

Two-photon resonance on the D_2 line of Cs enhanced by optical pumping

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Abstract. Sub-natural-width N -type resonance, observed in the D_2 line of Cs vapor has been reported recently. Three-photon, bi-chromatic excitation of Cs atomic vapor with 20 Torr of Neon was performed and a sub-natural-width resonance was observed, superimposed on the absorption profile of a set of hyperfine transitions starting from single hyperfine ground level. Here we report the study of a different three-photon, couple-probe- excitation scheme that allows the observation of a sub-natural-width resonance situated on a flat background, well outside the D_2 line absorption spectrum of Cs. The lack of a noisy background could be advantageous for many applications related to the narrow N resonance. In addition, a new broader feature of enhanced transparency is demonstrated that is centered at the ground state level that does not interacting with the coupling beam. This enhanced transparency resonance can be attributed to the atomic population loss in a narrow spectral interval occurring due to excitation of energy levels situated above the 6^2P levels.

1. Introduction

Coherent laser spectroscopy of alkali vapor contained in thermal optical cells is widely used for various applications, including frequency references, atomic clocks, precise optical magnetometers, slow and stored light, and photonic sensors based on coherent population trapping (CPT) and electromagnetically induced transparency (EIT) resonances, etc. For those applications, various research groups are still exploring suitable approaches aimed at a profitable combination of the advantages of CPT/ EIT with other processes. Optical systems based on Rb atoms, in which narrow band N -type resonances appear, have been extensively studied recently [1-4]. Such N -resonances can be considered as a type of three-photon resonance where a two-photon Raman excitation is combined with a resonant optical pumping field.

A sub-natural-width N -resonance has recently been observed in Cs vapor [5]. The two-photon Raman excitation in Cs atoms is less effective than in Rb vapor due to the higher ground-state hyperfine splitting in Cs. Nevertheless, a high-contrast N -resonance have been observed in Cs vapor that can be attributed to the higher rate of the optical pumping process in Cs than in Rb. Three-photon, bi-chromatic excitation of Cs atomic vapor, contained in a 10 mm long cell with 20 Torr of neon has been performed and successfully applied to measurement of the magnetic field over a large range [5]. A coupling laser was fixed at frequency 9.2 GHz lower than the position of the absorption profile of the $F_g = 4$ set of transitions, while the probe laser was tuned over the D_2 line ($\lambda = 852$ nm).



A narrow high-contrast enhanced absorption N -resonance has been observed in the probe light profile, superimposed on the absorption profile of the $F_g = 4$ set of transitions.

In this communication, we report a systematic study and analysis of a less explored scheme that has been applied to Rb vapor [1]. In the case of the D_2 line of Cs, such an approach allows the observation of a sub-natural-width resonance on a flat background, well outside of the D_2 line spectrum. Here, bi-chromatic excitation is also used, but the coupling laser frequency is fixed within the absorption profile of the $F_g = 3$ or $F_g = 4$ set of transitions, and the transmission of the coupling beam is measured while tuning the probe beam frequency over the D_2 line.

2. Experimental set up for sub-Doppler and sub-natural width resonance study

Figure 1 shows a schematic diagram of the experimental setup we used. The two narrow-band distributed feedback (DFBs) diode-lasers systems used in our experiment, are built as in Ref. [5] using two DFB diodes, both with an emission spectral width of about 2 MHz (FWHM). The two

