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# The computer simulation of the scattering of fullerenes from a graphite surface: Energy partitioning and vibrational spectra

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## Abstract

Molecular dynamics computer simulation is used to investigate the scattering behaviour of  $C_{60}$ ,  $C_{76}$  and  $C_{84}$  fullerene molecules from a graphite surface. In particular the energy partitioning into translational, vibrational and rotational motion is determined and the vibrational frequency spectra are calculated and the principal structure identified. At the glancing angle of incidence, studied here, it was found that the fraction of retained kinetic energy after impact increases to about 90% as the initial impact energy increases. The fraction of energy retained as translational motion remains constant as a function of impact energy. Only a small fraction ( $\sim 1\%$ ) of the energy goes into rotational motion. The rotational frequency remains constant regardless of impact energy ( $\sim 5 \times 10^{11}$  Hz). A number of vibrational modes can be identified easily from the vibrational spectra. In particular the C–C and C=C bonds and a “breathing” vibrational mode are seen. © 2001 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

It has been shown recently how the scattering of  $C_{60}$  from graphite gives rise to anomalous non-specular scattering at glancing angles [1]. This work concentrated on the scattering of  $C_{60}$  at  $15^\circ$  to the surface with an interest mainly in the angular distributions of the scattered molecules. It was shown there how the angular distributions become narrower and closer to specular as the energy of the scattering event is reduced. The work presented here looks at even shallower scattering events and we look at the effects of changing the initial shape of the scat-

tering particle by using a  $C_{76}$  and  $C_{84}$  molecules in the simulations. In particular the  $C_{76}$  molecule is non-spherical. The simulation data has been analysed to show how the energy of the scattering event is partitioned into translational movement of the centre of mass of the molecule, rotational movement, and vibrational movement of the constituent atoms of the scattered molecule. We have also made a preliminary look at the vibrational spectra of the scattered molecules.

## 2. Molecular dynamics simulations

The simulation model used for this investigation has been described in detail elsewhere [2,3] and only a short description of the important

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details is given here. The simulation employs many body potentials in the Brenner formulism [4] which are known to model the carbon system in these energy ranges quite well. The simulation system uses 4 atomic layers of a graphite lattice of dimensions  $300 \times 40 \text{ \AA}$ , containing in total about 20,000 atoms. The target was longest in the direction of motion of the ion. The temperature of the lattice was 0 K. Free boundaries were used in conjunction with a large lattice to avoid reflection of energy back from the sides of the simulation cell before the molecule had left the surface. The scattering events were followed for 2 ps. Each energy and scattering molecule was averaged over a set of 29 independent trajectories, little differences were found between different trajectories in each set. Due to the large diameter of the fullerene molecules little difference was expected in changing the impact point. However, further studies need to be made to investigate the possibilities of out of plane scattering for the non-spherical  $C_{76}$  molecule as the trajectory set was too small to gain reliable data on this.

### 3. Results and discussion

Simulations were performed for an incidence angle of  $80^\circ$  to the surface normal for a range of energies for the three fullerene molecules  $C_{60}$ ,  $C_{76}$  and  $C_{84}$ . Fig. 1 shows the mean total scattering angle of the scattered fullerenes as a function of

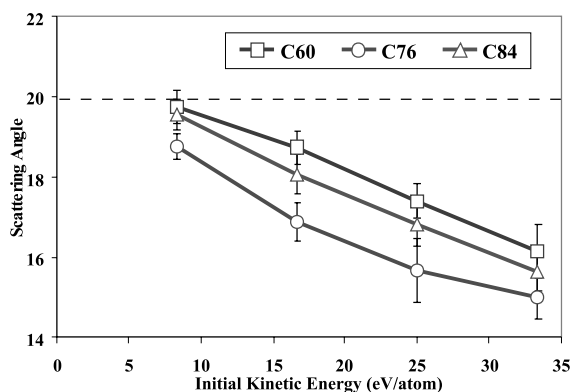


Fig. 1. Total scattering angle as a function of energy.

the impact energy. The total scattering angle is such that specular reflection from the surface is achieved for this angle of incidence at  $20^\circ$ , indicated by the dashed line in Fig. 1. Individually the molecules exhibit very similar behaviour to the previous analysis [1] at  $75^\circ$ . As the energy is reduced the scattering angle gets closer to specular and the distributions become narrower. Compared together, however, shows that the non-spherical  $C_{76}$  molecule seems to undergo a greater deviation from the specular conditions than either  $C_{60}$  or  $C_{84}$ . This would suggest that the shape of the molecule is a factor in determining the behaviour and not just the response of the graphite surface.

In Fig. 2 we show the fraction of kinetic energy retained in the molecule after the impact for the three molecular species as a function of impact energy. There is very little difference between the molecules and all show that the fraction of retained energy increases as a function of energy but seems to saturate at about 90%. At low energies a higher fraction of the initial energy is given up to deforming both the target and molecule on impact.

