Journal de physique et de chimie des étudiants

Volume 1. No. 2. - Décembre 2016
Pages 11 - 16

SATURATED ABSORPTION SPECTROSCOPY

DAVID DUMONT

Saturated absorption spectroscopy improves the resolution of the atomic transmission spectra by giving access to the hyperfine transitions and, contrary to classical spectroscopy, it is not limited to the fine structure. Furthermore, it overcomes the limit of Doppler-broadening of the transitions due to the kinetic motion of atoms. The method exploits two counter-propagating beams, a strong one to saturate the atoms of a vapour cell in order to reduce the absorption of a weak beam. This scheme is widely implemented to stabilise precisely the lasers frequency.

1. INTRODUCTION

Measuring atomic energy spectra is crucial for many fields in chemistry and atom physics to determine precisely the atomic structure but also in physics to stabilise the frequency of lasers. However, the resolution of the spectra measured using this classical method is limited by several phenomena, and mainly by Doppler-broadening due to the frequency shift for moving atoms. The atoms in a gas move permanently and their velocity follows a Gaussian distribution, which implies a broadening of the spectral lines. This broadening is a real limitation for laser locking: the laser frequency can be set to a specific transition (reference transition) by means of various methods [1], [2]. The precision of this laser locking, i.e. the variation range of the laser frequency called *laser linewidth*, is given by the linewidth of the reference transition. Consequently, the linewidth of a laser locked to a Doppler broadened spectral line will be larger.

The saturated absorption spectroscopy improves this resolution by revealing not only the fine structure of the atoms as for simple spectroscopy but their whole hyperfine structure. Then, the main limiting parameter is the power broadening. A good estimation of the resolution is provided by the ratio of the linewidth of the spectral line ($\Delta\omega$) divided by its frequency (ω_0). In numerical terms, for the D2 transition [1] of Cesium, a classical spectroscopy measurement has a ratio $\frac{\Delta\omega_D}{\omega_0} \approx 10^{-6}$ while the saturated absorption spectroscopy has a ratio $\frac{\Delta\omega_{hole}}{\omega_0} \approx 10^{-8}$: the spectra resolution is improved by a factor 100. Thus, the laser linewidth is reduced when the laser is locked to that sub-Doppler spectrum.

The following article will explain what the Doppler broadening is before developing the principle of the saturated absorption spectroscopy. Finally, we will quickly compare the theoretical behaviour with our experimental spectra.

2. DOPPLER BROADENING

When an atom is moving, it does not see the same radiation than when it is fixed. In fact, as shown on Figure 1, a radiation with an angular frequency ω in the laboratory frame is seen, by an atom moving with a velocity v, like a radiation with an angular frequency given by:

$$\omega' = \omega \pm kv, \tag{1}$$

where k is the wave vector amplitude of the radiation. The sign ± depends on the propagation direction of the radiation compared to the direction of the atom movement. Let's assume that the radiation and the atom are moving in the same direction, i.e $\omega' = \omega - kv$. Besides we suppose that $\boldsymbol{k}$ and $\boldsymbol{v}$ are collinear, i.e $\boldsymbol{k}.\boldsymbol{v} = kv$, so in the following paper we will remove the vector sign.

We suppose that all the atoms absorb the radiation with an angular frequency ω_0 in their rest frame: that means we only consider a two level system with only one available

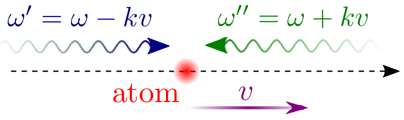

Fig. 1 Doppler effect for a moving atom. In it rest frame, radiation is shifted by $\pm kv$ among the direction of the atom movement.

---

[1] The electronic configuration for Cesium is $[Xe]6s^1$. In spectroscopy, the transition $6s \rightarrow 6p$ is widely studied and it is usually called *D2 transition*. It corresponds to an infrared radiation at 352 THz or 852 nm.

