Chemical and nuclear catalysis mediated by the energy localization in crystals and quasicrystals

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Outline

- Localized Anharmonic Vibrations: history and the state of the art
- LAV role in chemical and nuclear catalysis
- MD simulations in crystals and quasicrystalline clusters
In the summer of 1953 Enrico Fermi, John Pasta, Stanislaw Ulam, and Mary Tsingou conducted numerical experiments (i.e. computer simulations) of a vibrating string that included a non-linear term (quadratic in one test, cubic in another, and a piecewise linear approximation to a cubic in a third). They found that the behavior of the system was quite different from what intuition would have led them to expect. Fermi thought that after many iterations, the system would exhibit thermalization, an ergodic behavior in which the influence of the initial modes of vibration fade and the system becomes more or less random with all modes excited more or less equally. Instead, the system exhibited a very complicated quasi-periodic behavior. They published their results in a Los Alamos technical report in 1955.

The FPU paradox was important both in showing the complexity of nonlinear system behavior and the value of computer simulation in analyzing systems.
Localized Anharmonic Vibrations (LAVs)  
A. Ovchinnikov (1969)

Two coupled anharmonic oscillators

\[
\begin{align*}
\ddot{x}_1 + \omega_0^2 x_1 + \varepsilon \lambda x_1^3 &= \varepsilon \beta x_2 \\
\ddot{x}_2 + \omega_0^2 x_2 + \varepsilon \lambda x_2^3 &= \varepsilon \beta x_1
\end{align*}
\]

Localization condition

\[
A_0 > \frac{4 \beta}{3 \lambda} \implies \tau \to \infty
\]

Phase diagram
**Sine-Gordon** standing breather is a swinging in time coupled kink-antikink 2-soliton solution.

**Large amplitude moving sine-Gordon breather.**
1D crystal — Hirota lattice model
(nonlinear telegraph equations, 1973)

Equation of motion of Hirota lattice

\[
\frac{m\ddot{u}_n}{1 + \frac{\pi^2}{4} \frac{\dot{u}_n^2}{s^2}} = \frac{2}{\pi}\gamma d_0 \left\{ \tan \left[ \frac{\pi}{2} \left( \frac{u_{n-1} - u_n}{d_0} \right) \right] - \tan \left[ \frac{\pi}{2} \left( \frac{u_n - u_{n+1}}{d_0} \right) \right] \right\}
\]

\[
H = \left( \frac{2}{\pi} \right)^2 ms^2 \sum_{n=-\infty}^{+\infty} \left\{ \frac{1}{2} \ln \left[ 1 + \tan^2 \left( \frac{\pi}{2} \frac{p_n}{ms} \right) \right] + \frac{1}{2} \ln \left[ 1 + \tan^2 \left( \frac{\pi}{2d_0} \left( u_{n-1} - u_n \right) \right) \right] \right\}
\]

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Standing weakly localized DB

Bogdan, 2002

Standing strongly localized DB
\[ u_n^{(b)} = \frac{2d_0}{\pi} \arctg \left[ \frac{\sh\left(\kappa d_0/2\right) \cos\left(knd_0 - \omega t\right)}{\sin\left(kd_0/2\right) \ch\kappa\left(nd_0 - Vt\right)} \right], \]

\[ \omega = \frac{s}{d_0} 2 \ch\left(\frac{\kappa d_0}{2}\right) \sin\left(\frac{kd_0}{2}\right), \quad V = \frac{s}{\kappa d_0/2} \cos\left(\frac{kd_0}{2}\right). \]

Bogdan, 2002

Moving strongly localized DB
The concept of LAV in regular lattices is based on *large anharmonic* atomic oscillations in Discrete Breathers excited outside the phonon bands.

Existence of breathers (1994)

Nonlinear coupled oscillators

\[ V = \sum V(X_n) + C W(X_n - X_{n+1}) \]

- Exact, periodic and localized solution

Phonons

- Frequency band \( \omega_{ph}^2 = \omega_0^2 + 4C \sin^2 q/2 \)
- Non localized states

\[ \omega_0 \]

\[ \omega = 0 \]
FIG. 1. The pair potential $V(r)$ of Ni (solid line) and its approximation by the fourth-order polynomial (dashed line). The inset shows an expanded view.

