

1 Introduction

The discovery of atomic clusters has greatly extended the field of application of many body systems, influencing physics, chemistry, the applied sciences, and other fields of science. In particular, there has been a general extension of many body studies in the research of nuclear fission and fusion reactions to many body studies of cluster fission (dissociation) and fusion reactions. This paper is an attempt to expand this extension by drawing parallels between many body studies of internuclear potentials in nuclear physics and interatomic potentials in cluster physics through collision experiments, that is, from elastic collisions between nucleons in a nuclei to strong collisions between atoms in a cluster at a few keV [1].

Buckminsterfullerene (C_{60}), discovered in 1985 by Kroto *et al.* [2], has been used easily in many research fields since its successful refinement and mass production by Krätschmer *et al.* [3] in 1990. Scientists have made much progress in characterizing the new chemical properties and the electron structure of fullerene. Fullerene has received much attention in many body research because it has a small number of atoms and high symmetry. Many collision experiments on C_{60} with molecules, electrons and photons have been performed, and fragmentation and dissociation of excited C_{60} has been well studied as will be shown below.

Many of the collision experiments with molecules are performed by projecting a C_{60} beam onto a gas molecule. R. J. Doyle *et al.* performed the collision of C_{60} cations accelerated to a few keV with an O_2 gas target [4], and E. E. B. Campbell *et al.* performed a similar experiment with noble gas targets [5]-[7]. Moreover, Campbell *et al.* have extended their collision techniques to include $C_{60}^{q+} + C_{60}$ collision experiments in recent years [8]-[12]. In these cases, it is easy achieve a low collision energy (~ 100 eV) in the center of mass system. P. Hvelplund *et al.* injected a higher energy, 50-200 keV, C_{60} cation beam into a noble gas [13]-[16]. In all of the above experiments, only collision products scattered in the beam direction were able to be measured because, in general, when a heavier C_{60} cluster is injected into lighter gas

molecules, all the collision products scatter into a limited forward angular range.

In other experiments using C_{60} beams, surface-induced dissociations (SID) of C_{60} were performed. R. D. Beck *et al.* [17]-[20] observed the SID changing from fragmentation by successive C_2 loss into fragmentation by fission of the C_{60} into two or more greater parts as the energy of impact of C_{60} on the surface increased. And, I. V. Hertel *et al.* studied the rebound of the C_{60} from the surface; the recoil velocity distributions as a function of the recoil angle were measured [21]-[28].

In contrast, there have been other collision experiments with C_{60} as the target. Ionization reactions with C_{60} , plasmon excitations, capture reactions of beam atoms in the cage of C_{60} , and so on, have all been studied. In experiments using MeV-order ion beams, T. LeBrun *et al.* [29, 30], Y. Nakai *et al.* [31] and J. Opitz *et al.* [32] reported on ionization and fragmentation of C_{60} , while S. L. Anderson *et al.* [33]-[39] performed collision experiments using ion beams with energies less than 100 eV. They paid particular attention to endohedral complex species ($X@C_{60}$; X indicates any kind of atom) produced by atomic capture reactions between low energy ions and the whole of a C_{60} molecule.

These experimental researches have motivated theoretical physicists to understand the experimental results. The collision of a C_{60} and an atom promote the C_{60} into an excited state. The statistical distribution of the vibration energy of the atoms in C_{60} , that is, its internal energy, is related to the sequential C_2 evaporation of C_{60} , and the statistical Rice-Ramsperger-Kassel-Marcus (RRKM) model successfully describes the experimental results [20],[40]-[42]. Several computer simulation studies have been performed and compared with experimental results. For example, a Molecular Dynamics (MD) simulation is being developed by R. Ehlich *et al.* [43] of the collision dynamics of C_{60} with noble gas atoms, and simulations of the fusion of two C_{60} molecules and fragmentation in $C_{60} + C_{60}$ collisions have been reported by Y. Xia *et al.* [44] and R. Schmidt *et al.* [45]. Simulations of the rebound-

ing of C_{60} on a surface have been reported by R. C. Mowrey *et al.* [46] and by Z. Y. Man *et al.* [47].

Further experiments, involving the collisions of multi-charged ion beams with C_{60} at keV energies, were performed by C. W. Walter *et al.* [48], B. Walch *et al.* [49], P. Scheier *et al.* [50], N. Selberg *et al.* [51], J. P. Briand *et al.* [52] and C. W. Walter *et al.* [53], among others. In the experiments, a multi-charge exchange reaction, that is, a transfer of many electrons at a time from C_{60} to an ion, occurs. The corresponding cross sections were represented by the classical over-barrier model [54, 55]. Jin *et al.* [56] used a more highly charged projectile, Bi^{44+} , to produce C_{60}^{9+} , the most highly charged free C_{60}^{i+} ion reported up to 1996. For charged states $i < 9$, Jin *et al.* [56] found C_{60}^{i+} ion to have lifetimes of at least 5 μ sec. This remarkable stability of C_{60}^{i+} indicates considerable charge shielding in these ions and supports the delocalized charge model.

