

## High-resolution photoabsorption cross section measurements of SO<sub>2</sub>, 2: 220 to 325 nm at 295 K

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[1] High-resolution, ultraviolet SO<sub>2</sub> photoabsorption cross section data are required for modeling density and temperature profiles of SO<sub>2</sub> in solar system atmospheres. We report measurements of such cross sections on SO<sub>2</sub> at 295 K for the  $\tilde{B}^1B_1 - \tilde{X}^1A_1$  and  $\tilde{C}^1B_2 - \tilde{X}^1A_1$  bands over the wavelength range 220 to 325 nm. Resolving powers of up to 550,000 were employed in order to study the congested spectrum of sharp SO<sub>2</sub> features in parts of this region. Our data are compared to earlier values obtained with lower resolving power. *INDEX TERMS*: 0343 Atmospheric Composition and Structure: Planetary atmospheres (5405, 5407, 5409, 5704, 5705, 5707); *KEYWORDS*: ultraviolet absorption spectra, planetary atmospheres, photoabsorption cross sections

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### 1. Introduction

[2] SO<sub>2</sub> is a major component of several solar system atmospheres and is important in a number of terrestrial atmospheric processes. *Ballester et al.* [1994], *Clarke et al.* [1994], *Lellouch* [1996], *Hendrix et al.* [1999], and *McGrath et al.* [2000] have examined the temperature and density profiles of SO<sub>2</sub> on Io. Until recently, models of the atmosphere of Io have relied on photoabsorption cross section data measured with low resolving power,  $R$  ( $R = \lambda/\Delta\lambda$ , where  $\Delta\lambda$  is the wavelength resolution). The use of low-resolution data has led to the modeling of SO<sub>2</sub> ultraviolet (UV) absorption as a continuum when it is, in fact, a dense line spectrum.

[3] Several authors, e.g., *Belton* [1982], have pointed to cross sections measured at low resolution as a possible source of systematic error in models of planetary atmospheres. In the case of Io, high-resolution laboratory data could be used to improve atmospheric models and provide insight into the relative contributions of volcanic outgassing and surface-frost sublimation as sources of SO<sub>2</sub>.

[4] SO<sub>2</sub> is also a significant component in the Venusian atmosphere [*Barker*, 1979; *Stewart et al.*, 1979; *Esposito et al.*, 1988]. High-resolution photoabsorption cross sections are of increasing importance as improved spectra of Venus have become available [*Esposito et al.*, 1997]. Studies of SO<sub>2</sub> in the terrestrial stratosphere as a result of volcanic activity [*Krueger*, 1983; *Hansen et al.*, 1997; *Eisinger and*

*Burrows*, 1998] would also be furthered by better photoabsorption cross section data for SO<sub>2</sub>.

### 2. UV Spectroscopy of SO<sub>2</sub>

[5] The UV spectrum of SO<sub>2</sub> is characterized by two major absorption systems. The first, with densely packed lines extending from approximately 170 to 230 nm, comprises a series of vibrational bands assigned to the  $\tilde{C}^1B_2 - \tilde{X}^1A_1$  electronic transition. A region of relatively low cross section separates these bands from a similar but somewhat weaker progression that stretches from roughly 240 to 338 nm and is attributed mainly to the  $\tilde{B}^1B_1 - \tilde{X}^1A_1$  transition, although a number of other states ( $^3B_1$ ,  $^3B_2$ , and  $^1A_2$ ) are also involved in absorption at these wavelengths [*Kullmer and Demtroder*, 1985].

[6] Analysis of transitions within the  $\tilde{B} - \tilde{X}$  system has been performed [e.g., *Herzberg*, 1966; *Brand and Nanes*, 1973; *Hamada and Merer*, 1975; *Baskin et al.*, 1995; *Hegazi et al.*, 1998]. A summary of the progress in assigning transitions within the  $\tilde{C} - \tilde{X}$  system and the effects of predissociation on the bands is given in our first report of SO<sub>2</sub> cross sections measured with high resolving power [*Stark et al.*, 1999]. However, theoretical analysis of both band systems is incomplete.

