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Calculation of Heavy-Ion Tracks in Liquid Water[1]

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RUNNING HEAD: Heavy-Ion Tracks

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ABSTRACT

Detailed Monte Carlo calculations are presented of proton and alpha-particle tracks in liquid water. The computations treat the interactions of the primary particle and all secondary electrons on a statistical, event-by-event basis to simulate the initial physical changes that accompany the passage of an ion through water. Our methods for obtaining the cross sections needed for such calculations are described. Inelastic scattering probabilities (inverse mean free paths) are derived from a complex dielectric response function constructed for liquid water, based on experimental and theoretical data. Examples of partial cross sections for ionization and excitation by protons are shown. The computation of electron transport and energy loss includes exchange, elastic scattering, and a scheme for the delocalization of energy shared collectively by a large number of electrons in the condensed medium. Several examples of calculated proton and alpha-particle tracks are presented and discussed. The meaning and significance of the concept of a "track core" is briefly addressed in the light of this work. The present paper treats only the initial, physical changes produced by radiation in water (in $\sim 10^{-15}$ sec in local regions of a track). The work described here is used in calculations that we have reported in other publications on the later chemical development of charged-particle tracks.

INTRODUCTION

One of the central problems of radiation research is trying to understand and correlate the chemical and biological effects produced by radiation in terms of the physical and subsequent chemical changes that take place. Monte Carlo procedures, which can, in principle, be used to calculate statistically in complete detail these event-by-event changes, offer a powerful analytic tool for attacking this important problem. To this end, we have developed a Monte Carlo computer code, called OREC, to calculate the passage of a heavy charged particle or electron and all secondary electrons through the initial physical stage of interactions ($\sim 10^{-15}$ sec) in liquid water. At this time, the products formed by the radiation - the ionized and excited species, H_2O^+ and H_2O^*, as well as subexcitation electrons - are distributed along the path of the incident particle. The present paper will focus briefly on the physical basis for these calculations and then on some results obtained for protons and alpha particles at times $\approx 10^{-15}$ sec. We have reported elsewhere (1,2,3) on additional calculations of the subsequent chemical evolution of tracks, both for electrons and heavy ions, to times $\approx 10^{-6}$ sec. Most recently, we have made some preliminary calculations with segments of DNA in solution (4).

DERIVATIONS OF CROSS SECTIONS USED IN OREC

In this section, we summarize the procedures we have used to obtain the cross sections needed for carrying out the Monte Carlo computations with OREC. Additional details can be found in the references cited. Our approach has been to develop a complex dielectric response function $\varepsilon(\omega,q)$ for liquid water, where $\hbar\omega$ and $\hbar q$ are the energy and momentum transferred by a charged particle to the medium. By using $\varepsilon(\omega,q)$,

collective effects in the condensed phase are included a priori. The macroscopic cross section, or inverse mean free path (IMFP), for any kind of inelastic interation can be obtained directly from $-\mathrm{Im}(1/\varepsilon)$, the negative of the imaginary part of $1/\varepsilon$.

Ideally, Monte Carlo calculations can be based directly on experimental measurements. However, data for liquid water are very scant. Our starting point has been to use the optical measurements of Heller et al. (5), which give directly the function $\varepsilon(\omega,0)$ up to $\hbar\omega = 26$ eV. As described elsewhere (6,7,8), we constructed a dielectric response function for liquid water that utilizes data on the isolated H_2O molecule (gas phase), fits the liquid data of Heller et al., is properly constrained by quantum-mechanical sum rules on total oscillator strength, and gives the Bethe ridge for large ω and q.

In contrast to the vapor, energy can be shared collectively by large numbers of electrons in liquid water. An algorithm for treating initially delocalized excitations in the liquid is used, based in part on considerations given earlier (9,10). These excitations are treated as collective in character. They are assumed to exist coherently and to be centered about the track of the causative ionizing particle. De-excitation occurs through the several possible channels for single-particle and many-particle decay. In the present application, the probability of a given mode of de-excitation is taken to be proportional to the imaginary part of the partial dielectric function and to depend on the distance from the particle track. The rationale for this procedure rests on the similarity of the electric field of a long-wavelength collective excitation to the electric field of a photon with the same energy. Specifically, we apply the delocalization

algorithm to any energy loss $\leq$50 eV. Such an event is "moved" laterally away from the track by a distance picked from an appropriate distribution and not exceeding 100 Å.

