

1 **Revision 1**

2 **Effect of orientation on ion track formation in apatite and zircon**

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17 **Abstract**

18 Fission track (FT) thermochronology is essentially based on empirical fits to annealing data of
19 FTs revealed by chemical etching, because, until now, unetched, latent FTs could not be
20 examined analytically at the atomic-scale. The major challenge to such an analysis has been
21 the random orientation of FTs and their extremely small diameters. Here we use high-
22 energy ions (2.2 GeV Au or 80 MeV Xe) to simulate FT formation along specific
23 crystallographic orientations. By combining results from transmission electron microscopy
24 (TEM) of single tracks and small angle X-ray scattering (SAXS) for millions of tracks, a
25 precise picture of track morphology as a function of orientation is obtained. High-
26 resolution analysis reveals that orientation affects the shape of tracks in apatite and zircon
27 through the preferential creation of damage along directions with highest atomic density.
28 However, track radius does not depend on orientation, contradicting previous reports.
29 Independent of track orientation, track radii, as measured at each point along the entire
30 length of 80 MeV Xe ion tracks in apatite, can be understood using the thermal spike model
31 of Szenes. Thus, the well-known track annealing anisotropy of apatite is not due to track

32 radius anisotropy. The combination of ion-irradiations with TEM and SAXS analysis
33 provides a unique opportunity to understand and model track formation and annealing
34 under a variety of geologic conditions.

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37 **Keywords:** ion tracks, fission tracks, apatite, zircon, orientation effects, thermal spike,
38 TEM, SAXS

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Introduction

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68 Swift heavy ions (SHIs) create a narrow track of damage in an irradiated solid along the
69 trajectory of the ion. Such SHI irradiations have widespread applications in
70 nanoengineering (Akcoltekin et al. 2007; Mara et al. 2004), and particle damage in
71 electronics (Dodd and Massengill 2003). More interestingly, SHI irradiations can be used to
72 simulate the formation of fission tracks (FTs), damage trails that were caused by
73 spontaneous fission events of uranium impurities in minerals that have been widely used
74 for age dating and thermochronology (Donelick et al. 1999; Gleadow et al. 2002;
75 Jonckheere and Chadderton 2002; Yamada et al. 1995). Generally, tracks in transverse cross
76 section are circular, but in a few cases (Bursill and Braunshausen 1990; Hebert et al. 1998;
77 Paul and Fitzgerald 1992; Vetter et al. 1994), they have a tendency to facet along specific
78 crystallographic directions, with an important indication that orientation may play a role in
79 track formation. However, details of how orientation affects track formation remain
80 elusive, because there are very few systematic studies of the shape and radius of latent,
81 unetched tracks as a function of orientation at the atomic-scale. According to a highly cited
82 transmission electron microscopy (TEM) study (Paul and Fitzgerald 1992), the mean
83 radius ($\sim 4.5 \pm 3.7$ nm) of latent FTs $\parallel c$ (or the c axis) of the hexagonal apatite
84 $[\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2]$ is significantly larger than that ($\sim 2.5 \pm 2.8$ nm) of FTs $\perp c$. The anisotropy of
85 track radii in apatite has gained general acceptance (Carlson 1993; Jaskierowicz et al. 2004;
86 Rabone et al. 2008; Schouwink et al. 2010), and has been used to explain (Carlson 1993;
87 Paul and Fitzgerald 1992) the well-known track annealing anisotropy (Donelick et al. 1999;
88 Gleadow et al. 2002; Green and Durrani 1977), i.e., tracks $\parallel c$ anneal more slowly than

89 tracks $\perp c$, however, a molecular dynamics simulation concluded that track size was
90 insensitive to orientation (Rabone et al. 2008). Because of the random orientation of FTs,
91 TEM imaging of the track radius requires considerable effort to accurately determine the
92 orientation of any individual track that is measured, essentially requiring the alignment of
93 the electron beam parallel to major zone axes of the grain. In this study, we overcome this
94 problem by investigating parallel tracks produced by the irradiations of SHIs (2.2 GeV Au
95 or 80 MeV Xe) along known zone axes of apatite ($P6_3/m$) and zircon ($ZrSiO_4$: $I4_1/amd$), two
96 minerals that have anisotropic structures and figure prominently in FT thermochronology.
97 By combining the results from TEM analysis of single tracks and small angle X-ray
98 scattering (SAXS) for millions of tracks (Afra et al. 2011), a precise picture of track
99 morphology as a function of orientation is obtained. High-resolution TEM analysis reveals
100 how orientation affects track shape and size at the atomic-scale. Using the microtome
101 cutting method and the thermal spike model, we compare variations in radius at each point
102 along the entire length of 80 MeV Xe ions in apatite $\parallel c$ and $\perp c$.

