Heavy Ion Passive Dosimetry With Silver Halide Single Crystals

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SUMMARY

A method of detecting radiation damage tracks due to heavy particles in large single crystals of the silver halides is described. The tracks, when made visible with simple electrical apparatus, appear similar to tracks in emulsions. The properties of the crystals, the technique of printing out the tracks, and evidence concerning the threshold energy for registering particles indicates that this method may find application in heavy ion dosimetry. The method has been found to be sensitive to stopping He nuclei and relativistic M group cosmic rays. Some impurities strongly influence the "decoration" or printout of the tracks, and the effects of these impurities are discussed.

INTRODUCTION

The hazard from the heavy particle component of space radiation has been considered for some time, and interest in this component has recently increased, particularly due to reports of "light flashes" in the closed eyes of the Apollo astronauts. The particles of charge greater than one in the galactic cosmic radiation and from solar flares require special emphasis in measurement due to a number of factors: The mode of biological damage due to the densely ionizing heavy particles is different from that due to the much more abundant $Z = 1$ particles, and the radiation hazard from the heavy particles has been predicted to be very significant (ref. 1); the heavy particles must be measured in the presence of a flux of electrons and protons which is often many orders of magnitude more abundant, causing saturation in detectors sensitive to $Z = 1$ particles; due to the high energy of the cosmic ray flux, typical spacecraft shielding is not very effective, and predictions from transport calculations on the heavy particles is not very accurate due to uncertainties in fragmentation parameters. Also in a spacecraft, each crew member encounters a different shielding situation which varies with time, requiring individual heavy ion dosimeters on extended space missions.

We will discuss a technique for detecting radiation damage tracks in large single crystals of the silver halide. This method has features which make it a potentially attractive candidate for measurements on the heavy ions.

The radiation damage tracks produced by energetic heavy particles in silver halide single crystals can be made microscopically visible, and the tracks appear superficially similar to those produced by heavy primary cosmic rays in the nuclear track emulsions. The process by which these tracks are registered and made visible has been investigated for some time (refs. 2 and 3), but the method has not been generally used for heavy charged particle identification due to past inconsistencies in the printout of the tracks in different samples of crystals. Recently the study of the effects of impurities and other factors affecting track registration has advanced to the point that reproducible results now appear feasible.

In lead-doped silver chloride, the track registration is completely insensitive to electrons and recent evidence indicates that stopping protons are not registered. Stopping He nuclei and relativistic nuclei of the CNO group have been observed in the crystals. The radiation damage tracks may be erased by annealing, and the tracks may be "decorated" or printed out in a short time with simple electronic apparatus raising the possibility of a detector that will allow the heavy particle flux to be observed for a definite time period.

The ability to decorate the heavy particle tracks in silver halides depends upon the nature of the radiation damage tracks, the properties of electrons in silver halides, and impurities in the crystals. We will briefly discuss these topics, a procedure for preparation of silver chloride crystals for track detection, and summarize the experience with radiation damage tracks in lead-doped silver chloride single crystals.

PROPERTIES AND PREPARATION OF SILVER CHLORIDE CRYSTALS

Silver chloride is an ionic conductor, transparent in the visible region, and has a density of 5.56 gm/cm^3. Its index of refraction of 2.07 (5890A) increases microscope working distances by 35% compared to emulsions. It melts at 455°C. and has a hardness of 1.3 while lead has a hardness of 1.5.

Our crystals are grown by the Bridgman method in quartz crucibles 2 cm x 2 cm x 25 cm. The starting material contains only one detectable impurity, iron in concentrations less than .07 ppm. The dopant is lead which is added to the molten AgCl in the crucibles. The melt is then treated by bubbling through it a N_2-Cl_2 mixture, the crucible sealed in its Pyrex envelope, and placed in the Bridgman furnace for growth at 1 or 2 mm/hr. Samples 8 mm thick are cut perpendicular to the crystal growth axis. These samples are polished on silicon carbide polishing papers until about a 2 mm depth is removed from each of the two largest surfaces. They are then etched with a 3% KCN solution to give transparent

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surfaces. The samples are placed on quartz plates and annealed in air at 425°C. for 12 hours, followed by cooling to room temperature at 4% per hour.

PRODUCTION OF POSITIVELY CHARGED IMPERFECTIONS BY RADIATION

The localized energy deposited by a heavy charged particle passing through the crystal may produce what Seitz and Koehler have termed "thermal spikes" (ref. 4). These spikes are regions in which some of the localized energy loss is converted into heat and the material is heated to several hundred degrees and then rapidly thermal quenched. This heating process takes place in less than 10^{-10} seconds and produces "large" concentrations of point defects which may form stable clusters during the subsequent rapid cooling. In addition, these defects can produce a disordering which causes a local volume change. This volume change plus the intense temperature gradients produce a stress field which results in plastic flow near the spike and thus forms permanent imperfections (dislocations) at distances much greater than the radius of the molten core of the spike.

