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Structural Principles in Liquids and Glasses: Bottom-Up or Top-Down

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The conventional approach to elucidate the atomic structure of liquid and glass is to start with local structural units made of several atoms, and to use them as building blocks to form a global structure, the *bottom-up* approach. We propose to add an alternative *top-down* approach in which we start with a global high-temperature gas state and then apply interatomic potentials to all atoms at once. This causes collective density wave instability in all directions with the same wavelength. These two driving forces, local and global, are in competition and are mutually frustrated. The final structure is determined through the compromise of frustration between these two, which creates the medium-range-order. This even-handed approach on global and local potential energy landscapes explains the distinct natures of short-range order and medium-range order, and strong temperature dependence of various properties of liquid.

Keywords: liquid and glass, atomic structure, short-range order, medium-range order, pseudopotential

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INTRODUCTION

Liquids and glasses have strongly disordered atomic structures which are difficult to characterize with precision. Explaining the origin of such structures is an even more daunting task, which has been a subject of discussion for a long time (Egelstaff, 1967; Croxton, 1974; Hansen and McDonald, 1976; Debenedetti and Stillinger, 2001; March and Tosi, 2002; Parisi and Zamponi, 2010). In the absence of symmetry, the most popular approach is the *bottom-up* approach, starting with local structural units, such as the nearest neighbors of an atom (short-range order—SRO) and building up the global structure with preferred local structures (Miracle, 2004; Sheng, et al., 2006; Robinson et al., 2019). The preferred structures, such as icosahedral clusters, are often geometrically frustrated as a building unit (Sadoc, 1981; Nelson, 1983; Sethna, 1983; Tarjus, et al., 2005), resulting in a disordered global structure. However, in this approach the frustration condition depends on details of the cluster geometry, and the universal principle of structure formation remains deeply hidden.

We propose a holistic concept to depict this frustration through global and local features of potential energy landscape, by going beyond the idea of geometrical frustration. On top of the bottom-up approach we add a *top-down* approach, in which we start with a high-density gas state and apply interatomic potentials to all atoms at once. We show that a high-density gas state is unstable against the global density wave state, once the interatomic potential is introduced in reciprocal space. The local force to form atomic units and the global force to form density waves are in conflict against each other. The real structure results from the competition and compromise between these two frustrated driving forces. The medium-range order (MRO) in the oscillation in density correlations beyond the nearest neighbors reflects this competition. The competition depends on temperature. At high temperatures the local forces are dominant, whereas as

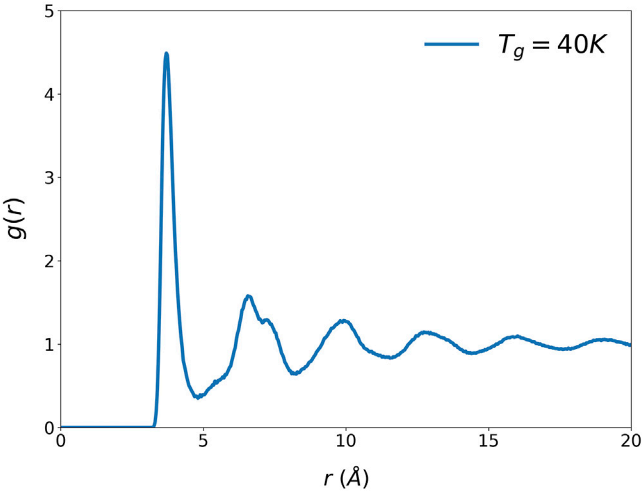

FIGURE 1 | The PDF, $g(r)$, of liquid Argon at T_g = 40 K.

