Frequency-modulation saturation spectroscopy of molecular iodine hyperfine structure near 640 nm with a diode laser source

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In a frequency-modulation spectroscopy experiment, using the radiation from a single frequency diode laser, the spectra of molecular iodine hyperfine structure near 640 nm were recorded on the transition \( B^3\Pi_{u} - X^1\Sigma_{g}^+ \). The frequency reference given by the value of the modulation frequency (12.5 MHz in given experiment) allows determination of the frequency differences between hyperfine components with accuracy better than 0.1 MHz using the fitting procedure in experiment with only one laser.

I. INTRODUCTION

The dense spectrum of molecular iodine is widely used for optical wavelength reference in for laser spectroscopic applications and laser frequency stabilization in a wide region of the optical spectrum from the green (500 nm) to the near infrared (900 nm). High frequency stability of He-Ne laser at 543 nm, 612 nm, 633 nm, 640 nm, Nd:YAG laser at 532 nm (second harmonic), Ar\(^+\) laser at 514.5 nm is achieved by use of sub-Doppler techniques such as saturation spectroscopy for locking to an iodine molecular transitions. Seven of the 20 recommended wavelengths for the realization of the metre are based on the hyperfine transitions wavelengths of \( ^{127}\text{I}_2 \) [1–2].

The precise frequency locking onto the iodine hyperfine transitions is used now for the determination of the nuclear electric quadrupole interaction and the nuclear spin-rotation interaction parameters of iodine molecules. The \( ^{129}\text{I}_2 \) and \( ^{127}\text{I}^{129}\text{I} \) molecules are the attracting objects for these investigations. The very promising set of iodine transitions for laser frequency stabilization includes the wavelengths 502 nm (\( ^{127}\text{I}_2 \) R(51) 68-0), 633 nm (\( ^{127}\text{I}_2 \) R(33) 6-3), 793 nm (\( ^{127}\text{I}_2 \) R(92) 0-15). The additional references could be created with transitions of \( ^{127}\text{I}^{129}\text{I} \) molecules.

The standard approach to the experiments with saturated absorption resonances is based on the use of two identical lasers stabilised to different hyperfine structure components and on the measurements of the beating frequencies of two laser radiations. In this work, we report on the Doppler-free saturation spectroscopy of the molecular iodine hyperfine lines at 640 nm using much simpler experimental setup. We have used so-called frequency-modulation spectroscopy [3-4] for which the laser field is phase modulated at a frequency higher than the resonance linewidth. When the mean laser frequency is close to some hyperfine component due to saturation of the absorption and dispersion the phase modulation is transformed to amplitude modulation. The amplitude modulation is detected by lock-in amplifier and the output signal consists of the saturated absorption or saturated dispersion depending on the reference oscillator phase. The important feature which we use in our approach is that the FM-resonances are a sum of individual resonances shifted on half of the modulation frequency what gives the frequency reference. The frequency reference given by the value of the modulation frequency (12.5 MHz in given experiment) allows determination of the frequency differences between hyperfine components with accuracy better than 0.1 MHz using the fitting procedure in experiment with only one laser.

II. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. The single frequency diode laser (Toptica Model DL100) emitting 640-nm radiation with 40-mW output power has been used. Within the frequency tuning range (more than 5 nm) of the laser more than 1000 strong rovibrionic lines of the \( ^{127}\text{I}_2, ^{129}\text{I}_2 \) and \( ^{127}\text{I}^{129}\text{I} \) molecules can be observed.

To reduce the laser-frequency drift induced by temperature changes the laser temperature was stabilized with...

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accuracy about 0.01 C with laser drift better than 10 MHz per hour. A controllable frequency tuning without mode jumps was achieved in range about 20 GHz by means of the electronic module DigiLock with use of a piezoelectric actuator mounted on the external laser mirror. The probe beam of 7 mW and the pump beam of 16 mW with beam diameter of 2 mm counterpropagate inside a 8-cm-long iodine cell. The iodine vapor pressure was kept constant by keeping the cold-finger temperature within 0.001 C. Phase modulation of the laser output radiation is produced by a laser diode injection current modulation at the frequency 12.5 MHz. This frequency is higher than the iodine resonance linewidth (about 1-2 MHz), it is much higher than the characteristic technical noise frequencies and it is comparable with with frequency differences between hyperfine components. The photomultiplier PMT detects the iodine fluorescence, the photodiode PD detects the probe beam power. The spectral control of the laser output provide the scanning Fabry-Perot interferometer (FPI) with finesse about 250 and FSR 2 GHz, the spectrometer USB2000 with resolution 0.5 nm. The absolute wavelength of the laser can be determined by help of optogalvanic effects in the Ne filled lamp with hollow cathode. In the described experiments we have used the optogalvanic resonance with the 0.6403 nm Ne line. The signal from the output of the numerical lock-in amplifier built in DigiLock was processed by the PC.

III. EXPERIMENTAL RESULTS

Here we present some preliminary results of study of the hyperfine structure of the $^{127}\text{I}^{129}\text{I}$ line closed to the 0.6403 nm Ne line. The figure 2 shows an example of the hyperfine structure obtained with FM spectroscopy technique (resonances of saturated absorption). The $^{129}\text{I}_2$ P(90) 5-3 and $^{127}\text{I}_2$ P(10) 8-5 line contributes in the displayed spectrum. The figure 3 shows an example of approximation of the two hyperfine line by theoretical formula describing FM spectroscopy. The fitting are very close to the experimental data. As one can see from Fig.3c, the frequency difference of the displayed components is 30.8 MHz.

In a frequency-modulation spectroscopy experiment, using the radiation from a single frequency diode laser, the spectra of molecular iodine hyperfine structure near 640 nm were recorded on the transition $B^3\Pi_{u^+} - X^1\Sigma^+$. The frequency reference given by the value of the modulation frequency (12.5 MHz in given experiment) allows determination of the frequency differences between hyperfine components with accuracy better than 0.1 MHz using the fitting procedure in experiment with only one laser.

IV. CONCLUSION

In a frequency-modulation spectroscopy experiment, using the radiation from a single frequency diode laser, the spectra of molecular iodine hyperfine structure near 640 nm were recorded on the transition $B^3\Pi_{u^+} - X^1\Sigma^+$. The frequency reference given by the value of the modulation frequency (12.5 MHz in given experiment) allows determination of the frequency differences between hyperfine components with accuracy better than 0.1 MHz using the fitting procedure in experiment with only one laser.

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