Effect of ion velocity on the creation of point defects halos of latent tracks in alkali-halides

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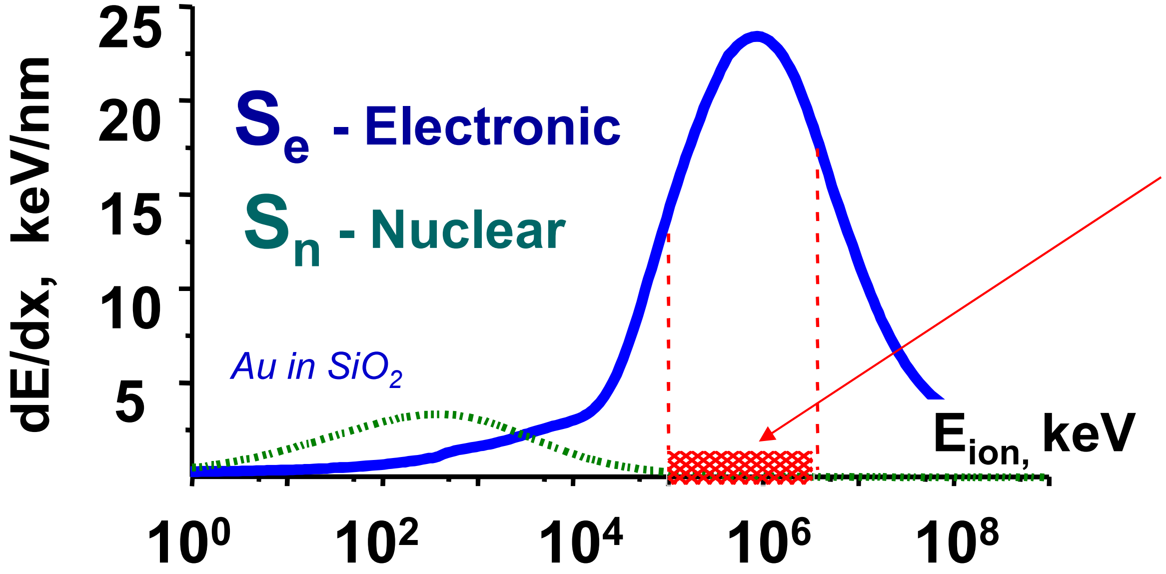
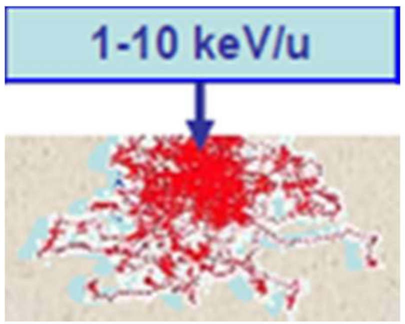
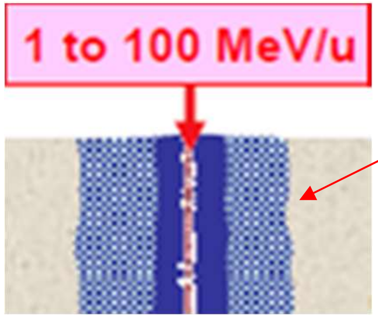
Swift Heavy Ions



1. **Bragg Peak:**

$S_n / S_e < 0.01$





2. $M_{ion} > 20\ m_p$

$E_{ion} > 1$ MeV/u

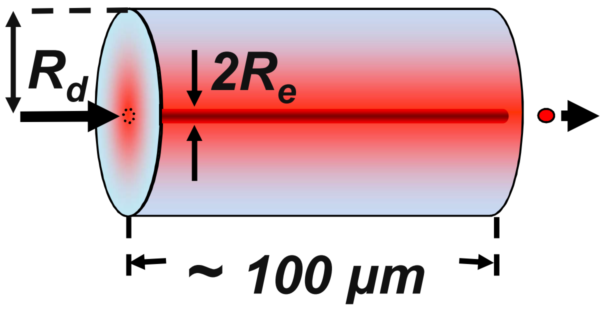
$S_e > 1$ keV/nm

$O^+ > 20$ MeV

$Xe^+ = 100 – 300$ MeV

$U^+ = 1 – 2$ GeV

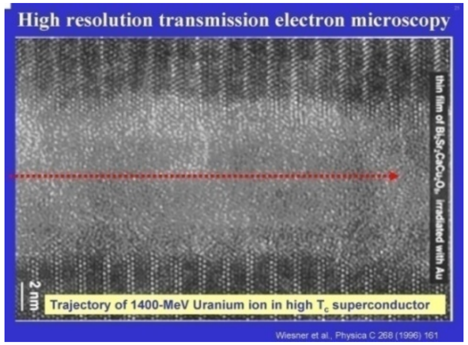
A new tool for nanostructuring



Initial

excitation :

R_e ~ 1nm



High resolution transmission electron microscopy

thin film of $Bi_2Sr_2CaCu_2O_8$ irradiated with Au

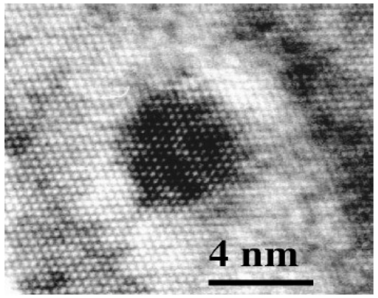
Trajectory of 1400-MeV Uranium ion in high T_c superconductor

Wiesner et al., Physica C 268 (1996) 161

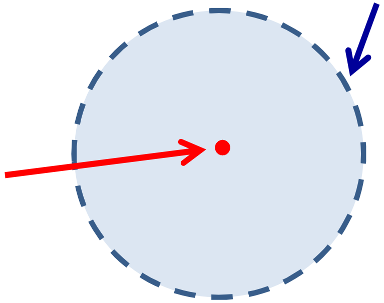
Structure

transformations:

R_d~10nm



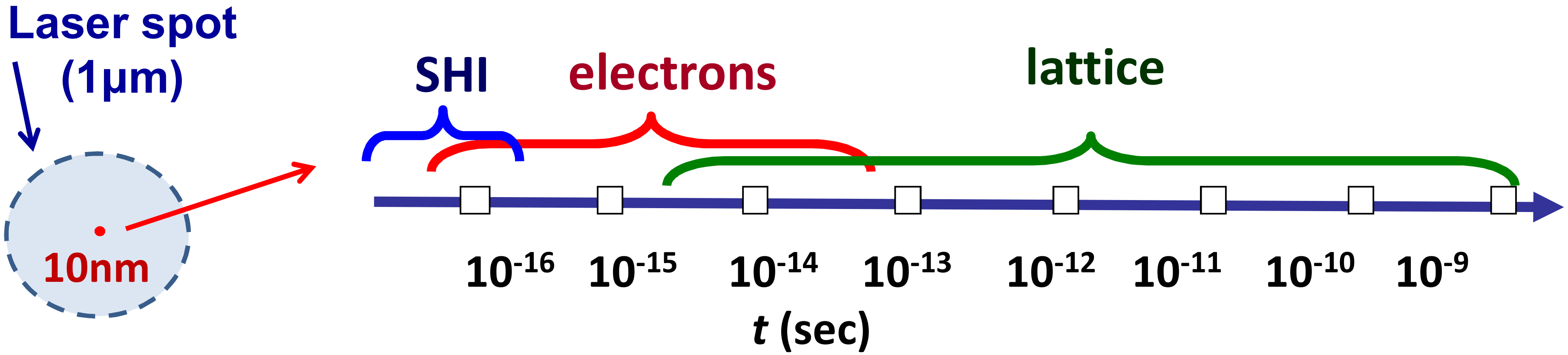
Laser spot (1µm)



- **Nanometric radial scale (nanoelectronics)**
- **Extremely high length/radius ratio (100 µm / 50 nm).**
- **Uniform tracks. Bulk effect**
- **Negligible disturbance of a surrounding matrix**
- **Direct one-step process of irradiation**
- **Elastic collisions are too week to produce detected damage**

Fundamentals

Nanometric spatial and Femto-picosecond temporal scales



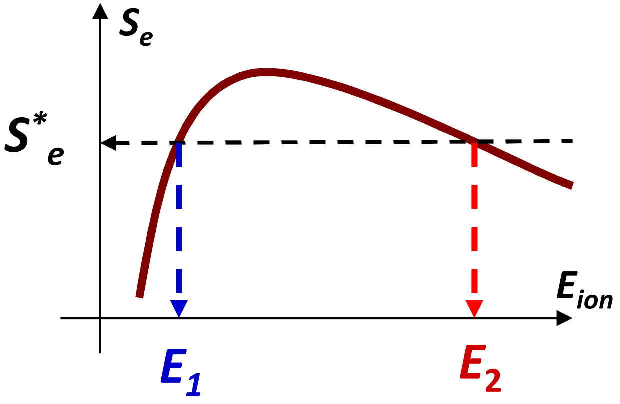
too fast, too small, too large excitation levels

Unusual pathways of the track kinetics which can not be described by macroscopic models

Conceptions of temperature, local equilibrium, heat diffusion, phonons etc. are applied hardly for the most important stages of the track kinetics

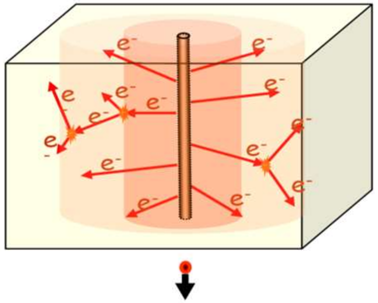
Velocity Effect

Peak-like form of stoppings



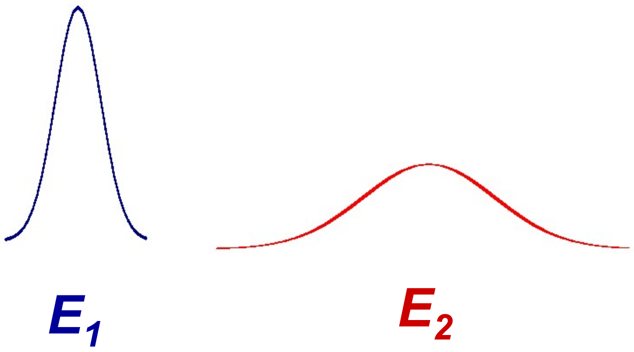
$$S^*_e(E_1) = S^*_e(E_2)$$

Two different energies (velocities) of an ion produce the same electronic stopping



$$E^{\delta}_{max} \sim (4m_e / M_i)\cdot E_i$$

Ranges of faster electrons may differ by an order of magnitude (3 nm vs 30 nm)

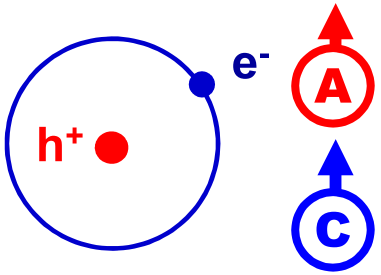


Larger density of deposited energy results in more pronounced structure transformations in tracks of slower ions

Does the velocity effect always appear the same way?

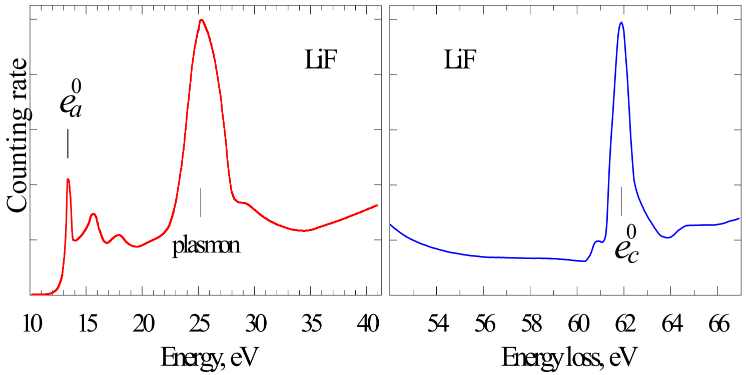
Alkali – Halides (LiF, NaCl…)

