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Electron impact ionization of large krypton clusters*

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We show that the detection of ionization of very large van der Waals clusters in a pulsed jet or a beam can be realized by using a fast ion gauge. Rapid positive feedback electron impact ionization and fragmentation processes, which are initially ignited by electron impact ionization of the krypton clusters with the electron current of the ion gauge, result in the appearance of a progressional oscillation-like ion spectrum, or just of a single fast event under critical conditions. Each line in the spectrum represents a correlated explosion or avalanche ionization of the clusters. The phenomena have been analysed qualitatively along with a Rayleigh scattering experiment of the corresponding cluster jet.

Keywords: atomic clusters, ion gauge, electron impact ionization
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1. Introduction

Research of ionization and fragmentation of van der Waals clusters has long been a subject of interest.[1,2] However, the data on very large clusters are scarce. In recent years, renewed interest has been devoted to the ultra-short intense laser interaction with large rare gas clusters.[3−11] In these studies the clusters represent certain unique media intermediate between atoms, molecules and solids. As a result, a wealth of experimental findings were reported for cluster sizes of a few hundred to about 10^6 atoms, including keV x-ray generation, production of very energetic electrons and ions, highly charged states and the table-top nuclear fusion, etc. Different theoretical models have been proposed to account for the findings.[3−5,7] It is generally agreed that the direct optical ionization is responsible for the initial ionization of atoms in a cluster which interacts with an intense laser pulse; however, controversies arise regarding the following ionization and expansion processes of the cluster. For example, the electron–ion collisional ionization mechanism is proposed and believed to be of significance.[5] So, it would be interesting to see what happens in the electron impact ionization of clusters in a gas. During the course of preparation and size characterization of large rare gas clusters (Ar, Kr and Xe) with sizes ranging from 10^2–10^6 atoms, we have studied experimentally the electron impact ionization of large clusters in a dense gas jet or a low-density beam using a fast ion gauge (FIG). Some years ago, following the pioneering work of Gentry and Giese,[12] we employed a FIG detector to diagnose the intensity and profile of an intense pulsed molecular beam.[13] However, the situation is changed dramatically when a gas of clusters is used instead of a gas of individual molecules. We find, to the best of our knowledge for the first time, that the FIG detector provides an informative response to a gas of large clusters, depending sensitively on the cluster source conditions while the FIG's parameters are kept identical. It has been revealed that a rapid ionization process of the clusters within the gauge occurs as soon as the electron density, which is the sum of the gauge's emission electron current and that resulting from ionization of atoms of the clusters, reaches a critical value. Whenever a rapid ionization event is dealt with, the intra-cluster as well as inter-cluster processes must be considered. Along with the study of electron impact ionization of clusters, experiments for cluster size characterization

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are also conducted in this lab by using a Rayleigh scattering method. A surprising analogy of a spectrum of the time-resolved Rayleigh scattered signal from large krypton clusters with the profile of an ion spectrum obtained under specified conditions of the corresponding krypton clusters was found.

2. Experimental setup

The electron impact ionization is used to investigate the ionization process of a gas flow of large size van der Waals clusters. As shown in Fig.1, the clusters, generated in the adiabatic supersonic expansion of a high-pressure gas into vacuum through a conical nozzle (300μm, 10° half opening angle, 28mm long), are detected by an ion gauge, which provides an electron current variable from less than 0.1 to 200μA, and the energy of the electrons is controlled by adjusting V_g, the electric potential of the grid of the ion gauge, ranging from 0 to 300V. The gauge has two opposite windows on its cylindrical collector so that the gas flow is allowed to pass through it. The ions produced during electron impact ionization of the clusters within the gauge are collected by the gauge collector. In order to study electron impact ionization of large clusters in a dense gas jet, the gauge was positioned about 3cm downstream from the nozzle exit. An alternative experimental scheme is also arranged (not shown in detail in Fig.1) to investigate the electron impact ionization of large clusters in a low-density flow in which the ion gauge is located after a skimmer and is about 80cm downstream from the nozzle.