Two figures will indicate the kind of information we thus derive for calculating heavy-ion transport in liquid water. We divide the electronic transitions into six specific excitation and five ionization events. Figure 1 shows, as an example, the inverse mean free paths we calculate for ionization from the $1b_1$ band and for diffuse-band excitation by a proton in liquid water as a function of its energy. IMFPs for the other electronic transitions look similar (except for the oxygen K-shell ionization, which has a threshold of 540 eV). As another example, Figure 2 shows the spectra of energy losses in a single collision of a 1-MeV proton, leading to either a $1b_1$ or a diffuse-band transition. The spectra for the other transitions and for other proton energies are similar in appearance (except for the K-shell ionization).

Some additional factors must be considered in obtaining the cross sections needed for the transport of secondary electrons (as well as for the transport of a primary electron) in water. First, we compute the angular distribution of electrons produced in inelastic collisions by a simple algorithm, giving an isotropic distribution at small values of energy transfer and a "free-electron" distribution when the energy transfer is large. Second, in contrast to heavy ions, the direction of travel of an electron can be appreciably changed by elastic scattering, especially at low energies. We compute elastic scattering on the basis of a phase-shift analysis at low energies, joined to a Thomas-Fermi model at higher energies (7). Third, also in contrast to heavy ions, exchange is taken into account explicitly.

HEAVY-ION TRACKS

Figure 3 shows some examples of proton and alpha-particle track segments calculated with OREC. Each dot represents the position of an inelastic collision by the heavy ion or a secondary electron. Thus, either an H_2O^+ or H_2O^* species is located at each dot. All secondary electrons are transported in the computations until their energies fall below the assumed threshold of 7.4 eV for electronic transitions in liquid water. The examples in Fig. 3 show, on each line, the tracks of the ions traveling at the same velocity, from left to right. The secondary-electron spectra are the same, at the same speeds, and the alpha-particle stopping power is four times that of the proton. The dots in this and later figures represent a "picture" of the track at local times $\approx 10^{-15}$ sec, before the water has begun to respond to the initial changes produced directly in it by the passage of the ion and the slowing down of its secondary electrons. We have used the detailed information obtained with OREC at this time stage as input for our calculations of the later stages of track development (1,2,3).

Various concepts used in modeling chemical and biological effects of radiation can be analyzed for tracks calculated with OREC. Consider, for example, the microscopic deposition of energy and formation of reactive species along an ion's path. The upper line in Fig. 4 shows the distribution of H_2O^+ and H_2O^* formed initially by the 1-MeV proton from Fig. 3 over a distance of 1000 nm. The clustering of events, evident in the top line, is clearly seen on a scale of 10 nm in the bottom line, which shows a 10-fold blowup of a central portion of the segment. The subsequent chemical development within such clusters, or spurs, can be analyzed from such calculations.

The two track segments from Fig. 4 are shown end-on, both on the same scale, in Fig. 5. The entire 1000 nm segment in the upper portion of the figure gives the appearance of having a central "core," surrounded by the events produced by energetic delta rays. No "core" is evident in the lower view of the 100 nm segment.

One of the reasons for developing OREC is to provide a tool for use in analyzing and trying to understand radiochemical effects in water and biological effects of radiation (1,2,3). Most recently, we have made preliminary calculations of reactions with a model for a heavy-ion track in the vicinity of a straight segment of double-stranded DNA in aqueous solution (4). Figure 6 shows a representation of a 20 nm segment of the DNA in the vicinity of a calculated track segment of a 1-MeV proton at 10^{-15} sec. The cylindrical array of dots represents the alternating bases and sugars in each strand of the DNA. The development of such tracks and their interactions with DNA can be calculated to later times, e.g., to $\sim 10^{-9}$ sec with scavengers present in the water, when chemical reactions are essentially complete (4). One can then compare different models of strand breakage with differences in the track structure of different ions.

SUMMARY

We have outlined the derivation of the cross sections we use in the computer code, OREC, for Monte Carlo calculations of the initial physical interactions of a heavy charged particle or an electron in liquid water. The information obtained with OREC has been used by us as a basis for calculating the subsequent chemical development of charged-particle tracks. In this paper, we have presented several examples of calculated track segments of protons and alpha particles.