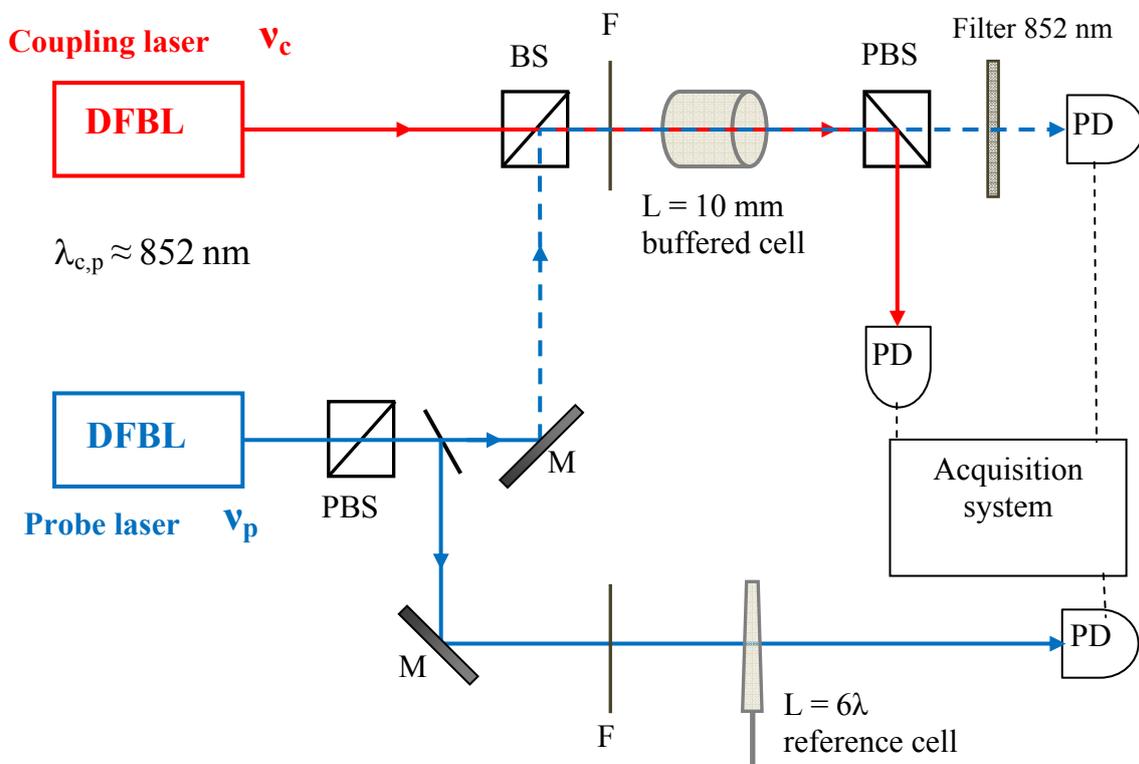


Fig.1 Schematic diagram of the experiment: DFBL - distributed feedback diode-laser: coupling and probe, PBS -polarizing beam splitters, PD - photo diodes, M-mirrors, F - neutral optical filters, interference filter of a wavelength of 852 nm, and acquisition system. Sub-natural-width resonance is registered in the transmission of the $L = 10\text{mm}$ buffered cell. The $L = 6\lambda$ cell provides a reference spectrum.

laser systems show a mode-hop-free laser frequency detuning that largely exceeds the absorption spectrum of the Cs D_2 line. The coupling laser has a fixed frequency ν_c , while the probe laser with frequency ν_p is tuned over the D_2 line. Both laser beams have spot size $3 \times 2\text{mm}$ and are carefully superimposed on a beam splitter BS and then directed at the cell containing atomic vapor. The intensities of both lasers falling on the $L = 10\text{ mm}$ cell are equal, $I = 500\text{ mW/cm}^2$. The polarizations of the coupling and probe lasers are linear and mutually orthogonal. The $L = 10\text{ mm}$ long optical cell

is filled with atomic Cesium with 20 Torr of neon gas added that serves as a buffer. Part of the probe radiation is directed to an additional cell with thickness $L = 6\lambda$, which is used as a frequency reference [5]. In the experiments, the transmission of the coupling beam is registered with the probe beam scanned in a large frequency interval over the D_2 line absorption. The probe beam is separated from the coupling laser by a polarizing beam splitter. The broad transmission spectrum of the coupling laser is measured with a sub-natural-width two-photon resonance superimposed on it, for different Cs vapor densities and frequency positions of the coupling beam situated within the $F_g = 4$ transmission profile.

3. N_p resonance within absorption profile, observed in the probe beam transmission

For clarity, the formation of the N -type resonance reported in Ref. [5] is briefly discussed. A schematic diagram of the relevant atomic levels and optical transitions is shown in Fig.2. The lower levels used for formation of the required Λ system are the two ground $F_g = 3, 4$ levels and the upper level is $6P_{3/2}$ which consists of four hyperfine levels $F_e = 2, 3, 4, 5$. The probe laser frequency ν_p is tuned over the $F_g = 4 \rightarrow F_e = 3, 4, 5$ set of transitions and the coupling laser frequency ν_c is fixed. In the probe beam transmission spectrum, an N -type resonance involving two-photon process has been observed when the two-field frequency difference was $\nu_p - \nu_c = \Delta$, where Δ is the splitting ($\Delta = 9.2$ GHz) of the electronic ground state hyperfine levels. Due to the fact that the resonance is observed in the probe beam transmission, further on we denote it as N_p resonance.

