

LASER INDUCED FLUORESCENCE MEASUREMENTS OF VIBRATIONALLY EXCITED OXYGEN PRODUCED BY RECOMBINATION OF O ATOMS

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Vibrationally excited oxygen and nitrogen have been long recognized to be of critical importance in nonequilibrium high-enthalpy flows, primarily encountered behind hypersonic shock waves. To date, no direct experimental verification of vibrational state resolved dissociation rates predicted by different kinetic models has been undertaken, in large part due to the difficulty of measuring these rates directly. We present a framework where state specific recombination rates can be inferred from the time-resolved measurements of O₂ vibrational populations, such that the state specific dissociation rates can be obtained from the detailed balance. In the present approach, recombination reactions of atomic oxygen are monitored in the afterglow of a diffuse ns pulse discharge burst in an O₂-Ar mixture. The time evolution of O₂(v) populations in the recombining mixture are measured by ps Laser Induced Fluorescence (LIF) in the O₂(B³Σ_u⁻, v'=0 ← X³Σ_g⁻, v'') Schumann-Runge bands, with absolute calibration by NO(A²Σ⁺, v'=0 ← X²Π, v''=0) LIF. By varying the output wavelength of the ps laser / Optical Parametric Oscillator (OPO) system used for the excitation in the 250-450 nm range, levels from v''=7 to 21 have been detected. Two-photon Absorption LIF (TALIF) at 226 nm has been used to measure the atomic oxygen concentration, with absolute calibration by Xe. Within ~1 ms after the discharge burst, a rapid decay of O₂(v) is observed, indicating vibration-vibration (V-V) and vibration-translation (V-T) relaxation of vibrational states populated by electron impact and by quenching of the excited electronic states of Ar. After the rapid initial decay, the vibrational populations level off and remain nearly constant, or exhibit a transient rise, on the timescale of about 10 ms, suggesting the presence of a persistent source of vibrational excitation due to chemical reactions. In addition to O atom recombination to form vibrationally excited O₂ molecules, at low temperatures atomic oxygen may also recombine with O₂ and form ozone. The latter may well generate vibrationally excited O₂ in O + O₃ → O₂ + O₂ reaction, which would therefore affect the present results. Ozone formation is controlled by preheating the excited mixture up to 600 K, as well as increasing the pressure and reducing the O₂ mole fraction, when the production of ozone is reduced significantly.