Cross-correlation TOF Method by Pseudorandom Pulse Discharge

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A new cross-correlation time-of-flight (CC-TOF) method for pulsed metastable atom beam is demonstrated. In this method, we modulate the grid voltage for the metastable atom formation with pseudorandom pulse sequence instead of mechanical chopping. The TOF spectrum convoluted from the observed CC-TOF spectrum is found to nicely reproduce the conventional TOF spectrum, indicating useful applicability of the new method.

Translational energy dependence of reaction cross section in molecular beam studies is important to understand detailed reaction dynamics. In crossed beam experiments, only weak signal intensity is unavoidably expected. CC-TOF method has been known as a powerful method to improve S/N ratio.[1,2] A mechanical beam chopper mostly has been used in CC-TOF method. Comsa and co-workers pointed out the following demerits in using a mechanical beam chopper.[1] (1) instability of rotation frequency and short endurance of the motor (2) the machinery limitation of the slit's correctness. Recently, Kishimoto and co-workers overcome such demerits and give a excellent result by using a mechanical beam chopper.[3] In general sense, it is desirable to develop a complementary CC-TOF methods without mechanical chopper.

In this letter, we demonstrate a new method that the pseudorandom pulse sequence of metastable Ar($^3P_{0,2}$) (Ar*) beam is directly prepared by the modulation of grid voltage in the formation by glow discharge. The experimental apparatus is shown as Figure 1. When the grid voltage is positive with respect

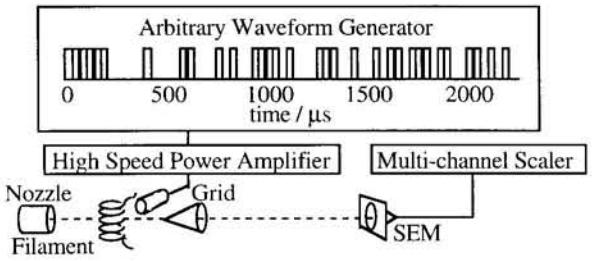

Figure 1. Schmetic experimental apparatus with CC-TOF method by pseudorandom pulse discharge.

to the filament, Ar* beam is excited by glow discharge between grid and filament. Therefore, the grid voltage-on corresponds to the open state in mechanical chopping. The response time of the glow discharge to the grid pulse was measured to be less than 2μs by the comparison of time profile of the stray light from glow discharge with the grid pulse. This response time is estimated to result from both the flight time of ion species and the lifetime of the emitting excited species in the glow discharge. The fluctuation was not measured with time resolution of 40ns. Therefore, the actual fluctuation of the pulse width is expected to be less than 0.4% for the 10μs pulse. It would be possible to improve the response time by changing the distance between the filament and the grid electrode. The desired distance between filament and grid electrode for the grow discharge also depends on the kind of gas and it's stagnation pressure. In the present study, the distance was set to be 8mm with the stagnation pressure of 60 Torr for Ar beam. The endurance of CC-TOF method is significantly improved as compared with CC-TOF method with a mechanical beam chopper. Continuous operation at least 6 months is achieved in the present stage. It is also convenient that the time resolution can be changed easily by changing the pseudorandom pulse sequence in the arbitrary waveform generator.

A practical problem in the new method is the effect from finite size of the glow discharge region. Figure 2 shows the

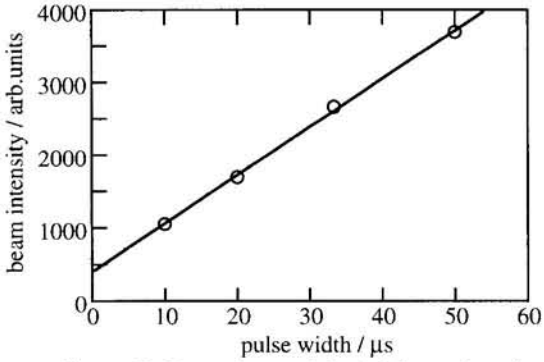

Figure 2. Dependence of the Ar* beam intensity on the pulse width of glow discharge.

dependence of the Ar* beam intensity on pulse width of one pulse discharge. Although very nice linear relationship is obtained between the Ar* beam intensity and the pulse width, the extrapolated line does not pass through the origin. The intercept would be attributed to the finite size of the glow discharge region. Thus, the time resolution in new method is mainly limited by the size of the glow discharge region. This excessive discharge time, t_0 due to the finite size of the discharge region depends on the discharge condition such as kind of gas, stagnation pressure, and the setup structure electrodes. In order to minimize the size of the glow discharge region, the grid electrode is made by a stainless rod of 2mmϕ in diameter. In the present condition, t_0 is the time interval of 5 μs, which corresponds to ca.3mm length of discharge region. The glow discharge was found to be somewhat unstable for the pulse width shorter than 5μs. In a practical sense, the limit of the time resolution in new method is better than 15μs.

