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Spectroscopy of the Mg $^1S_0 - ^3P_1$ intercombination transition in a luminescent cell with walls at room temperature

V.I. Baraulya, A.E. Bonert, A.N. Goncharov, A.M. Shilov

Abstract. Absorption at the $^1S_0 - ^3P_1$ intercombination transition in magnesium atoms is studied experimentally. The saturated absorption resonance of thermal magnesium atoms in a compact low-pressure absorption cell with walls at room temperature was recorded for the first time by using the spatial trapping of excited magnesium atoms in the detection region and the time separation of luminescence excitation and detection.

Keywords: intercombination transition in magnesium, high-resolution spectroscopy, optical frequency standard.

1. Introduction

Thermal absorption cells are widely used in spectroscopy for recording absorption lines of chemical elements and compounds in gaseous state. The application of these cells in the Doppler-free laser spectroscopy provides a high spectral resolution, which is limited by the collision and time-of-flight line broadening. The studies of optical frequency standards based on intercombination transitions in alkaline-earth Mg [1, 2] and Ca [3, 4] atoms stimulated investigations aimed at the development of thermal cells for these atoms.

The principal features of a new generation of optical frequency standards based on cold atoms captured in magneto-optical and optical traps are a small width ($\Delta\nu \sim 1 - 100$ Hz) of the ‘clock’ transition used to stabilise the laser frequency and the separation in time of the recording process related to the alternation of the trapping, cooling, and measurement cycles. The frequency of the measurement cycle is approximately a few tens of hertz. Therefore, the short-term frequency stability during averaging times shorter than 0.1–1 s should be provided by the preliminary stabilisation of the laser frequency, for example, with the help of a Fabry–Perot interferometer isolated from external acoustic and vibrational perturbations. Such a frequency stabilisation by the transmission band of a

high- Q interferometer can provide a narrow emission line-width down to fractions of a hertz. However, due to uncontrollable temperature variations and aging of the interferometer base material, it is difficult to provide the frequency drift smaller than $1 - 10$ Hz s^{-1} . To reduce this frequency drift and perform spectroscopic studies of cold atoms confined in a trap, it is useful to stabilise preliminary the radiation frequency, for example, by employing saturated absorption resonances at the ‘clock’ transition in an external absorption cell as the natural frequency reference. Such frequency stabilisation is also of interest for the development of mobile secondary frequency standards based on the intercombination transitions in magnesium and calcium atoms.

The operation principle of the known intercombination-transition magnesium and calcium cells is based on the recording of the absorption resonance using the transmitted beam power. But, taking into account the parameters of intercombination transitions and physicochemical properties of alkaline-earth element vapours, this method requires the use of large cells heated to high temperatures, continuous vacuum pumping, and a complicated protection of the cell windows.

In this paper, we studied the possibility of the luminescence recording of saturated absorption resonances at the weak two-level transition at low optical densities for the development of a compact sealed off cell used as the reference at the $^1S_0 - ^3P_1$ transition in magnesium.

2. Features of intercombination-transition absorption cells

To obtain the optical density of the order of unity at weak intercombination transitions in Mg and Ca in a cell of length 1–2 m, the vapour pressure should be 10–30 mTorr. Such a pressure for the magnesium vapour is obtained at temperatures 400–500 °C, and therefore the cell walls and windows should be heated to higher temperatures. However, the magnesium atom vapour actively interacts with the material of vacuum windows (oxides like glass, quartz, sapphire) even at moderate temperatures, beginning from ~ 250 °C, resulting in a drastic degradation of their transparency. The use of fluoride windows (MgF_2 , CaF_2 , BaF_2) is also complicated because the soldering technology of such windows with metals restricts their application at temperatures above 200–250 °C. The cell windows can be protected against degradation with the help of a buffer gas [5]; however, the gas pressure should be in

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the range from 1 to 10 Torr, which results in a considerable broadening of saturated absorption resonances. Thus, the use of conventional thermal alkaline-earth element cells for recording saturated absorption resonances at the weak intercombination transition is complicated.

The designs of vacuum cells for calcium and magnesium atoms with double windows and internal mirrors were proposed and realised in [6–8]. Unfortunately, these cells are bulky and require constant pumpdown; as a result, their application in compact laser systems is complicated.