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Experimental methods

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Two types of SHIs were used in this study. For the 2.2 GeV Au ion irradiation
107 experiments, the ion range is ~ 70 and $85 \mu\text{m}$, as calculated by the SRIM code (SRIM 2012),
108 for zircon and apatite, respectively (Fig. 1). The electronic energy loss per unit length, or
109 stopping power, $(-dE/dx)_e = S_e$, as a function of target depth was calculated by SRIM simulation
110 with the function of Calculate Quick Range Table. The densities of apatite and zircon were set as
111 3.2 and 4.52 g/cm^3 , respectively. The total number of runs (or ions) was set $10,000$ to calculate
112 ion distribution and damage along the ion trajectories. The SRIM simulation shows that the

113 angle of each track deviated from the original trajectory for the 2.2 GeV Au ion is negligibly
114 small, especially at the lower target depths ($< 40 \mu\text{m}$). The samples were pre-polished to a
115 thickness $\sim 40 \mu\text{m}$ to ensure that the GeV projectiles completely penetrate the sample
116 thickness, inducing an approximately uniform energy deposition, $-dE/dx$, and producing a
117 nearly constant track radius throughout the entire sample thickness (Li et al. 2013) . In
118 contrast, as the 80 MeV Xe ions, which have a typical mass and energy of fission fragments,
119 slow down, $-dE/dx$ decreases (Fig. 1), resulting in a significantly smaller track radius in
120 apatite toward the end of the track (Li et al. 2012). Variations in radius along the entire
121 length (up to $8.1 \mu\text{m}$) of ion tracks along the c -axis of apatite were first obtained by the
122 microtome method (Li et al. 2012). For a fission fragment (e.g. 80 MeV Xe ion), the track
123 range that can be observed by TEM is $\sim 2 \mu\text{m}$ (Li et al. 2012) less than the ion range because
124 the energy at its very end falls below the energy threshold required to produce an
125 observable track. Although the energy of 2.2 GeV Au is significantly larger than that of a
126 typical fission fragment, the difference in $-dE/dx$ is small at the point of impact (for apatite:
127 $\sim 27 \text{ keV/nm}$ for 2.2 GeV Au vs. $\sim 16 \text{ keV/nm}$ for 80 MeV Xe) (Fig. 1). More importantly, this
128 difference in $-dE/dx$ does not result in significant differences in the track morphology (Li et
129 al. 2010).

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131 Tracks ($\parallel c$ or $\perp c$) were produced in bulk samples $\sim 40 \mu\text{m}$ thick by exposing (0001) and
132 ($5\bar{1}\bar{4}0$) surfaces of Durango fluorapatite, and (001) and (100) surfaces of synthesized
133 single crystal zircon, respectively, to 2.2 GeV Au ions in 5×10^{10} ions/cm² at normal
134 incidence at the UNILAC accelerator, GSI, Darmstadt, Germany. Without further preparation,
135 bulk samples were investigated after irradiation using transmission SAXS at the Australian

136 Synchrotron in Melbourne, Australia. See Kluth et al. (2008) and Li et al. (2013) for details
137 of the SAXS measurements. TEM studies were completed with a JEOL 3011 electron
138 microscope. The irradiated bulk samples were crushed and suspended on a carbon film
139 supported by Cu grid for the TEM studies. To simulate FTs, tracks $\parallel c$ or $\perp c$ of apatite were
140 produced by irradiating a single crystal $\sim 200 \mu\text{m}$ thick with 80 MeV Xe ions to a fluence of
141 5×10^{10} ions/cm² at the Cyclotron DC-60 at the National Nuclear Centre in Kazakhstan.
142 Cross-sectional TEM specimens containing parallel tracks were prepared by cutting the
143 irradiated sample along their ion trajectories ($\parallel c$ or $\perp c$) with a diamond knife in a
144 microtome to show variations in track radius along the entire track. See Li et al. (2012) for
145 details of the microtome method.

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Results and discussion

2.2 GeV Au Ions

149 High resolution TEM (HRTEM) analysis of 2.2 GeV Au tracks was obtained in transverse
150 cross section, i.e., tracks are parallel to electron beam, by carefully tilting a crushed
151 specimen to align its zone axis ($\parallel c$ or $\perp c$), which is parallel to the tracks in this case, to the
152 incident electron beam. In apatite, the central core region of the tracks has the same bright
153 contrast as the free space (Figs. 2a,2b and 3a), indicating the highly porous nature of tracks,
154 as has been discussed in detail in (Li et al. 2010). This is significantly different from the
155 amorphous core region of tracks in zircon (Figs. 2c, 2d, and 3b). For each track $\parallel c$ of
156 apatite, there is clear hexagonal faceting (Fig. 2a) (Paul and Fitzgerald 1992), as compared
157 with the circular tracks $\perp c$ (here along $[5\bar{1}\bar{4}0]$) (Figs. 2b and 3a). In a fast Fourier
158 transformation (FFT) image, a vector pointing from (0000) to a diffraction spot represents