The distance between the nearest atoms in Ni at room temperature is $r_0 = 2.49$ Å and longitudinal sound velocity is $v_l = 5266$ m/s. These values give $\tilde{K}_2 = 2.75$ eV/Å$^2$ (as expected, $\tilde{K}_2 > K_2$) and $\tilde{\kappa} \approx 1.2$. The distance $r_0$ increases.

FIG. 2. Phonon density of states and three ILM spectral signatures for Ni. Phonon spectrum (dashed line) and spectrograms (solid line) of the different ILM's: The frequencies are 5.58, 5.86, and 6.07 ($10^{13}$ rad/s) and the amplitudes of vibrations of the central bond are 0.18, 0.31, and 0.42 Å.
Standing DB in bcc Fe: $d_0=0.3$ Å
D. Terentyev, V. Dubinko, A. Dubinko (2013)
Moving DB in bcc Fe: $d_0=0.4 \, \text{Å}, \, E=0.3 \, \text{eV}$

D. Terentyev, V. Dubinko, A. Dubinko (2013)
Visualization of the PdH fcc Lattice (NaCl type)
Visualization of the PdH fcc Lattice Oscillations at T=100 K
Visualization of the PdH fcc Lattice Oscillations at T=1000K
Gap breathers in NaCl type lattices, Dmitriev et al. (2010)

NaCl-type $M_H/M_L = 10$ at temperatures $T = (a) 0$, (b) 155, (c) 310, and (d) 620 K.

Phonon Gap

DOS for $\text{PdD}_{0.63}$ and $\text{PdH}_{0.63}$: $M_H/M_L = 50$; 100 GPa pressure at $T=600$ K.

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MD modeling of gap DBs in diatomic crystals at elevated temperatures

$A_3B$ type crystals $M_H / M_L = 10$

In NaI and KI crystals Hizhnyakov et al has shown that DB amplitudes along $<111>$ directions can be as high as 1 Å, and $t^*/\Theta \sim 10^4$

Lifetime and concentration of high-energy light atoms increase exponentially with increasing $T$

$K_{B,n}/\bar{K} \approx 5.1$

$t^*/\Theta \approx 70$  $\bar{K} = 0.1eV \geq 1000K$
MD modeling of gap DBs in diatomic crystals at elevated temperatures

A$_3$B type crystals, Kistanov, Dmitriev (2014),

A$_3$B compound based on fcc lattice with Morse interatomic potentials. Grey atoms are **50 times lighter** than yellow (similar to the PdD crystal).

DB is localized on a **single light atom** vibrating along <100> direction with the frequency of **227 THz**, which is **inside the phonon gap**. Shown is the x-displacement of the light atom as the function of time. DB has very large amplitude of **0.4 angstrom**, which should be compared to the lattice parameter a=1.35 angstrom.
LAV effect (1): periodic in time modulation of the potential barrier height
Reaction-rate theory with account of the crystal anharmonicity

\[ U(x,t) = U(x) - (V \cdot x/x_m) \cos(\Omega t) \]

Kramers rate is amplified:
Bessel function \( I_0 \left( \frac{V_m}{k_B T} \right) \)
How extend LAV concept to include Quantum effects, Tunneling?

The Kramers theory is extended in order to take into account the action of the thermal and zero-point oscillation (ZPO) energy.

\[ R_K = \frac{\omega_0}{2\pi} \exp \left[-\frac{E_0}{D(T)}\right] \]

\[ D(T) = E_{ZPO} \coth \left(\frac{E_{ZPO}}{k_B T}\right) \approx \begin{cases} E_{ZPO}, & T \rightarrow 0 \\ k_B T, & T \gg \frac{E_{ZPO}}{k_B} \end{cases} \]

T – temperature is a measure of thermal noise strength

\[ E_{ZPO} = \frac{\hbar \omega_0}{2} \] - ZPO energy is a measure of quantum noise strength
When we heat the system we increase temperature, i.e. we increase the *thermal* noise strength

Can we increase the *quantum* noise strength, i.e. ZPO energy?
Stationary harmonic potential

\[ \langle E \rangle_n = \hbar \omega_0 \left( n + \frac{1}{2} \right) \]

