

Cross section and hyperfine structure of the atomic iodine ($^2P_{1/2}$ - $^2P_{3/2}$) Raman transition

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The cross section for anti-Stokes Raman scattering from inverted atomic iodine is calculated by a semiempirical procedure and shown to be dominated by the quadrupole component. Relative intensities for F_2 - F_1 transitions (F_2 and F_1 are the total angular momenta of the initial and final hyperfine split states) in the hyperfine split multiplets are calculated, and maximal intensity of 25% is found for the 2-4 transition. However, in stimulated Raman scattering maximal gain is expected at the position of the 3-2 line, where collisional broadening and Stark splitting lead to a strong overlap of three lines. The resulting effective cross section is about 33% of the total cross section. For scattering with pump radiation at $\lambda=1.06\ \mu\text{m}$ an effective cross section of $d\sigma/d\Omega=0.71\times 10^{-28}\ \text{cm}^2$ is obtained, with an estimated accuracy of 20%. This is somewhat lower than the estimate $d\sigma/d\Omega=10^{-28}\ \text{cm}^2$ which has been published recently.

I. INTRODUCTION

Electronic Raman scattering from atomic iodine is of interest both for theoretical and practical reasons. The latter include the use of stimulated anti-Stokes Raman scattering for frequency up-conversion of high-power lasers,^{1,2} and recent proposals to increase the gain in the iodine laser system by the technique of collision-induced transitions.³ In spontaneous and stimulated Raman scattering important parameters like transition rate, cross section, and gain in inverted media can be calculated in terms of the scattering tensor⁴ for the system considered. Estimates of the Raman cross section for atomic iodine have been published before,¹ however the approximations made in such estimate were not made explicit. Furthermore, it was not apparent to what extent the hyperfine structure of the atomic iodine Raman line and the effects of Stark splitting had been taken into account. While the validity of both the up-conversion scheme as such and the order of magnitude for the cross-section estimates have been demonstrated experimentally² recent plans for more detailed experimental study⁵ made it desirable to reconsider the theoretical work done including the points mentioned above.

In this paper we estimate the scattering tensor for atomic iodine by a semiempirical procedure, including both the electric quadrupole and the magnetic dipole component. This estimate employs both experimental line-strength values and general sum rules for those contributions where no experimental

values are available.

In Sec. I we derive a general formula for the reduced matrix elements of the atomic scattering operator valid for all three components (trace, magnetic dipole, and quadrupole component), and show how to evaluate these in terms of single-electron radial integrals for the special case of the ($^2P_{1/2}$ - $^2P_{3/2}$) transition of atomic iodine. General formulas of a similar type have been developed by Vriens⁶ for polarized and depolarized scattering. For our purpose however, the distinction between the three irreducible scattering components is more appropriate since they are associated with different types of selection rules and different relative intensities in hyperfine split multiplets.

In Sec. II we give quantitative estimates for both the electric quadrupole and the magnetic dipole component, showing that the latter is negligible for pump frequencies of the order of magnitude of the ground-state fine-splitting interval. In Sec. III we calculate relative intensities for F_2 - F_1 transitions (F_2 and F_1 are the total angular momentum of the initial and final hyperfine split states) in the hyperfine split multiplets. All levels intercombine, and the 2-4 component is found to be strongest with a relative intensity of 25%. However, due to collisional broadening and Stark splitting maximal gain in an actual anti-Stokes experiment is expected at the position of the 3-2 line, where the three closely neighboring 3-1, 3-2, and 3-3 lines with a total relative intensity of 46% strongly overlap. This and possible sources of error involved in our approximation are discussed in the final Section.

I. SCATTERING TENSOR AND LINE STRENGTH FOR RAMAN SCATTERING

The transition rate dW_{21} for spontaneous anti-Stokes Raman emission into the solid angle $d\Omega$ at frequency $\nu = \nu_0 + \nu_{21}$ between atomic levels 1 and 2 is given by (Ref. 4, Secs. 45 and 60)

$$dW_{21} = \frac{(2\pi\nu)^3}{hc^3} \frac{1}{2J_2 + 1} \times \sum_{m_1 m_2} \left| \sum_{\sigma\rho} e_\sigma A_\rho \alpha_{\sigma\rho}(m_1 m_2) \right|^2 d\Omega, \quad (1.1)$$

where J_i is the angular momentum of level i ($i = 1, 2$), e_σ is the component of polarization vector of emitted radiation ($\sigma = 1, 2, 3$), and A_ρ is the component of amplitude of incoming radiation field $F_\rho(t)$ ($\rho = 1, 2, 3$),

$$F_\rho(t) = A_\rho e^{2\pi i \nu_0 t} + A_\rho e^{-2\pi i \nu_0 t} \\ = 2A_\rho \cos 2\pi \nu_0 t,$$