[7] Table 1 summarizes previous measurements of UV photoabsorption by the SO<sub>2</sub> molecule. *Manatt and Lane* [1993] compiled and compared the work of a number of groups to produce a low-resolution photoabsorption cross section dataset extending from 106 to 403 nm. *Prahlad et al.* [1996] and *Wu et al.* [2000] studied wavelength regions similar to that discussed here over a range of temperatures but with resolving powers up to two orders of magnitude lower than used in our work. *Koplow et al.* [1998] employed a tunable, narrow-band, frequency-quadrupled diode laser to reach a very high resolving power, considerably beyond that required to resolve the Doppler widths of individual lines, but they studied only

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**Table 1.** Published SO<sub>2</sub> Absorption Datasets

Reference	Resolving Power	Spectral Range, nm	Temperatures, K
Present work	90,000–550,000	220–325	295
<i>Stark et al.</i> [1999]	450,000	198–220	295
<i>Wu et al.</i> [2000]	5,200	208–295	400, 295, 200
<i>Koplow et al.</i> [1998]	$1.5 \times 10^8$	215.21–215.23	295
<i>Prahlad et al.</i> [1996]	3,000	280–320	220–300
<i>Vandaele et al.</i> [1994]	16,600	250–370	295
<i>Manatt and Lane</i> [1993]	2,500	106–403	295
<i>Ahmed and Kumar</i> [1992]	1,250	188–231, 279–320	295
<i>Martinez and Joens</i> [1992]	2,200	197–240	295
<i>Hearn and Joens</i> [1991]	4,725	228–339	300
<i>McGee and Burris</i> [1987]	10,400	300–324	295, 210
<i>Freeman et al.</i> [1984]	200,000	172–240	213
<i>Wu and Judge</i> [1981]	4,560	208–228, 299–340	295

a small spectral range. The work of *Vandaele et al.* [1994] comes closest to that reported here, but their resolution was a factor of 5 to 30 less than that used for our measurements.

[8] Because of the concern about the limits of atmospheric models that employ low-resolution photoabsorption cross section data, we have undertaken a number of measurements with resolutions at or near the Doppler width of the SO<sub>2</sub> absorption features. Our previous paper [*Stark et al.*, 1999] presents high-resolution cross section data for SO<sub>2</sub> at  $\sim 295$  K over the wavelength range 198 to 220 nm. This paper extends the wavelength range to 325 nm; preliminary results at lower temperature have been discussed by *Smith et al.* [2000].

### 3. Experimental Procedure

[9] Measurements of the photoabsorption cross sections of the SO<sub>2</sub> band systems between 220 and 325 nm were made using the Imperial College Fourier transform spectrometer (ICFTS) in a manner similar to that of our earlier measurements [*Stark et al.*, 1999]. The ICFTS [*Thorne et al.*, 1987] is capable of resolving powers that are more than sufficient to resolve the 295 K Doppler profiles (about 4 mÅ FWHM at 260 nm) of the narrowest SO<sub>2</sub> absorption features in this spectral region. However, high-resolution but limited bandwidth studies (J. W. Brault, personal communication, 1999) and preliminary low-  $S/N$  (signal-to-noise) ratio studies with the ICFTS showed that there are large variations of the natural line widths and in the magnitudes of the photoabsorption cross sections of SO<sub>2</sub> features across the 220 to 325 nm spectral range. Therefore we divided the spectral range into a number of smaller regions so that both resolving power and column densities could be selected to optimize the  $S/N$  ratio in the time available for the measurements. The smaller regions were selected from the 300 W xenon arc background continuum using a zero-deviation, zero-dispersion monochromator [*Murray*, 1992] with a bandpass of approximately 20 nm. Widths of spectral features become somewhat greater than the 295 K Doppler widths shortward of 291 nm, possibly because of line blending. Below this wavelength, measurements could be made at resolutions greater than the Doppler width without loss of accuracy. Only the region 220 to 230 nm, where the SO<sub>2</sub> spectral features are again very narrow, was under-resolved (resolution = 16 mÅ, while Doppler width = 3.5 mÅ); the parameters chosen for this region reflect a com-

promise between resolution and  $S/N$  ratio in the time available.