An example of an experimentally observed N_p resonance is presented in Fig. 3. Here, the frequency of the coupling laser is fixed in the lower-frequency wing of the D_2 line absorption and the transmission of the probe laser is measured as a function of its frequency detuning. The absorption spectrum of $L = 6\lambda$ cell was also measured and used as a frequency reference. The probe beam frequency was scanned in a spectral region broader than 30 GHz, thus providing the transmission spectrum of the D_2 line and the frequency position of the coupling beam. A good contrast, sub-

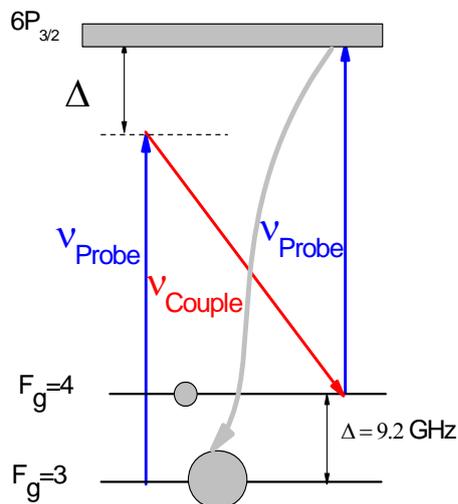


Fig.2 Relevant transitions of D_2 line of Cs. Probe frequency ν_p is scanned over the $F_g = 4 \rightarrow F_e = 3, 4, 5$ transitions, while the coupling frequency ν_c is fixed at a lower frequency [5].

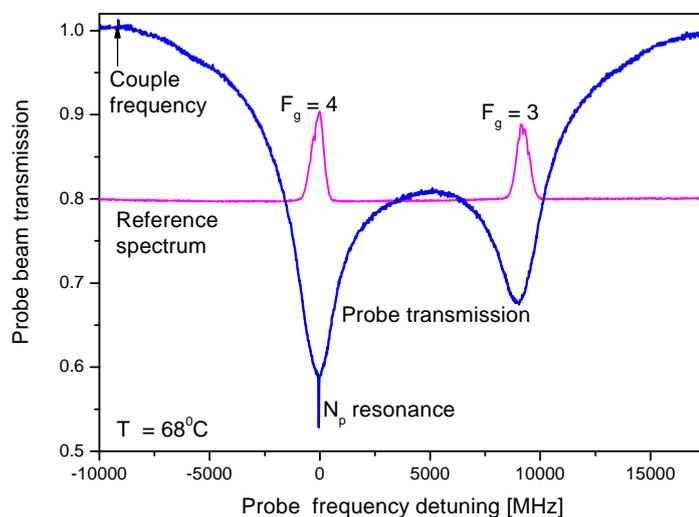


Fig.3 Large detuning of the probe beam frequency over the coupling-probe beating signal (showing the fixed-frequency position of the coupling beam) and both sets of hyperfine transitions starting from $F_g = 4$ and $F_g = 3$ levels (blue). Transmission spectrum of the D_2 line involving the N_p resonance situated within the $F_g = 4$ absorption set (magenta). The spectrum of the $L = 6\lambda$ cell is used as a frequency reference only.

natural (~ 1.5 MHz) line width N_p resonance is observed at the center of the absorption profile. The N_p -resonance frequency position is shifted from that of the coupling laser by the electronic ground-state hyperfine level splitting Δ . The atomic source temperature is kept at 68°C . Under this condition the density of cesium atoms was approximately 1.7×10^{12} atoms/cm³.

4. Formation of narrow coherent resonance in the transmission of the fixed-frequency coupling beam

In the experimental study presented here, the frequency of the couple beam ν_c is fixed within the absorption profile of the $F_g = 4$ or $F_g = 3$ set of transitions. Now again the frequency of the probe beam is scanned over a large spectral interval. However, the transmission of the fixed-frequency coupling beam is measured.

Mainly, we consider the case where the coupling laser frequency is within the $F_g = 4$ absorption profile. The corresponding atomic levels, as well as the two-photon and the single-photon transitions, are shown in Fig. 4. The coupling beam is in resonance with the absorption profile of the $F_g = 4$ set of transitions. Under this condition, Cs atoms are accumulated by the coupling laser in the $F_g = 3$

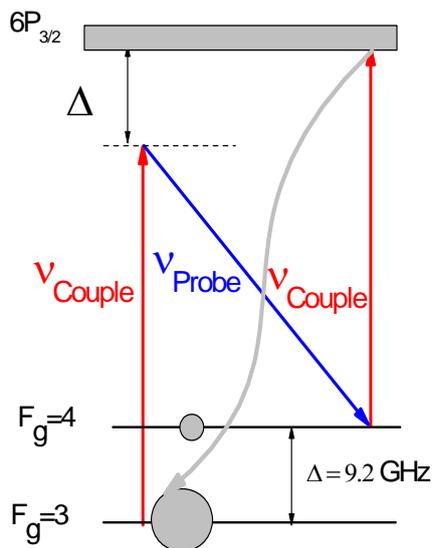


Fig. 4 Probe frequency ν_p is scanned over a large spectral interval, while ν_c is fixed within the $F_g = 3,4$ set of transitions.

