First-principles study on high speed collisions between neon atom and carbon cluster

T. Morisato[1], K. Ohno[2], Y. Kawazoe[1], T. Miura[3,*] and S. M. Lee[3]

[1]Institute for Materials Research (IMR), Tohoku University, Katahira, Aoba-ku, Sendai 980-8577. Japan
Fax: 81-022-215-2052, e-mail: morisato@imr.edu, kawazoe@imr.edu
[2]Department of Physics, Faculty of Engineering, Yokohama National University, Tokiwadai, Hodogaya-ku,
Yokohama 240-8501, Japan
Fax: 81-045-339-4254, e-mail: ohno@ynu.ac.jp
[3]Institute of Physics, University of Tsukuba, Tennodai, Tsukuba, Ibaraki 305-8577, Japan
e-mail: lee@tac.tsukuba.ac.jp

Recently Miura *et al.* carried out an experimental study of the collision dynamics of the monatomic noble-gas ions on CH_4 and C_{60} molecule with ~keV incident energies. They reported that the differential cross section for the collision of noble-gas ions with a carbon atom in CH_4 molecule is different from that in the case of C_{60}. We thought that this may be caused by a strong screening effect working in the case of targets having C-C network structure like C_{60}. In order to ascertain whether this is true, we carried out the total energy calculation of system as a function of interatomic distance by first-principles calculation. From results of calculations we conclude that C-C network structure makes screening effect stronger to some extent.
Key words: all-electron mixed-basis approach, interatomic potential, C_{60}, scattering differential cross section, atomic cluster

1. INTRODUCTION

In the study of atomic cluster the collision of atomic clusters with atoms is one of the main subjects. Especially as for atom-C_{60} collisions, many experimental and theoretical studies carried out [1-8]. A C_{60} molecule [9] is one of atomic clusters and consists of 60 carbon atoms with a truncated-icosahedral symmetry structure.

Recently as one of such studies Miura *et al.* carried out an experimental study of the collision dynamics of the monatomic noble-gas ions on an isolated C_{60} molecule with ~keV incident energies [7]. They have also conducted a supplementary experiment for comparative study, i.e., the collision of the noble-gas ions on CH_4 molecules. For these experiments they reported as follows:

The experimental result of the scattering differential cross section for the collision of noble-gas ions with a carbon atom in a CH_4 molecule agrees with the curve made from the Moliére potential, therefore the influence of hydrogen on the collision is negligibly small. It is well known that the Moliére potential successfully describes various types of binary ion- and atom-atom collisions phenomenologically [10,11]. But with C_{60} the Moliére potential can't describe the collision interaction between the noble-gas ion and a carbon atom in C_{60} very well.

As for CH_4, one can expect this result easily because C-H bonding energy is negligibly small compared with the incident energy. But as for C_{60}, one may think that this result is strange because C-C bonding energy is also negligibly smaller than kinetic energy of noble-gas ion. So, as one of possibilities, we thought that this may be caused by a strong screening effect working in the case of targets having C-C network structure like C_{60}.

In order to know whether the C-C network structure makes screening effect stronger, we carried out the total energy calculation of system as a function of interatomic distance between noble-gas atom and target by first-principles calculation. For this calculation, we used a Ne atom as an incident noble-gas atom, and a CH_4 molecule and a graphite sheet as target. The reason why we used graphite sheet instead of C_{60} is as follows: C_{60} is too big to calculate and graphite has periodic structure, so we can use smaller unit-cell than C_{60}. Moreover graphite has C-C network as well as C_{60}.

Generally, if an atomic beam with a ~keV kinetic energy per atom is irradiated to molecules, two nuclei come close to each other (typically 0.1-0.5 Å) in atomic collisions, and not only valence but also core electron orbitals deform largely from atomic orbitals. To study such collisions by *ab initio* techniques, an adequate description of deformed core orbitals is inevitable and *ab initio* methods using muffin-tin potentials or pseudopotentials are powerless. Because of these reason, we used the all-electron mixed-basis approach [12,13]. As is usual, it is based on a density-functional framework [14,15] within the local density approximation

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[16,17] to the exchange and correlation interactions. This approach uses plane waves (PWs) and atomic orbitals (AOs) as basis functions and can present a reliable description of the deformed core orbitals. This approach has following advantages:

(1)　It can very efficiently describe both extended and localized electronic states because of the usage of plane waves in addition to atomic orbitals.

(2)　An adequate description of core states is possible even when another atom comes close into core regions, if an enough number of PWs is adopted in basis functions

(3)　The matrix elements between plane waves, which are the largest part in the Hamiltonian matrix, are not necessarily stored in computer memory as well as in the case of the standard plane-wave approach.

(4)　Since atomic orbitals are confined within non-overlapping atomic spheres, there is no matrix element between atomic orbitals of two adjacent atoms. Therefore, the calculation of matrix elements does not cause any unnecessary numerical errors, and moreover the problem of overcompleteness is reduced.

It is one of the purpose of this study that we ascertain whether the all-electron mixed basis approach is usable as a method of simulation of atom-cluster collision.