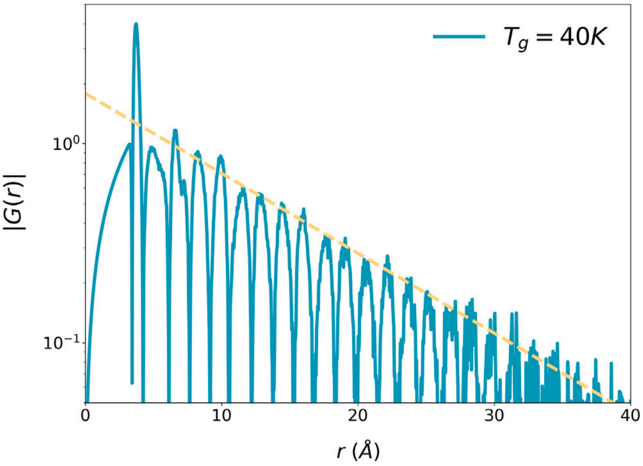

FIGURE 2 | The absolute values of the reduced PDF, $|G(r)|$, of liquid Argon at T_g = 40 K. The dashed line shows the exponential decay by **Eq. 1**.

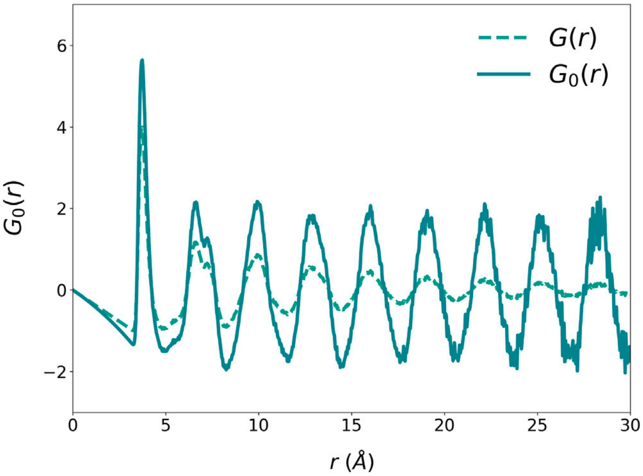

FIGURE 3 | The $G_0(r)$ of liquid Argon at T_g = 40 K obtained by **Eq. 1**.

temperature is reduced the importance of global force increases. This change in their roles explains strong temperature dependence in various properties of liquid.

NATURE OF THE MEDIUM-RANGE-ORDER

The atomic structure of liquid and glass is usually described in terms of the pair-distribution function (PDF), $g(r)$, which shows the distribution of distances between two atoms. The PDF is obtained by Fourier-transforming the structure function, $S(Q)$, where Q is the momentum transfer in scattering, determined by x-ray or neutron diffraction (Warren, 1969). **Figure 1** shows an example of the PDF, that of Argon liquid model with the Lennard-Jones (LJ) potential (Rowley, et al., 1975), with 34,461 atoms in the cubic supercell of the edge size, L = 112.01 Å. The first peak of the PDF describes the distribution of the distances to the nearest neighbor atoms from a central atom and describes the radial SRO. The oscillations in the PDF beyond the first peak describes the MRO, and they decay with the form,

$$G(r) = 4\pi r\rho_0\left[g(r) - 1\right] = G_0(r)\exp\left(-r/\xi_s\right) \tag{1}$$

where ρ_0 is the macroscopic number density of atoms and ξ_s is the structural coherence length, as shown in **Figure 2**.

The form of **Eq. 1** was first suggested by Ornstein and Zernike. (1914), but it can be derived by a more general argument (Ryu and Egami, 2021). Also, whereas the OZ theory connects the SRO directly to the MRO and regards the MRO as a consequence of the SRO, we found that the nature of the MRO is substantially distinct from that of the SRO. For instance, the variation of the MRO with temperature shows a clear change at the glass transition temperature, T_g, but the SRO is continuous through T_g (Ryu and Egami, 2021). The first peak of the PDF is relatively narrow (<1 Å) and describes the atom-atom distances to the nearest neighbors, which number in the order of ten. But the MRO peaks are wider (~1 Å), and cover hundreds of atomic distances. Therefore, they describe the correlation between the center atom and aggregates of atoms, or density fluctuations (Egami, 2020). In other words, the SRO describes the *point-to-point* correlation, whereas the MRO describes the *point-to-set* correlation (Berthier and Kob, 2012).

