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## Fossil Tracks in Doped Muscovite Crystals

F M Russell

October 1992

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### Abstract

Positively charged particles can perturb the lattice in crystals of muscovite to precipitate impurities as a separate phase to record the particle tracks. It is well known that intense neutral lattice perturbations, such as shocks, also can trigger phase transitions. It is shown that many previously unexplained lines in muscovite crystals can be interpreted as the tracks of intense pulse-like disturbances of the lattice caused by coupling of energy to the lattice from energetic heavy ions. These intense pulse-like disturbances exhibit properties similar to those of solitary waves or some kinds of soliton.

The potential importance of these pulse-like lattice disturbances arises because, in principle, they could transport significant amounts of energy, in the 1eV to 30eV range, through a crystal in packets which are very localized in space. This energy range spans that for the creation of many types of static crystal defects found in radiation damage in solids.

In perfect or nearly perfect regions in crystals these pulse-like disturbances propagate without loss of energy or dispersion. However, in damaged or disturbed regions of the lattice, such as at grain boundaries or dislocations, the conditions for solitary wave or soliton propagation no longer exist and the pulse-like disturbance can lose energy to the lattice.



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## Fossil tracks in doped muscovite crystals.

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### Abstract.

Positively charged particles can perturb the lattice in crystals of muscovite to precipitate impurities as a separate phase to record the particle tracks. It is well known that intense neutral lattice perturbations, such as shocks, also can trigger phase transitions. It is shown that many previously unexplained lines in muscovite crystals can be interpreted as the tracks of intense neutral pulse-like disturbances of the lattice caused by coupling of energy to the lattice from energetic heavy ions. These intense pulse-like disturbances exhibit properties similar to those of solitary waves or some kinds of soliton.

### Introduction.

Natural crystals of muscovite mica often exhibit internal defects in the form of a hatchwork of linear markings or lines which create complex patterns. These lines consist mainly of thin ribbons of magnetite ( $\text{Fe}_3\text{O}_4$ ), lying in the (001) plane of easy cleavage, with lengths up to the dimensions of the crystal. It is evident from even a cursory examination that these lines do not occur at random and so must have a causal origin which creates nucleation sites leading to precipitation of impurity atoms from the muscovite to form the lines. Some of these lines exhibit unusual and unique properties which defy explanation in terms of known types of crystal defects but which can be understood in detail if they are interpreted as the fossil tracks of charged particles.[1,2] Although muscovite mica is a common mineral its PT stability curve shows that for large crystals to form a pressure in excess of 1500 bar at 900K is needed. A long growing time for large crystals, high pressure and expensive equipment have so far precluded experimental verification of lepton track recording in muscovite. However, the track hypothesis has been used to predict results that have subsequently been confirmed in the laboratory. The discovery that some of these lines can be explained as 'fossil tracks of charged leptons has prompted studies of the track recording and decorating processes because they provide a window for the study of events occurring at the atomic level. Of special interest is the possible origin of the majority of the lines which can not be explained in terms of known types of charged particles but which, presumably, also are the decorated records of events that occurred at the atomic level.

### Track recording and decorating processes.

Usually, tracks are decorated with magnetite,  $\text{Fe}_3\text{O}_4$ , derived from an impurity of iron initially held in solid solution. The extent of decoration is proportional to the rate of energy loss  $dE/dx_1$  by ionization.[3] For this process to occur  $dE/dx_1$  must exceed on average about 10KeV/cm or 1eV per 1000 unit cell lengths along a track. A second type of decoration involving a phase change was predicted from the track hypothesis and occurs in crystals containing an excess of calcium, which precipitates as epidote.[4] The extent of decoration with epidote is independent of  $dE/dx_1$  in that the parts of positron tracks decorated with epidote have uniform width, although the actual width varies from track to track. This leaves the moving positively charged particles as the most probable cause for initiating the phase transition to epidote. It is already known that energetic ions can influence phase transitions in metallic solids.[5] Since ionization is quickly neutralized in a metal these results imply that it is the presence of positive charge, both as a moving charged particle and as the net positive part of ionization in an insulator, which enables a transition to occur leading to precipitation of a new phase. It should be noted that in this recording process the track is delineated by release from the lattice of the exothermic energy associated with the phase transition instead of by deposition of energy to form a track.

Both of these recording processes are inhibited by lattice disorder such as dislocations and so do not occur near fractures. This is evident in long tracks caused by high energy muons which span a fracture as a gap in the track, the gap in recording and decorating extending across the damage zone surrounding the fracture. However, significant structural defects such as grain boundaries and dislocations can act as nucleation sites for precipitation of impurities, the dislocations having the appearance of curvilinear arrays of decoration spots. The general appearance of these various kinds of decorated lines is illustrated in Figure 1.