Fig. 3 shows the fraction of the retained kinetic energy that goes into translational motion. This has been calculated by analysing the velocity of the centre of mass of the molecule and converting this back into kinetic energy. For all three molecules this remains a constant fraction as a function of energy. The differences between molecules is due to the extra number of atoms in the scattering

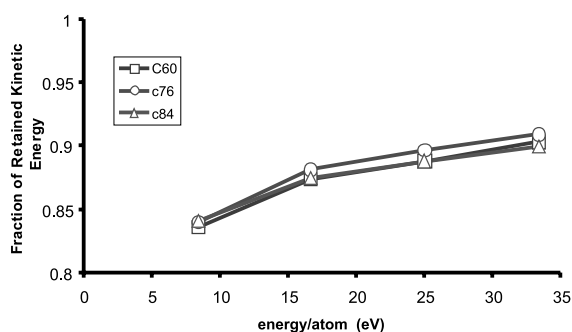


Fig. 2. Retained kinetic energy after impact as a function of impact energy.

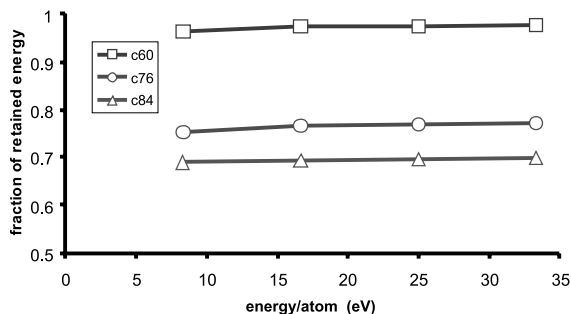


Fig. 3. Fraction of retained energy going into translational energy as a function of impact energy.

molecules, which take up extra energy in vibrational motion. There are 90 bonds in the C<sub>60</sub> molecule, 114 bonds in the C<sub>76</sub> and 126 bonds in the C<sub>84</sub> molecule. The larger molecules take up a higher fraction of kinetic energy into vibrational motion. The differences between molecules shown in Fig. 3 can be accounted for just be the increase in the number of extra vibrating pairs in the bigger molecules.

At this angle of incidence the scattered molecules can be seen to rotate nearly a full half turn in the 2 ps of the simulation. This makes it quite simple to calculate the rotational frequency of the scattered molecules by looking at the rotation of a set of points on the surface of the molecule across the centre of mass. In doing this we find that the rotational frequency of the molecules remains constant as a function of im-

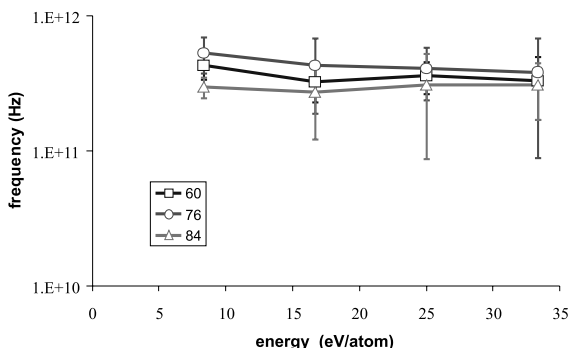


Fig. 4. Rotational frequency after collision as a function of impact energy.

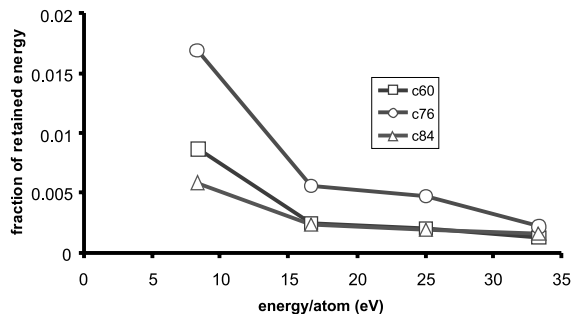


Fig. 5. Fraction of retained energy going into rotational motion as a function of impact energy.

pact energy as shown in Fig. 4. The C<sub>76</sub> molecule consistently shows a higher rotational frequency than C<sub>60</sub> which is the inverse of the case for the C<sub>84</sub>. This would seem likely to be an effect of the shape of the C<sub>76</sub> again. Interestingly though the rotational velocity (frequency) does not appear to change despite increasing the energy by a factor of 4. By converting these rotational velocities into energy we find that the fraction of energy going into rotation is less than 1% and decreases as the impact energy increases as shown in Fig. 5. By subtracting the rotational energy and translational energies away from the total kinetic energy we are left with the energy going into vibrational motion. This also remains a fairly constant fraction as the energy of impact increases and is shown in Fig. 6. As the energy of rotation is so small it looks largely like the inverse of Fig. 3.

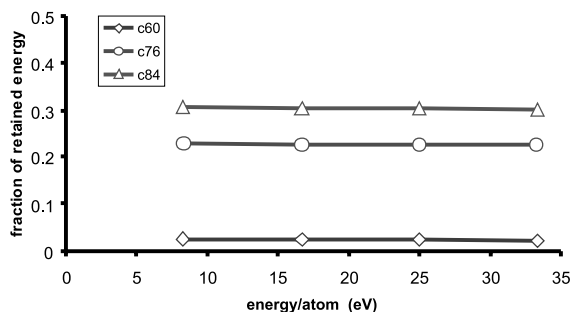


Fig. 6. Fraction of retained energy going into vibrational motion as a function of impact energy.