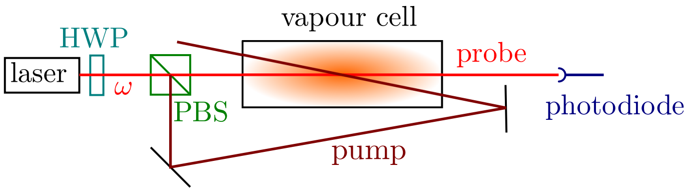

Fig. 2 General setup of the saturated absorption spectroscopy. The laser beam is split by the polarising beam splitter (PBS) into a probe and a pump beams. The pump/probe intensity ratio is adjusted by a half-wave plate (HWP). The probe beam light is recorded by a photodiode after crossing the vapour cell. The probe and the counter-propagating pump beams intersect or better overlap in the vapour cell. The laser frequency ω sweeps over a few hundred of MHz around the D2 transition to measure the transmission spectrum.

transition. Thus, a radiation is absorbed when $\omega' = \omega_0$ so when the detuning to the transition $\delta_\omega = \omega - \omega_0 = kv$. Therefore $k = \frac{\omega_0}{c}$ so an atom absorbs the radiation only if it has the following velocity:

$$v_{abs} = \frac{\delta_\omega c}{\omega_0}. \tag{2}$$

Here, we are working with a Cesium gas cell (called *vapour cell*) at room temperature, and all the atoms are moving. The velocity distribution in the cell is given by:

$$\begin{aligned} f(v)dv &= \sqrt{\frac{M}{2\pi k_B T}} \exp\left(-\frac{Mv^2}{2k_B T}\right) dv \\ &= \frac{1}{\sqrt{\pi} u} \exp\left(-\frac{v^2}{u^2}\right) dv, \end{aligned} \tag{3}$$

where $u = \sqrt{\frac{2k_B T}{M}}$ is the most probable velocity of the atoms, T the temperature and M the atomic mass. At a temperature of 300 K, the Cesium atoms are moving with a typical velocity of 200 m/s. The absorption function $g(\delta_\omega)$ is defined by:

$$g(\delta_\omega) = \int_{\mathcal{R}} N(v)\sigma(v, \delta_\omega) dv, \tag{4}$$

with $N(v)$ the number of atoms per unit of volume in the velocity class v to $v + dv$ and $\sigma(v, \delta_\omega)$ the absorption cross-section. As only atoms moving with the velocity v_{abs} absorb light, we have $\sigma(v, \delta_\omega) = \frac{c}{N\omega_0}\delta(v - v_{abs})$. Besides, considering a two-level system, $N(v) = Nf(v)$ where N is the total number of atoms in the ground state per unit volume. So the absorption function is:

$$\begin{aligned} g(\delta_\omega) &= Nf(v_{abs})\frac{c}{N\omega_0} \\ &= \frac{c}{\sqrt{\pi}\omega_0 u} \exp\left(-\left(\frac{c}{u}\frac{\delta_\omega}{\omega_0}\right)^2\right). \end{aligned} \tag{5}$$

It is also a gaussian shaped function. The maximum value is achieved for $\delta_\omega = 0$ and $g(\delta_{\omega_{1/2}}) = g(0)/2$ for $\delta_{\omega_{1/2}} = \frac{u\omega_0}{c}\sqrt{\ln 2}$. The Doppler-broadened line has a linewidth $\Delta\omega_D = 2\delta_{\omega_{1/2}}$ equal to:

$$\frac{\Delta\omega_D}{\omega_0} = 2\sqrt{\ln 2}\frac{u}{c} \approx 10^{-6} \text{ for Cesium.} \tag{6}$$

Thus, for the D2 transition at 352 THz usually used to stabilise the laser frequency, the Doppler broadening implies a linewidth of a few hundred of MHz. Nonetheless, a proper frequency stabilisation scheme requires a linewidth of a few hundred of kHz. For a higher resolution of a spectrum, we need to implement a sub-Doppler method. The saturated absorption spectroscopy is mainly implemented to frequency stabilise lasers.