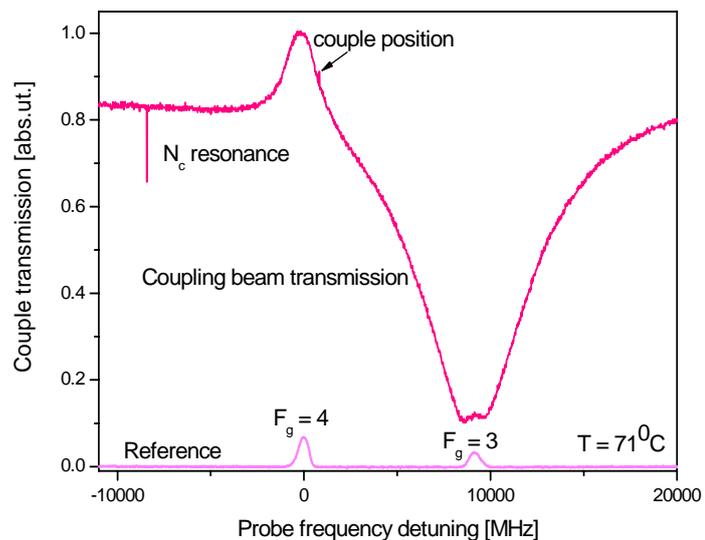


Fig. 5 Transmission spectrum of the fixed-frequency coupling beam, as probe is scanned. The N_c resonance is observed on a flat background, outside the absorption profile.

level due to the hyperfine optical pumping. During the probe beam detuning, when the Raman condition is not fulfilled, the $F_g = 4$ level is significantly depleted by the coupling beam. In case of two-photon Raman resonance, the atoms accumulated in the $F_g = 3$ level are transferred back to the $F_g = 4$ level by the coherent two-photon process, and are detected as enhanced absorption of the coupling beam. However now, the sub-natural-width enhanced absorption feature is observed for probe light frequency much lower than the frequencies of the $F_g = 4$ set of transitions, that is, outside the D_2 line absorption profile.

In Fig. 5, the experimentally observed sub-natural-width resonance is shown for cell temperature of $T = 71^\circ\text{C}$ (i.e. Cs atom density is 2.1×10^{12} atoms/cm³). Starting from the left side of the figure, first the very narrow resonance N_c can be seen, which is the enhanced absorption experienced by the coupling beam. The observed N_c resonance is well outside of the absorption profile of the $F_g = 4$ set

of transitions. The frequency ν_p of the probe beam at which the N_c resonance is observed can be determined by the expression $\nu_p = \nu_c - \Delta$.

When the probe beam is tuned in resonance with the $F_g = 4$ set of transitions, the coupling beam experiences almost 100% transparency, due to the simultaneous depletion of the $F_g = 4$ level by the coupling and probe beams. Further increase in the probe frequency results in a reduction of the coupling beam transmission. The minimum transmission (i.e. maximum absorption) of the coupling beam is observed when the probe frequency is tuned to the $F_g = 3$ set of transitions (Fig. 5).

Note that the complete transmission resonance of the coupling beam, centered at $F_g = 4$, is narrower than the absorption resonance observed when the probe beam is detuned across the $F_g = 3$ set. The absorption resonance is attributed to the strong repumping of the $F_g = 4$ level caused by the probe light during its detuning over the $F_g = 3$ set of transitions. Thus, the $F_g = 4$ level depletion is within a narrower frequency interval than its repopulation by the probe light scanned around the $F_g = 3$ transitions. Such a difference can be related to velocity-changing collisions [6] between Cs and Ne atoms that take place during the fluorescence decay of Cs atoms from excited states to the $F_g = 4$ level after being excited from the $F_g = 3$ level by the probe beam. Our experimental study has shown that the spectral width of the enhanced absorption spectral region is strongly influenced by the Cs atom density (Fig. 6). The following measurement is performed for the coupling frequency fixed

