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Sub-Doppler spectroscopy of molecular iodine around 541 nm with a novel solid state laser source

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Abstract

We report high resolution sub-Doppler spectroscopy of molecular iodine hyperfine transitions around 541 nm. A novel solid state source, based on a diode laser, a Yb^+ fiber amplifier and a periodically-poled $KTiOPO_4$ crystal, was developed, having a continuous tunability range of 1.2 THz. The performance of this crystal for doubling 1083 nm laser radiation is demonstrated for the first time, as well as the use of a broad band Yb-doped fiber amplifier for Doppler-free spectroscopy. Preliminary results for the frequency stability of the 1083 nm diode laser locked to an iodine hyperfine line are reported. This source can be used as a compact frequency standard for precision measurements of the atomic helium and determination of the fine structure constant α. © 2000 Elsevier Science B.V. All rights reserved.

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Electronic transitions of the iodine (I_2) molecule are well known frequency standards in the visible range and have been successfully used to stabilise laser sources as, for instance, frequency-doubled cw Nd–YAG sources at 1.064 μm at the level of a few parts in 10^{-14} in 1 s [1,2].

In this near-infrared spectral region, highly stable sources are required for the measurement of the 2^3P_0–2^3P_1 energy splitting for atomic helium, for which a few kilohertz accuracy was recently achieved, by using diode laser sources at 1083 nm [3], in view of an accurate determination of the fine structure constant α.

One of the limiting factors to improve the accuracy of these frequency measurements is the lack of accurate and stable frequency standards in the near infrared region. In the 1083 nm region, we have previously proposed sub-Doppler Cs_2 transitions [4] or sub-Doppler helium transitions [5]. However, a frequency stability better than a few parts in 10^{-11} in 1 s cannot be reached, due to the broad linewidth of the reference signals, and to the long-term depen-

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dence of the transition frequency on the environmental conditions of both references. To exploit the better stability of an iodine-based reference, we characterised frequency doubling of a fibre-amplified Distributed Bragg Reflector (DBR) laser at 1083 nm using birefringent phase-matching in a semi-monolithic MgO:$LiNbO_3$ crystal mounted in an enhancement cavity [6].

In the present paper we report, for the first time to our knowledge, saturated-absorption spectroscopy of