These recording processes can be understood in terms of a spinodel decomposition model: impurities make the muscovite polyphasic and metastable. After growth muscovite crystals cool until they become saturated or even super-saturated with dissolved impurities but since phase transitions to a lower energy state do not occur spontaneously they must be inhibited by a potential energy barrier. Lowering of this barrier locally enables the exothermic phase transitions to occur.

Clearly, the recording process in muscovite is **very** sensitive to perturbations of the lattice since it can respond to even a single positron moving at near relativistic speed to create a decorated track. It is likely that other types of perturbation of the lattice also could alter the height of the potential energy barrier  $V$  and so perhaps enable a transition to occur quickly. The magnitude of  $V$  can be estimated from the fact that, for example, the transition to epidote is inhibited even when the muscovite is cold but can occur at 900K in the presence of a positive charge, giving  $V > \approx 0.2\text{eV}$ . Indeed, it is well known that phase transitions can be induced by intense shocks in other metastable materials, for example, as seen in some meteoric impact material. It is plausible that in doped muscovite crystals phase transitions could be initiated by large relative movements of the lattice atoms alone in absence of positive charges. The natural

speed of a large amplitude lattice disturbance would be much closer to that of sound than to that of a high energy lepton so the dwell-time of a disturbance in crossing a unit cell would be increased greatly, thereby increasing the probability for the transition to be initiated. *It is proposed here that some of the unexplained (U) lines in mica crystals might result from the passage of intense but highly localized Pulse-like Lattice Disturbances (PLDs) resulting from coupling of energy to the lattice by energetic particles such as fission fragments or  $\alpha$ -recoil nuclei.* Since the unexplained U-lines are very long relative to the atomic spacing any PLD must propagate with minimum or no dispersion, have a low probability for scattering by small random lattice defects and propagate without loss of energy between scatterings. Also, as the recording and decorating of tracks only occurs at high temperatures the PLDs must be stable against small random perturbations of the lattice caused by thermal motion of the nuclei. The PLDs of interest here have large amplitudes of motion relative to the interatomic spacing so that non-linear effects can be expected. Such behaviour and properties suggests a PLD might be some kind of solitary wave or soliton.

#### **Solitary wave or soliton models.**

As the U-lines occur only in the (001) basal plane and have similarities with decorated positron tracks, which are known to propagate in the monatomic potassium (K) sheets, it is possible that the proposed PLD also propagate in the quasi-two dimensional K-sheets. PLDs might consist of large transverse or longitudinal relative motions of the nuclei in the K-sheets. A simplistic classical case is illustrated in Figure 2 in which the atoms K1 and K2 provide a focussing action on the atom K3 which can have a large amplitude of motion in the direction between the atoms K1, K2. This simple model suggests that PLDs might propagate along chains of nuclei in the K-sheets in the [100] direction or at multiples of  $30^\circ$  from that direction. In fact, the U-lines are found to lie exactly in these directions. A PLD propagating along a chain would have one space and time (1+1) dimensions, similar to a Toda lattice soliton.[6] Alternatively, the disturbance might start in two space and time (2+1) dimensions, suggesting a description in terms of the Kadomtsev-Petviashvili [7] or Zakharov-Manakov [8] equations.

A problem for such descriptions is that rapid lateral spreading of the disturbance by coupling to nuclei in adjacent chains in the K-sheets could occur, leading to spreading of the initial energy over a wide wave-front.[9] At present it is not known how to deduce anything specific about the internal details of the proposed PLDs from the observable properties of the decorated lines despite there being clear differences in the detailed nature of the decoration on PLD tracks relative to charged particle tracks; see later, in Figure 7. For this reason it is proposed to study the propagation of different types of large amplitude, and therefore non-linear, PLDs in muscovite by molecular dynamic computer simulation methods.[10] It is hoped these studies will assist in identifying which type of solitary wave or soliton solution best describes the observed and computed results or point to some other solution. It is interesting that a recent Monte Carlo computational study of atomic cascades in Copper crystals has shown that particle momentum directions associated with energy

transport through the lattice depend only on the crystallography and that this directionality is not destroyed by thermal motion even at high temperatures.[11]