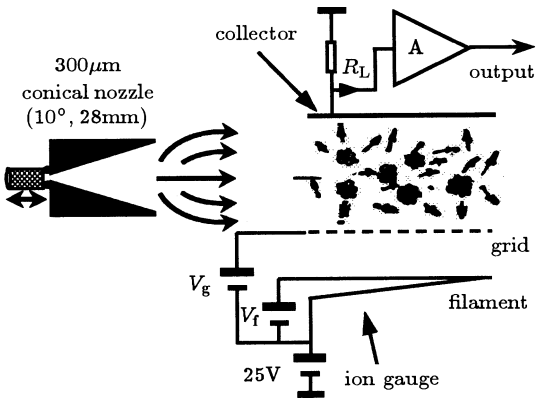


Fig.1. Experimental configuration of electron impact ionization of large krypton clusters. A denotes a preamplifier.

3. Results and discussion

Figures 2(a) and 2(b) show the time evolution of the charge yield of ions produced within the ion gauge during the electron impact ionization of a dense krypton cluster flow. For the ionization process in Figs.2(a) and 2(b) to be understood more reasonably, in Fig.2(c) a Rayleigh scattering experimental result is given to show the time and size characteristics of the corresponding krypton cluster flow. The two-plateau structure of the curve in Fig.2(c), which only occurs for krypton or xenon clusters, could be considered to result from a second gas injection into the nozzle due to a bounce of the valve plug from the rubber O-ring seal when it is forced to close the valve.[14,15] The other possibility which is responsible for the observation may be the cascade supersonic gas expansion because there is a small room ($\sim$15mm^3 in volume) just before the nozzle orifice (not shown in Fig.1). Because the second gas injection into the nozzle is very limited, the atomic density before clustering should be much lower for the latter plateau than the former one. Considering the scattered light intensity $S_R \propto n_0 \bar{N}_c (n_0$ is the atomic density before clustering and $\bar{N}_c$ is the average number of atoms per cluster), we can see that the average cluster size at the latter plateau is about one order of magnitude larger than that at the former one. In our experiment of electron impact ionization with krypton clusters, the backing pressure is 6×10^5Pa, the average size of the clusters at the first plateau is measured to be about 4×10^3 atoms,[14] and, then, that of the clusters at the second plateau should be about 4×10^4 atoms per cluster. Clearly, the size increase of the clusters is at the sacrifice of the number density of the clusters with the same factor.

In Fig.2(a), the curve shows a usual gas pulse profile featuring the cluster flow. There is no evidence that a second gas injection exists as seen in Fig.2(c), indicating that the amount of gas flow is very limited in the time period of the second plateau. However, an oscillation pattern of the ions is suddenly built up when the electron current of the gauge filament is just increased a very little (Fig.2(b)). In Fig.2(a), the curve is easy to understand: when the clusters pass through the ion gauge and are bombarded by the electrons, for various cluster sizes, there is not too much difference for the 'effective' ionization cross-section, i.e. the total ionization cross-section of a cluster divided by the number of the cluster constituents.[16] The ions generated are approximately proportional to the number of atoms within the gauge volume. It can be seen from Fig.2(a) that the ion current remains almost changeless in the time period from 1 to 4 ms, indicating that this time span is the opening time of the valve. During this period, the atomic density re-

mains unchanged but the cluster size is getting larger. After this period, the valve is closed and the atomic density decreases gradually; as a result the ion current signal decreases correspondingly. This character is also manifested in Fig.2(c). When the valve reopens due to the bounce of the valve plug as we have suggested above, because the leaked gas would be very limited, its influence cannot be seen in Fig.2(a). It is interesting to note that the time evolution of the oscillation pattern in Fig.2(b), including the appearance and disappearance of the oscillation pattern, is consistent with the positions of the two plateaus in Fig.2(c). Furthermore, the profiles of the curves in Figs.2(b) and (c) demonstrate an obvious resemblance with each other although the principles of the Rayleigh scattering experiment and the electron impact ionization experiment are basically different. Especially, Fig.2(b) shows again the second increase in signal intensity as seen in Fig.2(c), however, in a dramatically sensitive and fascinating way.

