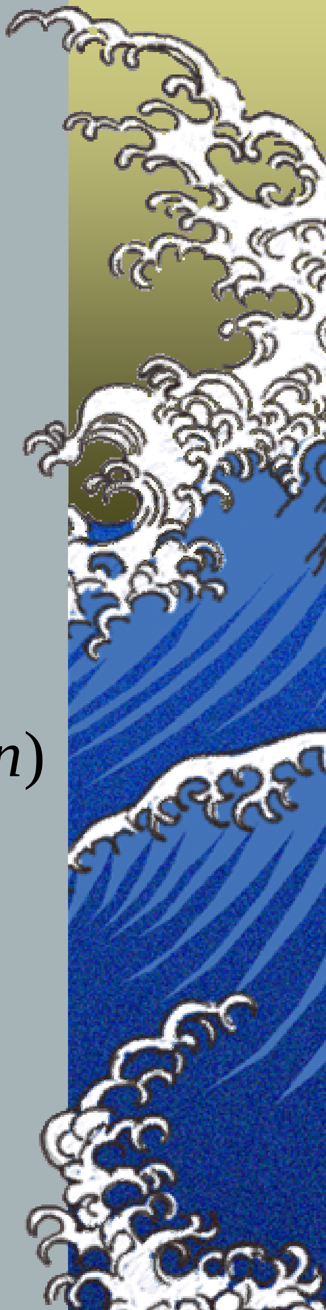
k_1, k_2

$$\dot{n}_0 = -\sum_{i=1,2,3} w_i(n_0 - n_i) + \sum_{i=1,2,3} \gamma_i n_i + k_1 m \sum_{i=1,2,3} n_i - \lambda_a (n_0 - n)$$

$$\dot{n}_i = w_i(n_0 - n_i) - \gamma_i' n_i - k_1 m n_i - k_2 m^* n_i$$

$$\dot{m}^* = (k_1 m - k_2 m^*) \sum_{i=1,2,3} n_i - \lambda_m m^*$$

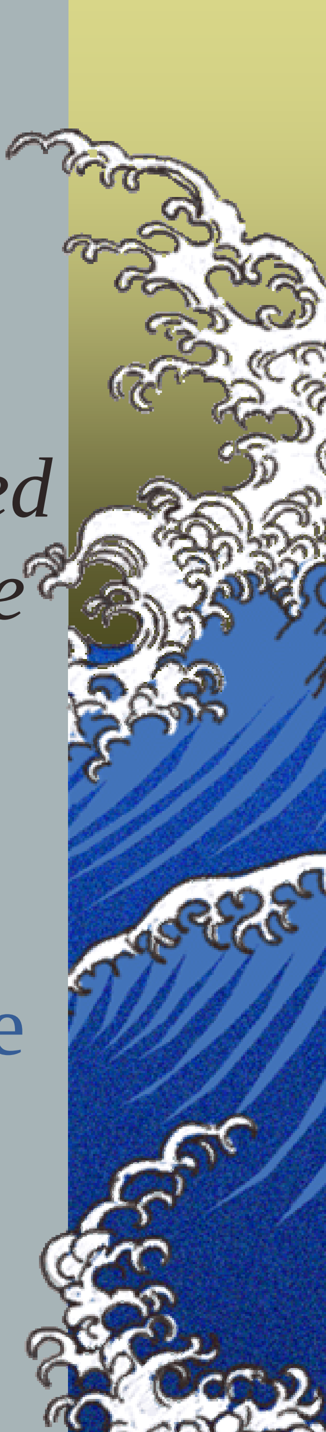
$$\dot{m} = -k_1 m \sum_{i=1,2,3} n_i$$



Computations with the model described above were made by assuming $m=\mathrm{const}$,