Until now all of the experimental measurements have been of products of large impact parameter collisions, that is at $\sim 0^\circ$ scattering. The reason for this is that in MeV energy collisions, the cross sections of large scattering collisions (elastic scattering) between the ion and a carbon are much smaller than that of the electron excitation reaction. In contrast, in eV energy collisions, the projectile reacts with the whole C_{60} molecule, and studies of the large-angle scattering of particles are of no use. However, in the middle energy region, at a few keV, atom-atom elastic scattering at large angles occurs in C_{60} , which is well known in radiation physics. This is an exciting energy region and reaction mechanism that allows us to study the fundamental processes of atom-atom collisions in a many body system.

In order to study these fundamental processes, we came up with a new idea for an experiment – injecting a noble gas ion into a C_{60} molecule at a few keV and measuring the angular distribution of the scattered atoms. When a keV energy atom collides with a carbon atom in C_{60} with a small impact parameter, the projectile and an individual

carbon are strongly scattered out of the C_{60} at large angles (elastic scattering) because the binding energy of the carbon atoms (covalent bond energy $\sim 4\text{eV}$) is smaller by several orders of magnitude than the kinetic energy of the collision. Such a direct scattering reaction between a keV energy atom and an individual carbon in C_{60} is dominant as indicated from cross section calculations using the interatomic potential (see next paragraph). We wanted to investigate the large-angle scattered atoms rather than the $\sim 0^\circ$ scattered atoms. Therefore, we performed experiments in which noble gas atoms which have been accelerated to a few keV are injected into a single isolated C_{60} molecule.

The atomic collision theory for a few keV is important in the present study. The interaction between two atoms is a many body problem involving the Coulomb interactions among two atomic nuclei and the electrons bound to each nuclei. Many approximate models of the interatomic potentials for two atoms have been proposed [57]. The simplest and best approximation is a model emphasizing the screening of the Coulomb force by the electrons. L. H. Thomas [58] and E. Fermi [59] developed a simplified model, the Thomas-Fermi model, for a many-electron atom based on the Fermi-Dirac statistics of a free electron gas. The Thomas-Fermi potential is represented as a simple Coulomb field reduced by a factor called the Thomas-Fermi screening function. There are several approximate equations of the Thomas-Fermi screening function. G. Molière represented the Thomas-Fermi function as the sum of three exponential components [60]. For two atoms, the Thomas-Fermi model had to be modified. The screening radius was made dependent on the atomic number of the two atoms, and the Coulomb potential extended to that between two atoms rather than between an electron and its nucleus, by O. B. Firsov [61] for interatomic distances less than 1\AA . In general, this screened interatomic potential is simply called the Molière potential. Since the 1950s many experiments on atomic collisions have been performed at different energies with a variety of target and incident atom combinations [62]. From these experimental results a correction to the screening radius involving a

function of the atomic numbers of the collision partners was derived by D. J. O'Connor and R. J. MacDonald [63]. With this correction, the Molière potential is more highly accurate for greater interatomic distances, as verified in two-atom collisions, and it has been used in computer simulations of cascades of atomic collisions in the keV range, for example, in radiation damage studies [64].

According to calculations using the Molière potential, the partial cross section of large angle scattering of atoms (elastic collision) becomes a large part of the total reaction cross section which is almost equal to the geometrical cross section of C_{60} ; therefore, it is the dominant reaction in atom- C_{60} collisions at a few keV. Generally speaking, the dynamics governing the processes in the atom-cluster collisions are expected to have features similar to those in simpler atomic collisions which have been more extensively studied so far and are well understood, as mentioned above. However, more complicated situations may appear in atom-cluster collisions. Indeed, there should be many kinds of nontrivial effects because of the larger number of degrees of freedom. Unfortunately, there are only a few experimental or theoretical studies of elastic collision of atoms in cluster, and no studies at all of the screening effects in many-atom systems.

In the present study, we have carried out an experimental study of the collision dynamics of noble gas atoms He (4.0 keV), Ne (3.2 keV), Ar (4.2 keV) and Xe (9.8 keV) on an isolated C_{60} molecule (Figure 1). Such combinations of fullerene and noble gas have several advantages over other complicated combinations. It is preferable to keep the collision dynamics as simple as possible. The C_{60} is a rather simple object, which consists of 60 carbon atoms, and has a well established geometrical structure with a concatenated icosahedral symmetry, while a noble gas molecule is a monoatomic. Therefore, we have not had to struggle with both structural variations of a cluster and extra interactions among the atoms in a molecule. In addition, we have conducted a supplementary experiment for comparative study - the collision of noble gas atoms He, Ne, and Ar on CH_4 molecules instead of C_{60}

at the same incident energies. To compare this with the C_{60} case, it is possible not only to obtain some features of the atom- C_{60} collisions, but also to check the accuracy of the experiments. In order to obtain data over a wider energy range for a single noble gas, the collision of Ar gas atoms on C_{60} and CH_4 at different incident energies from 4.2 keV to 10.0 keV were performed.