For metallic liquids the structural coherence length, ξ_s, which characterizes the MRO, obeys the Curie-Weiss law (Ryu, et al., 2019),

$$\xi_s(T) = \frac{aC}{T - T_{IG}} \tag{2}$$

where a is the nearest neighbor distance, and T_{IG} (<0) is the ideal glass temperature. At $T = T_{IG}$, ξ_s diverges and $G(r, T_{IG}) = G_0(r)$. Therefore, $G_0(r)$, **Figure 3**, describes the structure of an ideally coherent glass state (Ryu, et al., 2019), which has long-range

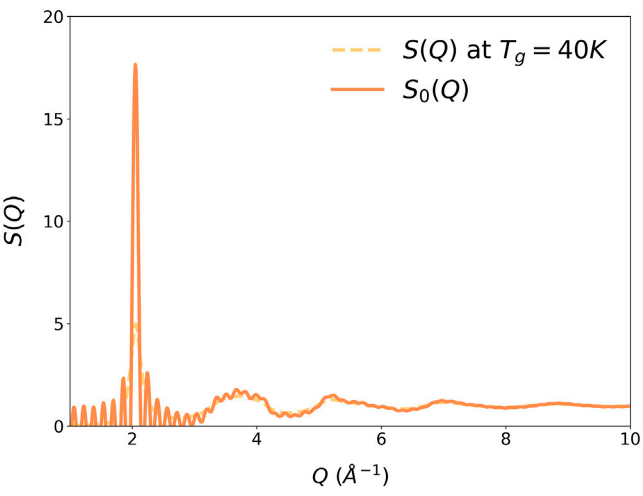

FIGURE 4 | The $S(Q)$ of liquid Argon at $T_g = 40$ K. The height of the first peak depends on the model size, and extrapolates to a δ-function for a macroscopic model.

positional correlation without periodicity. Its Fourier-transform, the structure function $S_0(Q)$ shown in **Figure 4**, has a Bragg-like sharp first peak at Q_1, of which height depends on the model size. Small sharp peaks are noise due to the finite model size. Because the structure is isotropic, in three-dimensions the first peak forms a Bragg sphere.

INTERATOMIC PSEUDO-POTENTIAL AND DENSITY WAVE

The ideally coherent glass state exists only in extrapolation, because T_{IG} is negative and the structure freezes at T_g. However, the Curie-Weiss temperature dependence of the structural coherence length, ξ_s, suggests that there is a driving force for a liquid towards this state. We suggest that the origin of such a force is the attractive interatomic potential. Let us consider a simple monoatomic liquid of atoms interacting with a two-body spherical potential, $\phi(r)$. We express the local atomic density, $\rho(\boldsymbol{r})$, in terms of the density waves,

$$\rho(\boldsymbol{r}) = \int \rho(\boldsymbol{q})\, exp(i\boldsymbol{q}\cdot\boldsymbol{r})d\boldsymbol{q} \tag{3}$$

In order to make the density real, we assume, $\rho(-\boldsymbol{q}) = \rho^*(\boldsymbol{q})$. The $\rho(\boldsymbol{q})$ is a complex number,

$$\rho(\boldsymbol{q}) = |\rho(\boldsymbol{q})|\, exp(i\delta(\boldsymbol{q})) \tag{4}$$

and the phase factor, $\delta(\boldsymbol{q})$, has a nearly random distribution to avoid wave pile-up which would put more than one atom at a place. The total potential energy is given by

$$U = \int |\rho(\boldsymbol{q})|^2 \phi(\boldsymbol{q})d\boldsymbol{q} = \rho_0^2 \int S(\boldsymbol{q})\phi(\boldsymbol{q})d\boldsymbol{q} \tag{5}$$

where $\phi(\boldsymbol{q})$ is the Fourier-transform of $\phi(\boldsymbol{r})$,

$$\phi(\boldsymbol{q}) = \int \phi(\boldsymbol{r})\, exp(-i\boldsymbol{q}\cdot\boldsymbol{r})d\boldsymbol{r} \tag{6}$$

