III. MOLECULAR ENERGY TRANSFER AND SPECTROSCOPY^{*}

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RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

1. Resonance Fluorescence Spectroscopy

a. Quenching and vibrational energy transfer cross sections have been determined for electronically excited iodine in V' = 43 (excited by the Argon laser 5145 Å line) and V' = 51 (excited by the cadmium 5086 Å line). Fluorescence depolarization measurements are being readied for the former system.

b. Inelastic scattering model calculations are being carried out in reference to the multiquantum vibrational energy transfer observed in the V' = 43 state of iodine.

c. The fluorescence of iodine vapor at high temperatures, in the presence of a significant concentration of free iodine atoms, is being investigated. It is probable that an $(I_3)^*$ collision complex may be found, and we want to find the influence of such complexes on inelastic processes occurring in the excited states.

d. The monochromatically excited resonance fluorescence method is being applied to the species Se_2 , Te_2 , and TII. The first two systems can exist in several spin states, and we want to investigate collisional transfer between these states. The last system is a good example of a molecule with long-range polar forces in the interaction potential, and we want to find the influence of these terms on inelastic processes.

2. Optical and Infrared Double Resonance

a. An infrared-infrared double resonance experiment has been completed on sulfur hexafluoride. We found that the upper vibrational levels are very rapidly equilibrated, and relaxation proceeds through the lowest (v_6) vibrational quantum. A similar experiment on boron trichloride is under way, and an infrared-ultraviolet double resonance experiment on propynal (HC = C - CHO) is being prepared.

b. The v_3 absorption in sulfur hexafluoride at 940 cm⁻¹ is being studied at very low temperatures, in collaboration with Professor K. N. Rao of Ohio State University. This work should provide a contour, free of hot bands, and suitable for computer synthesis.

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The results of this part of the analysis can then be used in the interpretation of some ultrahigh-resolution spectra that are being obtained at Lincoln Laboratory, M.I.T.

3. Dye Solution Laser

a. A flashlamp-pumped dye solution laser has been constructed and successfully operated with rhodamine and fluorescein. The temporal dependence of the output will be investigated. Also, the laser will be used as a pumping source for fluorescence life-time measurements on several molecular systems, such as I₂ and NO₂, in the vapor phase.

b. The absorption and emission spectra of some laser dyes have been obtained in both rigid plastic matrices and EPA solution, down to 4.2 °K. The data will be used to interpret the gain characteristics of the dye laser in terms of molecular energy states and associated relaxation processes.

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