AD 711 614

Quarterly Technical Report

The research carried out under this contract is divided into four tasks, indicated as follows in the work statement.

(1) Application of currently available close coupling codes to the calculation of cross sections of interest to the molecular lasers under consideration (initially N_2, N_2O, CO_2, and CO).

(2) Extension of the existing computer program for the vibrational population distribution of a binary mixture of gases to include excitation and de-excitation processes specific to the laser mixtures of interest, making use of the results of the close-coupling calculations, as they become available.

(3) Application of the spectroscopic observation of side light and small signal gain measurements to the determination of vibrational and rotational population distributions in the molecular laser systems under study (initially CO_2 and CO).

(4) Preliminary computer modeling of the laser system, to include the effects of spatial variation of electron density and temperature and gas temperature.

Progress to date has been as follows:

1. Cross Section Calculations

In the previous quarterly report,[1] it was pointed out that calculations made of the inelastic cross sections for vibrational excitation of CO by collision with rare gas atoms yielded results which were in excess of the experimental results by an order of magnitude. It was felt that this discrepancy was

due to the use of a Lennard-Jones potential. An empirical potential has been developed, which corresponds to the experimental form given by Jordan et al [2] at short range, and to the Lennard-Jones potential for separations greater than the potential minimum. The case of CO-Argon collisions was chosen as a test case to study the improvement in agreement with experiment brought about by the use of this potential. The Lennard-Jones parameters for this case were taken from Hirschfelder et al.[3]

In Fig. 1, we see the specific form of the CO-Ar interaction potential. For separations less than 3.8 (in units of a_o), the potential is undefined. For $3.8 \leq r \leq 5$, the potential is taken from the measurements of Jordan et al [2], and is given by $V(r) = 47529/r^7$, in eV. In the intermediate region, $(5 \leq r \leq 7.5)$, the potential is represented by a fourth order polynomial, with coefficients chosen to ensure continuity of the potential and its first derivative at the region boundaries, and to pass through the zero of the Lennard-Jones potential. In this region, the analytic form is given by $V(r) = 0.0422\ (\sigma-r) + 0.0684\ (\sigma-r)^2 + 0.0422\ (\sigma-r)^3 + .00692\ (\sigma-r)^4$, where $\sigma = 6.79$ for CO-Ar. In the region $r \geq 7.5$, the potential is given by the Lennard-Jones form, $V(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6]$, with $\epsilon = .0087$ eV for CO-Ar.

Clearly the potential cannot validly be employed for collision energies greater than 4.2 eV. However this is more than sufficient for our present purposes.

A FORTRAN subroutine to generate this potential has been written and tested. Currently it is being incorporated in the general scattering code in order to calculate the vibrational excitation cross sections.

