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A Dense Grid of Reference Iodine Lines for Optical Frequency Calibration in the Range 571–596 nm

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A high precision dense grid of reference lines in the hyperfine structure of the B–X system of molecular iodine ($^{127}I_2$) is presented. A simple parametrization has been derived, predicting the hyperfine line positions of the ‘‘*t*’’ components for vibrational bands (13-1) up to (18-1) and rotational quantum numbers $J = (9–140)$. The analysis in this paper is based on Doppler-free saturation spectroscopy spectra used for calibration of more than 100 new components in the hyperfine structure of $^{127}I_2$. The data presented contains a prediction (with 2 MHz accuracy) for the positions of 1584 ‘‘*t*’’ components in the rotational structure of iodine, covering the wavelength interval 571–596 nm. © 1998 Academic Press

1. INTRODUCTION

In a large part of the optical spectrum, extending from the green to the near infrared (500–900 nm), the absorption spectrum of molecular iodine (*1–3*) has become a convenient reference standard for calibration purposes and is widely used in laser spectroscopic studies. Throughout the entire range this Doppler-broadened absorption spectrum (with an absolute accuracy of 60 MHz (*4*)) provides a suitable accuracy for studies in which pulsed dye lasers are employed. Particularly the fact that iodine has several lines per cm^{-1} makes this molecule such a useful standard. However, for precision measurements with cw lasers that have typical bandwidths of 1 MHz, the accuracy provided by the Doppler-broadened lines is insufficient. Even in precision experiments where pulsed-dye amplified lasers of 100-MHz bandwidth are used (*5–8*) the I_2-reference lines are the limiting factor to the accuracy.

For higher precision experiments the iodine molecule remains a good candidate for a wavelength standard. Individual hyperfine components of rotational lines measured by saturation spectroscopy provide a reliable and reproducible standard that is even used in the *mise en pratique* of the definition of the Metre (*9*). Over the years several of such lines were calibrated with sub-MHz accuracy, usually for specific purposes in precision experiments (*10–19*). Recently Sansonetti (*20*) reported the calibration of 102 I_2-hyperfine components in the yellow–red range of the spectrum with sub-MHz accuracy, thus providing a useful tool for calibration. Still this collection is insufficient for on-line calibration in many laser spectroscopic studies, mainly due to the large (up to 50 cm^{-1} in Ref. (*20*)) gaps between the lines. Most cw lasers allow for continuous scans of only one or few cm^{-1}; moreover the accuracy is lost when multiply overlapping scans are necessary to reach a calibrated line.

In the present study we present a grid of reference iodine ($^{127}I_2$) lines which is accurate (2 MHz absolute accuracy) and dense (at least one reference line in each cm^{-1}). The accuracy of this standard is dependent on previous absolute calibrations as well as on the unperturbed molecular structure of the B–X system of the $^{127}I_2$ molecule. The rotational structure and the hyperfine structure in the (13-1), (14-1), (15-1), (16-1), (17-1) and (18-1) bands of the B–X system is analyzed for $P(J)$ and $R(J)$ lines up to $J = 140$. Molecular parameters are derived that predict line positions of individual hyperfine components to within 2 MHz. If only a single hyperfine component per rotational line is counted, this procedure yields more than 1500 useful reference lines in a range of about 700 cm^{-1}, covering the wavelength range 571–596 nm. This standard represents more than an order of magnitude improvement over the accuracy of the I_2 atlas and should be useful in many studies where high precision is required.

2. EXPERIMENTAL

The experimental setup is schematically shown in Fig. 1. A stabilized cw ring-dye-laser (Spectra Physics 380D) running on Rhodamine 6G and continuously tunable over 1 cm^{-1} supplies the narrow bandwidth (∼1 MHz) radiation. The laser beam is split, as shown in Fig. 1, and about 10 mW is used for recording I_2-saturation spectra. Differential absorption is monitored on two photodiodes, where one of the probe beams is overlapped in the I_2 cell (∼10-cm length)