Another type of spike concept has been formulated by Brinkman (ref. 5). He proposed that since the time of the molten spike is greater than the mechanical relaxation time, there is sufficient strain energy, released after density fluctuations have relaxed, to raise the temperature even higher and thus extend the period of existance of the liquid state. This temperature extension produces turbulent motion so that most of the atoms will occupy new lattice sites. Such a region which has undergone melting and resolidification is a "displacement spike."

Regardless of which model might best describe the processes involved in radiation effects in silver chloride crystals, the particle's path will be surrounded by a core of positively charged clusters of point imperfections and arrays of line dislocations which are stable at room temperature.

DECORATION OF TRACKS

Figure 1 shows the apparatus used in the laboratory for decorating tracks. The crystal is placed between blocking electrodes (E) on quartz plates (Q) and forms the major dielectric of a capacitor. The top electrode is a quartz plate covered with an ultraviolet transmitting electrically conducting thin film. That film is connected to the positive terminal of a high voltage supply (2,000 volts) which charges the pulse-forming network (P.F.N.). Above the ultraviolet transmitting electrode is a mercury flash lamp. This flash lamp is connected in series with the network and the plate of a hydrogen thyratron. The sequence of events is:

1. The high voltage supply charges the pulse-forming network and produces an external electric field on the crystal. The crystal polarizes, resulting in the surface toward the transparent electrode having negative surfaces charges.
2. When the charging cycle of about 1,000 microseconds is completed, a trigger pulse is applied to the thyratron grid discharging the network through the lamp and removing the external field on the crystal.
3. The lamp gives a 10 microsecond light pulse which forms photoelectrons at the crystal surface. These photoelectrons are then forced towards the opposite surfaces by the decaying internal polarization field.
4. Some of the electrons are trapped at the positively charged imperfections produced by the ionizing particle. These trapped electrons may then capture an interstitial silver ion, resulting in formation of silver atoms.
5. The newly formed silver atoms can capture other electrons so the process of silver atom formation continues until the particle's path is delineated by microscopic silver grains. This process continues until the silver grain size is limited by the mechanical stress in the crystal.

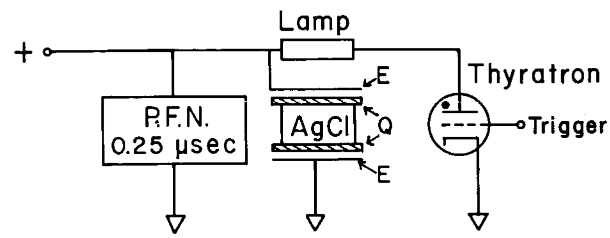

Fig. 1. Apparatus For Decorating Tracks in Silver Chloride Crystals

Since the silver grains reach a saturation size, it is possible to decorate tracks which occur after the initial decoration without affecting the original tracks.

In a crystal 5 mm thick, tracks of heavy primary cosmic rays can be made visible in 2 hours at a pulse frequency of 10^3/sec with the laboratory apparatus described above. In principle, the pulse repetition rate could be increased to around 10^5/second (limited by the electron lifetime) and the crystals decorated to saturation in a few minutes.

OBSERVATION OF TRACKS

The experience with radiation damage tracks in silver halide crystals has been too limited to allow an accurate determination of the minimum LET observable. In addition, it is yet uncertain to what extent impurities may influence the threshold for observable track decoration. We will summarize here some of the data available for silver chloride crystals.

Tracks have been observed at the surface of AgCl crystals resulting from alpha particle exposures (ref. 6). The tracks have consistent ranges of 16μ corresponding to the 5.3 MeV polonium alpha.

Tracks in the interior of lead-doped silver chloride crystals have been generated by exposing them to high energy proton and pion beams and generating "stars," and by some limited exposures of the crystals to the primary cosmic rays on balloons (ref. 2).

Figure 2 is a photograph of stars produced by 1.8 GeV/c π^- mesons which indicates tracks due to evaporation alpha particles and other heavier fragments.

Figure 3 shows the distribution of the ranges of all visible tracks from 50 stars produced by a 1.8 GeV/c π^- beam. The tracks can be attributed to He nuclei and other heavier fragments (silver evaporation He^4 nuclei of 16 MeV would have a range of 92μ). Evaporation protons from silver (8 MeV) would have a range of 290μ, and a peak is clearly not observed there, indicating that the crystals are not sensitive to protons of 8 MeV and greater.