$\alpha_{\sigma\rho}(m_1 m_2)$ is the scattering tensor for the transition considered:

$$\alpha_{\sigma\rho}(m_1 m_2) = \sum_r \frac{\langle 1J_1 m_1 | M_\rho | r \rangle \langle r | M_\sigma | 2J_2 m_2 \rangle}{E_r - E_2 - E_0} + \frac{\langle 1J_1 m_1 | M_\sigma | r \rangle \langle r | M_\rho | 2J_2 m_2 \rangle}{E_r - E_1 + E_0}. \quad (1.2)$$

$$\sum_{m_1 m_2} \left| \sum_{\sigma\rho} e_\sigma A_\rho \alpha_{\sigma\rho}(m_1 m_2) \right|^2 = [R^{(1)} \frac{1}{2} \sin^2 \theta + R^{(2)} \frac{1}{2} (1 + \frac{1}{3} \cos^2 \theta)] \sum_\rho A_\rho A_\rho. \quad (1.4)$$

The coefficients $R^{(k)}$ are the Raman scattering analog of the line strength in ordinary dipole radiation. They are related to the reduced matrix elements of the scattering operator:

$$R^{(k)} = \frac{1}{2k+1} \left| \langle 1J_1 || \hat{\alpha}^k || 2J_2 \rangle \right|^2. \quad (1.5)$$

From Eqs. (1.1) and (1.4) we obtain the differential cross section for linearly polarized Raman scattering:

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^4 \lambda^{-4}}{(2J_2 + 1)} [R^{(1)} \frac{1}{2} \sin^2 \theta + R^{(2)} \frac{1}{2} (1 + \frac{1}{3} \cos^2 \theta)] \\ = \sigma^{(1)} \frac{1}{2} \sin^2 \theta + \sigma^{(2)} \frac{1}{2} (1 + \frac{1}{3} \cos^2 \theta). \quad (1.6)$$

The reduced matrix element of the scattering operator can be calculated from Eq. (1.2) in a straightforward manner by using standard tensor operator techniques [Ref. 7, p. 77 (4.120), p. 69 (4.88)]

$$\langle 1J_1 || \hat{\alpha}^k || 2J_2 \rangle = \sqrt{2k+1} (-1)^{J_1 + J_2 + k} \sum_r \langle 1J_1 || M^1 || rJ_r \rangle \langle rJ_r || M^1 || 2J_2 \rangle \begin{Bmatrix} J_2 & J_1 & k \\ 1 & 1 & J_r \end{Bmatrix} \\ \times \left[\frac{1}{E_r - E_2 - E_0} + (-1)^k \frac{1}{E_r - E_1 + E_0} \right]. \quad (1.7)$$

$E_r = h\nu_r$ and $E_i = h\nu_i$, $i = 1, 2$ are the energies of the atomic levels r and 1, 2 and $E_0 = h\nu_0$ is the energy of the incoming photon. M_ρ are the components of the dipole-moment operator. Here and in the following equations the labels 1 and 2 in matrix elements stand for all but angular momentum quantum numbers labeling the atomic states involved in the transition. The label r for the intermediate state stands for all quantum numbers, including angular momentum.

The scattering tensor $\alpha_{\sigma\rho}(m_1 m_2)$ may be considered as the matrix element of some tensor operator $\hat{\alpha}_{\sigma\rho}$ between the atomic states involved:

$$\alpha_{\sigma\rho}(m_1 m_2) = \langle 1J_1 m_1 | \hat{\alpha}_{\sigma\rho} | 2J_2 m_2 \rangle. \quad (1.3)$$

