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AIAA 82-4148

Absorptivity of Water Vapor for 10.6 μm Radiation

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IN a recent paper, Fowler[1] presents measurements of the absorptivity of water vapor to 10.6 μm laser radiation. The experiments were performed in an axisymmetric chamber, where a mixture of water vapor and hydrogen was heated by a cw CO_2 laser, and a CO_2 probe laser was used to measure absorptivity.

Fowler obtained rather surprising results. His values of absorptivity are a factor of 50 to 100 higher than the values calculated by Ludwig and co-workers,[2-4] who used a model fitted to low-resolution spectral data. Fowler comments that "one possible explanation is radiative excitation of the H_2O molecule causing its vibrational population distribution to be highly excited, thereby making possible absorption of probe beam energy by high lying H_2O energy levels."

The purpose of this Note is to call attention to recent measurements of this absorptivity, using shock-heated H_2O/H_2 and H_2O/Ar mixtures, and a probe CO_2 laser.[5] These measurements give values about a factor of 2 *lower* than Ludwig's low resolution values. We will also present an argument that Fowler's high values are unlikely to be caused by excited water molecules.

The experiments described in Ref. 5 were performed behind incident and reflected shocks near the end wall of a heated shock tube. They encompassed a range of temperatures from 600 to 3700 K, at pressures from 1 to 40 atm, in mixtures of 2, 5, and 10 mole percent water vapor in argon. A limited number of runs were made in a 10 mole percent mixture of water vapor with either hydrogen or nitrogen.

The effects of pressure broadening were also investigated by varying conditions at several temperatures.

Care was taken to assure that the water vapor did not condense on the walls or windows. The gas conditions were calculated using the measured shock speed. Absorptivity was measured by detecting the fraction of an incident CO_2 probe laser beam which was transmitted through the shock tube along a diameter. The carefully controlled conditions provided well-characterized gas conditions of pressure, temperature, and composition. The details of the measurements are presented in Ref. 5.

The probe laser was operated at the P(16), P(18), and P(20) transitions, which bracket the line used by Fowler. Within this narrow spectral range of 944-948 cm^{-1}, no significant variation in absorption coefficient (suitably normalized as described below) was observed as a function of laser line, water vapor concentration, total pressure, or diluent gas.

The results are presented in Fig. 1 in the form of absorptivity α vs temperature. The units of α are cm^{-1}/atm of water vapor existing in equilibrium at the temperature of the measurement. This is in contrast to the values presented by Fowler in Fig. 9 of Ref. 1, where absorptivity is given in cm^{-1} for a mixture whose unheated composition is 0.3 atm H_2O in 13 atm H_2. His values have been converted to the present units by using the equilibrium amount of water vapor present in his mixture at each temperature.

Figure 1 shows that the shock tube data of Ref. 5 is about a factor of 2 *below* the Ludwig calculations while the data of Ref. 1 is a factor of 50 to 100 *above* those calculations.

The slight reduction of the present high resolution values below those of Ludwig is not unexpected. The low resolution data used by Ludwig show considerable structure and the laser probe lines would be unlikely to fall on a major absorption line.

Fowler's suggestion that a highly excited vibrational population distribution of water vapor enhanced the absorptivity seems unlikely. In the steady-state plasma, the excitation of vibrational states is balanced by collisional de-excitation with other molecules, according to

$$\alpha I = nc_p(T_v - T)/\tau$$

where I is laser intensity, n the number density of water vapor molecules, T_v their effective vibrational temperature, T the gas temperature, and τ the vibrational relaxation time. Taking $c_p \sim R$ we find

$$I = \frac{0.1 p_{H_2O}(\text{atm})(T_v - T)/T}{(p_{H_2O}\tau)(\alpha/p_{H_2O})} \frac{\text{W}}{\text{cm}^2}$$

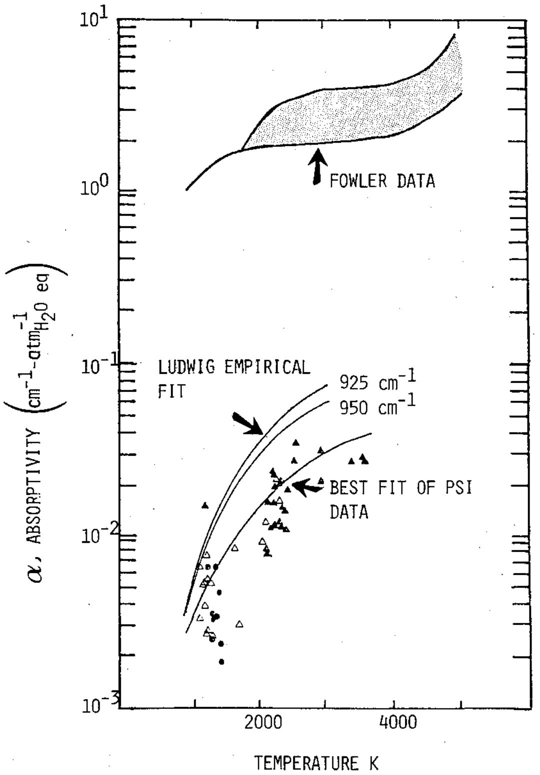

Fig. 1 Absorptivity vs temperature.

