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Propagation of a 4μm free electron laser beam through sulphur dioxide gas

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ABSTRACT

Propagation of a free electron laser beam of wavelength 4μm through sulphar dioxide is studied. The frequency interval considered is from 2523.336 to 2524.727 cm^{-1}. Values of transmittance, averaged over intervals of 0.1 cm^{-1}, are calculated for absorber thickness 0.01, 0.1 and 1 atm-cm by the help of quasi random model of molecular band absorption. From generated absorptance values intensities of the high resolution absorption lines in the above interval of SO_2 can be simulated. There is a scope to apply this work in atmospheric optics.

Keywords. Sulphar dioxide, transmittance, quasi-random model.

INTRODUCTION

Sulphur dioxide is an important astrophysical and atmospheric molecule. Natural sources of sulphur dioxide (SO_2) include release from volcanoes, oceans, biological decay and forest fires. In 1983 the United Nations Environment Programme estimated a figure of between 80 million and 288 million tonnes of sulphur oxides per year. Sulphur dioxide emissions also result from combustion of fossil fuels due to varying amounts of sulphur being present in these fuels. Lasers are increasingly used in pollution control studies and monitoring the environment. Recently a linear relationship between concentration of sulfur dioxide and optical parameter (OP) is established using the Beer–Lambert law [1]. A precise knowledge of the spectra of sulphur dioxide is very important for accurate measurements involving the passage of a laser beam through this gaseous medium. In this work, a free electron laser is considered tuned to 4μm. The frequency interval in which the propagation of this free electron laser beam is considered, 2523.336 – 2524.727 cm^{-1}, lies in the combination vibration – rotation $\nu_1+\nu_3$ band of SO_2. The combination vibration – rotation $\nu_1+\nu_3$ band of SO_2 has been recorded under Doppler limited and atmospheric conditions with 3* 10^{-4} cm^{-1} instrumental resolution using a difference-frequency laser [2]. The $\nu_1+\nu_3$ band of SO_2 is strategically located in the 4 μm atmospheric window which is convenient for monitoring SO_2 in the air or observing extraterrestrial SO_2 through the atmosphere. The method used in this work, the quasi-random model [3] of molecular band absorption, is a variant of one of the methods described by Goody and Yung [4]. In their monograph, Goody and Yung have contrasted the use of random models with the line by-line method, and concluded that in some circumstances the random models might be sufficient, and require much less computer time. Infrared transmittances, based on the quasi-random model, have been calculated for H_2O and CO_2 and the results fitted with experimental measurements [5,6]. Using this model, simulations of intensities of absorption lines have been done for p-benzoquinone-H4 vapour [7] in the region 17800 - 24900 cm^{-1}, for water vapour [8] around 1.15 μm, and for nitrogen [9] around 575 nm. Potential use of this model in developing rapid models for accurately calculating atmospheric transmittances has been indicated [10]. Simulating the intensities of high-resolution lines of nitrogen around 570 nm, applicability of this model in optics of the atmosphere, especially of the upper atmosphere, has been shown [11]. Intensities of the high resolution absorption lines of sulphur dioxide are simulated in the frequency interval 2499.0115-2499.9910 cm^{-1}[12]. The frequency interval under consideration in the present study is another convenient sector in the SO_2 spectrum for the investigation of propagation of a laser beam through it.

Method of calculation

High-resolution near-infrared absorption spectrum of sulphur dioxide is considered frequency interval 2523.336 – 2524.727 cm^{-1}. The maximum relative intensity is normalized to unity and other values of intensity are taken relative to this one. The lines along with the assigned intensities are given in Table 1. The entire spectrum is divided into frequency intervals $\Omega = 0.1$ cm^{-1} wide. These Ωs are the intervals over which the average transmittances have been computed. Each interval is further divided into smaller intervals $\delta = 0.025$ cm^{-1}. The quasi-random model localizes each line within an error defined by the interval size δ. The transmittance at a frequency ν, as affected by n_p lines within the interval δ_p is computed from the expression [13]

$$\Im(\nu) = \prod_{i=1}^{5} \left\{ (1/\delta) \int_{\delta_p} \exp\left[-\frac{S_i u \alpha / \pi}{(\nu - \nu_i)^2 + \alpha^2} \right] d\nu_i \right\}^{n_i} \qquad (1)$$

where n_i represents the number of lines within the intensity range i, which itself is characterized by an average intensity S_i, α is the half width at half maximum, u is the absorber thickness, and ν_i refers to the centre of the line. For three different masses per unit area, u = 0.01, 0.1 and 1.0 atm-cm, and taking the half-width as $\alpha = 0.015$ cm^{-1}, equation (1) is evaluated with the help of a computer program based on Simpson's rule of numerical integration. First, the transmittance values are calculated at the centres of 0.1 cm^{-1} intervals. Transmittances by the wings of lines at the left and right adjacent intervals are also included. The transmittance at the centre of an interval is finally obtained as [14]

$$\Im = \Im_j \prod_{i \neq j} \Im_i \qquad (2)$$

Next, transmittance values are obtained for another set of frequency intervals whose centres are shifted by half the interval size (0.05 cm^{-1}) from the original positions of the centres of the intervals. This is done in order to minimize the error associated with the occurrence lines at frequencies near the edges of a given interval. The results for the shifted and un-shifted intervals are averaged, and thus we obtain the average transmittance over a 0.1 cm^{-1} interval.

