RAPID COMMUNICATION

Learning physical states of bulk crystalline materials from atomic trajectories in molecular dynamics simulation

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Melting of crystalline material is a common physical phenomenon, yet it remains elusive owing to the diversity in physical pictures. In this work, we proposed a deep learning architecture to learn the physical states (solid- or liquid-phase) from the atomic trajectories of the bulk crystalline materials with four typical lattice types. The method has ultra-high accuracy (higher than 95%) for the classification of solid–liquid atoms during the phase transition process and is almost insensitive to temperature. The atomic physical states are identified from atomic behaviors without considering any characteristic threshold parameter, which yet is necessary for the classical methods. The phase transition of bulk crystalline materials can be correctly predicted by learning from the atomic behaviors of different materials, which confirms the close correlation between atomic behaviors and atomic physical states. These evidences forecast that there should be a more general undiscovered physical quantity implicated in the atomic behaviors and elucidate the nature of bulk crystalline melting.

Keywords: melting phase transition, crystalline materials, physical states, deep learning, molecular dynamics simulation

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Melting of crystalline material is a common physical phenomenon that occurs when the free energy of solid is equal to that of liquid, yet it remains elusive owing to the diversity in physical pictures of melting behavior in a variety of substances.[1–3] Large-scale molecular dynamics simulation is an effective and widely approved method for in-depth understanding of melting at the atomic level.[4–7] The atomic coordinates and velocities are the primary outputs describing atomic spatiotemporal state, and the pursuit to interpret the physical states of matter from atomic behavior is critical for understanding the essence of solid melting. Yet, till date finding a general physical quantity for interpreting the connotation of physical states has remained challenging.

Usually, physical states of a simulation system can simply be determined via atomic local structure information, i.e., order of atomic arrangement. The common atom types of crystalline systems mainly include face-centered cubic (FCC), body-centered cubic (BCC), hexagonal close packed (HCP), and icosahedral ones. Once subjected to thermal load, crystalline materials with regular lattice will lose their atomic order. According to this simple geometric change, the crystal structure recognition method, such as common neighbor analysis[8] (CNA) and its extended model adaptive-CNA (a-CNA),[9] bond-orientational order parameter,[10] and the diamond structure[11] (IDS) can be used to judge whether the crystal melts.[6,12] Recently, the polyhedral template matching (PTM)[13] was proposed based on the topology of the local atomic environment, which provides great reliability of atomic identification against thermal vibration. Although these methods can detect the melting phenomenon of crystalline materials, they cannot perform quantitative characterization due to the sensitivity to temperature or atom strain. Notably, the disorder of the atomic configuration is not the essence of the liquid phase.

The Lindemann criterion,[14] proposed as early as the early 20th century, has been widely accepted as an orthodox theory to elucidate the melting of solid via atomic vibrations. According to the Lindemann melting theory, melting stems from the mechanical instability due to enhanced atomic vibration. Solid melts when the magnitude of atomic thermal vibration exceeds a certain proportion of interatomic spacing, e.g., 0.05–0.2.[15] The thresholds of different materials need to consider additional factors acting as prior knowledge, such as crystal structure,[16,17] crystal surface,[18] dimension,[19] and the periodic table.[20]

Deep learning has attracted intensive attention to deal with complex scientific issues in many research fields.[21–24] Recently, a new modeling method "machine learning embedded with materials domain knowledge"[21] was proposed to reconcile the major contradictions[22] in applying machine

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learning to the materials community. Research shows that neural networks can explore the fundamental laws of classical mechanics.[23] Many scholars identified the transition of solid–liquid phase via deep learning,[25–28] where the atomic interaction potential surface[29] was applied to construct the learning feature. In fact, the dynamic behavior of atoms or particles is closer to the physical essence to characterize the melting phase transition of material systems.[30] From this point of view, atomic dynamic information, rather than atomic local structure, should be better to characterize the physical states. However, learning physical state of matter from atomic behavior for unlocking the essence of melting is still an open topic.

Here, we put forward a strategy mapping atomic behavior to physical states for crystalline material via convolution-based deep learning, where the temporal and spatial information of the atomic behavior, i.e., 3D atomic trajectories, are used as the inputs for training, validation, and prediction. The results show that the proposed method has excellent ability to identify solid and liquid atoms of bulk crystal materials in the first-order phase transition with high accuracy. The cross-prediction demonstrates that the atomic behavior can be used to predict crystal phase transition. The proposed method exhibits the intrinsic characteristics against thermal shock noise.