The pseudorandom pulse sequence edited in an arbitrary waveform generator is amplified to 60 V by a high speed power amplifier. The pulse width is set to be 36 μs per pulse. The pseudorandom pulse sequence which consist of 63 pieces is used on the basis of the ref. 4. We take into account t_0 in editing the pseudorandom pulse sequence. That is, a pulse of 36 μs must be edited as the combination of 31 μs pulse and 5 μs

pulse with no voltage. The discharge time over all sequence must be longer to cover a total time of TOF distribution of Ar* in conventional TOF experiments. To confirm TOF spectrum with CC-TOF method, the conventional TOF is also measured using one pulse discharge of 31 μs width.

Ar* beam is detected by a secondary electron multiplier (SEM) with a hole of 1mmϕ in diameter at the distance of 1100mm from the nozzle and the time profile from the SEM is measured by a multi-channel scaler. The Ar* beam intensity is measured to be ~10^{15} atoms $sr^{-1}s^{-1}$.

Since 5ms-pulsed Ar beam is discharged using only one cycle pseudorandom pulse sequence in the present experiment, a general correlation function which is obtained under the cyclic continuous pseudorandom sequences, can be constructed by summing up the time evolution function with that which is shifted by one period of discharge cycle. TOF spectrum can be convoluted by Hadamard transformation procedure using the correlation function. In Figure 3, the TOF spectrum convoluted from the

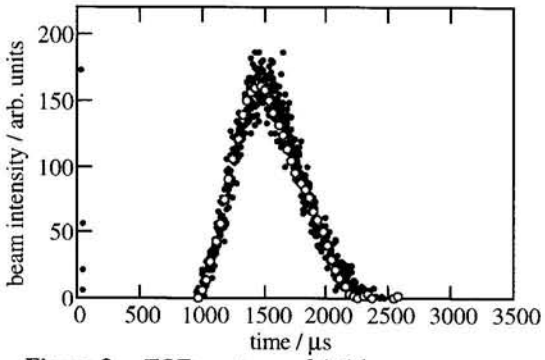

Figure 3. TOF spectrum of Ar* beam.
the filled circles; conventional TOF method, the open circles; present CC-TOF method.

CC-TOF method is shown with the open circles together with the conventional TOF spectrum. The TOF spectrum convoluted from the CC-TOF method is found to nicely reproduce the spectrum obtained by a conventional TOF method, indicating usefulness of this new method.

By applying the present technique to a crossed beam experiment with an effusive molecular beam, we measured the translational energy dependence of the reaction channel having less than $10^{-21}m^2$ cross section with the flight length of 900mm. As an example, the translational energy dependence of CClF(A) formation from the reaction of Ar*+CCl_3F is shown in Figure 4.[5] Furthermore, by using a supersonic molecular beam under

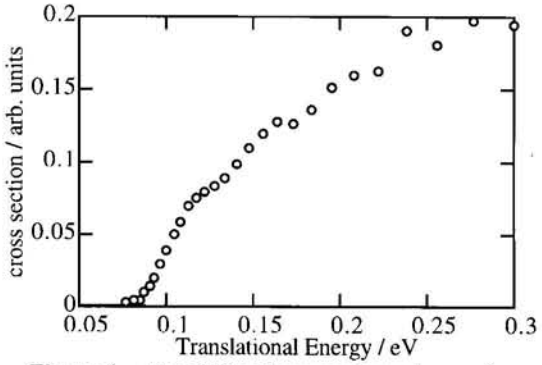

Figure 4. Translational energy dependence of CClF(A) formation cross section in the reaction of Ar*+CCl_3F

the short flight length, the reaction channel having less than $10^{-23}m^2$ cross section will be studied within the suitable time resolution.

In future, translational energy dependence of steric opacity function would be measured by the application of this CC-TOF method to oriented molecular beam experiments.[6,7]

References and Notes

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