Generative Latent Space Dynamics of Electron Density

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Electron density contains rich but *expensive* information for quantum mechanical properties

Modeling the time-dependent evolution of electron density is essential for understanding the quantum mechanical behaviors of condensed matter and unlocking predictive simulations for spectroscopy, excited states dynamics, charge transport, and ultrafast sciences. Yet, while various ML methods have advanced static density prediction from nuclei coordinates to bypass expensive electronic structure optimization, **modeling the spatiotemporal dynamics of electron density remain largely unexplored**.

Two common (simple) governing equations:

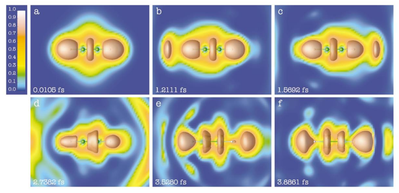
$$\left(-\frac{\hbar^2}{2m_e}\nabla^2 + V_{\text{eff}}(\mathbf{r})\right)\psi_i(\mathbf{r}) = \varepsilon_i\psi_i(\mathbf{r}),$$
$$\rho(\mathbf{r}) = \sum_i |\psi_i(\mathbf{r})|^2$$

Kohn-Sham Density Functional Theory (KS-DFT)

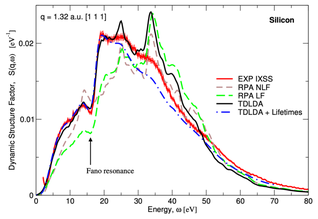
$$i\hbar\frac{\partial}{\partial t}\psi_i(\mathbf{r},t) = \hat{H}(\mathbf{r},t)\psi_i(\mathbf{r},t)$$
$$\rho(\mathbf{r},t) = \sum_i |\psi_i(\mathbf{r},t)|^2$$

Time-Dependent Density Functional Theory (TD-DFT)

Electron density dynamics can be used to calculate various quantities, such as dynamics structure factor, optical absorption, inelastic X-ray scattering, UV-Vis spectra, dielectric properties, and more.



Time-dependent electron localization (TD-ELF) for the excitation of acetylene (C_2H_2) [1]



Silicon dynamic structure factor at q=1.32 a.u. along [111] [2]

Our contributions

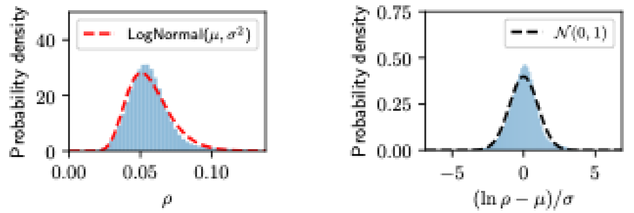
- We introduce a hybrid generative architecture combining **3D convolutional autoencoders** and **latent diffusion model (LDM)** to learn electron density trajectories from *ab-initio* molecular dynamics (AIMD) simulations.
- We propose a transformation scheme to map electron density from **lognormal distribution to normal distribution**.
- We use **scaled Jensen-Shannon divergence (sJSD)** loss to regularize the prediction distribution for better long-range spatial correlation.

Methods

Dataset and state representation

We generate **an AIMD trajectory of 32 lithium atoms in liquid state at 800 K for 10 ps**. We found that the distribution of electron density follows log-normal-like distribution, which can be transformed into normal distribution for model training. This approach also alleviates the positivity constraint on the model output space and therefore stabilizes the training and autoregressive prediction.

$$\tilde{\rho}(\mathbf{r},\tau) = \frac{\ln\rho - \mu}{\sigma} \in \mathbb{R}^{N_1\times N_2\times N_3}$$



To have complete representation of the current state, the state of electron density is described by both the number density and its time derivatives:

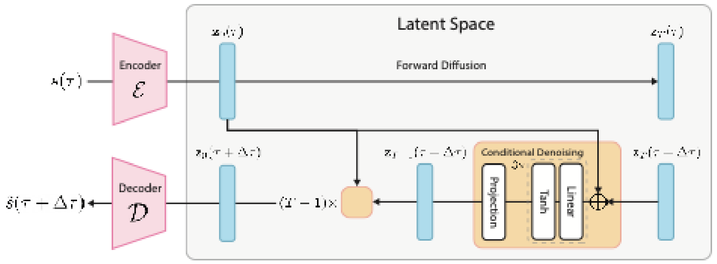
$$s(\tau) = (\mathbf{A}(\tau), \rho(\mathbf{r},\tau), \dot{\rho}(\mathbf{r},\tau)) \in \mathbb{R}^{3\times 3} \times \mathbb{R}^{N_1\times N_2\times N_3}_{\geq 0} \times \mathbb{R}^{N_1\times N_2\times N_3}.$$

