

Atomic Spectroscopy Laboratory

Master's degree in Quantum Engineering

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1 Absorption spectroscopy on an atomic vapor

One of the easiest arrangements to probe an optical atomic transition, it to shine a collimated beam through the atomic sample and measure the transmitted intensity **(I)** with a photodetector. This is shown in Fig. 1. If the laser frequency is known, by tuning the laser frequency, we can scan the spectral region when the atoms absorb and thus determine the atomic resonance frequency. Alternatively, when the atomic transition is already well-known, we can use the atoms as an absolute frequency reference, and stabilize the laser to the atomic transition. The latter case is a typical experimental tool that is used in many Atomic and Molecular Physics experiments, such as atomic clocks.

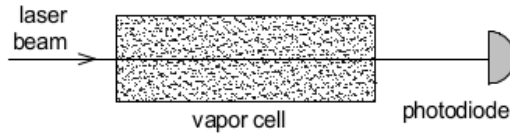


Figure 1: Basic arrangement for ordinary laser absorption spectroscopy.

1.1 Atomic absorption

An atomic gas or vapor will absorb laser light close to a resonant transition linking its ground state to an excited state. For a two-level atom model (see Fig. 2) with a ground state $|g\rangle$ and an excited state labeled $|e\rangle$ (which is a good description close to resonance) we can write the attenuation of the laser beam intensity I along the laser propagation axis z as [1]:

$$\frac{dI}{dz} = -\kappa(\omega_L)I = -n\sigma(\omega_L)I \quad (1)$$

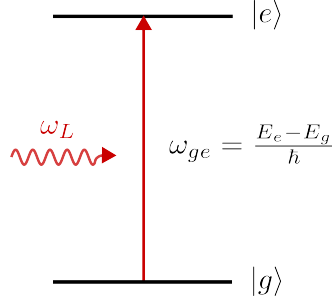


Figure 2: Energy scheme of a 2 level atom. The atoms will absorb when the photon energy $\hbar\omega_L$ matches the energy difference between the two states.

In Eq.1 we have defined an absorption coefficient κ , that is a function of the laser angular frequency ω_L , the atomic density n (number of atoms per unit of volume) and the atomic cross-section $\sigma(\omega_L)$. $n\sigma\Delta z$ can be interpreted as the fraction of the target area that is covered by the atoms, and thus the probability that an atom will absorb a photon when crossing the volume $V = A\Delta z$.

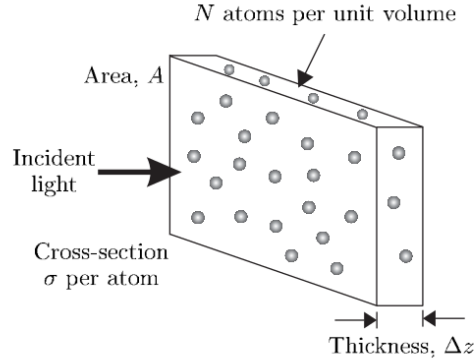


Figure 3: Pictorial illustration of the atomic cross-section. Taken from [1].

If we integrate along z , we obtain the Lambert-Beer extinction law:

$$I(\omega_L, z) = I(\omega_L, 0)e^{-\kappa(\omega_L)z} \quad (2)$$

that gives an exponential decay of the input intensity as the laser propagates into the atomic medium. The atomic absorption coefficient is related to the atomic properties and in absence of broadening mechanisms is given by:

$$\kappa(\omega_L) = \frac{g_2}{g_1} \frac{\pi\omega_{ge}d_{ge}^2 n}{\epsilon_0 \hbar c} g(\omega_L, \omega_{ge}) \quad (3)$$

where ω_{ge} is the atomic transition angular frequency, d_{ge} the transition dipole moment, g_1 and g_2 the degeneracy of the ground and excited state respectively, and

$$g(\omega_L, \omega_{ge}) = \frac{1}{2\pi} \frac{\Gamma}{(\omega_L - \omega_{ge})^2 + (\Gamma/4)^2} \quad (4)$$

is a lineshape factor, coming from the finite lifetime of the excited state (Γ is the decay rate of level $|e\rangle$). The latter expressions are valid for very weak input laser intensities, since we have assumed that almost all the atoms remain in the ground state ($n_g - n_e \simeq n$). At higher intensities, saturation effects must be accounted for, and the saturation effects must be taken into account [1].

