THE BREATHER STATISTICS IN CRYSTALS FAR AWAY FROM EQUILIBRIUM

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The statistics of discrete breathes (DBs) formed by thermal spikes (TSs) in solids under irradiation with swift particles is considered, and the corresponding reaction rate amplification factors are derived.

Radiation-induced formation of DBs is shown to change mechanical properties of materials under irradiation, which is confirmed by experimental data.

The reaction rate amplification factor increases strongly with increasing DB lifetime and the maximum DB energy that depend on the system.

The present model shows that the effects due to the crystal anharmonicity are of both fundamental significance and of considerable technological importance in the fields of nuclear engineering and radiation effects.
Relaxation of an initially strongly heated lattice part (thermal spike)  
Ivanchenko, Kanakov, Shalfeev and S. Flach  
Discrete breathers in transient processes and thermal equilibrium  
Radiation-induced thermal spikes (TS)  
[Lifshits, Kaganov, Tanatarov, 1959]

\[ c_e \frac{\partial T_e}{\partial t} = \kappa_e \Delta T_e - \alpha (T_e - T_p) \]

\[ c_p \frac{\partial T_p}{\partial t} = \kappa_p \Delta T_p + \alpha (T_e - T_p) \]

Extremely short lifetimes ~ 1-100 ps
TS -induced reaction rates (LKT-theory)

\[
\langle \dot{R}(T) \rangle_T = \frac{1}{0} \int \dot{R}(\bar{T} + T(W)) dW, \quad W(T) \approx \left( \frac{T^*}{T} \right)^2 \dot{R} = R_0 \exp\left(-\frac{E_a}{k_b T}\right)
\]

\[
(T^*_{e,p})^2 \equiv \frac{1}{16\pi} \left( \frac{\epsilon_{e,p}}{L} \right)^2 \frac{\varphi}{\chi_{e,p} (c_{e,p})^2}
\]

- adiabatic regime [LKT, 1959]

\[
(T^*_{a})^2 \equiv \frac{1}{16\pi} \left( \frac{\epsilon_a}{L} \right)^2 \frac{\varphi}{\chi_a c_a^2}
\]


\[
\langle \dot{R}(T) \rangle_T \rightarrow R_0 \exp\left(-\frac{E_a}{k_b T}\right) + 2R_0 \left(\frac{k_b T^*_a}{E_a}\right)^2
\]
TS -induced formation of DBs

Temperature dependence of the mean concentration of DBs formed at thermal equilibrium and in thermal spikes

\[ \varphi = 10^{14} \text{ ion/cm}^2 \text{s} \quad \varepsilon_e + \varepsilon_p = 1 \text{ MeV} \]

\[ K_B^0 \tau_B^0 = 1 \quad z = 9 \]

\[ E_B^{\text{min}} = 0.2 \text{ eV} \quad E_B^{\text{max}} = 1 \text{ eV} \]

Energy distribution of DBs under irradiation

\[ \langle f_B \rangle_{TS}(E) = 2K_B^0 \tau_B^0 \left( \frac{T_a^*}{T} \right)^2 \left( \frac{E}{E_{\text{min}}} - 1 \right)^z E^{-2} \]

Energy distribution of DBs under thermal equilibrium

\[ f_B(E) = K_B^0 \exp(-E) \tau_B^0 \left( \frac{E}{E_{\text{min}}} - 1 \right)^z , \]
DB-induced reaction rates

\[ \langle \dot{R}(T) \rangle_{B,TS} = \hat{R}_K \int_{E_{\text{min}}}^{E_{\text{max}}} \langle f_B \rangle_{TS}(E)I_0(E)dE = \langle A \rangle_{B,TS} \hat{R}_K \]

**Figure a**
- Thermal equilibrium DBs
- Arrhenius' low

**Figure b**
- Radiation-induced DBs
- Radiation-induced TSs
- Arrhenius' low

\[ R_0 = 10^{13} \text{ s}^{-1}; \quad E_B^{\text{max}} = E_a = 1 \text{ eV} \]
DB -induced amplification factor

\[
\langle \hat{R}(T) \rangle_{B,TS} = \langle A \rangle_B \langle \hat{R}(T) \rangle_{TS}
\]

\[
\langle A \rangle_B = K_B^0 \varepsilon_B^0 \left( \frac{E_a}{E_{B \text{min}}} \right)^2 \exp\left( -\frac{E_a}{k_B T} \right) \left( \int_1^{E_{\text{mod}}/E_{\text{min}}} y^{-2} (y-1)^z I_0(yE_{\text{min}}) \, dy + \int_{E_{\text{mod}}/E_{\text{min}}}^{E_{\text{max}}/E_{\text{min}}} y^{-2} (y-1)^z I_0(yE_r) \, dy \right)
\]

**Graphs:**

- **Graph a:**
  - DB amplification factor vs. DB decay exponent, \( z \)
  - \( E_B^{\text{max}} = E_a = 1 \) eV