159 the corresponding crystal direction in the imaging plane. Thus, the preferential faceting
160 directions in Fig. 2a can be determined as the six equivalent, close packed
161 $\langle 11\bar{2}0 \rangle$ directions, which are parallel to the six diagonals of a hexagonal projection onto
162 (0001) for etched tracks $\parallel c$ (Jonckheere and VandenHaute 1996; Paul and Fitzgerald
163 1992). In zircon, tracks along [001] appear to be approximately square in shape (Fig. 2c)
164 with the diagonals parallel to the four equivalent $\langle 100 \rangle$ directions, as this tendency can be
165 further confirmed in a low magnification image (Fig. 3b), which shows multiple square
166 tracks with the diagonals pointing in the same directions. Within the (001) plane, the
167 atomic density is highest along the $\langle 100 \rangle$ directions, similar to the case in the close packed
168 directions. Tracks along [100] of zircon appear to be elliptical with the long axis in the
169 $\langle 001 \rangle$ direction, which have the highest atomic density within the (100) plane (Fig. 2d).
170 The ratio (1.106) of the long axes to the short axes (7.8 nm vs. 7.1 nm) in Fig. 2d
171 corresponds to the reciprocal ratio of the unit cell constants ($a/c = 1.105$), as well as the
172 corresponding d spacings ($d_{200}/d_{002} = 1.105$). This is similar to the case for GeS, where the
173 long axis of the elliptical tracks are parallel to the close packed $\langle 001 \rangle$ direction (Vetter et
174 al. 1994). Thus, the atomic-scale resolution reveals that the shape of tracks in apatite and
175 zircon depends on track orientation in the crystal as a result of preferential creation of
176 radiation damage along the directions with highest atomic density.

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178 As the scale bar can be precisely calibrated by the known d spacings (Fig. 2), the mean
179 track radius can be measured reliably: 4.0 ± 0.2 nm for tracks $\parallel c$ vs. 3.9 ± 0.4 nm for tracks
180 $\perp c$ in apatite; 3.8 ± 0.4 nm for tracks $\parallel c$ vs. 3.7 ± 0.4 nm for tracks $\perp c$ in zircon, which is
181 consistent with a previous HRTEM measured radius (~ 4 nm) for 2.9 GeV Pb ion induced

182 tracks $\perp c$ of zircon (Bursill and Braunshausen 1990). The mean radius was determined by
183 averaging the radii of approximately 5 tracks for each orientation ($\parallel c$ or $\perp c$) in apatite and
184 zircon. Within the errors, the HRTEM measured track radii are actually indistinguishable for
185 the different orientations. However, the track shape changes significantly for the different
186 orientations.

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188 In addition to the images of tracks in transverse cross section, the structure and
189 morphology of tracks can be viewed in longitudinal cross section, i.e., tracks are
190 perpendicular to the electron beam by the under focus low magnification images (Fig. 4).
191 For apatite, the average radius of tracks $\parallel c$ and $\perp c$ is 4.6 ± 0.4 nm and 4.6 ± 0.5 nm,
192 respectively. For zircon, the average radius is 4.3 ± 0.4 nm $\parallel c$, close to 4.1 ± 0.5 nm for $\perp c$.
193 Ten tracks were measured for each orientation. Thus, the mean values of the track radii are
194 insensitive to orientation, which is consistent with the HRTEM measurements.

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196 As compared with the limited number of tracks in a localized area as observed by TEM,
197 the strong scattering oscillations, as detected by SAXS from a very large number of well
198 aligned, identical tracks in a bulk sample, provide an extremely reliable means for
199 determining the mean track radius. The scattering intensities of tracks and corresponding
200 analytical fits to the hard cylinder model (Afra et al. 2011; Li et al. 2013) are shown in Fig.
201 5. The GeV Au ions induce tracks $\parallel c$ of apatite with a mean radius of 4.81 ± 0.03 nm, which
202 is slightly larger than that (4.72 ± 0.03 nm) of tracks $\perp c$. In zircon, the mean track radius \parallel
203 c is 4.43 ± 0.03 nm, slightly larger than that (4.32 ± 0.03 nm) of tracks $\perp c$. The SAXS

204 measurements are consistent with the TEM measurements in that the mean radii of tracks
205 $\parallel c$ are very close to those of tracks $\perp c$.

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207 **80 MeV Xe Ions**

208 In addition to the GeV ions, the influence of track orientation on track radius was
209 investigated by TEM comparing the damage at each point along latent tracks created by 80
210 MeV Xe ions $\parallel c$ or $\perp c$ of apatite (Fig. 6). Using the microtome method (Li et al. 2012), the
211 track profile was obtained by taking a series of images along the ion paths (Figs. 6a-6d).
212 Three examples of under focus TEM images of tracks $\perp c$ measured at different depths ($x =$
213 0, 3, and 6.7 μm) from the ion-irradiated surface are shown in Figs. 6a-6c. Approximately
214 10 tracks were measured at each target depth. The changes of track radii along the ion
215 paths $\parallel c$ and $\perp c$ in apatite are plotted in Fig. 6e. Within experimental error, the mean radii
216 for tracks $\perp c$ are very close to the radii of tracks $\parallel c$ at almost all the depths. This provides
217 direct evidence that latent ion tracks in apatite do not have different sized radii as a
218 function of orientation, in contrast to the previous observation of significantly larger FTs
219 $\parallel c$ than those $\perp c$ in apatite (Paul and Fitzgerald 1992). This discrepancy is due to the
220 difficulty of determining the precise orientation of any individual track [of the total 293
221 randomly orientated FTs that were measured (Paul and Fitzgerald 1992)]. For complex
222 materials (e.g., apatite) where different zones may have similar diffraction patterns, the
223 incorrect indexing of the zone axis of a grain is possible. Furthermore, because it is
224 impossible to determine the target depth of a randomly orientated FT by TEM, the decrease
225 in radius along a FT, as clearly shown in Fig. 6, could not be considered, resulting in
226 significantly larger errors.