With the *nice* property and transformation above, as well as simplification of fixed volume under isochoric-isothermal ensemble (NVT), the state representation reduces to

$$s(\tau) = \left(L, \tilde{\rho}(\mathbf{r},\tau), \dot{\tilde{\rho}}(\mathbf{r},\tau)\right) \in \mathbb{R} \times \mathbb{R}^{N_1\times N_2\times N_3} \times \mathbb{R}^{N_1\times N_2\times N_3}.$$

Conditional latent space generation as autoregressive prediction

At each physical time step, the model conditions the latent denoiser with the current latent state to predict the next latent state. The decoder then maps the latent state back to the physical space.



Latent space generative process to emulate the dynamics of electron density

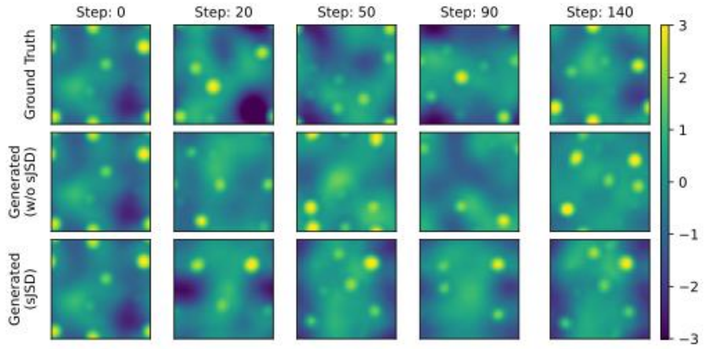
Experiments

Loss and sJSD regularization

$$\mathcal{L} = \mathcal{L}_{\text{AE}} + \lambda_1\mathcal{L}_{\text{LDM}} + \lambda_2\mathcal{L}_{\text{sJSD}}$$

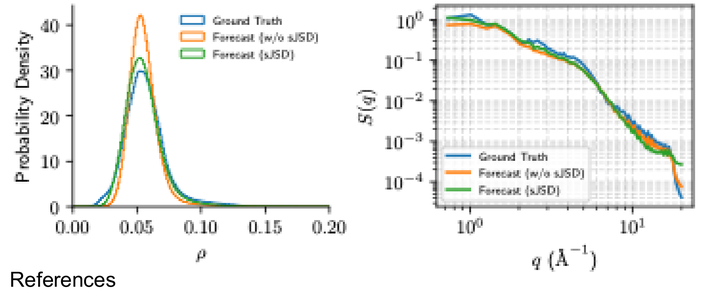
$$\mathcal{L}_{\text{sJSD}} = \frac{1}{Z}D_{\text{JS}}(P\,\|\,Q) = \frac{1}{2Z}\left[D_{\text{KL}}(P\,\|\,M) + D_{\text{KL}}(Q\,\|\,M)\right], \quad \text{where} \quad M = \frac{P+Q}{2}.$$

LDM generates and evolves electron density qualitatively consistent with unseen test trajectory of Li atoms at 800 K. The model trained with sJSD loss has less delocalized electron distribution similar to the ground truth.



Autoregressive trajectory rollouts with and without sJSD loss regularization in comparison with AIMD ground truth

We further compare the distributional and structure factor similarities between two generated and ground-truth trajectories. The model trained with sJSD loss clearly demonstrates more similar distribution to test trajectory than the one without sJSD regularization.



References

[1] Olevano, V., Palummo, M., Onida, G., & Del Sole, R. (1999). Exchange and correlation effects beyond the LDA on the dielectric function of silicon. Physical Review B, 60(20), 14224.

[2] Weissker, H. C., Serrano, J., Huotari, S., Bruneval, F., Sottile, F., Monaco, G., ... & Reining, L. (2006). Signatures of Short-Range Many-Body Effects in the Dielectric Function of Silicon for Finite Momentum Transfer. Physical review letters, 97(23), 237602.