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FIGURE CAPTIONS

Fig. 1. Inverse mean free paths (IMFP) for $1b_1$ ionization and diffuse-band excitation (multiplied by 10) by protons in liquid water as functions of proton energy.

Fig. 2. Single-collision energy-loss spectra for $1b_1$ ionization and diffuse-band excitation (multiplied by 0.1) for 1-MeV proton in liquid water. Ordinate gives probability per eV of energy loss shown by the abscissa. (Both curves have unit area.)

Fig. 3. Examples of calculated track segments of protons and alpha particles at the same speed at local times $\approx 10^{-15}$ sec. Each dot shows the location of an inelastic collision made by the heavy ion or one of its secondary electrons, resulting in the formation of an H_2O^+ or H_2O^* species at that location. The ions travel from left to right.

Fig. 4. Upper portion shows the 1000 nm segment of the 1-MeV proton from Fig. 3. Lower portion shows 10-fold blowup of a central portion of the segment, 100 nm in length.

Fig. 5. Track segments from Fig. 4, viewed end-on, both on the same scale.

Fig. 6. Segment of double stranded DNA near portion of track segment of the 1-MeV proton from Fig. 4. Dots in cylindrical array represent alternating bases and sugars in the two strands of the DNA.

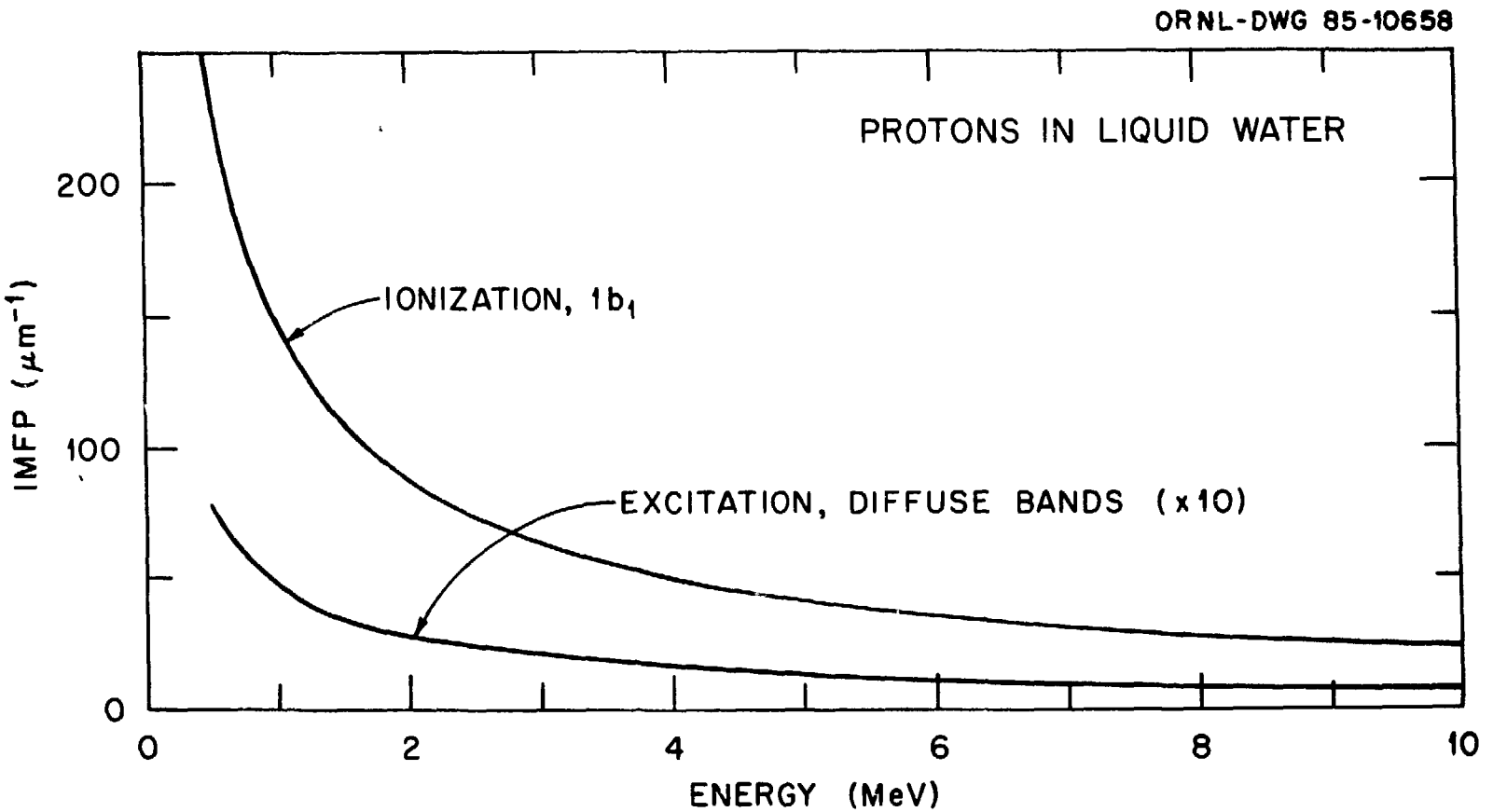
ORNL-DWG 85-10658
PROTONS IN LIQUID WATER
IONIZATION, 1b1
EXCITATION, DIFFUSE BANDS (x10)
IMFP (μm⁻¹)
200
100
0
0
2
4
6
8
10
ENERGY (MeV)