For a cell with length L , the light intensity measured at the exit of the cell, as a function of the laser frequency is:

$$I(\omega_L, L) = I(\omega_L, 0)e^{-\kappa(\omega_L)L} \quad (5)$$

and the absorption coefficient κ can be calculated by knowing the cell length L and by measuring the relative absorption close to the atomic transitions:

$$\kappa(\omega_L) = -\frac{\ln\left(\frac{I(\omega_L, L)}{I(\omega_L, 0)}\right)}{L} \quad (6)$$

1.2 Doppler broadening

When we consider an hot atomic vapor contained inside a glass cell, each atom will have a different velocity, and in the reference frame when it is at rest, it will see a laser frequency:

$$\omega'_L = \omega_L - \vec{k} \cdot \vec{v} \quad (7)$$

where \vec{k} is the laser wavevector with magnitude $k = \omega_L/c$ and \vec{v} the atomic velocity. Since each absorber has a different velocity, each atom will be resonant and absorb at a different frequency. This means that to obtain the absorption profile Eq.(3) we must average over the whole velocity distribution. This leads to a broadening of the line. This is a typical example of “inhomogeneous” broadening, where each absorber exhibits a different shift and the collective effect is a broadening of the transition.

For atoms at thermal equilibrium, the velocity distribution is described by the Boltzmann distribution:

$$f(v)dv = \sqrt{\frac{M}{2\pi k_B T}} \exp\left(-\frac{Mv^2}{2k_B T}\right) dv \quad (8)$$

Where M is the atomic mass and T is the cell temperature in Kelvin. Here we can define $\sigma_v = \sqrt{k_B T/M}$, that is the standard deviation of the Gaussian distribution, and is linked to the most probable atomic velocity ($u = \sqrt{2}\sigma_v$).

When integrating over all the velocities, in the limit $ku \gg \Gamma$, Eq.(3) becomes:

$$\kappa(\omega_L) = \frac{g_2}{g_1} \frac{\pi \omega_L d_{ge}^2 n}{\epsilon_0 \hbar c} g_D(\omega_L, \omega_{ge}) \quad (9)$$

with the resonance profile given by a Gaussian function:

$$\begin{aligned} g_D(\omega_L, \omega_{ge}) &= \sqrt{\frac{Mc^2}{2\pi k_B T \omega_{ge}^2}} \exp\left(-\frac{Mc^2(\omega_L - \omega_{ge})^2}{2k_B T \omega_{ge}^2}\right) \\ &= \frac{1}{\sqrt{2\pi}\sigma_D} \exp\left(-\frac{(\omega_L - \omega_{ge})^2}{2\sigma_D^2}\right) \end{aligned} \quad (10)$$

where we have defined $\sigma_D = \sqrt{k_B T / Mc^2 \omega_{ge}}$.

1.3 Saturated absorption spectroscopy

Saturated absorption can be exploited to obtain a resonance signal that is Doppler-free. If we introduce a second laser beam with higher intensity, counterpropagating with respect to the probe beam, we will obtain a non-negligible transfer of atomic population into the excited state. If we rewrite the absorption coefficient including the population of the excited beam, we obtain:

$$\kappa(\omega_L) = \int_{-\infty}^{\infty} (n_e(v) - n_g(v)) \sigma(\omega_L - kv) dv \quad (11)$$

where v is the atoms velocity projection along the propagation axis of the probe and pump beam. The effect of the pump beam is to create a population imbalance, for the resonant atoms, that possess a velocity $v = (\omega_L - \omega_{ge})/k$ inside the inhomogeneously broadened profile. This is often referred to as “hole burning”. The width of the hole is close to the natural linewidth of the transition, that is only broadened by saturation effects ($\Delta_{hole} = \Gamma\sqrt{1+s}$, where $s = I/I_s$ is the saturation parameter and is the ratio between the pump laser intensity and the saturation intensity I_s which depends on the particular transition). For the Cs D₁ line $I_s = 2.5 \text{ mW cm}^{-2}$. If the laser is off-resonance with respect to the atomic transition, the pump and probe beam will interact with different velocity classes (with $v = \pm(\omega_L - \omega_{ge})/k$). No effect will be visible by observing the transmission of the probe beam. Instead, for $\omega_L \simeq \omega_{ge}$, atoms with $v \simeq 0$, will interact both with the pump and the probe beams. The net effect is that in the Doppler broadened profile, a dip in the absorption is dug, centered around the atomic resonance. This small dip can be used as a Doppler-free spectroscopic tool to determine more precisely the atomic frequency, or to lock the laser to a narrower feature. Finally, it has to be noted that if the pump and probe beam are not exactly collinear, but slightly tilted, the Doppler cancellation is not perfect and the width of the saturation dip can be broadened by a residual Doppler effect.

