High Entropy Alloys, Fractional Calculus, and Anomalous Diffusion

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ABOUT ME

Major: Physics

Goals: In the long term I would like to pursue a position in academia or a National Lab. In the short term I would like to go to graduate school in physics, studying nonequilibrium statistical physics and QFT.

Why LLNL? Scientists at LLNL are national leaders in nonequilibrium statistical physics. I enjoy being in places with many experts in diverse fields and LLNL is such a place.

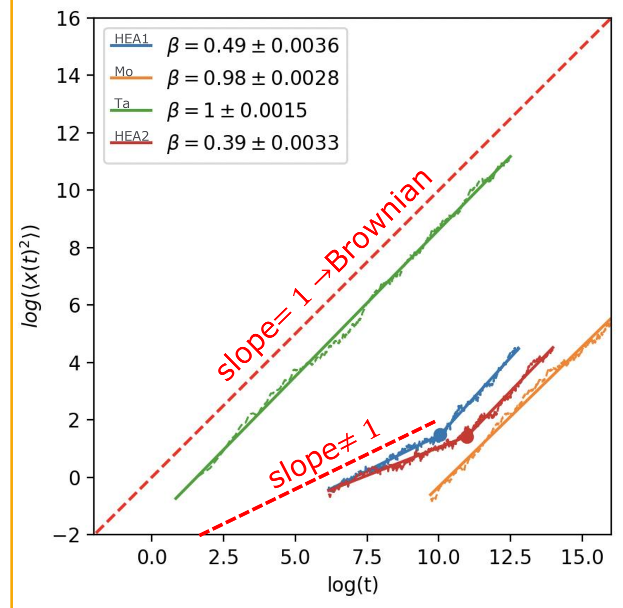
What else do I do? In my spare time I build cyclotrons &c. See: thecyclotronproject.github.io

SUMMER RESEARCH

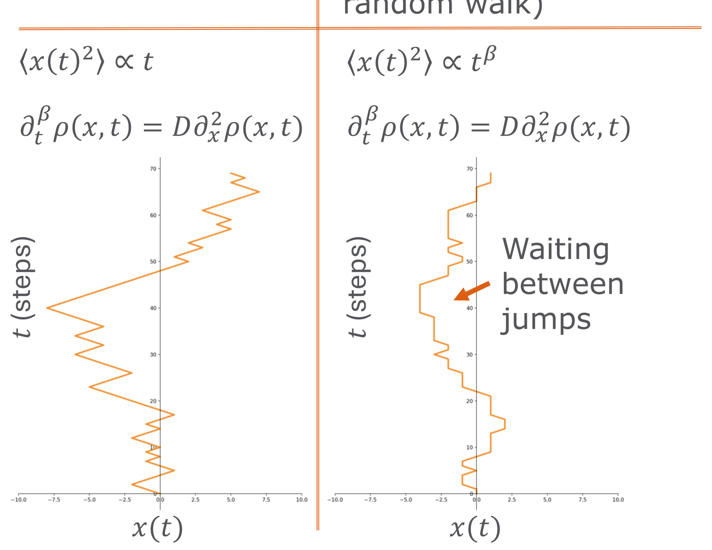
Abstract

Diffusion of lattice vacancies though high entropy alloys (HEAs) is modeled using a time fractional diffusion equation. This will be used to build a first passage kinetic Monte Carlo (FPKMC) simulation to optimize material properties of fusion reactor first walls.

Random Walks



Brownian motion (random walk)	Time-changed Brownian motion (continuous time random walk)
$\langle x(t)^2 \rangle \propto t$	$\langle x(t)^2 \rangle \propto t^\beta$
$\partial_t^\beta \rho(x,t) = D\partial_x^2 \rho(x,t)$	$\partial_t^\beta \rho(x,t) = D\partial_x^2 \rho(x,t)$



Ergodicity Time

$$\langle T_{ergodic} \rangle = \langle J_i \rangle \sum_{i=1}^{N_{cfg}} \exp\{\beta(E_{max} - E_i)\}$$