Figure 1 shows the time convolution neural network (TCNN) based architecture to forecast the physical state of crystalline materials during solid–liquid phase transition process which consists of two steps: (1) learning features from atomic trajectories [Figs. 1(a)–1(c)] and (2) predicting the solid–liquid phase transition of crystalline solid system [Figs. 1(c)–1(d)]. A defect-free bulk Au (FCC) is taken as an example to illustrate the architecture. Figure 1(a) shows a bulk Au arranged in a periodic box. Figure 1(b) shows the expanded trajectories of two atoms, signifying that the deep learning module exists three input channels. Figure 1(c) shows the module mainly including two parts: (1) the convolution layers for learning features from the atomic trajectories and (2) the fully connected neural network for mapping the features stemming from the first part to two output nodes indicating the atomic physical states, i.e., solid and liquid. Detailed configuration of this learning module and the corresponding parameters are provided in S1, where the inception module[31] was employed to build the convolution layers. The atoms that are identified as solid or liquid phase are called solid-like or liquid-like atoms for distinguishing the physical states in practical sense. It is noted that a single atom has no concept of physical state, but a group or system has real physical state, i.e., solid, liquid and gas. Here we evaluate the physical state of the crystal material system by counting the proportion of liquid-like atoms predicted by the model, as shown in the upper panel of Fig. 1(d). The lower panel of Fig. 1(d) shows the corresponding error curve.

In this paper, four single bulk crystalline solids, i.e., Au (FCC), Fe (BCC), Mg (HCP), and Si (diamond), and the Cu–Ni alloy with an initial FCC state were studied. The potential function of bulk Au is the multi-body potential function EAM.[32] That of bulk Fe is the multi-body potential function EAM/FS.[33] That of bulk Mg is the multi-body potential function EAM/FS.[34] That of bulk Si is the multi-body potential function SW.[35] The potential function of bulk Cu and Ni is the multi-body potential function EAM/ALLOY.[36] All molecular dynamics calculations were performed using the large-scale atomic/molecular massively parallel simulator (LAMMPS).[37] See S2 for the simulation details and S3 for the training settings and loss functions.

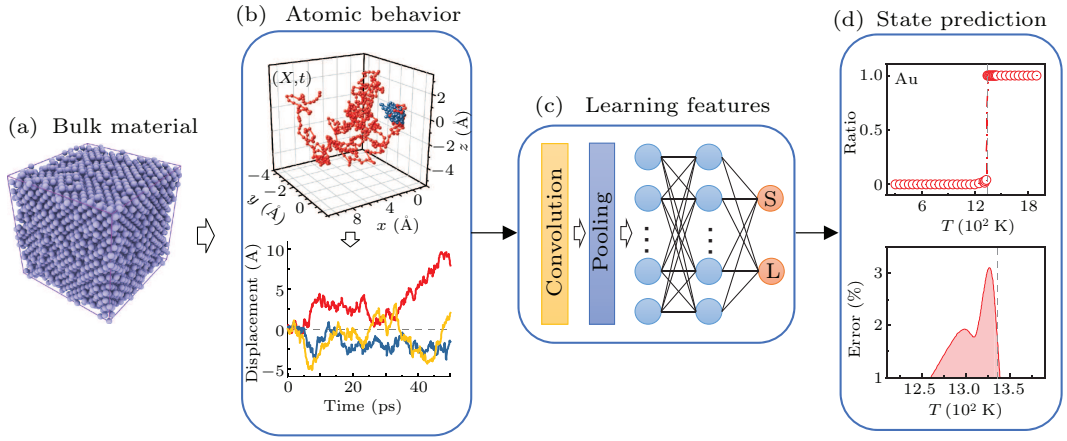


Fig. 1. Architecture for probing the melting process of bulk crystalline solids. (a) A bulk Au solid. (b) Atomic trajectories in 3D space. (c) TCNN-based module comprising convolution layers and full connection layers. (d) The phase transition curve defined as the variation of the liquid-like atoms (upper) and the prediction error of the atomic physical states (lower).