1.4 Vapor density

The density of an atomic vapor in thermodynamical equilibrium with its liquid or solid reservoir, can be estimated from the vapor pressure, using the ideal gas

law. For Cs atoms the vapor pressure was measured accurately for example in [2], giving:

$$\log_{10} P_S = 12.6709 - 1.00 \log_{10} T - 4150/T \quad \text{for } T < 302K \quad (12)$$

$$\log_{10} P_L = 13.178 - 1.35 \log_{10} T - 4041/T \quad \text{for } T > 302K \quad (13)$$

Were S stands for “solid”, and must be used when the reservoir is solid (below the melting point), whereas L stands for “liquid”. Once the vapor pressure is calculated (or taken from tabulated data), it can be converted into numerical density by the ideal gas law:

$$n = \frac{P_{S,L}}{k_B T} \quad (14)$$

1.5 ^{133}Cs properties

^{133}Cs is the only stable isotope of Cs, thus it has a natural abundance close to 100%. Moreover, it presents a low melting point and high vapor pressure, easing the operation of a vapor cell near room temperature. The level scheme of interest for the laboratory experience is reported in Fig. 4. They refer to the D_1 line, linking the $|L = 0, J = 1/2\rangle$ ground state to the $|L = 1, J = 1/2\rangle$ excited state. This transition is used in many quantum-sensors, such as optically-pumped magnetometers, or in compact and chipscale Cs clocks. Both the excited and the ground state are further splitted by the interaction with the nuclear spin (hyperfine splitting). For cesium the nuclear spin has a value $I = 7/2$, thus the total angular momentum $F = J \oplus I$ can take the value $F = 3$ or $F = 4$ for both the ground and the excited state. Thus, we have effectively four different allowed transitions centered around 894.59 nm.

The absorption coefficient κ defined in Eq. (9) can be calculated for the four transitions linking the groundstates $|F_g\rangle$ and the excited states $|F_e\rangle$. Since each F state is further splitted into magnetic sublevels m_F , we need to account for the degeneracy, and the absorption coefficient for each $|F_g\rangle \rightarrow |F_e\rangle$ transition is:

$$\kappa_{ge}(\omega_L) = \frac{\pi \omega_{ge} n}{\epsilon_0 \hbar c} \frac{S_{ge}}{2(2I+1)} g_D(\omega_L, \omega_{ge}) \quad (15)$$

Recalling the expression for g_D from Eq. (10), at resonance, i.e. at the peak of the absorption profiles, the coefficients become:

$$\kappa_{ge}(\omega_L = \omega_{ge}) = \frac{\omega_{ge} n}{\epsilon_0 \hbar c} \frac{S_{ge}}{2(2I+1)} \sqrt{\frac{\pi}{2}} \frac{1}{\sigma_D} \quad (16)$$

In equations (15) and (16), we have defined the line strength S_{ge} [4, 5]:

$$S_{ge} = g_2 d_{ge}^2 = \sum_{m_{F_g}, m_{F_e}} |\langle F_g, m_{F_g} | e\vec{r} | F_e, m_{F_e} \rangle|^2 = c_F^2 |\langle L_g || e\vec{r} || L_e \rangle|^2 \quad (17)$$

that already takes into account the degeneracy of the excited state g_2 , whereas the term $2(2I+1)$ comes from the degeneracy of the ground state g_1 .