#### Decay and propagation of PLDs.

Let us assume that the majority of the U-lines are the tracks of large-amplitude PLDs propagating in the K-sheets and that these PLDs are created during the slowing down of energetic particles passing through the mica. It is to be expected that as a result of scattering at crystal defects the PLDs will eventually degrade into high energy phonons. Consequently, the energy  $E_{PLD}$  and momentum  $p_{PLD}$  of a PLD can be found, to first order, by summing over the energies and momenta of the residual phonons:

$$\begin{aligned} E_{PLD} &\approx \sum h\nu_n & 1 \\ p_{PLD} &= \sum h/\lambda_n & 2 \end{aligned}$$

where  $\nu_n$  and  $\lambda_n$  are the frequencies and wavelengths of the  $n$  phonons comprising the residue from the degraded PLD. In a large tree-like pattern of PLD tracks upwards of 100 branches can be counted before the secondary PLDs cease to produce further decorated tracks. Hence, the energy of a PLD can be found if the threshold energy of a PLD to just produce a decorated track can be estimated. This can be done as follows. Since mica crystals are not filled with small decorated spots from multiple phonon pile-up it is reasonable to suppose that the threshold lies well above the disturbance caused by pile-up of more than 10 energetic phonons. Assuming the maximum energy and momentum of a phonon at a prevailing temperature of about 900K is of order  $5 \times 10^{-3} \text{eV}$  and  $2 \times 10^{-2} \text{mDc}$ , then energetic PLDs could have energies and momenta in the region of 5eV and 20mDc, respectively.

By the de Broglie postulate any entity which has momentum  $p$  also has an associated wavelength  $\lambda = h/p$ . For a phonon of wave vector  $k$  the (crystal) momentum  $p(\text{ph})$  is  $h(k/2\pi)$  and so the de Broglie  $\lambda$  is  $2\pi/k$ , yielding minimum wavelengths in the same range as the atomic spacing. Similarly, for a PLD of (crystal) momentum  $p_{PLD}$  the associated de Broglie wavelength  $\lambda_{PLD}$  is  $h/p_{PLD}$  and from equation 2

$$\lambda_{PLD} = 1/\sum (1/\lambda_n) \quad 3$$

indicating that  $\lambda_{PLD}$  can be a few orders of magnitude shorter than the atomic spacing. For the energetic PLD considered above  $\lambda_{PLD}$  is of order  $10^{-2} \text{\AA}$  or less. Such entities should be diffraction scattered strongly by the lattice to give preferred propagation in specific crystallographic directions. The smallness of  $\lambda_{PLD}$  suggests that a PLD should be treated more as a quantum mechanical entity than as a classical mechanical disturbance propagating in the lattice. This might help resolve the problem of lateral spreading, as the PLD would show particle-like as opposed to wave-like behaviour. Alternatively, PLDs might experience the equivalent of ballistic phonon focussing.[12]

The above estimate for the momentum of a primary PLD is compared in Figure 3 with the maximum momenta available from several possible sources. The comparison is made as a function of the speeds of the particles  $v(p)$ , PLDs  $v(\text{PLD})$  and phonons  $v(\text{ph})$ . The transfer of energy and momentum from a

particle to the lattice to create a PLD is expected to be most efficient when the speed of the particle is similar to that of the PLD, as this maximises the interaction time and encourages resonant coupling. To first order, the interaction time will decrease as  $v(\text{PLD})/v(p)$  as  $v(p)$  tends to  $c$ . As  $v(p)$  decreases below  $v(\text{PLD})$  both the interaction time and the momentum available also decrease. Figure 3 shows that the most probable origins for the PLDs are fragments from spontaneous fission, nuclear recoils from  $\alpha$ -emission, high energy muons and, least likely, nuclear recoils from the decay of  $K^{40}$  nuclei. No PLD tracks would be expected to arise from the positrons and electrons emitted in the  $K^{40}$  decays, a result which is consistent with the observed occurrence of the U-lines.

### Experimental results.

Having outlined the hypothesis that PLDs are the cause of many of the U-lines observed in muscovite a compendium of experimental results is now assembled against which this or any other hypotheses can be tested. For example, it has been suggested that all of the lines are simply decorated dislocations.[13] It is explained below why the results presented here are wholly inconsistent with a dislocation origin.

By progressive splitting of mica crystals the complex arrays of overlapping lines can be resolved into contiguous patterns. After eliminating all lines attributable to known types of charged particles, all isolated or irregular markings and the arrays of spots associated with dislocations, various measurements have been made on the remaining U-lines. No evidence has been found for coordinated interaction between U-lines in different (001) planes, that is, the patterns are 2 and not 3-dimensional.

The frequency of occurrence of U-lines varies between crystals. Typically, it is about  $500/\text{cm}^2$  measured in a plane perpendicular to the (001) plane. Assuming the duration of the sensitive time for recording the U-lines is similar to that for charged particles then the U-lines are produced at an average rate of about  $0.03/\text{cm}^2$  per year. When the U-lines have been fully resolved by splitting, they typically range from less frequent strongly decorated central or primary lines from which secondary lines of weaker intensity branch away to more frequent single isolated lines.