Fig. 1

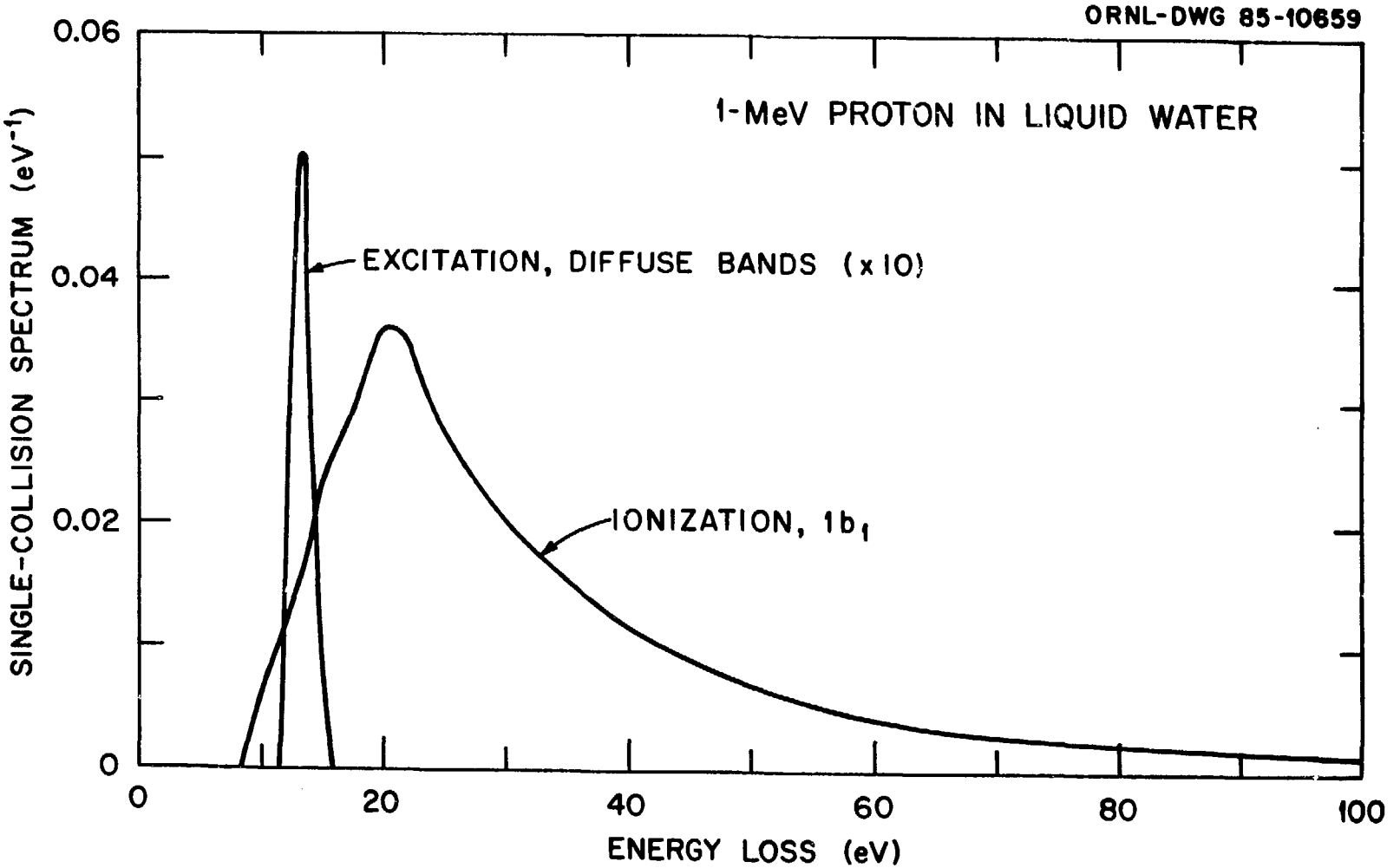
ORNL-DWG 85-10659
1-MeV PROTON IN LIQUID WATER
EXCITATION, DIFFUSE BANDS (x 10)
IONIZATION, 1b1
SINGLE-COLLISION SPECTRUM (eV-1)
ENERGY LOSS (eV)
0.06
0.04
0.02
0
0
20
40
60
80
100