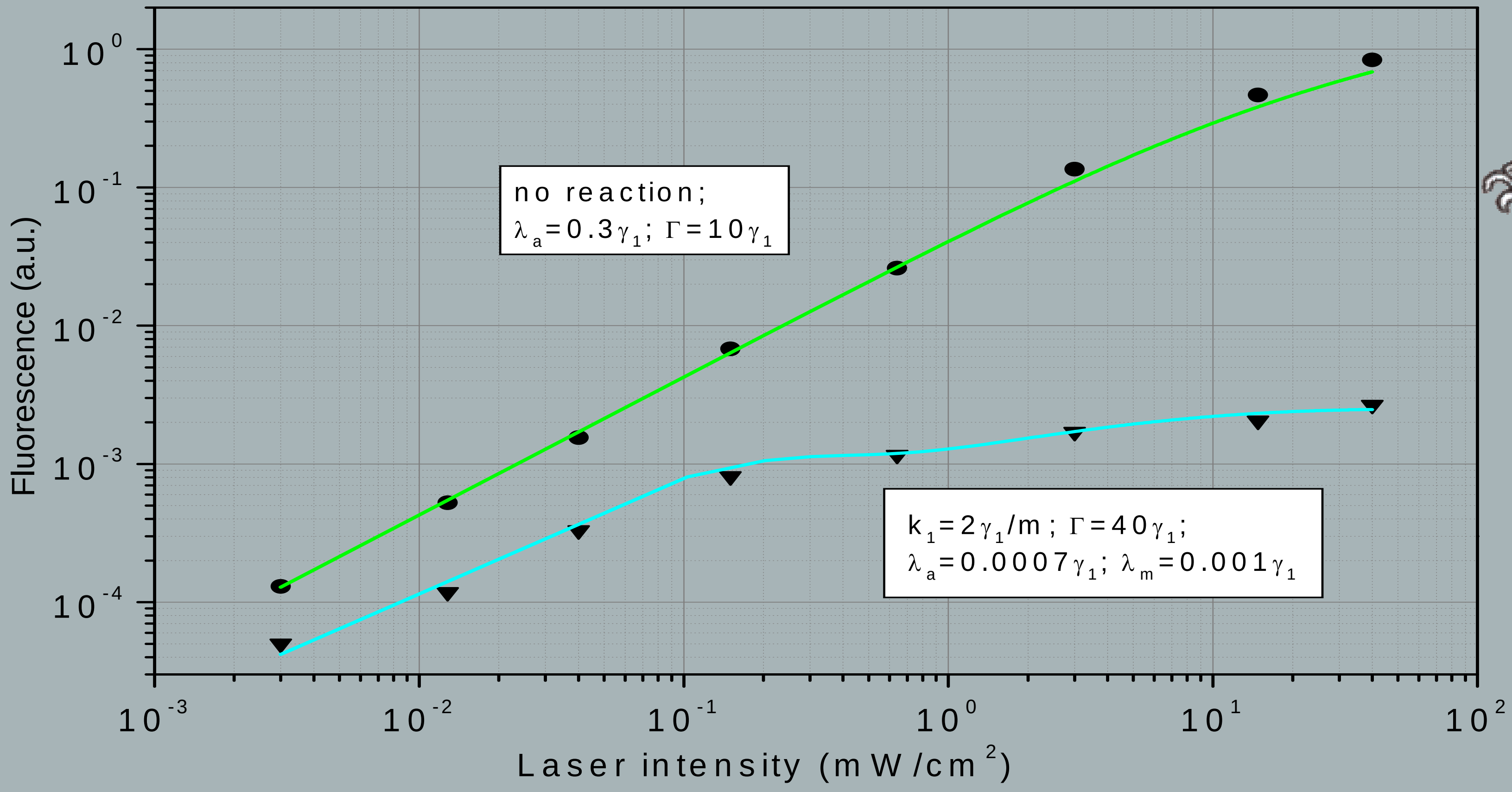
since the number density of hydrogen molecules exceeds by many orders of magnitude that of cesium atoms and the excited hydrogen molecules. We computed from these equations the integral fluorescence as a function of the laser intensity.

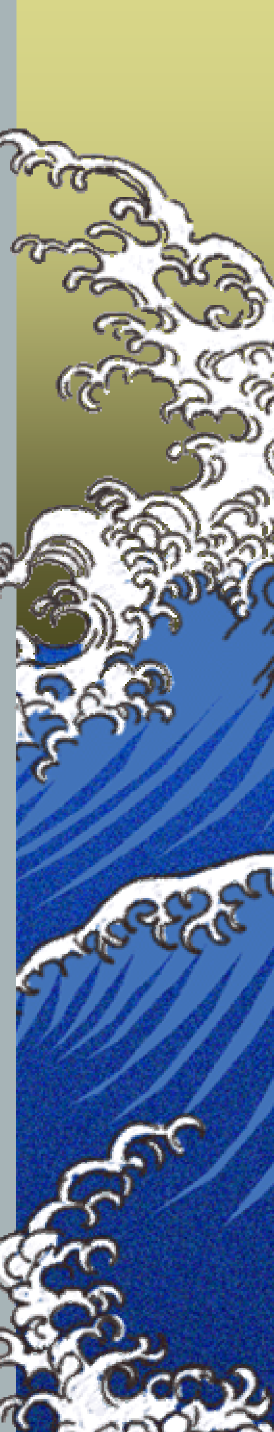
In case of pure atomic vapor we have $m=0$, $k_{1,2}=0$, and the collisional contribution into the overall linewidth is negligible. The presence of hydrogen increases the overall linewidth and reduces the leaving rates.



