1. Introduction
The phenomenon that has come to be known as the Ring effect was first noted by Grainger and Ring\textsuperscript{1} as a filling in (broadening and reduction of depth) of solar Fraunhofer lines when viewed from the ground in scattered sunlight. Various processes have been proposed as contributing to the effect, including scattering with fluorescence from aerosols and from the ground.\textsuperscript{2,3} However, the predominance of molecular scattering as the major cause was established by Kattawar \textit{et al.},\textsuperscript{4} who analyzed the Ring effect contributions from rotational Raman scattering and inelastic Rayleigh–Brillouin scattering. The Rayleigh–Brillouin component is not of primary importance in atmospheric scattering situations. The atmospheric scattering situation is nicely defined by Young\textsuperscript{5}: “To summarize: molecular scattering consists of Rayleigh scattering and vibrational Raman scattering. The Rayleigh scattering consists of rotational Raman lines and the central Cabannes line. The Cabannes line is composed of the Brillouin doublet and the central Gross or Landau–Placzek line. None of the above is completely coherent. The term ‘Rayleigh line’ should never be used.” Note that the vibrational Raman contribution results in lines so widely separated from the frequency of the incoming light that they are not normally considered part of the Ring effect, although in recent applications the Ring effect has developed a somewhat broader definition that includes substantial interfering structure in observations, rather than the initial effect that was limited to the broadening of partially resolved lines.

Understanding the Ring effect has become more important in recent years with the increase in ultraviolet and visible spectroscopic observations of the Earth’s atmosphere from the ground\textsuperscript{6,7} and from satellites.\textsuperscript{8–12} To retrieve abundances of trace species from such observations, it is necessary to take the Ring effect into account. Methods have been developed to do so by pragmatic means: by measuring the polarization of scattered sunlight\textsuperscript{6} and by modeling the effect directly from molecular scattering processes.\textsuperscript{7,10} It has been proposed to use the Ring effect with selected Fraunhofer lines (in particular, the Ca II H and K lines) to determine cloud parameters in conjunction with satellite-based observations of $O_3$.\textsuperscript{11} Joiner \textit{et al.}\textsuperscript{10} also determined that the contribution to the Ring effect from the Rayleigh–Brillouin scattering process is negligible for most geometries used in satellite observations.

The use of visible bands of $O_2$, in particular the 762-nm $A$ band, for determination of cloud parameters is being developed for proposed and existing satellite instruments that monitor the atmosphere with
emphasize on tropospheric measurements. This includes the European Space Agency’s global ozone monitoring experiment (GOME) and the upcoming scanning imaging absorption spectrometer for atmospheric chartography (SCIAMACHY). The Ring effect has a substantial influence on such observations; work is under way in our institution to refine and model the effects. In this case, the effect is due to inelastic scattering in molecular absorption lines themselves rather than in the solar Fraunhofer lines. Similar effects have been noted for the detailed retrieval of trace species including O₃ and NO₂.6,7,12

To examine the detailed Rayleigh and rotational Raman scattering properties, including their relative intensities and the scattering phase functions, we begin with Table I of Kattawar et al.,4 which describes the relative intensities and angular behaviors for the Rayleigh–Brillouin and rotational components and their sum. They are given for various input polarizations, including unpolarized light (the predominant contribution for many atmospheric observations for which single scattering is the major contributor to the Ring effect). For unpolarized light, the depolarization ratios (defined in each case as the ratio of the horizontally polarized component to the vertically polarized component at a 90° scattering angle) can be determined directly. Table I of Kattawar et al.4 is reproduced here as Table 1, with the addition of depolarization ratios for three cases: Rayleigh–Brillouin (the central Cabannes component C); rotational Raman (the wings W); and the sum of the two (T for total). The phase functions for scattering can also be derived for each case. They are (normalized over solid angle to 1)

\[
\Phi_0^C = \frac{3}{160\pi} \left[ \frac{(180 + 13\epsilon) + (180 + \epsilon)\cos^2\theta}{18 + \epsilon} \right],
\]

\[
\Phi_0^W = \frac{3}{160\pi} (13 + \cos^2\theta),
\]

\[
\Phi_0^T = \frac{3}{80\pi} \left[ \frac{(45 + 13\epsilon) + (45 + \epsilon)\cos^2\theta}{9 + 2\epsilon} \right],
\]

where \(\epsilon = \left(\gamma \alpha\right)^2\), \(\gamma\) is the anisotropy of the polarizability, and \(\alpha\) is the average polarizability. In each