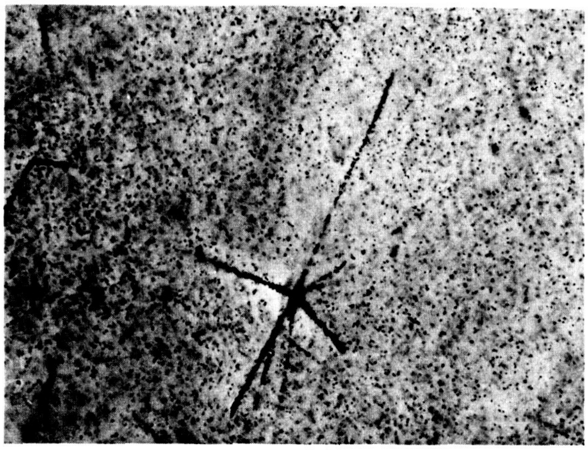

Fig. 2. A Star Produced By 1.8 GeV/c π^- Mesons.

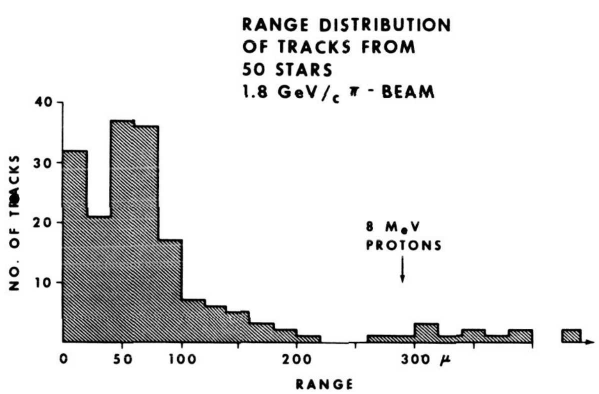

Fig. 3. Range Distribution of Tracks From 50 Stars 1.8 GeV/c π^- Mesons.

Recently we have exposed some samples of AgCl crystals dopes with 4 ppm lead to a stopping proton beam. The 70 MeV beam entered the side of a 0.5 cm x 2 cm x 2 cm crystal and stopped within the crystal. Control samples were exposed to 158 MeV protons to generate stars and check for consistency. Although tracks consistent with alpha ranges were observed, no tracks of the stopping protons were found.

Small crystals (2 cm x 2 cm) with nuclear track emulsions attached have been exposed to the heavy primary cosmic flux on balloon flights in Texas. Due to the small area-time factor, the number of observed events has been small. Figure 4 shows the tracks of two heavy primary cosmic rays in G 5 emulsions and tracks of the same particles in silver chloride crystals. The clearness of the background and the lack of delta rays in the crystals are obvious. It was also observed that the track density in the crystals varied consistently with the track width in the emulsions. In these exposures, tracks due to relativistic CNO group nuclei were observed, but no minimum ionizing He nuclei were seen.

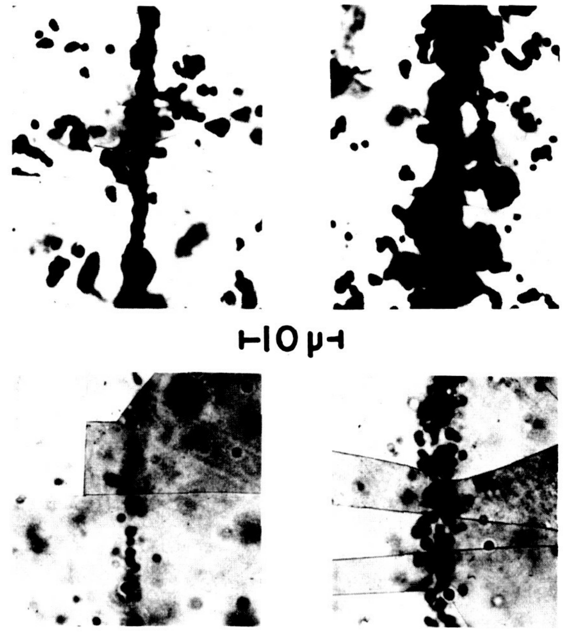

Fig. 4. Two Different Relativistic Primary Cosmic Ray Particles as Seen in G 5 Emulsions (Top) and Silver Chloride Crystals (Bottom) (ref. 8).

