RESEARCH: RARE GAS CLUSTERS

Explosions and Dissociations

Henry Hutchinson

Clusters containing many identical atoms are of considerable interest because they form a bridge between individual atoms and molecules on the one hand and the bulk solid-state form of matter on the other. Structures of up to many thousands of atoms can be made quite readily, and studying them gives some valuable information about how the properties of matter change as one progresses from a single atom to the solid state. In particular, the way in which clusters interact with light is of particular interest because, although the density of the atoms is similar to that of the solid, the overall size of the assembly is less than the wavelength of light and so all the atoms see essentially the same optical electric field. This results in behavior that is quite different from atoms or solids. In particular, free electrons that are driven by the oscillating electric field of the light wave can exhibit collective motion producing a so-called "giant" dipole. An indication of the remarkable interaction between clusters and light has recently been demonstrated by Lezius *et al.* (*1*) who have studied the explosion of rare gas clusters when irradiated with intense laser radiation.

Early experiments by Rhodes and his coworkers at the University of Illinois at Chicago showed that, when clusters of noble gases were irradiated with short pulses of ultraviolet laser radiation with durations of less than 1 ps at very high intensities ($>10^{17}$ W cm^{-2}), x-rays were generated with intensities and photon energies that were much higher than would be expected from individual atoms (*2*). These results were quite unexpected, and Rhodes *et al.* attributed them to the recollision of energetic electrons (which were produced by photoionization of the atoms) with the atoms within the cluster, which coupled strongly to the inner atomic shells to eject electrons to produce "hollow" atoms. This work stimulated interest from other groups, not only in the anomalous x-ray emission (*3*) but in harmonic generation (*4*) and the dissociation dynamics of the clusters after they are irradiated (*1*, *5*) with high-intensity laser light.

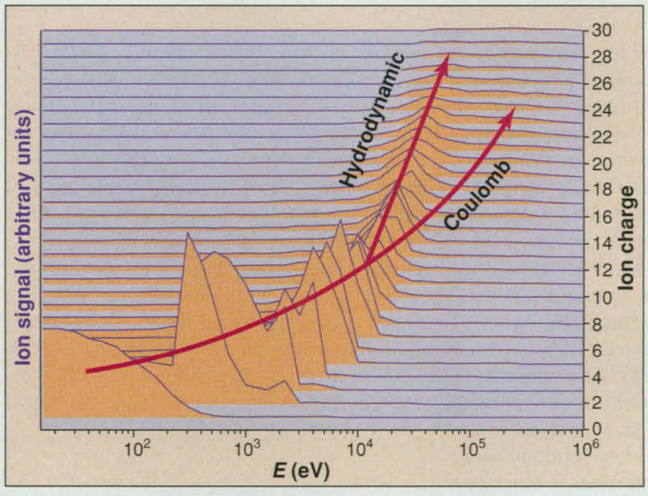
When an intense, short pulse of laser light interacts with a cluster, the constituent atoms are ionized either by multiphoton absorption or, if the intensity is sufficiently high (more than about 10^{15} W cm^{-2}), by tunneling of the electron through the electrostatic potential barrier, which has been modified by the electric field of the laser light. The liberated electrons cannot easily leave the cluster and are held by the space-charge attraction of the parent ions. They are forced to oscillate by the light field, and, because of the very close proximity of the atoms, they collide frequently, receiving more energy from the light field by the process of inverse bremsstrahlung. As a result, the electrons and, ultimately, the ions acquire very large amounts of energy, resulting in the inevitable explosion of the cluster into its electron and ion fragments.

Several studies have been done to try to understand the mechanism by which this absorption and explosion take place (*1*, *5*). The experimental technique is quite simple. Clusters of the noble gases (xenon, krypton, and argon) are formed by the adiabatic expansion of a high-pressure gas through a pulsed nozzle or jet. When the gas cools rapidly, the atoms coalesce into clusters, bound together by van der Waals forces. A short laser pulse of typically 100-fs duration is focused to an intensity of about 10^{16} W cm^{-2} into the stream of clusters. The explosion fragments are detected, and their energies are measured by time-of-flight analysis. Such studies show a quite remarkable result, namely, that very highly stripped ions (for instance, Xe^{30+} to Xe^{35+}) with extremely high energies of up to 1 MeV can be produced. For similar charge states to be produced from individual atoms by electron tunneling would require intensities about 1000 times higher than were used.

There are two mechanisms that might be responsible for this behavior. The first relies on the fact that the space-charge attraction is sufficient to hold most of the electrons within the cluster while it is heated to very high temperatures. This heating process may be made yet stronger by the collective motion of the electrons, the frequency of which decreases as the ionized cluster expands and passes through a resonant frequency defined by the frequency of the light wave (*5*). The result is that the plasma temperature rises to values similar to the interior of stars and the system explodes under the enormous hydrodynamic pressure that is generated.

The second possible mechanism is the so-called Coulomb explosion, which is known to be the mechanism by which small diatomic molecules dissociate when subjected to intense laser light. In this mechanism, a sufficient number of electrons escape from the cluster for a high positive charge to be built up from the densely packed, highly ionized atoms. In this case, the electrostatic repulsion is sufficiently high to rip the cluster apart and for the ions to be accelerated to very high kinetic energies.

The recent work of Lezius *et al.* (*1*) has sought to clarify this question of the relative importance of these two mechanisms by measuring both the kinetic energy and the charge state of the explosion fragments. The technique used was time-of-flight mass spectrometry in which a stream of clusters of a noble gas is irradiated with a short, intense laser pulse and the resulting ions are extracted electrostatically and then allowed to drift along a measured flight tube to a detector. However, in addition, a uniform magnetic deflection field was applied perpendicular to the ions' motion, resulting in a sideways deflection that depends on the ion charge and its kinetic energy. A measure-



The curve of kinetic energy. Distributions of kinetic energy *E* of xenon clusters with different kinetic energy and ion charge. Note the change in dissociation mechanism for energies around 10^4 eV. [Adapted from (*1*)]

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ment of the flight time and the deflection of the ions enables their kinetic energy and charge state (Xe^{q+}) to be determined.