The most noticeable feature of U-lines is that they are mostly parallel to crystallographic directions of low Miller index in the (001) plane, as shown in Figure 4. Where a secondary U-line joins a primary U-line there is no measurable change in direction of the primary U-line, the same being true for higher order branches. These two features indicate that the entities responsible for the U-lines must interact strongly with the lattice.

The length distribution  $L_p$  of the primary U-lines, Figure 5, shows that the majority are limited by the size of the crystal in the (001) plane, with most primary lines extending to a crystal boundary or edge, giving lines of 500mm or more in length in large crystals. The length distribution  $L_s$  of branch U-lines, however, is quite different, as shown also in Figure 5. The most probable length is about 20 mm.

Within isolated tree-patterns the average U-line length decreases as the branch order increases. The secondary U-lines occur randomly along primary and higher order U-lines. The frequency distribution of the length intervals  $L_1$  between secondary U-lines along primary U-lines, Figure 6, indicates an average interval of  $14 \pm 2$  mm. Occasionally a secondary U-line crosses another U-line in the same tree-like pattern. These results suggest that the mean free path (MFP) between scatterings decreases as a PLD loses energy by inelastic scattering. This is an important result because PLDs are expected to degrade eventually to phonons which have short MFPs at high temperatures.

The extent of lateral decoration of a primary U-line is similar to that of a positron track in the low energy region where the speed of the positron is not vastly different from that of sound. Higher order U-lines show progressively weaker decoration. The extent of lateral decoration of any U-line on average remains approximately constant over the whole line length but often shows a periodic modulation on a smaller scale. The frequency distribution of spacings between decoration maxima peaks at about 3mm, and does not vary greatly from one crystal to another nor within a single crystal. Closer examination of the decoration shows that there is often a systematic difference in appearance between the decoration on charged particle tracks and that on U-lines. As seen in Figure 7, the decoration on U-lines seems to relate more closely with the underlying crystal structure.

Some primary U-lines are found to be contiguous with high energy muon tracks, occasionally with electron shower tracks but never with the tracks of positrons from  $K^{40}$ . Moreover, chemical etching of the ends of primary U-lines within a crystal sometimes reveals fission fragment tracks in close spatial association with the U-lines. The probability for U-lines to be contiguous with particle tracks by chance is very small and pinning by impurity atoms would prevent them becoming contiguous by migration through the lattice. These findings are consistent with the primary U-lines being generated as the result of coupling energy to the lattice by energetic charged particles of momentum of order  $100m_0c$  or more, where  $m_0$  is the electron mass and  $c$  is the speed of light. It is not sufficient for the charged particles just to have high energy, they must also carry a substantial amount of momentum to create U-lines.

A small fraction ( $\approx 10^{-3}$ ) of U-lines show a sudden large change in direction  $\theta$  in the (001) plane away from a direction of low Miller index, often in association with a branch U-line, as shown in Figure 8. The initial large deflections lie apparently at random in the range  $0 < \theta < \pi/6$ . These deflected U-lines show multiple small changes in direction, analogous to multiple-scattering, until they regain a low-index direction which they follow thereafter. The extent of decoration of a primary U-line is not significantly changed by such deflections. The frequency distribution of the angular changes along these scattered U-lines shows that small deviations are most probable.

#### Interpretation of results.

As noted above, it has been suggested that all the lines, or at least the U-lines, might be decorated dislocations. As far as is known, no evidence

has been offered to support this suggestion and there is much evidence against it. Firstly, dislocations can not start or stop inside a single crystal but must form loops or end on a boundary or edge, in complete contrast to the measured properties of the lines and tracks. Secondly, the high concentration of impurities in these muscovite crystals would pin any dislocations and make them immobile, so they could not have been formed elsewhere and then migrated to the observed positions of the lines. Thirdly, if the lines were decorated dislocations then decoration would have occurred on all dislocations present and not just on those formed during the sensitive time for recording and decoration. The measured value of  $\approx 500/\text{cm}^2$  for the average density of U-lines would imply that the density of dislocations in natural crystals of muscovite containing substantial amounts of impurities is many orders of magnitude less than achieved in carefully grown pure crystals, which is most unlikely. Fourthly, major crystal defects such as fractures often are associated with arrays of decorated spots forming curvilinear lines which do present features characteristic of dislocations. These arrays of spots do not join on to the U-lines or particle tracks. Fifthly, there is no known type of dislocation or other type of large scale lattice defect that mimics the unique features of Coulomb scattering (Rutherford and multiple), lattice diffraction scattering and ionization-dependent decoration that have been observed by measurements on mica crystals. Clearly, a dislocation origin for the lines is incompatible with the evidence.