\( E_{ZPO} = \frac{\hbar \omega_0}{2} \)
Time-periodic modulation of the **double-well** shape changes (i) eigenfrequency and (ii) position of the wells.
Quasi-energy in time-periodic systems

Consider the Hamiltonian which is periodic in time.

\[ i\hbar \frac{\partial \psi}{\partial t} = \hat{H} \psi \]

\[ \hat{H}(t + T) = \hat{H}(t) \]

It can be shown that Schrodinger equation has class of solutions in the form:

\[ \psi_\alpha(t + T) = \exp(-i\alpha) \psi_\alpha(t) \]

where \[ \alpha = \frac{\hbar \omega}{T} \]

Is the quasi-energy

\[ i\hbar \frac{\partial}{\partial t} \psi(x,t) = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial x^2} \psi(x,t) + \frac{m\omega^2(t)}{2} x^2 \psi(x,t) \]

\[ \epsilon_n = \left(n + \frac{1}{2}\right) \lambda(\omega(t)) \]

Time-periodic driving of the harmonic oscillator with non resonant frequencies \( \Omega \neq 2\omega_0 \) renormalizes its energy spectrum, which remains equidistant, but the quasi-energy quantum \( \lambda(\omega(t)) \) becomes a function of the driving frequency.
Time-periodic modulation of the double-well shape changes (i) eigenfrequency and (ii) position of the wells
DB frequency and eigenfrequency of the potential wells of neighboring D ions in PdD (Dubinko, ICCF 19)

DB polarized along the close-packed D-D direction $<110>$
Parametric resonance with time-periodic eigenfrequency $\Omega = 2\omega_0$

\[
i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m} \frac{\partial^2 \psi}{\partial x^2} + \frac{m\omega^2(t)}{2} x^2 \psi
\]

Schrödinger equation

\[
\psi(x_0, t_0 = 0) = \frac{1}{\sqrt{4\pi\sigma_0}} \exp\left(-\frac{x_0^2}{4\sigma_0}\right)
\]

Initial Gaussian packet $\sigma_0 = \frac{\hbar}{2m\omega_0}$

Parametric regime $\Omega = 2\omega_0$:

\[
\ddot{x} + \omega_0^2 \left[1 - g \cos(2\omega_0 t)\right] x = 0
\]

g $\ll$ 1 – modulation amplitude

\[
\sigma_x(t) = \sigma_0 \cosh\left(\frac{g\omega_0 t}{2}\right) \left[1 + \tanh\left(\frac{g\omega_0 t}{2}\right) \sin(2\omega_0 t)\right]
\]

dispersion

ZPO energy:

\[
E_{ZPO}(t) = \frac{\hbar \omega_0}{2} \cosh \frac{g\omega_0 t}{2}
\]

ZPO amplitude:

\[
\Lambda_{ZPO}(t) = \sqrt{\frac{\hbar}{2m\omega_0} \cosh \frac{g\omega_0 t}{2}}
\]
Non-stationary harmonic potential with time-periodic eigenfrequency $\Omega = 2\omega_0$

\[ \Lambda_{ZPO}(t) = \sqrt{\frac{\hbar}{2m\omega_0}} \cosh \frac{g\omega_0 t}{2} \]

\[ E_{ZPO}(t) = \frac{\hbar \omega_0}{2} \cosh \frac{g\omega_0 t}{2} \]
\[ \langle E \rangle_{\text{theor}}(t) \approx \hbar \omega_0 \left( n + \frac{1}{2} \right) \cosh \frac{g \omega_0 t}{2} \]

\[ \langle E \rangle_{\text{num}}(t) = \frac{\hbar \omega_0}{2} \left( n + \frac{1}{2} \right) \left[ \frac{\dot{Y}^2 + \omega_0^2 \dot{Z}^2}{\omega_0^2} + \frac{\omega^2(t)}{\omega_0^2} \left( Y^2 + \omega_0^2 Z^2 \right) \right] \]

\[ \omega^2(t) = \omega_0^2 \left[ 1 - g \cos(2\omega_0 t) \right] \]

\[ g = 0.1, \quad n = 0 \]