227 For both track directions, the decrease in track radius per unit depth, $-dR/dx$, is relatively
228 low at shallower depths ($0 \leq x \leq 3.5 \mu\text{m}$). The track radii at shallower depths (Figs. 6a and
229 6b) can be described by a relation of $R \sim S_e^{1/2}$ (Fig. 6e), as this relation was applied to tracks
230 in $\text{Y}_3\text{Fe}_5\text{O}_{12}$. However, at greater depths ($x \geq 3.5 \mu\text{m}$) (Fig. 6c), R falls below the curve of
231 $R \sim S_e^{1/2}$, as the $-dR/dx$ is obviously higher than that at the shallower depths (Fig. 6e). The
232 dE/dx as a function of target depth x was simulated by the SRIM code (2012), and R was
233 measured at different depths by TEM (Fig. 6e). Therefore, R as a function of $-dE/dx$ for both
234 directions can be obtained, and these values are plotted in Fig. 6f. These data were
235 compared with the theoretical prediction from the thermal spike model of Szenes (1995).
236 Depending on the value of S_e , the track radius R (the maximum radius of the melt during
237 the heating-cooling process) can be determined by two different expressions (Szenes
238 1995). For $S_e \geq 2.7S_{et}$, the melted zone expands and reaches its maximum,

$$239 R^2 = [(a(0)^2/2.7)](S_e/S_{et}), \quad (1)$$

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241 where S_{et} is the threshold energy for the production of continuous tracks. $a(0)$ relates to
242 thermal diffusivities, but it can be determined by fitting. This expression is consistent with
243 our analysis shown in Fig. 6e that $R \sim S_e^{1/2}$ for tracks at shallower depths. For $2.7 \geq S_e/S_{et} \geq 1$,
244 the melted zone shrinks with time, and it has the maximum radius at $t = 0$ when the cooling
245 spike begins,

$$246 R^2 = a(0)^2 \ln(S_e/S_{et}). \quad (2)$$

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248 This regime ($2.7 \geq S_e/S_{et} \geq 1$) corresponds to tracks at greater depths where the values of S_e
249 are smaller (Fig. 6c). There is a smooth transition from the logarithmic to linear regimes, as
250 both Eqs. 1 and 2 provide the same R^2 at $S_e = 2.7S_{et}$. In this study, the transition point is at x
251 $\sim 3.5 \mu\text{m}$ in Fig. 6e. Thus, the corresponding two parameters for the fits ($R = a(0) = 3.6 \text{ nm}$,

252 $S_e = 2.7S_{et} = 1065 \text{ eV/\AA}$) can be directly determined from the transition point in Fig. 6e. As
253 shown in Fig. 6f, all experimental data for 80 MeV Xe ions follow a single curve as a good
254 approximation in both the logarithmic and linear regimes. The data for the 2.2 GeV Au
255 induced tracks (measured from Figs. 4a-4b) are below the curve (Fig. 6f). This is due to the
256 velocity effect (Meftah et al. 1993), which describes the decrease of track radius with the
257 ion speed at certain high energy regimes.

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Remarks

261 The hexagonal apatite has an open anion channel || *c* (Calderin et al. 2003). The exact details
262 of how structural anisotropy affects anisotropic annealing behavior remain unclear, although
263 there is certainly a connection between the two types of anisotropy (Gleadow et al. 2002;
264 Green and Durrani 1977). As mentioned above, the slower annealing rate of tracks || *c* was
265 attributed to the larger tracks || *c* (Paul and Fitzgerald 1992). Besides the weaker
266 intermolecular bonds along the *c*-axis (Paul and Fitzgerald 1992), smaller dielectric
267 constants || *c* were used to explain the larger tracks along this direction (Rabone et al.
268 2008). In this study, except for the orientation-related difference in track shape as
269 discussed above, the detailed TEM and SAXS experimental evidence on parallel tracks with
270 well-defined orientations in the structure clearly demonstrates that the track radii have no
271 apparent dependence on track direction in apatite, which is in agreement with a previous
272 molecular dynamics simulation (Rabone et al. 2008). Based on the direct observation of *in*
273 *situ* thermal annealing of unetched FTs in apatite (Li et al. 2011), the annealing anisotropy
274 as observed in chemically-etched tracks can be attributed to the preferential motion of FT

275 segments || c during annealing. Similarly, no obvious track radius anisotropy was detected
276 in zircon by the TEM and SAXS measurements, although zircon is structurally anisotropic
277 with a chain of alternating edge-sharing SiO_4 tetrahedra and ZrO_8 triangular dodecahedra
278 extending || c (Robinson et al. 1971).