[10] Samples of SO<sub>2</sub> were taken from a cylinder of 99.9% pure gas supplied by BOC and admitted at room temperature to a 10.0 cm long fused silica absorption cell. The sample pressures used ranged from 1.02 to 32.8 Torr, which are equivalent to column densities of  $3.34 \times 10^{17}$  to  $1.07 \times 10^{19}$  cm<sup>-2</sup>, respectively (see Table 2). The pressures were recorded with 10 Torr (MKS Baratron) and 100 Torr (Edwards) capacitance manometers.

[11] The experimental procedure took advantage of the dual output beams of the ICFTS. The absorption cell was positioned in one of the beams, which was used to record absorption cross section data; the other was used to monitor the intensity of the background continuum,  $I_0$ . Before and after each set of absorption scans, empty-cell values of the two output channel signals were recorded; absolute cell transmittances were obtained by dividing the full-cell ratios by the empty-cell ratios. Measurements in all spectral regions were performed at two or more pressures.

### 4. Results and Analysis

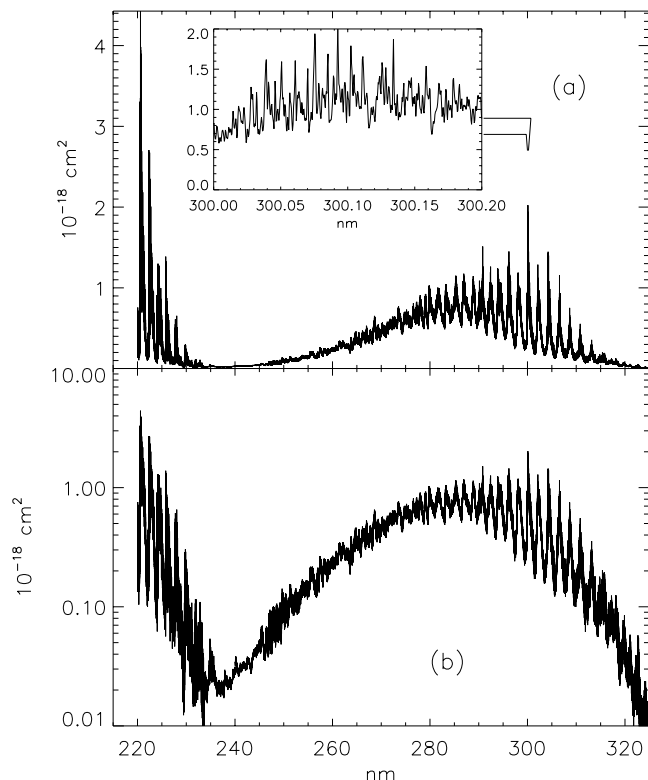
[12] The measured SO<sub>2</sub> absorption spectra were converted to photoabsorption cross sections,  $\sigma$ , through application of the Beer-Lambert law,

$$\sigma(\lambda) = \frac{1}{N} \ln \left[ \frac{I_0(\lambda)}{I(\lambda)} \right], \quad (1)$$

**Table 2.** Parameters for SO<sub>2</sub> Absorption Measurements

Label	Region, nm <sup>a</sup>	Observation Time, hrs	Pressure, Torr	Resolution, mÅ
A	308.4–325.0	6	6.9–33.	4
B	294.8–308.4	8	1.5–9.1	4
C	282.9–294.8	6.6	1.6–2.5	4
D	282.9–294.8	3	1.6–2.5	8
E	263.5–282.9	3	1.9, 3.5	8
F	246.9–263.5	3	7.0, 8.7	8
G	233.6–246.9	2	12.6, 30.	32
H	220.0–233.6	2.5	16.3	16
I	220.0–226.7	4	1.5	16

<sup>a</sup>The wavelength range given here is the portion of the bandpass of the prism monochromator that was used to produce the final set of cross section data. The actual bandpasses of the monochromator extend about  $\pm 2$  nm beyond the ranges given here, so the data sets, in fact, overlap.