However, the U-lines can be understood in some detail if they are assumed to be the tracks of intense PLDs which initiate a phase transition as they propagate through the lattice to form tracks. It might be supposed that the PLDs consist of rare superpositions of many high energy phonons but this origin is eliminated by the observation that some PLD tracks show large angular deflections, as seen in Figure 8, which would require some unknown kind of cooperative action between many phonons to cause their identical scattering. Nevertheless, if PLDs are interpreted as solitary waves or solitons then their stability may well depend on the amplitude of the disturbance. In that case, it might be possible for a random superposition of phonons to interact anharmonically in a small volume to create an occasional PLD. Irrespective of how a PLD is created it would be expected to degrade eventually into phonons and very energetic PLDs also may cause static lattice defects such as atomic displacements if it leads to a lower energy state for the lattice.

There is also the possibility that a PLD could trigger the release of energy stored in a strained lattice leading to a structural rearrangement but without a compositional change or a combination of structural and compositional changes.

The potential importance of PLDs arises because, in principle, they could transport significant amounts of energy, in the 1eV to 30eV range, through a crystal in packets which are very localized in space. This energy range spans that for the creation of many types of static crystal defects found in radiation damage to solids and in the growth of micro-cracks.

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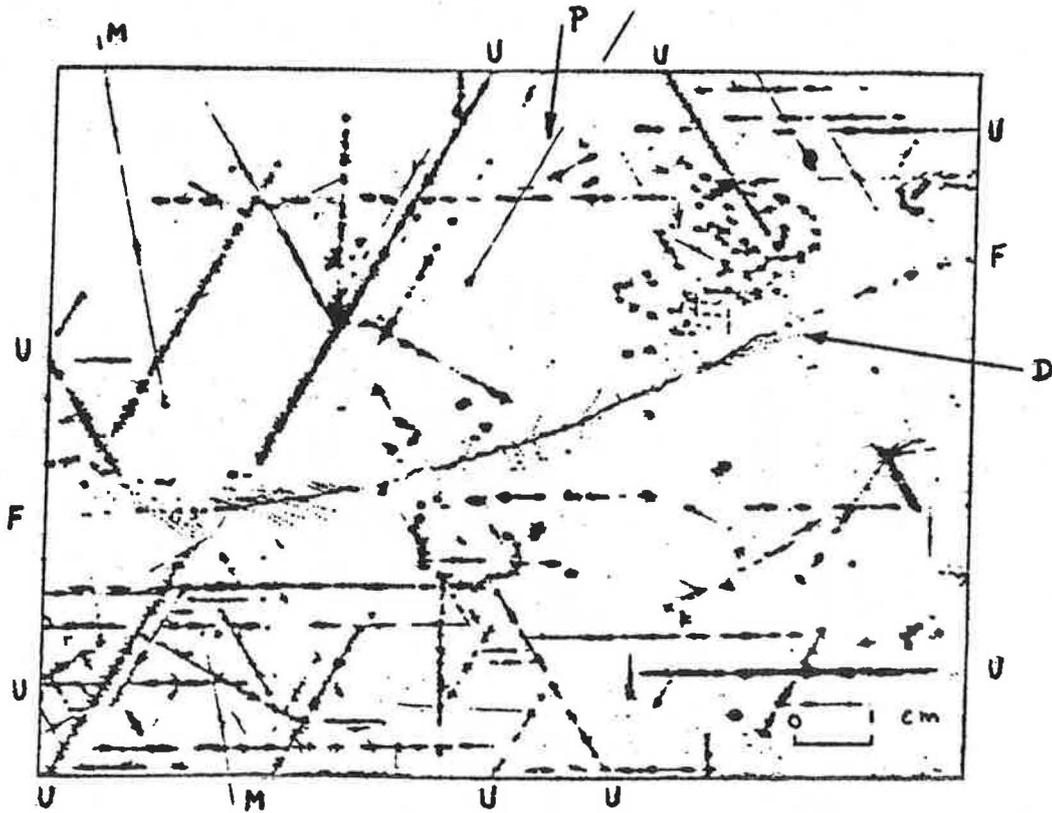


Figure 1. Photograph of a thin ( $\approx 0.05\text{mm}$ ) sheet of doped muscovite mica split in the (001) plane of easy cleavage from a single crystal, showing various types of decorated defects. Note the 1cm scale. A curved crystal fracture F extends through the sheet, from which micro-fractures and dislocations emerge. Two charged particle tracks are shown, a muon M and a positron P. The numerous lines labeled U form the sub-set of lines studied in this paper.

