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Photon emission under MeV carbon cluster impact for probing cluster–solid interactions

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Abstract

First experimental results on the luminescence induced in thin molecular samples by the impact of single cluster ions C_n^+ ($n = 2$–10) at 1 MeV/atom have been reported. The cluster ions were produced and accelerated at the Erlangen tandem accelerator. The samples were a 1 μm CsI layer deposited onto a 2 μm aluminized polyester foil and a 100 nm POPOP layer prepared onto a 20 nm formvar film. The bombarding ions could traverse the samples but the cluster constituents did not remain in proximity in all cases. The luminescence was registrated by time-correlated single photon counting using either secondary electrons or transmitted clusters for starting the measurement. The relative photon yield obtained with clusters of various size increases linearly with the number of cluster constituents. The increase is less pronounced in case of the thin sample. This experimental result can be explained by the fact that the cluster ions leave the exit side of the sample as intact entities and that the photon emission is reduced in regions of high energy density.

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1. Introduction

The impact of a cluster ion on a solid can be described as the nearly simultaneous arrival of several atoms at the surface within close distances. As a consequence, the deposited local energy density is very high and leads to cooperative effects, e.g., to an enhanced secondary ion emission [1,2] or to a sublinear electron emission effect [3]. From the mechanisms discussed in this context the paving-the-way effect, which is based on the fact that atoms are pushed away from the track by in front running cluster atoms, and the sweeping-out-electron effect considering that fewer electrons are available for the interaction with behind running cluster atoms may be quoted here [3,4].

The analyis of the luminescence produced in solid samples by incident energetic atomic ions yields valuable information on the ion-solid interaction [5,6]. The luminescence has been found to come from radiative transitions within the bulk

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of the sample due to relaxations of the electronic system. It originates from the fluorescence of intact molecules or from the decay of self-trapped excitons. The luminescence yield obtained with different ions is proportional to the energy deposited into the electronic system only at constant initial ion velocities. Calculations based on the energy deposition by the secondary electrons yielded that the luminescence is produced in the region of low energy deposition at larger radial distances from the ion track [6].

In this paper, we present first experimental results on the photon emission induced in thin molecular samples (POPOP or CsI) by single cluster ions C_n ($n = 2$–10) at 1 MeV/atom. The cluster ions were produced in a conventional sputter source and injected into the Erlangen tandem accelerator [7–9]. The luminescence was registrated by time-correlated single photon counting using either secondary electrons or transmitted clusters for starting the measurement.

2. Experimental

2.1. Production of MeV carbon cluster ions

The carbon cluster ion beams were produced at the Erlangen tandem accelerator with energies of 1 MeV/atom. First, singly charged negative cluster ions C_n^- ($n = 2$–10) were produced in a conventional sputter source and then injected into the tandem accelerator. They were accelerated toward the accelerator terminal biased with a voltage up to 5 MV. A small fraction of the C_n^- ions was stripped at the terminal to singly charged positive cluster ions while passing a gas channel (about 10^{-5} mbar N_2). After passing the terminal the C_n^+ ions were accelerated to the high energy end of the machine. The cluster ions were then magnetically analyzed (deflection through 3°) and injected into the recipient. The flux applied was between 100 and 500/s.

2.2. Setup for starting with electrons (Experiment I)

In the setup "Experiment I" depicted in Fig. 1, electrons produced by the impact of the cluster

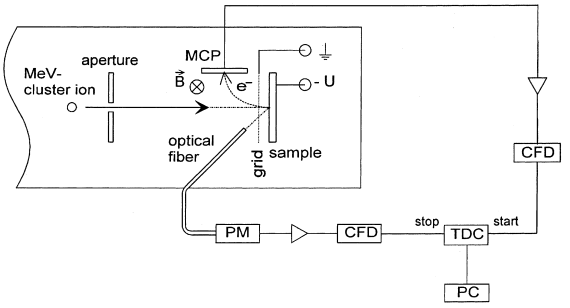

Fig. 1. Setup "Experiment I" in which the start signals were obtained from secondary electrons leaving the sample surface. MCP: micro channel plate, $\vec{B}$: magnetic field, $-$U: voltage (about – 100 V), PM: photo multiplier, CFD: constant fraction discriminator, TDC: time-to-digital converter, PC: computer.

