2 Experiment

The experiments were performed by bombarding C_{60} gas and CH_4 gas targets with noble gas ions at a few keV of incident energies. The angular distributions of the scattered atoms were measured in a wide angular range. The experimental setup was as follows.

2.1 Source of Noble Gas Ions

First, an ion beam was produced by a Colutron ion source system [66]. A schematic diagram of the Colutron ion source is shown in Figure 3. The Colutron ion source includes an ion source chamber, a voltage difference electrode for ion acceleration, an Einzel lens for beam focusing and a Wien-filter for selection of the mass-charge-ratio. The noble gas atoms He, Ne, Ar and Xe were ionized in a standard plasma ion source. The plasma ion source chamber was made of Boron-Nitoride which is a high-heat-resistant and insulator. There are only a helical filament and an anode plate in the source chamber. The voltage difference between the filament and the anode was set between 50 to 150 V. When the filament current is up to 11 A for a 15 mil filament, thermal electron emission occurs and electrons are accelerated to the anode plate by the voltage difference. At this time noble gas flows into the ion source chamber, then an arc discharge plasma is produced in the ion source chamber which discharges between the filament and the anode at several 100 μA. In the plasma, the noble gas atoms become positive ions. Charge numbers up to +3 for the ions. The larger the charge number, the smaller the ion density and ion current. The ions were accelerated up to 10 keV. The intensity of the ion beam was on the order of a μA D.C. out of the Wien-filter

2.2 Beam Pulsing System

The experimental setup for the angular distribution measurements for the atomic collisions of beam atoms with target gas atoms is shown in Figure

4. Before the ion beams are injected into the target, they are pulsed by a beam pulsing system. It is composed of two sets of ion deflectors with high voltage controlled by a square wave pulse generator and several slits. The deflectors' lengths along the ion beam array are 30 mm and 10 mm. The gaps between the two sets of electrodes are 10 mm. There are two slits locate in front of the deflectors and two slits in back of them. The upstream ones collimate the ion beam, and the downstream ones chop the beam. The DC beam was usually deflected by the statistical electric field of the deflector. The first deflectors and second produce electric fields opposed to each other. When both of them were reversed at the same time, the ions' trajectories were changed depending on their location at the time of the reversal. Only the ions located at 10 mm from the entrance of the first deflector are able to exit the last pair of slits and become a pulsed beam. The control signal of the high-voltage pulse generator is used as a pulse timing trigger. The time width of the pulsed beam can be controlled by changing the magnitude of the voltage. The time width was adjusted to maintain balance between the time resolution and a high reaction rate. Usually we chose a time width of several 10 ns with which we had an enough time resolution for the our experiments. The flight length from the ion source to the target was about 1.5 m, and the background gas pressure was kept less than 10^{-6} Torr.

2.3 Supersonic Molecular Beam Source for CH_4 Gas Target

A molecular-beam-type supersonic gas flow was developed as the CH_4 gas target (see Figure 5). The supersonic gas flow is oriented dominantly in the forward direction because of its strong cooling with adiabatic expansion. Therefore the gas density can be kept high with a comparatively low background pressure. CH_4 gas molecules are accelerated to the exit of the nozzle in Figure 5 by a pressure difference $(P_0 - P_d)$. The flow may reach sonic speed, i.e., a Mach number of 1. A skimmer slit ($d_s = 0.5$ mm) sets in front of the Mach disk at a distance L_s from the exit of the nozzle and directs

the supersonic flow to a collision point within the noble gas ion beam. The Mach number of the supersonic flow at the skimmer (M_s) can be expressed approximately as [67]

$$M_s \sim 2.75\sqrt{\frac{L_s}{d_n}} \tag{1}$$

where d_n is the nozzle diameter. This formula is in good agreement with the experimental results [69, 70]. As the molecular beam ejected through the skimmer spreads in transverse direction, the gas density (n_t) at a distance L_t from the skimmer is evaluated by the equation [67, 68],

