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Comb-assisted Lamb-dip spectroscopy of the mercury intercombination line at 253.7 nm for metrological applications

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Summary. — The saturated absorption spectrum of mercury vapors is measured in coincidence with the $6s^2\ ^1\mathrm{S}_0 \to 6s6p\ ^3\mathrm{P}_1$ intercombination transition at 253.7 nm. Coherent radiation in the deep-UV region is produced by means of a double stage of second harmonic generation of a 1014.8 nm external cavity diode laser (ECDL) in a pair of nonlinear crystals. The ECDL frequency is tightly locked to the nearest tooth of a self-referenced optical frequency comb synthesizer, traceable to a GPS-disciplined time base signal provided by a Rb-clock. We present the measurement of the pressure-broadening coefficient due to Hg-Hg collisions by analyzing the Lamb-dip profile at different values of the mercury vapor pressure. This study is significant for ongoing experiments that employ the mercury intercombination line for temperature metrology as well as tests of fundamental symmetries.

1. – Introduction

The pursuit of high precision in spectroscopic measurements is a cornerstone in various scientific research domains such as atomic and molecular physics, atmospheric chemistry, material sciences, and fundamental metrology. Saturation spectroscopy emerges as a pivotal technique within the realm of precision spectroscopy, capitalizing on the velocity-selective saturation of Doppler-broadened atomic or molecular transitions. Beyond its Doppler-free advantage, this method finds motivation in addressing challenges related to signal-to-noise ratios and sensitivity, crucial for applications demanding high-fidelity measurements. This paper describes Doppler-free saturated-absorption spectroscopy of mercury atoms at 253.7 nm, in coincidence with the intercombination transition. The immediate motivation for our work comes from the field of temperature metrology, as this transition has been recently chosen for an ongoing experiment in Caserta aimed at developing a new primary thermometer based on Doppler-broadened precision spectroscopy of mercury atoms in the UV region [1]. Doppler-broadening thermometry provides a promising avenue for the practical realization of the new kelvin unit. By examining the broadening of spectral lines caused by the thermal motion of atoms or molecules in any

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gas sample at the thermodynamic equilibrium, this method offers a direct way to thermodynamic temperature determinations [2]. The present state of Doppler-broadening thermometry involves advanced experimental setups and analytical methodologies [3]. The analysis of the Doppler-broadened profiles is conducted with sophisticated theoretical models, to carefully take into account collisional broadening and narrowing effects within the gas medium [4].

We report on the measurement of the self-broadening coefficient of the 6s^2 ^{1}S$_0$ →6s6p ^{3}P$_1$ transition, a key parameter aiding in characterizing the collisional dynamics contributing to the width and shape of the line.

2. – Experimental apparatus

The spectrometer is based on a dual-stage second harmonic duplication of a 1014.8 nm external-cavity diode laser (ECDL), as detailed in refs. [5, 6]. The first doubling stage (1014.8 nm → 507.4 nm) involves a fiber-coupled periodically poled lithium niobate crystal waveguide. The green beam power is amplified using the injection locking technique in a seed laser. For the second doubling stage (507.4 nm → 253.7 nm), we employ a home-made bow-tie build-up cavity where a beta barium borate crystal is situated. A laser beam intensity stabilisation system has been devised that incorporates an acousto-optic modulator as an adjustment component within the intensity control feedback loop. This system is complemented by a silicon carbide (SiC) photodetector, which monitors a portion of the zero-order beam. The residual UV radiation is sent to a 1-mm-long mercury vapor cell equipped with a pair of far-UV quartz windows. The Hg reservoir placed in the middle of the cell is submerged in a custom liquid-bath thermostat utilizing Peltier cells. We used a standard pump and probe setup for saturation spectroscopy. In this arrangement, the laser beam travels through the thin cell and then it is retroreflected toward the cell by a plane mirror. The laser beam is then directed and focused onto a preamplified SiC detector connected to a $6\frac{1}{2}$-digit multimeter. The near-infrared ECDL laser is tightly locked to an optical frequency comb synthesizer, following the scheme of ref. [6]. The comb repetition rate, f_{rep}, and carrier-envelope offset frequency, f_{ceo} are precisely stabilized to a GPS-disciplined Rb clock. Calibrated frequency scans across the intercombination line are obtained with a continuous variation of the f_{rep} frequency, while the ECDL is locked to the comb. Throughout the scanning process, we maintain continuous monitoring of the ECDL frequency using a nine-digit wavemeter to ensure the absence of mode-hops in the near-infrared laser. Simultaneously, we monitor the radiation at 507 nm on an optical spectrum analyzer to check the quality of the injection locking.