The tensor operator $\hat{\alpha}_{\sigma\rho}$ decomposes into irreducible components $\hat{\alpha}^k$ of rank $k = 0, 1$, and 2. Traditionally these are called the trace ($k = 0$), magnetic dipole ($k = 1$), and quadrupole ($k = 2$) component:

$$\hat{\alpha}_{\sigma\rho}^0 = \delta_{\sigma\rho} \frac{1}{3} \sum_\tau \hat{\alpha}_{\tau\tau}, \\ \hat{\alpha}_{\sigma\rho}^1 = \frac{1}{2} (\hat{\alpha}_{\sigma\rho} - \hat{\alpha}_{\rho\sigma}), \\ \hat{\alpha}_{\sigma\rho}^2 = \frac{1}{2} (\hat{\alpha}_{\sigma\rho} + \hat{\alpha}_{\rho\sigma}) - \hat{\alpha}_{\sigma\rho}^0.$$

In our case $J_1 \neq J_2$ and the trace component does not contribute because of the selection rule $\Delta J = 0$. The magnetic dipole and quadrupole component give contributions to the rate dW which may be distinguished by their dependence on the angle θ between the directions of linear polarization of the incoming and the scattered radiation (Ref. 4, Sec. 61):

Here $\langle ||M^1|| \rangle$ are the reduced matrix elements of the electric dipole moment operator and $\{ \}$ is a $6j$ symbol.

Let us now apply the general formula (1.7) to Raman transitions in the ground-state fine-structure splitting multiplet of atomic iodine. In this case

$$J_1 = \frac{3}{2}, \quad J_2 = \frac{1}{2}, \quad E_2 - E_1 = 6.93 \times 10^{-2} \text{ Ry}$$

The first resonance level contributing to the sum (1.7) over intermediate states is at an energy of $E_2 - E_1 = 0.511 \text{ Ry}$. On the other hand, both the fine-structure splitting intervals in LS multiplets and the differences in term values belonging to the same electron configuration are of the same order of magnitude as the ground-state fine-structure splitting $E_2 - E_1$. We shall hence neglect these differences in the energy denominators of Eq. (1.7). This allows us to use LS wave functions and hence to express the reduced transition matrix element of the scattering operator by a diagonal matrix element for the ground state LS wave function ($L = 1, S = \frac{1}{2}$)

$$\begin{aligned} |\langle 1J_1 || \hat{a}^k || 2J_2 \rangle|^2 &= |\langle L || \hat{a}^k || L \rangle|^2 (2J_1 + 1)(2J_2 + 1) \begin{Bmatrix} L & J_1 & S \\ J_2 & L & k \end{Bmatrix}^2 \\ &= \frac{2}{9} |\langle L || \hat{a}^1 || L \rangle|^2 \quad k=1 \\ &= \frac{2}{3} |\langle L || \hat{a}^2 || L \rangle|^2 \quad k=2. \end{aligned} \quad (1.8)$$

Furthermore, by summing over all states corresponding to the same electron configuration we can express the dipole-moment matrix element in terms of one-electron radial integrals:

$$\begin{aligned} \langle L || \hat{a}^k || L \rangle &= 5\sqrt{2k+1} \sum_{\beta > \alpha} \langle \alpha j_\alpha || M^1 || \beta j_\beta \rangle \langle \beta j_\beta || M^1 || \alpha j_\alpha \rangle \\ &\quad \times \begin{Bmatrix} j_\alpha & j_\alpha & k \\ 1 & 1 & j_\beta \end{Bmatrix} \left[\frac{1}{E_\beta - E_\alpha - E_0} + (-1)^k \frac{1}{E_\beta - E_\alpha + E_0} \right], \end{aligned} \quad (1.9)$$

$$\begin{aligned} \langle \alpha j_\alpha || M^1 || \beta j_\beta \rangle &= \begin{cases} \sqrt{j_\alpha} R_\alpha^\beta & j_\beta = j_\alpha - 1 \\ -\sqrt{j_\alpha + 1} R_\alpha^\beta & j_\beta = j_\alpha + 1, \end{cases} \\ R_\alpha^\beta &= e \int_0^\infty dr r \bar{\chi}_\alpha(r) \chi_\beta(r). \end{aligned} \quad (1.10)$$

Here α and β label the states of the shell and the excited optical electron with radial wave functions χ_α and χ_β and angular momenta $j_\alpha = 1$ and $j_\beta = j_\alpha \pm 1$. We can restrict our attention to electrons in the open $5p$ shell, since electrons in closed shells do not contribute to the ($k = 1$) and ($k = 2$) components.