Fig. 2

ORNL-DWG 85-10660

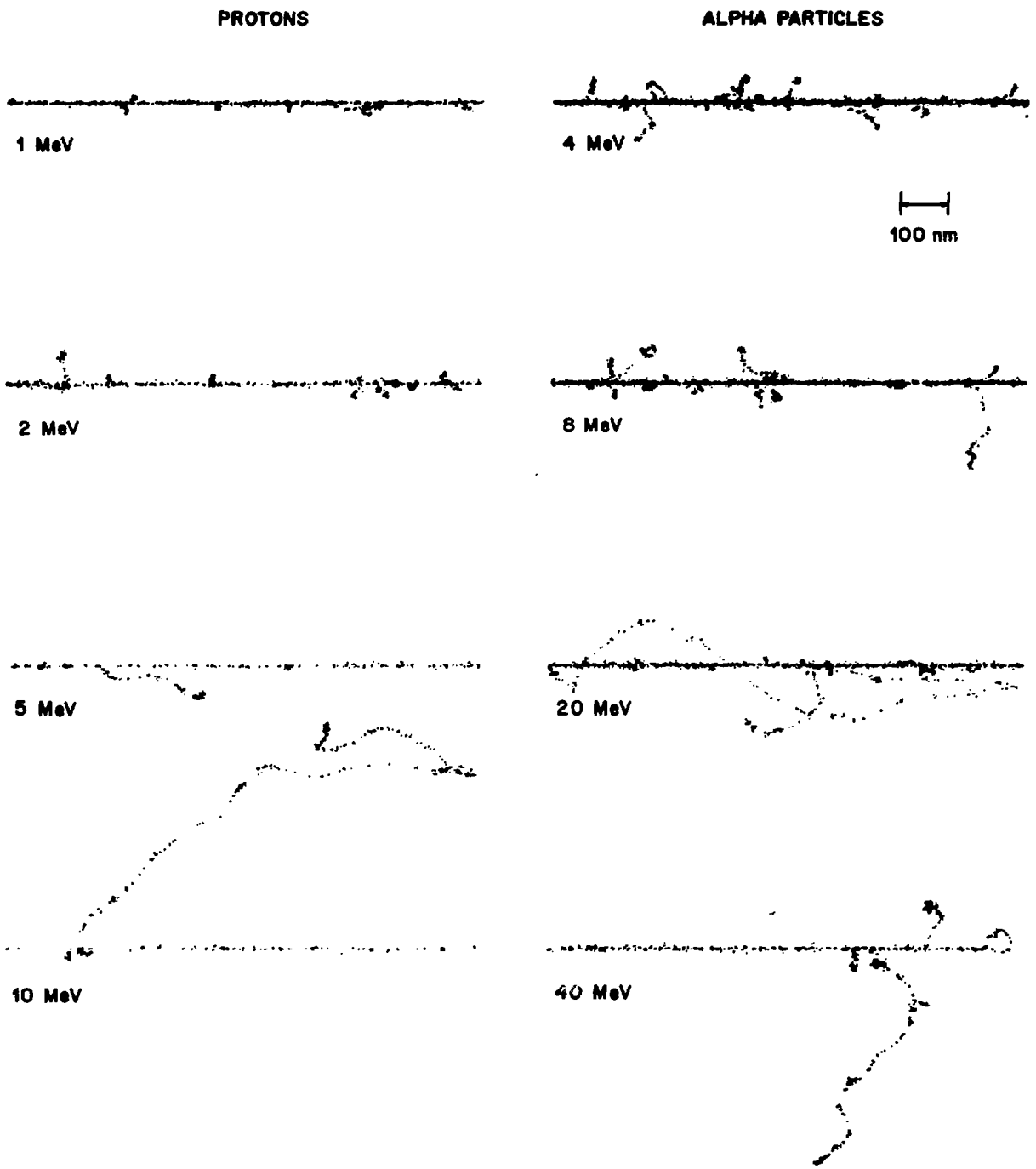

Fig. 3