General case: \( n = 0,1,2, \ldots \)
\[ \frac{\langle E \rangle_n}{\hbar \omega_0} \]

\[ g = 0.1 \]
Non-stationary harmonic potential with time-periodic shifting of the well position at $\Omega = \omega_0$

$$\langle E \rangle = \frac{\hbar \omega_0}{2} + \left( g_A A_{ZPO} \right)^2 m \omega_0^2 \left[ \omega_0^2 t^2 + \omega_0 t \sin 2\omega_0 t + \sin^2 \omega_0 t \right]$$

$$\lambda(t) = \frac{g_A A_{ZPO}}{2} \omega_0 t \left( \cos \omega_0 t - \frac{\sin \omega_0 t}{\omega_0 t} \right)$$
Well-known and well-forgotten quantum mechanics


Uncertainty Relations (UR)
Heisenberg (1927)

\[ \Delta x \Delta p \geq \frac{\hbar}{2} \]


Generalization of the UR
Schrödinger (1930); Robertson (1930)

\[ \sigma_x \sigma_p - \sigma_{xp}^2 \geq \frac{\hbar^2}{4}, \]

\[ \sigma_x = \langle (x - \langle x \rangle)^2 \rangle \]
\[ \sigma_p = \langle (p - \langle p \rangle)^2 \rangle \]
\[ \sigma_{xp} = \langle \hat{x}\hat{p} + \hat{p}\hat{x} \rangle/2 - \langle x \rangle\langle p \rangle. \]

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Correlator
Correlation coefficient

\[ \sigma_x \sigma_p \geq \frac{\hbar^2}{4(1 - r^2)}, \]
\[ r = \frac{\sigma_{xp}}{\sqrt{\sigma_x \sigma_p}} \]

Effective Plank constant

\[ \hbar_{ef} = \frac{\hbar}{\sqrt{1 - r^2}}. \]

Can CORRELATIONS make the barrier transparent ?!


\[ G_{ef} \approx \exp \left\{ - \frac{2}{\hbar_{ef}} \int_{R_0}^{R_c} dr \sqrt{2\mu (V(r) - E)} \right\} \]
\[ \Downarrow \quad r \to 1, \hbar_{ef} \to \infty \quad 1 \]
Correlations Coefficient for the parametric resonance $\Omega = 2\omega_0$

$$r_{xp} = r_{xp}(t) = \frac{\sinh\left(\frac{g\omega_0 t}{2}\right)\cos(2\omega_0 t)}{\sqrt{1 + \left[\sinh\left(\frac{g\omega_0 t}{2}\right)\cos(2\omega_0 t)\right]^2}} + O(g)$$

$g = 0.1$

$$T = \frac{2\pi}{2\omega_0}$$
**Tunneling:** Numerical solution of Schrödinger equation

Stationary: $t_{\text{Kramers}} \sim 10^5$ cycles at $V_{\text{barrier}} = 12E_0$

Time-periodically driven: $\Omega = 1.5 \omega_0$, $g = 0.2$
Extreme example – Low Energy Nuclear Reactions (LENR)
Why LENR is unbelievable?


\[ G \approx \exp \left\{ -\frac{2}{\hbar} \int_{r_0}^{R_c} dr \sqrt{2\mu(V(r) - E)} \right\} \quad \text{Gamow factor} \]

\[ r_0 \sim 3 \text{ fm} \quad \text{Nuclear radius deduced from scattering experiments} \]

\[ V(R_0) = \frac{e^2}{r_0} \approx 450 \text{ keV} \quad \text{Coulomb barrier} \]

At any crystal Temperature:

\[ E \ll V(r_0) \Rightarrow G \approx 10^{-2760} \]

\textbf{However, is} the Coulomb barrier that huge in the lattice?
Willis Eugene *Lamb*
Nobel Prize 1955