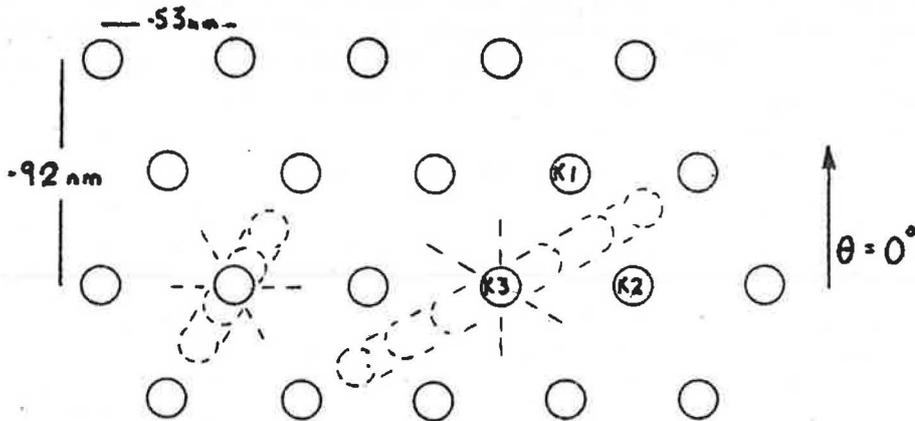


Figure 2. The arrangement of potassium atoms in monatomic sheets in the (001) plane showing the large interatomic spacings in the sheets. These potassium sheets are sandwiched between two layers consisting of sheets of closely spaced oxygen atoms coordinated by silicon atoms. The oxygen sheets are spaced  $\approx 0.15\text{nm}$  either side of the potassium sheet. Potassium atoms can have large amplitudes of quasi-focussed motion in certain directions in the plane of the sheets, as shown.

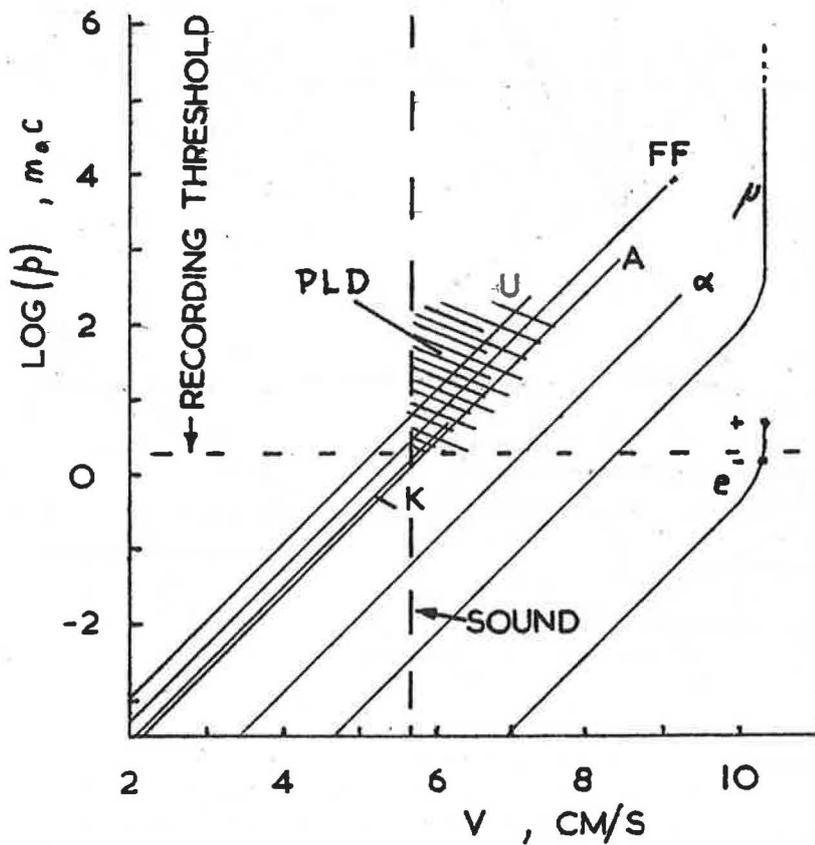


Figure 3. Momentum and velocity diagram for several types of energetic particles. The probable region for Pulse-like Lattice Disturbances (PLDs) is indicated, together with the estimated threshold for recording PLD tracks in mica. Nuclear recoils from  $\alpha$ -decay of uranium U, fission fragments from spontaneous fission FF and very energetic muons  $\mu$  are all capable of generating PLDs whereas energetic positrons  $e^+$  from potassium decay have insufficient momentum.