II. QUANTITATIVE RESULTS FOR THE ATOMIC IODINE (${}^2P_{1/2} - {}^2P_{3/2}$) TRANSITION

We shall now estimate the Raman line strength for the atomic iodine (${}^2P_{1/2} - {}^2P_{3/2}$) transition from

Eq. (1.9). In this case

$$\begin{aligned} \langle \alpha j_\alpha | &= \langle 5p | \\ \langle \beta j_\beta | &= \begin{cases} \langle ns |, & j_\beta = 0 \\ \langle nd |, & j_\beta = 2; n > 5 \end{cases} \end{aligned}$$

Here we use the symbol n for the radial quantum numbers labeling states both in the discrete and the continuous spectrum.

Let us first treat the quadrupole component ($k = 2$). Equation (1.9) then gives

$$\begin{aligned} \langle L || \hat{a}^2 || L \rangle &= -\frac{5\sqrt{5}}{3} \sum_{n > 5} (R_{5p}^{ns})^2 \left[\frac{1}{E_{ns} - E_{5p} - E_0} + \frac{1}{E_{ns} - E_{5p} + E_0} \right] \\ &\quad - \frac{\sqrt{5}}{12} \sum_{n > 5} (R_{5p}^{nd})^2 \left[\frac{1}{E_{nd} - E_{5p} - E_0} + \frac{1}{E_{nd} - E_{5p} + E_0} \right]. \end{aligned} \quad (2.1)$$

The radial integrals in Eq. (2.1) satisfy the sum rule (Ref. 8, p. 257):

$$\sum_n (R_{5p}^{ns})^2 = \sum_n (R_{5p}^{nd})^2 = \int_0^\infty dr r^2 |\chi_{5p}(r)|^2 = 7.20. \quad (2.2)$$

All numerical values given are in atomic units, unless stated otherwise. Radial integrals for core and shell electrons have been adopted from or calculated from Hartree-Fock wave functions published in Ref. 9. Calculations show that contributions to the sum rule (2.2) from the core electrons are negligible for d electrons and are almost exclusively due to the $5s$ electrons. For the radial integral of the $5p-6s$ transition the experimental value of Ref. 10 has been adopted. The figures obtained are given in Table I.

It is apparent from these figures that the $6s$ resonance states contribute about 60% of all excited ns states ($n > 5$) to the sum rule (2.2). In estimating the sum (2.1) we shall hence calculate this contribution separately. For the higher s states and for all excited d states we shall replace the energy denominators by a fixed average energy ΔE_s and ΔE_d . These energies are calculated from the oscillator-strength sum rule as $\Delta E_s = 2.46$ Ry, $\Delta E_d = 0.69$ Ry. Finally we consider the case where the energy E_0 of the incoming photon is small compared to the resonance energy and can hence be neglected in the energy denominator. In this approximation we obtain

$$\begin{aligned} \langle L || \alpha^2 || L \rangle = & -\frac{5\sqrt{5}}{3} \left[(R_{5p}^{6s})^2 \frac{1}{E_{6s} - E_{5p}} \right. \\ & \left. + \sum_{n>6} (R_{5p}^{ns})^2 \frac{1}{\Delta E_s} \right] \\ & - \frac{\sqrt{5}}{12} \sum_{n \geq 5} (R_{5p}^{nd})^2 \frac{1}{\Delta E_d} \\ = & -27.7 \end{aligned} \quad (2.3)$$

in atomic units. The magnetic dipole component ($\kappa=1$) vanishes in the limit $E_0 \rightarrow 0$ which corresponds to a static electric field. For small E_0 (see above) we expand the energy denominator up to first order in E_0 and obtain

$$\begin{aligned} \langle L || \alpha^1 || L \rangle = & E_0 \frac{10\sqrt{3}}{3} \left[\sum_{n>5} (R_{5p}^{ns})^2 \frac{1}{(E_{ns} - E_{5p})^2} \right. \\ & \left. - \sum_{n \geq 5} (R_{5p}^{nd})^2 \frac{1}{(E_{nd} - E_{5p})^2} \right] \\ = & -E_0 53.5 \end{aligned} \quad (2.4)$$

TABLE I. Squares of radial integrals.

nl	$5s$	$6s$	ns ($n > 6$)	nd ($n > 5$)
$(R_{5p}^{nl})^2$	4.84	1.44	0.92	7.20

in atomic units. Here the same approximations have been used as in the calculation of the quadrupole component. Note that the energy E_0 of the incoming photon has to be expressed in Ry. For an energy of $E_0 = 5 \times 10^{-2}$ Ry which is about 10% of the resonance energy the magnetic dipole component gives less than 10% of the quadrupole contribution, which is of the same order of magnitude as the error involved in our approximations.