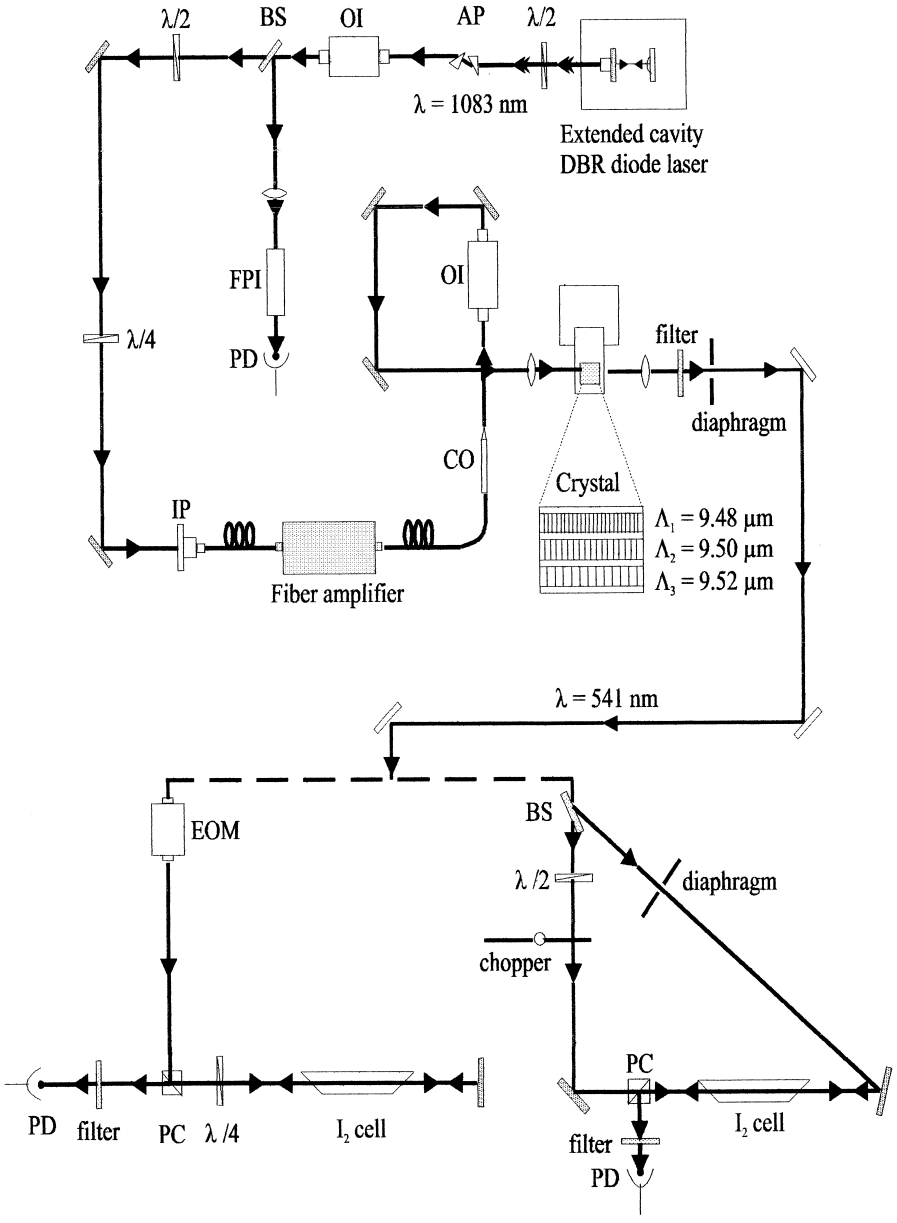

Fig. 1. Experimental set-up for 541 nm laser source and saturation spectroscopy of hyperfine I_2 components. (a) Amplitude modulation experiment. (b) Low frequency modulation experiment. AP: anamorphic prims, OI: optical isolator, BS: beam splitter, IP: input port, CO: collimator, PC: polarising cube, PD: photodiode.

I_2 hyperfine components around 541 nm, using a novel tunable laser source. The set-up includes an Yb^+-doped fiber amplifier with a maximum output power of about 1.2 W for 1083 nm radiation and an efficient periodically-poled $KTiOPO_4$ (PP-KTP) frequency doubler [7]. These components provide sufficiently high second harmonic power for saturation spectroscopy by single-pass frequency doubling, thus avoiding any complication due to enhancement cavities. Preliminary results for frequency locking of the frequency-doubled DBR laser on I_2 hyperfine components are reported, too.

The experimental layout is shown in Fig. 1. The DBR diode laser at 1083 nm was mounted in a 100 mm long extended-cavity configuration [5], with a 50% transmitting output mirror. In this way the laser linewidth was narrowed down to 300 kHz and the frequency of the laser emission could be controlled by changing the cavity length with a piezoelectric transducer (PZT), on which the output mirror was glued. The laser light was amplified using a Yb^+-doped fiber amplifier (IPG model YAM-1000). The output power exceeded 1 W with an injected saturation power of about 1 mW. A quarter wave-plate ($\lambda/4$) and a half-waveplate ($\lambda/2$) mounted before the fiber injection input were used, in order to obtain at the fiber output a linear polarisation parallel to the z direction of the nonlinear crystal. A study of the amplified laser spectrum showed that no significant frequency broadening is added, which means that the amplified radiation can be used in high resolution spectroscopy without any further frequency control of the fiber amplified light [8]. In the same reference, it is found that residual amplitude modulation (RAM) of the laser injected in the amplifier should be carefully taken into account, because it can induce very strong amplitude noise in the amplified radiation. Since this noise severely limits the use of these amplifiers with frequency modulated diode lasers, we directly phase-modulated green radiation by an electro–optic modulator (EOM in Fig. 1).