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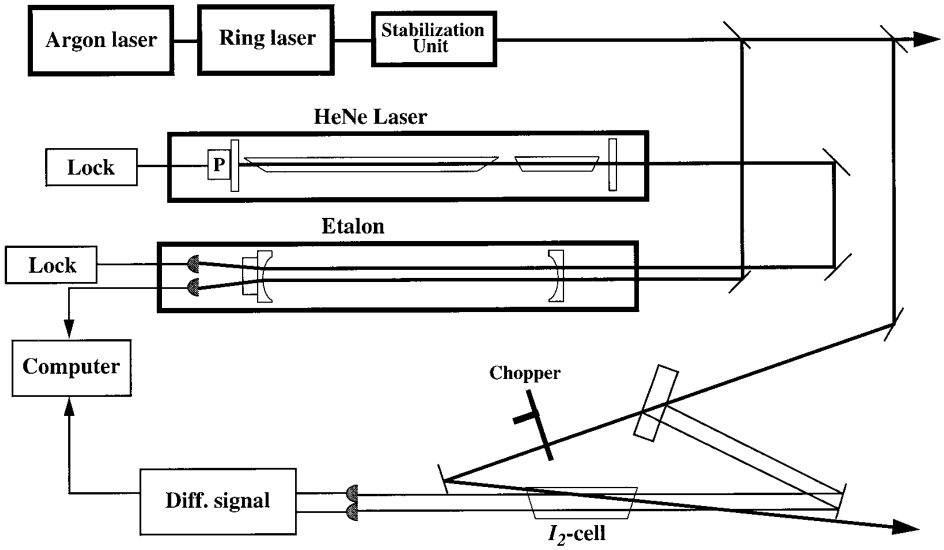

FIG. 1. Scheme of the experimental setup.

with a mechanically chopped (800 Hz) saturating beam. The differential signal is electronically filtered and stored in a computer. The sealed I_2 cell is used at room temperature vapor pressure. From the kHz-precision studies of Bordé *et al.* (*21*) we conclude that at our measuring conditions aiming at an accuracy of 2 MHz no pressure-induced or intensity-induced effects play a role.

Apart from the purpose of a coarse tuning needed to identify the resonances, no absolute wavelength measurements were performed in our experiment. The concept of the present method involves only relative frequency measurements with a stabilized (75-cm length) etalon with respect to calibrated absolute frequency points derived from the literature (*18, 20*). The étalon is pressure and temperature stabilized and further actively locked to the output of a HeNe-laser (Fig. 1). A $^{3}He^{22}Ne$ laser is actively locked to the *B* component of $^{129}I_2$ following the procedures of Schweitzer *et al.* (*22*). Before each measurement session the free spectral range (FSR) of the étalon is determined, resulting in a value of (148.9560 ± 0.0005) MHz, with small day-to-day variations. For this purpose accurately calibrated I_2 lines (*13, 18, 20*) were used. It is essential for a derivation of absolute frequencies that both the HeNe-laser and the étalon are kept in lock during an entire measurement session.

Subsequently computer-controlled scans of I_2 lines were recorded in saturated absorption with simultaneous detection of marker fringes of the étalon. In some cases overlapping scans were used to record the desired spectral line. After linearization of the scan by fitting a spline function through the marker positions, the relative position (between two markers) of a fixed component of each I_2 line is determined by means of a computerized fitting and interpolation procedure. In this way well-calibrated hyperfine components (''reference'' lines) and new lines were recorded. While the integer number of fringes separating these lines from reference lines is determined with the aid of the I_2 atlas the fractional values (to within 1% of FSR) give an improved calibration. Only when the separation exceeds several cm^{-1} does the error in the FSR contribute to the final uncertainty. In two cases long overlapping scans of 10 cm^{-1} were performed with the goal of calibrating an I_2 component close to a frequency of special interest. First, the frequency of a component at 1/10 of the $1^1S–2^1P$ resonance line of helium (*6, 7*) was recorded in overlapping scans starting at the $R99$''*i*''(15-1) line accurately calibrated by Grieser *et al.* (*18*). Second, the I_2 component $R100$''*t*''(14-0) at 1/6 the frequency of the *Q* branch of the $L–X(0, 0)$ band of CO (*8*) was recorded with respect to the $P98$''*t*''(14-0) line provided by Sansonetti (*20*). Data from these procedures are included in the present analysis.

