

de l'Imprimerie Royale, MDCXCVI). It was also translated and published in 1730 by E. Stone under the title *The Method of Fluxions both Direct and Inverse* (The Former being a translation from... and the later supply'd by the Translator). Newton's original work was first published by John Colson in English in 1736, in a book entitled *The Method of Fluxions and Infinite Series* with its Application to the Geometry of Curve-Lines, By the Inventor Sir Isaac Newton, Kt., Late president of the Royal Society. Translated from the author's Latin Original not yet made publick. London, Printed by Henry Woodfall; M.DCC.XXXVI.

⁸¹D. J. Struik, "The origin of l'Hospital's rule," *Math. Teacher* **56**, 257–260 (1963).

⁸²G. W. Leibniz, *Nova Methodus pro Maximis et minimis*... *Acta Eruditorum*, 467–473 (Anno MDCLXXXIV).

⁸³Hooke's biographer Waller informs us that after about 1681/2 "...From this time, or rather something before, he began to be more reserv'd than he had been formerly, so that altho' he often made Experiments, and shew'd new Instruments and Inventions, and read his

Cutlerian Lectures, yet he seldom left any full Account of them to be enter'd designing, as he said, to fit them himself for the Press, and then make them publick, which he never perform'd. This is the reason that I am oblig'd to be the shorter in the remaining part of his Life; and shall only touch upon some few of his Performances, since the bare naming of them, or mentioning their Titles, will but create an uneasy Curiosity in the Reader without any satisfaction... In the beginning of the Year 1687, his Brothers Daughter, Mrs. Grace Hooke dy'd, who had liv'd with him several Years, the concern for whose Death he hardly ever wore off, being observ'd from that time to grow less active, more Melancholly and Cynical..." Ref. 22, p. 24. Grace was his mistress for some time, and her death followed shortly after the publication of the *Principia*, printed on July, 1686.

⁸⁴At the time these words were written, I. Newton was president of the Royal Society, a post he accepted only after Hooke died, and Richard Waller was the secretary of the Society.

⁸⁵I am indebted to D. W. Dewhirst for help in deciphering some words in Hooke's text.

Iodine molecular constants from absorption and laser fluorescence

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Absorption and laser-induced fluorescence spectra of the iodine molecule are compared as sources of molecular constants. The comparative simplicity of the fluorescence spectrum and the increased precision provide a vivid and direct understanding of the iodine molecular ground state.

I. INTRODUCTION

The iodine molecular spectrum was one of the first to be analyzed successfully and provides an ideal case for demonstrating the basic characteristics of diatomic spectra.^{1–3} It possesses a long absorption series in the visible region associated with the $B-X$ states which correlate at large separations with the well-known atomic fine structure states, as shown in Fig. 1. This spectrum has remained important in the development of new techniques in molecular spectroscopy following the advent of lasers. Methods of optical pumping in molecules and the development of saturation and polarization spectroscopy were pioneered in iodine and many of its molecular constants are known with considerable precision.^{4–8}

The most striking element of the iodine absorption spectrum is the long series of the $B-X$ bands in the visible. The electronic potential energy curves which give rise to the electronic-vibrational-rotational levels involved in the bands are shown as full lines in Fig. 1. The B level is the first member of these electronically excited levels. Levels other than the ground X level which correlate to the ground atomic configuration at large separation are shown

as broken lines, although the repulsive states are not yet well established. Both B and X states have $\Omega=0$, (i.e., zero electronic angular momentum along the molecular axis) in standard molecular notation as specified in Chap. IV of Ref. 2. The visible system of $B-X$ bands can be studied in straightforward absorption spectroscopy and yields a great deal of information even when studied with an instrument of modest resolving power.

Absorption experiments on the $B-X$ system have been carried out in student laboratories for many years (see, e.g., Ref. 9) and, while we summarize the information that can be obtained by conventional spectroscopy, the aim of this paper is to describe a laser fluorescence experiment which gives a greatly simplified spectrum. Due to fortuitous coincidences with molecular absorptions, fluorescence from iodine can be stimulated by light from an inexpensive He-Ne laser. The fluorescence spectrum is simpler to analyze than the absorption spectrum and gives immediate evidence for the J -selection rule, anharmonicity of the lower state, and the difference between the bond lengths of the B and X states.