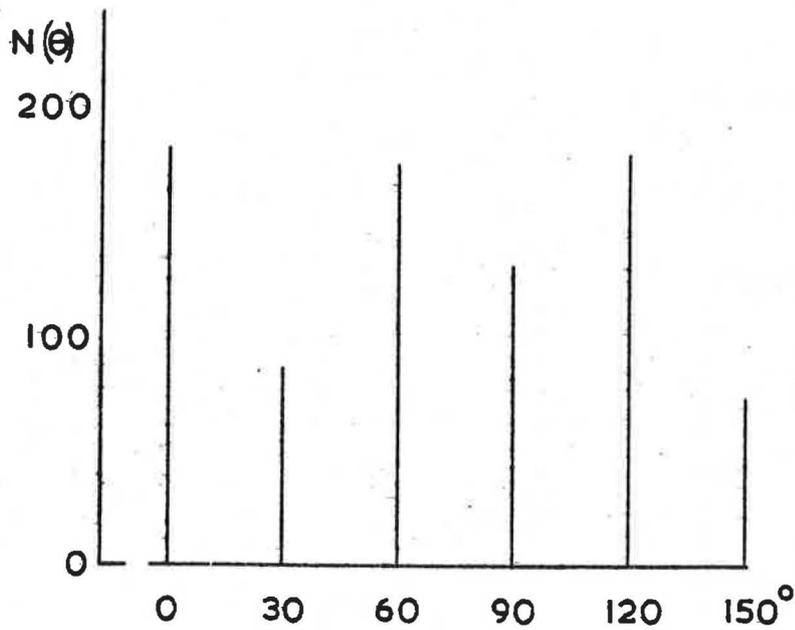


Figure 4. Plot of the angular distribution of the U-lines in the  $\langle 001 \rangle$  plane.

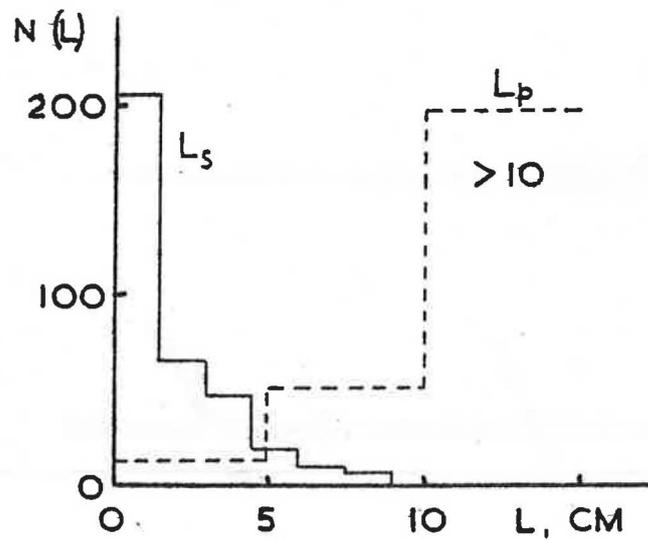


Figure 5. Length distributions of the primary U-lines  $L_p$  and of secondary or branch U-lines  $L_s$ . The primary U-lines frequently extend to a crystal edge. The extent of decoration on a branch U-line is less than that on the U-line from which it branches. In a large tree-like pattern of U-lines there can be up to about 100 branches. On average the length of U-lines decreases for higher order branch U-lines.

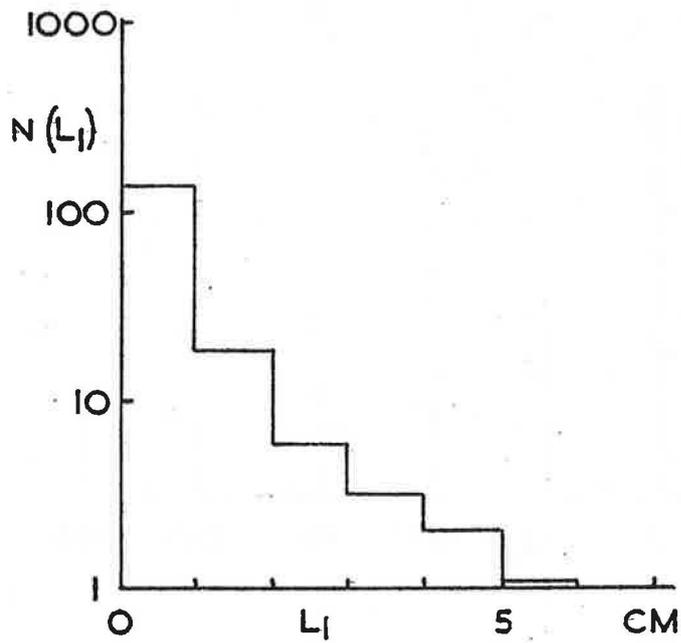


Figure 6. The distribution of length interval  $L_1$  between branch U-lines along strong primary U-lines.

