

cal evidence exists for bound excited states in this energy range.¹⁵ It is therefore difficult to interpret the two sharp structures at 8.02 and 8.25 eV in the usual fashion, and no clear-cut answer can be provided. The possibility exists that the two resonances result from an avoided crossing of two O_2^- potential-energy curves, which may lead to a well of sufficient depth to support two vibrational modes.

Condon¹⁶ has shown that Franck-Condon factors associated with repulsive-type curves can exhibit undulations which he called "diffraction bands in the continuous spectrum." These have been recently confirmed theoretically^{13, 17} and observed experimentally.^{13, 17, 18} In fact, electron beam experiments also could be interpreted in this fashion.^{19, 20} The absorption peaks resulting from "undulations" and measured by Bixon, Raz, and Jortner¹³ are indicated in Fig. 3 by the vertical solid lines and the strongest absorption peaks of Tanaka¹⁴ by the vertical dashed lines. Fair agreement exists between the absorption peaks and the structure found in the present experiment. The two sharp structures at 8.02 and 8.25 eV seem to be exceptions, and their sharpness would indicate that they do not result from "diffraction" effects.

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Complete Hyperfine Structure of a Molecular Iodine Line*

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The complete hyperfine structure of the $P(117)$ line of the 21-1 band of the $B-X$ transition of $^{127}I_2$ is observed at 568.2 nm. A resolution in excess of 10^8 is obtained by a sensitive technique of saturation spectroscopy using a krypton-ion laser. The spectrum can be fitted by a model which includes both a nuclear electric quadrupole and a magnetic hyperfine interaction.

The hyperfine structure in the visible absorption spectrum of molecular vapors is usually masked by Doppler broadening.¹ The techniques of laser-saturated absorption²⁻⁴ offer a way,

however, to overcome this difficulty. By studying inverted Lamb dips in a 632.8-nm He-Ne laser with an iodine-vapor absorption cell placed inside the cavity, Hanes and Dahlstrom⁵ were

able to detect and resolve fourteen of the hyperfine components⁶ of a single rotational vibrational line of $^{127}\text{I}_2$.

This paper reports the observation of the complete hyperfine structure of the $P(117)$ line of the 21-1 band of the $B^3\Pi_{ou}^+ - X^1\Sigma_{og}^+$ transition in $^{127}\text{I}_2$. This resonance falls within the 5500-MHz tuning range of one (568.2 nm) of the eight output wavelengths of the cw krypton-ion laser. A sensitive and convenient technique of saturation spectroscopy was used to resolve the ~1400 MHz wide [full width at half-maximum (FWHM)], inhomogeneously broadened line into 21 components of ~10-MHz width. This represents a resolution in excess of 10^8 .

In the present technique, two beams of equal frequency are sent in opposite directions through an external iodine-vapor absorption cell. One beam is periodically chopped and the intensity modulation of the other (probe) beam, which results from the nonlinear interaction in the absorbing gas, is detected. The transmission of the probe beam is enhanced by the saturation of the absorption produced by the other beam if the laser frequency ω is tuned close to the line center Ω_{ab} of one of the Doppler-broadening transitions. Both light waves then interact with the same molecules, those within the narrow range of axial velocity centered at $v_x = 0$.

For moderate light intensity and large Doppler width a simple hole-burning model is adequate to describe the transmission increase. Such a model predicts that the change of the partial absorption coefficient α_{ab} for the probe beam due to a saturating beam of intensity I is

$$\frac{\Delta\alpha_{ab}}{\alpha_{ab}} = -\frac{1}{2} \frac{I}{I_{\text{sat}}} \frac{\gamma_{ab}^2}{\gamma_{ab}^2 + (\Omega_{ab} - \omega)^2}, \quad (1)$$

where γ_{ab} is the natural linewidth of the transition,⁷ and

$$I_{\text{sat}}^{-1} = \frac{4\pi}{\hbar^2 c} \frac{|\mu_{ab}|^2}{3} \gamma_{ab}^{-1} (T_a + T_b - A_{ab} T_a T_b). \quad (2)$$

The saturation parameter I_{sat} depends on the matrix element μ_{ab} of the transition, on the lifetimes of the two levels involved, T_a and T_b , and on the rate A_{ab} of the direct and cascade spontaneous transitions connecting the upper and lower states.