Our previous studies have shown that at room temperature no interaction of magnesium atoms with cell window materials resulting in their degradation was observed. Moreover, the adhesion coefficient for magnesium atoms to windows at the vapour pressure $10^{-5} - 10^{-6}$ Torr proves to be so small that the transparency of windows is not degraded for hundreds hours of operation. This allowed us to fabricate a compact sealed off absorption magnesium cell for the $^1S_0 - ^1P_1$ transition in magnesium with the internal source of magnesium atoms and walls at room temperature [2]. However, this cell cannot be used directly at the $^1S_0 - ^3P_1$ transition due to a low absorption coefficient $\alpha = \sigma nL \approx 10^{-6}$, where $\sigma = 1.8 \times 10^{-17} \text{ cm}^2$ is the effective absorption cross section for the $^1S_0 - ^3P_1$ transition; $n \approx 10^{10} \text{ cm}^{-3}$ is the density of magnesium atoms in the cell; and $L = 10 \text{ cm}$ is the cell length.

The minimal detectable absorption limited by the shot noise upon the direct detection of saturated absorption resonances in the power of a probe counterpropagating wave is $\alpha_{\min} = \sqrt{2e\Delta f/SP}$, where e is the electron charge; Δf is the detection bandwidth; S is the detector sensitivity; and P is the probe beam power. The minimal detectable absorption at a wavelength of 457 nm of the $^1S_0 - ^3P_1$ intercombination transition for the maximal possible $S = 0.37 \text{ A W}^{-1}$ (the quantum efficiency of the photodetector is $\eta = 1$), for $P = 1 \text{ mW}$, $\Delta f = 1 \text{ Hz}$, and the signal-to-noise ratio equal to unity is $\alpha_{\min} = 3 \times 10^{-8}$. The contrast of saturated absorption resonances for the saturation parameter and optical density of the cell equal approximately to unity is ~ 0.1 . Thus, the possibility of direct recording of nonlinear resonances is at the limit of the sensitivity of the direct detection even in the ideal case of the shot noise.

3. Possibility of a Mg luminescent cell development

The signal-to-noise ratio can be considerably increased by recording absorption by using a luminescence signal because in this case only the signal proportional to the power absorbed in the cell is detected, without a considerable contribution from the unabsorbed power. The increase in the signal-to-noise ratio compared to direct recording method in the case of the shot noise is $(K_r/\alpha)^{1/2}$, where K_r is the recording coefficient equal to the ratio of the number of detected photons to the number of excited atoms. For resonance transitions with the short lifetime τ of the upper level ($\tau \leq \tau_{\text{fl}}$, where τ_{fl} is the flight time of atoms through the laser beam), the diameter D of the luminescence region is of the order of the probe laser beam diameter, and in this case $K_r \approx \Omega\eta$ (where Ω is the solid angle). The diameter of the luminescence region for the $^1S_0 - ^3P_1$ transition of magnesium in a low-pressure cell greatly exceeds that of the excitation region because

$\tau = 5 \text{ ms}$ and $D = 2\langle v \rangle\tau = 8 \text{ m}$, where $\langle v \rangle = 800 \text{ m s}^{-1}$ is the average thermal velocity of magnesium atoms. Taking into account that at the centre of the absorption line the atoms are excited which intersect the laser beam at an angle of $\theta \sim 90^\circ$, the luminescence region has a cylindrical shape with diameter D , and the signal is mainly determined by the contribution from the inner region of diameter approximately equal to the distance from the photodetector to the laser beam. Then, $K_r \approx (d/D)^2\eta$, where d is the photocathode diameter. For typical values $d = 5 \text{ cm}$ and $\eta = 0.1$, we have $K_r \approx 10^{-6} \approx \alpha$. Thus, it is impossible to increase directly the signal-to-noise ratio in a magnesium luminescent cell compared to an absorption cell.

The luminescence detection sensitivity is also limited in our case by the fact that excitation and luminescence occur in the two-level scheme, i.e. at the same wavelength. Therefore, optical filters cannot eliminate the probe beam radiation scattered from cell windows and incident on the photodetector. It was pointed out in [9] that the sensitivity of luminescence spectroscopy at forbidden two-level transitions is mainly limited by scattering of laser light from cell windows. The level of the parasitic signal is determined by the scattering coefficient of cell windows, the screen emissivity, and the geometrical factor. Estimates and experiments showed that in our case of a cell of limited size, it is difficult to obtain the useful-to-parasitic signal ratio at the 10^{-3} level.