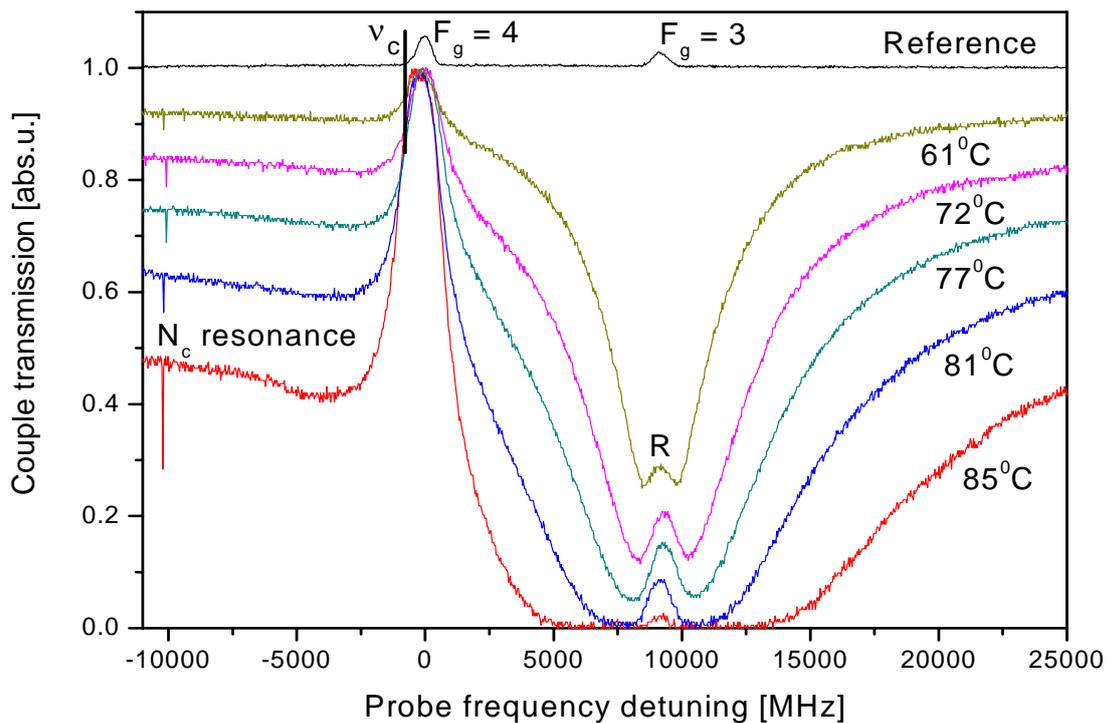


Fig. 6 Transmission spectra of the coupling beam when the probe is detuned over a spectral interval of more than 30 GHz. The coupling frequency ν_c is red detuned from the $F_g = 4$ absorption profile by 900 MHz. Here, in addition to the N_c resonance, a new much broader resonance (R) of reduced absorption appears superimposed on the coupling beam transmission, when the probe is resonant with the $F_g = 3$ level transitions. The spectra are shown for different atomic source temperatures; Cs atom densities vary from $(1 \div 5.6) \times 10^{12}$ atoms/cm³.

close to the absorption maximum of the $F_g = 4$ set of transitions (ν_c is detuned by 900 MHz lower than the $F_g = 4$ absorption maximum). The narrow N_c resonance is observed for a large interval of

atomic source temperatures, with strongly enhanced amplitude (up to 20% in transmission) for higher atomic density.

As the probe frequency approaches the absorption profile of the $F_g = 3$ set of transitions, and the atomic source temperature increases, the coupling beam experiences complete absorption over a very large spectral interval. The complete absorption of the coupling beam can be explained by the strong repumping to the $F_g = 4$ level caused by the probe beam.

In Fig. 6, we see a new broad resonance R of enhanced transparency centered at the $F_g = 3$ group. The behavior of the new resonance was investigated at different temperatures of the cell. Our experiments show that as the atomic density decreases, the amplitude of the new R resonance first increases significantly but then decreases at even lower temperatures, eventually disappearing. From Fig. 6, the following conclusion can be drawn: while the coupling absorption profile experiences strong broadening with increasing Cs atom density (for probe detuning around $F_g = 3$ set of transitions), the spectral width of the R resonance is practically unaffected by the atomic source temperature.

After studying the effect of the temperature, the next experiment was performed with a constant high atomic density. Here, the coupling frequency was changed, setting it at different frequencies lower than that of the $F_g = 4$ absorption maximum. Significant reduction of the N_c resonance amplitude takes place with the ν_c detuning from the $F_g = 4$ absorption profile maximum (Fig. 7). The amplitude of the N_c resonance is larger when the frequency ν_c is closer to the absorption profile