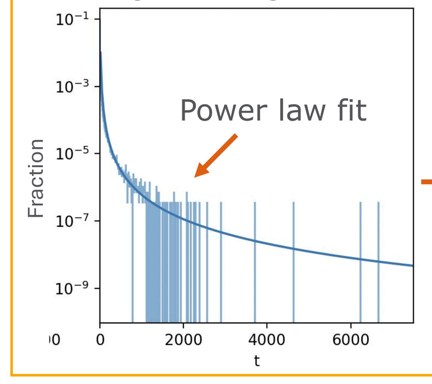
Number of local environments

Mean waiting time

Difference in site occupation energy

Fully resolved lattice kinetic Monte Carlo code demonstrates the abnormality of diffusion in HEAs. Scaling at a rate less than t (until $T_{ergodic}$) the mean-squared displacement (MSD) is consistent with an ensemble of continuous time random walks.

After $T_{ergodic}$, the vacancies have explored a representative fraction of their energy landscape and enter the ergodic regime, with MSD scaling like t.



CTRWs with power law waiting times are modeled in the statistical limit by time-fractional diffusion equations.

Fractional Diffusion Equation

Caputo Derivative

$$\partial_x f(x) = \lim_{h\to 0} \frac{\Delta f(x)}{h} \qquad \Delta f(x) = f(x) - f(x-h)$$

$$\partial_x^2 f(x) = \lim_{h\to 0} \frac{\Delta^2 f(x)}{h^2} \qquad \Delta^2 f(x) = f(x) - 2f(x-h) + f(x-2h)$$

$$\vdots$$

$$\partial_x^n f(x) = \lim_{h\to 0} \frac{\Delta^n f(x)}{h^n} \qquad \Delta^n f(x) = \sum_{m=0}^{\infty} (-1)^m \binom{n}{m} f(x-mh)$$

$$\beta \notin \mathbb{Z} \qquad \vdots$$

$$\partial_x^\beta f(x) = \lim_{h\to 0} \frac{\Delta^\beta f(x)}{h^n} \qquad \Delta^\beta f(x) = \sum_{m=0}^{\infty} (-1)^m \binom{\beta}{m} f(x-mh)$$

$$\binom{\beta}{m} \longrightarrow \frac{\Gamma(\beta+1)}{\Gamma(m+1)\Gamma(\beta-m+1)}$$ By analytic continuation $\quad n! \to \Gamma(n+1)$

Fundamental solution

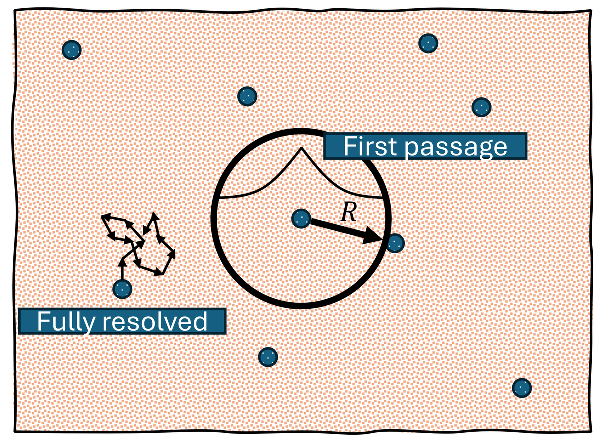
$$\partial_t^\beta \rho(x,t) = D\,\partial_x^2 \rho(x,t) \xrightarrow{\boxed{\mathcal{L} \circ \mathcal{F}}} s^\beta \bar{\rho}(k,s) - s^{\beta-1} = -Dk^2 \bar{\rho}(k,s)$$

$$\bar{\rho}(k,s) = \frac{s^{\beta-1}}{s^\beta + Dk^2} \xrightarrow{\boxed{\mathcal{F}^{-1} \circ \mathcal{L}^{-1}}} \rho(x,t) = \frac{1}{2\sqrt{Dt^\beta}} M_{\frac{\beta}{2}}\left(\frac{|x|}{\sqrt{Dt^\beta}}\right)$$

$$\underbrace{M_{\frac{\beta}{2}}(z) = \sum_{j=0}^{\infty} \frac{(-z)^j}{j!\,\Gamma(-\frac{\beta}{2}(j+1)+1)}}_{\text{M-Wright Function}}$$

The M-wright function is a generalized Gaussian distribution and the Green's function for the time fractional diffusion equation (i.e. the field response to a δ –function source).

