Results in isotonic NaCl were qualitatively similar, except that the increase in phase c was very small, or almost absent.

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## FAST BIOCHEMICAL REACTIONS

### IN THIN FILMS INDUCED

## BY NUCLEAR FISSION FRAGMENTS

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Fission track dynamics in thin films. The passage of nuclear fission fragments through thin solid films produces a fission track characterized by a high power density ( $\sim 10^{13}$  W/cm<sup>2</sup>), a diameter of  $\sim 100$  Å, and a length of  $\sim 10~\mu m$  (1). Measurement of the kinetic energy and angular distribution of ion (2) emitted from the track give evidence for the formation of a superradiant state containing a high density of molecules simultaneously excited by the intense electromagnetic field associated with the fission fragment (3). The radiation emitted in two narrow cones at 0° and 180° to the fission fragment direction develops strong hypersonic pulses by stimulated Brillouin scattering (4). These nonlinear processes have two effects: molecules in the fission track are electronically and vibronically excited and can undergo fast chemical reactions in the excited state, and reaction products formed on the surface have a high probability of being emitted when the hypersonic pulse reaches the surface. If the reaction products are charged, they can be characterized by mass spectrometry.

Electron transfer reactions. This reaction has been studied with chlorophyll-a as a model compound. Molecular aggregates of chlorophyll produce a singlet exciton state characterized by an electron exchange current within the aggregate (5). We have verified this directly by observing the breakup of this state into (Chl-a)<sup>+</sup> and (Chl-a)<sup>-</sup> ions. We also have detected dimers, trimers, and tetramers of chlorophyll and have shown that the presence of Mg is essential for the stability of oligomer formation.

**Proton transfer reactions.** Molecules that form aggregates in the solid state, mediated by hydrogen bonding where the  $H^+$  is weakly bound (acidic), give evidence of proton charge transfer in the singlet exciton states. Reaction products emitted from those states included  $(M + H)^+$  and  $(M - H)^-$  ions. Amino acids, peptides, and small oligonucleotides exhibit this reaction.

Cationization. Molecules forming aggregates that do not produce a charge delo-

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calization do not form separated ion pairs in the excited state. The reactions that do occur involve charge exchange  $(-OH + Na^+ \rightarrow -ONa + H^+)$  and cation attachment  $(M + Li^+ \rightarrow MLi^+)$ . Alpha cyclodextrin has been a good model molecule for these studies. These processes occur for large biomolecules that do not form charge transfer bonds, either because of absence of acidic hydrogens or random orientation of molecular aggregates. Maytansine, a tumor inhibitor that contains sugar moieties and a peptide chain, is an example of a molecule that undergoes this reaction.

Summary. Fission fragments produce tracks in thin films of biological molecules that form a superradiant state because of the high excitation density. Molecular excitation is similar to picosecond laser irradiation, in that nonlinear effects are observed. The added feature is that the acoustic pulse that follows the excitation gives a means of directly identifying products of fast chemical reactions that occur.

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# NONHOMOGENOUS CHEMICAL KINETICS IN PULSED PROTON RADIOLUMINESCENCE

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The spatial distribution of absorbed energy influences the yield of chemically active species in a medium exposed to radiation. Diffusive motion of molecules rapidly destroys the initial distribution of primary species in most liquids; consequently, stroboscopic techniques must be used to observe the effects of nonhomogenous chemical kinetics directly. Pulsed radiolysis with electrons has been used extensively to prove the subnanosecond time region. However, the application of this method to other types of radiation, in particular radiations with high linear energy transfer (LET), is limited by the requirement of a large dose per pulse to achieve significant absorption by the chemical species under investigation (1). The use of fluorescence, rather than absorption, to detect the presence of chemical species circumvents this difficulty. By time-resolve emission spectroscopy, the evolution of a small population of excited states can be studied under varied radiation conditions with subnanosecond time

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