$$n_t = \frac{P_0}{k_B T_0}\left(\frac{d_s}{2L_t}\right)^2\left(\frac{3}{2}+\frac{\gamma}{2}M_s^2\right)\left(1+\frac{\gamma-1}{2}M_s^2\right)^{-1/(\gamma-1)} \tag{2}$$

where k_B is the Boltzmann constant; T_0 is the gas temperature in the nozzle, usually assumed to be at room temperature; d_s is the diameter of the skimmer slit; and γ is the specific-heat ratio which is 1.31 for CH_4 gas. Eqn.(2) obviously indicates that n_t increases as M_s decreases; therefore, M_s should be as small as possible. The target gas passing through the collision point is pumped down through a duct effectively by a Turbo Molecular Pump (TMP-2).

To keep the main chamber pressure $\leq 10^{-6}$ Torr, differential pumping must be introduced within the chamber between the nozzle and the skimmer by a Mechanical Booster Pump (MBP), whose effective pumping speed (S_d) is 9 ℓ/s. Gas leaking out of the supersonic flow around the skimmer slit is pumped down by the main chamber pump (TMP-1, effective pumping speed S_{bg}=200 ℓ/s). The gas flow conductance of the sonic nozzle is given by $C_n \simeq \alpha_n d_n^2$, and the molecular flow conductance of the skimmer slit is $C_s = \alpha_s d_s^2$. The relation between P_0 and the background pressure (P_{bg}) around the collision point is

$$P_0 \simeq \frac{S_d S_{bg}}{\alpha_n \alpha_s d_n^2 d_s^2} P_{bg} \tag{3}$$

where $\alpha_n = 0.21$ mm$^2 \cdot \ell$/s for CH_4 [71] and $\alpha_s = 0.12$ mm$^2 \cdot \ell$/s at room temperature.

We adjusted L_s from 0.5 to 3.5 mm. For small L_s, the Mechanical Booster Pump doesn't work well. As L_s increases, the Mach number becomes larger, and the target density becomes smaller, as indicated by Eqn. (2). Therefore an L_s of 1.5 mm provided the best performance in our experiments. We adjust the parameters to P_0 = 1.0 Torr; T_0 = 300 K, which is room temperature; and L_t = 10.0 mm. From Eqn. (2) the target density was estimated to be 2.54×10^9 CH_4·molecules/mm^3. The background pressure was estimated using Eqn. (3) to be 8.75×10^{-7} Torr, and it was measured to be 1.0×10^{-6} Torr. So, apparently the gas target apparatus works well. The ambient gas percentage relative to the CH_4 gas density is 1.6% or so. The target gas extension at the ion beam (D_t) is 5.2 mm in diameter, and the target gas density distribution becomes uniform due to a property of supersonic gas flow [70], i.e., $N_0 = n_t$ and $n(x) = 1$ in Eqn. (10).

2.4 Effusive Molecular Beam Source for C_{60} Gas Target

For the C_{60} target, a gas beam target was developed using a vaporizing oven. A schematic illustration of the C_{60} gas target system is shown in Figure 6. To prepare the C_{60} gas target, a C_{60}/C_{70} (ratio is 89 to 11) powder was heated to a temperature between 500 to 580 °C, usually 530 °C, in an aluminum oven located at the center of the scattering chamber. The vapor pressure of C_{60} has been measured over the temperature range 400-600 °C by J. Abrefah *et. al.* [72]. The vapor pressure is evaluated by an equation fitted with a least squares estimated to their data, i.e.,

$$\log P = 0.0312 \times T - 16.3 \tag{4}$$

where P (in mTorr) is the vapor pressure, and T (in °C) is the temperature of the C_{60}. According to Eqn. (4), at 530 °C, the vapor pressure of C_{60} is about 1.27 mTorr. The evaporated C_{60} is emitted as effusive flow into the scattering chamber at 10^{-7} Torr through a thin orifice which a diameter of 3 mm. A shielding cover with a 10 mm diameter hole is located 6 mm from

the orifice, and the distance between the orifice and the beam line, y, is 9 mm, so the spread radius of the C_{60} gas target at the beam line is 7.5 mm.