These results confirmed the presence of the highly stripped, very energetic ions observed previously for clusters with about 2000 atoms (5) and also provided additional information on the acceleration mechanism. In the case of the Coulomb explosion, the energy of repulsion is expected to scale as the square of the ionic charge, whereas, in the case of hydrodynamic explosions, the energy is purely thermal and scales linearly as the ionic charge and the electron temperature. The studies by Lezius *et al.* of argon clusters that contained typically 10^5 atoms indicated that the dominant mechanism was Coulombic but that for xenon clusters that each contained about 10 times more atoms, lower charge states ($q < 6$) were Coulombic but the majority of the higher charge states ($q > 10$) were produced by a hydrodynamic expansion. These results for xenon ions of up to $q = 30$ are summarized in the figure and show the transition in mechanism as the ionic charge and the kinetic energy increase. However, the most energetic ions with energies approaching 1 MeV appear to be produced by Coulomb explosion.

By the elegant adaptation of a simple experimental technique, Lezius *et al.* have demonstrated the complexity of the processes responsible for the dissociation of clusters. Further progress will require more detailed theoretical models to be developed to elucidate further the physical mechanisms responsible.

References

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OCEANOGRAPHY

Microbial Control of Oceanic Carbon Flux: The Plot Thickens

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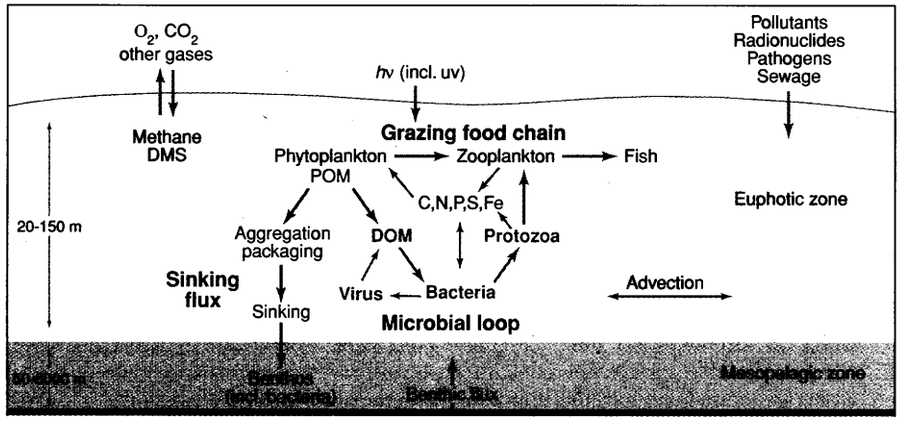
Photosynthesis fixes carbon into organic matter in the ocean. Biological forces then paint intricate flux patterns for carbon in ocean space and in time, as it flows through the food web, becomes stored in the sediments and exchanged with the atmosphere. Predicting how these carbon flux patterns might respond to global change (or to human manipulation) is a primary reason for learning more about the workings of the ocean's carbon cycle. The flux patterns are a result of intricate interactions of a diverse biota with a physically and chemically complex pool of organic matter. It now seems that things will get even more complicated before they get simpler. New fundamental findings on the roles of microbes in the fate of organic matter and, recently, on the nature of the organic matter itself (*1–4*) must be properly assimilated before we can hope to construct ecosystem models to predict the patterns of carbon flux. This impetus could lead to a powerful new synthesis.

What biological forces act on photosynthetically produced organic matter in the ocean? Historically, the paradigm has been that essentially all primary production stays within the particle phase (*5*), it is eaten by herbivores, and the fate of carbon is determined by the "grazing food chain" (see the diagram in the figure below). Little dissolved organic matter is spilled for bacteria to use. It had, therefore, been implicitly assumed to be safe to ignore bacteria, protozoa, and viruses in studying the fate of organic matter—they were too sparse and not active enough (*5*). This is now changed (*5–8*): Major fluxes of organic matter, often eclipsing the grazing food chain in quantity, move via dissolved organic matter into bacteria and the "microbial loop" (*7, 8*) (figure below). Previous methods had missed >99% of microorganisms and had grossly underestimated their metabolism. Now we know from extensive field studies that in most of the ocean, organic matter flux into bacteria is a major pathway; one-half of oceanic primary production on average is channeled via bacteria into the microbial loop (*7, 8*)—a major biological force in the ocean.

Ocean basin–scale biogeochemical studies now routinely quantify organic matter fluxes into bacteria in conjunction with other major flux pathways: grazing food chain, sinking flux, and dissolved organic matter "storage." The fraction of primary production used by bacteria (F_b) is highly variable over various time and space scales (*7–10*). The magnitudes and variability of the fluxes are large enough to cause variability in flux partitioning between competing pathways (see figure below)—the microbial loop, the grazing food chain, sinking fluxes, and storage of dissolved organic matter (*8, 9*). Fish production in the eastern Mediterranean was diminished by a dominant microbial loop ($F_b = 0.85$) (*11*). In an earlier study (*12*), the richness of the fishery in



The microbial loop: classical version. Modern view of the pelagic food web, emphasizing the microbial loop as a major path for organic matter flux. Competition between the three main flux paths—grazing food chain, microbial loop, and sinking—significantly affects oceanic carbon cycle and productivity. DOM, dissolved organic matter; DMS, dimethylsulfide.

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