Measurement of cesium resonance line self-broadening and shift with doppler-free selective reflection spectroscopy

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The density dependence of the line width and resonance frequency of the cesium D_1 line is studied with Doppler-free frequency modulated selective reflection spectroscopy. The experimental coefficient of self-broadening $\gamma_{col}/N = 2\pi \times 7.5(1.1) \times 10^{-8}$ cm^3 Hz is in good agreement with theoretical predictions. Also is observed a density-dependent shift with the coefficient $\Delta\nu_{col}/N = -0.9(5) \times 10^{-8}$ cm^3 Hz. In the limit $N=0$ a frequency shift of $-3.0(4)$ MHz remains that may be attributed to long-range atom–surface interactions. The experimental results can be used in measurements of the local field-induced frequency shift at high atomic densities.

1. Introduction

The optical properties of dense atomic vapors have received notable attention in recent years, partly motivated by possible applications in quantum electronics, such as intrinsic optical bistability [1].

In the present work we make use of Doppler-free frequency modulated selective reflection spectroscopy (FM-SR) to investigate how the homogeneous line width of the cesium D_1 line depends on the atomic density. We also analyse the density dependence of the frequency shift relative to an unperturbed transition obtained by saturation spectroscopy in a low density cesium cell. As will be shown below, we find a linear dependence both for the collisional broadening γ_{col} and for the frequency shift $\Delta\omega_{col} = 0.12 \, \gamma_{col}$. A small red shift of approximately 3 MHz, which is observable for very low densities, is characteristic for selective reflection spectroscopy, since this technique allows the monitoring of an interaction between the atoms and the reflecting surface.

In dense atomic vapors the interaction between identical atoms may modify the resonance frequency

and the line width of optical transitions. The dominant effect is brought about by collision-induced exchange of excitation due to a resonant dipole–dipole interaction between an excited and an unexcited atom, giving rise to a large effective cross section [2–4]. While the relaxation of optical coherence of an excited atom in a collision causes a homogeneous line broadening (self-broadening or resonance broadening), the transfer of optical coherence from an excited to a ground state atom can lead to a shift of the resonance line [5]. The shift-to-width ratio depends on the angular momenta of the ground state and the excited state. Besides the second order dipole–dipole interactions there are also asymmetries in the $\pm C_3 R^{-3}$ potentials of the resonance states that should contribute to the shift [6,7]. Other mechanisms are expected to produce a density dependent line shift, too, e.g. in addition to the van der Waals interaction between two separated atoms, the interaction energy connected with the overlap of the atomic wave functions in "hard" collisions is supposed to make a contribution [8].

For not too high densities, i.e. when the probability for three-body collisions is still small, the dominant term for self-broadening and for the different line shifting mechanisms is linear in the atomic density N. Hence the self-broadening can be written as

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$\gamma_{col}/2\pi = kN$, where k is the self-broadening coefficient, and the different collision-induced line shifts can be summed up to $\Delta\nu_{col} = \alpha N$, where α is some constant.

In addition to the line shifts mentioned above, there is another contribution to the shift that is connected with the local field correction. In a dense atomic vapor illuminated by a light field, an atom is not only subject to the incident field in vacuum, but also to the radiation generated by all other atoms in the sample. An approximate description of this situation is possible with the concept of the local field, defined as the effective field that acts on a typical atom in the vapor. The local field being different from the incident field, a modification of the dielectric constant ensues, that gives rise to a line shirt proportional to the density of resonant atoms (Lorentz shift, [8]). Hence if a vapor with predominant inhomogeneous broadening is illuminated by a narrow-band laser source, only a homogeneously broadened fraction of atoms contributes to the local field. In that case the effective atomic density that enters the expression for the Lorentz shift is equal to N reduced by the ratio of homogeneous (γ) to Doppler broadening ($\Delta\omega_D$), $\Delta\nu_{loc} = \beta(\gamma/\Delta\omega_D)N$, where β is the Lorentz shift coefficient.

Recently the density dependence of the line shift has been studied in high density potassium vapor ($N = 10^{17}$ cm^{-3}), where the homogeneous width exceeds the Doppler width [8]. Under these conditions the Lorentz shift is proportional to the total atomic density and cannot be distinguished experimentally from the collisional shift in the regime of linear interaction of light field and sample. At moderate densities however, where the Doppler broadening is larger than the homogeneous broadening, the difference in the density dependences of $\Delta\nu_{col}$ and $\Delta\nu_{loc}$ may be exploited for the experimental separation of Lorentz and collision-induced shift, since γ itself is a function of N.