279 Currently, FT thermochemistry is based almost entirely on empirical fits to annealing
280 data of FTs revealed by chemical etching. One of the main obstacles to quantitative modeling
281 of FT behavior has been the need to investigate the morphology of latent tracks as a
282 function of temperature and time. For a single track created by a fission fragment pair, the
283 radius is largest near the midpoint of track where the fission event occurs, and decreases
284 toward each end as the S_e decreases, as shown in Fig. 1. At elevated temperatures, the
285 etchable track length is gradually reduced from each end of a FT towards the midpoint
286 because the track ends have much smaller radii, and anneal much more rapidly, which was
287 observed by *in situ* TEM (Li et al. 2012). As a result, only the sections with smaller S_e (as
288 well as R) near each end of the FT make a major contribution for the length measurements
289 of etched tracks and mathematical fitting [see Ref. (Li et al. 2012) for details]. Therefore,
290 the description of the two different regimes with a single curve, especially the description
291 of the regime with smaller S_e by a simple logarithmic equation (i.e., Eq. 2), provides an
292 important physical form that has long been recognized as needed (Carlson 1990; Gleadow
293 et al. 2002; Jonckheere and Chadderton 2002) for the development of physical models of
294 FT formation and annealing. Besides the precise picture of morphologies of FTs, as
295 demonstrated in this study, the combination of ion-irradiation with TEM and SAXS can be
296 readily used in order to obtain a deeper understanding of the gradual recovery of radiation
297 damage during annealing at the atomic-scale.

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408 **Figure Captions**

409 **Figure 1.** The electronic energy loss per unit length, $(-dE/dx)_e = S_e$, as a function of target
410 depth, x , for the irradiations of 2.2 GeV Au ions in zircon, 2.2 GeV Au ions in apatite, and 80
411 MeV Xe ions in apatite, respectively.

412 **Figure 2.** Transverse cross section HRTEM images of tracks induced by exposure to 2.2
413 GeV Au ions $\parallel c$ or $\perp c$ of apatite and zircon. The track orientation (or the zone axis of the
414 target material) was determined by indexing the inset of fast Fourier transform image for
415 each figure.

416 **Figure 3.** Conventional transverse cross section TEM images (under focus and bright-field)
417 show multiple tracks in **(a)** apatite and **(b)** zircon. The areal densities of the 2.2 GeV Au
418 ions induced tracks for both materials are consistent well within the nominal ion density
419 (5×10^{10} ions/cm²). The track orientation (or the zone axis of the target material) was
420 determined by indexing the inset of diffraction pattern for each figure. A track in **b** marked
421 with the white arrows and square as a guide to the eye, is a representative of multiple
422 square tracks with diagonals pointing in the $\langle 100 \rangle$ directions.

423 **Figure 4.** Conventional TEM images (under focus and bright-field) show tracks (viewed in
424 longitudinal cross section) induced by exposure to 2.2 GeV Au ions $\parallel c$ or $\perp c$ of apatite or
425 zircon, as marked in each image.

426 **Figure 5.** SAXS measurements showing scattering intensities of 2.2 GeV Au ion induced
427 tracks $\parallel c$ or $\perp c$ of apatite and zircon as a function of the scattering vector q , and
428 corresponding fits with the hard cylinder model (solid lines). The errors of track radii
429 correspond to the uncertainty of the fitting.

430 **Figure 6.** A comparison of variations in radius at each point along the tracks induced by the
431 irradiation of 80 MeV Xe ions || c and $\perp c$ of apatite. (a) to (c) show three examples of
432 conventional TEM images (under focus and bright-field) of tracks $\perp c$, taken at different
433 depths from the ion irradiated surface along the whole length of a microtome section, as
434 marked in a lower magnification image (d) A thin layer of gold was deposited on the
435 irradiated surface as a surface marker. (e) Mean track radii, R , almost overlap along the
436 tracks of the two directions. $R \sim S_e^{1/2}$ at smaller target depths. (f) Variations in track radius
437 versus S_e normalized to the threshold value of S_{et} can be understood in terms of the Szenes
438 Model.

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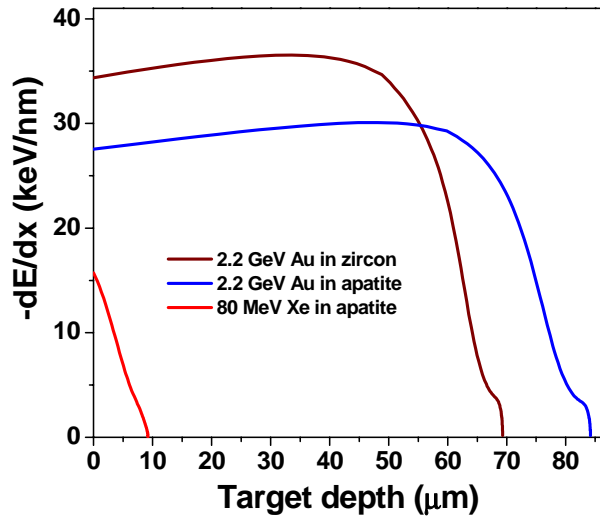
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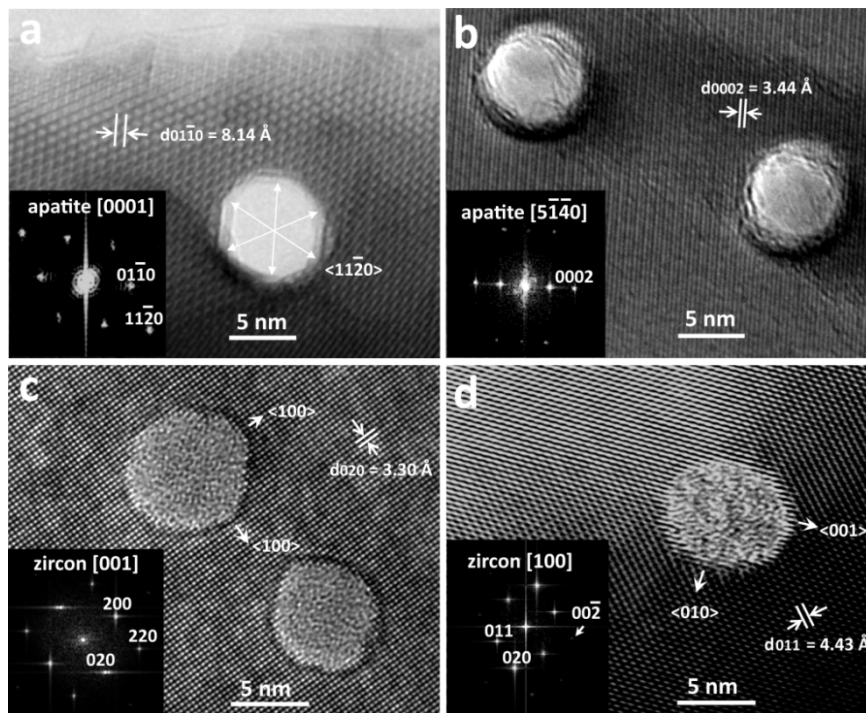
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Fig. 1