To improve considerably the useful-to-parasitic signal ratio, we propose here to separate in time luminescence excitation and detection and restrict the drift rate of excited atoms from the luminescence collection region by using their collisions with noble gas atoms. Such collisions, as shown in [10], do not affect considerably the radiative relaxation time of the 3P_1 level. In the case of the sufficient density of noble gas atoms, excited magnesium atoms can be localised in the luminescence collection region during the lifetime of the 3P_1 level. We do not know the diffusion coefficient of magnesium atoms in the 3P_1 state in argon gas and, therefore, will assume that it is equal to the diffusion coefficient D_{dif} of argon atoms in argon gas. According to [11], $D_{\text{dif}} \approx 10^2/p \text{ cm}^2 \text{ s}^{-1}$ at room temperature, where p is the argon pressure in torr. The diffusion length $(D_{\text{dif}}\tau)^{1/2}$ at the pressure of $p \sim 10^{-2}$ Torr is 7 cm, which is comparable with the size of the luminescence collection region in a compact cell of size $L \sim 10 \text{ cm}$.

Of course, the undesirable broadening of the transition line by argon will take place. However, it should not restrict considerably the practical value of a luminescent cell. According to the data obtained in [6], the line broadening by the intrinsic magnesium pressure is 12 MHz Torr^{-1} ; therefore, the line broadening in the cell at the argon pressure 10^{-2} Torr will be, probably, about 100 kHz.

4. Experimental results

Figure 1 shows the scheme of the experiment. Luminescence was excited by a 457-nm, 200-mW Ti:Al₂O₃/KNbO₃ laser [12] with the linewidth less than 10 kHz. A luminescent magnesium cell was a stainless steel vacuum chamber of diameter 120 mm with quartz windows with AR coatings at $\lambda = 457 \text{ nm}$. Luminescence was detected with a FEU-97 photomultiplier located in one housing with a preamplifier. The time constant of the photomultiplier for a weak light signal was $\sim 100 \text{ ns}$. The parasitic light incident on the

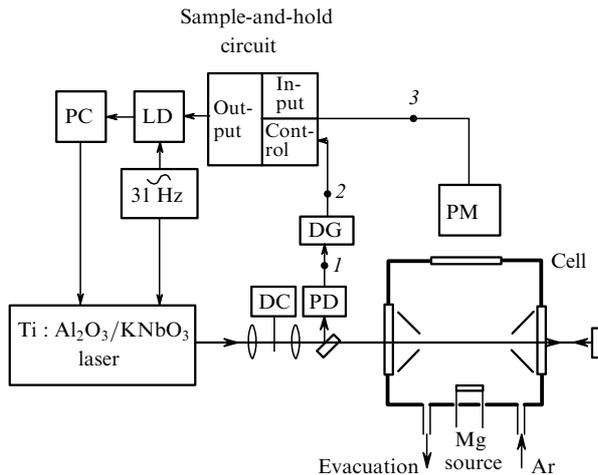


Figure 1. Scheme of the experiment: DC: disc chopper; PD: photodiode; DG: delay generator; LD: lock-in detector; PM: photomultiplier with a preamplifier; PC: personal computer; (1) photodiode output; (2) delay generator output; (3) PM output.

photomultiplier due to scattering of the laser beam from cell windows and walls was reduced with the help of conic apertures mounted inside the vacuum chamber. The parasitic signal caused the saturation of the photomultiplier, but did not produce irreversible consequences; the relaxation time τ_{rel} of the photomultiplier sensitivity after the laser beam was blocked did not exceed 30 μs .

A cloud of magnesium atoms was produced by using a direct-heating thin-wall stainless steel capsule with a ~ 0.3 -mm hole filled with metal magnesium. The vacuum chamber was pumped out to a pressure of $\sim 10^{-5}$ Torr, and the buffer argon gas was supplied upon continuous evacuation. The density of magnesium atoms averaged over the chamber length was measured by the absorption of laser radiation at the resonance 285-nm line and was maintained at the level $\sim 4 \times 10^{10} \text{ cm}^{-3}$ over the entire argon pressure range ($1 \times 10^{-4} - 5 \times 10^{-2}$ Torr).