For a spherical potential, $\phi(\boldsymbol{r}) = \phi(r)$, $\boldsymbol{r} = |\boldsymbol{r}|$, and $\phi(\boldsymbol{q}) = \phi(q)$, $\boldsymbol{q} = |\boldsymbol{q}|$. The $\phi(q)$ is dominated by the strong repulsive part of $\phi(r)$ at small r. However, the strongly repulsive part of the potential with the energy much larger than k_BT is irrelevant, because atoms never come so close to each other. Therefore, we define a "pseudopotential," $\phi_{\mathrm{pp}}(r)$, in which the strongly repulsive part of $\phi(r)$ is removed and $\phi_{pp}(\boldsymbol{r}) = \phi(\boldsymbol{r}_c)$ is assumed for $r < r_c$, where r_c is the cut-off distance. This is equivalent to disregarding the inaccessible high energy states from the potential energy landscape. It is also similar to the concept of the pseudopotential in the quantum scattering theory: A strong attractive potential simply adds a multiple of 2π to the scattering phase shift, and taking just the principal part of the phase shift corresponds to replacing it with the pseudopotential (Wu and Ohmura, 1962). For electrons removing the redundant phase shift is equivalent to reinforcing orthogonality to the core states (Phillips and Kleinman, 1959). For a repulsive potential we remove the

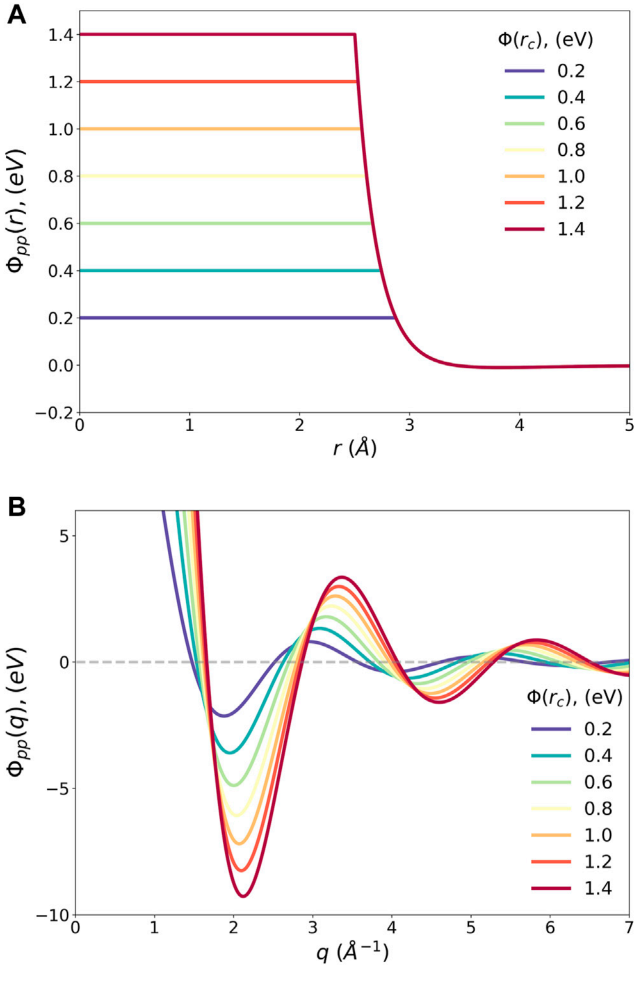

FIGURE 5 | (A) The pseudopotential of the Lennard-Jones potential with various cut-off levels, $\phi_{\mathrm{pp}}(r)$, and **(B)** their Fourier transforms, $\phi_{\mathrm{pp}}(q)$.