2. CALCULATION

As mentioned above, we calculated total energy of two systems, i.e., Ne -CH$_4$ and Ne-graphite as functions of interatomic distance. In this chapter, we describe how calculate these cases in detail.

2.1 Total energy calculation of Ne-CH$_4$ system

As shown in figure 1, we calculate the total energy of this system by the all-electron mixed basis approach, changing the interatomic distance from 0.3Å to 2.116Å. The cut-off energy of PWs is 121Ry in this calculation. We adopted super-cell approximation for this calculation, and cell size is 4.233Å×4.233Å×4.233Å. Next, we got a function of interatomic potential from the results of calculation. The total energy function is

$$E(r) = E_0 + P(r),$$

where r is Ne-C interatomic distance and P(r) is the interatomic potential function given by fitting to the calculated point. We took E$_0$ as reference of interatomic potential. Next, we calculate the differential cross section from this interatomic potential function by assuming that this system is 2 -body system, i.e., Ne -C.
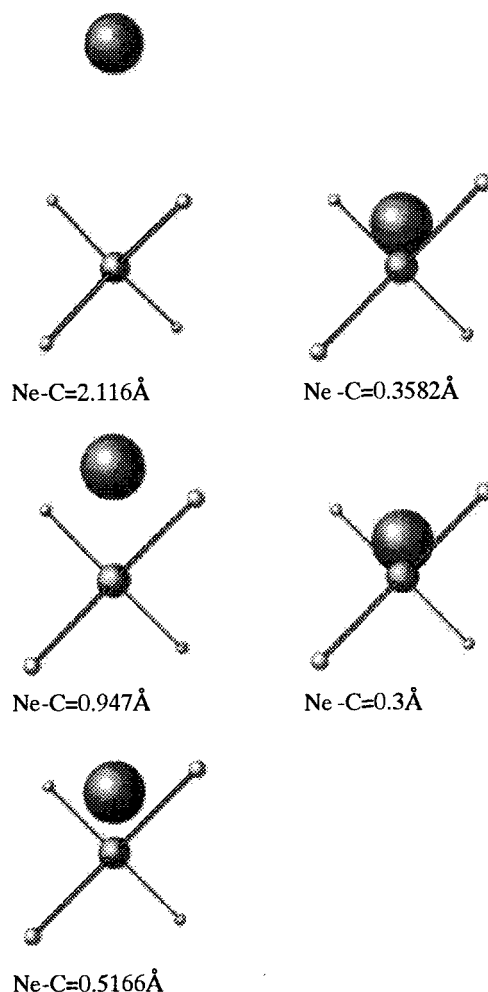

Ne-C=2.116Å   Ne -C=0.3582Å

Ne-C=0.947Å   Ne -C=0.3Å

Ne-C=0.5166Å

Fig. 1 The each locations of a Ne atom and a CH$_4$ molecule in the calculation of total energy that is a function of Ne-C interatomic distance.

2.2 Total energy calculation of Ne-graphite system

Figure 2 shows the way that a Ne atom approach to a C atom when we calculated the total energy of Ne-graphite system as a function of interatomic distance. As shown in Fig.2, we moved a Ne atom to a C atom in graphite along the line perpendicular to the graphite sheet. We use super-cell approximation also in this system, and cell size is 4.912Å×4.912Å×4.912Å and 4.819Å×4.819Å×4.819Å. The first cell size is for normal graphite that C-C bond length is 1.42Å, and the second cell size is the size that C-C bond length is 1.39Å, which is one of the C-C bond length of C$_{60}$. In the first cell the cut-off energy of PWs is 103 Ry and in the second cell the cut-off energy of PWs is 107 Ry. Also in this case we calculated the differential cross section from the interatomic potential function given by fitting to the results of the total energy calculations. This differential cross section is calculated under the same assumption as the case of Ne-CH$_4$, i.e., we thought of Ne-graphite system as 2-body system of Ne-C (in graphite).
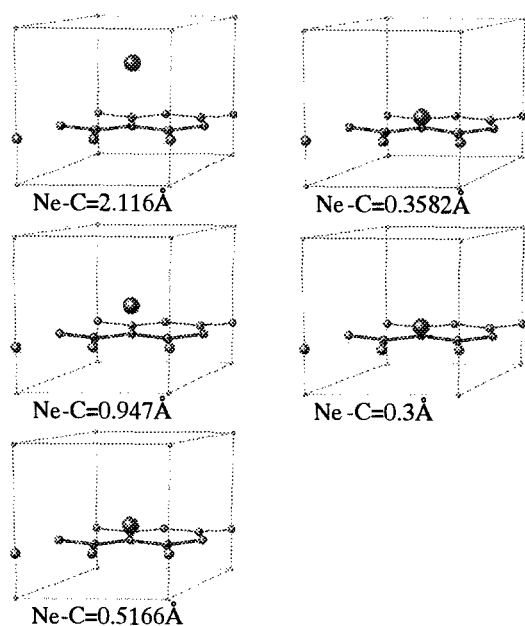
Fig. 2 The each locations of a Ne atom and a graphite sheet in the calculation of total energy that is a function of Ne-C interatomic distance.