We briefly summarize the analysis of the visible absorp-

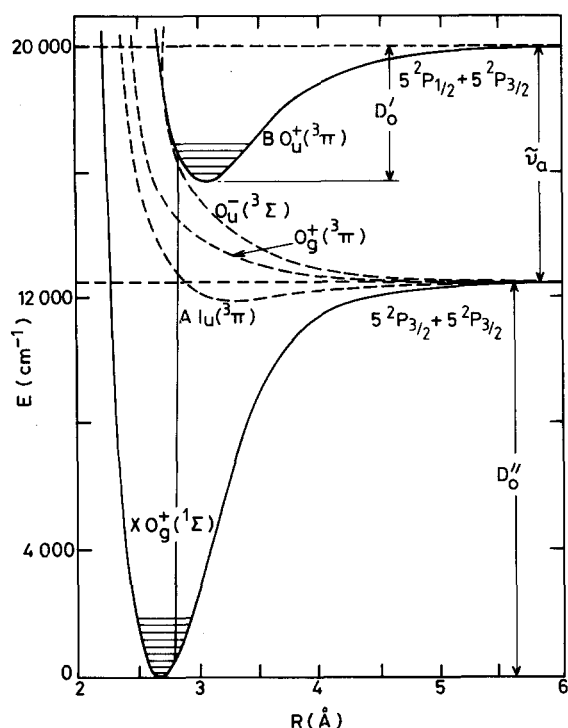


Fig. 1. Iodine molecular potential energy curves for the lowest lying states.

tion spectrum to highlight the complementarity of the fluorescence and absorption spectra and show how the combined results can characterize both the *B* and *X* states of the molecule.

II. EXPERIMENT

A. Absorption spectra

In Fig. 2 we show the absorption spectrum of iodine in the region 500–630 nm. The continuum was provided by a

quartz-iodine lamp, the room temperature absorption tube was approximately 1 m in length, and the spectrum was recorded photoelectrically by means of a Spex 1/4 m monochromator. Detection was by an IP28 photomultiplier and the voltage developed across the anode resistor was recorded directly vs time on an *x-t* chart recorder as the monochromator was scanned. The resolution is of the order of 0.1 nm and the spectrum is very nearly linear in wavelength. Positions of suitable calibration lines from a mercury-cadmium lamp are also indicated.

Sections of three series of electronic-vibrational bandheads can be seen in the absorption spectrum in Fig. 2. They arise from absorption from the three lowest vibrational levels of the *X* ground state and are labeled by vibrational quantum numbers for the upper state, *v'*, and the ground state, *v''*. The series towards the short and long wavelength ends of the spectrum arise from the *v''*=0 and *v''*=2 levels, respectively, and there are regions of overlap between both these series and the series originating in the ground *v''*=1 level, which lies in the middle of the spectrum. All bands are shaded towards long wavelengths due to unresolved rotational structure. A straightforward analysis of the vibrational spectrum can be made with the assumption that the observed bandheads have a constant, or at least slowly varying relation to the band origins. The Birge plot in Fig. 3 gives an immediate analysis of the vibrational energies of the *B* state. At small wave numbers the vibrational energy differences tend to about 125 cm⁻¹, corresponding to the frequency of the harmonic potential of the *B* state at the equilibrium position.

This harmonic potential is a parabola fitted to the bottom of the potential while the real potential becomes progressively wider than that as the vibrational energy increases. Since such a widening of the well corresponds to weaker harmonic forces the vibrational energy intervals become correspondingly smaller as dissociation is approached. This anharmonicity in the upper *B* state is immediately obvious in Fig. 3 with the interval rapidly approaching zero at large wave numbers. Since there is no long range potential barrier in the *B* state of iodine a mono-