2. Method and line shape

In dense atomic vapors, only the wings of a resonance line can be studied with conventional absorption laser spectroscopy because the vapor is essentially opaque and reabsorption of the fluorescence light strongly distorts the line center. However it is possible to analyse the line shape in the central region by means of the resonant reflection of light at a dielectric–atomic vapor interface (selective reflection, SR). In the vicinity of an atomic absorption line the reflectivity R of the interface is governed by the Doppler-broadened, anomalous dispersion of the vapor. At gas pressures for which the homogeneous width is less than the Doppler width, however, an additional sub-Doppler contour appears in the SR spectrum [9]. Its origin is due to depolarizing collisions of gas atoms with the window of the cell, which quenches the contributions of atoms with velocity components parallel to the laser beam.

The Doppler broadened background of the spectrum can be eliminated if, in addition, the laser frequency is modulated and the signal recorded with a phase-sensitive detection scheme (FM-SR) [10]. The frequency dependence of the reflectivity transforms the frequency modulation of the incident light into amplitude modulation of the reflected light, with a modulation depth proportional to the frequency derivative of the reflectivity $dR/d\omega$. Since the Doppler broadened part of the spectrum varies much less with frequency than the sub-Doppler contour, the signal $dR/d\omega$ is practically Doppler-free.

In the limiting case of weak incident field, where saturation of the atomic transition may be neglected, theory predicts, to lowest order in the detuning $\Delta\omega \ll \Delta\omega_D$, a dispersive lorentzian, $dR/d\omega \propto \Delta\omega/(\Delta\omega^2 + \Gamma^2)$, with Γ equal to the homogeneous line width γ [10,11]. However, the observed line shapes differ from the antisymmetric dispersive lorentzian shape. Recently Ducloy and Fichet have modelled these distortions theoretically by taking into account induced long-range atom–surface interactions, which leads to shifted, asymmetric lines [12]. They characterize both shift and line asymmetry by a single, dimensionless coupling constant A that measures the surface-induced frequency shift, in units of the dipole decay rate $\gamma/2$, for an atom at the distance of a reduced wavelength $\lambda/2\pi$ from the interface.

3. Experimental setup

A schematic diagram of the experimental arrangement is shown in fig. 1. We use a tunable single-mode
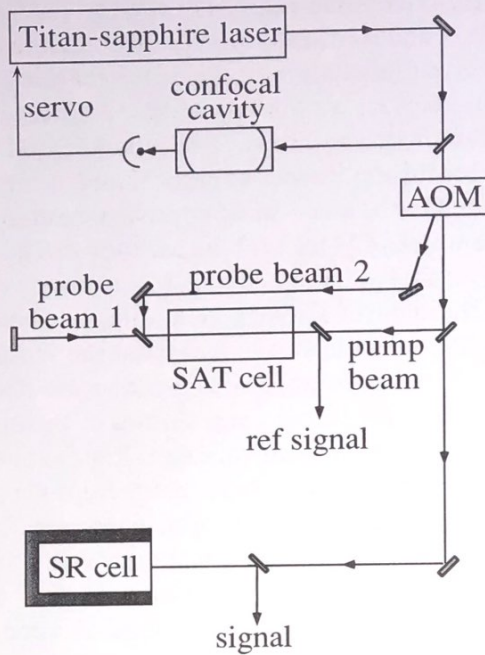
Fig. 1. Schematic diagram of the experimental setup. AOM is an acousto-optic modulator, SAT cell is a cesium cell at room temperature, SR cell is a heated cesium cell.

titan–sapphire laser that is frequency-stabilized onto the resonance of an external confocal reference cavity. The laser line width of less than 1 MHz is sufficient to easily resolve the natural line width of the atomic transition. We modulate the laser frequency with 1 kHz.

The cell used for SR is made of pyrex and has a wedged window to avoid interference from the inner and outer surfaces. We use the cell near normal incidence (angle <2 mrad), and the diffraction angle of the beam does not exceed 1 mrad. To avoid nonlinear effects like optical pumping, we work with low beam intensity (<200 μW/cm^2). The atomic density inside the cell is calculated from the temperature of the coldest spot of the cell by means of the Langmuir–Taylor relation [13]. This temperature is measured with a commercial platinum resistance temperature detector (Pt 100 class A). The accuracy is limited by the digital display and by the exact position of the detector. The total uncertainty in the number density near the cell window, due both to errors in temperature determination and possible inaccuracies of the Langmuir–Taylor relation, is estimated to be 10%. For temperatures between 90°C

and 170°C we obtain atom concentrations between 10^{13} cm^{-3} and 3×10^{14} cm^{-3}.