The luminescence signal was detected in the time-selection regime, as shown in the diagram in Fig. 2. Signal (1) is the meander of probe laser pulses obtained by using a disc chopper placed in the waist formed by two lenses of a telescope (see Fig. 1). The duration of the laser pulse front for a chopping frequency of 2 kHz was 2 μs . Strobe pulses (2) were formed by a delay generator. The time delay T_d was chosen according to the condition $T_d > \tau_{\text{rel}}$ ($\tau_{\text{rel}} \approx 30 \mu\text{s}$). Output signal (3) of the photomultiplier was supplied to a sample-and-hold circuit controlled by strobe signal (2). The output signal of the sample-and-hold circuit was supplied either to an ADC (upon recording linear absorption) or to a lock-in detector (upon recording saturated absorption resonances). The laser wavelength was controlled with a λ meter, which allowed us to tune preliminarily the laser frequency to the centre of a Doppler absorption line with an accuracy of ~ 50 MHz.

The separation of the luminescence signal at the centre of the absorption line is demonstrated in Fig. 3 which shows the time dependence of the output signal of the sample-and-hold circuit upon switching on the magnesium source followed by filling with argon. At the beginning of recording, only radiation scattered from chamber windows is incident on the photodetector, but it does not make a noticeable contribution to the output signal. The voltage is

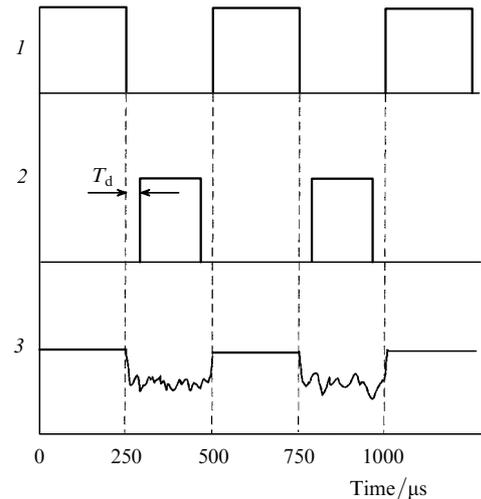


Figure 2. Time diagram for selecting luminescence signals (see Fig. 1).

applied to the magnesium source at the instant t_1 , which is followed by the increase in the signal due to the thermal radiation from the source. The instant t_2 corresponds to the beginning of filling the chamber with argon. The argon pressure continuously increases up to 5×10^{-2} Torr by the moment t_3 and the luminescence signal increases and then saturates. The leak is closed at the moment t_3 , and the signal decreases with decreasing argon pressure. The magnesium source is switched off at the instant t_4 . The maximum intensity of the luminescence signal was approximately 10^{-3} of the signal caused by laser radiation scattered from chamber windows. In the control experiment upon frequency detuning by 10 GHz from the absorption line centre, only a decrease in the radiation emission of the magnesium source was observed upon filling the chamber with argon, which was caused by its cooling by argon (the dashed curve in Fig. 3). The thermal emission signal from the magnesium source was completely suppressed with a SZS-22 filter mounted in front of the photomultiplier, the luminescence signal being reduced only by $\sim 10\%$.

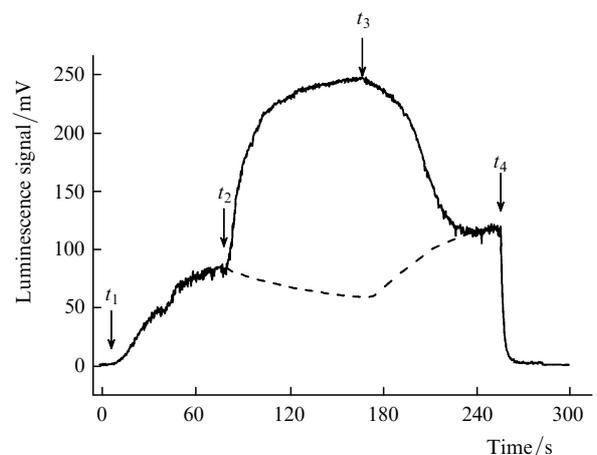


Figure 3. Time dependence of the luminescence signal at the absorption line centre. The arrows denote the instants of switching on (t_1) and switching off (t_4) of the magnesium source and the beginning (t_2) and end (t_3) of the argon admission; the dashed curve shows the emission of the magnesium source.