To perform a experiment on atomic scattering for large angles with C_{60} targets successfully, the C_{60} molecule must be isolated in space. To achieve this we developed a molecular-beam target. Because C_{60} is very stable and easily vaporizes, it was easy to produce a high density gas target for scattering experiments.

The noble gas atoms at a few keV are able to approach a carbon atom to a range of less than 0.5\AA , as estimated from calculations with the Molière potential. The average distance between nearest carbon atoms in C_{60} is 1.4\AA , so the distance of closest approach between the noble gas atom and a carbon is one order of magnitude smaller than the lattice spacing of C_{60} . Therefore, there must be some effects due to neighboring carbon atoms when a quasi-elastic collision occurs. As I mentioned before, in this energy region the interatomic potential is not only determined by the Coulomb potential caused by the nuclei of the atoms, but also by the screening of the Coulomb force by the electrons. More precisely, screening effects from electrons in neighboring carbon atoms should influence the interatomic potentials between the noble gas atom and the dominant carbon atom target of the C_{60} molecule. In order to see such a many body effect directly, the angular distributions of scattered atoms for large angles in quasi-elastic collisions with a carbon in C_{60} were measured (Figure 2). We were the first to deduce the scattering differential cross sections from the angular distribution as has been reported in ref. [65].

The goal of the present study is to extract the details of the interaction potential between the noble gas atom and the C_{60} experimentally by measuring the differential cross sections for quasi-elastic scattering. In contrast with the studies of collisions between two atoms that are well described by the Molière potential based on the Thomas-Fermi model, the primary interest in

our study is to see whether the interatomic potential between the incident atom and the carbon atoms are modified or not by the incident atom being embedded within the C_{60} . There were several motivations to search for such modifications. We had expected that there would be a difference between ion-cluster collisions and ion-atom collisions because of the three following reasons.

First, the electron distribution of the C_{60} molecule is different from that of an isolated carbon atom. In C_{60} there are not only the core electrons (1s) binding each carbon and three σ -bond electrons in each carbon, but also 60 π electrons spread all over the C_{60} . The diversity of electron states and the singular non-localization of the π electrons are well known. The 60 π electrons are able to move over the C_{60} molecule freely just as in a metal. When any atom collides with a carbon in the C_{60} , there is a possibility that these non-local free π electrons play a role in some special screening effects for the atomic interactions, screening effects that are different from those of local electrons in an atom.

Second, the boundary conditions for a carbon atom will be substantially altered in a cage of C_{60} from those for an isolated carbon because C_{60} is a kind of small crystal of carbons. For example, the electric field should be zero at the mid-point between adjacent carbon atoms because of the structural symmetry of C_{60} , while it becomes zero at infinity in the case of an isolated carbon atom. Since the noble gas atom moves rather slowly compared with its electrons, the electric field around it will also be largely affected by the same boundary conditions when it comes near the C_{60} cage. Therefore, the electron distributions of the noble gas atom and the carbons are modified by the same boundary conditions, and this must affect the electronic screening effects on the interatomic potentials.

Third, the new additional electronic screening effects should be attributable to the electron distribution about the noble gas atom and some carbons near the collision point. Firsov studied the electron gas density distribution around two nuclei and extracted the interatomic potential between

any two atoms as a function of the distance between two atoms, r ; that is to say, he formulated an isotropic potential depending on only r . In atom- C_{60} collisions, to extract the screening function, we must consider the electron density structure in the many body system, and we cannot make use of the special symmetry in the two-atom collision to reduce the problem to a one dimensional problem in r . The electron density distribution for a given set of initial conditions could perhaps be calculated using approximations such as the Local Density Approximation or the *ab initio* methods; however, it would be difficult to perform the calculations for all the various initial conditions.

In conclusion, we expected to find a different electronic screening effect in the collisions of the noble gas atoms with a carbon in the C_{60} – on effect different from that in collisions with an isolated carbon atom that is described well by the Molière potential – due to the difference of the electron density distributions around the carbon atom in the two types of collisions. That is to say, the interatomic potentials for the atomic collisions in the C_{60} must be different, but this was not known before our experiments. Therefore, we measured the differential cross sections of elastic atom-atom collisions in C_{60} to extract the interatomic potentials experimentally.

The report is organized as follows. Section 2 explains in detail our experimental setup, some techniques for measuring the angular distributions of scattered atoms with Time of Flight (TOF) mass identification, and describes the gas-phase molecular targets and our analysis of the differential cross sections from the TOF spectra. Section 3 gives a brief introduction into some properties of C_{60} and interatomic potentials including the electronic screening effect, and describes the experimental and theoretical results. Section 3.3 and 3.4 discuss the analysis of the scattering differential cross sections and subsequent deduction from them of the interatomic potential modified by an additional screening function. Some concluding remarks will be given in the last section, Section 4.