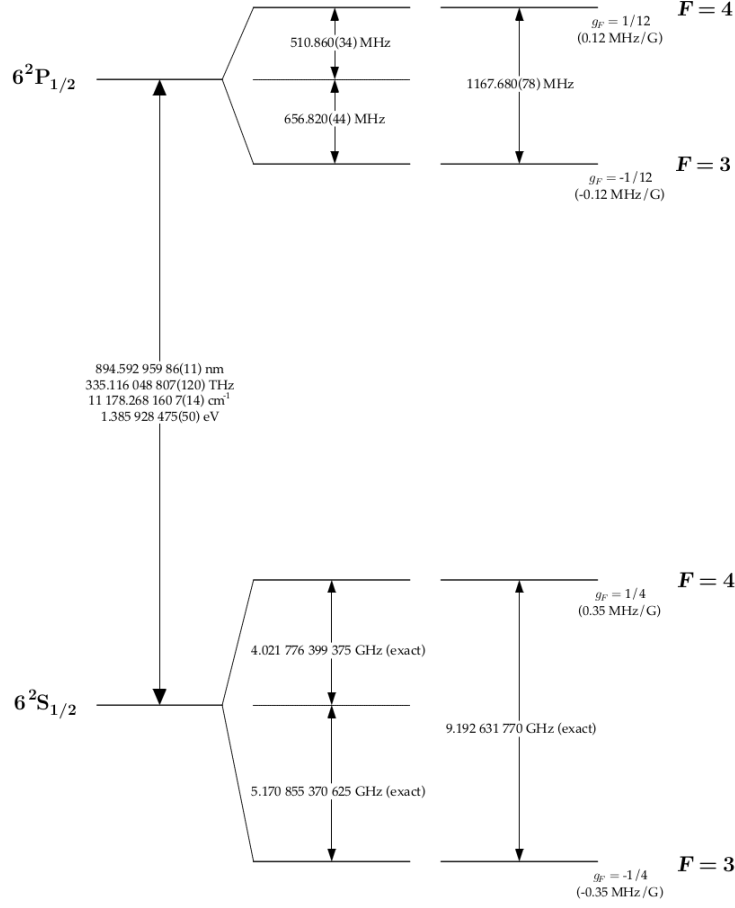


Figure 4: Energy levels of ^{133}Cs for the D_1 line (from [3]).

The term $\langle L_g \| e\vec{r} \| L_e \rangle$ can be calculated for the ground and excited state of interest, from the corresponding angular momentum and from the natural linewidth of the transition Γ (which in turn is related to the lifetime of the excited state τ , since $\tau = 1/\Gamma$). For the Cs D_1 line:

$$\langle L_g \| e\vec{r} \| L_e \rangle = \sqrt{3} \sqrt{\frac{3\pi\epsilon_0\hbar c^3\Gamma}{\omega_{ge}^3}} \quad (18)$$

and $\Gamma = 2\pi \times 4.56$ MHz. The transition strengths factors c_F^2 are instead listed in Table 1.

Transition	c_F^2
$ F_g = 4\rangle \rightarrow F_e = 3\rangle$	7/12
$ F_g = 4\rangle \rightarrow F_e = 4\rangle$	5/12
$ F_g = 3\rangle \rightarrow F_e = 3\rangle$	7/36
$ F_g = 3\rangle \rightarrow F_e = 4\rangle$	7/12

Table 1: Transition strengths factors for the Cs D₁ line.

2 Experimental activity description

The goal of this experimental activity is to perform a spectroscopic study of the cesium D₁ transition. From the analysis of the Doppler profiles, interesting properties such as the vapor density can be retrieved.

2.1 Laser characterization and wavelength tuning

The first task is to find the correct working point for the laser diode. The laser emission (wavelength and power) depends on the driving current and on the diode temperature.

The laser diode temperature is stabilized using a temperature controller (Thorlabs TED200), while the current is provided by a constant current generator (Thorlabs LDC210C). The controller reads an internal temperature sensor (a 10 k Ω Negative Temperature Coefficient (NTC) resistor). The display of the instrument shows the measured resistance R , so for obtaining the actual temperature, the Steinhart-Hart empirical formula can be used:

$$\frac{1}{T} = A + B \ln R + C (\ln R)^3 \quad (19)$$

where T is the temperature in Kelvin, R the resistance in Ohm, and A , B , C are characteristic coefficients of the thermistor that can be obtained from the laser datasheet.

The first operations are:

- Place the powermeter at the laser output.
- Stabilize the laser temperature near room temperature (e.g. 25 °C).
- Activate the current generator, slowly rising the current setpoint starting from zero mA up to the point where some laser emission is measured.