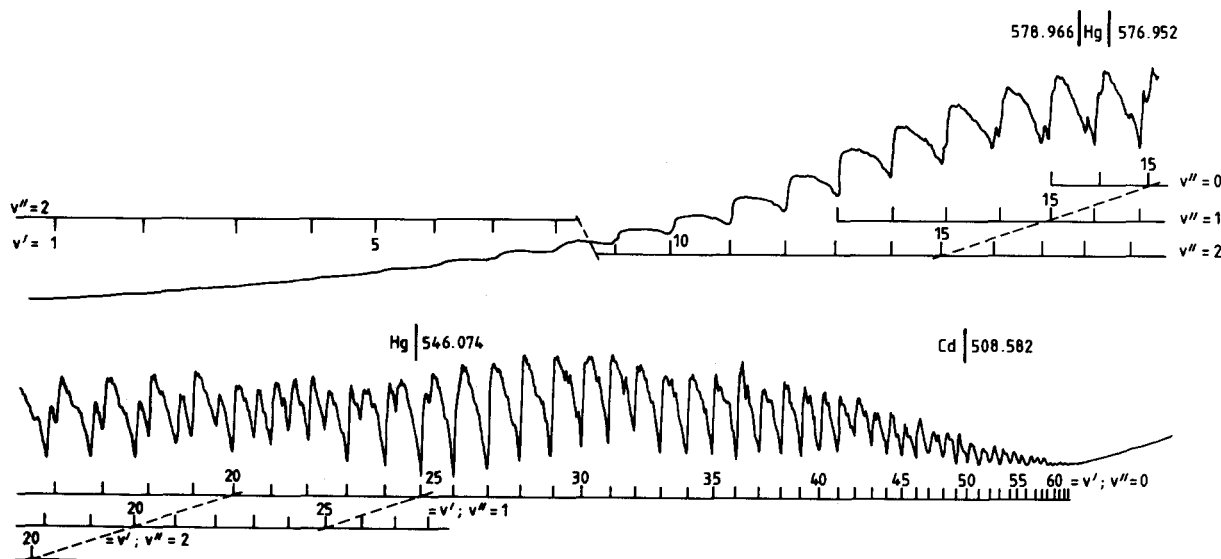


Fig. 2. Absorption spectrum of molecular iodine in the visible. The positions of calibration lines from a mercury/cadmium lamp are indicated and the three vibrational series are assigned.

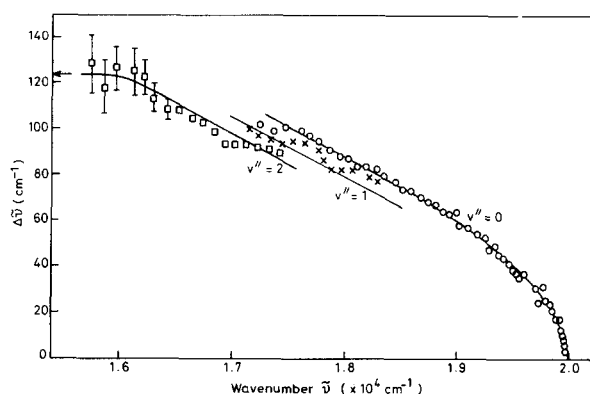


Fig. 3. Birge plot for the B - X system of molecular iodine, $\tilde{\nu}$ is the larger of the two wave numbers associated with the interval.

tonic extrapolation on this plot can be made to the short wavelength limit of the $v''=0$ series at about $20\,000\text{ cm}^{-1}$. Subtraction of the atomic energy difference,¹⁰ 7603.15 cm^{-1} , ν_a , gives the dissociation energy of the ground electronic state D_0'' , in good agreement with the accepted value in Table I. This plot displays the assignment of the bandheads to the three series which are progressively shifted to smaller energies by the ground state vibrational intervals as v'' increases. The region of overlap indicates that this vibrational interval is somewhat larger than 200 cm^{-1} . The progression of vibrational intervals allows a correlation of vibrational quantum numbers between the three series so that the separation of pairs of bandheads with the common upper vibrational number v' can be obtained directly from the spectrum with considerable precision (Table I). The slight oscillatory behavior of the intervals is the result of unresolved structure in the region of the bandheads. The ground state anharmonicity is only marginally evident from the difference in the $\Delta(v''=0-v''=1)$ and $\Delta(v''=1-v''=2)$ intervals. The close similarity of this interval to kT at room temperature explains why so few absorption series are visible and the scale on which the rotational structure degrades the bands.

Table I. Molecular constants for the B and X states of iodine. All quantities in cm^{-1} . (Standard deviations from random errors are quoted. Absolute values of vibrational and rotational constants are subject to a further systematic error from the calibration factor, of the order of 0.5%.)

Value obtained from	Absorption	Laser fluorescence	Literature
$X\ O_2^+(^1\Sigma)$			
D_0''	12 600(250)	...	12 440.9(1.1) ^a
$\omega_e''(C_{v1})$	213(1)	212.5(5)	213.695 ^b
$x\omega_e''(C_{v2})$	-2(2)	-0.58(2)	-0.608 ^b
$B_v''(C_{r1})$...	0.038(2)	0.0371 ^b
$B\ O_2^+(^3\Pi)$			
D_0'	4400(50)	...	4391 ^c
$\omega_e'(C_{v1})$	130(2)	...	125.18 ^b
$x_e\omega_e'(C_{v2})$	-0.985(15)	...	-0.744 ^b

^aReference 16.

^bReference 7.

^cReference 15.