Figure 2 shows the average atomic potential energy (PE) and the ratio of liquid-like atoms predicted by TCNN as a function of temperature for the Au, Fe, Mg, and Si bulk crystal solids. The phase transition processes predicted by TCNN are consistent with those via PE curves. Since the quasistatic simulation method was adopted to anneal the crystalline solid with a gap of 10 K near the phase transition point, we calculated the melting points by averaging the two points before and after

the phase transition. The predicted melting point is 1335.0 K for Au, 2000.4 K for Fe, 1075.2 K for Mg, and 2313.5 K for Si. In fact, there is a certain deviation of melting points between calculations and experiments, which mainly depends on the potential function, material defects, heating rate and simulation method. For all cases, the ratio of liquid-like atoms is almost at the level of 0.0 before the phase transition, and abrupt increases to 1.0 after the phase transition. The former means that the system is solid, while the latter is liquid. The results are also verified from the Lindemann law in S4, where the threshold to identify solid-like or liquid-like atoms is not unique for different materials. The results are confirmed by the distribution of atomic diffusion coefficient as shown in S5. These coincidences are not surprising, because atomic trajectory contains atomic thermal vibration and diffusion behavior, which should be learned via deep learning.

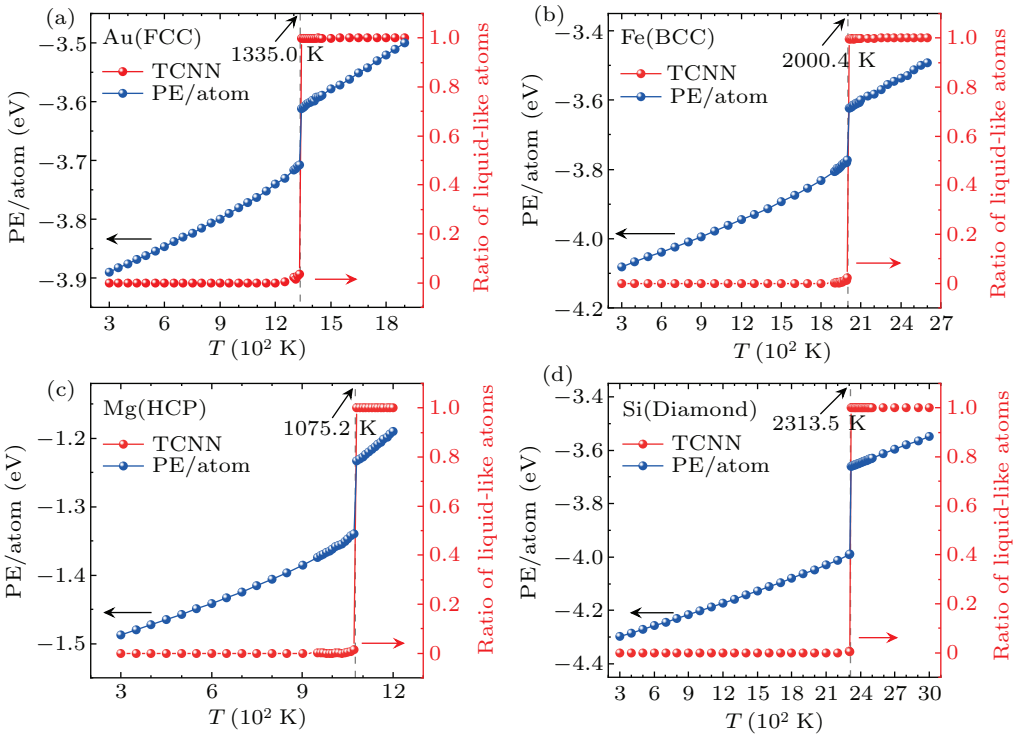


Fig. 2. The first-order phase transition curves represented with average atomic potential energy (blue) and ratio of liquid-like atoms by TCNN (red). (a) Au, (b) Fe, (c) Mg, and (d) Si.

Figure 3 shows the robustness of the model against thermal oscillation. For comparative analysis, TCNN, a-CAN, and PTM were used to identify the atomic state of bulk Au, Fe, and Mg, and TCNN and IDS were employed for bulk Si. The first column displays the liquid-like atom variation curves, which show that the results predicted by TCNN exhibit the highest accuracy. Even within the superheated state near the phase transition point, the error of bulk Au is less than 4%, bulk Fe is less than 3%, bulk Mg is less than 2%, and bulk Si is almost negligible, shown as the middle one. The results of bulk Au, Fe, and Mg classified using a-CNA and PTM comprised large errors. The errors at the time of impending phase transition afforded when using a-CNA and PTM are 88.5% and 38.1% for bulk Au, 79.7% and 34.5% for bulk Fe, and 80.1% and 23.6% for bulk Mg, respectively. The a-CNA and PTM usually employ atomic local information to identify the type or specific structure of atomic crystals; hence, they are easily affected by the violent thermal oscillation. The results of bulk Si identification through TCNN and IDS methods comprised negligible errors, which is attributed to the thermal stability of the diamond lattice. The right panels show several snapshots captured at the moments before and after the phase transition, where solid-like and liquid-like atoms are denoted with blue and orange colors, respectively.

