Reaction-rate measurements of cold ion-polar molecule reactions using a combined Stark-velocity-filter–ion-trap apparatus†

K. Okada,*1,*2 T. Furukawa,*2 T. Takayanagi,*2 H. A. Schuessler,*3 and M. Wada*1

Cold molecules and their ions are attractive subject of research in the fields of fundamental physics and cold chemistry. With regard to cold chemistry, the reaction-rate constants of cold ion-molecule reactions are important information for studying the chemical evolution of interstellar clouds[1]. Recently, we have developed a combined Stark-velocity-filter–ion-trap apparatus for measuring the reaction rate between cold trapped ions and slow polar molecules under ultra-high vacuum conditions[2]. We experimentally measured the reaction rates between sympathetically cooled N_2H^+ ions and velocity-selected polar molecules, namely CH_3CN.

The measurement procedure is as follows. First, we produce a Ca^+ Coulomb crystal in a linear Paul trap. Then a nitrogen gas of about 1×10^{-7} Pa is introduced into the vacuum chamber, and an electron beam is incident to the center of the ion trap in order to produce N_2^+ ions by electron impact ionization. Because the mass of the nitrogen molecular ion is lighter than that of Ca^+, the molecular ions are more tightly bounded by the trapping potential and accumulate near the trap axis. After the preparation of cold N_2^+ ions, a hydrogen gas of about 6×10^{-6} Pa is introduced into the vacuum chamber. All N_2^+ ions change into N_2H^+ ions via the reaction of $N_2^+ + H_2 \rightarrow N_2H^+ + H$ in a reaction time of 240 s[3].

After the preparation of cold N_2H^+ ions, we irradiated the velocity-selected CH_3CN molecules to the two-species Coulomb crystal containing Ca^+ and N_2H^+ ions. Figure 1(a) shows the snapshots of the laser-induced fluorescence (LIF) images of the Coulomb crystal at several reaction times. The dark area containing N_2H^+ progressively decreases with increasing reaction time owing to the progress of $CH_3CN + N_2H^+ \rightarrow CH_3CNH^+ + N_2$ reactions. We also observed an increase in the sparse dark area in the outer peripheral region of the Ca^+ Coulomb crystal because a part of the reaction products (CH_3CNH^+) is trapped. Under the present experimental conditions, the average reaction energy is estimated to be approximately 3 K[2].

In order to obtain the reaction rate, we determine the relative number of molecular ions from the volume of the dark area in the observed fluorescence images under the assumption of a constant number density at 0 K. Figure 1(b) shows the decay curve of the relative number of N_2H^+ ions as a function of the reaction time. In this example, the reaction rate is determined to be $2.4(4)\times10^{-3}$ s^{-1}. We performed 9 measurements and obtained an averaged reaction rate of $2.0(2)\times10^{-3}$ s^{-1}. Using the number density of the velocity-selected CH_3CN, which was separately determined, the reaction-rate constant was also determined to be $1.7(6)\times10^{-8}$ cm^3 s^{-1}. The main reason for the error is considered to be the uncertainty in the number density of CH_3CN[2]. The present reaction-rate constant is consistent with the estimated capture rate, $k_{ts} = 3.6\times10^{-8}$ cm^3 s^{-1}, using the trajectory-scaling formula[4], which is considered to be the maximum value of the reaction-rate constant. In the future, the present velocity filter combined with a cryogenic trap apparatus will enable us to perform systematic measurements of cold ion-polar molecule reactions, which are important problems from a fundamental viewpoint and contribute to astrochemistry.

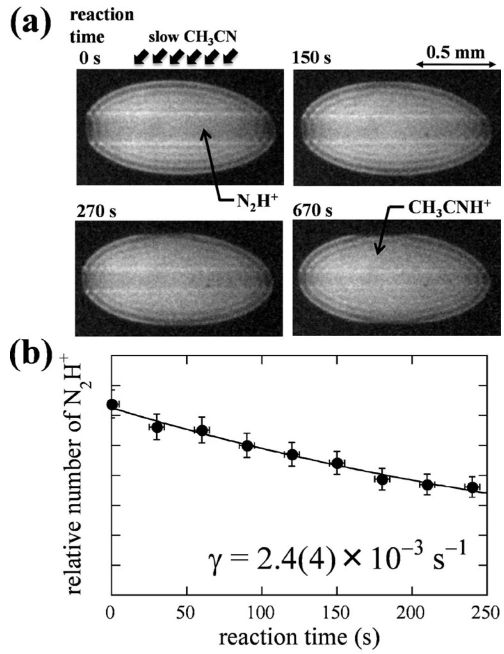

Fig. 1. (a) Sequential LIF images of the two-species Coulomb crystal containing Ca^+ and N_2H^+ during $CH_3CN + N_2H^+ \rightarrow CH_3CNH^+ + N_2$ reactions. (b) Plot of the relative number of N_2H^+ ions as a function of the reaction time.

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*1 RIKEN Nishina Center
*2 Department of Physics, Sophia University
*3 Department of Physics, Texas A&M University

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