An example is shown in Fig. 2 displaying a single scan over two rotational lines, $R17$ of (16-1) band and one of the reference lines $P12$(16-1), measured by Sansonetti (*20*) with accuracy of 1 MHz (the ''*t*'' components are denoted by *). The scan axis is calibrated to the reference line using the FSR of the etalon. In this way the absolute frequency of $R17$''*t*''(16-1) is determined with an accuracy of about 2 MHz.

Through frequency separation measurements hyperfine components of selected $P(J)$ and $R(J)$ lines in the (13-1) to (18-1) bands were recorded and calibrated (Table 1). The estimated absolute accuracy varies in the range 1–5 MHz. Except for the two wavelength ranges mentioned above lines were always recorded close to calibrated lines

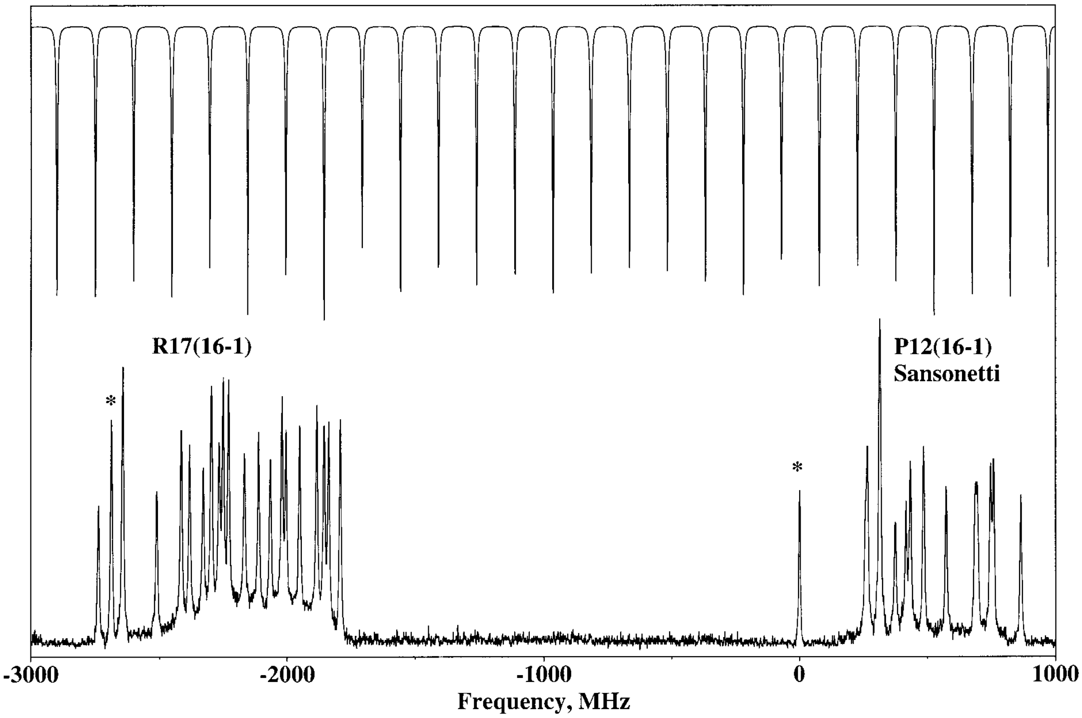

FIG. 2. Simultaneous scan over two lines (*R*17 and *P*12) in the *B*–*X* (16-1) band of I_2 (lower trace) and the fringes of a stabilized etalon (upper trace). The ''*t*'' components are denoted by *. Note that for the *R*17 line all 21 transitions corresponding to $\Delta F = \Delta J = +1$ are resolved.

presented in literature (*11, 13, 18, 20*). In all cases the ''*t*'' component was selected for calibration. For convenience and to avoid any confusion, two typical hyperfine spectral structures, for odd and even *J*, are displayed in Fig. 3 with the identification of hyperfine components (''*a*''–''*u*'') adopted from Sansonetti (*20*). For low *J* ($J < 20$) these typical shapes of the rotational lines undergo changes and more hyperfine components become resolved (see Fig. 2, where all 21 hyperfine components of *R*17(16-1) can be seen).