Therefore, the identification result by the TCNN-based method is insensitive to temperature and the errors for all crystal bulk materials were less than 4%, which can be explained from the statistical properties of the method itself. Theoretically, atomic vibration is a behavior contained in the trajectory of atoms. From this perspective, it should be beneficial to the recognition accuracy of atomic types, as described by Lindemann's theory: the physical state of matter can be characterized using atomic thermal oscillation. Note that whether the thermally activated oscillation characteristic plays a positive role needs to be further researched due to the black box characteristic of neural networks.

Last, cross-prediction experiments were designed to illustrate the generality of predicting the phase transition of crystalline solids using atomic trajectories. Figure 4(a) shows the variation of the ratio curves of liquid-like atoms of bulk Au,

Fe, Mg, and Si with temperature, which were obtained by the approach that the physical state of each atom experiencing the phase transition process was predicted using different models trained with the data of other elements. Taking bulk Au as an example, the model parameters were first separately trained with the training data of Au, Fe, Mg, or Si, and given the well-trained network parameters, i.e., models; then, these models were utilized to predict the atomic physical state of each atom of bulk Au with additional data prepared using different random seeds. It shows that all the cross-prediction results for each case can accurately capture the temperature point of the phase transition, which agree well with the prediction results using target elements. However, the correctness loss before and after phase transition differs on a case-to-case basis. For bulk Au, the cross-prediction accuracies before phase transition exhibit little deviation, while those after phase transition decrease to a certain extent. Surprisingly, the accuracy increases with temperature, which is almost coincident once the temperature exceeds 1800 K. The cross-prediction results of the other elements (Fe, Mg, and Si) are consistent with each other; only the red curves (Au models) shown in the panels of Fe and Mg are not consistent, where a slight deviation ocurrs in a small range before the phase transition.