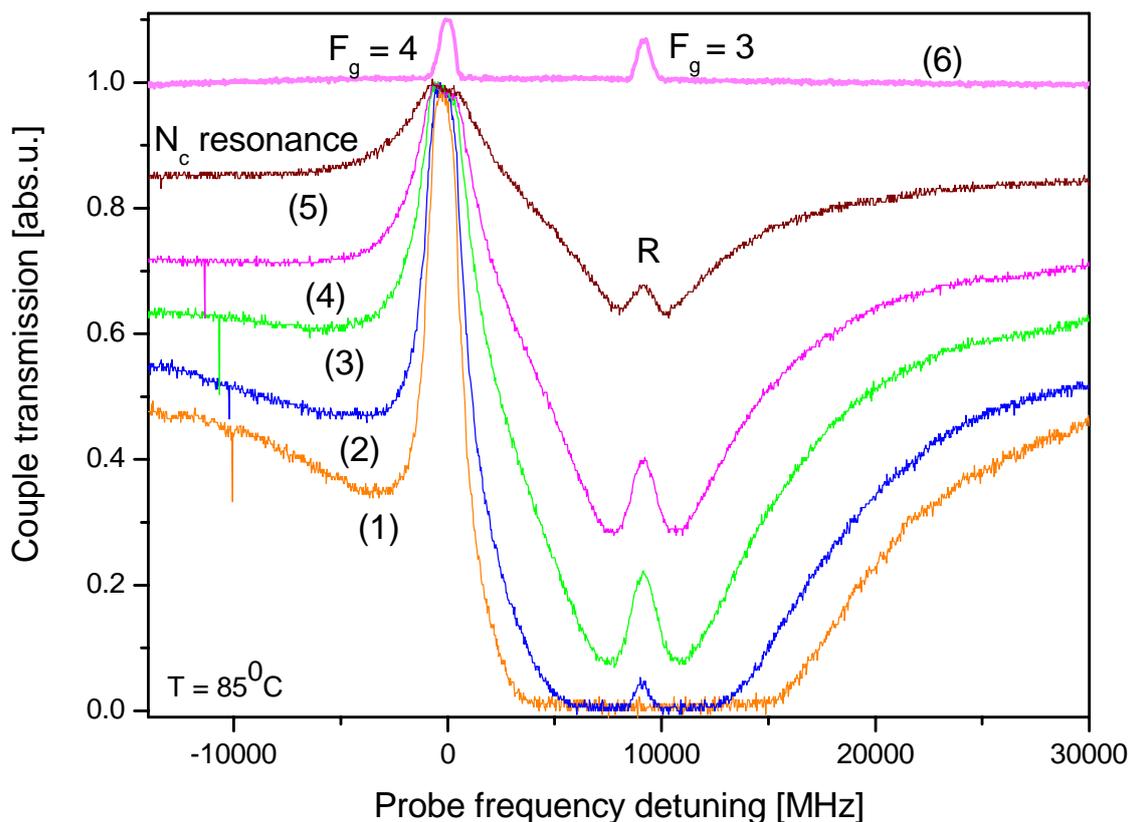


Fig. 7 Transmission spectrum of the coupling beam when the probe is scanned over a spectral interval of more than 40 GHz. The amplitude of the new R resonance depends on the coupling frequency ν_c detuning: (1) -869 MHz, (2) -1026 MHz, (3) -1484 MHz, (4) -2162 MHz, (5) -4214 MHz, (6) reference spectrum. The density of cesium atom is approximately 5.6×10^{12} atoms/cm³.

maximum of the $F_g = 4$ group, but at the same time the slope of the coupling transmission background around the N_c resonance increases.

The position of the coupling frequency influences also the complete transparency resonance shape and amplitude. This resonance is observed when the probe beam frequency ν_p is tuned to the $F_g = 4$ set. In the case of ν_c detuning - 4214 MHz (i.e. large detuning of the coupling beam), the absorption of the couple beam is the lowest (hence the transmission is the highest, Fig.7, curve 5) and the depletion of the $F_g = 4$ level by the probe light results in low amplitude of the complete transparency resonance. The opposite is the case when the ν_c detuning is - 869 MHz, where the absorption rate of the coupling beam is much higher. This is the reason for the observation of the highest absorption background around the N_c resonance (Fig.7, curve 1). However, when the probe beam is in resonance with the $F_g = 4$ group, this ground level is strongly depleted and the coupling transmission approaches 100%. Thus, the highest amplitude, complete transparency resonance is observed.

In addition, a rather fast reduction of the coupling absorption around the $F_g = 3$ transitions occurs with the coupling frequency detuning. Let us first consider a small detuning of the coupling frequency (- 869 MHz). Here, complete absorption of the coupling beam is measured over a very large region, for probe frequency detuning around the $F_g = 3$ set of transitions. Under condition of high Cs atom density, the $F_g = 3$ level is effectively populated by the coupling beam, due to the hyperfine optical pumping. However, a strong repopulation of $F_g = 4$ level by the probe beam also takes place. In this way, Cs atoms are cycled between both ground hyperfine levels.

The situation is different for large detuning (- 4214 MHz) of the coupling frequency from the $F_g = 4$ group. In this case, the optical pumping of the $F_g = 3$ level by the coupling beam is much less effective, and hence $F_g = 4$ level repopulation by the probe light is lower. This results in significantly lower absorption of couple beam.

The new R resonance is also influenced by the coupling detuning. At small detuning (Fig.7, curve 1), the R resonance is completely washed out, probably due to complete absorption of the coupling beam over a large interval of probe detuning. Then as the detuning increases, the R resonance appears and significantly enhances its amplitude (Fig.7, curves 3, 4). At even greater detuning, the R resonance amplitude decreases (Fig.7, curve 5).