**Figure 1.** Photoabsorption cross section of SO<sub>2</sub> at 295 K measured with a resolution of 4 to 32 mÅ. (a) Linear scale, with structure in a 0.2 nm region shown in the inset. Note that the  $S/N$  ratio for this measurement was about 100, so the structure shown in the inset is real. (b) Log scale.

where  $N$  is the SO<sub>2</sub> column density and  $I(\lambda)$  is the transmitted intensity through the absorption cell at wavelength  $\lambda$ . Wavelength calibration was achieved through comparison of SO<sub>2</sub> absorption features with the well-determined wavelengths of Fe emission lines produced in a hollow cathode discharge [Lerner and Thorne, 1988]. This method allowed calibration of our SO<sub>2</sub> wavelength scale to an accuracy better than 10 mÅ.

[13] Our results are displayed on linear and log scales in Figure 1. In spectral regions that have a large range of absorption cross section, namely those denoted A, B, G, H, and I in Table 2, the final data are based on a combination of measurements: high cross section values were obtained from measurements with lower column density and vice versa.

[14] An analysis of uncertainties in our photoabsorption cross section measurements using the ICFTS was discussed in our previous paper [Stark et al., 1999] where it was shown that

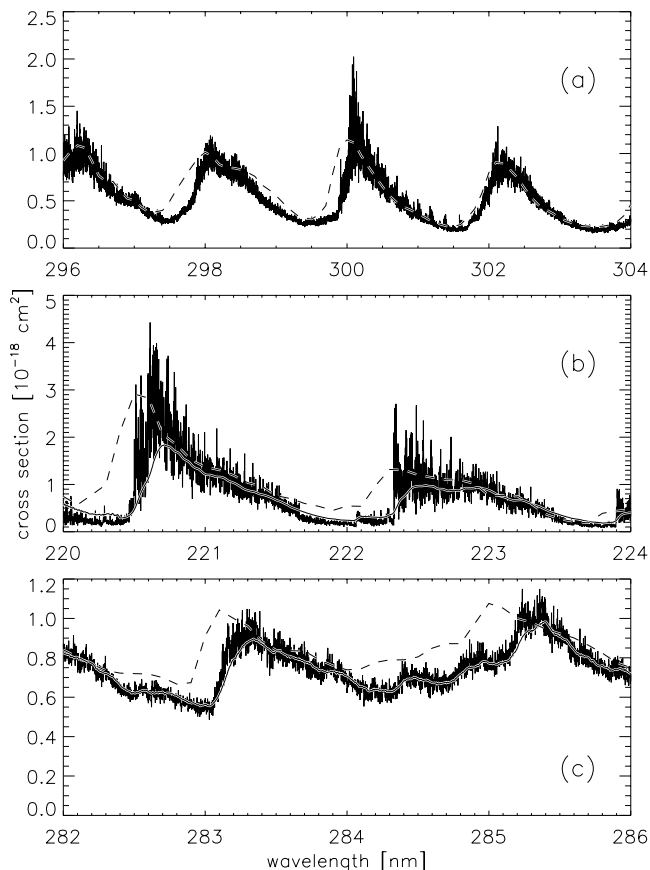
$$\frac{\Delta\sigma}{\sigma} = \left[ \left( \frac{\Delta N}{N} \right)^2 + \left( \frac{1}{(S/N \text{ ratio})N\sigma} \right) \{1 + e^{2N\sigma}\} \right]^{0.5}, \quad (2)$$

where the two terms are the fractional uncertainty in the column density and uncertainties associated with the  $S/N$

ratio and the absorption optical depths ( $N\sigma$ ) of the measurement. We estimate the uncertainty in the column densities to be 3% (one standard deviation). The  $S/N$  ratio was 50 to 100. By comparing overlapping datasets for various wavelength regions we determined that the statistical uncertainty was about 3%. Overall, we estimate that the one standard deviation uncertainty in the results is about 5%.

