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Repulsive Rydberg States and Dissociative Ionization LUTZ HUWEL, HONG CHEN, Wesleyan University — We have performed experimental studies of the competition between direct and dissociative ionization of Na₂ in single ro-vibrational levels of the $2^{1}\Pi_{q}$ state. Excitation and ionization of the molecules is achieved in a molecular beam and with a two-color, three-photon optical-optical double resonance (OODR) technique. Total excitation energies range from about 700 to about 1400 cm-1 above threshold for ground state dissociative ionization. Discrimination of fragment atomic Na^+ from molecular Na_2^+ ions is accomplished with a linear Time-of-Flight spectrometer. We observe $Na^+: Na_2^+$ ion ratios ranging from about 0.05 to a about 0.15. We have developed a semi-classical model that accounts reasonably well for the observed behavior of the ratio. The model is based on the assumption that dissociative ionization occurs in competition between autoionization and dissociation along repulsive neutral Rydberg states converging to the Na⁺₂ $1^{2}\Sigma^{+}_{u}$ potential. Single adjustable model parameter is the autoionization lifetime. A chi-square procedure yields quite reasonable agreement with the experimental data suggesting autoionization lifetimes of the order of the corresponding Bohr orbit time. Details of the experiment and the model will be presented.

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