To find an optimal current working point for performing the atomic spectroscopy, we need to know the threshold current of the diode and the laser characteristic (output power vs driving current). To this end, the next activity is:

- Measure the output power as a function of the driving current, staying well below the maximum rating for the diode reported in the datasheet.

From this characterization, the threshold current is retrieved (by fitting data with a linear function), and the current setpoint can be chosen roughly between the threshold and the maximum rating.

- After the current setpoint is fixed, the temperature setpoint can be modified to tune the laser wavelength close to the atomic transition (894.59 nm, using the characterization found on the datasheet as a guidance).
- Next, the students need to complete the beam path in order to deliver a collimated beam towards the vapor cell and onto the photodetector (see Fig. 5). The setup needs to be compatible with the future integration of a counterpropagating pump beam with adjustable power.
- In order to observe the atomic resonance, a current triangular modulation is applied to the laser current by using a frequency generator. In this way, a frequency ramp is applied to the laser. The current modulation needs to be wide enough to scan a few GHz. For data acquisition, it is advantageous to trigger the oscilloscope with the function generator.
- The transmitted intensity is measured with an amplified Si photodiode (Thorlabs PD36A2) and the signal is visualized on a digital oscilloscope.

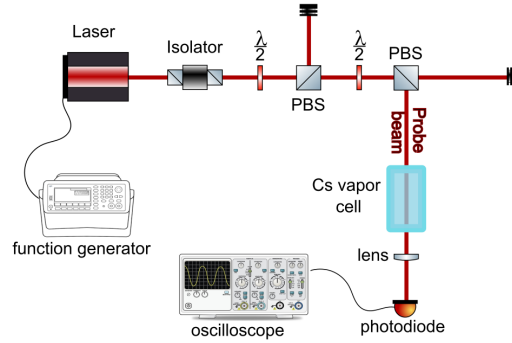


Figure 5: Sketch of the optical breadboard used to perform absorption spectroscopy on a Cs vapor cell. The first half-waveplate+PBS system is needed to reduce the optical power of the laser to work in safe conditions ($P < 0.5$ mW). The second waveplate+PBS is needed to adjust the relative power between probe and pump beams. PBS: Polarizing Beamsplitter.

2.2 Doppler spectroscopy

Next, the students will acquire the absorption signal of the four Doppler profiles corresponding to the Cs D₁ transitions. The signal can be acquired on a digital oscilloscope that needs to be set with the appropriate sampling rate and vertical scale. To increase the resolution, the same signal can be acquired simultaneously on two channels, set in AC and DC mode respectively. By combining DC and AC data, we can exploit at best the vertical resolution of the oscilloscope (on the AC data) and keep the information about the signal level (on the DC data).

- Measure the temperature and the length of the cell. It will be necessary during the data analysis.
- Acquire at least 2 measurements of the 4 absorption peaks in the same ramp at different input power (staying below 20 μW , that is $I/I_{\text{sat}} < 0.2$).

2.3 Sub-Doppler spectroscopy

Sub-Doppler spectroscopy can be performed by integrating a counterpropagating pump beam to the optical setup. An example of the optical scheme is reported in Fig. 6. In this way, the sub-Doppler dips can be used to have a

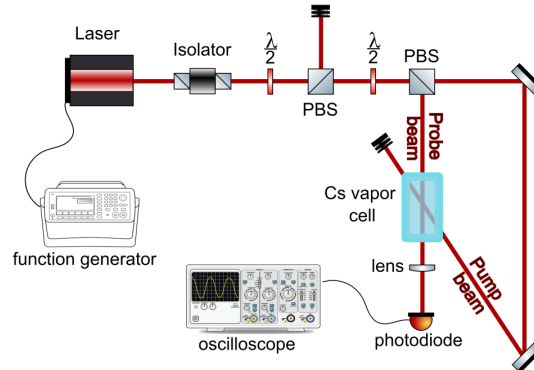


Figure 6: Experimental setup, including the pump beam to perform saturated spectroscopy.

more precise estimate of the center of the absorption profiles, and thus a more accurate calibration of the frequency axis.

- The students need to integrate the optical setup to generate the counter-propagating optical beam. The saturated peak are maximized when the pump and the probe beams are well overlapped within the cell and as collinear as possible, considering the beams geometry.