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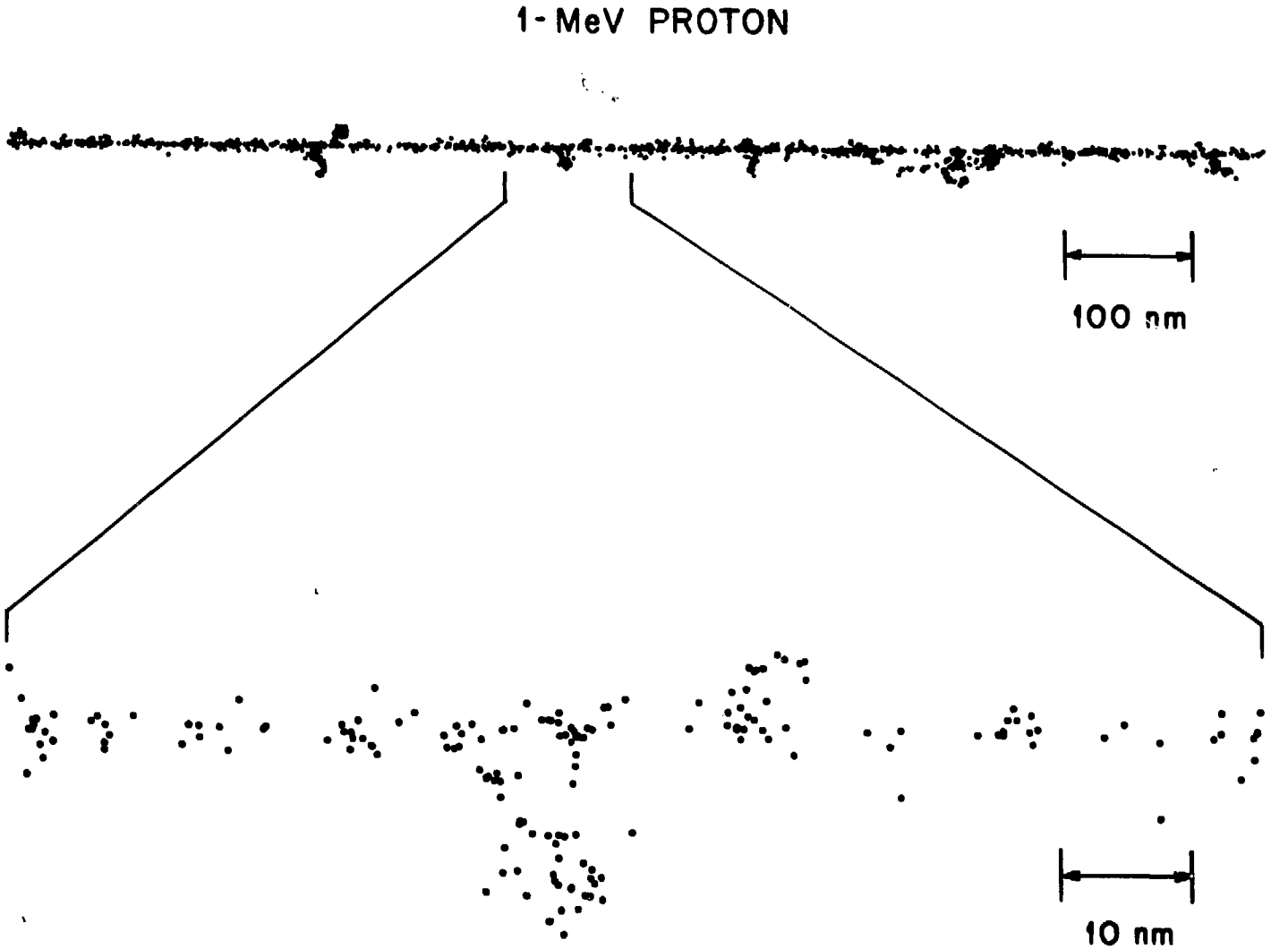