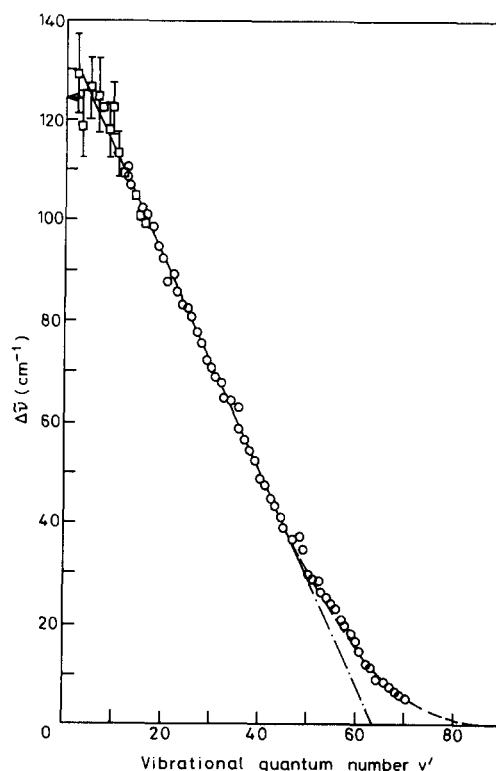


Fig. 4. Birge-Sponer plot for the B - X system of molecular iodine indicating the closely linear anharmonicity of the B vibrational energies. v' is the larger of the two quantum numbers.

The absolute numbering of the bands can be determined from a study of the long wavelength end of the spectrum. In Fig. 2 the last clearly visible bandhead is $v''=5$ but greater amplification of the long wavelength part of the spectrum enables all bandheads to $v''=0$ to be seen. The numbering from the $v''=2$ series can then be transferred to the other series, as indicated by the dotted lines in Fig. 2, so that the $v''=0$, $v'=25$ band lies in near coincidence with the mercury 546.1 nm line.

This numbering can be used in a Birge-Sponer plot (Fig. 4) where upper vibrational intervals from all three series are plotted against v' . This plot indicates that the anharmonicity is closely linear in vibrational number v' . As noted above the extrapolation to the dissociation limit is not invalidated by the presence of any long range potential barrier as in so many cases, but the linearity of this Birge-Sponer plot is modified by the nature of the long range van der Waals forces. The broken line in Fig. 4 indicates the deviation from linearity for $v' > 50$ which was found by LeRoy and Bernstein¹¹ up to the last bound level with $v'=87$. This nonlinearity is not so evident in the plot in Fig. 3 and does not affect the derivation of D_0'' from the extrapolation, within the 250 cm^{-1} accuracy of this experiment. The area under the Birge-Sponer curve (Fig. 4) is the sum of all vibrational energies of the excited B state, i.e., the dissociation energy D_0' (Table I).

B. Laser fluorescence

1. Experimental considerations

A much simpler spectrum can be obtained in laser fluorescence excited by the He-Ne line 632.8 nm . The exci-

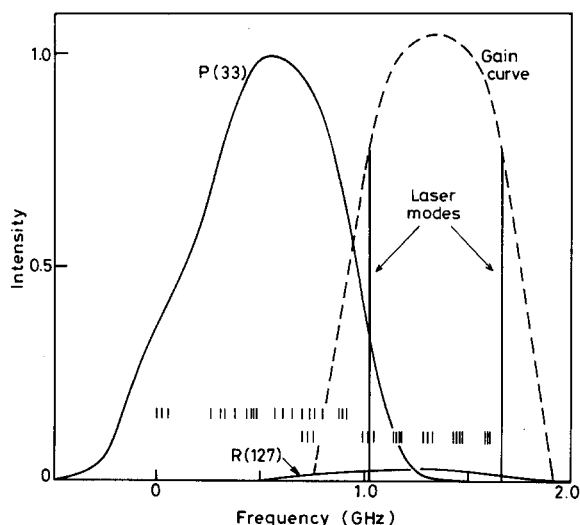
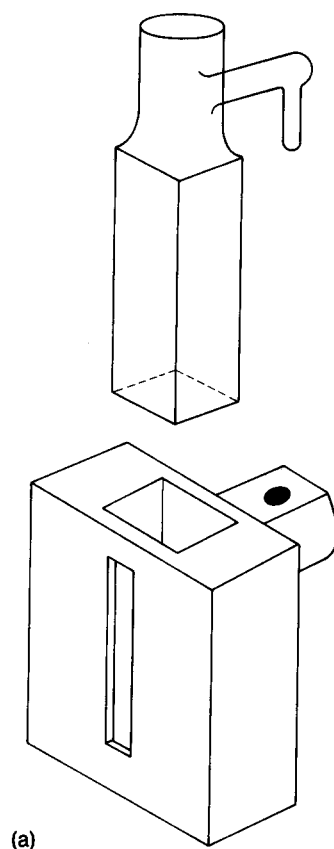