Table 1: SO_2 lines affecting the propagation of the 4 μm laser beam

Frequency (cm^{-1})	Intensity	Frequency (cm^{-1})	Intensity
2523.336	0.42	2524.078	0.48
2523.345	0.61	2524.107	0.32
2523.3603	0.66	2524.1303	0.63
2523.3697	1	2524.188	0.43
2523.4394	0.59	2524.197	0.43
2523.5122	0.50	2524.274	0.40
2523.570	0.50	2524.3924	0.56
2523.5821	0.76	2524.424	0.32
2523.6553	0.76	2524.431	0.32
2523.744	0.34	2524.443	0.46
2523.752	0.46	2524.447	0.43
2523.775	0.52	2524.488	0.32
2523.824	0.44	2524.496	0.18
2523.8510	0.97	2524.519	0.24
2523.881	0.44	2524.5785	0.66
2523.9610	0.58	2524.5887	0.47
2523.971	0.48	2524.693	0.35
2524.0025	0.61	2524.727	0.27

Table 2: Absorption of a free electron laser beam for three different amounts of SO_2

Frequency (cm^{-1})	Absorptance (%) for path length 0.01 atm-cm	0.1 atm-cm	1 atm-cm
2523.336	23.45	23.63	25.45
2523.436	30.09	30.38	33.29
2523.536	25.24	25.6	29.15
2523.636	26.93	27.66	34.43
2523.736	31.8	33.55	48.55
2523.836	39.43	61.75	84.11
2523.936	47.26	89.88	100
2524.036	44.83	89.48	100
2524.136	38.66	68.43	87.05
2524.236	25.45	27.7	45.78
2524.336	28.47	29.11	35.17
2524.436	34.63	34.97	38.23
2524.536	37.52	37.8	40.55
2524.636	31.41	31.56	33.06
2524.736	20.9	20.98	21.77

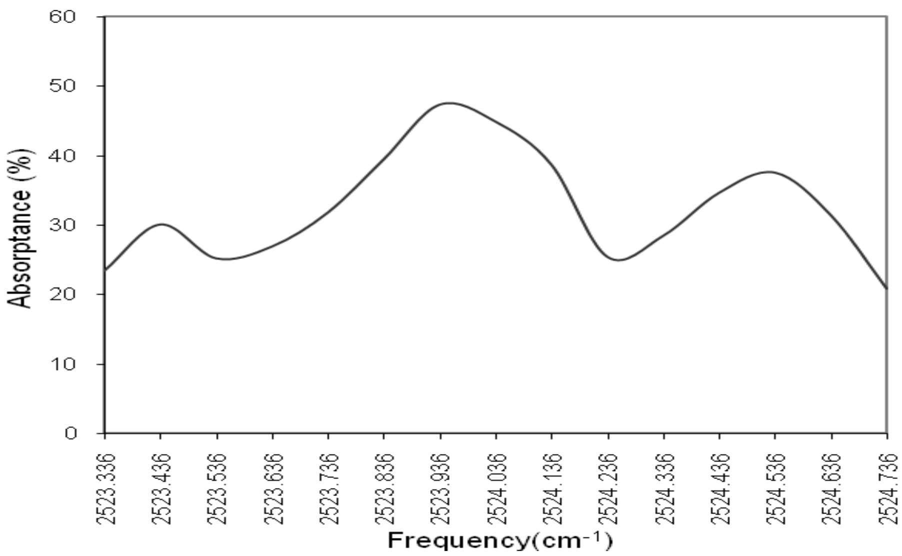

Figure 1: Absorptance of a 4 μm free electron laser beam for a 0.01 atm-cm path length of SO_2.