IMPURITY EFFECTS

It has been found that the decoration of tracks and the visible background is very sensitive to trace impurities in the crystals. For example, in crystals containing one part per million (ppm) of either iron or copper, inherent line dislocations are made visible by sweeping in electrons, but no radiation tracks are made visible. On the other hand, tracks can be made visible in crystals containing 4 ppm lead, but no dislocations are observed. One feasible explanation of this difference in impurity effect is as follows.

Before exposure to radiation, samples are cut from the crystals and annealed at 425°C. to decrease the strain introduced during growth and cutting. Since this temperature is only 30°C. below the melting point, the impurities are relatively mobile. As the samples are cooled to room temperature, the impurities settle preferentially at dislocations. So, when the samples are at room temperature, the impurities are relatively immobile and remain at the dislocations. With an impurity concentration of 1 ppm of either copper or iron, the dislocations have a positive charge so they trap electrons, leaving few if any electrons for tracks.

On the other hand, lead impurity behaves differently than iron and copper. When lead is present in about 4 ppm, it too prefers to settle at dislocations but with this difference: through complex formation, the presence of the lead leaves the dislocation regions with a negative charge. The result is that few electrons are captured by the dislocations, leaving most electrons to be captured by the positively charged imperfections produced by the ionizing particle.

While a few ppm lead are required for good track decoration, it should not exceed its room temperature soluability limit of about 8 ppm. All lead greater than this concentration merely precipitates out of solution and increases the background as shown in Fig. 5.

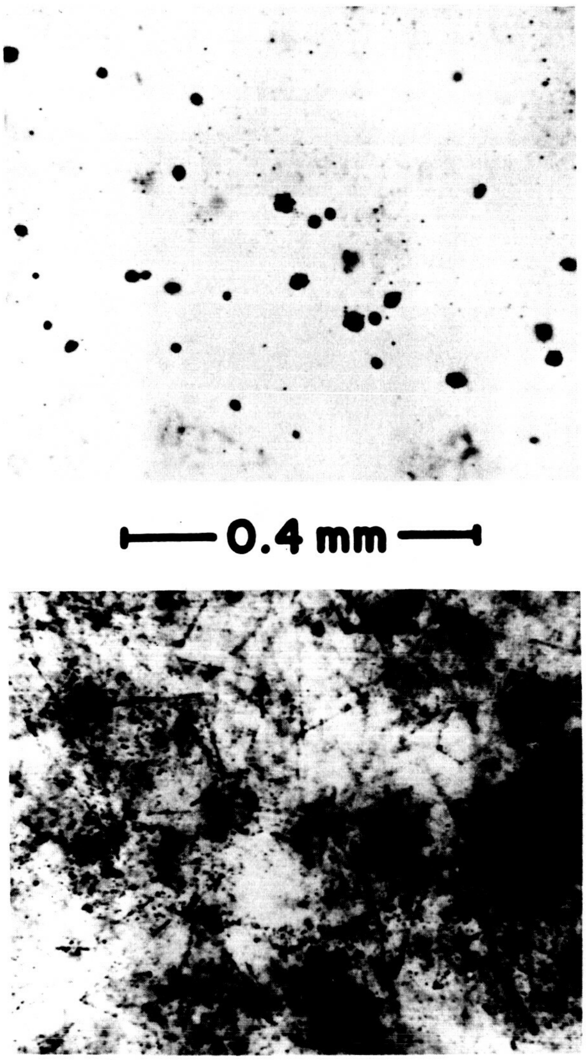

Fig. 5. Effect of Lead Concentration. Top photograph shows crystal containing 50 ppm lead after exposure to 1.5 GeV protons and decoration. Bottom photograph is crystal containing 15 ppm lead after a similar exposure and decoration.

ERASING PARTICLE TRACKS

The silver specks delineating the particle paths can be redissolved by heating the crystal to 300°C. to 400°C. Heating the crystal will cause the silver specks to dissolve and anneal out the imperfections produced by the ionizing particle. Thus with proper annealing, the crystals could be erased and reused for particle registration.

TRIGGERING CRYSTALS

Schopper (ref. 7) has proposed that it might be possible to trigger a crystal to particles having certain predetermined properties such as velocity and charge. His proposal is based on Henig's (ref. 8) studies of cadmium-doped crystals in which tracks could be made visible only immediately after exposure to particles. We are in agreement with Henig's observation since we have found that there is a cadmium ion complex relatively mobile at room temperature which would rapidly neutralize positively charged imperfections. The proposal of Schopper should be pursued with crystals containing various concentrations of cadmium and other impurities that produce a negative ion complex which is mobile at room temperature.

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