3. DATA ANALYSIS

In the analysis of their spectra Gerstenkorn and Luc (*1*) performed a fit to the rovibronic structure of the *B*–*X* system of I_2 with energy representations

$$E_{X\nu} = E_{\nu''} + B_{\nu''}J(J+1) - D_{\nu''}J^2(J+1)^2 + H_{\nu''}J^3(J+1)^3, \quad [1]$$

$$E_{B\nu} = E_{\nu'} + B_{\nu'}J(J+1) - D_{\nu'}J^2(J+1)^2 + H_{\nu'}J^3(J+1)^3 + L_{\nu'}J^4(J+1)^4 + M_{\nu'}J^5(J+1)^5 \quad [2]$$

for ground and excited states, respectively. The molecular parameters derived in (*1*) give an accurate description of transition frequencies up to $J \sim 160$ within the stated uncertainty of 60 MHz. The representation of Eqs. [1] and [2] will be used in the present study to denote the ''center of gravity,'' i.e., the energy level structure and transition frequencies with all hyperfine effects set to zero.

The hyperfine structure of I_2 has been studied previously for a large number of individual rotational lines scattered in the visible wavelength range. For the purpose of our analysis only the two most important terms in the hyperfine Hamiltonian (*23*) are included:

$$H_{hf} = eQq \cdot H_{\mathrm{EQ}} + C \cdot H_{\mathrm{SR}}, \quad [3]$$

where $eQq \cdot H_{\mathrm{EQ}}$ is the electric quadrupole term and $C \cdot H_{\mathrm{SR}}$ is the spin-rotation term (eQq and C are the conventional symbols representing the constants for the respective interactions). These terms are sufficient to represent the separations of $F = J$ hyperfine components with respect to the center-of-gravity at the 0.2 MHz level if $\Delta J = \pm 2$ contributions are included in the diagonalization of the hyperfine matrix. Following the discussion of Bordé *et al.* (*21*) we adopt for the $X\,^1\Sigma_g^+$, $\nu'' = 1$ ground state the hyperfine constants $eQq'' = -2452.6$ MHz and $C'' = 3$ kHz. For the excited vibrational levels the hyperfine constants (eQq and