The iodine molecule possesses several possible transitions with slightly different resonance frequencies. As long as these transitions are independent, the observed saturation spectrum will be merely a linear superposition of resonance

lines as given by Eq. (1).

The geometry of the experimental setup is shown in Fig. 1. The light source was a Coherent Radiation model 52K krypton-ion laser operated on the 568.2-nm line. An intracavity etalon (free spectral range 6.5 GHz) suppressed all but a single axial mode. The laser mode structure was continuously monitored by a scanning Fabry-Perot interferometer.

The output of the laser was split into two counterpropagating beams^{8,9} of 10 mW each, which crossed at a small angle (<2 mrad) within the 20-cm-long iodine-vapor cell. The calculated spot size within the cell was $w_0 = 0.34$ mm. The vapor was in equilibrium with the solid at a temperature of $22 \pm 1^\circ\text{C}$. At the corresponding vapor pressure of 250 ± 20 mTorr, 45% of the laser light was absorbed at the line center. The modulation of the probe beam, synchronous to the chopper frequency, was measured by means of a silicon photoelement and a phase-sensitive detector.

The laser frequency was scanned by applying a linear voltage ramp to a piezoelectric translator supporting the front mirror. The angle of the etalon was varied simultaneously to maximize the output at each frequency. In this manner the entire 5500-MHz gain band of the laser transition could be scanned in a few minutes. Thermal drifts were minimized by stabilizing the etalon temperature within a few millidegrees and by allowing the laser cavity to reach an equilibrium temperature before performing the experiment.

The spectra appeared as plots of the ac signal versus tuning voltage. The calibration of the frequency scale was confirmed both before and after each measurement by comparison with the known separation of the axial modes of the laser cavity. This was accomplished by applying the voltage ramp to the front mirror, leaving the etalon angle fixed. A particular iodine resonance structure appearing in successive orders was used as

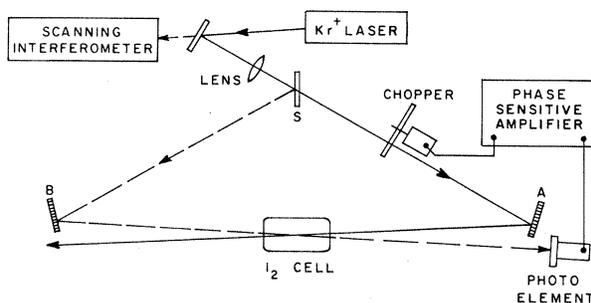


FIG. 1. The saturation spectrometer.

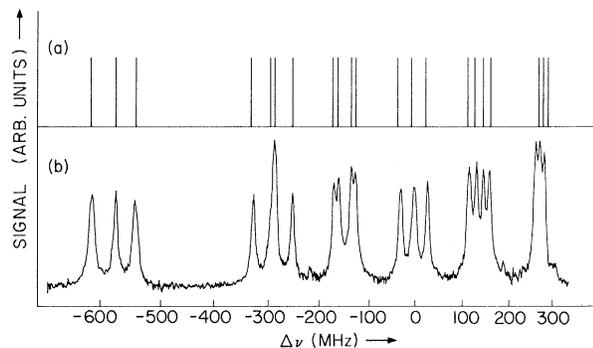


FIG. 2. Hyperfine structure of molecular iodine. (a) Theoretical spectrum with the coupling constants $\Delta e Qq = 940$ MHz and $C = 53$ KHz. (b) The observed structure of the $P(117) 21-1 B \leftarrow X$ transition. Except for the unresolved doublet peak at -290 MHz, the variation in peak signal intensity results from variations in laser power during the scan.

a frequency marker. Residual drifts limited the accuracy to 3%.

A typical experimental spectrum is shown in Fig. 2(b). 21 components are clearly visible, implying that the J value of the lower state is odd.⁶ At the maximum signal level the modulation of the probe beam at the chopper frequency was

$$H_{\text{hfs}} = \sum_{i=1}^2 \left\{ eQq \frac{3(\vec{I}_i \cdot \vec{J})^2 + \frac{3}{2}\vec{I}_i \cdot \vec{J} - |\vec{I}|^2 \cdot |\vec{J}|^2}{2J(2J-1)I_i(2I_i-1)} + C\vec{I}_i \cdot \vec{J} \right\}. \quad (3)$$

The first term within the braces describes to first order the interaction energy between the nuclear electric quadrupole moment Q and the effective field gradient at the nucleus due to the molecular charges. The second term gives the interaction energy between the nuclear magnetic moment and the molecular magnetic field.