Fig. 4

ORNL-DWG 85-10666

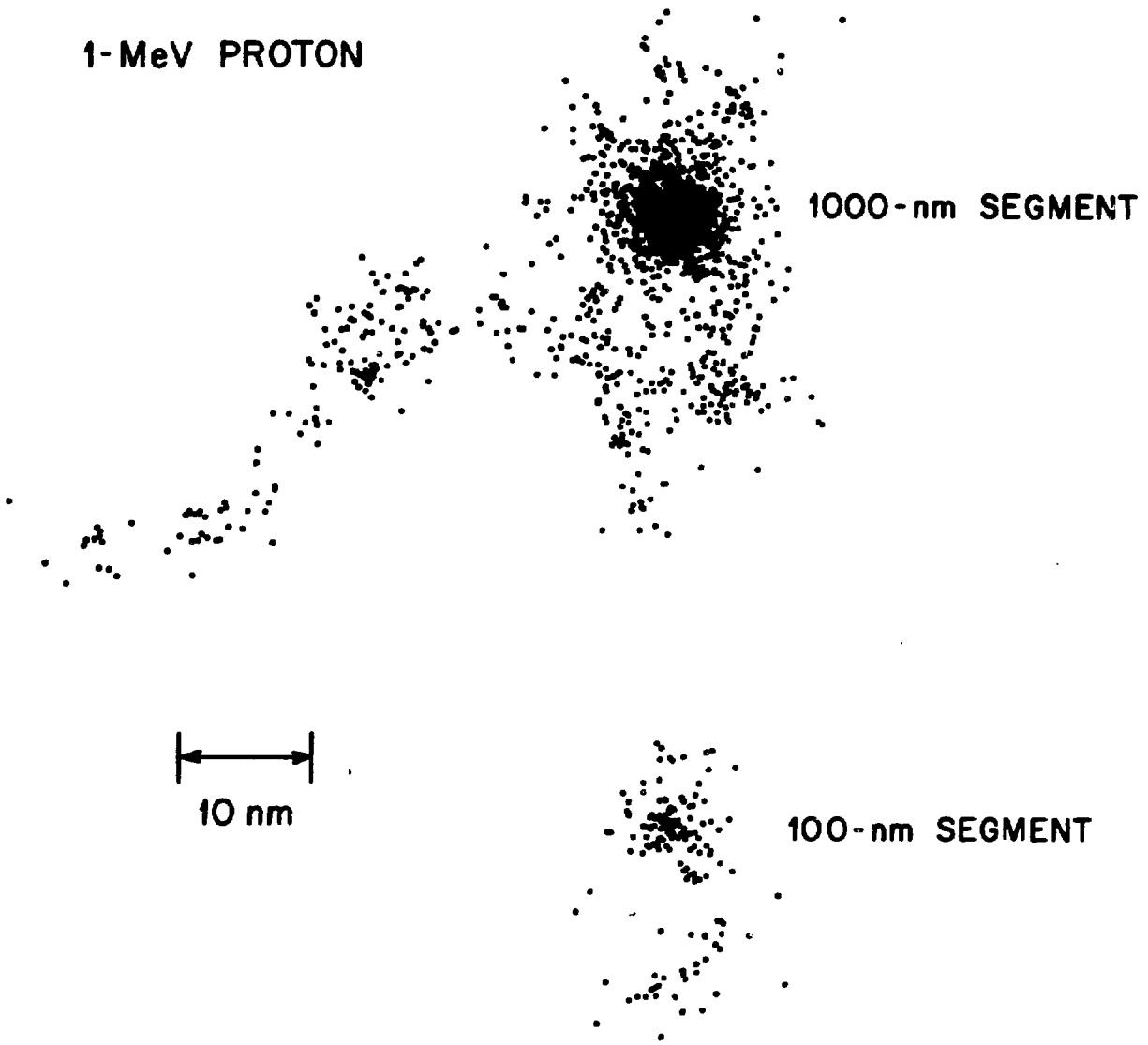