### Table I. Relative Rayleigh and Raman Scattering Intensities

<table>
<thead>
<tr>
<th>V Polarization in</th>
<th>H Polarization in</th>
<th>Sum (Natural Light in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rayleigh–Brillouin</td>
<td>(^{\nu}C_V = 3\epsilon)</td>
<td>(^{\nu}C_V = 180 + 7\epsilon)</td>
</tr>
<tr>
<td>(^{\nu}C_H = 3\epsilon)</td>
<td>(^{\nu}C_H = 6\epsilon + (180 + \epsilon)\cos^2\theta)</td>
<td></td>
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<tr>
<td>(^{\nu}C_0 = 6\epsilon + (180 + \epsilon)\cos^2\theta)</td>
<td>(^{\nu}C_H = 180 + 13\epsilon)</td>
<td></td>
</tr>
<tr>
<td>Raman</td>
<td>(^{\nu}W_V = 9\epsilon)</td>
<td>(^{\nu}W_V = 21\epsilon)</td>
</tr>
<tr>
<td>(^{\nu}W_H = 9\epsilon)</td>
<td>(^{\nu}W_H = 18\epsilon + 3\epsilon\cos^2\theta)</td>
<td></td>
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<tr>
<td>(^{\nu}W_0 = 21\epsilon)</td>
<td>(^{\nu}W_0 = 39\epsilon + 3\epsilon\cos^2\theta)</td>
<td></td>
</tr>
<tr>
<td>Sum</td>
<td>(^{\nu}T_V = 180 + 16\epsilon)</td>
<td>(^{\nu}T_V = 180 + 28\epsilon)</td>
</tr>
<tr>
<td>(^{\nu}T_H = 12\epsilon)</td>
<td>(^{\nu}T_H = 24\epsilon + (180 + 4\epsilon)\cos^2\theta)</td>
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</tr>
<tr>
<td>(^{\nu}T_0 = 180 + 28\epsilon)</td>
<td>(^{\nu}T_0 = (180 + 52\epsilon) + (180 + 4\epsilon)\cos^2\theta)</td>
<td></td>
</tr>
<tr>
<td>(^{\nu}C_V = 9\epsilon)</td>
<td>(^{\nu}C_H = 6\epsilon + (180 + \epsilon)\cos^2\theta)</td>
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<tr>
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<td>(^{\nu}W_V = 9\epsilon)</td>
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<td>(^{\nu}W_0 = 21\epsilon)</td>
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<tr>
<td>(^{\nu}T_V = 12\epsilon)</td>
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<td>(^{\nu}C_H = 9\epsilon)</td>
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<td></td>
</tr>
</tbody>
</table>

Mostly from Kattawar et al., 1981.
case, the phase function is given in terms of the respective depolarization ratio \( \rho_0 \) \( X = C, W, T \) by

\[
\Phi_x = \Phi_0 x = \frac{3}{8\pi} \left[ \frac{(1 + \rho_0^x) + (1 - \rho_0^x) \cos^2 \theta}{2 + \rho_0^x} \right].
\] (2)

The Rayleigh scattering cross section at standard temperature (273.15 K) and pressure (1 atm) is given by

\[
Q_R = \frac{32\pi^2(n - 1)^2F_K}{3N_0 \lambda^4},
\] (3)

where \( n \) is the index of refraction, \( F_K \) is the King correction factor,\(^{17} \) \( N_0 \) is Loschmidt’s number \((2.686763 \times 10^{19} \text{ cm}^{-3})\), and \( \lambda \) is the wavelength. The King correction factor is given by

\[
F_K = 1 + 2 \left( \frac{\gamma}{3\alpha^3} \right)^2 = \frac{6 + 3\rho_0^n}{6 - \rho_0^n} = 1 + \frac{2\epsilon}{9}.
\] (4)

The average polarizability can be determined from the relation

\[
|\alpha|^2 = \frac{(n - 1)^2}{4\pi^2N_0^2}.
\] (5)

3. Molecular Parameters

A. Rayleigh Scattering Cross Sections

The major source of improved data for the indices of refraction of \( O_2, N_2 \), and air versus wavelength, the King correction factors, and the anisotropies of the polarizabilities is Bates.\(^{16} \) He presented a comprehensive review of both measurements and theoretical calculations and derived a data set that was demonstrated to be better than 1% for all the data and parameterizations utilized in this study.

