Giant plasmon excitation in single and double ionization of C\textsubscript{60} by fast highly charged Si and O ions

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Abstract. We have investigated single and double ionization of C\textsubscript{60} molecule in collisions with 2.33 MeV/u Si\textsuperscript{q+} (q=6-14) and 3.125 MeV/u O\textsuperscript{q+} (q=5-8) projectiles. The projectile charge state dependence of the single and double ionization yields of C\textsubscript{60} are then compared to those for an ion-atom collision system using Ne gas as a target. A large difference between the gas and the cluster target behaviour was partially explained in terms of a model based on collective excitation namely the giant dipole plasmon resonance (GDPR). The qualitative agreement between the data and GDPR model prediction for single and double ionization signifies the importance of single and double plasmon excitations in the ionization process. A large deviation of the GDPR model for triple and quadruple ionization from the experimental data imply the importance of the other low impact parameter processes such as evaporation, fragmentation and a possible solid-like dynamical screening.

1. Introduction

Fullerenes are non-conventional targets for atomic collision studies due to their mesoscopic size intermediate between atoms and solid surfaces. This however, proves to be advantageous experimentally in probing the effect of large electron density on a microscopic level. Collisions of C\textsubscript{60} with energetic electrons, fast and slow heavy ions, and intense photons fields of high power lasers have provided greater insights in to the structural and dynamical properties of C\textsubscript{60}. The solid like electron density of C\textsubscript{60} fullerenes \cite{1} can be probed via inner shell x-ray detection, in single collision conditions, unlike the case with thin films. This offers a unique possibility of studying collective properties of large systems on a microscopic scale. The C\textsubscript{60} molecule is known to exhibit a collective mode of excitation of its loosely bound de-localized pi electrons, called giant dipole plasmon resonance (GDPR) \cite{2}. This collective excitation can play a major role in the ionization of C\textsubscript{60} molecule. The effect of this GDPR on the response of C\textsubscript{60} has been studied in photo absorption studies and intense laser induced ionization and fragmentation of C\textsubscript{60} \cite{3}. LeBrun et al. \cite{4, 5} proposed a simplistic model for the giant plasmon excitation of heavy ion impact ionization of C\textsubscript{60} molecule. Tuschida et al. \cite{6} tested the model with a proton beam at various velocities. This test was very limited since the model predicts very weak energy dependence. However the GDPR excitation probability is quite sensitive to projectile charge state q. Therefore a more stringent test of the model would be to study the q-dependence on a wide range. Nevertheless the effect of this process on single and multiple ionization of C\textsubscript{60} is not very well understood. The effect of this collective excitation mode on the x-ray emission following electron capture from C\textsubscript{60} has been reported earlier \cite{7}. It is only very recently that the q-dependence of single and multiple ionization of C\textsubscript{60} has been investigated by our group using highly charged fast oxygen ions as projectiles. Following the initial results, here we present the details
of such study along with the theoretical model on the giant plasmon excitations for the following
collision systems: 2.33 MeV/u Siq+ + C60 and 3.125 Mev/u Oq+ + C60.

2. Experimental Setup

The fast heavy ion beams of O and Si-ions were obtained from the 14 MV TIFR-BARC pelletron
accelerator facility at TIFR, Mumbai. The beam was made to pass through a post-acceleration stripper
carbon foil to obtain various charge states of the Si and O ions. The post stripped beam was further
charge state analyzed using switching magnet and finally collimated to approximately 2x2 mm2 using
a set of four jaw slits before entering the experimental chamber.