3. SATURATED ABSORPTION SPECTROSCOPY

It is possible to overcome the Doppler broadening by implementing a sub-Doppler spectroscopy method. The saturated absorption spectroscopy is a general sub-Doppler method.

Consider a radiation with an angular frequency ω travelling across a vapour cell. The density distribution of the atoms in the cell with velocities ranged between v and $v + dv$ is $N(v) = Nf(v)$ where N is the total number of atoms in the cell and $f(v)$ is the velocity distribution defined previously by Equation (3).

3.1. Principle

The basic idea is to select only atoms for the spectroscopy signal, which are at rest and do not show any Doppler shift. The general setup is reported on Figure 2: two counter-propagating beams, one strong called *pump beam* and another one weak called *probe beam*, overlap or intersect crossing a vapour cell. In fact, the laser beam is split into two beams by a polarising beam splitter (PBS) which separates the horizontally and the vertically po-

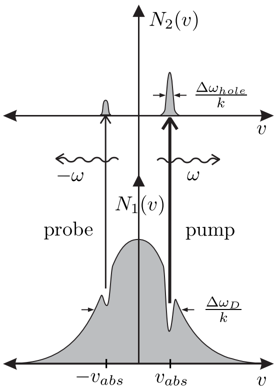

Fig. 3 Gaussian shaped velocity distribution of the ground and the excited states. The linewidth of the ground state distribution is $\frac{\Delta\omega_D}{k}$. The pump (thick arrow, on the right) and the probe (tiny arrow, on the left) excite the atoms from the lower to the upper level: this is called *hole burning*. The linewidth of the hole is $\frac{\Delta\omega_{hole}}{k}$ because of the power broadening. The pump excites more atoms than the probe because its intensity is higher. The probe and the pump are counter-propagating so they excite two opposite classes of atoms ($\pm v_{abs}$). The symmetrical scheme is also available. Drawing adapted from [3].

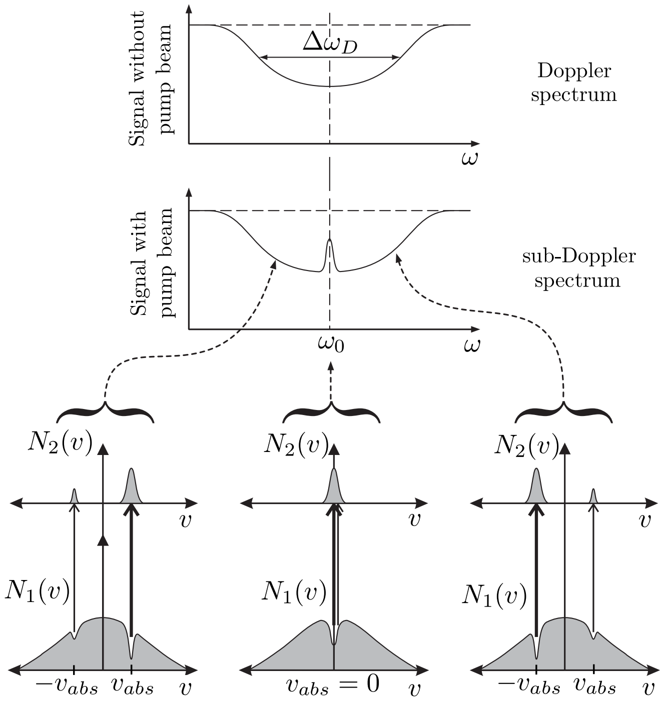

Fig. 4 Top: Probe intensity transmitted across the vapour cell with and without the pump. The sub-Doppler and the Doppler spectra are respectively observed. The linewidth of the Doppler broadened spectrum is $\Delta\omega_D$. Bottom: The corresponding population densities of the two levels for three different laser frequencies: below, equal to and above the atomic resonant frequency showing the probe and the pump effects. Far from the transition (on the sides), the probe and the pump interact with atoms from two different velocity classes $\pm v_{abs}$. Thus, the transmitted probe intensity is not affected by the pump and the simple Doppler profile is observed. On the transition, $v_{abs} = 0$ so the probe and the pump interact with the same atoms which are motionless. The pump excites a large part of these atoms: less atoms can be excited so the probe is less absorbed and a peak on the transmission spectrum is obtained. Drawing adapted from [3].

larised light. By adjusting an half-wave plate (HWP), we modify the pump/probe intensity ratio.