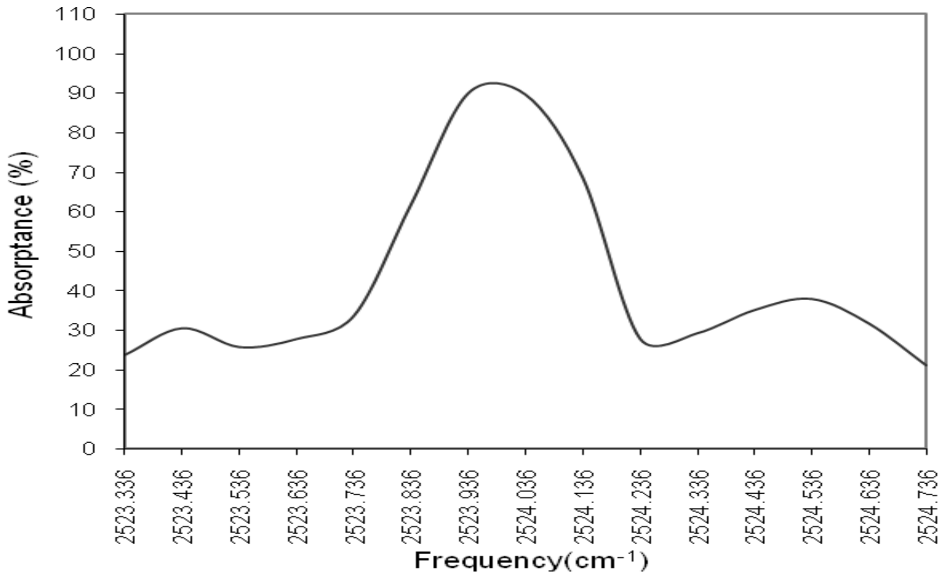

Figure 2: Absorptance of a 4 μm free electron laser beam for a 0.1 atm-cm path length of SO_2.

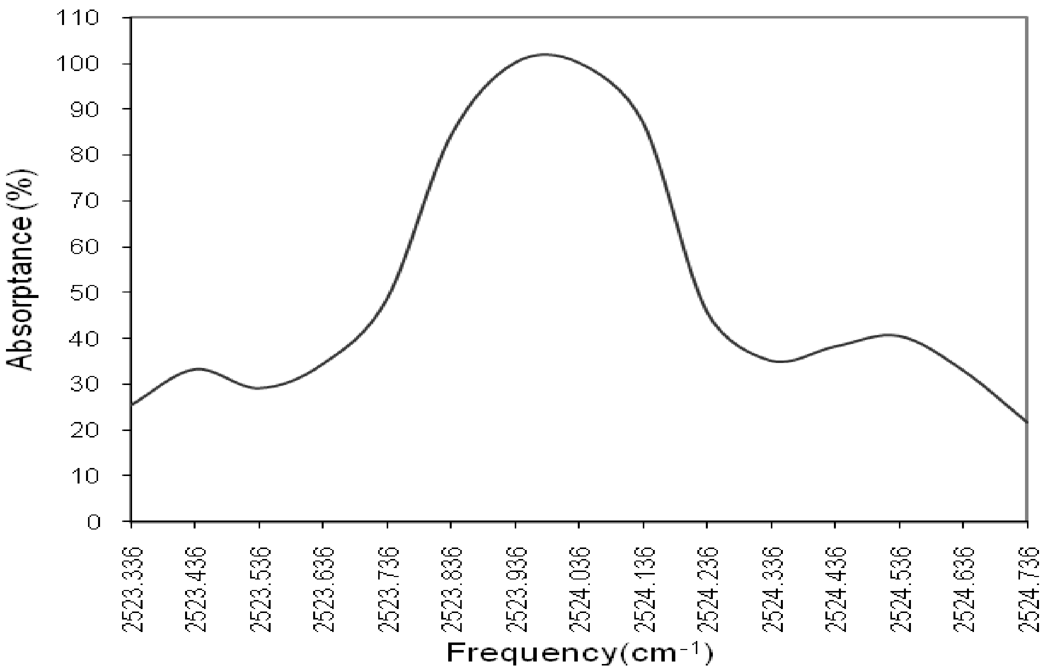

Figure 3: Absorptance of a 4 μm free electron laser beam for a 1 atm-cm path length of SO_2.

RESULTS AND DISCUSSION

Influences of 36 lines for the interval are worked out for 0.01, 0.1 and 1 atm-cm thickness of sulphur dioxide in the frequency intervals 2523.336 – 2524.727 cm^{-1}. The computational results for the propagation of a 4 μm laser beam through these three amounts of the absorber are presented in Tables 2 and shown in Figures 1 –3. From generated absorptance values intensities of the high resolution absorption lines in the above interval of SO_2 can be simulated. The absorption values agree well with the experimental data taken for this work. This verifies that the quasi-random model for simulating the intensity distribution by grouping the lines in a given frequency interval works reasonably well.

CONCLUSION

In this work, the broadening of the lines is assumed to be homogeneous, as the rotational lines are observed to be sufficiently fine. Therefore, there is a scope to generalize the model for inhomogeneous broadening as well. The application prospect of this model in atmospheric optics is quite bright. Till now, a large number of high resolution absorption spectra of other diatomic and polyatomic molecules have been reported; the present work could easily be extended to these spectra. Temperature and pressure dependence of the linewidth, and consequently of the absorptance, is the aspect that calls for further research. The scopes of error in these computations are (a) the size of the small intervals, δ, as mentioned in the description of the model, and (b) the number of divisions taken in the Simpson-rule based program, which is known to any programmer.

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