The density of the gas emitted from the orifice as effusive flow varies according the follow formula,

$$\rho(r,\theta) = \frac{n_0 A_s \cos\theta}{4\pi r^2} \tag{5}$$

where θ is the angle of emission from the orifice, n_0 is the number of atoms per unit volume inside the orifice, A_s the hole area of the orifice, and r the distance from the orifice [68]-[71]. From this formula, the density of C_{60} at a point x mm from the center of the target region along the beam line and y mm above the orifice is

$$\rho(x,y) = N_0 \cdot n(x), \tag{6}$$

where

$$N_0 = \frac{A_s n_0}{4\pi y^2} \tag{7}$$

and

$$n(x) = \left(\frac{y}{\sqrt{x^2+y^2}}\right)^3. \tag{8}$$

This equation is important in calculating the effective solid angle and target thickness. The value of y was set to 9 mm in our calculation, corresponding to the center of the noble gas beam line. The target density at the center, i.e., N_0 was estimated to be 2.4 $\times 10^8$ C_{60}·molecules/mm^3 at 530 °C. The C_{60} gas which has passed through the beam line is trapped on a copper plate cooled with liquid nitrogen. The background pressure is usually under 3×10^{-7} Torr (that is, 9.7×10^6 molecules/mm^3) when the oven heated up at 530 °C. By our estimates, the probability of multi-collision in the region of the C_{60} gas target was low. Moreover when the target temperature was changed from 500 °C to 580 °C, the number of collision events was in proportion to the beam intensity, indicating that primarily only single collisions were occurring.

2.5 Particle Detectors

The scattering particles were detected using a Micro Channel Plate (MCP) detector [73]. When a particle hits the MCP's surface, fast pulse-signal comes from an anode located behind the MCP. The average signal height is about -30 mV, and the width is less than 2 ns. We use a fast pre-amplifier, whose gain is 8, on the analog signal from the anode before input a discriminator which works to cut the electrical noises for threshold level and converts a signal in excess of the threshold into a for standard NIM level logic signal. A threshold must be set so that it removes the ground noise, but does not chop off the MCP's signal which has broad pulse height distribution. We checked the saturation level by changing the threshold level in the experiment as shown in Figure 7.

Usually it is difficult to evaluate the efficiency of an MCP detector. For neutral He atoms and He^+ ions there is a study about the efficiency [74]. According to this report, the detector efficiency for He atoms is almost uniform over the energy range from 1 to 10 keV. However, there is no experimental data for carbon atoms; therefore, we appraised the efficiency of the MCP for carbon atoms by comparing the experimental results for CH_4 with theoretical calculations. Because the CH_4 experimental data agreed with the theoretical calculations, we inferred that the detection efficiency of the MCP is no problem in these experiments.

The stability of the gas target density was observed with a Micro Sphere Plate (MSP) detector [75] located at a scattering angle of 45 ° during the measurements of angular distribution with the MCP. The MSP is a new type of electron multiplier. It is similar to the MCP; in fact, the way it operates is completely same. By dividing the particle counting yield of MCP for measuring angular distribution by the counting yield of MSP, Y_{45}, the instability of target density was canceled out in measurements for each angle.

2.6 Time of Flight Measurements

Using the pulsed beam trigger signal as a start signal and the MCP detection time as a stop signal, a Time of Flight (TOF) measurement has been done to identify the scattered particles. Typical TOF spectra of scattering particles at different angles are shown in Figures 8 and 9. In each of the measured TOF spectra of scattered particles, the peak structures are well defined. By assuming elastic collision, the energy of the scattered particle can be calculated from the TOF and vice versa, and, consequently, the particle can be identified. The results are shown in Figure 10. The position of the peaks in the TOF spectra (Figures 8,9) correspond correctly with Figure 10. From the measurements of TOF, it is clear that quasi-elastic collisions of atoms occur in C_{60} and CH_4. In order to obtain the total collision event rate for carbon, we integrated over the width of the carbon count peak.