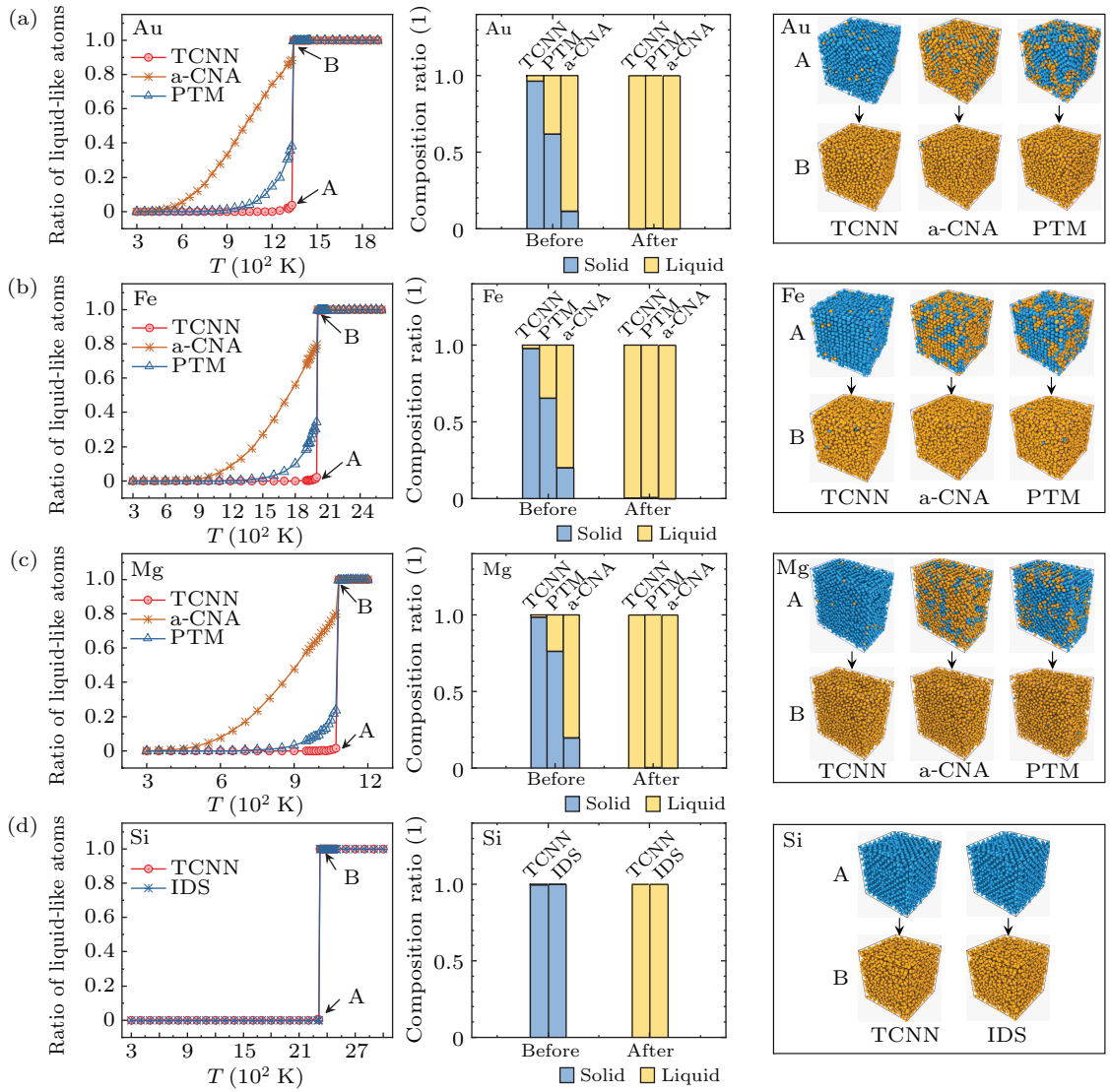


Fig. 3. Prediction accuracy of four types of bulk crystalline solids: (a) bulk Au (FCC), (b) bulk Fe (BCC), (c) bulk Mg (HCP), and (d) bulk Si (diamond) using TCNN, PTM, a-CNA, and IDS. The first column: ratio of liquid-like atoms as function of temperature; the middle column: the specific ratio of solid-like atoms with light blue and liquid-like atoms with pale yellow; the last column: snapshots corresponding to the middle column.

To understand these deviations, the previous results shown in Fig. 2 are reviewed. In the melting process, bulk Au hardly experiences overheating, while bulk Fe or Mg experiences a certain degree of overheating and Si endures a large degree of overheating. These seemingly coincidental phenomena indicate that phase transition under overheating induces high prediction accuracy. On the one hand, as the temperature increases, the liquid phase characteristics of atomic behavior (rapid diffusion, violent oscillation, etc.) become significant. On the other hand, when phase transition occurs in the ab-

sence of overheating or a small amount of overheating occurs, some atoms still afford local vibrations similar to solid-like atoms. Hence, relevant features can be captured by deep learning methods and retain in the model parameters.

We further demonstrated the generality of the TCNN-based method for predicting the evolution of physical states for solid alloys, where the 50%–50% copper–nickel alloy ($Cu_{0.5}Ni_{0.5}$) was considered, as shown in Fig. 4(b). The left panel shows that the model trained by each single element (Cu or Ni) not only can accurately predict the phase transition process of the alloy but also has high consistency in the ratio curves of liquid-like atoms at each temperature point. The right panel shows that the max error of Cu is about 2.5% and Ni is about 1.5%. The errors of the prediction results are almost the same (2%) near the phase transition line. It shows that the atomic behavior of different elements exhibits unified solid- or liquid-phase characteristics, which is independent of element or lattice types. The TCNN-based method using complete atomic trajectories affords a certain degree of consistency and universality, indicating that a more universal characterization quantity should be defined to identify the physical state of crystalline solid. This is consistent with the mean square displacement (MSD) theory that only considers the characteristics of atomic trajectories without distinguishing element types. Note that the average diffusion coefficient of particles is calculated by combing MSD with the Einstein theory.[38]