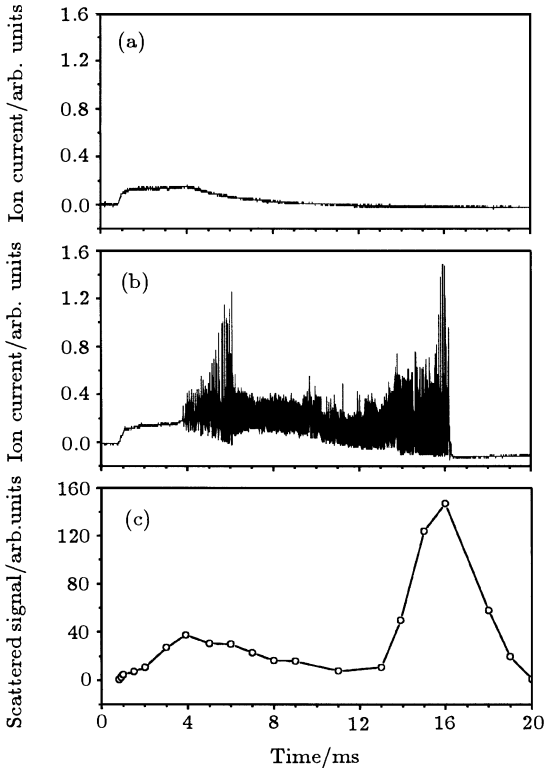


Fig.2. Measured charge yield of ions produced in electron impact ionization of large krypton clusters (average size $\bar{N}_c \approx 4\times10^3$ atoms) as a function of time with the gauge filament voltage being (a) 4.04V (the corresponding filament emission electron current is measured to be 0.07μA) and (b) 4.06V (filament emission current is just a little more than that in (a)). Figure 2(c) shows the time history of the corresponding krypton cluster flow obtained using a Rayleigh scattering method for comparison.

A number of experiments using He, Ar, and Xe gases have been conducted as well and all the data obtained lead to the conclusion that oscillation patterns of ion signals produced in electron impact ionization processes within an ion gauge are only related to a flow consisting of large clusters. In the process of electron impact ionization of a cluster flow, the cluster may produce more electrons via cascade ionization/fragmentation. These newly generated electrons will be accelerated in the electric field defined by V_g-ground and enhance the ionization of the clusters nearby. If the generated electrons are numerous enough compared with the filament emission, this 'positive feedback' and intercluster process will continue and finally lead to the occurrence of the avalanche-like ionization of the clusters. All the ions produced in a very short period of time have been evacuated by the static electric field completely, giving rise therefore to a sudden increase of the ion current shown as a line in the spectrum in Fig.2(b). The character that the first avalanche ionization, i.e. the left-most line of Fig.2(b) corresponds to the local size maximum of the cluster flow (Fig.2(c)) is an indication that the critical conditions of the correlation explosion of the clusters are just met. After the explosions some electrons will remain in the gauge in an oscillating manner by crossing the grid many times, resulting in an equivalent increase of the electron current of the gauge. Only with the aid of this increase of electrons does the next following correlation ionization of the clusters become possible. These progressional avalanche ionization events of the clusters proceed until the time about 16ms after the trigger of the pulsed valve when the second local maximum in cluster size is encountered.