We shall therefore neglect the magnetic dipole component altogether. From Eqs. (1.5), (1.8), and (2.3) we obtain the coefficient $\sigma^{(2)}$ in expression (1.6) for the cross section

$$\sigma^{(2)} = (2\pi)^4 \lambda^{-4} (\text{cm}^{-4}) 4.48 \times 10^{-48} \text{ cm}^2. \quad (2.5)$$

For anti-Stokes Raman scattering of light at the neodymium-laser wavelength ($\lambda_0 = 1.06 \mu\text{m}$) (Ref. 1) with scattered radiation at a wavelength of $\lambda = 0.59 \mu\text{m}$ we obtain an (energy scattering) cross section for $\theta = 0$ (in cm^2)

$$\frac{d\sigma}{d\Omega} = 3.84 \times 10^{-28} \text{ cm}^2.$$

The corresponding photon number cross section is given by (in cm^2)

$$\left[\frac{d\sigma}{d\Omega} \right]_N = \frac{d\sigma}{d\Omega} \frac{\lambda}{\lambda_0} = 2.14 \times 10^{-28} \text{ cm}^2.$$

III. SELECTION RULES AND RELATIVE INTENSITIES IN THE HYPERFINE SPLIT MULTIPLET

Owing to the spin $I = \frac{5}{2}$ of the I^{127} nucleus the ${}^2P_{3/2}$ and ${}^2P_{1/2}$ levels split into sublevels classified by the total angular momentum F (nuclear spin plus electronic angular momentum).^{7,11} The levels obtained and their energy shift ΔE (in wave numbers cm^{-1}) are given in Table II below.

The transition rate between sublevels F_2 and F_1 is proportional (Ref. 7, p. 84):

$$\frac{1}{2F_2+1} |\langle J_1 I F_1 || \hat{\alpha}^2 || J_2 I F_2 \rangle|^2 = \frac{1}{2J_2+1} |\langle J_1 || \hat{\alpha}^2 || 2J_2 \rangle|^2 (2J_2+1)(2F_1+1) \left\{ \begin{matrix} J_1 & F_1 & I \\ F_2 & J_2 & 2 \end{matrix} \right\}^2, \quad (3.1)$$

where $\{ \}$ is the $6j$ symbol. In our case $F_1, F_2 \geq 2$, and the selection rule associated with this transition is

$$\Delta F = 0, \pm 1, \pm 2 \quad (3.2)$$

showing that all sublevels intercombine. It follows that the iodine Raman line splits into eight hyperfine components in contrast to only six components for the luminescence line.

From (3.1) we find the relative intensity $q(F_2 F_1)$ of the $F_2 - F_1$ transition as observed in a scattering experiment:

$$q(F_2 F_1) = \frac{(2F_2+1)(2F_1+1)}{2I+1} \left\{ \begin{matrix} J_1 & F_1 & I \\ F_2 & J_2 & 2 \end{matrix} \right\}^2. \quad (3.3)$$

These relative intensities obey the sum rule

$$\sum_{F_1} q(F_2 F_1) = \frac{2F_2+1}{(2J_2+1)(2I+1)}, \quad (3.4)$$

where the right-hand side of (3.4) is just the statistical weight of the level F_2 .

Numerical values from (3.3) and for the relative intensities of the dipole line are summarized in Table III below. Both the Raman and the dipole spectrum are shown in Fig. 1.

We see from this that the 3-4 component which is strongest in the luminescence line is of only average strength in the Raman line. The strongest feature in the Raman line on the other hand is the 2-4 component which is completely absent in the luminescence line. It is interesting to note that this component also undergoes the strongest frequency shift.

TABLE II. Hyperfine split levels of the 2P_J ground-state multiplet (energy ΔE in cm^{-1} relative to the line center).