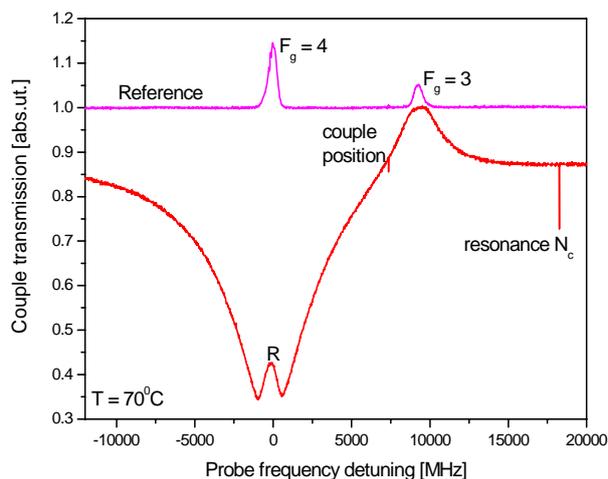


Fig. 8 Transmission of the coupling beam, for $\nu_c = 7360$ MHz. Here, the R resonance is formed at the $F_g = 4$ group. The sub-natural-width N_c resonance is situated at frequency higher than that of the $F_g = 3$ group, well outside the absorption profile and on a flat background. The density of cesium atom is $\sim 1.9 \cdot 10^{12}$ atoms/cm³.

It is worth stressing that the R resonance appears at the position of the $F_g = 3$ set of transitions and do not follow the shift of the coupling frequency that is performed in a frequency interval larger than 3 GHz.

Analyzing the R resonance behavior, it can be assumed that it occurs when the cycling of Cs atom population between the $F_g = 3$ and $F_g = 4$ levels is most intense. Such cycling process can also provide very high population of the excited $F_e = 2, 3, 4, 5$ levels. In fact, under such conditions, a negligible amount of Cs atoms can stay optically pumped and accumulate in each of the two ground state levels.

A similar experimental study was performed for the observation of the N_c resonance in the high frequency wing of transmission of the $F_g = 3$ group of transitions (Fig. 8). For this transition set, the sub-natural-width resonance N_c also appears with very good contrast and

superimposed on a flat background. The N_c resonance behavior with the atomic source temperature and coupling detuning is similar to that of the resonance observed in the lower frequency wing of the $F_g = 4$ group of transitions (see Fig.6 and Fig.7). The conditions for the formation the R resonance are also similar. Here it is observed when the frequency of the probe beam is tuned to the $F_g = 4$ set of transitions. Thus in this case also, the R resonance occurs when intensive cycling of Cs atoms between both hyperfine ground levels is provided due to the most efficient repumping of the $F_g = 3$ level, which takes place when the probe beam is in resonance with the $F_g = 4$ group of hyperfine transitions.

Special care should be taken to distinguish the R resonance from the EIT resonance which is based on a three-level Λ -system involving both hyperfine ground levels and an excited level. Hence the R resonance could be supposed to be an EIT one that can be strongly broadened under the conditions of our experiment.

In Fig. 9, the case of observation of all three (N_c , EIT, R) resonances is presented. Starting from low probe frequency, first the N_c resonance is observed at $\nu_p = \nu_c - \Delta$. After that, a very weak sub-