Fig. 5

ORNL-DWG 85-10662

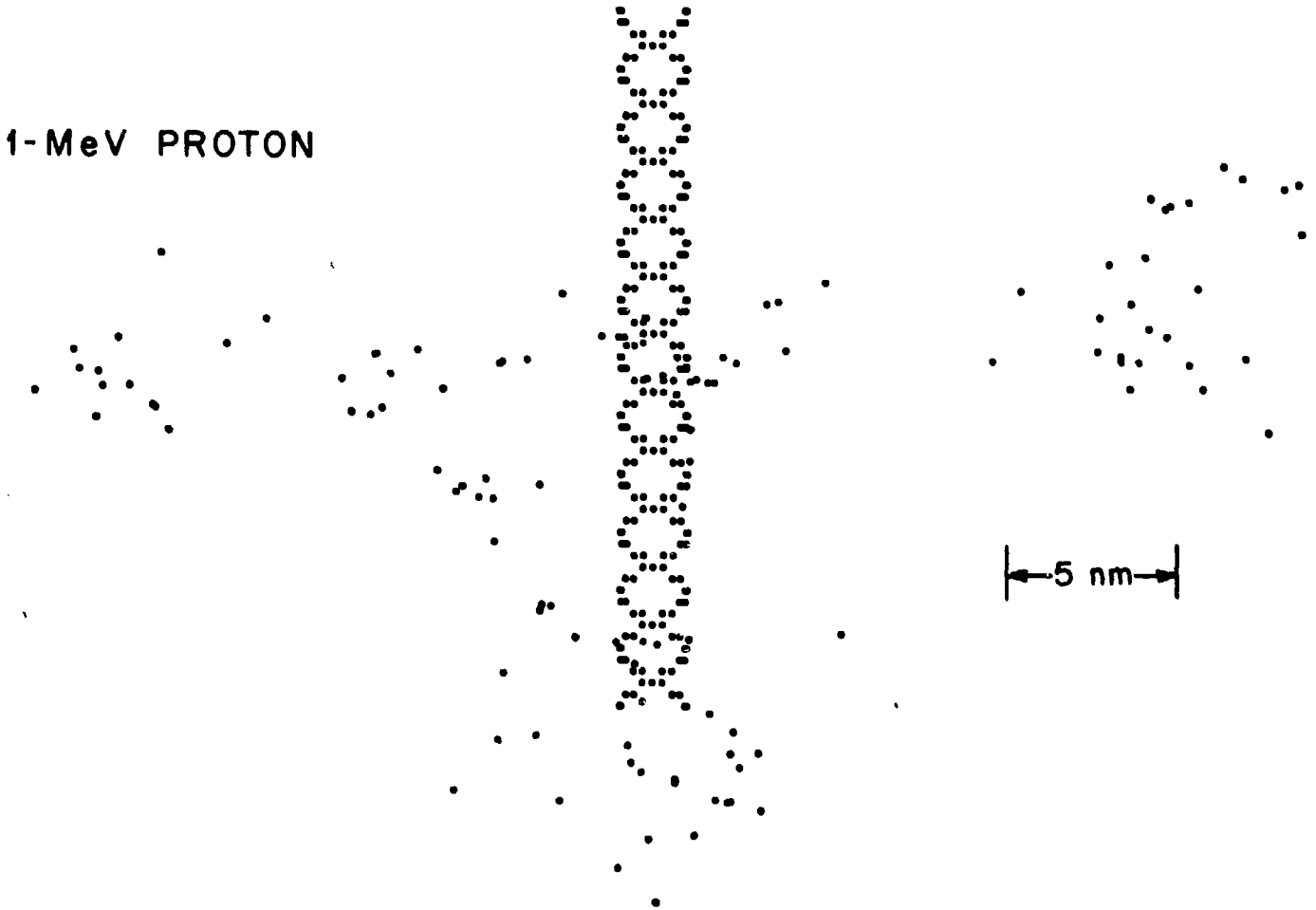

Fig. 6