Let's consider a two-level system: atoms which are in the ground state can be excited into the upper state by a radiation with an angular frequency ω_0 in their rest frame. $N_1(v)$ and $N_2(v)$ are the number densities in levels 1 and 2 respectively, for atoms in the velocity class v to $v + dv$. The pump beam excites most of the atoms into the upper state.

Figure 3 shows typical distributions for the ground and the excited states of the gas when the vapour cell is crossed by the pump and the probe. One notes that whatever the beams intensities, the integration of $N_i(v)$ over the velocity is equal to the number of atoms on the level i:

$$\int_{-\infty}^{\infty} N_i(v)dv = N_i \text{ for } i = 1, 2. \tag{7}$$

The probe and the pump are counter-propagating so using Equation (2), we show they interact with two opposite classes of atoms moving with the velocity $\pm v_{abs}$. We distinguish two behaviours at low and high intensities. At low intensities, i.e for the probe beam, almost all the atoms are in the ground state so $N_1(v) \approx N(v)$ and $N_2(v) \approx 0$. Thus, $N_1(v) = Nf(v)$ is the Gaussian described by Equation (3) as shown on the left side of Figure 3.

For higher intensities, i.e for the pump beam, the atoms distribution is affected by the strong beam: most of the atoms in the ground state and in the appropriate velocity class are excited into the upper level as represented on the right side of Figure 3. That is called *hole burning*. The width of the hole burnt into the ground state distribution is

$$\Delta\omega_{hole} = \Gamma\left(1 + \frac{I}{I_{sat}}\right)^{1/2}, \tag{8}$$

where I is the beam intensity and I_{sat} the saturation intensity defined so that if $I = I_{sat}$ half of the atoms of the velocity class are excited. That corresponds to the power-broadened natural linewidth: the higher the beam intensity is, the larger the hole width is.

We measure the probe intensity after crossing the vapour cell to obtain the transmission spectrum. The laser frequency sweeps over a few hundred of MHz around 352 THz, i.e the D2 transition. Figure 4 reports the spectra with and without the pump with the corresponding atom distributions for the two levels. When the laser emits a radiation with an angular frequency far from the resonant frequency, $|\delta_\omega| = |\omega - \omega_0| >> \Delta\omega_{hole}$, the two beams interact with different classes of atoms (those moving at $\pm v_{abs}$) so the probe intensity is not affected by the pump. The Doppler broadened spectrum is obtained as we can see on the left and right sides of the spectrum of Figure 4.

Close to resonance, $\omega \approx \omega_0$ ($\delta_\omega = 0$), the two beams interact with atoms with a velocity $v_{abs} = \pm\frac{\delta_\omega c}{\omega_0} \approx 0$. The pump beam excites most of these atoms into the upper level. So there are less atoms which can be excited and the probe beam absorption is reduced. Consequently, a peak appears on the transmission spectrum at the atomic resonance frequency ω_0.

3.2. Cross-over Peaks

We have only considered a two level system but in reality, atoms are multi-level systems which causes the apparition of new peaks, referred as *cross-overs*. We will show that starting from a three level system.