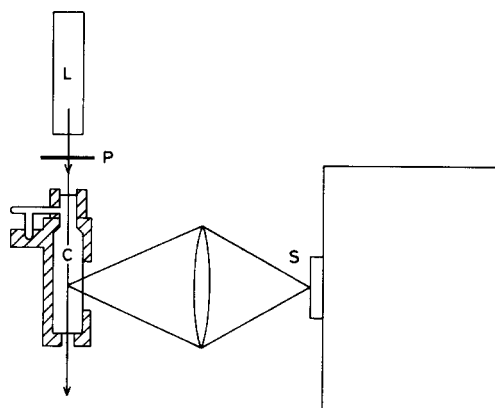
Fig. 5. The absorption spectrum of iodine molecule in the region of the He-Ne laser line 632.8 nm. Solid curves for the $P(33)$ and $R(127)$ lines are the total profile for the many hyperfine components for each transition, whose positions are indicated. The frequency scale is relative to the lowest of these components.

tation is due to an accidental coincidence between this line and two electronic-vibrational-rotational transitions in the iodine molecule as shown in Fig. 5.¹² The line $R(127)$ 11-5, in the usual notation, is in almost exact coincidence with the neon line at 632.8 nm while the $P(33)$ 6-3 line lies only 0.023 cm^{-1} (0.590 GHz) to lower frequency. The iodine Doppler width for room temperature is about 0.38 GHz, but each iodine line consists of many hyperfine components with approximately equal intensities⁸ so that the total absorption profiles are those that are indicated as full lines in Fig. 5, with a full width of approximately twice the Doppler width.¹³ The order of magnitude difference in the absorption by the two lines is determined by the Boltzmann factor for the two vibrational quanta difference in the lower levels, which corresponds to approximately $2kT$ at laboratory temperatures. These absorption lines are in the region of very weak absorption to the extreme long wavelength end of Fig. 2.

The lower levels of both these transitions are considerably less populated than the lower levels from which the absorption spectrum, discussed in the previous section, was obtained. It is therefore necessary to increase the iodine vapor pressure by heating the fluorescence cell. The cell is shown in Fig. 6(a) and is a modified Optilas silica cell 65 mm long and with square cross section of side 12.5 mm. The cylindrical end section is closed off with a silica window and a narrow sidearm for filling, with a reservoir, is attached between the window and the cell. When filling the cell iodine was progressively distilled under vacuum along a series of four such reservoirs, only the last reservoir being left attached when the cell was sealed off. The body of the cell is encased in a Dural block in which a 200 W cartridge heater (Diemould 1 1/2 in. \times 1/4 in.) is also fitted. The cell temperature was monitored by a thermocouple attached to this block. A block temperature of about 20 K above room temperature (about 20 °C) gave suitable vapor pressure at which the fluorescence from the laser light was clearly visible in the darkened laboratory. Since this is, however, too faint to be used for aligning the fluorescence with the



(a)



(b)

Fig. 6. (a) Silica fluorescence cell and oven. (b) Optical system for observation of molecular fluorescence, in vertical section. L—laser, P—linear polarizer, C—fluorescence cell, S—spectrometer slit.

monochromator axis, the alignment is carried out with light scattered from a copper wire inserted in the laser beam which is then replaced by the cell. The optical system is given in Fig. 6(b).

We have used a Spectra Physics Series 136 He-Ne laser which provides 1.1 mW of light. The gain curve of the laser is indicated as the broken line in Fig. 5 and is about 1.5 GHz wide, approximately equal to the Doppler width of neon at laboratory temperatures. The laser output consists of two longitudinal modes separated by 0.59 GHz and with negligible width on our scale. These modes drift within the gain curve as the laser warms up and finally settle down to positions symmetric about the line center, as indicated in Fig. 5. The two modes are linearly polarized orthogonally

