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Archives of Physics Research, 2011, 2 (3):23-28
(http://scholarsresearchlibrary.com/archive.html)

Scholars Research Library
ISSN : 0976-0970
CODEN (USA): APRRC7

Application of quasi-random model in laser beam propagation through SO_2

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ABSTRACT

The quasi random model is one of the elementary models of molecular band absorption. We show its application in the interaction of a free electron laser beam of wavelength 4μm with high resolution lines of the sulphar dioxide gas. 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 0.05, 0.5 and 5 atm-cm by the help of quasi random model. From generated absorptance values intensities of the high resolution absorption lines in the above interval of SO_2 can be simulated. The application prospect of this work in atmospheric optics is quite bright.

Keywords: Sulphar dioxide lines, transmittance, free electron laser, quasi-random model.

INTRODUCTION

The passage of a laser beam through the earth atmosphere presents an important transmission problem, a novel aspect of which is the almost monochromatic nature of a laser beam. 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 intervals 2499.0115-2499.9910 cm^{-1}[12] and 2523.336 - 2524.727 cm^{-1} [13]. This work can be treated as extended part of the previous work in the frequency interval 2523.336 - 2524.727 cm^{-1}. Here amount of absorbers are taken five times than in earlier work to see the effects.

MATERIALS AND METHODS

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 Ω = 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 δ = 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 [14]

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

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.

The gases of the atmosphere are usually measured by the unit *atmosphere centimetre* (atm-cm). This measurement unit is used to define an atmospheric gas distributed along a path reduced to a layer at STP, provided the other gases are excluded. The resulting thickness is then expressed in atm-cm, given by $u = cLP$, where c is the fractional concentration of the absorber, L is the path length in cm and P is the pressure in atmosphere.

For three different masses per unit area, u = 0.05, 0.5 and 5.0 atm-cm, and taking the half-width as α=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

centre 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 [15]

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

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		
	0.05 atm-cm	0.5 atm-cm	5 atm-cm
2523.336	23.53	24.45	33
2523.436	30.22	31.69	44.73
2523.536	25.4	27.2	42.95
2523.636	27.26	30.76	56.85
2523.736	32.58	40.75	81.82
2523.836	52.72	79.16	96.29
2523.936	75.14	99.97	100
2524.036	74.07	99.97	100
2524.136	57.26	82.36	98.83
2524.236	26.47	36.65	79.03
2524.336	28.76	31.87	56.12
2524.436	34.78	36.44	50.87
2524.536	37.64	39.04	51.33
2524.636	31.48	32.23	39.25
2524.736	20.93	21.33	25.19

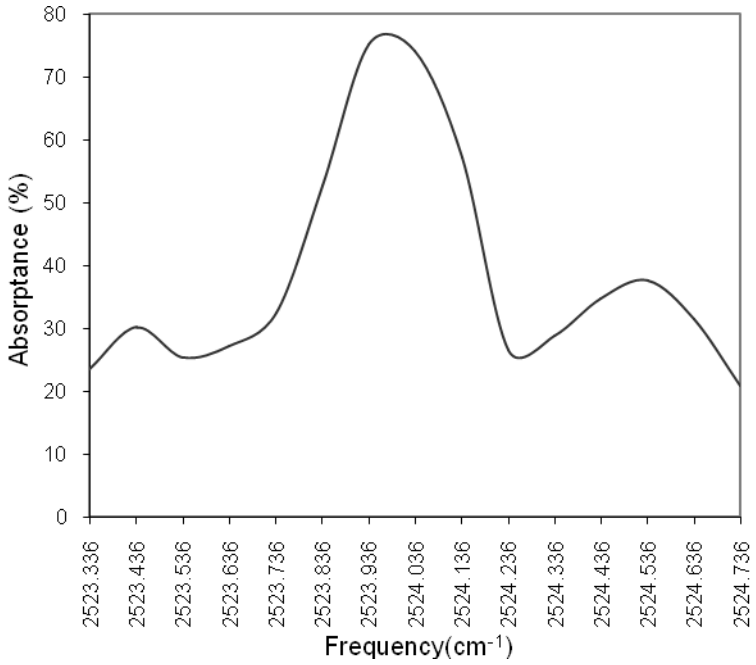

Figure 1: Absorptance of a 4 μm free electron laser beam for a 0.05 atm-cm path length of SO_2.