Now, we play with a ground state (1) and two excited states (2 and 3) with the corresponding atomic resonance frequencies ω_{12} and ω_{13} respectively. As previously, when the laser works with an angular frequency ω, a beam selects two classes of atoms: those which move with a velocity $v_{abs}^{12} = \pm\frac{\omega-\omega_{12}}{k}$ or with $v_{abs}^{13} = \pm\frac{\omega-\omega_{13}}{k}$ because we have two available transitions. The sign depends on the

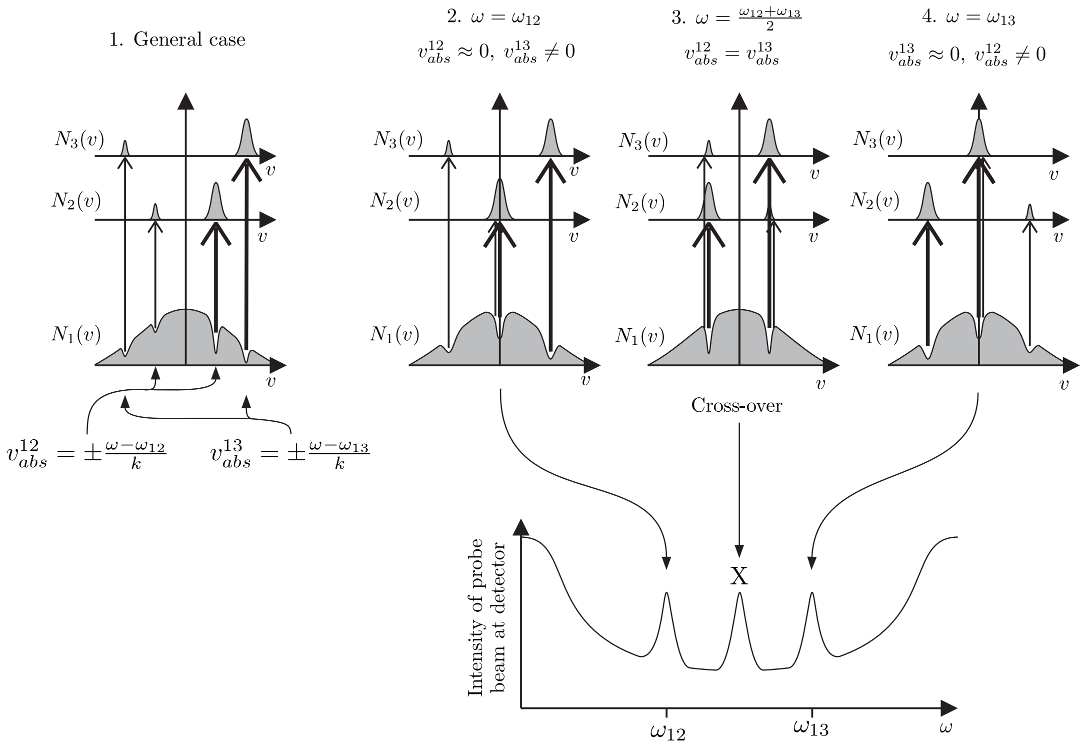

Fig. 5 Top: Population densities of the three levels for four situations: 1. the general situation: the probe and the pump excite two classes of atoms, i.e those which move at $\pm v_{abs}^{12}$ and $\pm v_{abs}^{13}$, 2. and 4. the radiation frequency corresponds to one transition: $\omega = \omega_{12}$ (respectively $\omega = \omega_{13}$): The pump and the probe excite three velocity classes of atoms $\pm v_{abs}^{13}$ and $\pm v_{abs}^{13} \approx 0$ (resp. $\pm v_{abs}^{12}$ and $\pm v_{abs}^{13} \approx 0$). So a peak at ω_{12} (resp. at ω_{13}) appears on the transmission spectrum as seen before. 3. the radiation frequency corresponds to the frequency midway: $\omega = \frac{\omega_{12}+\omega_{13}}{2}$ so $v_{abs}^{12} = v_{abs}^{13}$, i.e the pump and the probe interact with the same two velocity classes of atoms so a third peak is observed at the frequency midway $\frac{\omega_{12}+\omega_{13}}{2}$. This peak is called *cross-over*. Bottom: Corresponding transmission spectrum where the three peaks are visible. Drawing adapted from [3].

relationship between the propagation direction of the radiation and the direction of the atom movement.