Similarly, the peaks of the recoiled carbon atoms were integrated to deduce the differential cross sections. In Figure 11, the particle identification has been done easily because the carbon atoms were recoiled over a wide range of angles from 0° to 90° in the laboratory system under the kinematical conditions of our study and therefore, the quasi-elastically recoiled carbon atoms could make an well-defined peak structure at each angle.

The incident energies of the different noble gas atoms were chosen so as to keep the variation in the energy of the quasi-elastically recoiled carbon atoms small. This was because too much of a change in the energies of the recoiled carbon atom might have resulted in a significant change in the detection efficiency of the MCP. The energy of the recoiled carbon atom in the laboratory system E_r is given by

$$E_r = \frac{4m_1m_2}{(m_1+m_2)^2}\cos^2\theta_L \times E_0 \tag{9}$$

where m_1 is the mass of the projectile, m_2 is that of the target atom and E_0 is the energy of the projectile in the laboratory collision system. We chose the injection energy of the noble gas atoms as 3.2 keV for Ne, 4225 eV for Ar and 9818 eV for Xe. Since the energy of the scattered particle couldn't be

directly measured with our setup, the recoiled carbon energies were deduced from the flight times. In order to calculate the energy, the flight length (i.e., the distance between the target center and the MCP detector) and the collision time are needed. The results are shown for Ne (3.2 keV) + C in Figure 12, for Ar (4225 eV) + C in Figure 13 and for Xe (9818 eV) + C in Figure 14. The experimental results are well accounted for by the theoretical elastic scattering curves in the figures.

The width of the TOF peaks of recoiled carbon in the collision Xe^+ (9818 eV) + C_{60} are shown in Figure 15. The values of "full width at half maximum" (FWHM) of carbon peak are well accounted for by the solid curves estimated from the experimental setup and elastic-kinematics calculations. Therefore, elastic scattering adequately accounts for the results of the experiment within the sensitivity of our equipment.

2.7 Effective Solid Angle of the Detector

The number of scattering events, Y, is formalized as follows,

$$Y(\theta_L) = I \cdot \left(\frac{d\sigma}{d\Omega}\right)_L N_0 \cdot f(\theta_L) \cdot \eta \tag{10}$$

where θ_L is the scattering angle in the laboratory frame; I is the number of projectile atoms; $\left(\frac{d\sigma}{d\Omega}\right)_L$ is the differential cross section in the laboratory collision system; N_0 is the target gas density at the center of the target area (see Figure. 16); $f(\theta_L)$ is a function which includes the distribution of the target density $n(x)$ along the beam line, the effective range of the gas target, $R(\theta_L)$, and the solid angle $\Delta\Omega_L(x, \theta_L)$ from the collision point x; and η is the detector efficiency.

The solid angles at the collision points along the beam axis are different because the observable collision area is limited by the double slit in front of the MCP. A schematic of the double slit setup is shown in Figure 16. The beam axis is coordinated by as x with $x = 0$ the center of the target. The two slits are circular holes set with the same solid angle from the center

point, i.e.,

$$\frac{d_1}{\ell_1} = \frac{d_2}{\ell_2}. \tag{11}$$

The lengths ℓ_1 and ℓ_2 from the center to the slit are $\ell_1 = 40$mm and $\ell_2 = 120$mm. The hole diameters d_1 and d_2 of the slits are 1 mm and 3 mm for the CH_4 experiments and 3.3 mm and 10 mm for the C_{60} experiments. The solid angle at $x \neq 0$ is determined from the overlap of the second hole with a light spot formed by projecting light from the off-center point x through the first hole and onto the second hole. When the rotatable detector MCP is set at an angle θ_L, the solid angle becomes a function of x and θ_L.