TABLE 1
Measured Frequencies in This Study

Transition	"t" component, MHz	Transition	"t" component, MHz
P(9) 13-1	509601977.9(1.6)	P(65) 16-1	517185695.6(1.3)
R(9) 13-1	509632429.0(1.9)	R(69) 16-1	517217059.9(1.2)
R(13) 13-1	509609972.0(2.0)	P(93) 16-1	515659358.7(1.5)
P(15) 13-1	509545298.7(2.3)	R(116)16-1	514370607.9(1.1)
R(15) 13-1	509594978.1(1.6)	R(121)16-1	513979201.2(1.2)
P(18) 13-1	509508487.7(1.6)	P(124)16-1	513350047.8(1.1)
P(20) 13-1	509480808.2(1.9)	P(132)16-1	512647155.5(1.0)
R(20) 13-1	509546506.3(2.5)	R(136)16-1	512701376.6(1.1)
R(23) 13-1	509509886.3(1.7)	R(137)16-1	512610634.7(1.3)
R(25) 13-1	509482330.8(1.9)	P(8) 17-1	521586497.4(4.4)
P(27) 13-1	509364143.8(2.7)	R(14) 17-1	521575692.3(4.5)
R(32) 13-1	509366084.1(2.5)	R(15) 17-1	521567120.5(4.5)
R(38) 13-1	509241911.8(2.5)	P(22) 17-1	521418244.1(2.2)
P(39) 13-1	509092511.4(2.4)	P(42) 17-1	520948096.8(1.1)
R(44) 13-1	509095072.1(2.6)	R(54) 17-1	520705108.8(1.7)
P(44) 13-1	508952596.1(3.0)	P(64) 17-1	520117714.3(3.9)
P(46) 13-1	508892223.1(3.0)	R(68) 17-1	520143711.2(3.9)
R(49) 13-1	508955369.9(2.7)	R(6) 18-1	524475481.8(4.3)
R(51) 13-1	508895076.1(2.9)	R(8) 18-1	524468252.9(4.2)
P(65) 13-1	508192805.3(1.6)	R(18) 18-1	524390857.2(4.0)
R(75) 13-1	507974147.3(2.0)	P(19) 18-1	524319179.5(3.9)
R(79) 13-1	507785131.1(2.2)	R(24) 18-1	524311394.2(3.0)
R(83) 13-1	507585929.2(1.8)	P(27) 18-1	524177555.9(3.4)
R(87) 13-1	507376523.8(1.4)	R(31) 18-1	524187351.0(2.5)
R(91) 13-1	507156900.2(1.8)	P(31) 18-1	524090221.6(3.3)
P(99) 13-1	506369955.4(1.2)	R(40) 18-1	523978221.5(2.0)
R(104)13-1	506372344.2(1.3)	P(42) 18-1	523793188.2(1.8)
R(107)13-1	506175873.2(1.6)	R(46) 18-1	523807726.7(1.8)
R(111)13-1	505904887.9(1.6)	P(47) 18-1	523630566.8(1.3)
P(111)13-1	505550262.0(1.8)	R(50) 18-1	523680231.5(1.5)
R(116)13-1	505551619.9(1.9)	R(54) 18-1	523541657.9(1.2)
P(119)13-1	504952280.2(3.3)	R(56) 18-1	523468212.5(1.3)
P(123)13-1	504637769.6(1.1)	R(60) 18-1	523312993.5(1.8)
R(128)13-1	504637640.5(1.2)	P(61) 18-1	523083220.3(1.1)
R(130)13-1	504476197.9(1.6)	R(63) 18-1	523189287.3(1.6)
P(67) 14-1	511156922.0(1.1)	R(64) 18-1	523146661.6(1.1)
P(68) 14-1	511111745.3(1.1)	R(71) 18-1	522828790.3(1.5)
P(69) 14-1	511065923.3(1.1)	R(76) 18-1	522580821.6(2.0)
P(70) 14-1	511019455.2(1.4)	P(81) 18-1	522065155.8(3.3)
R(72) 14-1	511154541.7(1.2)	R(82) 18-1	522260185.6(2.5)
R(73) 14-1	511109285.1(1.3)	P(86) 18-1	521767018.7(2.0)
R(74) 14-1	511063381.3(1.1)	R(88) 18-1	521914325.2(1.5)
P(92) 14-1	509833399.7(1.1)	P(98) 18-1	520979920.4(3.2)
P(15) 15-1	515641846.2(2.4)	R(98) 18-1	521281620.3(1.4)
P(16) 15-1	515629854.0(1.2)	R(100)18-1	521146613.6(1.2)
R(19) 15-1	515651560.1(1.4)	P(101)18-1	520767302.1(1.4)
R(20) 15-1	515640121.4(1.3)	R(104)18-1	520868103.6(1.3)
R(21) 15-1	515628031.0(1.3)	P(104)18-1	520548328.3(2.2)
P(117)15-1	511044175.2(1.1)	P(105)18-1	520473922.7(2.2)
R(121)15-1	511103398.7(1.5)	R(110)18-1	520429053.2(1.3)
R(136)15-1	509850059.7(1.3)	R(113)18-1	520199924.4(1.5)
P(11) 16-1	518650204.9(1.3)	R(114)18-1	520122122.2(1.8)
R(17) 16-1	518637995.9(1.7)	R(116)18-1	519964376.7(1.7)
R(18) 16-1	518627631.3(1.5)	R(119)18-1	519722396.0(2.1)
P(21) 16-1	518525170.7(1.9)	P(123)18-1	519013099.8(3.6)
R(26) 16-1	518520828.6(1.3)	R(124)18-1	519304761.9(2.4)
P(30) 16-1	518355932.5(1.8)	R(132)18-1	518599137.6(1.4)
R(35) 16-1	518349878.8(1.8)		

Note. The second column gives the values for the ''*t*'' component along with the estimated accuracy of the measurement.