At large J values, the hyperfine splitting of a molecular state can be understood using a rather simple model: The rotational angular momentum may be treated as having a fixed direction in space, and the orientations of the nuclei are quantized independently along this same axis. In the classical limit ($J \rightarrow \infty$) the states $m_i = \pm|m_i|$ would be degenerate for a nuclear electric quadrupole interaction. For finite J values, however, the states with positive m_i are split slightly from those with negative m_i . An additional larger splitting results from the magnetic interaction. Since the orientation of the nuclei will remain unchanged in a radiative transition (selection rules $\Delta m_1 = \Delta m_2 = 0$), the splitting of a resonance line exhibits essentially the same structure as the splitting of the levels. The hyperfine structure

of the present resonance line may then be interpreted as follows: The outer lines of the triplet groups, centered near -580 , 0 , and 280 MHz, correspond to the nuclear orientation of $m_1 = m_2 = \pm\frac{5}{2}$, $\pm\frac{3}{2}$, and $\pm\frac{1}{2}$, respectively. The positions of the remaining lines are predicted at the average frequencies of each pair of these lines.

A more exact calculation of the hyperfine splitting is obtained according to Ref. 6 by calculating the matrix elements of H_{hfs} in the I_1, I_2, I, J, F, M_F representation and computing the eigenvalues which are then labeled by the quantum numbers ϵ, J, F . The selection rules become $\Delta\epsilon = 0$, $\Delta J = \Delta F = \pm 1$ in this treatment, and symmetry requires that J and I be both even or both odd for an even electronic state, such as the iodine ground state. This symmetry requirement is reversed for odd electronic states.

A theoretical spectrum, calculated in this way, is shown in Fig. 2(a). The zero point of the frequency scale was chosen according to the convention of Hanes and Dahlstrom.⁵ Two parameters were adjusted to fit the experimental line separa-

tion: The difference of nuclear electric quadrupole coupling constants for the two states is $\Delta eQq = -940 \pm 30$ MHz, and the magnetic coupling in the excited state is¹² $C = 53 \pm 5$ KHz.

The difference in quadrupole coupling constants, ΔeQq , is nearly the same for both the presently studied $P(117)$ line of the 21-1 band and the formerly observed $R(127)$ line of the 11-5 band,⁶ despite widely differing vibrational states. An estimate of the ground-state quadrupole coupling, extrapolated from heteronuclear molecules,¹ indicates that the hyperfine splitting in the electronically excited state is about four times smaller than in the lower state.

The experimentally determined spectrum agrees quite well with the prediction of the model. The present measurements, with their complete coverage, well-defined zero level, and good signal-to-noise ratio, reveal no structures other than the predicted 21 components. This confirms that the transitions where the nuclear spin orientations change do not occur and proves, moreover, that there is negligible cross relaxation between different molecular hyperfine levels.⁴ The saturation techniques utilized would also reveal a possible collisional cross relaxation in velocity space, as was demonstrated in a recent experiment investigating gas-laser plasma.⁸

The strict selection rules simplify the spectrum, but they also make it impossible to determine the hyperfine splittings of the levels separately: Only the difference is observed in experiments of this kind. However, the individual level spacings could be determined by applying a radio-frequency field to mix hyperfine levels.

The strong, narrow, iodine-vapor resonances reported in this paper may be used for long-term frequency stabilization of the krypton ion laser. Such a light source, with its excellent frequency

stability combined with significant output power, will certainly prove a very useful tool for a variety of applications.

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Anomalous Rapid Skin-Current Penetration and Heating in Pulsed-Plasma Experiments*

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Electrostatic instabilities in a plasma having electrons flowing along a magnetic field are generated by a shear in the electron drift velocity. This can cause an effective resistivity much higher than the Buneman value and consequently increase the rates of penetration of a skin current and of heating.

To explain the anomalous resistivity observed in several turbulent heating experiments,¹ the growth rate of the electron-ion two-stream instability^{2,3} has been interpreted as an effective

electron collision frequency. To get agreement between the observed conductivity¹ and the theory,^{2,3} the effective ion-electron collision time has been assumed equal to the e -folding growth