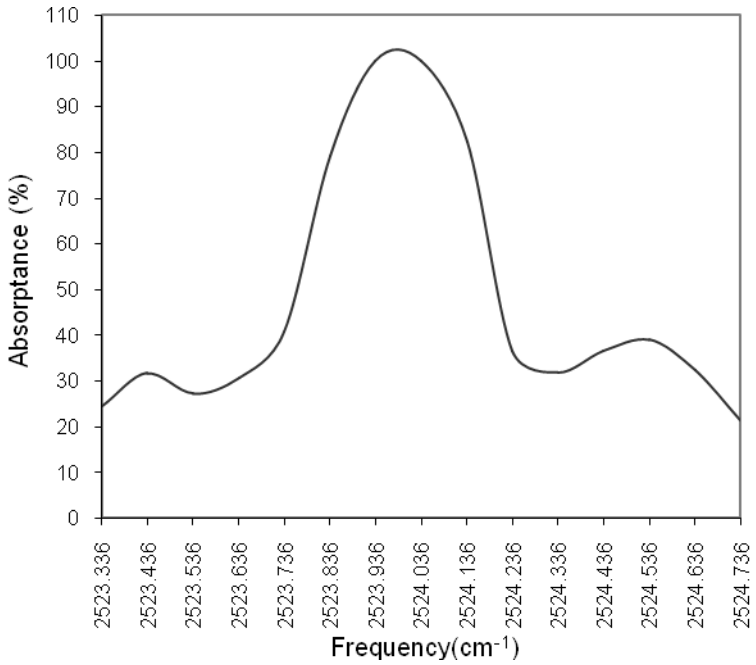

Figure 2: Absorptance of a 4 μm free electron laser beam for a 0.5 atm-cm path length of SO_2.

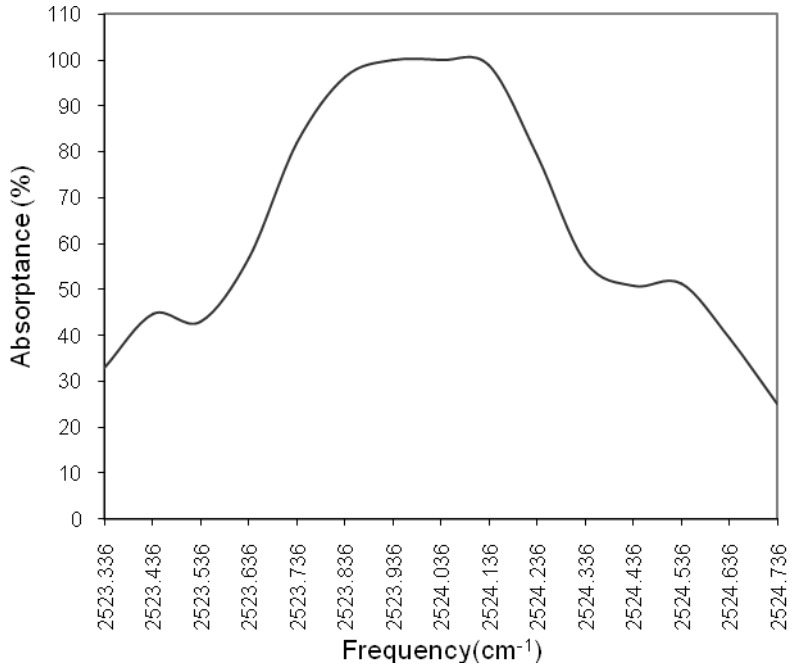

Figure 3: Absorptance of a 4 μm free electron laser beam for a 5 atm-cm path length of SO_2.

RESULTS AND DISCUSSION

Influences of 36 lines for the interval are worked out for 0.05, 0.5 and 5 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. By comparing these figures with the figures of the previous work of same interval [13], it reveals that the smaller the amount of the absorber the more marked is the variation. This is, perhaps, expected, as with greater amounts of the absorber the absorptance values tend to saturate. 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 concludes that the quasi-random model for simulating the intensity distribution by grouping the lines in a given frequency interval works reasonably well - a fact established in recent times for important atmospheric species like nitrogen sulphur dioxide and methane.

CONCLUSION

As the rotational lines are observed to be sufficiently fine, therefore in this work the broadening of the lines is assumed to be homogeneous. There is a scope to generalize the model for inhomogeneous broadening as well. Application prospect of this work 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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