Figure 5 shows the population densities of the three levels for four frequencies: far from the transitions, equal to the transitions ω_{12} and ω_{13} and equal to the frequency midway $\frac{\omega_{12}+\omega_{13}}{2}$. The corresponding transmission spectrum is also reported. Far from the transitions ω_{12} or ω_{13}, i.e on the sides of the spectrum represented on Figure 5, the two beams interact with four different classes of atoms (those moving at $\pm v_{abs}^{12}$ or $\pm v_{abs}^{13}$), so the simple Doppler broadened profile is observed.

Close to one transition, for instance for $\omega \approx \omega_{12}$, the two beams interact with only three classes of atoms. The pump excites most of the atoms with a velocity $v_{abs}^{12} \approx 0$ (and with $v_{abs}^{13} \approx \frac{\omega_{12}-\omega_{13}}{k}$) which reduces the absorption of the probe, so a peak at the atomic resonance transition, here at ω_{12}, appears. The same reasoning explains the peak at ω_{13}. However, when $\omega = \frac{\omega_{12}+\omega_{13}}{2}$, we show that $v_{abs}^{12} = v_{abs}^{13} = \frac{\omega_{12}-\omega_{13}}{2k} = v_{abs}^{co}$ so the probe and the pump beams select two classes of atoms: those which are moving with a velocity $v = \pm v_{abs}^{co}$. Thus, as usually the pump reduces the absorption of the probe which causes a peak at frequencies midway between the two transitions.

The height of the peaks depends directly of the number of atoms excited by the pump: the more excited atoms we have, the less the probe is absorbed by these atoms, so the more the probe is transmitted. The peaks at the atomic resonance transitions (ω_{12} and ω_{13}) are created by only one class of atoms (those which are fixed, $v_{abs}^{12} \approx 0$ or $v_{abs}^{13} \approx 0$). The cross-overs are produced by two classes of atoms (those which move with $\pm v_{abs}^{co}$), that is by twice more atoms than the transition peaks. Consequently, we expect to observe cross-overs twice higher than the transition peaks.

4. IMPLEMENTATION

In this section, we want to show some experimental spectra measured for Cesium following the saturated absorption spectroscopy method. We are using a laser at a

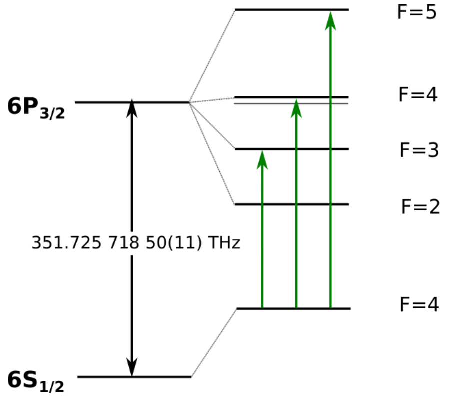

Fig. 6 Fine and hyperfine structure for the D2 transition for Cesium. In green, the achievable transitions for our laser. Data taken from [4].