3. – Doppler-free saturation spectroscopy of the ^{200}Hg transition at 253.7 nm

An example of the Lamb-dip spectrum for the $6s^2$ ^{1}S$_0$ → $6s6p$ ^{3}P$_1$ ^{200}Hg transition is shown in fig. 1 at the gas temperature of 289.75 K, corresponding to a vapor pressure of about 0.127 Pa [7]. As detailed below, a rigorous data analysis is conducted with the adoption of a Voigt-type profile. In the weak saturation regime, where the power broadening is negligible, the Lamb-dip line shape can be considered as the combined effect of transit-time broadening, expressed through a Gaussian component, and homogeneous line broadening due to the joint occurrence of natural broadening and damping (and dephasing) mechanisms, represented by a Lorentzian component. Moreover, the Voigt profile takes into account the residual broadening due to the UV radiation width. By

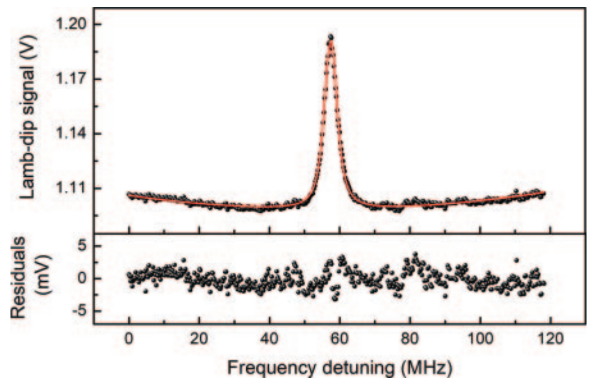

Fig. 1. – Example of Lamb-dip spectrum at the Hg vapor pressure of 0.127 Pa. The root-mean-square value of the absolute residuals amounts to ∼1 mV, while the signal-to-noise ratio is ∼100. The contrast of the sub-Doppler signal compared to the Doppler envelope is ∼5%.

employing a MATLAB code, each spectrum is fitted by the function

$$P(\nu) = [P_0 + P_1\nu + P_2\nu^2] \times e^{-Ag(\nu-\nu_0)}, \tag{1}$$

where ν_0 denotes the line center frequency, the parameters P_0, P_1, P_2 accounts for a quadratic background, A represents the frequency integrated absorbance and $g(\nu-\nu_0)$ is the normalized Voigt convolution. The parameters mentioned above, together with the Gaussian and Lorentzian widths of the lineshape function, are treated as free variables in the non-linear least-squares fitting procedure. The consistency between the model and the observed data is evidenced by the good residuals reported in the bottom of fig. 1. The full width at half maximum (FWHM) of the saturation dip spectrum is retrieved from the fit to be about 4.5 MHz.

We acquired the Lamb-dip signal at Hg temperatures from 285.75 K to 295.15 K, corresponding to a vapor pressure from 0.089 Pa to 0.203 Pa. The spectral analysis leads to the Lorentzian widths plotted in fig. 2 as a function of the vapor pressure. A linear fit of the data yields an intercept of (2.2 ± 0.2) MHz and a slope of (11 ± 2) MHz/Pa.

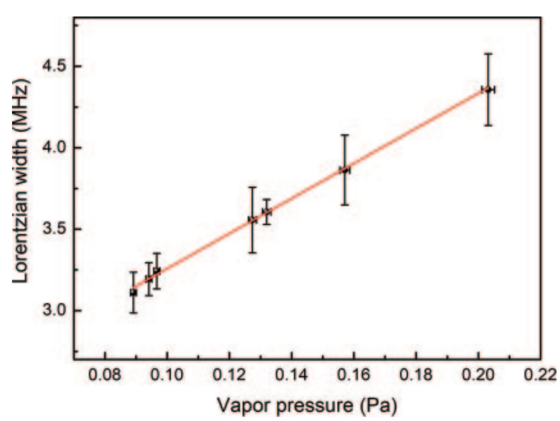

Fig. 2. – Lorentzian width of the Lamb-dip profiles as a function of the Hg vapor pressure. Error bars represent the statistical uncertainties.

The reported uncertainties are of statistical nature and result from the weighted linear fit including the errors on both variables. The slope of the line represents the pressure-broadening coefficient resulting from Hg-Hg collisions. The extrapolated zero-pressure Lorentzian width is consistent with the sum of the natural contribution ($\sim$1.3 MHz) [6, 8], the Lorentzian component of the UV radiation width ($\sim$70 kHz) and the Zeeman broadening due to the Earth magnetic field ($\sim$1 MHz).

4. – Conclusion

We have reported on comb-calibrated saturated absorption spectroscopy of the $6s^2\ {}^1\mathrm{S}_0 \rightarrow 6s6p\ {}^3\mathrm{P}_1$ intercombination transition of mercury in the deep-UV. By employing an accurate spectral analysis of the Lamb-dip profiles, we have measured the ^{200}Hg self-broadening coefficient. It is well known that pressure broadening in the saturated absorption regime is typically larger than that derived from conventional unsaturated Doppler-limited spectra [9]. From this consideration, it follows that the measured value can be considered an upper limit to pressure broadening in the spectral analysis of Doppler-broadening thermometry. However, the results of the present work are relevant for testing theories of collisional line broadening and for studying atomic force fields.

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