F_1	$^2P_{3/2}$	ΔE_1	F_2	$^2P_{1/2}$	ΔE_2
1		-0.118			
2		-0.094	2		-0.371
3		-0.028	3		+0.265
4		+0.113			

IV. DISCUSSION

From Table III we conclude that maximal gain in stimulated anti-Stokes Raman scattering will occur at the frequency of the 3-2 line, shifted by $\Delta E = +0.36 \text{ cm}^{-1}$ from the center of the hyperfine split multiplet. Although the 2-4 component is largest in relative intensity, the 3-3, 3-2, and 3-1 component will strongly overlap in a typical experiment² with collisional linewidth of about 0.05 cm^{-1} . These overlapping lines then contribute about 33% to the total cross section, yielding an effective cross section of

$$\left[\frac{d\sigma}{d\Omega} \right]_{\text{eff}} = 0.33 \frac{d\sigma}{d\Omega} = 0.71 \times 10^{-28} \quad (4.1)$$

in cm^2 , which is in fair agreement with estimates published previously.¹ Owing to Stark splitting of the levels this situation prevails even at low pressures where Doppler broadening is dominant with a linewidth of $0.01 - 0.02 \text{ cm}^{-1}$. At a pump intensity of 10^{10} W/cm^2 Stark splittings up to 0.05 cm^{-1} occur, leading to strong overlap of the lines considered.

This is in contrast to the normal quadrupole spectrum as observed both in emission¹² and absorption.¹³ There the 2-4 line gives the strongest contribution, and is in fact the only line which has been resolved so far.

Let us finally remark that our estimate (4.1) of the effective cross section for pumping at the neodymium laser wavelength ($\lambda = 1.06 \mu\text{m}$) is 30% below the estimate presented in Ref. 1. Possible

TABLE III. Relative intensities in the Raman and the dipole spectrum.

$F_2 - F_1$	$\Delta E_{21} (\text{cm}^{-1})$	$q(F_2 F_1)_{\text{Raman}}$	$q(F_2 F_1)_{\text{Dipole}}$
2-4	-0.484	0.250	
2-3	-0.343	0.117	0.130
2-2	-0.277	0.042	0.162
2-1	-0.253	0.008	0.125
3-4	+0.152	0.125	0.375
3-3	+0.293	0.175	0.162
3-2	+0.359	0.167	0.046
3-1	+0.383	0.117	

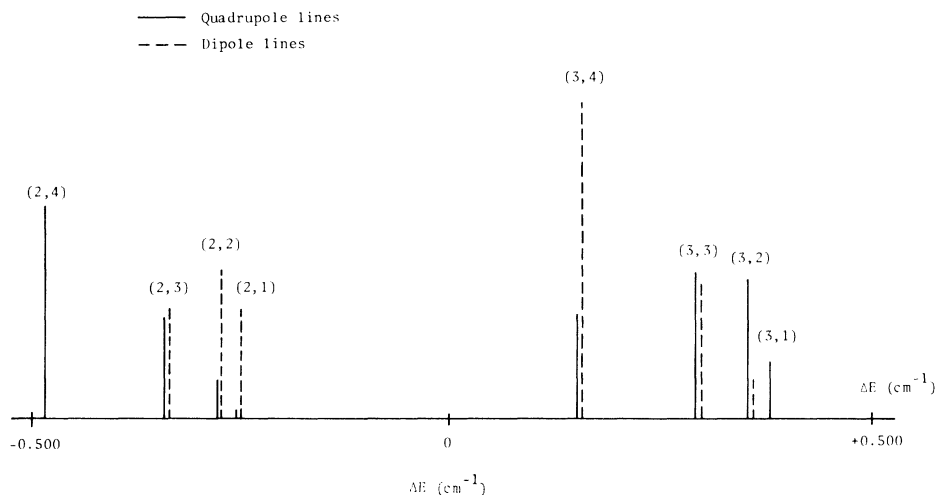


FIG. 1. Atomic iodine ($P_{1/2} - P_{3/2}$) hyperfine Raman spectrum.

sources of error in our calculation are the approximations involved, and the actual numerical values used for radial integrals. The fine-splitting differences we have neglected in the energy denominators are about 10% of the term values¹⁴; energy differences in the $6s$ multiplet are of the same order of magnitude. However the actual error is likely to be smaller, since most of the relative intensity for transition to the ground-state multiplet is concentrated on the two resonance states.

Finally the value of the radial integral for the $6s$ transitions enters both into the contribution of the $6s$ intermediate state and of the higher s states. This leads to partial cancellation of uncertainties in the experimental value used, which are quoted to be less than 9%.¹⁰

Our method of summing over the higher excited states and the d states by using average energies from the oscillator strength sum rule gives a lower bound to these contributions. Results from calculations of polarizabilities show that such an estimate usually comes within less than 20% of values obtained from more elaborate calculations. In our case these states contribute about 25% to the total Raman line strength, which reduces the weight of the error introduced by this procedure. From these considerations we expect an accuracy of about 20% for our estimates (2.5) and (4.1).

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