First Passage Kinetic Monte Carlo



Fully resolved Monte Carlo simulations address thermal fluctuations by a series of stochastic "jumps." This requires a simulation step on the thermal timescale of the system. Long simulations are therefore difficult.

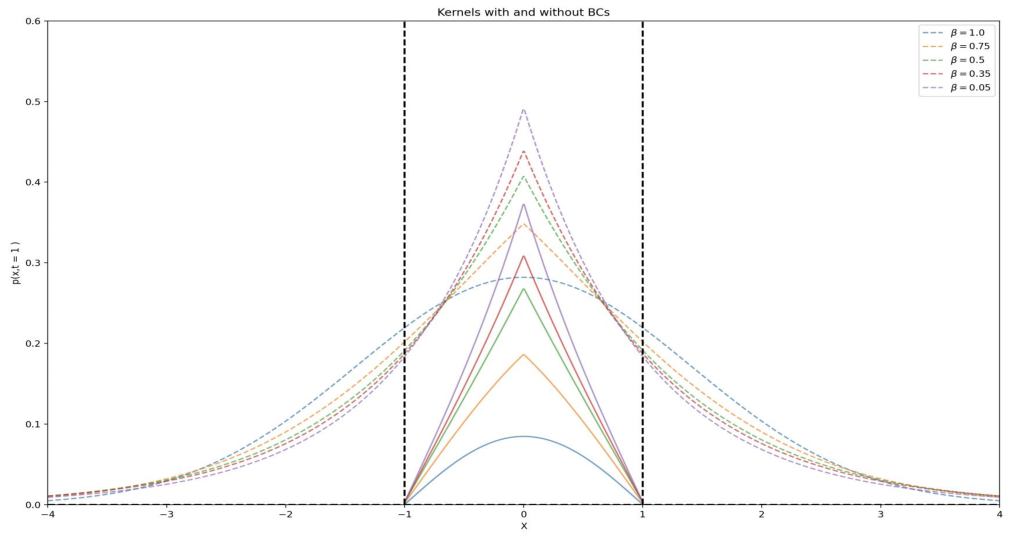
First passage methods are less granular, propagating particles by sampling analytic solutions for the ensemble. Many individual simulations steps can be skipped, drastically improving performance. In the context of HEAs, this allows for studying changes in material properties over months or years.

Proposed M-Wright Kernel and Boundary Conditions

$$G^\beta(x,t) = \frac{1}{2\sqrt{Dt^\beta}} M_{\frac{\beta}{2}}\left(\frac{|x|}{\sqrt{Dt^\beta}}\right) \xrightarrow{\text{Method of images}} K^\beta(x,t) = \frac{1}{\sqrt{Dt^\beta}} \sum_{n=0}^{\infty} M_{\frac{\beta}{2}}\left(\frac{|x-2nR|}{\sqrt{Dt^\beta}}\right)(-1)^n$$

To satisfy the boundary conditions of the first passage problem we need a bounded kernel. Such a kernel is constructed by superimposing many copies of $G^\beta(x,t)$ to force the kernel to vanish at some finite radius R (i.e. $K^\beta(\pm R, t) = 0$).

Right: dashed lines plot the bare Green's function $G^\beta(x,t)$ whereas the solid lines plot the bounded kernel $K^\beta(x,t)$ which is forced to vanish at $\pm R = \pm 1$.



LLF AWARD

The LLF fusion fellowship has supported my housing, transportation, and sustenance. It has allowed me to live near LLNL and commute to Berkeley weekly to continue my research there in preparation for a forthcoming publication.

Together, these projects have given me experience doing non-equilibrium statistical mechanics that I hope to transfer to graduate school.

The fellowship has also privileged me with the ability to explore northern California while remaining close to all the science I love.

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