The C60 heater source was placed at the bottom of the chamber perpendicular to the beam direction.
Target vapors were obtained by heating 99.9% pure C60 powder at approximately 400°C in the
vacuum chamber. The C60 powder was initially baked at 150°C for about 8 to 10 hours in vacuum to
remove all the adsorbed impurities. A Wiley-McLaren [8] type time-of-flight (ToF) mass spectrometer
placed perpendicular to the vapour target and beam axis was used to mass analyze the reaction
products. Pusher and puller plates of the ToF mass spectrometer were kept at ± 500V and the flight
tube was floated at -2500V. A micro channel plate (MCP) placed behind the flight tube was used to
detect the recoil ions where as a channel electron multiplier (CEM) was kept behind the pusher plate
to detect the electrons. The MCP front was floated at -4kV. A 10 mm x 1 mm rectangular slit was placed before the electron CEM to reduce electron count rate to a few kHz. The electron signal from the electron CEM was used as start trigger and signal from MCP was used as stop trigger to obtain the recoil ion ToF spectra. The beam current was kept typically about a few nA to maintain the electron and ion count rates below few kHz and hence to reduce the chance coincidence background in the ToF spectrum. The whole system was operated at a base pressure of better than 5x10^{-7} mbar. A SIMION simulation was done for the spectrometer geometry and the collection (i.e. geometrical) efficiency of the system was found to be unity for C_{60} ions. Intrinsic detection efficiency for the MCP was obtained using the empirical formula suggested by Itoh et al. [9]. Data was sent to a CAMAC ADC and stored in a PC. Relative cross sections for various ionization fragments were obtained by taking the area under each peak and normalizing with respect to the beam particle current and oven temperature. For this purpose oven temperature dependence of C_{60} vapor yield was investigated [10]. Relative error in the yields was therefore estimated to be approximately 10% except for C_{60}^{3+} where the error was more (~15%) due to statistical error and multiple peak fitting procedure. Figure 1 shows a typical ToF mass spectrum. The various reaction products can easily be identified. Multiple ionization peaks up to C_{60}^{4+} can be seen. We observe that double and triple ionization peaks are accompanied by corresponding daughter ions produced as a results of neutral C_{2} evaporation. No such evaporation peaks are seen in case of single ionization, suggesting that C_{60}^{1+} are dominantly produced in swift collisions at large impact parameter. A multiple peak fitting procedure was used to extract the contribution of C_{60}^{4+} ionization peak as shown in the inset of figure 1.

3. Theoretical model

Giant dipole plasmon resonance model: The GDPR model was proposed by Lebrun et al. [4, 5] under the frame work of linear response theory and dipole approximation. The model treated C_{60} electron cloud as a free electron gas and the plasmon excitations as oscillation of this free electron cloud against the C_{60} ion core analogous to harmonic oscillator excitation with a characteristic frequency \( \omega \) (\( h\omega = 20\text{eV} \)). The energy transferred from a projectile of charge \( Z \) and velocity \( v \) to a harmonic oscillator is calculated under dipole approximation [13] to be:

\[
\Delta E = \frac{2Z^2e^4}{mv^2b^2} \left[ \frac{\xi^2 K'_1(\xi)}{\xi} + \frac{1}{\gamma^2} \xi^2 K'_1(\xi) \right], \quad \xi = \frac{Eb}{\gamma h\nu}, \quad \text{and} \quad E = h\omega, 
\]

\( b \) is the impact parameter taken from the centre of the molecule. This transferred energy manifests as a plasmon excitation with oscillator strength function \( f(E) \). The effective no. of plasmon excitations \( N(b) \) is then given by,

\[
N(b) = \int_0^\infty dE \frac{f(E)}{E} \Delta E
\]

The oscillator strength function is approximated as a Gaussian [14]. The probability for n-plasmon excitation is given by:

\[
P_n(b) = \left[ \frac{N(b)}{n!} \right]^n \exp\left[ -N(b) \right]
\]

and finally, cross section for n-plasmon excitation is obtained by:

\[
\sigma_n (pl) = 2\pi \int bdb P_n (b)
\]

4. Results and discussion

As can be seen from figure 2(a) the single and double plasmon excitation probability for Si^{14+} peaks at impact parameter of 15 au and 12 au respectively, which is larger than the average radius of the C_{60}
Figure 2 (a). $P_n(b)$ versus $b$ ($b_{\text{max}}$ is the value of $b$ where $P_n(b)$ is maximum). (b) $b_{\text{max}}$ as a function of projectile charge state for single plasmon excitation.