Results of computations in these conditions:

solid lines – theory, points - experiment

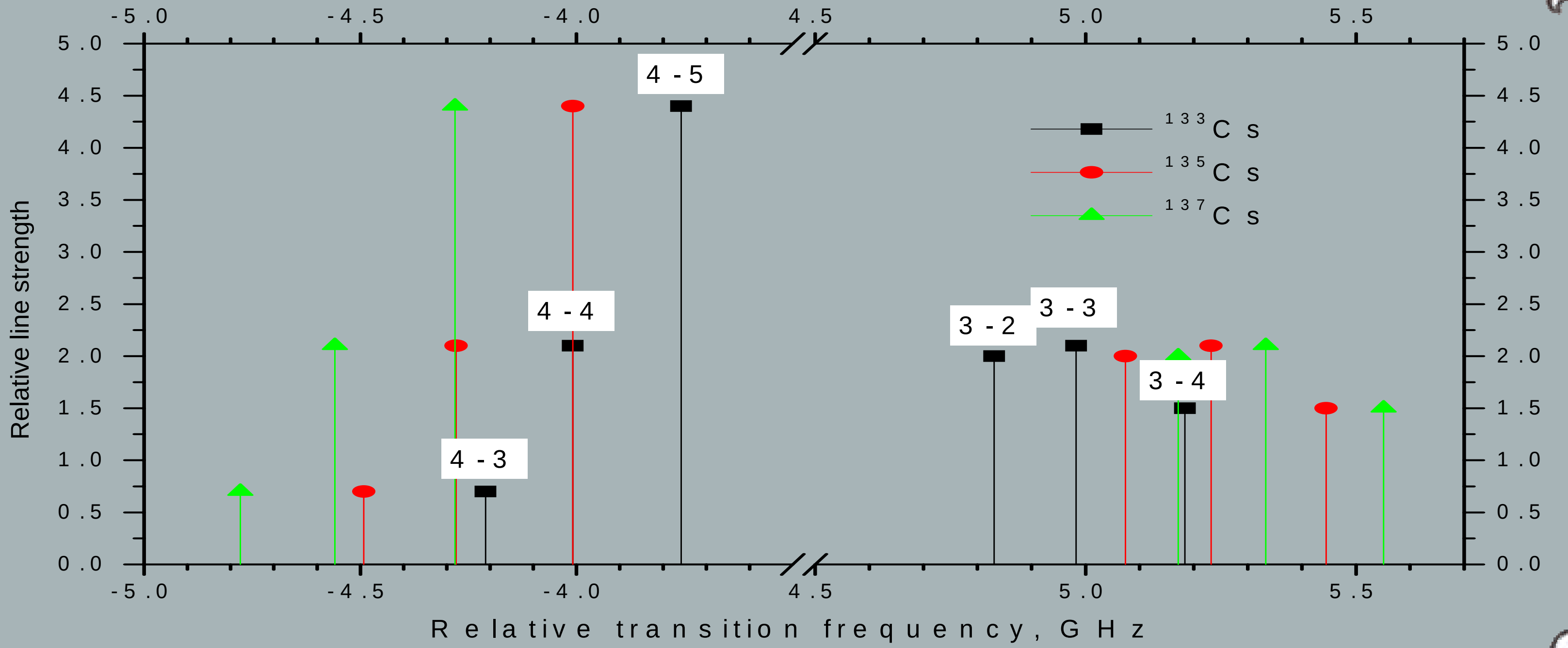




$$k_{eff}=(1\pm0.3)\cdot10^{-10}\ \mathrm{cm^3s^{-1}}$$

For future application of the presented results we calculated the energy levels of all Cs isotopes (since the data available in literature have discrepancies).