- By modulating the laser current (generating a ramp with the same properties of that utilized for the Doppler spectrum), observe the saturated spectroscopy.

Hint: To increase the resolution of the saturated peaks, it is possible to acquire also the spectrum of 2 transitions at a time, changing the time scale of the oscilloscope. If the signal is noisy, use the oscilloscope in “average mode”.

To optimize the contrast and width of the Lamb dips, the pump power can be varied.

- OPTIONAL: Acquire the saturated absorption spectra at different pump power and compare the results.

3 Data analysis

3.1 Calibration of the x-axis

The first operation for interpreting the data is a calibration of the x-axis. In fact, to analyze the atomic absorption, we need to convert the time axis of the oscilloscope data into units of laser frequency.

- Choose the saturated absorption data that presents the best contrast and width of the sub-Doppler peaks.
- Calculate the position of the center of two adjacent peaks and calculate the conversion factor $\Delta\omega_L/\Delta t$, taking into account the frequency separation of the atomic levels (see Fig. 4).

Note: The frequency response of the diode can be non perfectly linear.

OPTIONAL: Compare the calibration that is obtained fitting the position of the four atomic resonances with the calibration factors that are obtained considering only the two couples of nearest peaks separately.

Note: the conversion factor obtained using the saturated spectrum can be applied to convert all the datasets acquired if (and only if) the ramp used to modulate the laser current has not been modified.

3.2 Analysis of the Doppler profiles

- Combine AC and DC data to exploit at best the vertical resolution of the oscilloscope (on the AC data) and keep the information about the signal level (on the DC data).

The modulation of the laser current causes not only a modulation of the laser frequency, but also a modulation of the laser power. This has to be taken into account to retrieve the correct relative absorption of the atomic vapor. In case of a linear modulation the transmitted intensity is:

$$I(\omega_L, L) = I(\omega_L, 0)e^{-\sum_i \kappa_i(\omega_L)L} = (a + b \cdot \omega_L)e^{-\sum_i \kappa_i(\omega_L)L} \quad (20)$$

where i refers to the four transitions. The parameter a and b can be retrieved performing a linear fit, considering only the data far away from the atomic resonances.

- Calculate the relative transmission as $T(\omega_L, L) = I(\omega_L, L)/(a + b \cdot \omega_L)$.
- The logarithm of $T(\omega_L, L)$ can be fitted by a sum of 4 Gaussian profiles (see Eq. (9) and (10)):

$$\ln(T(\omega_L, L)) = -\sum_i \kappa_i(\omega_L)L \quad (21)$$

An example of a fit of the Cs D₁ transitions is shown in Fig. 7.

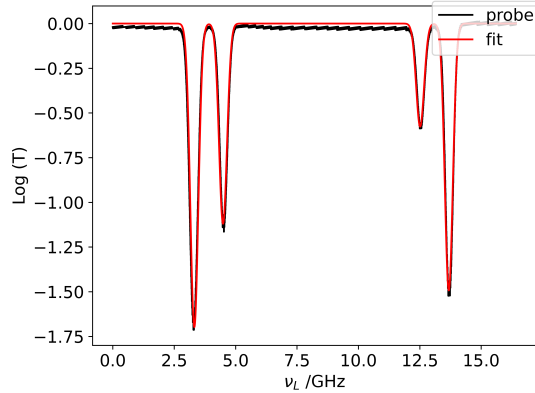


Figure 7: Example of a fit of the Cs D₁ transitions.

- From the fit parameters, calculate the Doppler width σ_D and the absorption coefficients κ_i . The experimental result of the Doppler width can be compared with the theoretical formula $\sigma_D = \sqrt{k_B T / M c^2} \omega_{ge}$.

3.2.1 Calculation of the atomic density

From the fit parameter also the density of the Cs vapor can be calculated.

- The students need to utilize the amplitude and the linewidth of the transitions to estimate the atomic density of the Cs vapor using Eq. (16).

- Compare the result of the atomic density obtained with the absorption measurement with the density estimated from the cell temperature, using Eq. (14).
- OPTIONAL: acquire several Doppler spectra at different temperatures by heating up the vapor cell. For each temperature, it is possible to experimentally measure the atomic density and compare the result with Eqs. (12)-(14).

References

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