Formation of lattice defects via decay of self-trapped excitons

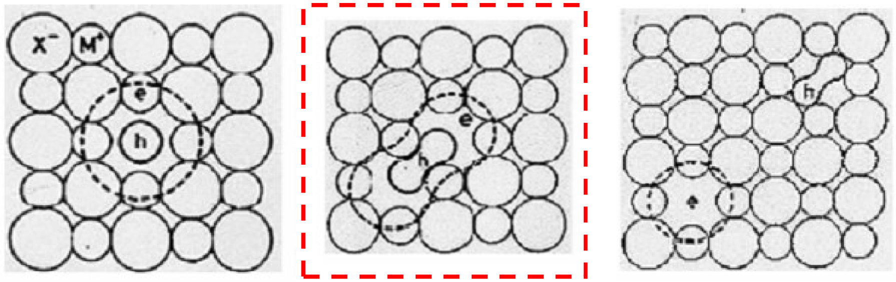


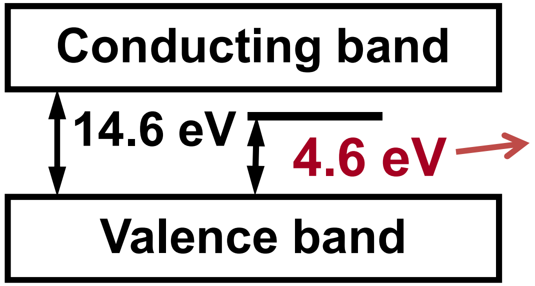
Anion exciton ~ 13 eV

Cation exciton 62 eV



Decay of self-trapped exciton *t* ~1-10 *ps*





Conducting band

14.6 eV

4.6 eV

Valence band

It is enough for creation of a Frenkel pair

Conversion efficiency *δ* ~ 0.01

Defects in alkali – halides

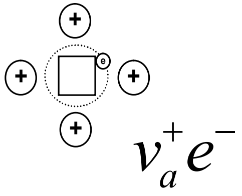
Electro-neutrality.