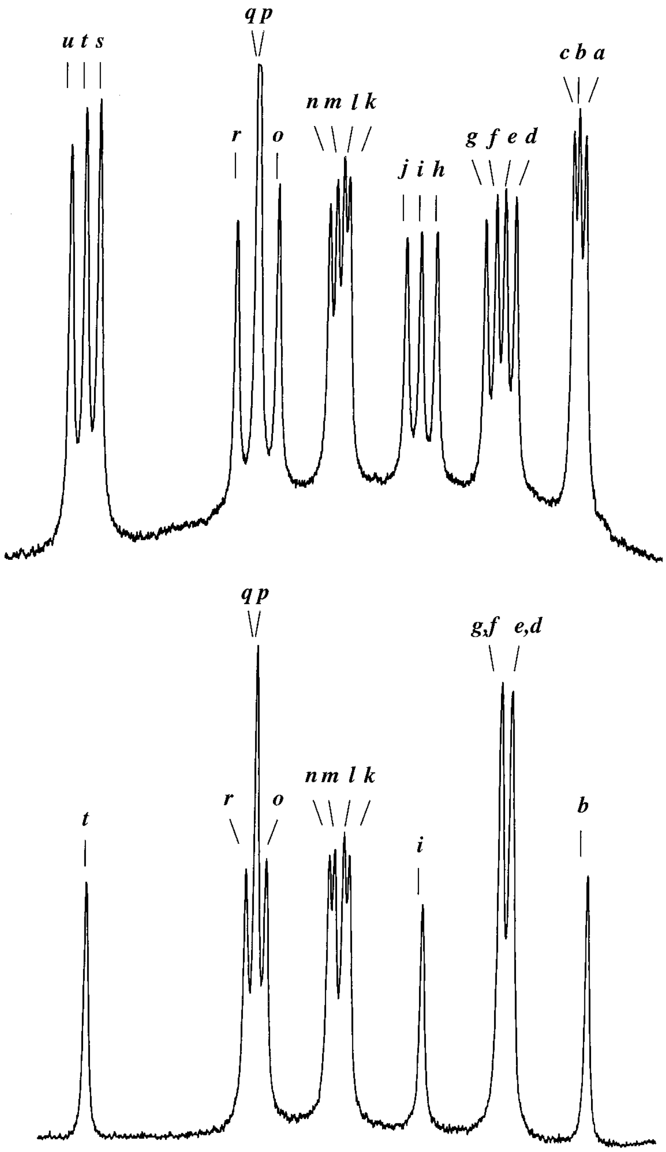

FIG. 3. Typical pattern of hyperfine structures in I_2. The upper trace is a recording of the $R69(16\text{-}1)$ line and shows the typical structure for an odd-J line with 21 $\Delta F = \Delta J$ main transitions. The lower trace shows the $R98(16\text{-}1)$ line with the typical 15 $\Delta F = \Delta J$ main transitions in an even-J line. In the present study we have adopted the identification of ‘‘*a*’’–‘‘*u*’’ components of Sansonetti (*20*).

C) have been reported for $\nu' = 15$ (*24*) and $\nu' = 17$ (*25*) only. In this work we adopted the hyperfine constants $eQq_{\nu'}$ and $C_{\nu'}$ ($\nu' = 13\text{–}18$) using the empirical formulae given by Razet and Picard (*26*), including the *J* dependence in $C_{\nu''}$. The uncertainty in the $eQq_{\nu'}$ is estimated at 0.3 MHz, which yields an uncertainty of less than 0.1 MHz in the position of the ‘‘*t*’’ component with respect to the center-of-gravity. Since the spin-rotation contribution to the eigenvalues for $F = J$ hyperfine components can be neglected at our level of accuracy, one of them, the ‘‘*t*’’ component, is preferably used in this study.