## 5. Comparisons and Conclusions

[15] Our SO<sub>2</sub> cross-section values at 295 K can be compared to previous results; the measurement parameters for these are listed in Table 1. Many of the differences in the published measurements can be related to instrumental resolution effects. Absolute cross sections of the very sharp and congested rotational structure in the SO<sub>2</sub> spectrum longward of about 291 nm and shortward of about 231 nm (see Figure 1) cannot reliably be determined from low-resolution measurements. Figure 2a compares results from the compilation of Manatt and Lane [1993], at a resolution of 1 Å, with our cross section data (4 mÅ resolution) in the 296 to 304 nm region. The peak cross



**Figure 2.** Comparison of our results (highly structured line) with those of Manatt and Lane [1993] (dashed line) and Wu et al. [2000] (smooth solid line with white border).

sections measured at high and low resolution in the sharp  $\bar{B} - \bar{X}$  band at 300 nm differ by almost a factor of two. In regions of lower absorption or where there are broader features the two data sets are in rough accord. There are, however, some wavelength scale discrepancies, which are most apparent in the contours of the bands at 298 and 300 nm.

[16] Three new SO<sub>2</sub> room temperature cross section results appeared following publication of the *Manatt and Lane* [1993] compilation: *Vandaele et al.* [1994] measured cross sections from 250 to 370 nm at a resolution of about 200 mÅ; *Prahlad et al.* [1996] covered the 280 to 320 nm region with 1 Å resolution; and *Wu et al.* [2000] measured the 208 to 295 nm region with 500 mÅ resolution. In the region of sharp band structure near 300 nm, our measured peak cross sections are about 30 to 50% higher than those of *Vandaele et al.* [1994] and about a factor of two higher than those of *Prahlad et al.* [1996]. Figures 2b and 2c show comparisons of our data with those of *Wu et al.* [2000] in regions of sharp structure (220 to 224 nm) and broader structure (282 to 286 nm). The two data sets show excellent agreement in the region of broader structure (Figure 2c) but, as expected from instrumental resolution considerations, differ significantly in the region of sharp and strong absorption features (Figure 2b). The data of *Manatt and Lane* [1993] are also shown in Figures 2b and 2c; again, positions of the broad absorption bands seem to be shifted shortward by about 0.2 nm.

[17] This paper presents absorption cross sections at 295 K measured at a resolution sufficient to reveal all rotational structure. However, the spectroscopic properties of SO<sub>2</sub> in the ultraviolet are still poorly understood and its absorption behavior with temperature cannot be predicted accurately. Thus to support planetary atmosphere research, high-resolution laboratory measurements at a range of temperatures are needed. *Wu et al.* [2000] complemented their low-resolution room temperature work with cross section measurements at 200 K and 400 K. Earlier low-resolution work over a range of temperatures in the wavelength region longward of 200 nm was published by *McGee and Burris* [1987] and *Prahlad et al.* [1996]; other low-resolution work is reviewed by *Wu et al.* [2000]. Our future work on SO<sub>2</sub> will concentrate on an extension of our high-resolution measurements in the 198 to 220 nm region [*Stark et al.*, 1999] to an SO<sub>2</sub> temperature of 160 K. Numerical tabulations of our SO<sub>2</sub> cross sections can be obtained from the authors and are also available at <http://cfa-www.harvard.edu/amdata/ampdata/cfamols.html>.

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