The luminescence intensity at the beginning of time intervals corresponding to the shut off laser beam remained invariable with decreasing the chopping frequency and decreased with time within these intervals. The decay time was close to the lifetime τ of the 3P_1 level, which demonstrates the diffusion nature of the motion of excited magnesium atoms in the absence of collision quenching. Thus, the increase in the luminescence signal can be estimated from the relation $(\langle v \rangle \tau / R)^2 \approx 10^4$, where $R \approx L/2 = 5$ cm is the radius of the detection region.

Saturated absorption resonances were recorded by using frequency-modulated counterpropagating waves with the synchronous detection of the luminescence signal. The laser beam power was ~ 40 mW and its radius was $w_0 \sim 0.45$ mm. The modulation frequency (31 Hz) was lower than the laser beam chopping frequency and the inverse decay time of the luminescence signal. We did not attempt to resolve the Zeeman components of the $^1S_0 - ^3P_1$ transition.

Figure 4 shows a saturated absorption resonance recorded for the modulation amplitude of 2.9 MHz. The approximation of the resonance shape by using the results obtained in [13] taking into account its broadening caused by the probe modulation (solid curve in Fig. 4) shows that the homogeneous width of the resonance is 1.3 MHz. This exceeds the time-of-flight width $\langle v \rangle / 2w_0 = 0.67$ MHz. It seems that the resonance is formed by the overlap of the unresolved central (π) and side (σ^\pm) Zeeman components. The frequency of the π component for the $^1S_0 - ^3P_1$ transition is independent of the magnetic field, and the frequency shift of the σ^\pm components is 2.1 MHz G^{-1} , which, taking into account that the magnetic field in our setup is ~ 0.5 G, can explain the discrepancy pointed out above. The influence of the magnetic structure on the resonance width can be eliminated by applying a bias magnetic field of ~ 10 G to shift the σ^\pm components by the value greatly exceeding the resonance width, and to record only the π component. The residual resonance broadening will be mainly determined by the time-of-flight and collision broadening, which can be estimated from the dependence of the resonance broadening on the argon pressure.

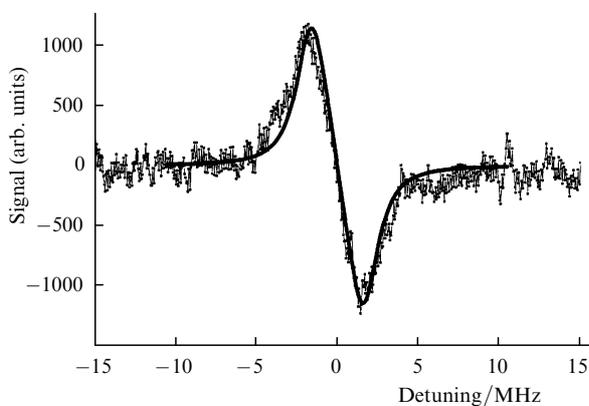


Figure 4. Shape of the saturated absorption resonance recorded with a lock-in detector with the integration constant $\tau = 0.1$ s; the solid curve shows the first derivative of the Lorentzian line shape of width 1.3 MHz for the modulation amplitude of 2.9 MHz.

5. Conclusions

We have demonstrated the recording of saturated absorption resonances at the $^1S_0 - ^3P_1$ transition in magnesium in a compact cell with walls at room temperature. The influence of the probe laser radiation scattered from the cell windows was completely eliminated by using time sampling. The addition of argon gas leads to the increase in the luminescence signal by 10^4 times due to the spatial localisation of excited magnesium atoms in the detection region during the lifetime of the 3P_1 level. Our estimates and experiments have shown that the spectral resolution achieved with this cell compares well with that of the known cells. We plan to use these results to stabilise the frequency of a 457-nm laser for the precision spectroscopy of cold magnesium atoms in a magneto-optical trap. The results of our paper can be used to study the collision-induced emission of magnesium atoms in the 3P_1 state in noble gases.

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