Bates’s Table 1 gives refractive indices, Rayleigh-scattering cross sections, and King correction factors versus wavelength for air from 200 to 1000 nm. We found that the following Edlen-type expression fits the index of refraction data to better than 0.1% for all values:

\[
(n_{\text{air}} - 1) \times 10^4 = 0.7041 + \frac{315.90}{157.39 - \sigma^2} + \frac{8.4127}{50.429 - \sigma^2},
\] (6)

where \( \sigma(\mu\text{m}^{-1}) = 1/\lambda (\mu\text{m}) \). The Rayleigh cross sections are reproduced to 0.3% rms (worst case, 0.5%) over the full 0.2–1-\( \mu \text{m} \) range by the expression

\[
Q_R \times 10^{24}(\text{cm}^2)
\]

\[
= 3.9993 \times 10^{-4}\sigma^4 \times \frac{(1 - 1.069 \times 10^{-2}\sigma^2 - 6.681 \times 10^{-5}\sigma^4}{1 - 1.069 \times 10^{-2}\sigma^2 - 6.681 \times 10^{-5}\sigma^4}.
\] (7)

Nicolet\(^{18} \) gave an expression for fitting the data of Bates to 0.5% over the 0.2–0.55-\( \mu \text{m} \) range; the current expression is slightly more accurate and extends over a larger wavelength range (also see Bucholtz\(^{19} \)). The King factor and depolarization ratio for any choice of wavelength within the 0.2–1.0-\( \mu \text{m} \) range can be accurately determined from the previous two equations.

B. Polarizability Anisotropies

The previous expansions are for air, including standard amounts of \( \text{Ar} \) and \( \text{CO}_2 \). For rotational Raman cross sections, we limit the calculations to \( O_2 \) and \( N_2 \). Bates gave segmented representations for the indices of refraction of \( O_2 \) and \( N_2 \) versus wavelength and expressions for the respective King correction factors. Tables giving the King correction factors, values of \( \sqrt{\epsilon} \), index of refraction \( \alpha \), and \( \gamma \) for \( O_2 \) and \( N_2 \) have been derived in this study. Values of \( \gamma_{O_2} \) from these data are fitted to 0.6% rms over this wavelength range (1% worst case) by

\[
\gamma_{O_2} \times 10^{24} = 0.07149 + \frac{45.9364}{48.2716 - \sigma^2}.
\] (8)

The \( \gamma_{N_2} \) values are fitted to 0.02% rms (0.04% worst case) by

\[
\gamma_{N_2} \times 10^{25} = -6.01466 + \frac{2385.57}{186.099 - \sigma^2}.
\] (9)

These equations should be considered merely as phenomenological fits over a particular wavelength range, rather than trying to attach physical importance to, for example, the negative value of the constant term in the \( N_2 \) equation. They should not be extrapolated to outside the 0.2–1.0-\( \mu \text{m} \) range.

C. Basic Spectroscopy

The ground state of \( O_2 \) is \( \Sigma_{g}^+ \); it has significant electronic structure in both its magnetic dipole rotational spectral and its rotational Raman spectrum. Rotational Raman spectra of both \( O_2 \) and \( N_2 \) have previously been approximated by simple expansions in the lowest rotational parameters (for positions) and by using \( T/c_B \delta_{0} \) for the rotational partition functions. Precise data are now available for the term energies, allowing line positions and Boltzmann factors to be calculated accurately and rapidly. The use of term values from the current HITRAN listing\(^{20} \) allows for calculations of line positions to 0.0001-\( \text{cm}^{-1} \) accuracy and for highly accurate calculation of statistical partitioning.

D. Placzek–Teller Coefficients

Cross sections for rotational Raman scattering are given by

\[
Q_{N,N'}(cm^2) = \frac{256\pi^5}{27(\lambda)^4} \gamma f_{N'} c_{P,T}(N, J, J', N'),
\] (10)

where \( f_N \) is the fractional population in the initial state. The quantum state-dependent factors in the cross sections \( c_{P,T}(N, J, J', N') \) are commonly known as Placzek–Teller coefficients from the initial derivations.\(^{21} \) These factors are given for a molecule in a \( \Sigma \) electronic state with one electronic spin angular mo-
momentum in the Hund’s case b coupling scheme (electron spin coupled to rotational angular momentum) by

\[
c_{PT}(N, J, J', J) = \frac{(2N + 1)(2N' + 1)(2J' + 1)}{(2J + 1)(2J + 3)},
\]

where \( J \) is the total angular momentum, \( S \) is the electronic spin angular momentum (1 for \( \text{O}_2 \) and 0 for \( \text{N}_2 \)), and \( L \) is the component of the 2nd rank polarizability tensor responsible for the interaction. \( L = 0 \) for the isotropic part of the polarizability (the Cabannes component), and \( L = 2 \) for the anisotropic part of the polarizability (Raman scattering). The standard definitions for 3-\( j \) and 6-\( j \) coefficients are used. Two near equivalents to this equation give the formula for line strengths. These include the initial state degeneracy.\(^{22,23} \) For \( S = 0 \) (i.e., \( \text{N}_2 \)), Eq. (11) reduces to the result of Eq. (7) in Ref. 15:

\[
c_{PT}(J \rightarrow J + 2) = \frac{3(J + 1)(J + 2)}{2(2J + 1)(2J + 3)},
\]

\[
c_{PT}(J \rightarrow J) = \frac{J(J + 1)}{(2J - 1)(2J + 3)},
\]

\[
c_{PT}(J \rightarrow J - 2) = \frac{3J(J - 1)}{2(2J + 1)(2J - 1)}. \tag{12}
\]

This derivation gives correct Placzek–Teller coefficients for \( \text{N}_2 \) but approximates those for \( \text{O}_2 \) by treating it as a pure Hund’s case b molecule. At low \( N \) and \( J \), the departures from the pure coupling case that are due to the electron spin-rotation interaction are enough to affect the spectrum significantly.\(^{22,24} \) In the classic study of the \( \text{O}_2 \) ground state, Tinkham and Strandberg\(^{25} \) include the correct eigenvectors of the secular determinant for levels to as high as \( J = 26 \) in the case b basis set (their Table V and Eq. 54). Above this level, the molecule is described by case b behavior to a high degree of accuracy. For \( J = 0 \) and all odd \( J \) levels, the case b description is exact. For even \( J \) levels, \( J \neq 0 \), the transformation from case b basis functions \( \phi_{N,J} \) to eigenfunctions \( \Psi_{N,J} \) is given by

\[
\Psi_{J-1,J} = b_J \phi_{J-1,J} - d_J \phi_{J+1,J}
\]

\[
\Psi_{J+1,J} = d_J \phi_{J-1,J} + b_J \phi_{J+1,J}. \tag{13}
\]

The \( b_J \) and \( d_J \) values from Ref. 25 are used here to calculate correct Placzek–Teller coefficients for values to as high as \( J = 10 \), above which the corrections become completely negligible. An almost identical result was determined by Altmann et al.,\(^{26} \) who calculated eigenvectors for the secular determinant and molecular parameters given in the slightly earlier work of Mizushima and Hill.\(^{27} \) The database calculated with the corrected eigenvectors is prepared with an intensity cutoff to include all rotational Raman lines with intensities at 296 K within 0.1% of the strongest line. Because of the mixing of states, this now includes two \( \Delta N = 4 \) transitions. Figure 1 gives rotational Raman cross sections for \( \text{N}_2 \) and \( \text{O}_2 \) calculated for a temperature of 250 K and an excitation wavelength of 440 nm, appropriate to investigations of the effect of the Ring effect on retrievals of atmospheric \( \text{NO}_2 \) concentrations. The cross sections in this figure do not include the relative abundances of \( \text{N}_2 \) and \( \text{O}_2 \) in air.

![Fig. 1. Rotational Raman cross sections for (a) \( \text{N}_2 \) and (b) \( \text{O}_2 \) calculated for a temperature of 250 K and an excitation wavelength of 440 nm, appropriate to investigations of the effect of the Ring effect on retrievals of atmospheric \( \text{NO}_2 \) concentrations. The cross sections in this figure do not include the relative abundances of \( \text{N}_2 \) and \( \text{O}_2 \) in air.](image-url)
these measurements as the basis for broadening in the present data set.

For $\text{O}_2$, the HITRAN 92 values corresponding to rotational transitions of the same $\Delta N$ are initially adopted. When multiple corresponding transitions exist, the pressure-broadening coefficients are averaged. For the two $\Delta N = 4$ transitions, the averages of the values for lines connecting the upper and lower states are taken. The resulting pressure-broadening coefficients are multiplied by 1.185, which is the average result for the ratio of measured air pressure-broadening coefficients for $\text{O}_2$ magnetic dipole rotational transitions to those given in HITRAN (see Ref. 29 for an explanation of this correction). For $\text{N}_2$, pressure-broadening values for the corresponding quadrupole lines of the vibrational fundamental are adopted. Values determined for the pressure-broadening coefficients of both $\text{O}_2$ and $\text{N}_2$ are at 296 K. The temperature dependence should be calculated using the recommended HITRAN 92 coefficient of $n = 0.5$ for $\text{N}_2$ lines$^{20}$ and a value of $n = 0.72$ for $\text{O}_2$ lines,$^{20}$ through the relation

