Coherent magneto-optical resonances in hot potassium vapor

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Abstract. In this communication, we present experimental results on the preparation of magneto-optical resonances in potassium. The magneto-optical resonances are registered in Hanle configuration, i.e. monitoring the atomic fluorescence vs the applied magnetic field scanned around zero value. The excitation of the second resonance line with a wavelength of 404.4 nm leads both to direct fluorescence at 404.4 nm and to a cascade decay with fluorescence at 770.1 nm and 764.5 nm (first resonance line transitions). This transfer is evidenced by the registration of a narrow magneto-optical resonance in the fluorescence. It should also be noted that even after multiple cascade transitions, the resonance observed on the infrared lines has a much better signal/noise ratio compared to that on the excited second resonance line. Therefore, the study of these sub-Doppler-width magneto-optical resonances in hot potassium vapor will be useful for the further development of high-resolution magneto-optical laser spectroscopy.

1. Introduction

Magneto-optical (MO) spectroscopy is a tool that is extensively applied in many scientific fields [1]. A key feature of this type of coherent spectroscopy is the possibility to register narrow MO resonances, whose width is determined by the ground-state hyperfine (hf) level lifetime. Potassium (K) vapor is used in the most sensitive optical pumping atomic magnetometers, which operate at high temperatures in order to achieve high alkali vapor densities and narrow optical resonances since for K the hf ground-state splitting is smaller than the Doppler width, the nonlinearity of the Zeeman splitting dependence on the magnetic field is more significant than that for other alkali atoms and the isotopic shifts of the two stable isotopes (³⁹K, ⁴¹K) are smaller than the Doppler width [2].

In a previous work, we have studied the excitation transfer from the second resonance line of ³⁹K (4s²S_1/2→5p²P_3/2 transition) with a wavelength of 404.4 nm to the first resonance line (4s²S_1/2→4p²P_1/2 transition) with a wavelength of 770.1 nm observed in hot atomic vapor. It was shown that besides the atomic population, atomic polarization is also transferred, and the signal-to-noise ratio of the resonances registered in fluorescence at the first resonance line is significantly higher than for those obtained at the excited second resonance line [3].
In this work, we present experimental investigations of coherent MO resonances on the first and the second resonance line in hot K vapor by exciting the second resonance line in a large interval of atomic densities. The influence is discussed of the spin-exchange collisions, radiation trapping, Rayleigh scattering, and geometry of registration on the shapes of the MO resonances. The results are providing new knowledge about the atomic coherence and its transfer dependence on the vapor density, as well as the collective optical processes in hot atomic ensembles and their application as a medium for quantum control and sensing.

2. Experimental setup
The scheme of the experimental setup is presented in figure 1. In this experiment, K vapor contained in an evacuated optical cell of diameter $D = 2.5$ cm and length $L = 5$ cm was excited by a tunable single-frequency, linearly polarized laser light, resonant with the 404.4 nm violet transition (figure 2). A single-mode extended-cavity diode laser (ECDL) with laser linewidth of the order of 1 MHz was used. The laser power used in the experiment was 2 mW, and the laser beam cross-section was 0.02 cm$^2$. The optical cell was shielded against stray magnetic fields.

The MO resonances are registered in the fluorescence in the so-called Hanle configuration, where the magnetic field $\vec{B}_{scan}$ is scanned in a direction perpendicular both to the laser propagation vector $\vec{k}$ and its electric vector $\vec{E}_{las}$. Depending on the polarization, two types of fluorescence signals are distinguished: perpendicular and parallel to the electric field vector $\vec{E}_{las}$ ($I_{orth}$ and $I_{par}$, respectively). The fluorescence profiles at 404.4 nm and 770.0 nm are registered by a photomultiplier System (PMS). Band-pass filters are used for registration of the violet (404.4 nm) and the infrared (770.1 nm) line separately. A polarizer is used for registration of the polarized fluorescence signals $I_{orth}$ and $I_{par}$.