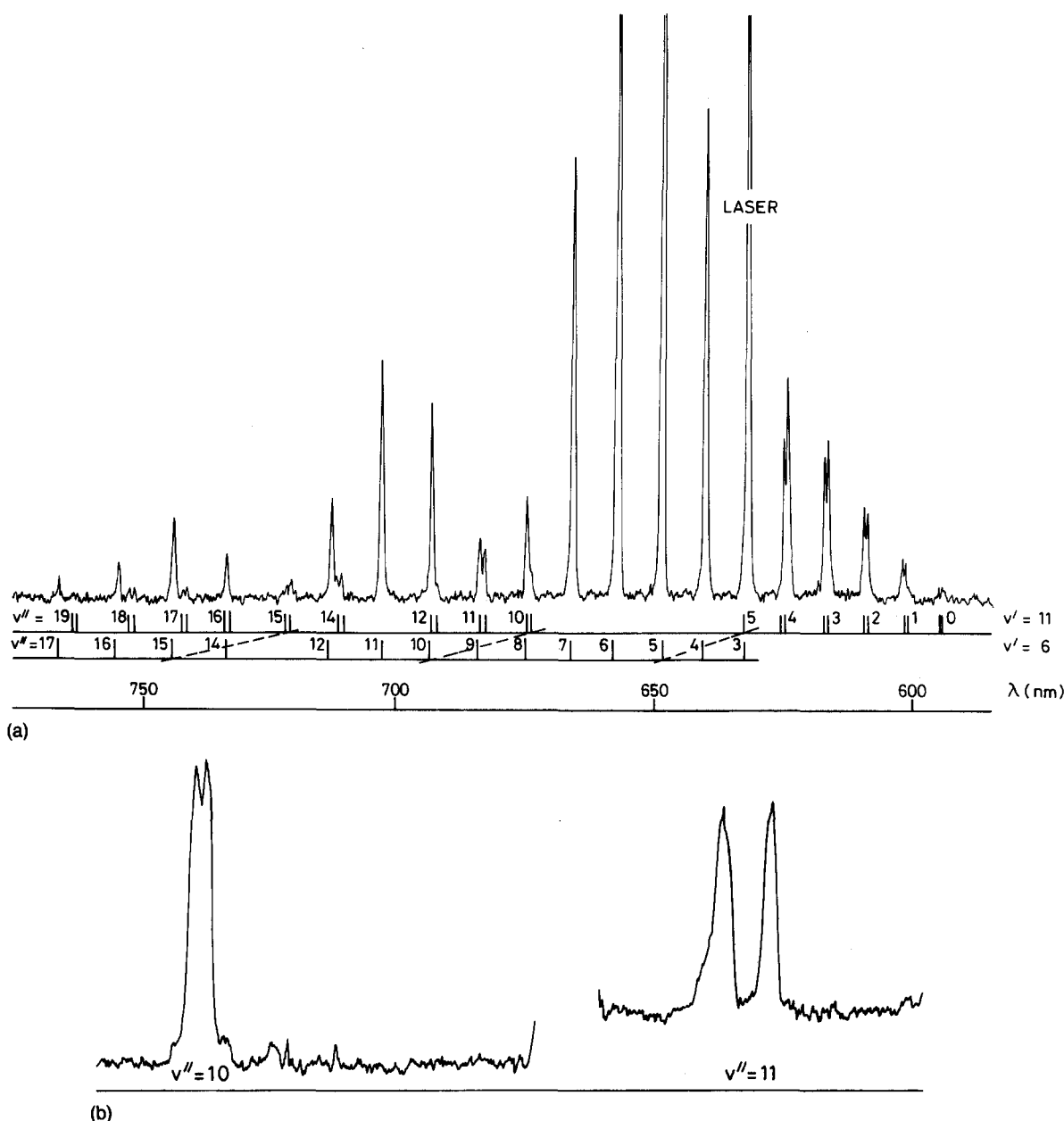


Fig. 7. Spectrum of molecular iodine fluorescence spectrum excited by He-Ne 632.8 nm. The series originating in the $v' = 3$ and $v' = 6$ levels are assigned. (a) complete fluorescence spectrum, (b) higher resolution trace of components of each of the two series, showing rotational splittings of 19.3 and 4.7 cm^{-1} , respectively.

to each other. Insertion of a linear polarizer between the laser and the cell can therefore be used to select one or the other mode thus modifying the relative absorption by the $P(33)$ and $R(127)$ lines, and so differentiating between the two fluorescent series. In this experiment we have used an EMI Trialkali photomultiplier to obtain good sensitivity and a spectrum extending well to the red of the laser wavelength. A neon discharge lamp provides many strong calibration lines in this spectral region.

2. The fluorescent spectrum

A typical fluorescent spectrum is shown in Fig. 7(a). It consists of the resonant fluorescence at the wavelength of the laser and a series of almost equally spaced lines some of which appear as doublets. The intensities exhibit a wave-like variation along the spectrum. Conventionally the lines

to the long wavelength side of the incident light are denoted as Stokes lines and those to the shorter wavelength side as anti-Stokes lines.

The spectrum consists of two series originating from the two absorption lines discussed above and can be understood with reference to the partial term diagram in Fig. 8. The two series result from the different excited levels $J' = 128$, $v' = 11$, and $J' = 32$, $v' = 6$, and may be distinguished by the different magnitudes of the rotational doublet splittings which are only resolved for the series from the $v' = 11$, $J' = 128$ level. This series is emitted after absorption from the $v'' = 5$ lower vibrational level and the fluorescence therefore contains lines to the five lower lying levels, $v'' = 0-4$, which constitute the five anti-Stokes lines in the spectrum. Only Stokes lines appear to originate from the upper $v' = 6$ level. Since the vibrational intervals are