To indicate the time evolution of the oscillation pattern clearly, Fig.3 shows the spectra of the ion current at three different time periods in Fig.2(b). Figure 3(a) shows the evolution process of the explosion at the beginning. After the first correlated explosion event of the clusters, the cluster flow continues to enter into the evacuated gauge volume. Favoured by an enhanced impact electron quantity, the second correlated explosion occurs as the newly entered cluster flow goes into the gauge to such a depth that the critical conditions are met. This process repeats again and again. As the process continues, the time interval between the adjacent explosions decreases, indicating that the depth of the cluster flow entering into the ion gauge, which is just needed for the correlated cluster explosion to occur, decreases continuously. From

the time about 6 to 13 ms after the trigger of the pulsed valve, the explosion process enters into a steady but quick oscillation state. This character is exhibited clearly in Fig.3(b). The comparatively smaller ion current and shorter time interval indicate that the cluster flow enters the gauge for only a limited depth when the explosion occurs. Comparing the leading edge of the second part in Figs.2(b) and 2(c), we see that the ion current in Fig.2(b) increases correspondingly with the increasing scattered signal, indicating that the size of the clusters should play a crucial role in the electron impact ionization. In this case the average gas density is very low; however, the avalanche ionization of the clusters results in an ion current which is several orders of magnitude larger than that in Fig.2(a). As shown in Fig.3(c), due to the continuous decrease of the average gas density, the time interval between two adjacent explosions increases, and finally the progressional explosions end when the critical conditions are no longer satisfied in the case that the cluster flow passes the whole gauge. From the time interval of the last two explosions (it is interesting to see that this time interval is equal to the one of the first two explosions in Fig.3(a)) and from the gauge dimension of 2.5cm, the velocity of the krypton cluster flow is estimated to be about 2.5×10^4cm/s, which is consistent with the theoretical limiting velocity ($\sim3.8\times10^4$ cm/s) of krypton atoms in a supersonic flow at room temperature. The extraction process of the ions, which depends on V_g and the gauge dimension, is estimated to be 5–10μs by using the widths of the lines in the spectrum, much shorter than the time interval of $\sim100\mu$s that the cluster flow needs to cover the gauge.

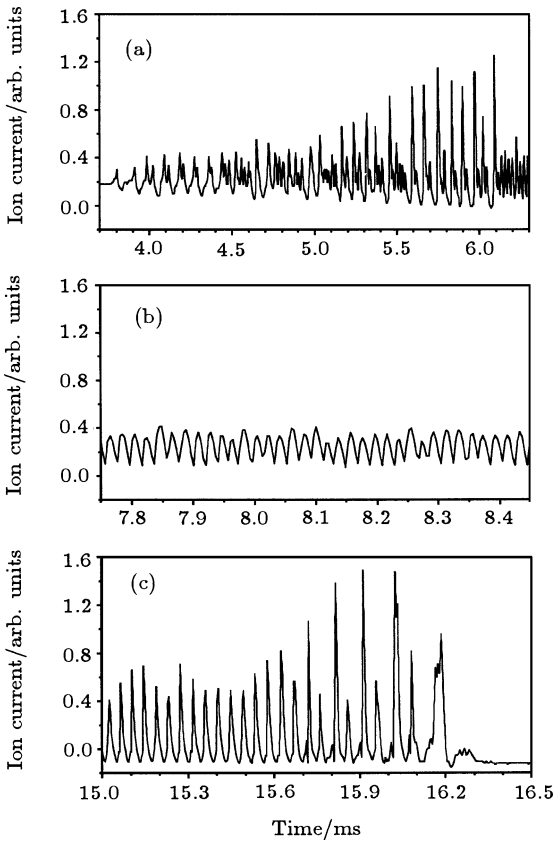


Fig.3. Expanded ion current signal in three different time spans of Fig.2(b).