Figure 3. Recoil ion charge state distribution for $C_{60}$ and comparison with GDPR model. (Open circle) represents the experimental ratio of single, double, triple and quadruple ionization to single ionization and (Solid line) represents the same ratio taken from GDPR model. Panel (a) and (b) are taken from reference [10].
molecule. In fact triple and quadruple excitation probability also has substantial contribution from impact parameters larger than 10 au. In Figure 2(b) we plot the impact parameter corresponding to maximum excitation probability as a function of projectile charge state. In the model it is believed that n-plasmon excitation leads to nth ionization of the molecule, which is also seen in intense laser induced ionization of C\textsubscript{60} [3]. However, this approximation is not necessary for present analysis since we shall be interested mainly in the relative cross section and their ratios. Another aspect of the model worth mentioning is that it is a perturbative approach and hence is valid for distant or large impact parameter collisions accompanying small energy transfer.

First we compare the ratios of multiple to single ionization with the predictions of GDPR model. Figure 3 shows the relative yields of Single and multiple ionizations of C\textsubscript{60} normalized at single ionization for various projectile ions. The yields for double and triple ionizations are taken excluding the evaporation yields as it is not expected that GDPR will lead to such evaporation fragments (for more details see reference [15]). It is evident that the normalized yields are in fairly good agreement with the prediction of GDPR model. The agreement is best for double to single ionization, which is in partial agreement with the findings of intense laser experiment [3]. The model over-estimates the data for triple and quadruple ionizations. However, given the complexity of the system the agreement is very
encouraging. In figure 4 we show projectile charge state dependence of C\textsubscript{60} ionization for single and double ionizations for 2.33 MeV/u Si\textsuperscript{q+}, and 3.125 MeV/u O\textsuperscript{q+} projectiles. As shown earlier [15, 16], the single and double ionization yields show fair agreement with the predictions of GDPR model. We observe that in both the cases single and double ionization yields increase linearly with q. These observations are partly consistent with the probability distribution as shown in figure 1. However, in the case of triple and quadruple ionizations a large deviation was observed for 3.125-MeV/u O\textsuperscript{-} projectiles [15] and it indicates that low impact parameter processes such as evaporation, multifragmentation and possibly dynamical screening of the charged projectile have to be considered along with the collective excitation models. However, in the case of high charge state projectile, such as Si, one finds a better agreement with the model as far as the single, double and triple ionization is concerned. Details can be found in [16].

For the sake of completeness, we also show projectile charge state dependence of ionization yields of typical atomic targets like Ne in the inset of figure 4(a) and (b). As evident from the figure, the behaviour of a typical atomic target is very different from that of C\textsubscript{60}. This implies that usual ion-atom theories are not adequate to explain the ionization behaviour of C\textsubscript{60}. For instance, the Bohr-Lindhard theory predicts a q\textsuperscript{2}-dependence for the single ionization [17]. Other models also predict a similar quadratic q-dependence for single and q\textsuperscript{4}-dependence for double ionizations and even higher orders for higher ionizations [18]. This is in sharp contrast to the C\textsubscript{60} ionization, which shows an almost linear q-dependence for single as well as higher ionizations.

5. Conclusion

In conclusion, we have measured the relative ionization cross sections of C\textsubscript{60} in collisions with 2.33 MeV Si\textsuperscript{q+} and 3.125 MeV O\textsuperscript{q+} projectiles and compared the results with a model based on the GDPR process. The single and double ionization yields are found to depend linearly on projectile charge state, which is contrary to that observed for a gaseous target. This behavior is in agreement with the predictions of the GDPR model. The increased deviation of the model for triple and quadruple ionizations is possibly due to the interplay between the large impact parameter process (such as GDPR) and low impact parameter processes, such as, fragmentation, evaporation and “solid-like” dynamical screening.

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