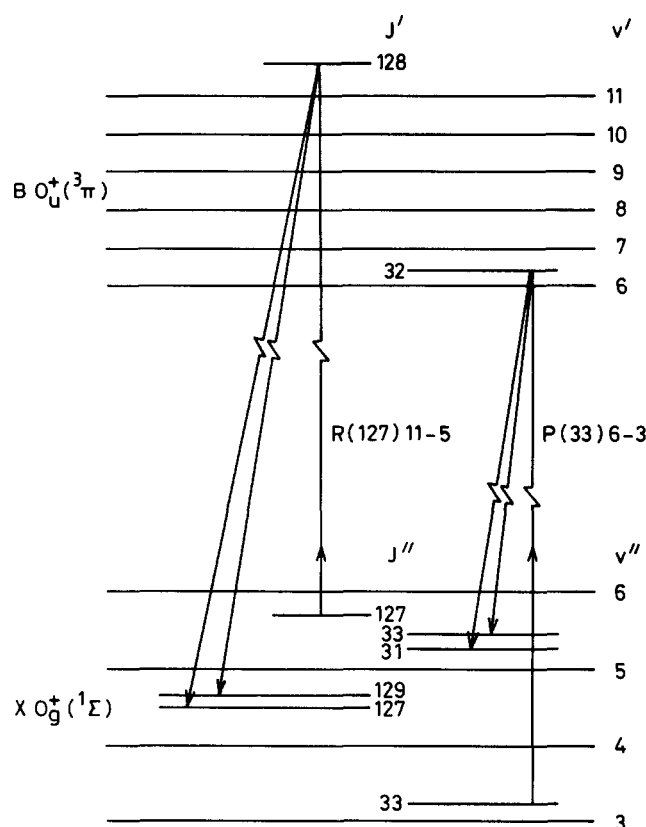


Fig. 8. Partial term diagram indicating the transitions involved in producing typical members of the two fluorescent series.

only weakly anharmonic, members of the two series lie very close together with the more intense obscuring the weaker in several cases.

The molecular state energies are given by

$$E(v, J) = T_e + G_v + B_v J(J+1) - D_v [J(J+1)]^2 + \cdots, \quad (1)$$

where T_e is the electronic energy relative to the ground state, G_v is the vibrational energy, B_v is the rotational constant, and D_v is the first correction for centrifugal distortion. For unperturbed levels G_v and B_v are smooth functions of v and can be expanded as Dunham series,

$$G_v = \sum_{i=1}^m C_{vi} (\nu + 1/2)^i, \quad (2)$$

$$B_v = \sum_{j=0}^n C_{rj} (\nu + 1/2)^j.$$

In Table I we also give the older and more commonly used, notation for these constants.

The spectrum in Fig. 7(a) is to be interpreted in terms of the differences (in cm^{-1}),

$$\tilde{\nu} = E(v'', J'') - E(v', J'), \quad (3)$$

where J' and v' are the values given above but J'' and v'' can take any value allowed by the selection rules. In the fluorescence spectrum there are therefore two series which display the vibrational-rotational energies of the X state relative to the position of the laser line, as indicated in Fig. 7(a). The two series are displaced by almost exactly two excited state vibrational intervals, anti-Stokes lines, to the

longer wavelength side of the laser, show a doublet structure which arises from the rotational selection rule $\Delta J = +1$ or -1 (the P and R components) in emission from the excited level with $v' = 11$, $J' = 128$. Only P and R components are seen since the Q ($\Delta J = 0$) component is forbidden for transitions between $\Omega = 0$ electronic states. The observed splittings are thus all between the $J'' = 127$ and $J'' = 129$ levels of the rotational states associated with the various lower vibrational states. From Eqs. (1) and (3) we obtain

$$\Delta\tilde{\nu} = 2B_{v''}(2J' + 1) - 4D_v(2J'^3 + 3J'^2 + 3J' + 1), \quad (4)$$

where $J' = 128$ is the rotational angular momentum of the upper level of the pair.

The standard estimate of the centrifugal distortion term, D_v , is $4B_v^3/\omega^3 = 4.4 \times 10^{-9} \text{ cm}^{-1}$ so that the last term is a correction of the order of the experimental uncertainty. The doublet splittings for this series is therefore close to $514 B_{11}$. The lines immediately to the long wavelength of the incident laser light arise from the $P(33)$ absorption, and are fluorescence from the $v' = 6$, $J' = 32$ level. They therefore have a much smaller splitting of $130 B_6$ and the centrifugal distortion is correspondingly smaller. While the splitting is not resolved in Fig. 7(a) it may be observed with a modest increase in resolving power. Figure 7(b) shows the two fluorescent bands, one from each series, taken with increased resolution and clearly exhibits the relative sizes of the splittings in the two series. At the precision of this experiment there is no observable dependence of the rotational splittings on v'' so that a single value of B is quoted for the two series in Table I.