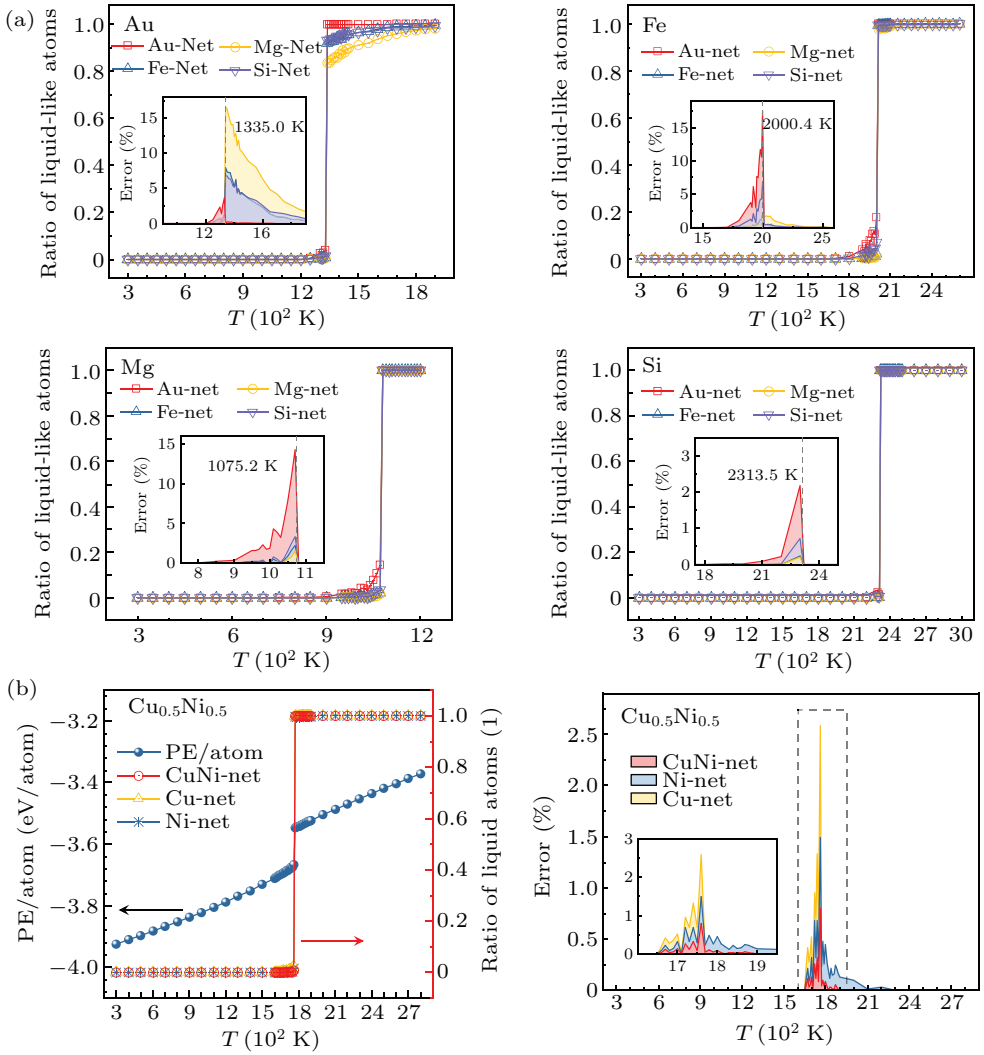


Fig. 4. Cross-prediction of melting phase transition for single-element crystalline solid and $Cu_{0.5}Ni_{0.5}$ alloy solid. (a) Ratio distribution of liquid-like atoms vs. temperature for bulk Au, Fe, Mg, and Si. (b) Ratio distribution of liquid-like atoms of $Cu_{0.5}Ni_{0.5}$ alloy vs. temperature.

This paper is limited to crystal bulk materials. However, the black box nature of the model limits us to deeply understand mechanism of the model extracting features and learning physical states. This interpretability problem should be solved by introducing domain expert knowledge. In addition, the applicability of our model needs further verification for more complex material systems with liquid-like atoms and solid-like atoms, such as nanoscale materials with significant scale effects, metallic glass materials and polymers with complex glass conversion processes.

Herein, a deep learning architecture based on the time-domain convolution neural network was proposed to predict

the phase transition process of bulk crystalline solids by probing the atomic physical state with atomic trajectories. For the bulk Au, Fe, Mg, and Si, the proposed architecture can accurately predict the physical states. i.e., solid and liquid. The predicted results are insensitive to temperature and the errors for all the crystal bulk materials are less than 4%. The cross-training and prediction analysis indicate that there should exhibit a lattice-independent generalized physical quantity for characterizing the physical state of crystal materials. Our study inspires future research to construct a more universal characterization quantity based on the atomic behavior to identify the atomic physical states of various materials.

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