Vacancies and interstitials must capture charge carriers

New electronic levels due to localization of carriers

Color Centers

Well detected in optical experiments

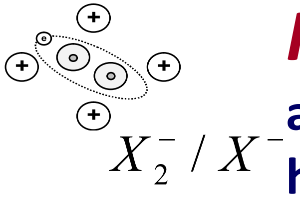


$v_a^+ e^-$

F **– center**

anion vacancy

electron center

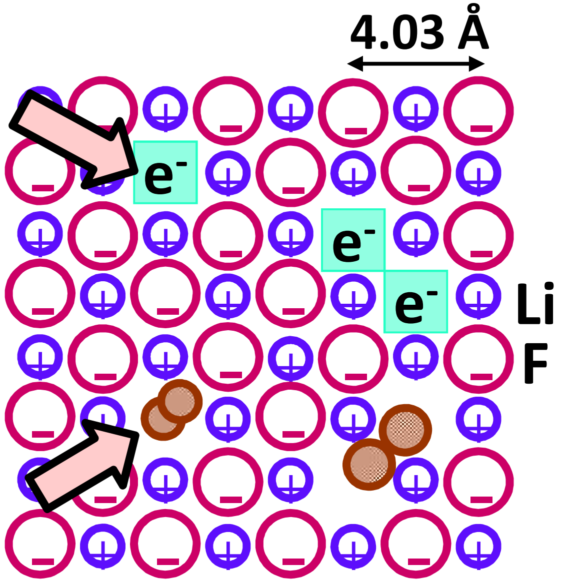


X_2^- / X^-

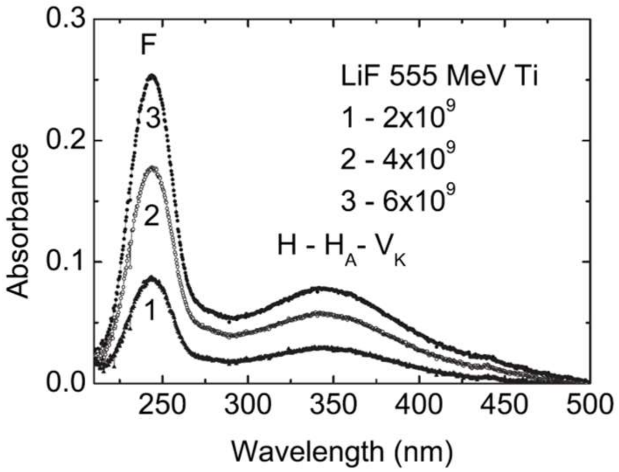
H **– center**

anion interstitial

hole center



fcc lattice, (100) plane



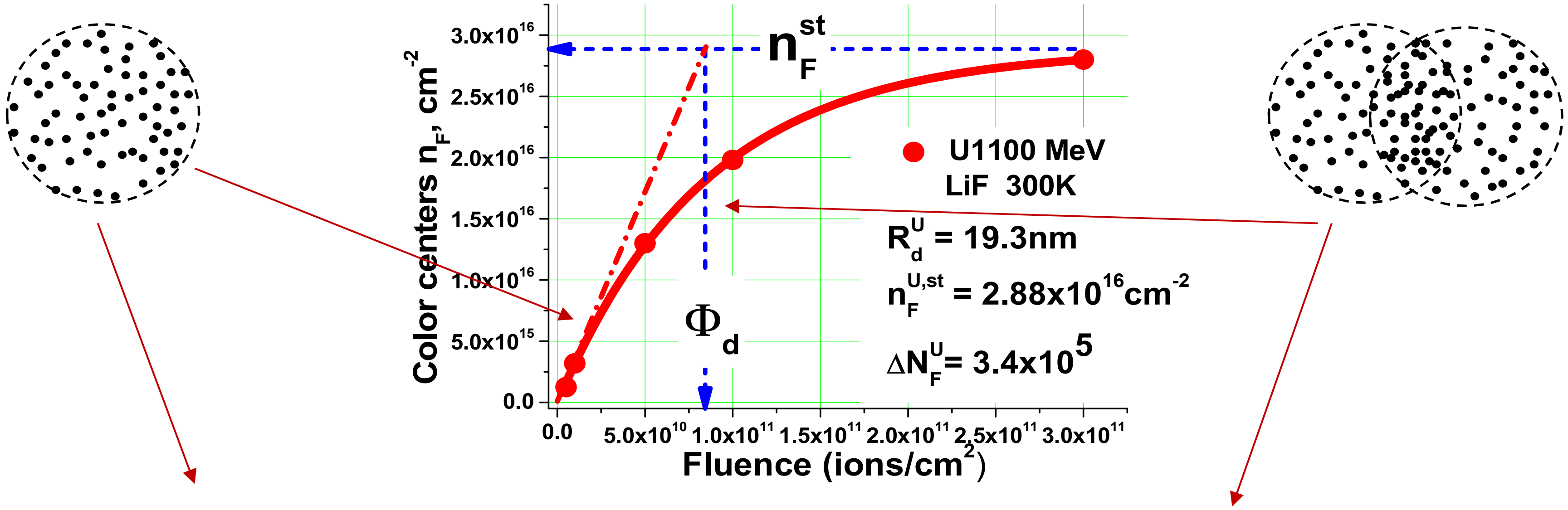
Parameters of Defect Halo of a track

K. Schwartz, A.E. Volkov, M. V. Sorokin, C. Trautmann, K.-O. Voss, R. Neumann, M. Lang, **Phys. Rev. B. 78 (2008) 024120**

$$n_F(\Phi) = n_F^{st}[1 - exp(-\Phi \pi R_d^2)]$$

1. Additive accumulation in isolated tracks at initial fluences $\Phi \ll \Phi_d$

2. Deviation from linear growth due to overlapping at $\Phi \geq \Phi_d$

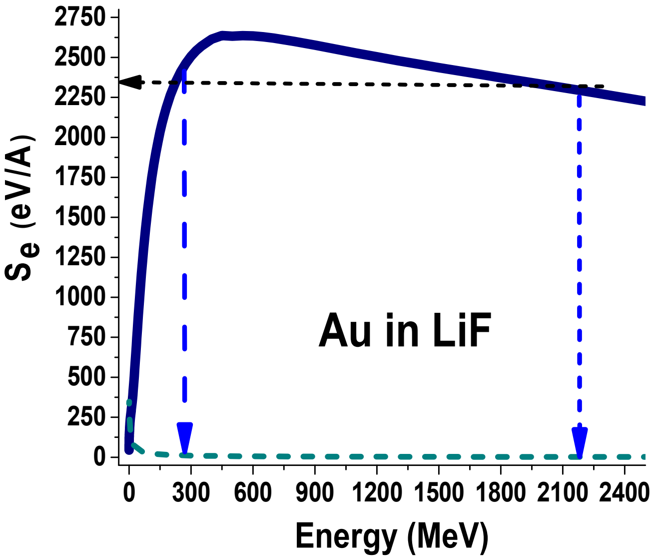


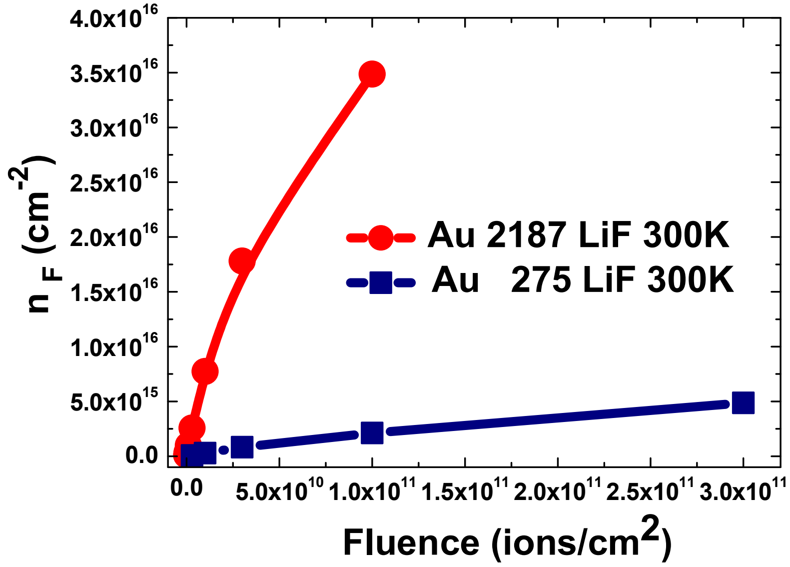
$$\Delta N_F \approx dn_F^{lin}/d\Phi = n_F^{st}\pi R_d^2$$

$$R_d = (\pi \Phi_d)^{-1/2} \sim 10–30\ \text{nm}$$

$$< C_F > = \Delta N_F / (\pi R_d^2 L)$$

Velocity effect in LiF: Au (272 MeV) vs Au (2187 MeV)