Julian Schwinger
Nobel Prize 1965

*Electron screening*

J. Schwinger, **Nuclear Energy in an Atomic Lattice** (1990)
*Lattice screening*
Effective Coulomb repulsion with account of zero-point oscillations

\[ 0 \langle V_c (r) \rangle_0 = \frac{e^2}{r} \sqrt{\frac{2}{\pi}} \int_0^r dx \exp \left( -\frac{1}{2} x^2 \right) \approx \begin{cases} r \gg \Lambda_0 : \frac{e^2}{r} \\ r \ll \Lambda_0 : \left( \frac{2}{\pi} \right)^{1/2} \frac{e^2}{\Lambda_0} \end{cases} \sim 100 \text{ eV (!!!)} \]

\[ T_0 \] is the mean lifetime of the **phonon vacuum** state before releasing the nuclear energy **directly** to the lattice (**no radiation!**):

\[
D-D \text{ fusion rate in Pd-D lattice: } \nu_{D-D} = \frac{1}{T_0} = \left( \frac{2\pi/\hbar}{\mathcal{E}} \right)_0 \langle V \delta(H - E)V \rangle_0
\]

\[
\frac{1}{T_0} \approx 2\pi\omega_0 \left( \frac{2\pi\hbar\omega_0}{E_{nucl}} \right)^2 \left( \frac{r_{nucl}}{\Lambda_0} \right)^3 \exp \left[ -\frac{1}{2} \left( \frac{R_0}{\Lambda_0} \right)^2 \right] \sim 10^{-19} \text{ s}^{-1} \div 10^{-30} \text{ s}^{-1}
\]

\[ \Lambda_0 = 0.1A \quad R_0 = 0.94A \div 2.9A \]


\[ V_{\text{eff}}(r) \approx \frac{m\omega_0^2}{2} r^2 + \frac{e^2}{R_0 - r} \exp \left( -\frac{R_0 - r}{\lambda_D} \right) \sqrt{\frac{2}{\pi}} \int_0^{R_0 - r} dx \exp \left( -\frac{1}{2} x^2 \right) \]

Dubinko, Laptev, *Chemical and nuclear catalysis driven by LAVs*, LetMat (2016)

\[
\frac{1}{T} \approx 2\pi \omega_0 \left( \frac{2\pi \hbar \omega_0}{E_{\text{nucl}}} \right)^{1/2} \left( \frac{r_{\text{nucl}}}{\Lambda} \right)^3 \exp \left[ -\frac{1}{2} \left( \frac{R_0}{\Lambda_0} \right)^2 \right]
\]

\[
\Lambda_0 = \sqrt[4]{\frac{\hbar}{2m\omega_0}} = \text{const} \quad \text{and} \quad \sqrt{2m\omega_0} \cosh \frac{g\omega_0 t}{2}
\]
LENR power density under $\text{D}_2\text{O}$ electrolysis

$$P_{\text{D-D}}(T, J) = K_{\text{DB}}^J \left( E_{\text{DB}}^*, T, J \right) E_{\text{D-D}}$$

Table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>D-D equilibrium spacing in PdD, b (Å)</td>
<td>2.9</td>
</tr>
<tr>
<td>DB excitation efficiency, $k_{\text{eff}}$</td>
<td>$10^{-10}$</td>
</tr>
<tr>
<td>Fusion energy, (MeV)</td>
<td>23.8</td>
</tr>
<tr>
<td>Mean DB energy, (eV)</td>
<td>1</td>
</tr>
<tr>
<td>DB oscillation frequency, $\omega_{DB}$ (THz)</td>
<td>20</td>
</tr>
<tr>
<td>Critical DB lifetime, $\tau_{DB}$ (ps/cycles)</td>
<td>10/100</td>
</tr>
<tr>
<td>Quodon excitation energy (eV)</td>
<td>0.8</td>
</tr>
<tr>
<td>Quodon excitation time, $\tau_{ex}$ (ps/cycles)</td>
<td>1/10</td>
</tr>
<tr>
<td>Quodon propagation range, $l_q$ (nm)</td>
<td>2.9</td>
</tr>
<tr>
<td>Cathod size/thickness (mm)</td>
<td>5</td>
</tr>
</tbody>
</table>

BNC can provide up to $10^{14}$ “collisions” per cm$^3$ per second

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Where to look for Nuclear Active Environment?
Small Energy Gap is required for LAV formation

Nuclear Active Environment
“Cracks and small particles are the Yin and Yang of the cold fusion environment” E. Storms

Structure of dimeric citrate synthase (PDB code 1IXE). Only $\alpha$-carbons are shown, as spheres in a color scale corresponding to the crystallographic B-factors, from smaller (blue) to larger (red) fluctuations [Dubinko, Piazza, 2014]
Atomic configuration of a Ni nanoparticle of 2899 atoms at $T = 1000$ K. The atoms are colored based on the potential energy and their size is proportional to Debye–Waller factor. Potential energy and DWF are time averaged over a 130 ps time window, corresponding to the time interval during which the strings show maximum length.