![Figure 1. Experimental setup. NBF – narrow band filters, P – polarizer, AS – acquisition system.](image1)

![Figure 2. Energy levels diagram of K. Excitation at 404 nm, MO resonances registration at 404 nm and 770 nm.](image2)

3. Results and discussion
3.1. 404 nm MO resonances at 404 nm excitation
The 404.4 nm excitation of K induces coherent superposition of the ground state Zeeman sublevels at $B = 0$, which is measured by the direct 404.4 nm fluorescence as a function of the magnetic field. The atomic polarization formed is also transferred to the atomic levels of the first resonance line.
The fluorescence profiles $I_{\text{orth}}$ and $I_{\text{par}}$ of the 404 nm MO resonances are both dark in the whole range of temperatures (117-157 °C). However, they are quite different in amplitude, shape and width. In figure 3 and figure 4 the normalized $I_{\text{orth}}$ and $I_{\text{par}}$ at 128 °C and 157 °C are shown. At equal temperatures the amplitude of $I_{\text{orth}}$ is higher, and the width is narrower. The difference $I_{\text{orth}}-I_{\text{par}}$ is Lorentzian in shape in the entire temperature range, while the wings of the MO resonances increase almost linearly with the temperature.

It is worth noting that the 404 nm fluorescence profiles are superimposed on a background of 404 nm scattered laser light, whose level in this case is 25% of the maximal fluorescence measured at $T_{\text{cell}} = 117$ °C. Moreover, the scattered light background shows a significant increase with the atomic density, when measured in the far wings of the respective Doppler profiles [3].

The Lorentzian difference and the scattered light could be related to Rayleigh scattering [4]. The shape of the resonances has a complex shape – a narrow structure superimposed on a power-broadened coherent population trapping (CPT) resonance. The fluorescence angular distribution agrees with the assumption that this narrow structure is due to atom interaction with the Rayleigh-scattered light.

The evaluated width of the Rayleigh scattered resonances in K at room temperature is 20 mG, however, at high temperatures the mean free path (MFP) in K is of the order of $10^3$ times smaller as a result of the higher density. In this case, the spin-exchange collisions [5] and photon reabsorption become significant [6,7]. The threshold atomic density for radiation trapping can be calculated from the following condition:

$$\frac{3}{8\pi} N\lambda^2 D \frac{\gamma_r}{W_d} > 1$$

where D is the diameter of the cell, N is the atomic density, $\gamma_r$ is the radiative decay rate and $\lambda$ is the transition wavelength. In our case, the evaluated threshold value for radiation trapping is $1.4 \times 10^{13}$ atoms/cm$^3$ ($T_{\text{RT}} = 151$ °C).

The contributions of radiation trapping and Rayleigh scattering to the fluorescence profiles have opposite signs. As a result, when the temperature is increased they become significant and compete. The radiation trapping leads to ground-state coherence relaxation and destruction of the CPT resonance. There is a change in the shape without inversion of the profile (figure 4). As it is shown, the resonance destruction is more effective in the wings of the $I_{\text{orth}}$ profile, while at its center the resonance is more resistant (figure 4 blue profile). Thus, resonance destruction can result in its narrowing [8]. At 128 °C, in $I_{\text{par}}$ there is a narrow, inverted structure, which is not visible at 157 °C.
3.2. 770 nm MO resonances at 404 nm excitation

Figure 5 presents the 770 nm MO resonances excited at 404 nm and registered in parallel and orthogonal polarization as a result of the cascade decay. The shapes of the MO resonances in the two orthogonal polarizations are similar (figure 5). When the photon reabsorption becomes significant ($T_{RT} = 151$ °C), their shapes invert for both types of polarization (figure 5a,b) and the fluorescence level of the wings starts to decrease due to the competition of the two processes (figure 5c).

![Figure 5a. Shape of the 770 nm MO resonances registered in ORTH normalized fluorescence.](image)

![Figure 5b. Shape of the 770 nm MO resonances registered in PAR normalized fluorescence.](image)

![Figure 5c. Level of fluorescence wings with ORTH and PAR polarization.](image)

Besides, at temperatures slightly higher than $T_{RT}$ (165 °C), the sum of the intensities of the MO signals in both polarizations (i.e. the unpolarized fluorescence, see section 3.3 and figure 6) has a narrow resonance on a pedestal. Its width is comparable to the width of the CPT resonances excited by 770 nm and registered at the same transition (figure 7). This transfer is evidenced by the registration of a narrow MO resonance in the fluorescence from the 4p$^2$P$_{1/2}$ IR line, which is with opposite sign.

