Experimental measurement of rotational energy distribution
in nitrogen molecular beam by REMPI

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Abstract Recently, the importance of the high Knudsen number flow, such as rarefied gas flows and micro/nano flows has increased significantly. In such flows, the gas-surface interaction affects the flow field. To investigate the gas-surface interaction, molecular beams have been widely used. The characteristics of molecular beams were mainly investigated on the velocity distribution by the time of flight method: there are still few reports on the rotational energy distribution. Conventional spectroscopic methods are difficult to be applied because of the low number density. In this study, therefore, Resonantly-Enhanced Multiphoton Ionization (REMPI) method, which is a high detection sensitivity technique, has been applied to obtain the rotational energy distribution in a nitrogen molecular beam. 2R+2 N²-REMPI process was used to detect the nitrogen molecular beam. The molecular beam was generated by the Kantrovitz-Grey type beam source with a 50 µm orifice at the stagnation pressure and temperature of 50 kPa and 293 K, respectively. The estimated translational temperature was 27.7 K under an isentropic assumption. The obtained REMPI spectrum was fitted with the theoretical spectrum by changing the rotational temperature as a parameter under the assumption of the Boltzmann distribution for rotational energy levels. The fitted rotational temperature was 29.3 K, which is slightly higher than the estimated translational temperature. This result indicates that the rotational temperature deviates from the translational temperature and is “frozen” before the beam arrives at the terminal state. Investigation of the degree of deviation by changing the value of the product of the stagnation pressure and the orifice diameter should give much more insight into the phenomenon.

1. Introduction

Recently, the importance of so-called “high Knudsen number flow,” where the Knudsen number, the ratio of mean free path to the characteristic length, is large, has increased significantly in both academic and industrial fields. The Knudsen number of the flow becomes high not only when the mean free path is large as in rarefied gas flows, but also when the characteristic length of the system is small such as gaseous flows around/inside micro/nano devices. In such high Knudsen number flows, the collision number of molecules with surfaces becomes much larger than that with other molecules. Therefore, gas-surface interaction is very important and largely affects the flow field. To investigate the detailed physics of the gas-surface interaction, the molecular beam scattering experiment (Scoles 1988) is one of the powerful and the popular tool. The characteristics of the molecular beam should be clear to use it as a probe for the gas-surface interaction, but there are still few reports on experimental analyses of the rotational energy distribution in the molecular beams. However, conventional spectroscopic methods such as LIF (Eckbreth 1988) and EBF (Muntz 1962) are difficult to be applied because of the low number density in molecular beams.

In this research, the Resonantly Enhanced Multiphoton Ionization (REMPI) method (Carleton 1985) has been applied to the rotational energy distribution measurement in a nitrogen molecular beam. In the 2R+2 N²-REMPI process, 4-photon ionization process of nitrogen molecules is divided into 2-photon excitation from ground (X²Σg⁻) state to the resonant (a¹Πg) state and additional 2-photon ionization from the resonant state. REMPI spectra reflect the rotational energy distribution in the ground state of neutral molecules directly. REMPI also has very high detection sensitivity because of the direct ion detection, and is a powerful tool for the detection of very low density gas.
2. Experimental Setup

Experimental apparatus is shown in Fig. 1. A frequency-doubled Nd:YAG-pumped dye laser is used for a laser source. The ionized nitrogen molecules are detected by a secondary electron multiplier. The signal intensity is averaged by a boxcar integrator and recorded on a personal computer. The molecular beam is generated by the Kantrovitz-Grey type beam source with a 50 µm orifice at the stagnation pressure and temperature of 50 kPa and 293 K, respectively. The experimental apparatus consists of a three-stage differentially pumped molecular beam line, and an ultrahigh vacuum detection chamber.

3. Results and Discussions

Figure 2 shows a obtained REMPI spectrum with the Fortrat diagram indicating the relation between each rotational level and the corresponding wavelength, where the experimental data is plotted by black dots. The translational temperature at the measuring point is estimated as 27.7 K for the terminal Mach number of 6.92 under an isentropic assumption. The theoretical REMPI spectrum for a rotational temperature of 27.7 K, which is in equilibrium with the translational temperature, is also plotted by a dashed line. As seen in Fig. 2, the experimental data seems to be well fitted by the theoretical spectrum. It should be noted that, when the experimental data was fitted with the theoretical spectrum by changing the rotational temperature as a parameter under the assumption of the Boltzmann distribution for rotational energy levels, the obtained value was 29.3 K, which is slightly higher than the estimated translational temperature. The REMPI spectrum indicates that the rotational temperature deviates from the translational temperature in the molecular beam. It is expected that the rotational temperature is “frozen” before the beam arrives at the terminal state and deviates from the translational temperature in a highly rarefied gas flow (Marrone...
Fig. 2 REMPI Spectrum obtained in N₂ molecular beam plotted with the theoretical simulated spectrum for the rotational temperature in equilibrium with translational temperature 27.7K with a line width of 6.8 cm⁻¹.

1967, Mori 2005). The estimated terminal rotational temperature following the results of free-jets (Marrone 1967) is about 32 K. Therefore, it is consistent with the fitted result mentioned above. The deviation between the rotational and the translational temperature is known to be affected by the value of \(p_0d\), where \(p_0\) is the pressure in a stagnation chamber and \(d\) is the diameter of the orifice. It is also interesting to investigate the degree of deviation by changing the value of \(p_0d\).

To analyze the rotational energy distribution in detail, the Boltzmann plot using the REMPI spectrum is very important, because the number density of the molecular beam is extremely low. Thus, the S/N of the experimental system should be improved. We also have to consider carefully that the obtained experimental data is evaluated as “broad” spectrum with a line width of 6.8 cm⁻¹, even though a line width of our laser is 0.1 cm⁻¹. Refinement of the system is important for further discussion.

4. Summary

The rotational energy distribution in a nitrogen molecular beam was successfully obtained by the 2R+2 N₂-REMPI method. The molecular beam was generated by the Kantrovitz-Grey type beam source. The REMPI spectrum of the molecular beam indicates the rotational temperature higher than the translational temperature estimated under the isentropic assumption. This result indicates that the rotational temperature is “frozen” before the beam arrives at the terminal state. Investigation of the degree of deviation by changing the value of the product of the stagnation pressure and the orifice diameter should give much more insight into the phenomenon.
Acknowledgements

The present work was supported by a grant-in-aid for Scientific Research from the MEXT and JSPS, Japan.

References