The 10 mm long and 0.5 mm thick flux-grown KTP crystal was poled at three different grating periods by the technique of low-temperature electric-field poling [9]. The poling period was calculated using the Sellmeier equations of Fan et al. [10]. Each grating period was 2 mm wide. The crystal was held inside a small copper holder whose temperature was controlled using a thermo-electric cooler. The infrared light coming out of the amplifier was focused into a spot size of 18 μm inside the crystal. Up to 6 mW were generated at the second harmonic frequency and the normalised internal conversion efficiency was about 0.84% W^{-1}. The optimal tem-

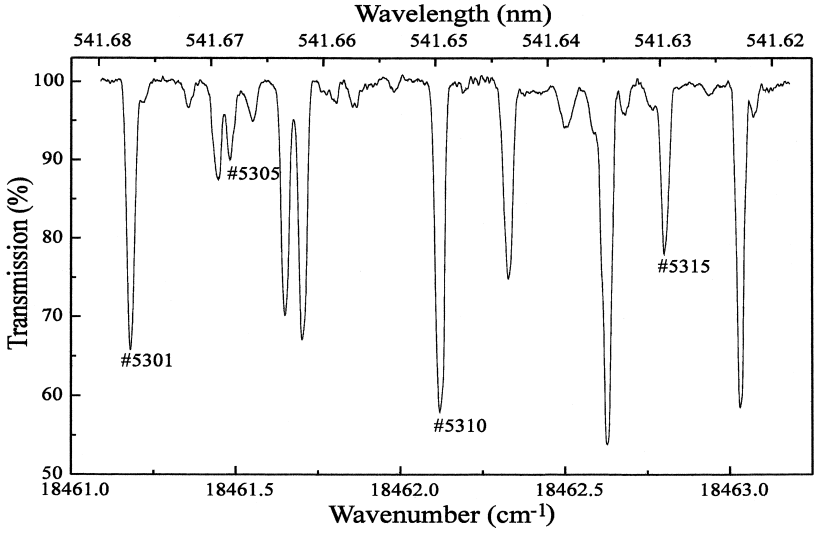

Fig. 2. Doppler-broadened absorption spectrum of I_2 lines around the second harmonic frequency of 1083 nm helium transitions. $L_{cell} = 8$ cm, $P_{iodine} = 50 \times 10^{-3}$ Torr.

peratures for quasi-phase-matched (QPM) frequency doubling using these gratings were near room temperature: for the 9.48 μm, 9.50 μm and 9.52 μm periods they were 47.1°C, 39.6°C and 29.7°C, respectively at $\lambda = 1083.272$ nm. The measured temperature acceptance bandwidth was 5°C. We have also experimentally determined the wavelength acceptance bandwidth to be 0.4 nm. Furthermore, the optimal wavelength could be tuned by changing the crystal temperature with a slope of 82 pm/K. A similar temperature tuning coefficient in PP-KTP was also measured at 1064 nm [7].

The second harmonic light was sent into a 8 cm long $^{127}I_2$ cell [1]. The vapor pressure in the cell was controlled by cooling the tip of the cell using a thermo-electrically cooled holder. Fig. 2 shows a 30 GHz (corresponding to 60 GHz in the green) wide scan, encompassing several Doppler-broadened lines of I_2. In this case, the DBR was operated without the external mirror, and the frequency was scanned by varying the bias current. Moreover, green radiation can be generated in a wide frequency range around 541.5 nm by controlling laser diode output wavelength and PP-KTP crystal temperature. In fact, to characterise the doubling crystal operation, we have generated up to 6 mW of green radiation in a 1.2 THz frequency range by tuning the laser wavelength between 1081.8 and 1084.2 nm with a QPM slope of 82 pm/K between 30 and 46.6°C. This green tunability number should be compared with the limited tunability range of other solid state sources like doubled Nd–YAG (about 60 GHz) [11]. Table 1 shows the line numbers and the measured frequencies, as taken from the I_2 atlas [12]. In addition, we have assigned the vibrational and rotational values of the observed transitions and calculated their frequencies using the I_2 vibrational bandhead energies and rotational constants [13].

The hyperfine structure of some of the Doppler-broadened lines was observed by using sub-Doppler saturation spectroscopy. Two different experimental approaches were used: amplitude modulation spectroscopy (AMS) and low frequency modulation spectroscopy (LFMS). In the first case, the green beam was split in a couple of pump-probe beams in a proportion 25:1, resulting $I_{pump} = 2.5$ mW/mm^2 and $I_{probe} = 0.1$ mW/mm^2 for a green output power of 2.5 mW and without any focusing in the I_2 cell. The pump beam was amplitude modulated by a chopper at 1.46 kHz and the detected light of the probe beam was lock-in demodulated at this frequency. The observed hyperfine components, belonging to the R(34) 27-0 I_2 line, are shown in Fig. 3(a). For LFMS, an electro–optic modulator at 541 nm was used, driven by a modulation frequency chosen in the 85–120 kHz range. Pump and probe beams were spatially separated according to their orthogonal polarisation, by using a polarising cube (PC) in combination with a quarter-waveplate ($\lambda/4$). In Fig. 3(b) a first derivative spectrum is shown for the R(34) 27-0 I_2 line, recorded using LFMS. As in the case of AMS, all hyperfine components, within the Doppler-broadened profile, are fully resolved. With the LFMS set-up, the intensities of the pump-probe beams were $I_{pump} = 2.5$ mW/mm^2 and $I_{probe} = 1.8$ mW/mm^2 for the same conditions indicated above. The baseline variations in Fig. 3(b) are caused by the Doppler envelope of the transition. They can be significantly reduced with different techniques, as for example, by chopping the pump beam and detecting the FM demodulated probe beam with a lock-in amplifier [2]. The

