SPECTROSCOPIC STUDIES OF SUPERSONIC SEMICONDUCTOR CLUSTERS

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ABSTRACT

Observation of silicon, germanium, and GaAs clusters produced by laser vaporization followed by supersonic expansion in a helium carrier is reported. Evidence that semiconductor clusters differ dramatically from similarly prepared metal clusters is discussed.

DISCUSSION

A detailed description of the apparatus used in this study has been given previously.[1] Since semiconductor elements are more readily available in the form of wafers than in the form of rods as required previously, a rotating disc laser-vaporization cluster beam source was developed. The 2nd harmonic output of a Nd:YaG laser (30 mj/pulse) was focused onto a 0.1 cm spot on the disc, producing a super-heated plasma entrained in a near-sonic flow of pulsed helium carrier gas. Free expansion of this cluster/helium mixture into a large vacuum chamber produced an intense supersonic beam, which was collimated by two skimmers. Subsequently, the cluster beams were characterized by F_2 (7.9 eV) and ArF (6.4 eV) excimer laser ionization accompanied by time-of-flight analysis.

An interesting observation of this study is that Si and Ge cluster beams produce almost identical mass spectra at moderate ArF excimer laser fluence (~0.5 mJ/cm^2), as shown in Fig. 1. Cluster ion signals in the size range of 6 and 11 atoms are particularly strong and show predominant contributions from fragments of higher clusters. This similarity means that the nascent cluster size distributions in the beam must be essentially the same for both elements. Furthermore, large Si and Ge clusters have almost the same fragmentation pattern and fragment primarily by fission into daughter ions in the 6-11 atom size range. This fragmentation pattern is quite different from that observed for metal clusters in the same apparatus. Time delay studies in two-color-experiments involving initial excitation with the SHG (2.4 eV) and THG (3.5 eV) of the NdYag laser followed by excimer laser ionization established that both Si and Ge clusters have excited electronic states with lifetimes of ~100 ns. The existence of such long-lived excited states indicates that there is probably an energy gap between the band of electronic states being excited and the ground state.

A pronounced even/odd alternation in the photoionization cross-section of Ga_xAs_y clusters, was observed depending only on the total number of atoms in the cluster.[2] Clusters in the 5-21 atom range with an odd number of atoms were one-photon ionized by low energy ArF excimer laser pulses (6.4 ev) [Fig. 2a]. The spectrum in Fig. 2b was taken with an F_2 excimer laser at low fluence, showing that all

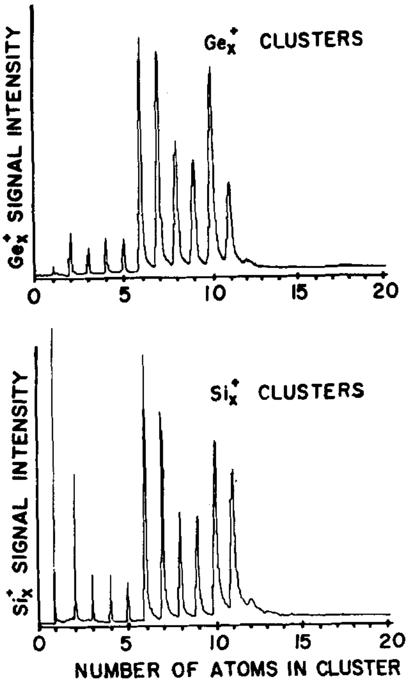

Fig. 1. Time-of-flight mass spectrum of Ge and Si clusters.

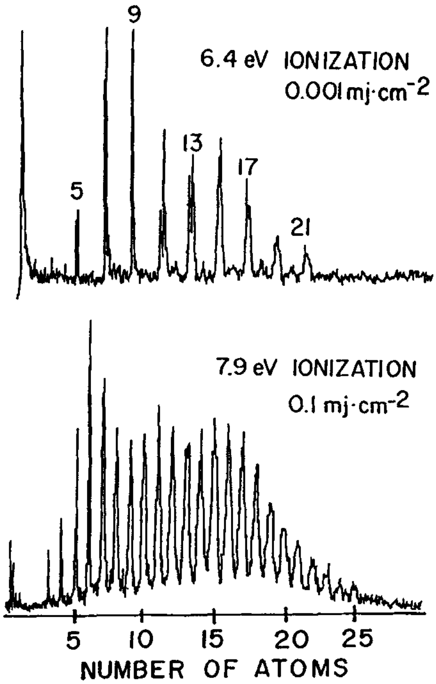

Fig. 2. Time-of-flight mass spectrum of Ga_xAs_y clusters ionized with ArF and F_2 excimer lasers.

clusters can be readily one-photon ionized at 7.9 ev. This even/odd alternation in ionization properties of clusters supports the view that the even clusters have fully paired singlet ground states with no dangling bonds, while the odd clusters have unpaired electrons which are more weakly bound.

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