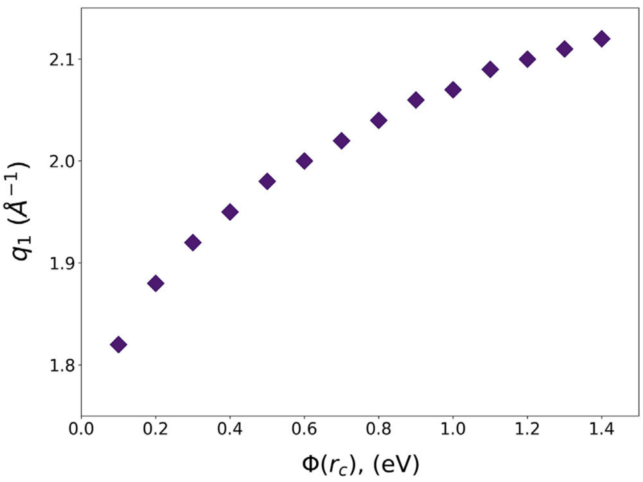

FIGURE 6 | The q_1 of liquid Argon pseudopotential as a function of the cut-off energy. Q_1 = 2.06 Å^{-1}.

strongly repulsive part. We separate $\phi(r)$ into two parts; $\phi_{pp}(r)$ and the repulsive part, $\phi_R(r)$,

$$\phi(\boldsymbol{r}) = \phi_R(\boldsymbol{r}) + \phi_{pp}(\boldsymbol{r}) \tag{7}$$

then, the potential energy is,

$$U = U_R + U_{pp} \tag{8}$$

$$U_R = \frac{1}{V}\iint \phi_R(\boldsymbol{r})\rho(\boldsymbol{r}')\rho(\boldsymbol{r}+\boldsymbol{r}')d\boldsymbol{r}d\boldsymbol{r}' \tag{9}$$

$$U_{pp} = \frac{1}{V}\iint \phi_{pp}(\boldsymbol{r})\rho(\boldsymbol{r}')\rho(\boldsymbol{r}+\boldsymbol{r}')d\boldsymbol{r}d\boldsymbol{r}' \tag{10}$$

where V is volume. The $\rho(\boldsymbol{r})$ of the ground state is determined by minimizing U. However, for the range $r < r_c$ where $\phi_R(r)$ is non-zero, there is no pair of atoms with such small separations because of the strong repulsion. Therefore,

$$U_R = 0, \quad U = U_{pp}, \tag{11}$$

and the structure is determined by minimizing U_{pp} with,

$$\phi_{pp}(\boldsymbol{q}) = \int \phi_{pp}(\boldsymbol{r})\,exp(-i\boldsymbol{q}\cdot\boldsymbol{r})d\boldsymbol{r} \tag{12}$$

The pseudopotentials of the Lennard-Jones potential for Argon with various cut-off values and their Fourier-transforms are shown in **Figure 5**. Interestingly $\phi_{pp}(q)$ has a minimum at q_1, which is not too far from the position of the maximum in $S(Q)$ at T_g, Q_1 = 2.06 Å^{-1}. The dependence of q_1 on $\phi(\boldsymbol{r}_c)$ is shown in **Figure 6**. The value of Q_1 decreases with increasing temperature, down to 1.93 Å^{-1} at T = 100 K. A minimum in $\phi_{pp}(q)$ at q_1 implies that a density wave with q_1 would be energetically preferred. Because both $S(Q)$ and $\phi(q)$ are dominated by the region in q around Q_1 (q_1), a density wave which minimize U must be that with q close to q_1. Thus, in the zero-th order the structure is dominated by the global density waves with q_1. In other words, a high-density gas state is unstable against the global density wave state with q_1.