$$\Gamma_T = \Gamma_{296} \left( \frac{296}{T} \right)^n. \quad (14)$$

4. Solar Reference and Ring Source Spectra

A solar reference spectrum for the range 230–800 nm, at 0.01 nm resolution, has been determined by combining ground-based measurements$^{30}$ and balloon measurements.$^{31}$ Both spectra were converted to vacuum wavelengths. The balloon data were recalibrated in wavelength using 20 selected atomic reference lines. The spectra were resampled at even 0.01-nm increments, employing a triangular filter of 0.01-nm half-width at half-maximum, and linearly merged over the 300–305-nm wavelength range. The result is a reference spectrum in vacuum wavelengths accurate to better than 0.001 nm above 305 nm and 0.002 nm below 300 nm. This spectrum was created specifically for GOME applications, in particular wavelength calibration studies and calculation of Ring effect contributions to GOME measurements. A sample of this Fraunhofer reference spectrum is given in Fig. 2(a); in Fig. 2(b), it has been convolved with a Gaussian slit function having a half-width at $1/e$ intensity of 0.212 nm, appropriate to the GOME instrument in this wavelength range. The solar spectrum, with GOME slit function, has been convolved with the rotational Raman cross sections to create a Ring effect source spectrum. The units are those of cross section $\times$ photons, corresponding to the Ring effect scattering source per air molecule at 250 K. (d) The ratio of the Ring effect source spectrum given in (c) to the Fraunhofer spectrum of (b), with a cubic polynomial subtracted off. This spectrum closely corresponds to the Ring effect spectrum used in previous studies for atmospheric spectrum correction, as discussed in the text.
Fig. 2(c). The units are those of cross sections times photons, corresponding to the Ring effect scattering source per air molecule at 250 K. In Fig. 2(d), the ratio of the Ring effect source spectrum to the Fraunhofer spectrum has been taken and a cubic polynomial, fitted to the region presented in the figure, has been subtracted off, for comparison with differential Ring effect spectra determined in previous studies. Atmospheric Ring spectra have been determined by several groups from polarized spectroscopic measurements of the zenith sky at high solar zenith angles. Measurements at polarizations parallel and perpendicular to the single-scattering plane are combined in similar formulations to obtain effective Ring effect cross sections.\(^6,7,12\) For a single-scattering Rayleigh atmosphere, this procedure could yield the rotational Raman cross sections directly. In practice, because of Mie and multiple Rayleigh scattering, what is determined is the ratio of the rotational Raman spectrum to the solar Fraunhofer spectrum. This can be confirmed by comparing Fig. 2(d) with the results of the above studies. Atmospheric radiative transfer calculations of Ring spectra\(^7,12\) also determine this quantity. It is distinct from the filling in calculated by Joiner et al.,\(^10\) who include both the rotational Raman source term and the Cabannes loss term in their calculations. The use of the ratio of rotational Raman to Fraunhofer has proved useful in fitting atmospheric spectra for minor constituent abundances; it is a good approximation to the quantity needed for Ring effect correction in some fitting techniques and measurement geometries. However, its use has limitations because of the distortion of the rotational Raman spectrum and its constant ratio to the Fraunhofer spectrum. The rotational Raman spectrum and Fraunhofer spectrum determined in this study should be more generally applicable to fitting of atmospheric spectra obtained from various measurement geometries, including satellite-based measurements.

5. Conclusions

The complete Rayleigh with rotational Raman-scattering database as described in the previous sections is available from the authors. It includes \(N_2\) and \(O_2\) term values and statistical partitioning (tables including the quantum numbers and term energies for \(O_2\) and \(N_2\) up to states allowing for partitioning to better than 0.01% accuracy and Boltzmann factors with nuclear spin degeneracies \(g_J\))\(^a\); tables giving the King correction factors, values of \(\epsilon\), where \(\epsilon = (\gamma/\alpha)\)\(^2\), index of refraction \(\alpha\), and \(\gamma\) for \(O_2\) and \(N_2\); Placzek–Teller coefficients, with examples of cross sections; pressure-broadening coefficients; the solar reference spectrum derived above; and the convolved solar and rotational Raman spectrum (Ring effect source function cross section calculations).

This database is a summary of the best available relevant data that were found in the literature and the calculations performed in this study. One item not included here is the additional broadening that is due to Rayleigh–Brillouin scattering.\(^4\) For the rotational Raman lines, this will provide an extra source of broadening, although the extent of the broadening is within the uncertainties in the pressure broadening for scattering in the troposphere. The effect of Rayleigh–Brillouin scattering for the filling in of the central Cabannes line for narrow Fraunhofer lines might be significant for some satellite measurement conditions and will be a topic for future investigations.

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