Received Sept. 29, 1981.

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As an example, take $T = 3000$ K and $p_{H_2O} = 0.3$ atm. Kung and Center[6] have measured $p_{H_2O}\tau$ as 6×10^{-9} s-atm. Putting in the measured shock tube value from Fig. 1 of $\alpha / p_{H_2O} = 3 \times 10^{-2}$ cm^{-1} atm^{-1}, we find

$$I = 10^8 \Delta T / T \quad \text{W/cm}^2$$

Thus very intense laser radiation would be required to obtain any appreciable vibrational non-equilibrium. The experiments of Ref. 1 apparently had an intensity of 10^4 W/cm^2, leading to $\Delta T / T = 10^{-4}$, and so making nonequilibrium effects very unlikely.

Acknowledgments

This work was sponsored by the Air Force Rocket Propulsion Laboratory under Contract F04611-80-C-0042 and the NASA Marshall Space Flight Center under Contract NAS8-33097.

References

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Errata: "A Comparison of Some Finite Element and Finite Difference Methods for a Simple Sloshing Problem"

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[AIAA J, **9**, 2094-2096 (1971)]

METHOD 3 of this Note is a *finite difference* method, not a finite element method. The variational principle was applied to show one way of deriving the differential equations and boundary conditions, which are then approximated by finite difference equations. The objective is to show that the finite element method generally requires more dependent variables than the finite difference method for fluid, thus requiring more computer time for the same accuracy. Note that the error in the section heading and in the text was introduced without my knowledge. Also Fig. 1 was omitted in error. This figure illustrates an finite element method by Hunt for rectangular domain which is ingeneous and yet inferior to method 3, a finite difference method. Method 3 is superior among second order methods based on truncation error, because of the combination of (extrapolated) central difference boundary conditions with the (central) difference form of the differential equation.

Received Oct. 1, 1981.

Errata: "Inviscid Solution for the Secondary Flow in Curved Ducts"

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[AIAAJ, **19**, 993-999 (1981)]

THERE was a mistake in the right-hand side of Eqs. (7) and (A10). The equations should read:

$$
\begin{aligned}
u\frac{\partial v}{\partial r} + \frac{v}{r}\frac{\partial v}{\partial \theta} + w\frac{\partial v}{\partial z} = \Big\{ & w\Big(u\frac{\partial \xi}{\partial r} + \frac{v}{r}\frac{\partial \xi}{\partial \theta} + w\frac{\partial \xi}{\partial z}\Big) \\
& + \Big[u\Big(\frac{\partial v}{\partial r} - \frac{v}{r}\Big) - v\frac{\partial u}{\partial r}\Big]\zeta + w\xi\Big(\frac{\partial u}{\partial r} + \frac{\partial w}{\partial z}\Big) \\
& + w\Big(\frac{1}{r}\frac{\partial w}{\partial \theta} - \frac{\partial v}{\partial z}\Big)\Big(\frac{v}{r} - \frac{\partial v}{\partial r}\Big) + v\Big[\frac{\xi}{r}\frac{\partial w}{\partial \theta} \\
& + \frac{\partial w}{\partial r}\Big(\frac{1}{r}\frac{\partial w}{\partial \theta} - \frac{\partial v}{\partial z}\Big) - u\frac{\partial \zeta}{\partial r} - \frac{v}{r}\frac{\partial \zeta}{\partial \theta} - w\frac{\partial \zeta}{\partial z}\Big]\Big\} \Big/ \zeta
\end{aligned}
$$

where

$$\zeta = \frac{\partial v}{\partial r} + \frac{v}{r} - \frac{1}{r}\frac{\partial u}{\partial \theta}$$

The error in Eqs. (7) and (A10) of our paper is purely a transcription error. The equation provided above is the equation actually used in the analysis and the results of the analysis are valid. The error resulted from trying to simplify the right-hand side of the equation for the purpose of publication only and was not used in the actual solution.

Received Jan. 26, 1982.

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