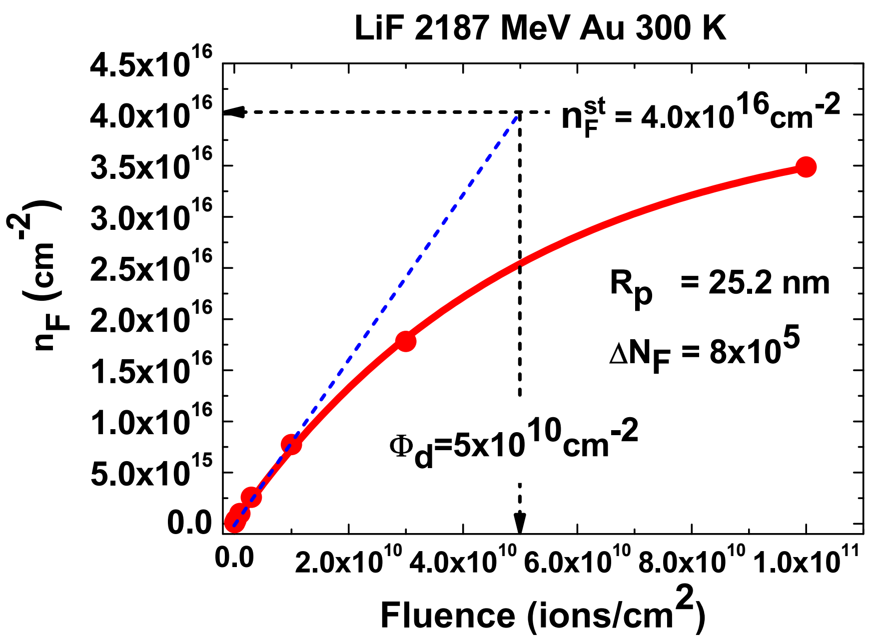


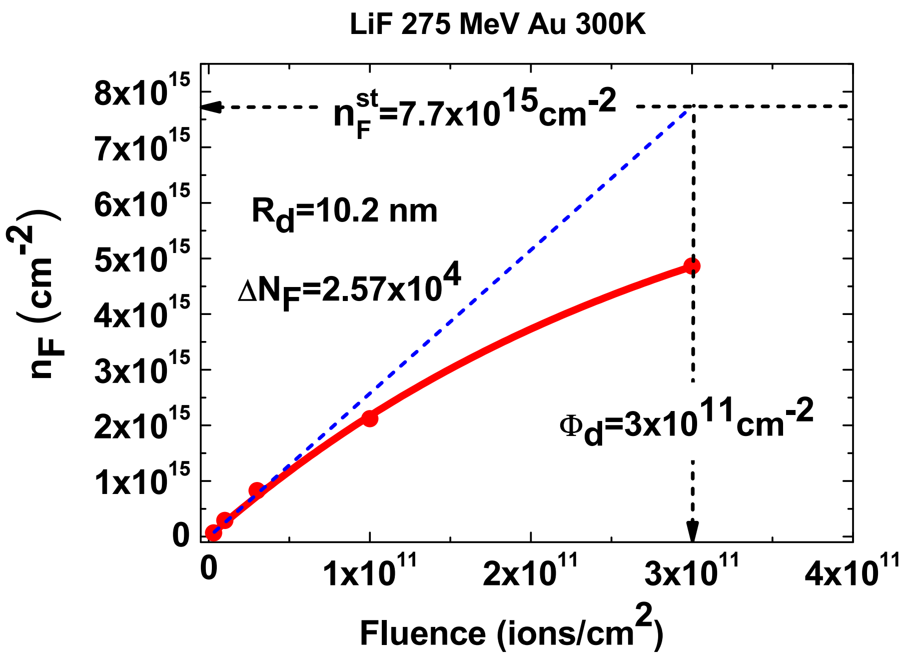


Faster ions produce much larger number of defects spreading at larger distances!

Velocity effect realizes differently for different modes of structure transformations in SHI tracks

Au (272 MeV) vs Au (2187 MeV) in LiF


LiF 2187 MeV Au 300 K


LiF 275 MeV Au 300K

Ion	S_e eV/Å	L µm	R_d nm	N_F	N_F/L cm^{-1}	<C $_F$> cm^{-3}
Au 275 MeV	2463	20.5	10.2	2.6 x10^4	0.13x10^8	2.8x10^{18}
Au 2187 MeV	2293	97.4	25.2	79.8x10^4	0.75x10^8	4.1x10^{18}

1. Size of defect halo is formed by spatial spreading of generated valence holes before their self-trapping

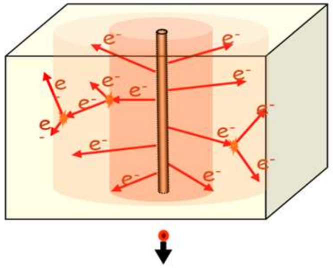
N.A. Medvedev, A.E. Volkov, K. Schwartz, C. Trautmann, Phys. Rev. B. 87 (2013) 104103

2. Larger concentration of self-trapped valence holes in tracks of Au 275 Mev vs Au 2187 MeV

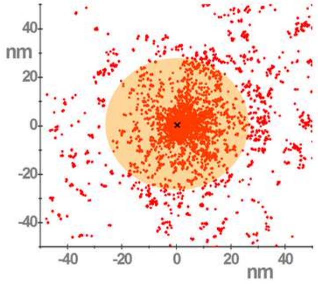
3. Shorter distances trigger intensive recombination of point defects decreasing their number in tracks of slower ions

Monte-Carlo describing electronic kinetics

TREKIS - Time-Resolved Electron Kinetics in SHI Irradiated Solids



$t = 10^{-17}$ s – 10^{-13} s



Collective response of a material is taken into account

$$\frac{\partial^2\sigma}{\partial k\,\partial(\hbar\boldsymbol{\omega})} \sim \frac{1}{n_{sc}}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(\boldsymbol{k},\omega)}\right]$$

1. SHI passage and generation of primary δ-electrons and holes ~10^{-17} s

2. Spreading of electrons and secondary electron cascading ~10^{-16} - 10^{-14} s

3. Auger decays of core holes ~10^{-15} s

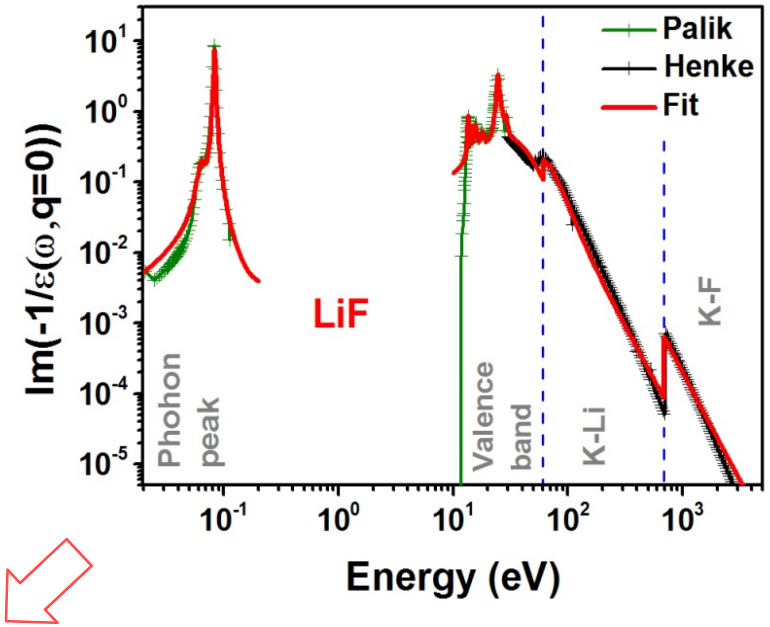
4. Kinetics of all generations of electrons and holes ~10^{-13}s