The diagram of energy levels and transition strengths for D_1 line hyperfine transitions of three of these isotopes – 133, 135, 137, are presented below



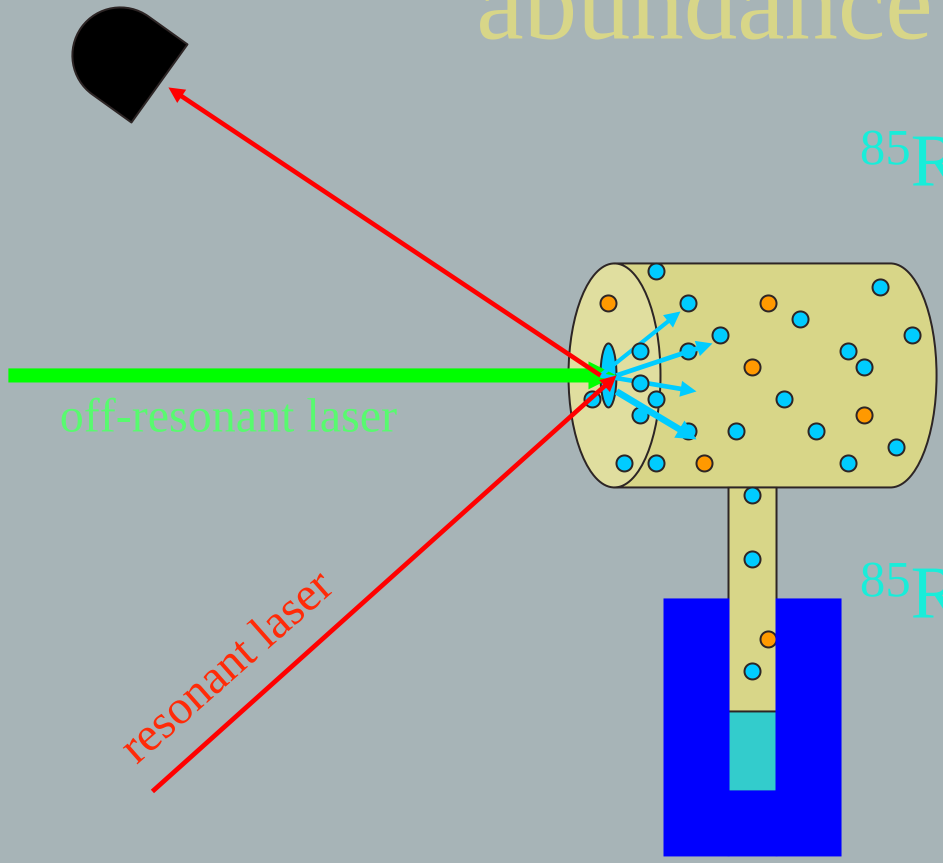
Note finally that for the determination of abundances of isotopes (Rb) and, thus, the enrichment degree, we elaborated and employ mainly two techniques

The first: *selective reflection from the deposited layer on the cell window with measuring the peaks caused by either isotope*

The second: *recording fluorescence from the close-to-window volume of the cell in conditions when a second, fixed-frequency strong laser destroys the deposited layer*

Preliminary results for Rb: abundance of ^{85}Rb may be raised up to 6 times

Selective reflection technique for determination of isotopic abundance



^{85}Rb/^{87}Rb = 72/28

^{85}Rb/^{87}Rb = 88.5/11.5

98.4/1.6

Conclusion

Formation of cesium hydride takes place at illumination of a room-temperature mixture of Cs vapor with hydrogen by a laser radiation tuned to the resonance D_2 line of atomic Cs. The rate constant of a two-step reaction is evaluated by comparing the experimental results for the laser-intensity dependence of the integral fluorescence with those of calculations based on rate-equation model.

Both the (extremely simple) experimental technique and the model may be used for evaluation of rate constants of chemical reactions between different interacting components.

So, if selective excitation condition is satisfied, the photochemical reactions may be directly used for the highly selective extraction of chosen alkali isotope from the mixture.

In heated rubidium vapor cells a solid deposition is observed on the inner surface of the cell under laser irradiation exciting rubidium atoms. This may also be used for isotope separation,

Thank you

very much

for attention