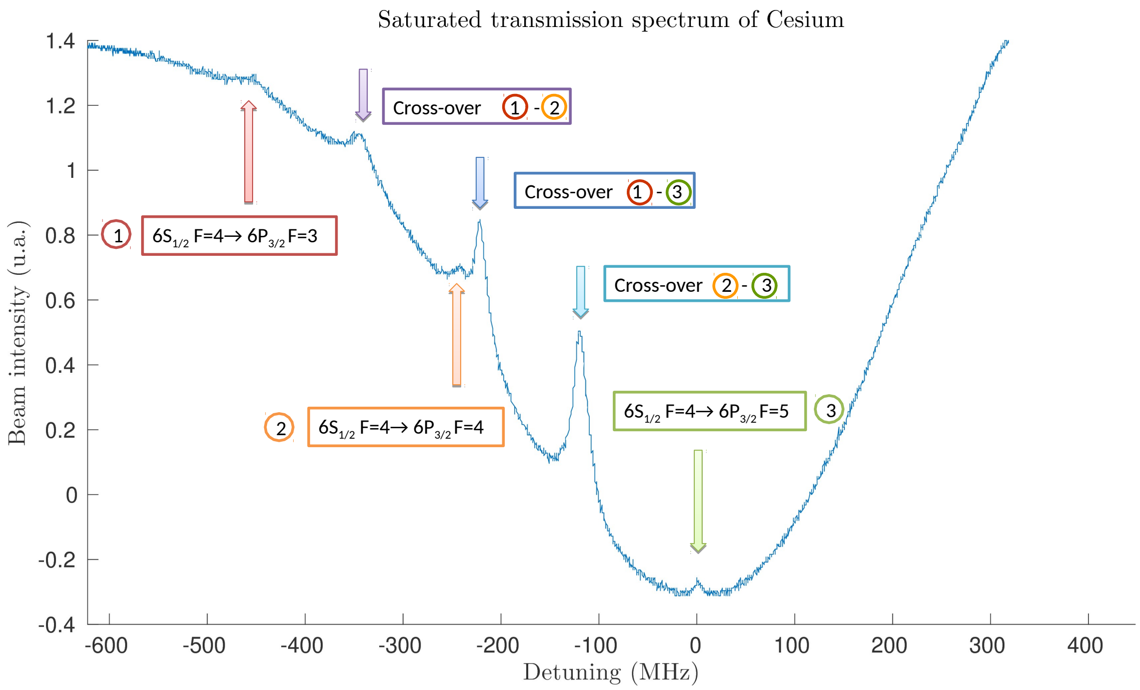

Fig. 8 Experimental sub-Doppler transmission spectrum of Cesium. Six peaks are observed and correspond to the three available transitions plus the three cross-overs. The x-axis calibration is calculated by using the known frequencies of peaks 1 and 3. Then, we have checked the positions of the other peaks.

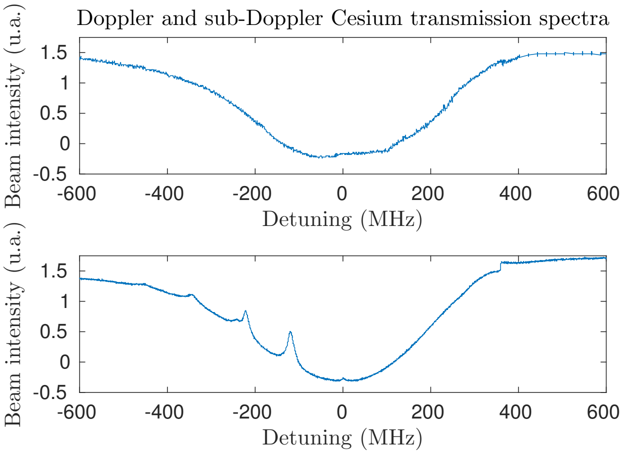

Fig. 7 Top: Doppler transmission spectrum of Cesium. The gap is due to the D2 transition which is Doppler broadened. Bottom: Sub-Doppler transmission spectrum. The main gap is due to the Doppler broadening of the D2 fine transition and the small peaks represent the hyperfine D2 transition. That illustrates the resolution improvement.

frequency of 352 THz, i.e an infrared laser at a wavelength of 852 nm, which allows us to achieve three atomic transitions from the hyperfine structure of the D2 transition as shown on Figure 6.

The setup of Figure 2 is implemented and our laser laser frequency sweeps over a few hundred of MHz around 352 THz which corresponds to the D2 transition. Thus, we measure the transmission spectrum of the Cesium which composes the vapour cell. Figure 7 compares a simple Doppler broadened transmission spectrum (on the top) to the experimental saturated transmission spectrum (on the bottom). The Doppler broadened spectrum is measured with the same setup but without the pump: by the way only one beam, i.e the probe, crosses the vapour cell.