MC code *TREKIS*

N.A. Medvedev, R.A. Rymzhanov, A.E. Volkov, ***J. Phys. D. Appl. Phys. 48 (2015) 355303***

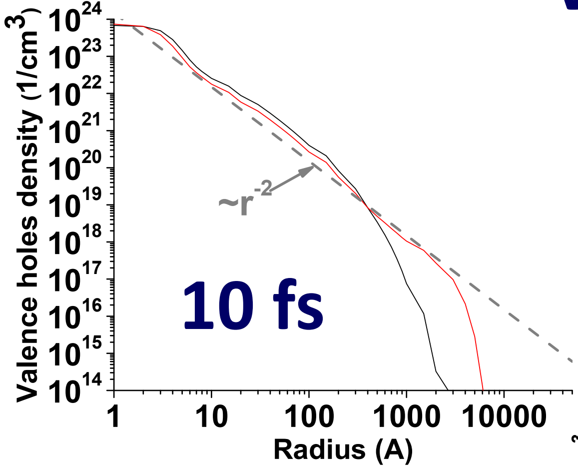
$$\frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} = \frac{2\left(Z_e(v,q)e\right)^2}{n_{sc}\pi\hbar^2 v^2}\frac{1}{\hbar q}\left(1-e^{\frac{\hbar\omega}{k_B T}}\right)^{-1}\mathrm{Im}\left[\frac{-1}{\varepsilon(\omega,q)}\right],$$

Experiment

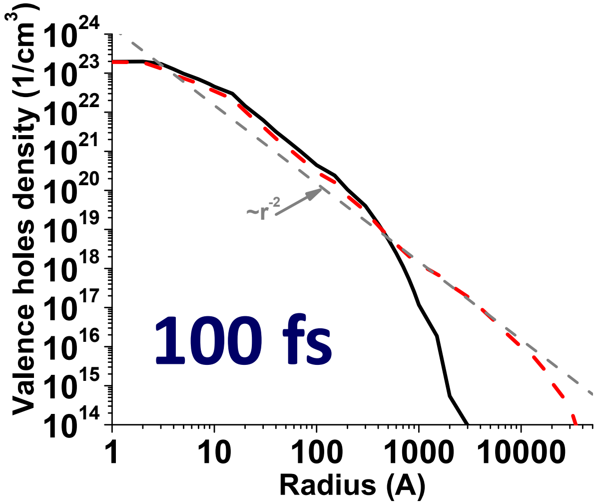


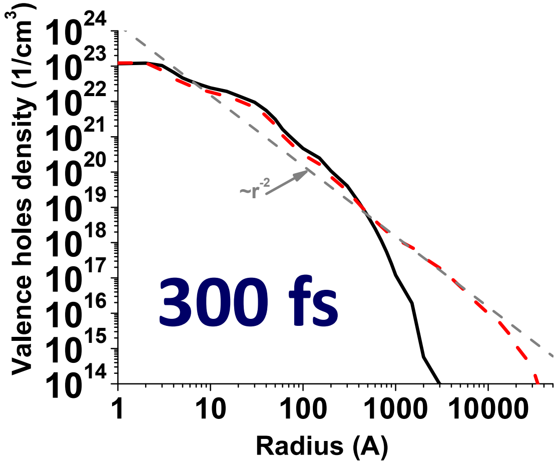
$$\mathrm{Im}\left[\frac{-1}{\varepsilon(\omega,q=0)}\right] = \sum_{i=1}^{N^{OS}} \frac{A_i\gamma_i\hbar\omega}{[\hbar^2\omega^2-(E_{0i})^2]^2+(\gamma_i\hbar\omega)^2}$$

Valence holes density



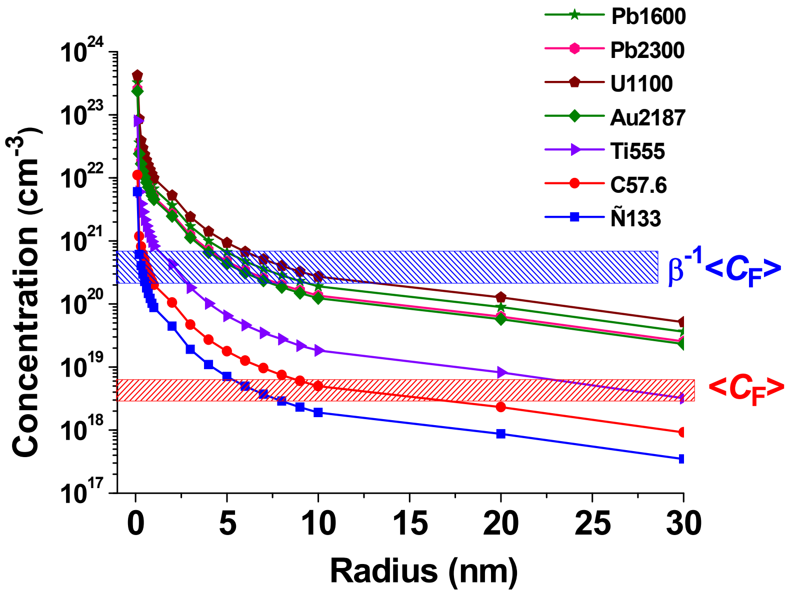
Au 275 MeV vs Au 2187 MeV in LiF





There is no difference at least from *t* > 10fs !

Initial concentrations of holes vs the concentration of defects



Decay of only β≈ 1% of holes results in defect production.