For all lines calibrated in the present study the centre of gravity of the rotational structure was determined by correcting the measured hyperfine components (Table 1) for the calculated hyperfine shift. All ‘‘*t*’’ components are red-shifted by approximately 483 MHz with slight variations over the six bands measured and over the rotational lines. The accurately calibrated lines cited in the literature (*9, 11, 18, 20*) were included in the analysis but not presented in Table 1. For each line an estimated uncertainty is listed as well, which depends on the measurement error and the uncertainty of the reference line used. For the collection of lines taken from Sansonetti the resulting uncertainty is less than 1.0 MHz, including a contribution of the hyperfine shift correction. For a number of previously reported lines calibrated by our group (*7, 8*) (not listed here), the accuracy varies from 3.0 to 5.0 MHz. The lines specifically calibrated for the present paper have an accuracy of 1.5–4.5 MHz depending on the measuring conditions.

For each band all available data on the center-of-gravity transition frequencies (calculated from Table 1 and from Refs. (*7, 8, 18, 20*)) were included in a weighted least squares fit to obtain a parametrization over the rotational structure in $B\text{–}X$ ($\nu' - 1$) bands ($\nu' = 13\text{–}18$). It turned out to be sufficient to vary only the band origin $E_0 = E_{\nu'} - E_1$, the rotational constant B_1, and the rotational constants $B_{\nu'}$, $D_{\nu'}$, and $H_{\nu'}$ of the excited state to find convergence with a resulting χ^2 of less than 1.0 per data point. The higher order molecular constants for the ground state ($\nu'' = 1$) were kept fixed at the values of Gerstenkorn and Luc (*1*): $D_1 =$

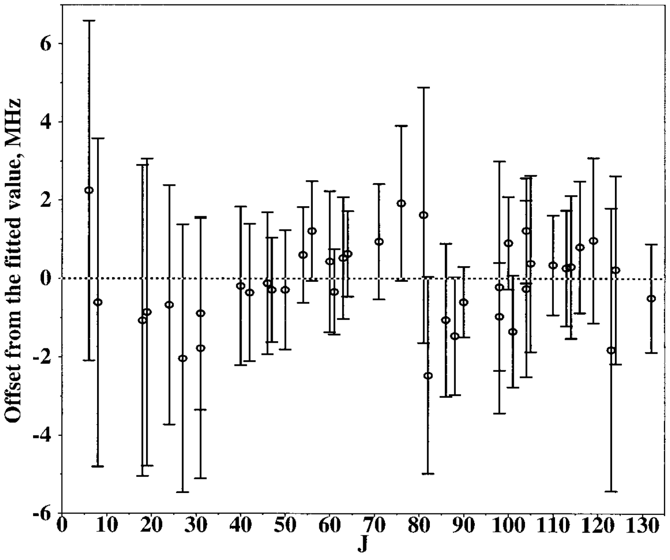

FIG. 4. The offset of the center-of-gravity frequencies, measured in the (18-1) band (the data points) from the fitted values (dashed line).

TABLE 2
The Derived Values for the Constants Appearing in Eqs. [1] and [2], Corresponding to (ν' − 1) Transitions

v'	$E_{v'} - E_1$	$B_1\times10^2$	$B_{v'}\times10^2$	$D_{v'}\times10^8$	$-H_{v'}\times10^{14}$	$-L_{v'}\times10^{19}$	$-M_{v'}\times10^{25}$
13	16999.931315	3.71968789	2.67324769	0.83217395	0.66855643	0.142729375	0.82943669
14	17102.554990	3.71970147	2.65407989	0.85592155	0.70580093	0.161026496	0.97512745
15	17203.371621	3.71969569	2.63449183	0.87997748	0.80130524	0.181804330	1.14494794
16	17302.358760	3.71969318	2.61450317	0.90621126	0.87175280	0.205409358	1.34313350
17	17399.494325	3.71964275	2.59404320	0.93415830	0.94165694	0.232226556	1.57482286
18	17494.756134	3.71967270	2.57323368	0.96427269	1.00923370	0.262682841	1.84624651

Note. $\nu' = (13–18)$. All the values are in wavenumbers.