ions were used for starting the measurements. Therefore, the incident ions must not necessarily traverse the sample. The photons were produced in a high vacuum recipient in which the accelerated cluster ions were injected. After passing a last aperture, the cluster ions impinged onto the target, a 1.0 μm CsI sample. Electrons originating from the ion impact were accelerated in the electric field between sample and grid, and deflected by a magnetic field ($\vec{B}$) onto a micro channel plate. The MCP-detector signal was amplified and then formed by a constant fraction discriminator (Ortec Quad CFD 934). The CFD output pulses were used to start a time-to-digital converter (CTN/M2, IPN, Orsay, France). For the collection of photons produced in the sample, an optical fiber (diameter: 1 mm, black coated) was inserted into the recipient. The front face of the optical fiber was directed to the center of the target at a distance of about 5 mm and under an angle of about 20° to the surface normal of the target (the fiber was protected against strikes of incident ions). The other end of the fiber was mounted at the entrance window of a photomultiplier tube (Hamamatsu R269) that was placed outside the recipient. The photodetector was kept at -20°C to reduce the thermal background (to about 30 counts/s). The accessible spectral range is 300–750 nm (corresponding to 1.7–4.1 eV) limited by transmission of the optical fiber and by detector response. The samples are nearly transparent with regard to the produced luminescence. The output signals of the photo-

multiplier were amplified and also formed by a CFD and then given to the TDC as stop pulses. By recording the time interval distribution into a personal computer via a direct memory interface (DMI, from IPN), the time profile of the photon emission was obtained. The relative photon yields were then determined by integrating the measured time profile.

2.3. Setup for starting with transmitted clusters (Experiment II)

In the setup "Experiment II" depicted in Fig. 2 cluster ions traversing the sample were used to start the measurements. The photons were produced in the same high vacuum recipient as in Experiment I in which the cluster ions were injected and after passing a last aperture impinged onto the target, a 100 nm POPOP sample. After traversing the sample the cluster or the remaining constituents hit a Si-detector (diameter of active area ≈5 mm) placed at a distance of about 5 cm behind the sample. The Si-detector provided a fast timing as well as a slow energy signal. Using the energy signal, it is possible to measure the energy spectrum of the incident ions by means of a pulse height analyzer. A typical energy spectrum obtained with C_8 clusters without (a) and with (b) transiting the POPOP sample is depicted in Fig. 3b. If an intact cluster hits the detector one observes the maximum energy value (see hatched area in Fig. 3b). For measuring photons which are correlated to starts produced by intact clusters, a

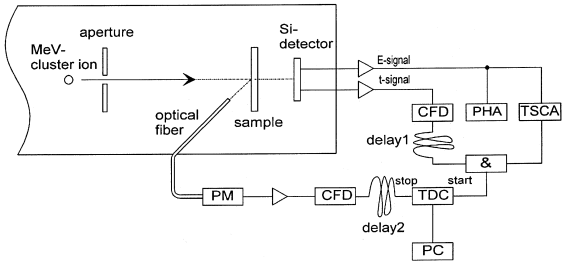

Fig. 2. Setup "Experiment II" in which the start signals were obtained from transmitted clusters. PHA: pulse height analyzer, TSCA: timing-single-channel analyzer, &: logic "and" gate, PM: photo multiplier, CFD: constant fraction discriminator, TDC: time-to-digital converter, PC: computer.

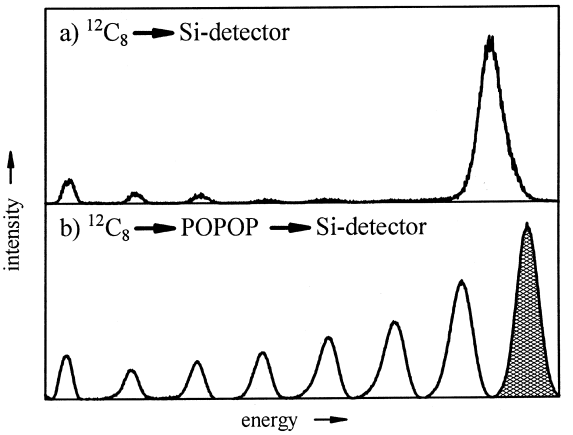

Fig. 3. Energy spectrum of a C_8 cluster beam without (a) and with (b) passing the POPOP sample.

fast-slow-coincidence was used. The energy signal was passing a timing-single-channel analyzer (TSCA) only when the pulse height was within the correct energy window. The timing signal was delayed (delay1) and a logic "and" gate (&) was used for gating the fast timing with the slow energy signal. The so processed signal started the measurement at the time-to-digital converter. For the collection and detection of photons induced in the sample by incident ions, the same procedure as in "Experiment I" was applied. But a delay (delay2) between CFD and TDC was used in order to compensate the time used for delaying the timing signal of the Si-detector. The determination of the relative photon yield was also carried out as in Experiment I.