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Fig. 2

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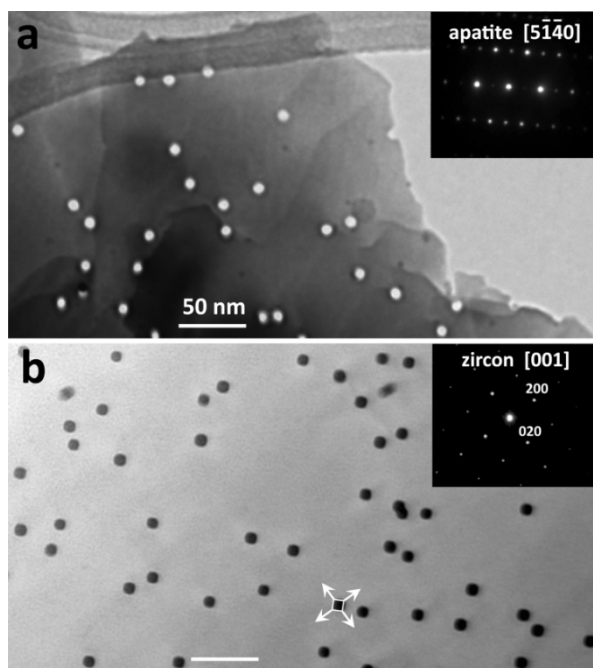
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Fig. 3

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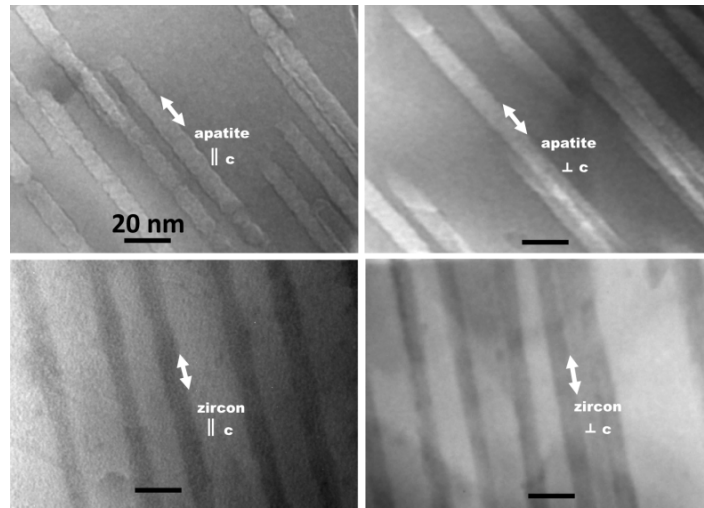
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Fig. 4

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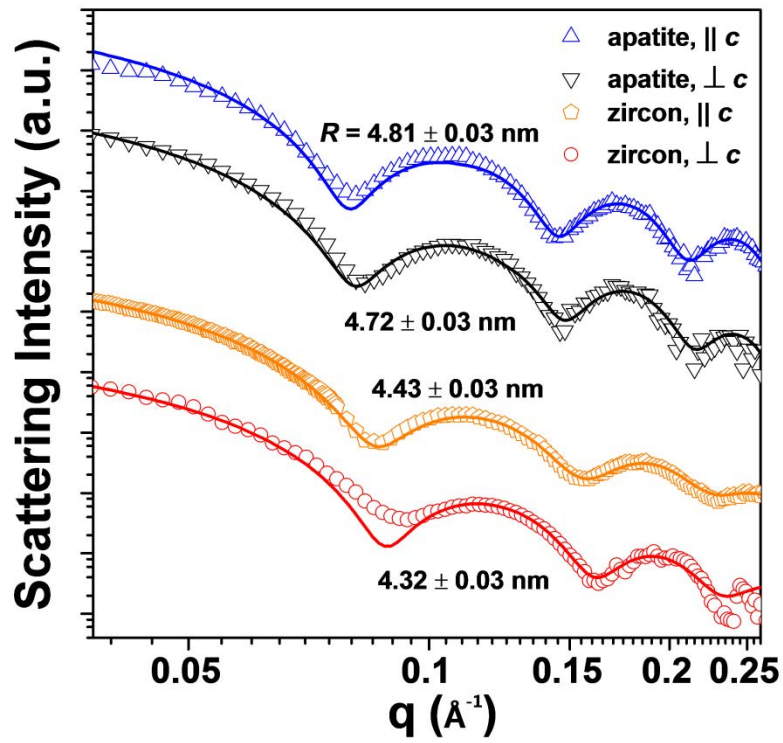
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Fig. 5