Besides the experiment performed in the near-nozzle configuration where a dense cluster flow and an extremely low electron current are used, far-nozzle experiments have also been conducted in which a low density cluster beam and a larger gauge electron current (1.2μA) are employed. The experimental apparatus is similar to Fig.1, but the ion gauge is placed about 80cm far from the nozzle (the average atomic density there is estimated to be $\sim1\times10^{12}$ atoms/cm^3), and with a skimmer located between the nozzle and the ion gauge. The ion gauge is movable along the cluster beam direction. By varying the distance between the nozzle and the ion gauge, we can study the dependence of the electron impact ionization process on cluster density. Figure 4 shows that under critical experimental conditions, a single avalanche process of the clusters is realized. The low atomic density in the flow and the specified low electron density serve to support the concept of the critical conditions under which only a single avalanche ionization of the cluster is established. The sharp character of the event in Fig.4 might provide a promising possibility of application of it in the diagnostics of a beam of large clusters, including the beam velocity, and perhaps the atomic density and the cluster size in the beam as well. For example, from the peaks obtained at different gauge positions and the corresponding times, a velocity of the cluster flow containing an average size of about 2×10^5 atoms per cluster can be readily calculated to be about 1.5×10^4cm/s, in good agreement with that estimated above. However, the diagnostics of atomic density and cluster size in the beam is an extremely difficult problem. An analytic method and calibration procedures would be required.

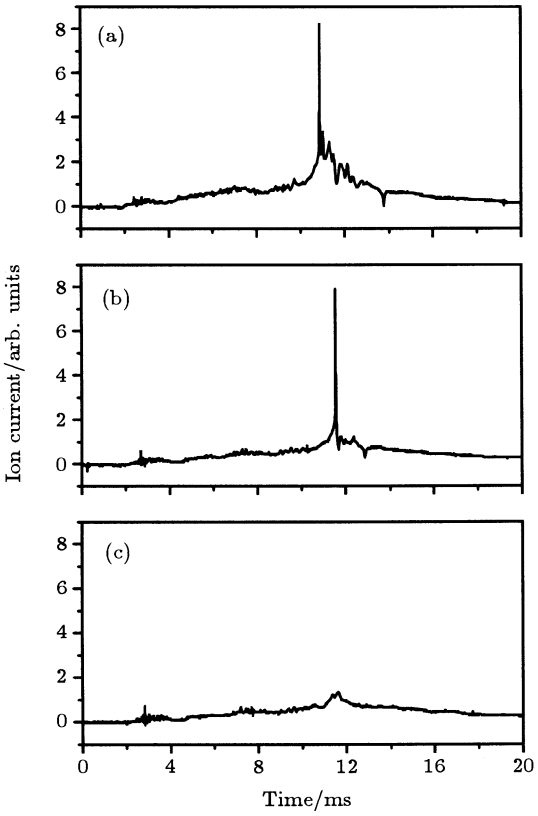


Fig.4. Measured ion signal of Kr clusters as a function of the distance between the nozzle and the ion gauge. (a) distance=80cm. (b) distance=90cm. (c) distance=92cm. The average cluster size is 2×10^5 atoms and the gauge's emission current is 1.2μA.

Experiments have also been carried out to study the dependence of the electron impact ionization of large clusters on cluster size and electron energy. The results indicate that larger clusters and higher electron energy favour the avalanche ionization process of the clusters. All data obtained provide evidence that as the density of the electrons, which are the sum of the electrons emitted by the ion gauge filament and those produced in the ionization process of clusters, reaches a critical value within the gauge, a sudden increase of ion generation will take place like an avalanche.

4. Conclusion

In summary, a fast ion gauge has been used to study electron impact ionization of large van der Waals clusters (10^3–10^5 atoms) in a dense cluster jet and in a low-density cluster beam. It is proposed that the ionization of the clusters within the ion gauge proceeds in an avalanche manner by a sequential feedback ionization process via fragmentation/explosion of the clusters. The ion gauge detection can be very sensitive in the sense that large ($\sim 10^4$ atoms) krypton clusters in an extremely low average atomic density ($\sim 1\times10^{12}$ atoms/cm^3) are strongly ionized under an impact electron current of as low as 1.2μA. Finally, the possibility of using this simple method for the characterization of a gas jet/beam of large van der Waals clusters is proposed.

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