Since $B = h/8\pi^2 Ic$, the bond length of the molecule in the X state can be derived from its moment of inertia, I , and reduced mass $\mu = 1.053\,766\,203 \times 10^{-25} \text{ kg}$, as $2.68(3) \times 10^{-8} \text{ cm}$.

The whole series of five anti-Stokes lines arising from the $R(127)11-5$ excitation can be seen in Fig. 7(a) and other members of the fluorescent series, with $v'' > 9$, appearing as Stokes lines to the long wavelength side of the laser. For the $P(33)6-3$ fluorescence most of the components between $v' = 11$ and 20 can be identified. The dependence of vibrational energy on v' is shown in the Birge-Sponer plot in Fig. 9 to be closely linear. The low intensity of the first members of the series make the measurement of these first few intervals relatively imprecise.

The variation in intensity along the fluorescent series is determined by the Franck-Condon factors, the squares of the overlap of the vibrational wave functions of the two states of the transition. In principle the constants in Table I specify the electronic potentials and can be used to generate these factors, but for the present we note that the variation of intensity displayed in Fig. 7(a) agrees qualitatively with the published variation of Tellinghuisen¹⁴ for the series originating in the $v' = 6$ level. In this case there is a monotonic rise from $v'' = 0$ to a maximum at $v'' = 6$ and oscillation at higher v'' with a period of between 4 and 5 quantum numbers. This pattern is also exhibited by the series originating from $v' = 11$. The wavelike variation of the intensities along these series is a striking demonstration of the quantum nature of the vibrational wave functions.

Table I indicates the level of precision which can be obtained for the molecular constants in this experiment. We also quote values from the literature, rounded to the next higher level of precision. We see that the two experi-

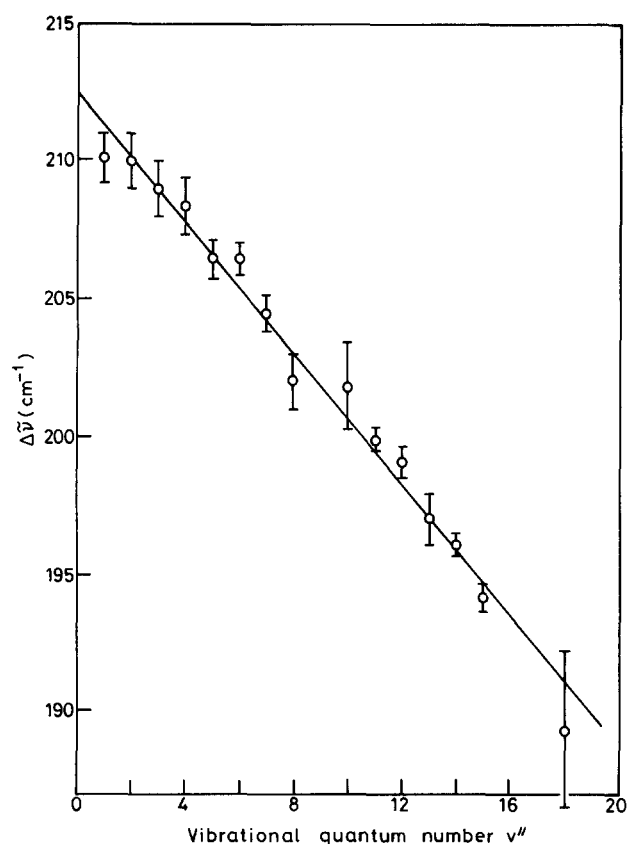


Fig. 9. Birge-Sponer plot for the fluorescent series. Vibrational intervals of the X state against v'' , the larger of the quantum numbers for the interval.

ments are very largely complementary in that the laser fluorescence spectrum provides data on the rotation and vibration of the *ground* state whereas the absorption gives less precise information on the vibrational intervals of the *upper* state and also yields the dissociation energies.

III. CONCLUSIONS

We have described simple laboratory experiments on the absorption and laser fluorescence in iodine which can yield a very comprehensive range of molecular constants for the diatomic molecule providing an insight into the molecular

structure. The two experiments are to some extent complementary in the information that they provide but the fluorescence data are considerably simpler to interpret, give greater precision, and demonstrate quantum selection rules and overlap integrals.

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THE CURIES, MARIE AND IRÈNE

Men and women who reverence Marie Curie for the scientific prowess that led to her winning the Nobel Prize on two occasions are apt to forget what is the most remarkable thing about her—that in spite of the intent and single-minded concentration that such a feat as she accomplished calls for, she raised a daughter, Irène, who, instead of, in the modern fashion, denouncing her parents and all their works and becoming a fashion model or dashing off to India to seek Enlightenment, became a Nobel Prize winner herself.

Peter Medawar, *The Limits of Science* (1984). (Reprinted by Oxford University Press, New York, 1987), p. 29.