The intensity of the light reflected from the glass–cesium vapor interface is detected with a photodiode and a lock-in amplifier. To obtain a frequency reference, we simultaneously record a saturation spectrum in a second cesium cell at room temperature. For frequency calibration we use two probe beams that are frequency shifted relative to each other by means of an acousto-optic modulator (AOM). Thus we obtain two identical saturation lines separated by half of the known AOM frequency.

4. Results

In the present work we study the self-broadening and frequency shift of the cesium D_1 line (wavelength $\lambda=894$ nm, natural width $\gamma_r/2\pi=5.3$ MHz). Since the hyperfine splittings of the $6S_{1/2}$ ground state and the $6P_{1/2}$ excited state exceed the Doppler width ($\Delta\nu_D\approx0.4$ GHz), the hyperfine components of the $6S_{1/2}(F=3, 4)\rightarrow6P_{1/2}(F=3, 4)$ transition appear as isolated lines. Here we concentrate only on the $(F=4)\rightarrow(F=3)$ hyperfine transition. Random samples of other hyperfine components show essentially the same behaviour.

A typical Doppler-free SR resonance recorded at low density ($N=10^{13}$ cm^{-3}, collision broadening $\gamma_{col}<\gamma_r$) is presented in fig. 2. We define the width Γ of the SR resonance by setting $\Gamma/2\pi$ equal to the spectral interval between the minimum and the maximum. A slight asymmetry is visible in the spec-
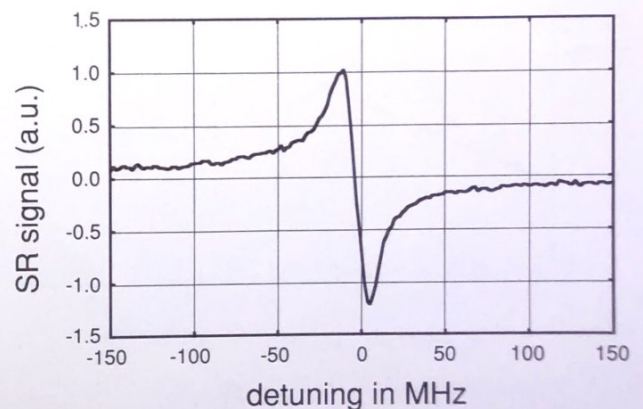

Fig. 2. Typical FMSR spectrum recorded at $N=10^{13}$ cm^{-3}.

trum: the blue wing is narrower and higher than the red wing. The resonance frequency of the unperturbed line has been obtained from a saturation spectrum recorded simultaneously at very low cesium density. Comparing our experimental line shapes with the shapes calculated by Ducloy and Fichet [12] for different values of the coupling constant A, we achieve good agreement, e.g. $A=0.05$ was estimated for $\Gamma=15$ MHz. Moreover, we find that the difference between Γ and the actual homogeneous line width γ, as used in the calculations of the theoretical line shapes, is less than 4%, which is below our measurement precision. In the following we do not distinguish between Γ and γ.

The density dependence of the line width Γ is shown in fig. 3. The experimental data are well approximated by a straight line, $\Gamma/2\pi=\Gamma_0/2\pi+kN$, with the slope $k=(7.5\pm0.3)\times10^{-8}$ cm^3 Hz and $\Gamma_0/2\pi=10.5$ MHz. The difference between Γ_0 and the natural linewidth γ_r may be attributed to residual buffer gas (N_2) in the cell. The constant k is equal to the self-broadening coefficient $(d\gamma/dN)/2\pi=(\gamma_{col}/N)/2\pi$, with a possible systematic error of 10% due to the uncertainty in the determination of the atomic density near the cell window.