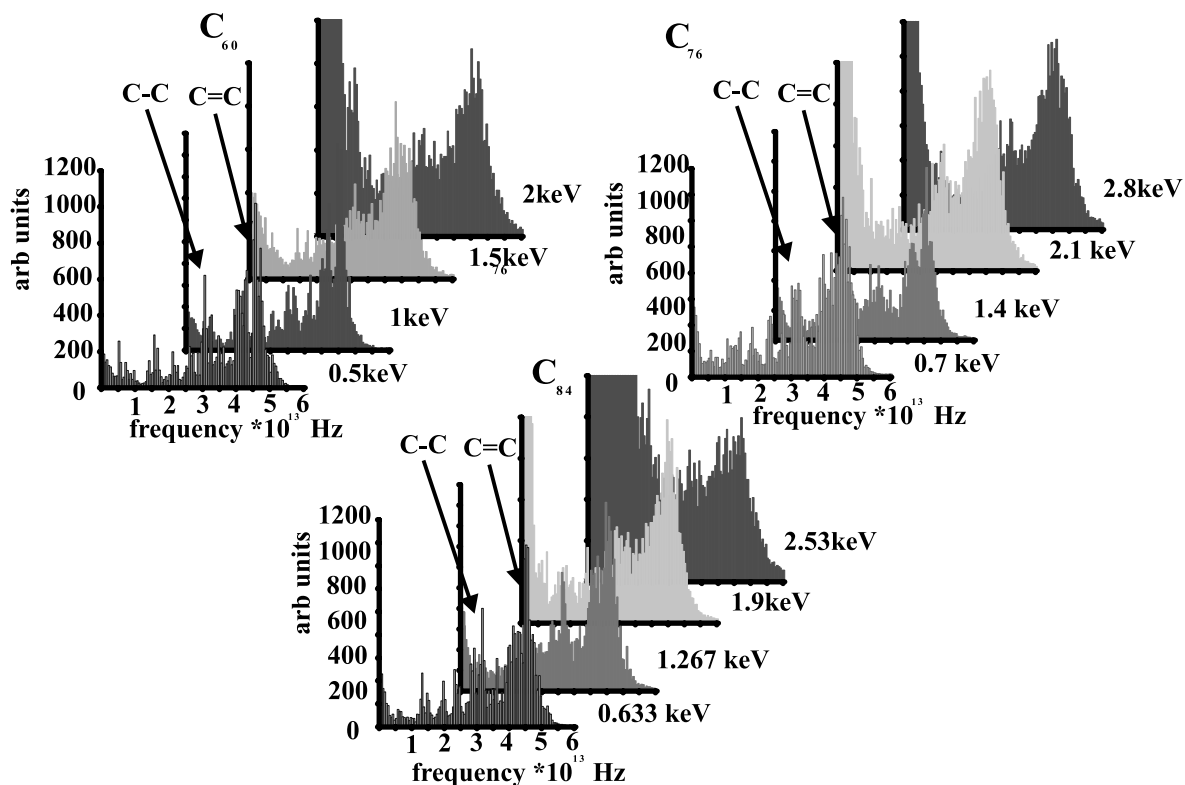


Fig. 7. Vibrational spectra after impact on a graphite surface for (a) C<sub>60</sub>, (b) C<sub>76</sub> and (c) C<sub>84</sub>.

Fourier analysis of all of the nearest neighbour pairs in the molecule as a function of time gives a vibrational spectrum for the scattered molecule as shown in Fig. 7. The vibrational spectra do not change very much as the scattering energy increases other than that they gain a low frequency background. This seems to be due to the compression and distortion of the molecule, indicating that we may need to extend the simulation to longer times to allow the molecule to stabilise for more time before recording the vibrational spectra. But the principal vibrational modes are evident in all of the spectra and look very similar regardless of the molecular species. Both the C–C and C=C vibrational modes are seen at  $3.4 \times 10^{13}$  and  $4.5 \times 10^{13}$  Hz, respectively. A peak at about  $1.5 \times 10^{13}$  Hz is seen which when analysed appears as a compression and expansion of the whole molecule – a “breathing mode”.

#### 4. Conclusions

It was found that some of the details of the scattering event seem to have a small dependence upon the shape of the molecule. In particular the behaviour of the scattering angle seems to be, not surprisingly, shape sensitive. The rotational frequency, and hence the amount of energy going into rotation, is, once again not surprisingly, shape sensitive. However the amount of energy partitioned into rotation is small and has little overall effect on the amount of energy put into the vibrational system. In this case it seems that the fraction of energy entering the vibrational system depends on the number of atoms in the system rather than on the shape. The results shown here seem to suggest that the rotational frequency of the scattered molecule is independent of the energy of the scattering event. A number of vibrational modes can be identified

easily from the vibrational spectra. In particular the C–C and C=C bonds and a “breathing” vibrational mode.

## References

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