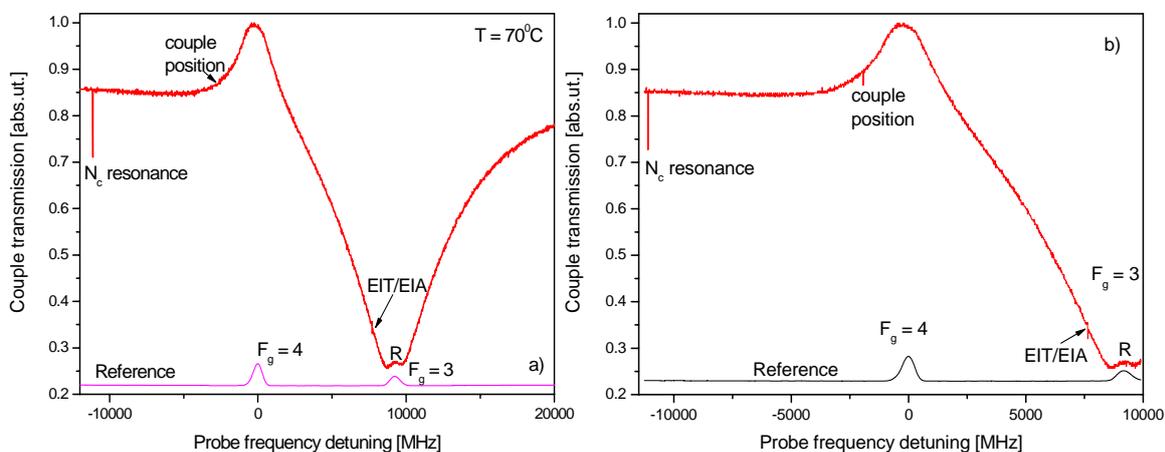


Fig.9. (a) Simultaneous observation of N_c , R resonance and EIT/EIA resonance in the couple beam transmission. (b) Zoom of (a) for a narrower interval of probe frequency detuning.

natural-width resonance can be seen at $\nu_p = \nu_c + \Delta$. Note that here, the EIT resonance is not characterized by the usual transparency window but instead has a dispersive shape. Such a dispersive shape can be observed due to the added buffer gas and the detuning of the probe and coupling frequencies from the center of the respective optical transitions [7]. From Fig. 9, it can be seen that the R resonance is situated at different values of ν_p from the EIT/EIA resonance. More specifically, for the EIT/EIA resonance formation the Raman condition should be satisfied, while the R resonance can be observed for different ν_c detunings provided the probe frequency ν_p is fixed at the maximum absorption of the $F_g = 3$ group of transitions. One can assume that under this condition, the $F_g = 4$ level repumping is the most effective.

Based on the experimental study of the behavior of the R resonance, it can be suggested that the physical process behind it could be related to optical transitions from the $F_e = 2, 3, 4, 5$ levels to higher Cs energy levels. Similar kinds of transitions have been realized in alkali atoms by the energy-pooling collision effect [8] which can be briefly summarized as follows. If hot Cs vapor is resonantly excited by high-intensity mono-mode laser light to the 6P levels, fluorescence from the 6D atomic levels will be observed. Note that the 6D atomic levels lie at nearly twice the frequency of the 6P levels. In Ref. [9], the population in the high-lying Cs levels was attributed to excited atom-excited atom collisions in which the two atoms pool their internal energy to produce one ground-state atom and one in a more highly excited state. Collisions of this type were mostly studied in Na, while there

has been little work on Cs. However, the energy-pooling collision study in cesium is of current interest because such collisions can be an important loss mechanism in ultracold laser traps, in which the atomic density is very high (10^{11} to 10^{13} cm⁻³).

The fluorescence produced by the energy-pooling collisions was systematically studied in Ref. [10]. There, by pumping the Cs D_2 line in a heated Cs atomic vapor cell, fluorescence containing about 30 spectral lines has been observed. The spectral lines were assigned to the transition lines of the cesium atom. In addition, the relative populations of the levels involved in the energy-pooling collision process have been determined. For instance, it has been shown that for Cs atomic density of 1.8×10^{14} /cm³, the relative populations of the $6D_{3/2}$ level compared to $6P_{3/2}$ is more than 1%. In our experiment the atomic density is less, but the light excitation is bi-chromatic so that it can provide more atomic population in the $6P_{3/2}$ levels. Thus, the R resonance can be considered as the beginning of population of energy levels higher than the $6P_{3/2}$ level. Further investigations will be needed in order to clarify if the bi-chromatic excitation of alkali hot atoms can result in more effective energy-pooling process.

5. Conclusion

Systematic experimental study of sub-natural-width resonance observed on the D_2 line of Cs vapor is presented. Three-photon, bi-chromatic excitation of Cs atomic vapor buffered by 20 Torr of neon is performed. The bi-chromatic excitation involves a fixed-frequency coupling beam and scanning the probe beam across the D_2 line. The transmission of the coupling beam is measured as a function of the probe frequency detuning. Such an approach allows the observation of very good contrast, sub-natural-width (less than 2 MHz) N_c resonance on a low signal flat background, which can be advantageous for many applications.

The probe light frequency is scanned in a large spectral interval (more than 30 GHz) that allows analyzing complex processes in the light-atom interaction such as velocity-changing collisions, hyperfine optical pumping and re-pumping, and their influence on the N_c resonance and the transmission of the constant frequency coupling laser.

The comparison between the N_c and EIT resonances shows that the N_c resonance contrast is much higher than that of the EIT resonance under the conditions of our experiment. A new spectral feature in the couple transmission, denoted as R resonance is observed and studied. Further theoretical analysis is needed to clarify the origin of the new resonance.

Acknowledgements: The authors greatly appreciate the fruitful discussions of experimental results with S. Gozzini, G. Grigoryan and Y. Malakyan. This work was supported by a Marie Curie International Research Staff Exchange Scheme Fellowship within the 7th European Community Framework Programme.

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