Literature values of the experimental and calculated selfbroadening coefficients are presented in table 1. The results marked [b] and [c] have been derived from line wing measurements. The present value agrees with these results by Niemax et al. [14] and by Chen and Phelps [15] within the limits of un-

Fig. 3. Self-broadening of the $(F=4)\rightarrow(F=3)$ hyperfine component of the cesium $6S_{1/2}\rightarrow6P_{1/2}$ line as a function of the atomic density N.

certainty. The value marked [a] for the D$_2$ line has been obtained by FMSR with a semiconductor laser. The ratio of this datum to our value for the D$_1$ line is $k[D_2]/k[D_1]=1.5\pm0.4$, which is in very good agreement with the ratio 1.4 in refs. [3] and [16], calculated in the impact approach, and in ref. [7], calculated in the quasi-static approach, but the value 1.9 from ref. [2] cannot be excluded. The ratio $k[D_2]/k[D_1]=1.5\pm0.4$ also agrees well with the ratio of the data of Niemax et al. and of Chen and Phelps. These groups have measured the D$_1$ and D$_2$ lines with the same apparatus, so the ratio does not include the rather large uncertainties of the number densities, which amount to about 20% for the data of Niemax et al. We conclude that there is very good agreement between the self-broadening coefficients derived from measurements in the line wings and the coefficients as measured at the line center.

The table also presents the results of some theoretical calculations. The datum [d] (Movre and Pichler [6,7,14]) is based on the quasi-static approach, while the data [e] (Vdovin and Dobrodeev [3]), [f] (Byron and Foley [16]) and [g] (Ali and Griem [2]) have been calculated using the impact approach. The present value agrees well with all of these theoretical results for the resonance broadening.

Furthermore we examined the frequency shift of the SR line center, taken as the average of the maximum and the minimum, $\nu_{centre}=(\nu_{min}+\nu_{max})/2$, and measured relative to the saturation line center. In fig. 4 the observed shift $\Delta\nu_{SR}$ is plotted against the atomic density N. Since the contribution of the Lorentz shift is reduced by the ratio of homogeneous to Doppler width, the Lorentz shift is negligible compared to the collisional shift in the density region studied here ($\gamma/\Delta\nu_D<0.1$). As expected for the collision-induced shift, we find good agreement with a linear function, $\Delta\nu_{SR}=\Delta\nu_0+\alpha N$, where $\Delta\nu_0=-(3.0\pm0.4)$ MHz accounts for the shift that remains for $N\rightarrow0$ and $\alpha=-(0.9\pm0.4)\times10^{-8}$ cm^3 Hz is the coefficient for the collisional shift. For the cesium D$_2$ line in the moderate density region ($N\leq10^{15}$ cm^{-3}), Chevrollier et al. have observed a density-dependent shift, that did not vary systematically and that was different for different hyperfine components [17]. The order of magnitude of the shift coefficient is the same as the one observed here. For the potassium resonance lines in the high density region
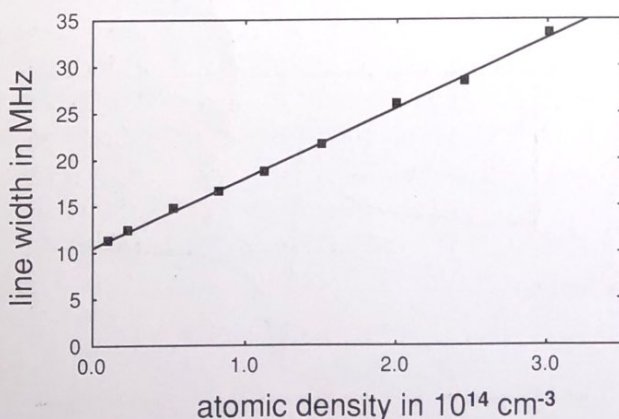
Table 1
Self-broadening coefficient $(\gamma_{col}/N)/2\pi$ for the cesium D_1 and D_2 resonance lines.

	D_1 line in 10^{-8} cm³ Hz	D_2 line in 10^{-8} cm³ Hz	Ratio $D_2:D_1$
Experiment (SR) [central region]	7.5±1.1 present	11.5±2.3 [a]	1.5±0.4
Experiment (abs.) [line wings]	8.4±2.5 [b] 8.6±1.3 [c]	12.6±3.8 [b] 15.1±2.3 [c]	1.5±0.2 1.8±0.3
Quasistat. theory	9.7 [d]	13.5 [d]	1.4
Impact theory	8.5 [e]	11.5 [e]	1.4
Impact theory	7.1 [f]	13.8 [f]	1.9
Impact theory	8.1 [g]	11.2 [g]	1.4