The region of upstream along the beam line from the center is called region 1 and the downstream, region 2 in Figure 16. In Eqn. (10), $f(\theta_L)$ is calculated from the geometry depicted in the schematic, as follows,

$$\begin{aligned} f(\theta_L) &= [n(x) \cdot R(\theta_L) \cdot \Delta\Omega_L(\theta_L)] \\ &= \int_0^a n(x) \frac{A(x,\theta_L)}{L_1(x,\theta_L)^2} dx + \int_0^b n(x) \frac{A(x,\theta_L)}{L_2(x,\theta_L)^2} dx \end{aligned} \tag{12}$$

where a is a limit point of area 1 and b is one for area 2, $A(x,\theta_L)$ is the solid angle at x and $L_{1,2}(x,\theta_L)$ is the distance between the collision point x and the detector. The value of a and b are described approximately as follows,

$$a \simeq \frac{\ell_1 d_2}{(\ell_1 - \ell_2)\sin\theta_L - d_2\cos\theta_L} \tag{13}$$

and

$$b \simeq \frac{\ell_1 d_2}{(\ell_1 - \ell_2)\sin\theta_L + d_2\cos\theta_L}. \tag{14}$$

But, if a or b is larger than the target spread radius, r_0, which is 2.6 mm for CH_4 and 7.5 mm for C_{60}, the value of a or b must be put at r_0.

The solid angle at x is calculated with the following formula,

$$A(x,\theta_L) = \frac{{d_2}^2}{2}(\phi - \sin\phi\cos\phi), \tag{15}$$

where

$$\phi = \cos^{-1}\left(\frac{h(x,\theta_L)}{d_2}\right), \tag{16}$$

and $h(x, \theta_L)$ is the distance between the center of the 2nd slit and the center of the light spot from the first slit, which is approximately described as follows,

$$h_1(x, \theta_L) \simeq \frac{x(\ell_2 - \ell_1)\sin\theta_L}{\ell_1 + x\cos\theta_L} \tag{17}$$

for area 1 and

$$h_2(x, \theta_L) \simeq \frac{x(\ell_2 - \ell_1)\sin\theta_L}{\ell_1 - x\cos\theta_L} \tag{18}$$

for area 2. The $L_{1,2}(x, \theta_L)$ is approximated with,

$$L_1(x, \theta_L) \simeq \sqrt{\ell^2 + x^2 + 2x\ell_2\cos\theta} \tag{19}$$

and

$$L_2(x, \theta_L) \simeq \sqrt{\ell^2 + x^2 - 2x\ell_2\cos\theta_L}. \tag{20}$$

The target density distribution $n(x)$ is given by Eqn. (8) for the C_{60} target, and $n(x) = 1$ for the CH_4 target. The results of calculations using these equations are shown in Figure 17 for the CH_4 target and Figure 18 for C_{60}. These values are also shown Table 1 for CH_4 and Table 2 for C_{60}.

2.8 Extraction of the Differential Cross Section

The differential cross section in the laboratory frame was deduced from measurements with Eqn. (10). However we couldn't measure the absolute value of the target density N_0. Therefore, in this study only the relative values of the differential cross sections were determined from the experiments. Measuring the total number of incident ions, I, for each different projectile experiments under the same target condition, allowed us to determine the relative differential cross section between different projectile experiments through Eqn. 10. To calculate the absolute values of the differential cross sections, the experimental data had to be normalized with a theoretical factor.

To deduce the value of the differential cross section in the center of mass (CM) frame from measurements in the laboratory frame, the solid angle

must be transformed as follows,

$$\frac{d\Omega_{CM}}{d\Omega_L} = \frac{\left[\sqrt{\xi^2 - \sin^2\theta_L} + \cos\theta_L\right]^2}{\xi\sqrt{\xi^2 - \sin^2\theta_L}} \tag{21}$$

for the projectile and

$$\frac{d\Omega_{CM}}{d\Omega_L} = 4\cos\theta_L \tag{22}$$

for the target particle, where ξ is the mass ratio m_2/m_1.