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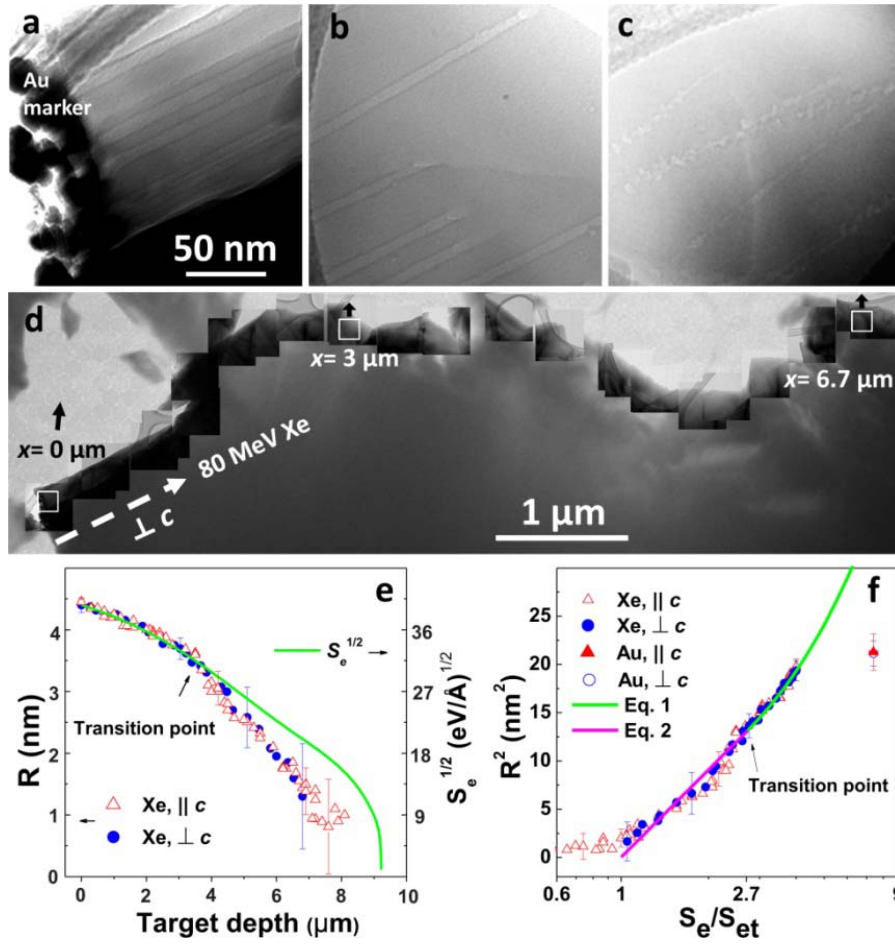
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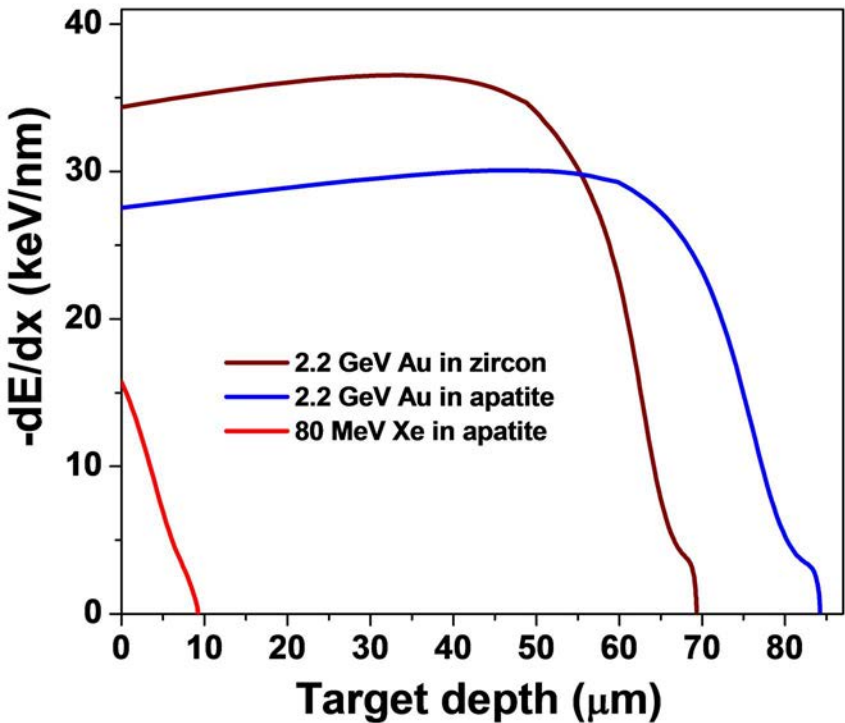
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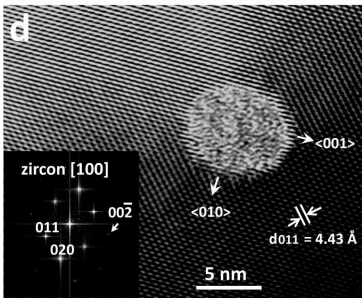
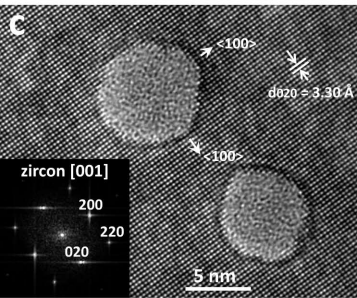
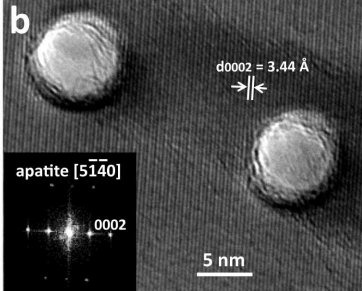
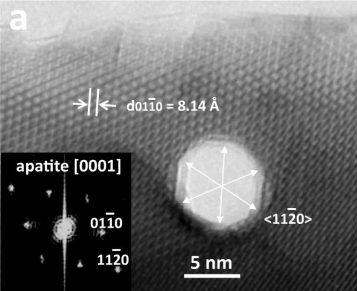