4.5722085×10^{-9} cm^{-1} and $H_1 = -5.3403223 \times 10^{-16}$ cm^{-1}. The higher order centrifugal distortion coefficients for the exited states ($L_{\nu'}$ and $M_{\nu'}$) were also kept fixed at the values of Ref. (*1*). Since the goal of the present study is to find a parametrization of the energy levels only rather than deriving new details on the molecular structure, no further attempts in this latter direction were made. The resulting parameters for the transitions with upper level $\nu' = (13–18)$ (see Table 2) were inserted in Eqs. [1] and [2] for calculation of the center-of-gravity frequencies.

The consistency of the fits is such that for all bands the resulting χ^2 is much lower than the number of data points. In all cases five parameters (described above) were optimized in the fitting routines. From the resulting χ^2 values we estimate that the final uncertainties of all center-of-gravity rotational frequencies are accurate within 2 MHz. This can be seen from Fig. 4, where the offsets of the calibrated lines in (18-1) band from the fit are presented. Most of the measured values lay in the interval $(-2, +2)$ MHz and the fit passes through all error margins. Consequently, the fit for the center-of-gravity frequencies can be used to predict the positions of the ''*t*'' hyperfine components for all lines with 2 MHz accuracy. This procedure will be discussed in Section 4.

An important assumption for the validity of the present procedure is the unperturbed nature of the B–X system of I_2. The excited B state is known to be perturbed by a repulsive state of $^1\Pi_{1u}$ symmetry, causing predissociation in the B-state (*27*). This holds for the region $\nu' = (13–18)$ in the B state. At much higher ν states also other states interact and even u–g symmetry breaking has been observed (*21, 27*). The coupling with the $^1\Pi_{1u}$ state may in principle cause energy shifts to the B-state energy levels. However, the coupling to the repulsive state gives rise only to global effects, slowly varying over the J-quantum number, while no local effects such as accidental shifts are expected. Indeed such phenomena were not observed, while care was taken that $P(J)$ and $R(J)$ transitions were included in the fit in a wide range of J values with gaps as small as possible. Possible perturbative effects by the $^1\Pi_{1u}$ state are therefore included in the present parametrization of the B state energy levels by the effective molecular parameters as derived in the fits (Table 2).

4. THE REFERENCE STANDARD

The molecular parameters derived from the present measurements and fitting procedures give a representation of center-of-gravity transition frequencies of $R(J)$ and $P(J)$ lines in the (13-1) to (18-1) bands for J up to 140 at an accuracy level of 2 MHz (Table 2). By invoking the theoretically known hyperfine structure, the frequency of the ''*t*'' component of each line can be calculated at the same accuracy level. A list of these calculated transition frequencies of ''*t*'' components is given in supplementary data, corresponding to R and P branches, respectively. The tables start at $J = 9$ because of the strong mutual overlapping of the rotational lines with lower J, which makes them unsuited for spectroscopic calibration. These frequencies form a secondary reference standard in the range 571–596 nm. It is a factor of 30 more accurate than the Doppler-broadened I_2-reference standard (*1*). Since a calibrated component is found in nearly all intervals of 1 cm^{-1}, this dense grid of reference lines should become a useful tool in laser spectroscopic studies.

5. CONCLUSIONS

In this paper we have shown that a simple parametrization (involving five molecular parameters) of the positions of the ''t'' hyperfine components in the rotational structure of $^{127}I_2$ is possible giving an accuracy of 2 MHz, suitable for precise spectroscopic measurements. Using this procedure we present a table, containing 1584 accurate wavenumbers (see supplementary data), that predicts the line-positions of the ''t'' components in the B–X system of $^{127}I_2$ for vibrational bands (13-1) up to (18-1) and rotational quantum numbers J from 9 up to 140 (both for R and P branches). This dense grid of high precision data ($\delta\nu/\nu \approx 4 \times 10^{-9}$) covers the wavelength range 571–596 nm, corresponding to a frequency span of 715 cm^{-1}. In a wavelength range near 790 nm a similar reference standard based on I_2 hyperfine components is being developed by Knöckel *et al.* (*28*).

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