Table 1
I_2 absorption lines near 541.6 nm

Line	Measured (cm^{-1}) [12]	Calculated (cm^{-1}) [13]	Assignment
5301	18461.1805	18461.1806	R(36)27-0
5302	18461.2168	18461.2190	P(154)32-0
5303	18461.3539	18461.3543	P(38)30-1
5304	18461.4409	18461.4407	P(125)30-0
5305	18461.4792	18461.4807	R(128)30-0
5306	18461.5491	18461.5460	R(41)30-1
5307	18461.6482	18461.6487	R(82)28-0
5308	18461.7032	18461.7039	P(32)27-0
5309	18461.7958	18461.7928	P(77)31-1
5310	18462.1232	18462.1199	R(35)27-0
5311	18462.3277	18462.3259	P(105)29-0
5312	18462.4977	18462.4934	P(37)30-1
5313	18462.6300	18462.6275	P(31)27-0
5314	18462.6806	18462.6810	R(40)30-1
5315	18462.8019	18462.8022	R(108)29-0
5316	18463.0315	18463.0317	R(34)27-0
5317	18463.0702	18463.0681	P(67)34-2
5318	18463.2342	18462.2348	P(78)28-0

[1] The I_2 cell is certified from the Bureau International des Poids et Mesures with reference number 26 B (5).