3. RESULTS & DISCUSSION

3.1 The calculated interatomic potential

Figure 3 shows the calculated interatomic potential of Ne-CH$_4$ system and 2 types of Ne-graphite system that have different C-C bond length each other, i.e., Ne-graphite1 and Ne-graphite2. The vertical axis shows interatomic potential and the horizontal axis shows Ne-C interatomic distance. In this figure we drew the Rutherford potential of Ne-C for reference. Of course, all these three calculated potentials are much more screened than Rutherford potential. Ne-graphite1 and Ne-graphite2 are more screened than Ne-CH$_4$. Therefore one can say that the C-C network structure makes screening effect of Ne-C interatomic potential stronger. And from comparison between Ne-graphite1 and 2, we can conclude that the system having short C-C bond length have stronger screening effect than that having long one.
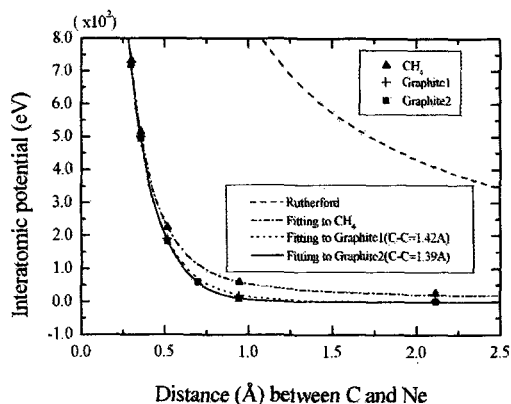

Fig. 3 The calculated interatomic potentials of Ne-CH$_4$, Ne-graphite1 (C-C=1.42Å) and Ne-graphite2 (C-C=1.39Å).

3.2 The calculated differential cross section

Figure 4 shows differential cross section curves calculated from the interatomic potential function in 3.1 under Ne-C 2-body approximation and experimental results by Miura *et al.*. The vertical axis shows the differential cross section and the horizontal axis shows scattering angle in center-of-mass coordinates.
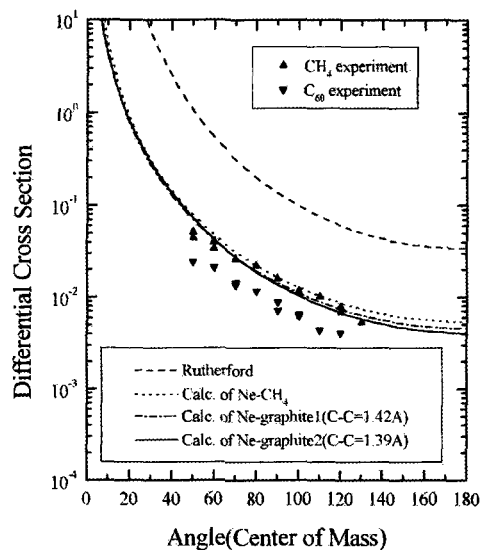

Fig. 4 The measured differential cross sections of Ne-CH$_4$ and Ne-C$_{60}$, and the calculated differential cross sections of Ne-CH$_4$, Ne-graphite1 (C-C=1.42Å) and Ne-graphite2 (C-C=1.39Å).

For Ne-CH$_4$ system the calculated differential cross section is in good agreement with experimental results. So we can say that the all-electron mixed basis approach is usable for such as a calculation of an atom-molecule collision. The calculated differential cross section of Ne-graphite1 system is below calculated Ne-CH$_4$ curve as well as the experimental results, so in this point one can say that the tendency of Ne-graphite1 agree with the experimental results, but one can also say that these calculated results still have some difference from the experimental results. As these experimental results of Ne-C$_{60}$ are below those of Ne-CH$_4$ in large scattering angle, one can say that the experimental results show that in Ne-C$_{60}$ system the screening effect is larger than that in Ne-CH$_4$ system, if such things are caused only by screening effect of Ne-C interatomic potential. In this point, the screening of the calculated interatomic potential of Ne-graphite1 system is smaller than that of the experimental potential of Ne-C$_{60}$ system. So, next we are going to see the calculated differential cross section of Ne-graphite2, which we conclude in 3.1 that Ne-graphite2 have larger screening effect than Ne-graphite1. The differential cross section of Ne-graphite2 is closer to the experimental results than Ne-graphite1. But even the result of Ne-graphite2 still deviates from the experimental results to some extent. We think that this is caused by the difference of structure between graphite and C$_{60}$, the assumption of Ne-C

2-body problem in the calculation of differential cross section and having no results calculated by moving a Ne atom along line different from the line perpendicular to the graphite sheet.

4. CONCLUSION

From all above, we can conclude for this study as follows.

(1) From the result of Ne-CH$_4$ system, the all-electron mixed-basis approach is usable as a method of simulation of atom-cluster collision.

(2) The C-C network structure makes screening effect of Ne-C interatomic potential stronger to some extent.

(3) The screening effect of Ne-C interatomic potential is large when the C-C bond length is short.

5.ACKNOWLEDGEMENT

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