[a] Akulshin et al. (1982). [b] Niemax et al. (1979). [c] Chen and Phelps (1968). [d] Movre and Pichler (1979). [e] Vdovin and Dobrodeev (1969). [f] Byron and Foley (1964). [g] Ali and Griem (1966).
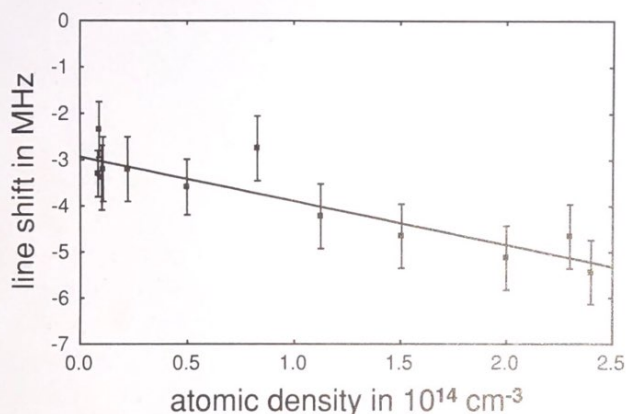

Fig. 4. Line shift of the $(F=4) \rightarrow (F=3)$ component relative to a saturation line as a function of the atomic density N.

$(N \approx 10^{17}$ cm$^{-3})$, a linear dependence of the shift on the density has recently been reported by Maki et al. [8]. At these densities the line is homogeneously broadened and only the sum of the collisional and Lorentz shift coefficients can be measured in the linear regime of interaction of light field and sample. However, the combination with experiments in the moderate density regime, where the Doppler broadening exceeds the homogeneous broadening, may separately yield the collisional and Lorentz contributions.

The red shift of approximately 3 MHz that remains for $N \rightarrow 0$ has already been observed for the rubidium D_1 line [18] and the cesium D_2 line [17]. We also note that a strong shift of the SR resonance was observed for the principal cesium line ($\lambda = 455$ nm) [17]. These shifts have been explained by long-range atom–surface interactions. Consistent with this interpretation is the fact that we have found no change in the red shift $\Delta\nu_0$ for intensities between 0.1 mW/cm² and 50 mW/cm².

5. Conclusion

FMSR spectroscopy has proved a powerful tool for analyzing the optical properties of dense atomic vapors. We have measured the self-broadening coefficient for the cesium D_1 line. Our findings are in good agreement with theoretical predictions and confirm previous measurements in the atomic line wings. Furthermore we have discovered a small density-dependent shift of the resonance line which is difficult to measure with absorptive techniques. It can be useful for the determination of the local field-induced shift at high atomic densities. In this work only the regime of linear interaction between atoms and light field has been studied. The extension to higher intensities should yield some interesting insight on optical pumping and hyperfine ground state relaxation processes.

Acknowledgements

References

[1] E.A. Hopf, C.W. Bowden and W.H. Luisell, Phys. Rev. A 29 (1984) 2591.

[2] A.W. Ali and H.R. Griem, Phys. Rev. 140 (1965) A1044.

[3] Yu.A. Vdovin and N.A. Dobrodeev, Sov. Phys. JETP 28 (1969) 544.

[4] E.L. Lewis, Phys. Rep. 58 (1980) 1.

[5] J. Cooper and D.N. Stacey, Phys. Lett. A 46 (1973) 299.

[6] M. Movre and G. Pichler, J. Phys. B 10 (1977) 2631.

[7] M. Movre and G. Pichler, J. Phys. B 13 (1980) 697.

[8] J.J. Maki, M.S. Malcuit, J.E. Sipe and R.W. Boyd, Phys. Rev. Lett. 67 (1991) 973.

[9] J.P. Woerdman and M.F.H. Schuurmans, Optics Comm. 14 (1975) 248.

[10] A.M. Akulshin, V.L. Velichanskii, A.S. Zibrov, V.V. Nikitin, V.A. Sautenkov, E.K. Yurkin, N.V. Senkov, Sov. Phys. JETP Lett. 36 (1982) 303.

[11] M.F.H. Schuurmans, J. Phys. (Paris) 37 (1976) 469.

[12] M. Ducloy and M. Fichet, J. Phys. II (Paris) 1 (1991) 1429.

[13] J.B. Taylor and I. Langmuir, Phys. Rev. 51 (1937) 753.

[14] K. Niemax, M. Movre and G. Pichler, J. Phys. B 12 (1979) 3503.

[15] C.L. Chen and A.V. Phelps, Phys. Rev 173 (1968) 62.

[16] F.W. Byron and H.M. Foley, Phys. Rev. 134 (1964) A625.

[17] M. Chevrollier, M. Fichet, M. Oria, G. Rahmat, D. Bloch and M. Ducloy, J. Phys. II (Paris) 2 (1992) 631.

[18] A.M. Akulshin, A.A. Celikov, V.A. Sautenkov, T.A. Vartanjan and V.L. Velichanskii, Optics Comm. 85 (1991) 21.