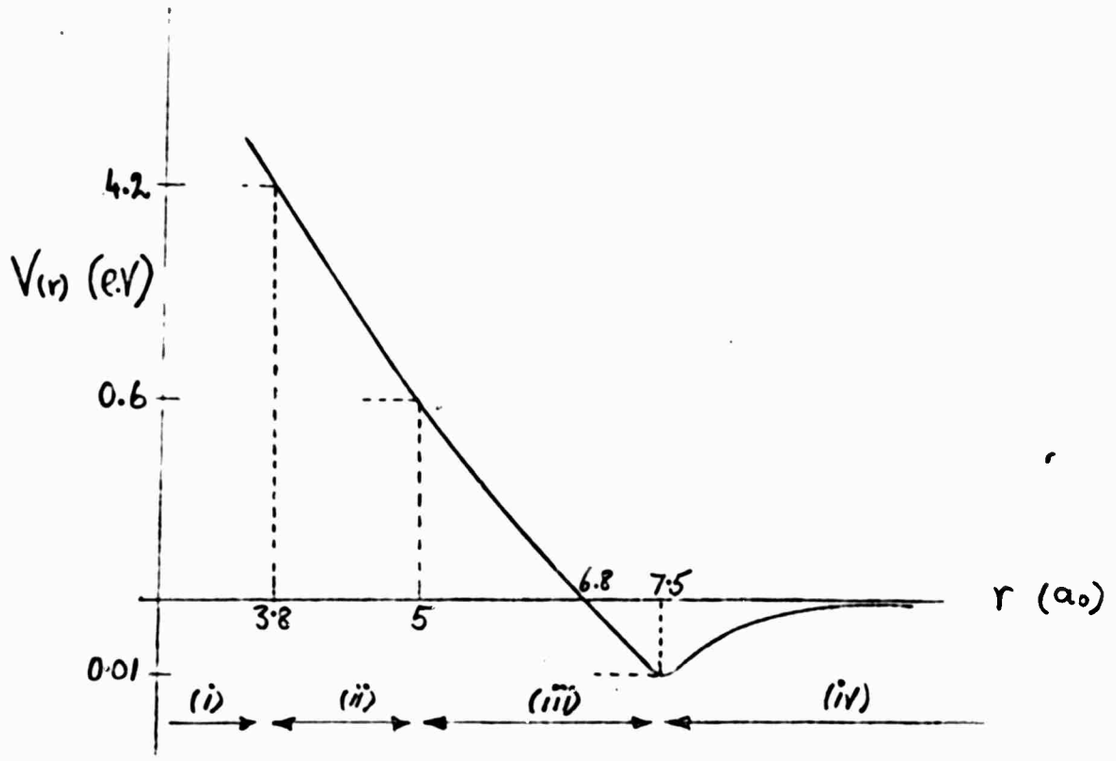

Figure 1. Empirical CO-Ar Interaction Potential.

Subsequently, these cross sections will be used to compute the effect of Ar on vibrational relaxation in CO. Comparison of these results with the available experimental data will give some indirect measure of the confidence which can be placed in the use of the constructed potential and in the application of similar approximations to collisions between CO and other rare gas atoms.

2. Vibrational Distributions

The binary mixture vibrational relaxation code has been modified to include the revised model for the vibration-vibration exchange rate coefficients and test calculations have been performed on the $H_2 + D_2$ reaction mixture. This system was chosen due to the recent experiments on raman excitation of the H_2(v=1) mode with the subsequent exchange reaction to form HD[4]. Vibrational excitation is believed to be necessary to initiate the reaction. Our preliminary calculations show that the vibration-vibration exchange processes are sufficiently rapid to redistribute the initial H_2 vibrational energy to other modes of H_2, i.e. v=2, etc. and also to D_2 (v=1,2, etc.). Thus, an unequivocal determination cannot be made of the relative importance of H_2 or D_2 vibrational energy as a reaction initiator by this experimental method.

3. Experimental Work

A pulse excitation circuit for the laser tube has been designed and constructed. Pulse durations of the order of 100 nsec, with rise times of less than 10 nsec have been obtained. An appropriate pulse forming network is being constructed. In CW operations, the output of the laser tube has been stabilized, and the gas handling system improved. Preliminary observations have been made of the infrared spontaneous emission from the 4.3 μm

fundamental of CO_2 in the CO_2 laser. Present work is directed towards improving our capabilities to make quantitative observations of various infrared emissions from the laser tube, as a diagnostic tool.

4. Plasma Properties

As a preliminary to developing a coupled electron-vibrational model program, a review of available electron-molecule excitation cross sections has been undertaken. These values will be needed in the program and the accuracy of the program results will be in a large part determined by the reliability of this cross section information.

REFERENCES

1) Technical Report No. 1, Contract No. DAHC04-70-C-0022, 25 May 1970.

2) J. E. Jordan, S. O. Colgate, I. Amdur, and E. A. Mason, J. Chem. Phys. 52, 1143 (1970).

3) J. O. Hirschfelder, F. C. Curtis, and R. B. Bird, "Molecular Theory of Gases and Liquids", New York (1954).

4) S. Bauer, Private Communication.