- **Graph b:**
  - DB amplification factor vs. DB maximum energy (eV)
  - \( z = 9 \)

**Key Points:****
- Thermal equilibrium vs. Thermal spikes

Radiation-Induced Softening: experiment
Radiation-Induced Softening: theory

\[
\dot{\varepsilon}_T = \dot{\varepsilon}_0 \exp \left\{ - \frac{E_a(\sigma)}{k_b T} \right\} \quad \text{Thermal} \quad \sigma_T(T, \dot{\varepsilon}_\text{ex}) = \sigma_{\text{in}} + \sigma_c \left( 1 - \sqrt{\frac{k_b T}{H_0}} \ln \left( \frac{\dot{\varepsilon}_0}{\dot{\varepsilon}_\text{ex}} \right) \right)
\]

\[
\langle \dot{\varepsilon} \rangle_{TS} \approx 2\dot{\varepsilon}_0 \left( \frac{k_b T_a^*}{E_a(\sigma)} \right)^2 \quad \text{TS} \quad \sigma_{TS}(\varphi, \dot{\varepsilon}_\text{ex}) = \sigma_{\text{in}} + \sigma_c \left( 1 - \left( \frac{2\dot{\varepsilon}_0}{\dot{\varepsilon}_\text{ex}} \right)^{1/4} \sqrt{\frac{k_b T_a(\varphi)}{H_0}} \right)
\]

\[
\langle \dot{\varepsilon} \rangle_{B,TS} = \langle A \rangle_B \langle \dot{\varepsilon} \rangle_{TS} \quad \text{DB} \quad \sigma_{DB}(\varphi, \dot{\varepsilon}_\text{ex}) = \sigma_{\text{in}} + \sigma_c \left( 1 - A_B^0 \left( \frac{2\dot{\varepsilon}_0}{\dot{\varepsilon}_\text{ex}} \right) \left[ \frac{k_b T_a(\varphi)}{H_0} \right]^2 \right)
\]

\[
\langle A \rangle_B \approx A_B^0 E_a^{3/2}
\]

\[
\delta \sigma_{\varphi} \bigg|_{TS} (\varphi, \dot{\varepsilon}_\text{ex}) = M \sigma_c \left( \frac{2\dot{\varepsilon}_0}{\dot{\varepsilon}_\text{ex}} \right)^{1/4} \sqrt{\frac{k_b T_a^*(\varphi)}{H_0}} \propto \rho_d^{1/4} \varphi^{1/4}
\]

\[
T_a^*(\varphi) = \sqrt{\frac{1}{16\pi} \left( \frac{\varepsilon_a}{L} \right)^2 \frac{\varphi}{\chi_a \xi_c^2}}
\]

\[
\delta \sigma_{\varphi} \bigg|_{DB} (\varphi, \dot{\varepsilon}_\text{ex}) = M \sigma_c A_B^0 \left( \frac{2\dot{\varepsilon}_0}{\dot{\varepsilon}_\text{ex}} \right) \left[ \frac{k_b T_a^*(\varphi)}{H_0} \right]^2 \propto \rho_d \varphi
\]
Radiation-Induced Softening: theory vs. experiment
Outstanding problems

1) Time evolution of breathers

\[
\frac{\partial f_B(E,t)}{\partial t} = K_B(E) - \frac{\partial}{\partial E} \left[ f_B(E,t) \frac{dE}{dt} \right] - S_B(E) \quad \frac{dE}{dt} = ?
\]

2) Quodons - high energy mobile longitudinal optical mode DBs that can transfer energy over great distances in atomic-chain directions [Russell, Eilbeck, 2007]

\[
p(E_r) dE_r = \frac{1}{\sqrt{2\pi}\sigma^2} \exp \left( -\frac{(E_r - \langle E_r \rangle)^2}{2\sigma^2} \right) dE_r \quad \text{probability distribution of the reaction potential barrier}
\]

\[
\langle \dot{R}(E_r) \rangle = \int_{-\infty}^{+\infty} \dot{R}(E_r) p(E_r) dE_r = R_0 \exp \left( -\frac{\langle E_r \rangle}{k_B T} \right) \exp \left( \frac{\sigma^2}{2k_B^2 T^2} \right)
\]

\[
\sigma^2 = \langle E_r^2 \rangle - \langle E_r \rangle^2 \quad \text{- dispersion = function (?) of the quodon statistics}
\]
Summary

• Reaction rate theory in solids has been modified taking into account intrinsic localized modes or discrete breathers that can appear in crystals with sufficient anharmonicity resulting in violation of Arrhenius law.
• The reaction rate averaged over large macroscopic volumes and times including a lot of DBs can be increased by many orders of magnitude depending on the DB statistics.
• The breather statistics in thermal equilibrium and under irradiation with swift particles has been considered, and the corresponding reaction rate amplification factors have been derived.
• Radiation-induced formation of DBs changes mechanical properties of materials under reactor conditions as compared to the surveillance samples in out-reactor tests after equivalent irradiation dose, which should be taken into account in forecasting the material service life.