Two orders of magnitude larger initial concentration of valence holes at 10 nm was obtained from MC than that necessary for creation of the observed defects

Diffusion of hot holes before self-trapping (0.5 – 1 ps)

$$D_h = \frac{C_h^o R_d^2}{4\tau_h < C_F >}$$

	Au 275 MeV	Au 2187
D_h , $cm^2 s^{-1}$	0.072	0.45

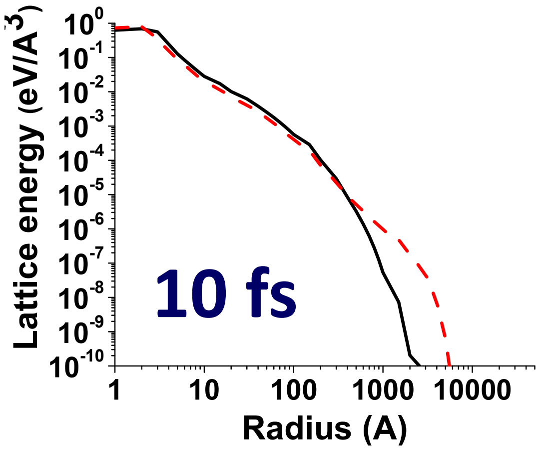
Three possible mechanisms were not taken into account

1. Preheating of lattice due to its interaction with relaxing electronic subsystem

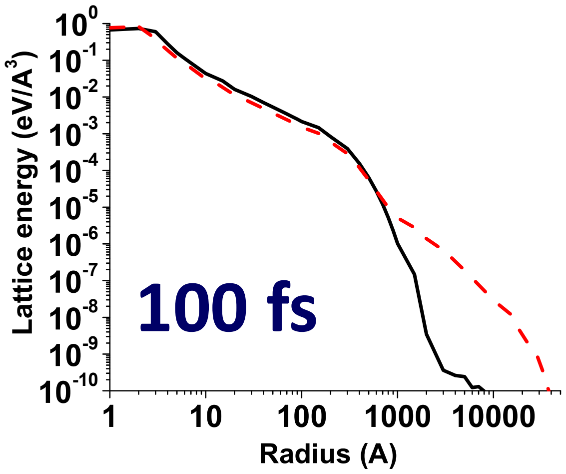
2. Modifications of the interatomic potential stimulated by high excitation of electronic subsystem - nonthermal melting

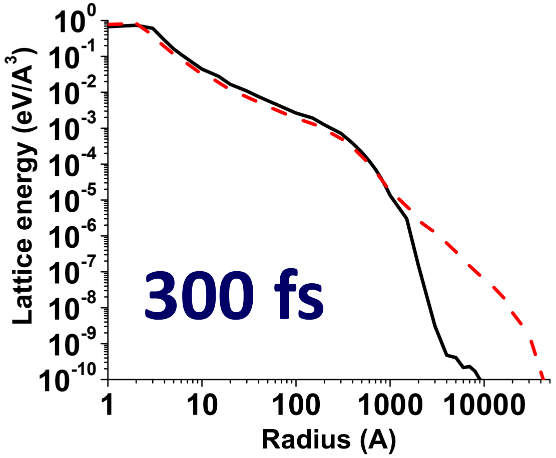
3. Difference in attraction between electrons and holes due to different spectra of electrons generated in tracks of Au 275 MeV and Au 2187 MeV ions

1. Preheating of lattice due to its interaction with relaxing electronic subsystem



Au 275 MeV vs Au 2187 MeV in LiF



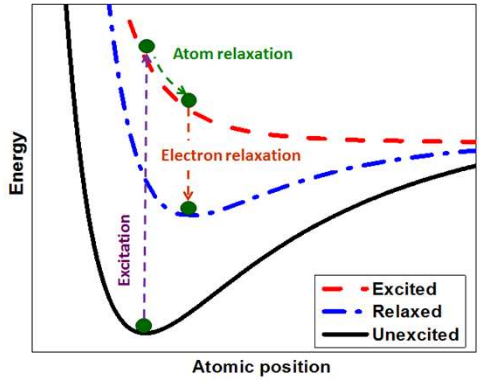


There is no difference at least from *t* > 10fs

Not important process

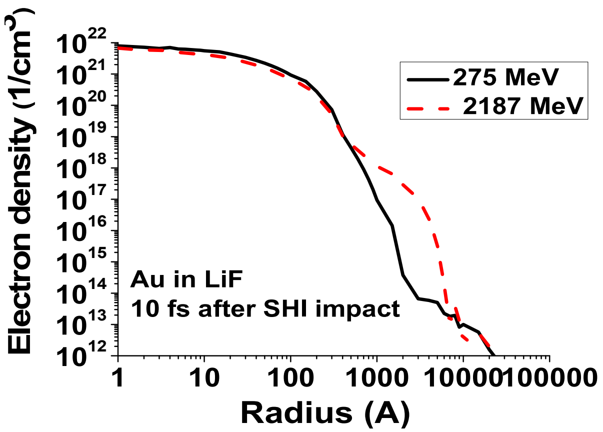
***) Only optical phonons were taken into account**

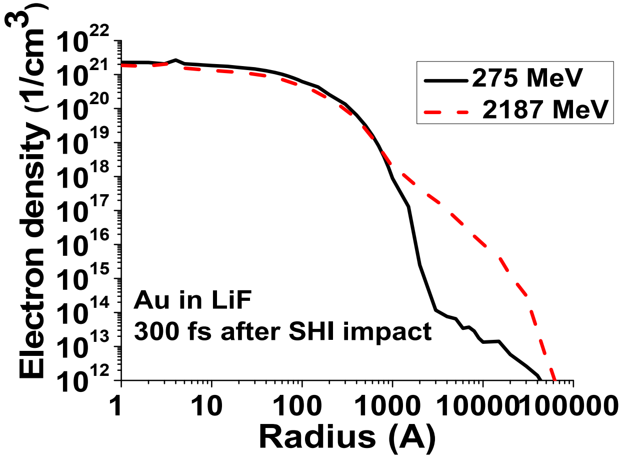
2. Modifications of interatomic potential due to high excitation of electronic subsystem



