LOW-ENERGY ELECTRON ATTACHMENT AND DETACHMENT IN VIBRATIONALLY EXCITED OXYGEN AND AIR

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Three-body electron attachment to O_2 molecules and electron detachment from O_2^- ions have been theoretically studied in vibrationally excited oxygen and O_2 -containing mixtures. It was assumed that electron attachment and detachment proceed via the formation of vibrationally excited temporary O_2^- ions (the mechanism of Bloch and Bradbery). This means that electrons detach in the following way. A stable O_2^- ion is excited into a vibrationally excited temporary negative-ion state $(O_2^-)^*$ in a collision with a molecule. Then, the excited ion autoionizes into a free electron and a neutral molecule: $(O_2^-)^* \rightarrow e + O_2$.

The rates of electron attachment and detachment were determined on the basis of the statistical approach for the vibrational transfer and relaxation in collisions between O_2^- ions and O_2 molecules. Preliminary results of this work were presented in [1] in which we used the simpest version of the statistical treatment when ignoring the conservation of total angular moment. In this work, we generalized the approach used in [1] to take into account this conservation rule.

The statistical treatment assumes strong coupling collisions between reactants with the formation of an intermediate complex that decomposes according to the relative phase space available to each product state, while total energy and angular momentum are conserved. The statistical approach is based on a hypothesis and the results must be subjected to comparison with experiment to test the validity of the hypothesis. To validate the statistical approach used, we calculated attachment and detachment rate constants under conditions under which measurements are available. The obtained attachment and detachment rate constants turned out to agree well with available drift-tube measurements in unexcited oxygen.

This method was extended to calculate attachment and detachment rates in vibrationally excited oxygen. It was shown that the effect of vibrational excitation on electron detachment is profound, whereas attachment of low-energy electrons to vibrationally excited O_2 is inefficient. The calculated vibrational distribution of stable O_2^- ions turned out to be non-equilibrium in an excited gas and the effective vibrational temperature of the ions was much lower than the vibrational temperature of molecules. Figure 1 shows the calculated detachment rate constant, k_d , as a function of the vibrational temperature of O_2 , T_ν , for the gas temperature T = 300 and 1000 K. For comparison the curve corresponding to the calculation at $T = T_\nu$ is also presented in the figure.

The calculated rate constants were used to simulate the formation and decay of an electronbeam-generated plasma in N₂:O₂ mixtures at elevated vibrational temperatures. Figure 2 compares measurements [2] and the calculated evolution in time of the electron density in atmosphericpressure air under the conditions typical to the experiment [2]. For comparison, the results calculated in unexcited gases are also given. Our calculations show that vibrational excitation of molecules leads to an increase by more than an order of magnitude (i) in quasi-steady electron density in the electron-beam-generated plasma and (ii) in the plasma lifetime. Under the conditions considered, the calculated time $\tau_{1/2}$ it takes to reduce initial electron density by half after the electron beam is turned off increased from 0.04 to 2 μ s as T_v increased from 300 to 2500 K and the electron temperature T_e increased from 300 to 5000 K. The value of $\tau_{1/2}$ measured in [2] at $T_v =$ 2500 K and $T_e = 5000$ K was around 5 μ s, in qualitative agreement with the calculation.



Fig. 1: The detachment rate constant as a function of vibrational temperature of O_2 for T = 300 and 1000 K and for $T = T_y$.



Fig. 2: The evolution in time of the electron density in atmospheric-pressure air plasma generated by an electron beam of duration 20 μ s. Solid curve corresponds to $T_v = 2500$ K, $T_e = 5000$ K and T = 560 K and dotted curve corresponds to $T_v = T_e = T = 300$ K. Symbols correspond to measurement [2].

References

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