Map of the local Debye–Waller factor showing the heterogeneity of the atomic mobility at a temperature of 1450 K. Regions of high mobility string-like motion are concentrated in filamentary grain boundary like domains that separate regions having relatively strong short-range order.
STEM images of LAVs of the decagonal Al\textsubscript{72}Ni\textsubscript{20}Co\textsubscript{8} at (a) 300 K and (b) 1100 K, according to Abe et al. Connecting the center of the 2 nm decagonal clusters (red) reveals significant temperature-dependent contrast changes, a pentagonal quasiperiodic lattice (yellow) with an edge length of 2 nm can be seen in (b).
(a) **LAV amplitude** dependence on temperature in Al$_{72}$Ni$_{20}$Co$_8$, fitted by two points at 300 K and 1100 K, according to Abe et al. The maximum LAV amplitude at 1100K = 0.018 nm.

(b) **LAVs give rise to phasons** at T > 990 K, where a phase transition occurs, and additional quasi-stable sites β arise near the sites α. The phason amplitude of 0.095 nm is an order of magnitude larger than that of LAVs.
(a) Structure of Pd-H cluster containing 147 Pd and 138 H atoms having minimum free energy configuration, replicated using the method and parameters by Calvo et al; (b) H-H-H chains in the nanocluster, which are viable sites for LAV excitation
Magic clusters are clusters of certain ("magic") sizes, which, due to their specific structure, have increased stability compared to clusters of other sizes.

In icosahedral clusters, each “k” layer consists of $10k^2+2$ atoms. So the total number of atoms in a cluster with “N” layers is given by

$$n = (2N + 1) + 10 \sum_{k=1}^{N} k^2$$

$$n = 13, 55, 147, 309, 561$$ for $N=1,2,3,4,5$
Magic icosahedral cluster of 55 Pd atoms

Consider a cluster of 55 Pd atoms with quasicrystalline 5th order symmetry axis.
**Initial conditions:**
at the initial time moment all particles have zero displacements from equilibrium positions.

Atom #1 has initial kinetic energy 1.5 eV in [00-1] direction.

Atom #12 has initial kinetic energy 1.5 eV in [001] direction

**Boundary conditions:** free surfaces of cluster
It is seen from the visualization, that Localized Anharmonic Vibration is generated. The observed LAV in the atomic cluster represents the coherent collective oscillations of Pd atoms along quasi-crystalline symmetry directions.
If the initial energy, given to cluster is large enough (greater then the cohesive energy) then the cluster is destroyed after a certain period of time (~ ps).
Conclusions and outlook

New mechanism of chemical and nuclear catalysis in solids is proposed, based on time-periodic driving of the potential landscape induced by emerging nonlinear phenomena, such as LAVs or phasons. The present mechanism explains the salient LENR requirements: (i, ii) long initiation time and high loading of D within the Pd lattice as preconditioning needed to prepare small PdD crystals, in which DBs can be excited more easily, and (iii, iv) the triggering by D flux or electric current, which facilitates the DB creation by the input energy transformed into the lattice vibrations. The model (under selected set of material parameters) describes quantitatively the observed exponential dependence on temperature and linear dependence on the electric (or ion) current. Atomistic modeling of LAVs and phasons in metal hydrides/deuterides is an important outstanding problem since it may offer ways of engineering the nuclear active environment.
Publications

8. V.I. Dubinko, O.M. Bovda, O.E. Dmitrenko, V.M. Borysenko, I.V. Kolodiy, Peculiarities of hydrogen absorption by melt spun amorphous alloys Nd$_{90}$Fe$_{10}$, *Vestink KhNU* (2016).
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