1. Lattice instability
2. Valence holes become unstable in a new band structure satisfying a new interatomic potential

Density of excited electrons must be above ~ 10^{21} cm^{-3}



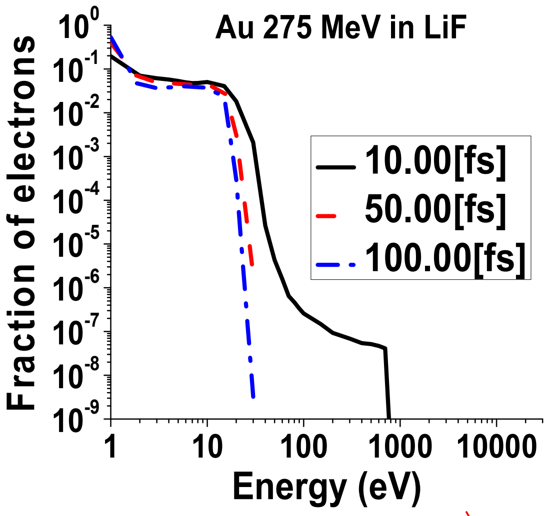


Electron density is too low after 100 fs

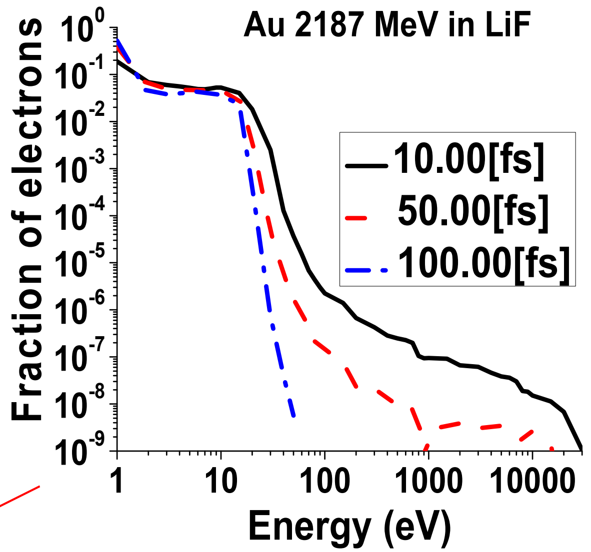
Not important process

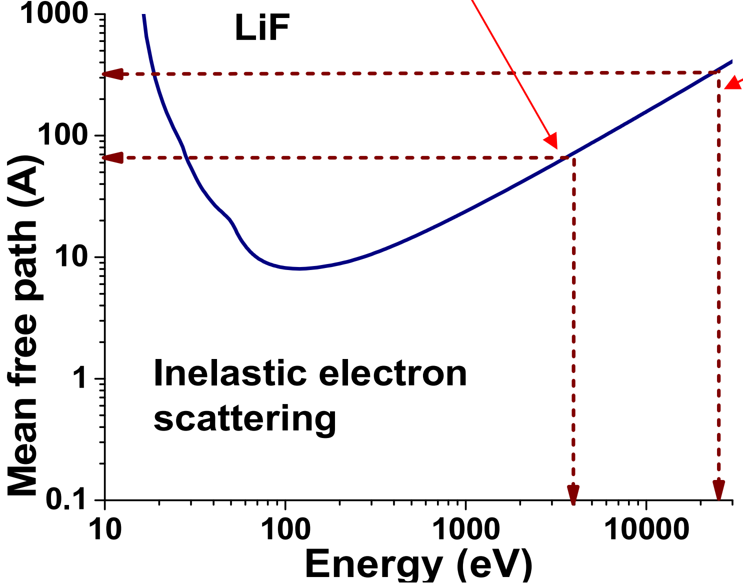
*) interactions between appeared valence holes and electrons where not taken into account

3. Interaction between excited electrons and holes in tracks of Au 275 MeV and Au 2187 MeV ions



different spectra of electrons

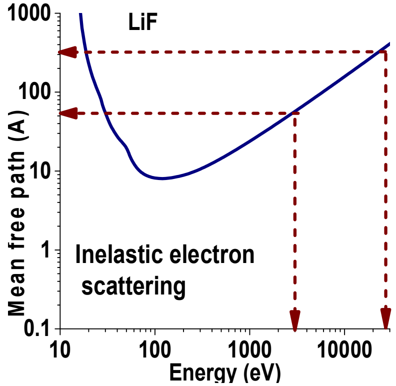




Five times difference in the mean free paths!

Effect of fastest electrons on spatial spreading of valence holes before their self-trapping

1. Au 2187 MeV ion tracks



Ranges of electrons can exceed defect halo radius

⇩

Transient uncompensated positive charge in defect halo

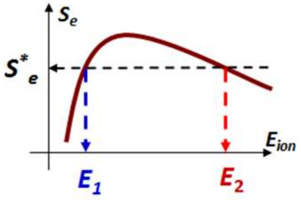
⇩

"Coulomb explosion" in valence holes ensemble

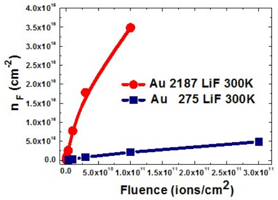
2. Au 275 MeV ions tracks

Fastest electrons cannot leave defect halo and quickly restore local electro-neutrality

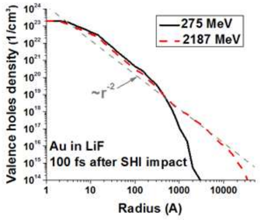
Conclusions



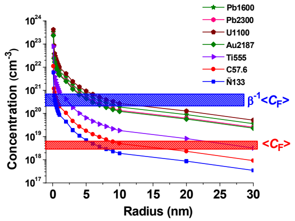
Effect of velocity realizes differently for different modes of structure transformations in SHI tracks:



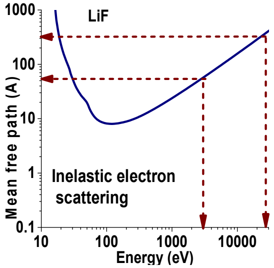
In LiF faster ions produce much larger number of defects !



MC simulations show no difference in radial distributions of valence holes for Au 275 vs Au 2187 tracks from *t* > 10fs !



Six times larger diffusion coefficient in tracks of Au 2187 MeV ions



Three possible mechanisms analyzed:

a) Lattice preheating by electrons
b) Interatomic potential changes due to high electronic excitation
c) Electrons-holes attraction: different spectra of excited electrons

“Coulomb explosion” in the valence holes ensemble

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