The improvement of the spectrum resolution brought by the saturated absorption spectroscopy is clearly visible: this method overcomes the Doppler broadening and detects the hyperfine transitions which give the peaks of the sub-Doppler spectrum. As expected, the sub-Doppler spectrum respects the general Doppler profile excepted on the hyperfine transitions where new peaks are observed. The Doppler broadening ratio $\frac{\Delta\omega_D}{\omega_0}$ is estimated at $1.4\ 10^{-6}$ which is consistent with the theoretical ratio of 10^{-6} expected. The 4% error on the x-axis calibration is too reduced to explain completely the difference. We have seen that $\frac{\Delta\omega_D}{\omega_0} = 2\sqrt{\ln 2}\frac{u}{c}$ so the previous difference could imply that the most probable velocity of the atoms u would be higher than predicted, that is the temperature in the cell would be higher than in the laboratory. This phenomenon could be due to the lasers who would heat the atoms. However, using the relationship $u = \sqrt{\frac{2k_BT}{M}}$, we show that the temperature in the cell would be equal to 736 K which is absolutely not realistic.

The experimental saturated transmission spectrum (the sub-Doppler spectrum) is also reported on Figure 8. The x-axis scale is deduced by assuming known the frequency values of peaks 1 and 3. Then, the positions of the other peaks have been checked comparing to the expected theoretical frequencies: as a result, we estimate the calibration error at $\pm 4\%$. Six peaks are observed: three of them correspond to the three atomic resonance transitions and the three others are the cross-overs. Indeed, with three transitions, it is possible to create three pairs of transitions (1+2, 1+3 and 2+3), which explains the number of cross-overs.

These cross-overs are higher than the transitions peaks themselves as the theory showed. However, we do not find factor 2 predicted between the cross-overs height and the other peaks height; the cross-overs are widely larger and the peaks corresponding to the hyperfine transitions are very tiny. In fact, we noted that the cross-overs height depends widely on the ratio probe/pump intensity. If the probe is too intense, the cross-over 1-2 recovers peak 2. This is consistent with the power broadening exposed previously: the linewidth $\Delta\omega_{hole}$ of a power broadened spectral line increases with the beam intensity.

5. Conclusion

The resolution of a simple transmission spectrum is mainly limited by the Doppler broadening. The saturated absorption spectroscopy exploits two counter-propagating beams to saturate the absorption of one weak beam. The hyperfine structure is achievable but new peaks are created at frequencies midway between pairs of close transitions.

This spectroscopy scheme is widely used for laser frequency stabilisation. Many setups have been developed to derive a signal from the saturated transmission spectrum at the atomic hyperfine transitions to lock precisely the lasers to this transition. The polarisation spectroscopy deals with the probe and the pump polarisations [5, 1] while the modulation transfer spectroscopy modulates the light frequency [2].

6. Acknowledgement

This work was achieved during an internship at the University of Strathclyde in Glasgow, Scotland. I would like to thank my supervisor Dr. Elmar Haller for correcting this paper and Camille Eloy for his enlightening advices.

References

[1] L. Qiang et al. “Polarization spectroscopy in cesium”. In: *Laser Physics and Laser Technologies (RCSLPLT) and 2010 Academic Symposium on Optoelectronics Technology (ASOT), 2010 10th Russian-Chinese Symposium on.* July 2010, pp. 200–203.

[2] D. J. McCarron, S. A. King, and S. L. Cornish. “Modulation transfer spectroscopy in atomic rubidium”. In: *Measurement Science and Technology* 19.10 (2008), p. 105601.

[3] C. J. Foot. *Atomic physics.* Oxford master series in physics. Oxford University Press, 2005, pp. 151–162. ISBN: 9780198506966.

[4] D. A. Steck. *Cesium D Line Data.* `http://steck.us/alkalidata/cesiumnumbers.1.6.pdf`. 2016.

[5] C. P. Pearman et al. “Polarization spectroscopy of a closed atomic transition: applications to laser frequency locking”. In: *Journal of Physics B: Atomic, Molecular and Optical Physics* 35.24 (2002), p. 5141.

☐ David Dumont
M2 Sciences de la Matière
Parcours FEADéP Physique
ENS de Lyon - Université Lyon 1
david.dumont@ens-lyon.fr