2.4. Sample preparation

The 1.0 μm layer of CsI used in Experiment I was evaporated onto an aluminized polyester foil (diameter: about 8 mm; thickness: 2.1 μm). The sample surface was rather plane allowing a precise determination of the thickness by optical interference. The POPOP sample used in Experiment II was prepared by depositing a layer of about 100 nm onto a 20 nm formvar foil, also by thermal evaporation. The thickness was determined by weighing using an on-line quartz-microbalance system and checked by measuring the energy loss of a ^{32}S beam.

3. Results and discussion

3.1. Bombardment of a 1.0 μm CsI sample

The 1.0 μm CsI sample was bombarded by C_n^+ ions ($n = 1$, 2, 4, 6, 8 and 10) with energies of 1 MeV/atom. The relative photon yields measured with setup Experiment I are plotted in Fig. 4 versus the number of cluster constituents n. As clearly shown in this figure, Φ increases linearly with the cluster size. The straight line does, however, not intersect the abscissa at the origin indicating that the yield per atom Φ/n is the largest for monoatomic carbon ions C_1^+. The photon yield per atom Φ/n is also plotted in Fig. 4 showing a clear decrease with n. The fact that Φ/n is nearly constant for $n \geqslant 4$ means that the photon yield results from n constituents which traverse the major part of the sample well separated, i.e. that the cluster ions break apart after passing only a small portion of the sample (for a scheme of this break-up process see Fig. 5). It has been shown that C_{10} clusters with an energy of 1 MeV/atom traversing a gold layer stay close together for a distance of roughly 140 nm before they break apart [10]. A similar behaviour is expected for 1 MeV/atom C_n clusters passing through a CsI sample. Since 140 nm represent only a small fraction of the C_n trajectory in the 1 μm thick CsI sample, the above interpretation of the Φ versus n curve is well justified. The

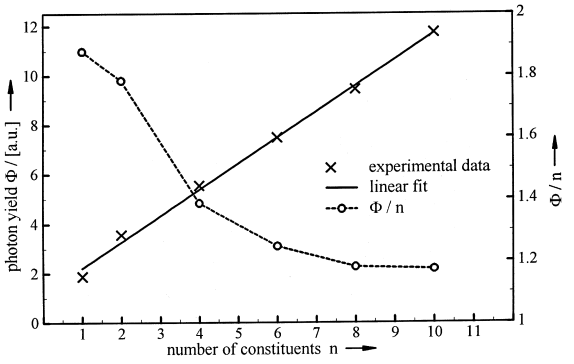

Fig. 4. Relative photon yield (crosses) versus number of carbon cluster constituents n; setup: Experiment I; sample: 1.0 μm CsI. The line represents a linear fit. Φ/n values are also depicted and joined by a dashed line. The error bars are smaller than the data points.

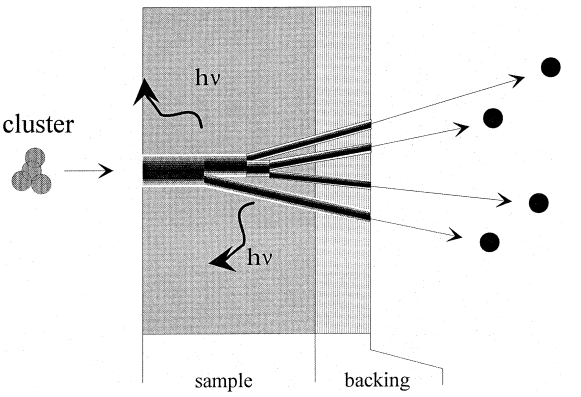

Fig. 5. Scheme of a C_4 cluster traversing the sample.

observation that the yield $\Phi(C_1)$ exceeds the Φ/n values for $n \geqslant 4$ reflects simply the fact that the cluster constituents remain in close proximity for a small distance d (small compared to the sample thickness). Therefore, the measured increase of the relative photon yield with cluster size is explained by a complete break-up of the cluster ions into separated individual atoms after getting into the sample which then traverse the CsI layer independent from each other and produce photons proportional to their number n along their trajectories.