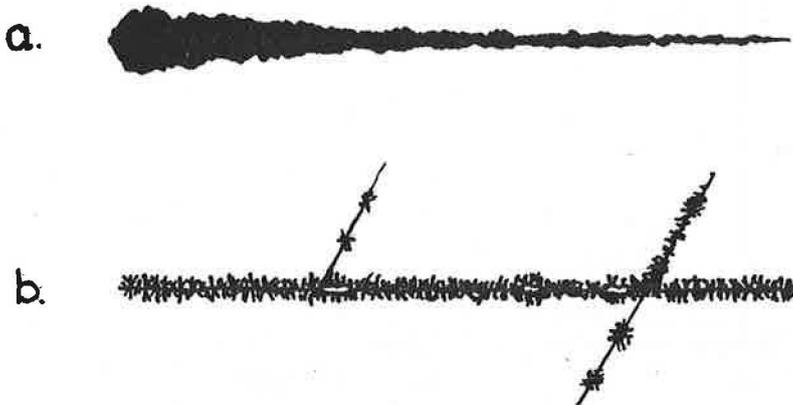


Figure 7. Micrographs of parts of decorated a) charged particle tracks and b) U-lines. The decoration on U-lines is influenced strongly by the crystal structure of muscovite.

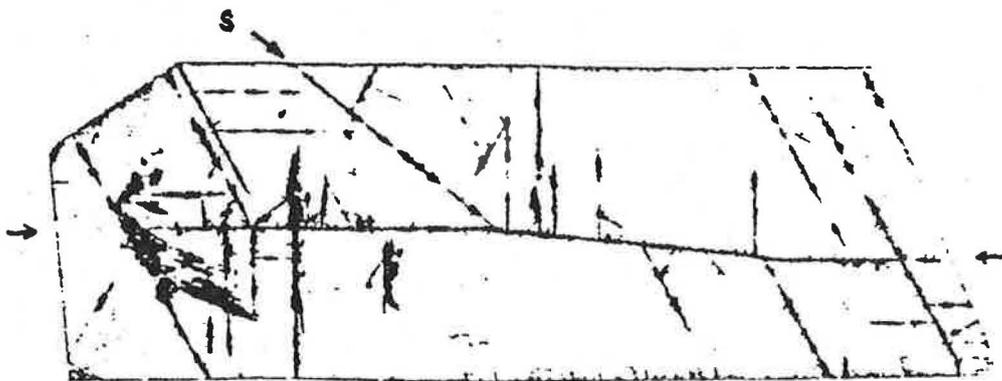
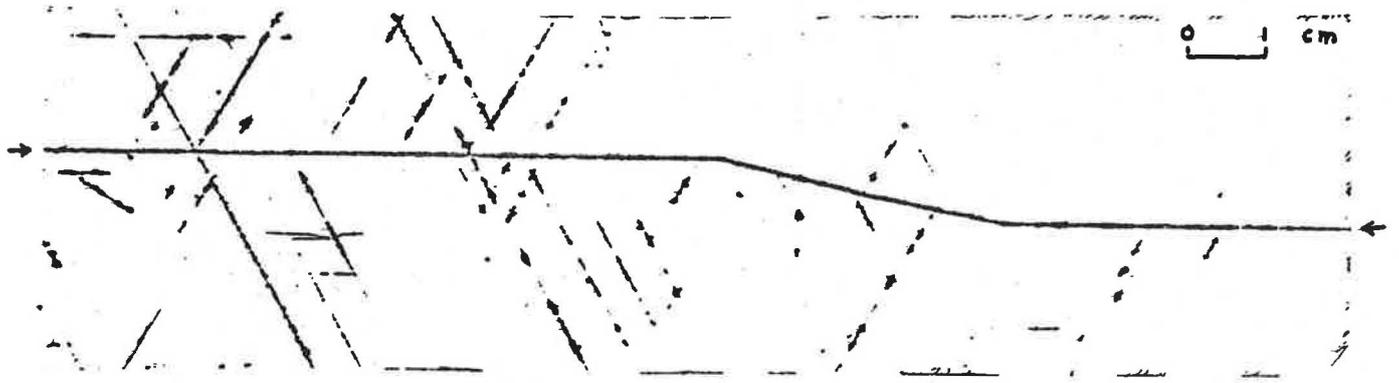


Figure 8. Photographs of strong primary U-lines which scatter away from a preferred crystallographic direction of low Miller index. The large angle scattering is sometimes accompanied by a branch U-line as indicated in the lower trace by S.

U-lines which have departed from a preferred direction show many small angular deflections before returning again to the preferred direction.