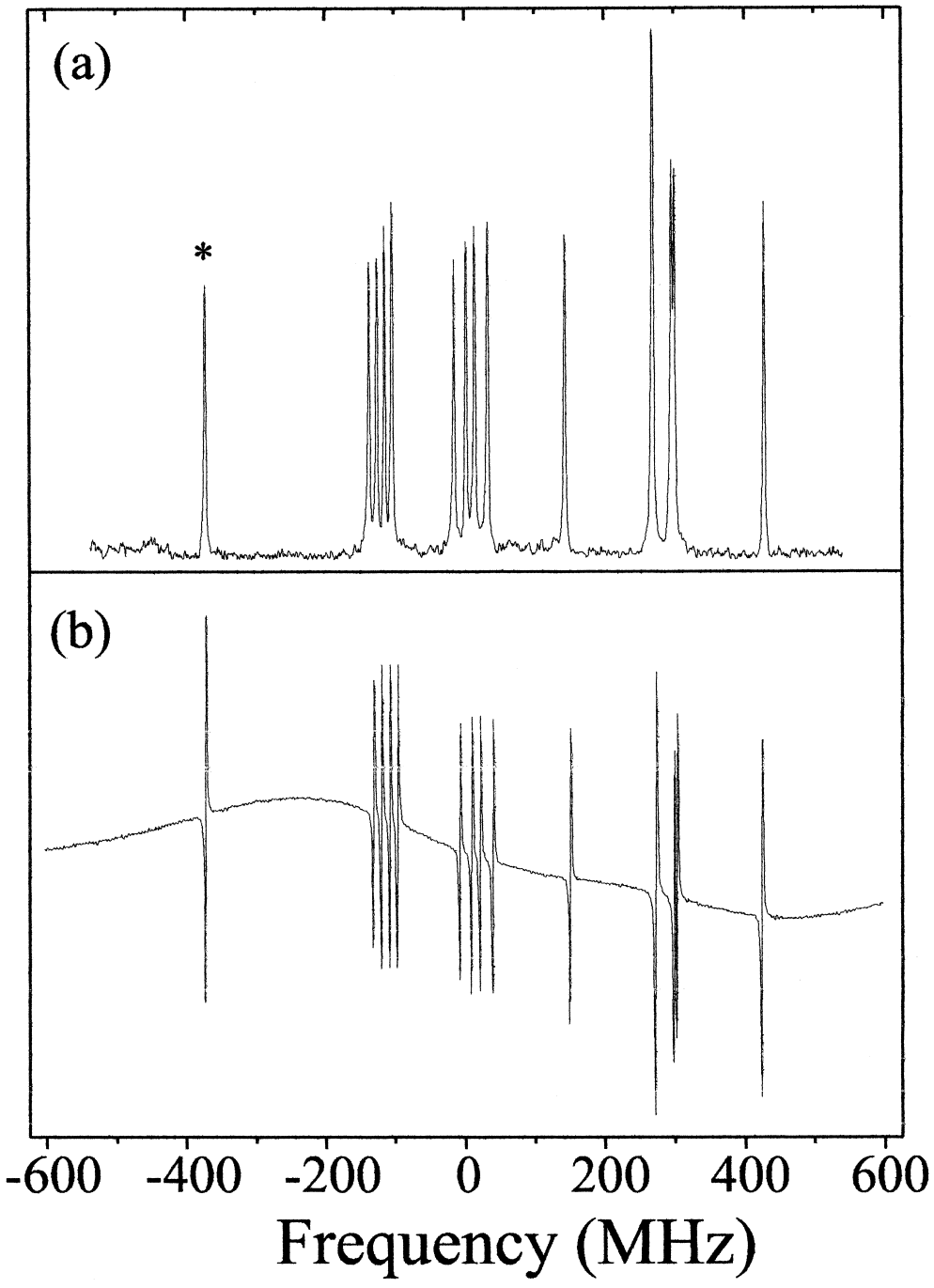

Fig. 3. Sub-Doppler saturation spectrum of the hyperfine components of the R(34)27-0 I_2 absorption line. (a) Amplitude modulation spectrum. (b) First derivative low frequency modulation spectrum. The large baseline modulation evident in recording (b) is due to the first derivative signal of the probe Doppler absorption. The number of points in both spectra is 2500.

linewidth of the more isolated component, in these spectra, (marked with * in Fig. 3) was 1.5 MHz for AMS and 1 MHz for LFMS, limited, for the latter case, by pressure broadening (50×10^{-3} Torr) and by the laser linewidth. The broader AMS linewidth is due to the little crossing-angle between pump and probe beams. This broadening can be reduced by choosing an appropriate optical set up. Finally, it is worth noting that the sampling rate of about 2 points/MHz used for the recordings in Fig. 3, underestimates the real S/N ratio.

The zero-crossing of the isolated line in Fig. 3(b) was used to lock the diode laser by feeding back the correction signal to the PZT transducer controlling the output mirror. The measured Allan variance, from the error signal at 1 s sampling in a 1000 s interval, allowed us to make a preliminary analysis of the laser frequency stability. It was 2 kHz in 1 s at 541 nm when the laser was locked with a lock-in amplifier time constant of 1 ms and S/N of about 900. It means that a 3.5×10^{-12} frequency stability for the 1083 nm diode laser is achieved, which is, at least, one order of magnitude better than previous results with other frequency references at this wavelength [4,5]. Since the Allan variance is estimated from the error signal, it does not take into account all the systematic sources for frequency instabilities. However, similar or even better levels of stability have been achieved with iodine-locked frequency references having comparable signal to noise ratio [2]. We plan to improve the stability figure by using frequency modulation spectroscopy or modulation transfer spectroscopy.

Summarising, we have observed Doppler-free hyperfine components of molecular iodine around 541 nm with a new laser source that combines three different laser optic technologies. That allows to build a very compact frequency standard, potentially extendible to a wide wavelength range. We are going to use these I_2 lines to stabilise the diode lasers at 1083 nm used for high precision spectroscopy. Moreover, using two lasers at 1083 nm simultaneously injected in the amplifier and hence doubled, measurements of the hyperfine constants in a new frequency interval and for a wide range of transitions having different rotational and vibrational numbers, made available thanks to the wide tunability of our set-up, can be done. Therefore, a more extended data base could be obtained, using very recent highly accurate measurements taken with limited tuning sources [14,15]. Single-pass doubling using efficient periodically-poled crystals can also be used to stabilise other near-infrared sources, and in particular the Nd:YAG laser.

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