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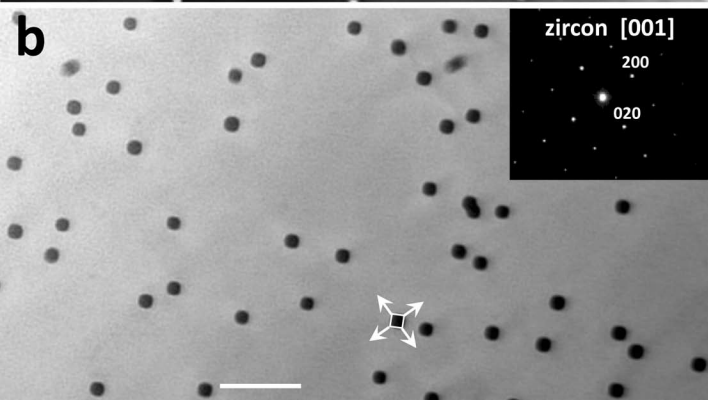
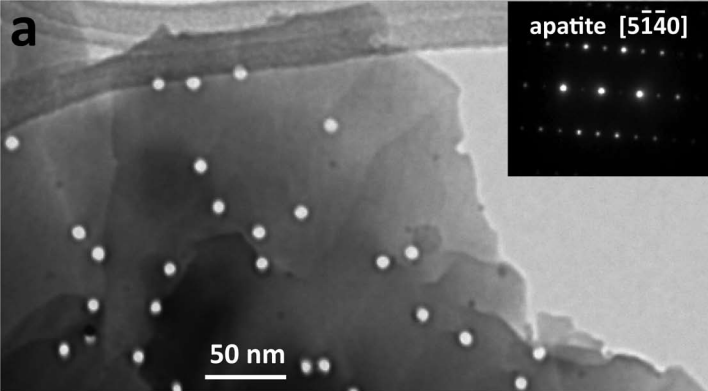
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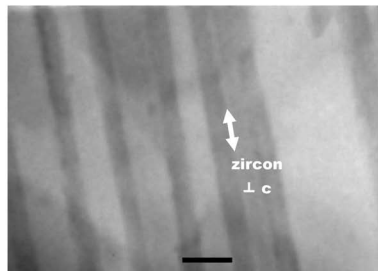
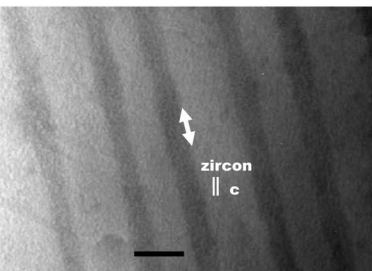
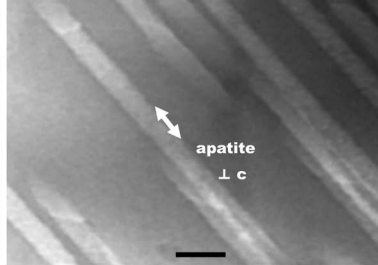
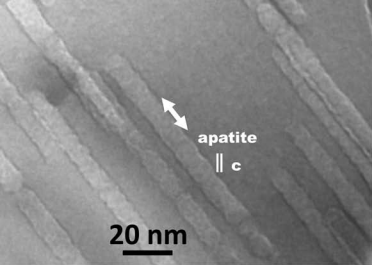
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Fig. 6









Scattering Intensity (a.u.)