![Figure 6. 404 nm MO resonances by 404 nm excitation ($I_{orth} + I_{par}$) at 165 °C.](image)

![Figure 7. 770 nm MO resonances by 770 nm excitation at 165 °C.](image)

3.3 Geometry of registration and interpretation of the experiments

In the experiments, the scanned magnetic field $\vec{B}_{scan}$ is orthogonal to the plane defined by the electric vector $\vec{E}_{las}$ and the direction of propagation of the laser radiation $\vec{k}_{las}$ (figure 1). For this geometry, the intensity of the MO fluorescence resonances is determined by the magnetic-field-dependent polarization moments $\langle P_{i}^{a} \rangle$ $i = 2$ linear, $i = 4$ nonlinear) and the observation tensor $T_{q}^{\kappa}(\vec{e}_{n})$ [9]; therefore we obtain:
\( I_{f \rightarrow f_\varphi}(\vec{e}_n) = \)

\[
C_0 \sum_{F_f} \frac{\|d_f\|^2}{3(2F_f+1)} \begin{pmatrix} (4)f_0^0 + (-1)^{F_f}f_\varphi^15 \sqrt{\left(2F_f + 1\right)} \begin{pmatrix} 1 & 2 \\ F_f & F_f \end{pmatrix} \end{pmatrix} + \begin{pmatrix} (4)f_2^2 T_2^2(\vec{e}_n) + (4)f_2^2 T_2^2(\vec{e}_n) + (2)f_2^2 T_2^2(\vec{e}_n) \end{pmatrix} \]

\( \) \( (2) \)

The multiplier term in front of the curly braces and the corresponding polarization moment \( f_\varphi^k \) are denoted by \((i)f_\varphi^k\) — the contribution of the polarization moment to the intensity:

\[
C_0 \sum_{F_f} \frac{\|d_f\|^2}{3(2F_f+1)} (i)f_\varphi^k = (i)f_\varphi^k
\]

We can rewrite equation 2 in the form:

\[ I_{\text{orth}} = (4)f_0^0 + 2K_{F_f}f_\varphi\left[(4)f_0^2 \right] \]  \( (3a) \)

\[ I_{\text{par}} = (4)f_0^0 - K_{F_f}f_\varphi\left[(4)f_2^2 + \frac{\sqrt{5}}{2}(2)f_2^2 + (4)f_2^2 \right] \]  \( (3b) \)

\[ I_{\text{nomp}} = I_{\text{orth}} + I_{\text{par}} = (4)f_0^0 + \frac{K_{F_f}f_\varphi}{2}\left[(4)f_0^2 - \frac{\sqrt{5}}{4}(2)f_2^2 + (4)f_2^2 \right] \]  \( (3c) \)

Within the framework of this model, the influence is considered of the observation geometry on the magnitude and shape of the recorded coherence resonances at the 404 nm transition. \( I_{\text{orth}} \) is described by the sum of the nonlinear population resonances and the longitudinal alignment (equation 3a), and \( I_{\text{par}} \), by their difference. In addition, the transverse alignment resonance is superimposed on the signal \( I_{\text{par}} \), which can also modify its amplitude and width (figure 3).

The coherence resonances created at the 404 nm transition and observed in the 770 nm fluorescence can also be interpreted within the framework of this description (equations 3a, 3b, 3c). As a result of the multi-level spontaneous transfer, they broaden and, moreover, the transverse alignment is destroyed faster. Considering only the first two terms in equations 3a, 3b, we can explain the small differences between \( I_{\text{orth}} \) and \( I_{\text{par}} \) observed in figures 5a and 5b.

4. Conclusions

The 404.4 nm excitation of K induces coherent superposition of the ground state Zeeman sublevels at \( B = 0 \), which can be measured by the direct 404.4 nm fluorescence as a function of the magnetic field. The atomic polarization formed is also transferred to the atomic levels of the first resonance line. This transfer is evidenced by the registration of a narrow MO resonance in the 770 nm fluorescence from the \( 4p^5P_{1/2} \), which is of opposite sign, for the case of hot K vapor. It should also be noted that even after multiple cascade transitions, the resonance observed on the infrared lines has a much better S/N ratio compared to that on the excited second resonance line. Therefore, the study of these sub-Doppler-width MO resonances in hot K vapor will be useful for the further development of the high-resolution MO laser spectroscopy.

The performed experimental investigations and evaluations of the MO resonances are needed to show the influence of the processes of excitation, spontaneous transfer, coherence and reabsorption on the shape of the registered MO resonances at different geometries of registration.

Acknowledgments

This work was supported by the National Science Fund of Bulgaria under Contract DO08-19/2016 “New coherent and cooperative effects in hot alkali vapor”, as well as Goszadanie: #3.4903.2017/6.7, AAAA-A17-117052210003-4”. S.T. acknowledges the “Post-Doctoral Students” National Program RMS no 271/2019.
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