3.2. Bombardment of a 100 nm POPOP sample

A 100 nm POPOP layer deposited onto a 20 nm formvar substrate was bombarded by C_n^+ ions ($n = 1$, 2, 4, 6 and 8) with energies of 1 MeV/atom. Due to the small thickness of sample and substrate, a considerable part of the cluster ions could traverse the sample without noticeable separation of their constituents as checked by the spectrum in Fig. 3b. The relative photon yields were measured using setup Experiment II allowing only starts correlated to transmitted clusters with no loss of constituents (see hatched area in Fig. 3b). The relative photon yield observed in this case is plotted versus the number of constituents n in Fig. 6. Obviously, the photon yield depends linearly on n as in the case of Experiment I. But the slope of the yield curve is significantly smaller. This is well demonstrated in Fig. 7, in which the results of both experiments are shown (both $\Phi(C_1)$ values are

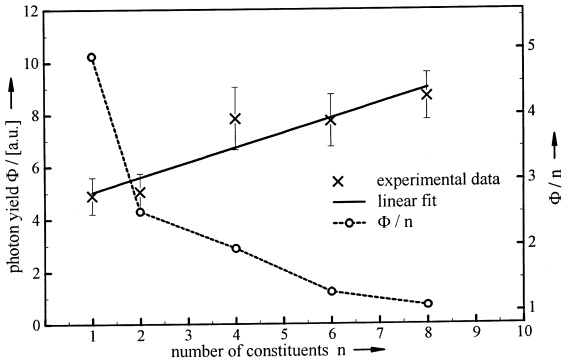

Fig. 6. Relative photon yield (crosses) versus number of carbon cluster constituents n; setup: Experiment II; sample: 100 nm POPOP. The line represent a linear fit. Φ/n values are also depicted and joined by a dashed line.

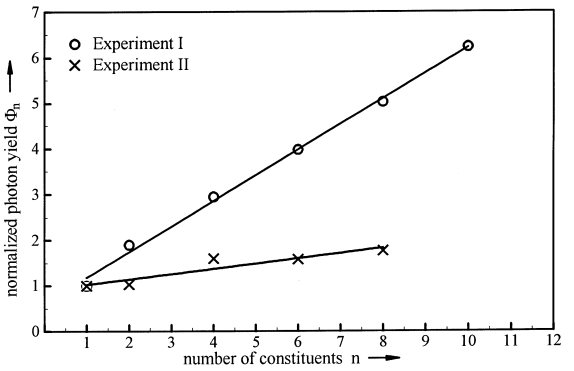

Fig. 7. Normalized photon yield versus number of carbon cluster constituents n; data taken from Experiment I and Experiment II. The straight lines represent linear fits through the data points. The data are normalized so that both C_1 yields have the value 1.

normalized to 1). The photon yield per atom Φ/n is also plotted in Fig. 6 showing a strong decrease with n. This decrease reflects that the cluster atoms stay close together for the major part of their trajectories through the 100 nm POPOP sample, and that no photons are produced in a region close to the ion path where the energy density is very high (leading e.g. to destruction of the molecular or crystal structure, or to exciton-exciton annihilation [11]). In fact, in case of the 100 nm POPOP sample one can safely assume that the clusters leave the exit side of the sample as intact entities (C_{10} clusters in gold start to break up after ≈140 nm [10]). This means that the energy density ϵ in a cylindrical volume around the trajectory of the intact cluster increases linearly with the number of constituents. This can be justified if one assumes that a major portion of the deposited energy goes into kinetic energy of the secondary electrons produced in the ion–matter interaction and if one notes that the range of the electrons is proportional to the square of the ion velocity. The maximum range of these electrons (assumed to evade perpendicular from the the ion path) determines the radius of the cylindrical volume into which the ion energy is deposited. Since this volume remains constant in case of cluster ions (all atoms have the same velocity), one obtains the linear increase of ϵ with n mentioned above. Thus, the yields plotted in Fig. 6 can be thought to be given as a function of ϵ instead of n, i.e. the dependence of photon emission on the energy density along the ion path is yielded by the measurements presented here. It is further interesting to note that luminescence measurements are suited to determine how far a cluster traversing a sample remains intact. In the case of Experiment II, one has simply to increase the sample thickness till a deviation from the measured Φ/n curve is observed.

4. Conclusion

First experimental results on the luminescence induced in thin molecular samples by the impact of single cluster ions C_n^+ ($n=2$–10) at 1 MeV/atom have been reported. The cluster ions were produced and accelerated at the Erlangen tandem accelerator. The samples were a 1 μm CsI layer deposited onto a 2 μm aluminized polyester foil and a 100 nm POPOP layer prepared onto a 20 nm formvar film. The bombarding ions could traverse the samples but the cluster constituents did not remain in proximity in all cases. The luminescence was registrated by time-correlated single photon counting using either secondary electrons or transmitted clusters for starting the measurement. The relative photon yield obtained with clusters of various size increases linearly with the number of cluster constituents. The increase is less pronounced in case of the thin sample. This experi-

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