However, the density wave with q_1 has a fairly long wavelength, which results in a wide first peak of the PDF, as wide as the higher order PDF peaks. This is in conflict with the real-space requirement that the first peak of $g(r)$ should be narrow around the bottom of $\phi(r)$ at $r = a$ to reduce the potential energy. Thus, the requirement to reduce the potential energy in q space by density waves and the same requirement in real space by better SRO are orthogonal, resulting in frustration. This is a more general energetic statement of the geometrical frustration discussed by many in more specific forms, such as formation of icosahedral clusters (Sadoc, 1981; Nelson, 1983; Sethna, 1983). This conflict also explains the distinct natures of the MRO and SRO (Ryu and Egami, 2021). The final structure results from the compromise between the two, requiring additional density waves with high q values.

DISCUSSION

The conventional approach to elucidate the structure of liquid is the *bottom-up* approach, in which the interatomic potential is applied to a small number of atoms to form a good local cluster, and the structure is extended by adding more clusters (Miracle, 2004; Sheng, et al., 2006; Robinson, et al., 2019). A major problem of this approach is that by starting with good clusters the boundaries between good clusters become strongly strained. This heterogeneous strain concentration increases the total strain energy. Actually, in an extended structure the peripheral atoms in one cluster are also central atoms in other clusters, thus the clusters are overlapping. Focusing just on "good" clusters is biased and dangerous. An orthogonal *top-down* approach is to consider all atoms at once, and to apply the potential simultaneously to all atoms. This can be done more effectively in reciprocal space. By removing the irrelevant strongly repulsive part of the interatomic potential to form the pseudopotential, the Fourier-transform of the potential, $\phi_{pp}(q)$, was found to have a minimum at q_1, close to the position of the maximum in $S(Q)$ at Q_1. This means that there is a driving force to form a density wave at q_1. A small set of coherent density waves can form the basis for a crystal structure (Alexander and McTague, 1978), but a large set of density waves with random phase factors can form a state with long-range correlation without periodicity, the structurally coherent ideal glass state. The nature of such a state will be discussed elsewhere. Quasicrystal was the first of such state with long-range correlation without periodicity, which has a periodic crystalline lattice in six-dimensions (Levine and Steinhardt, 1984). The ideal glass forms a crystal in infinite dimensions.

A model of the coherent ideal glass state can be constructed by using $G_0(r)$ as a guide. It was found the model has very diverse local structures (Ryu et al., 2019). For instance, the fraction of the icosahedral local environment is only 0.7%. Apparently, in order to maintain long-range density correlation the local structure is sacrificed. This is another proof of the conflict between the force to produce long-range density wave state and the force to create short-range atomic order. The final structure is determined through the compromise between these two conflicting forces. The balance between the two is reflected to the coherence length;

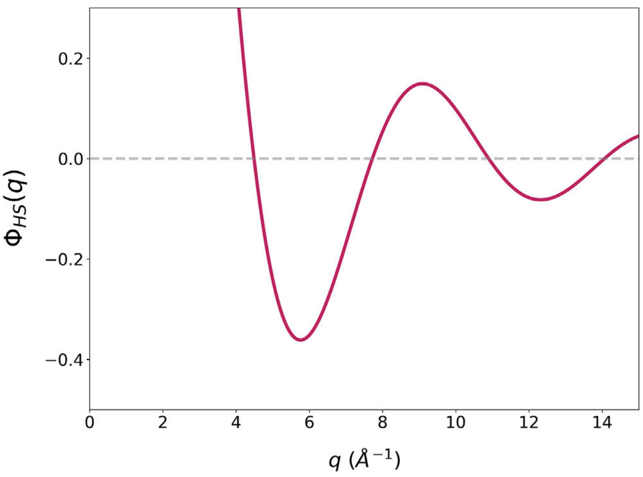

FIGURE 7 | The Fourier transform of the pseudopotential of the hard-sphere potential, $\phi_{\mathrm{HS}}(q)$, with the cut-off at 1 energy unit and D_{HS} = 1 Å.

the stronger the SRO is, for instance because of covalency, the shorter the ξ_s is. The extent of this balance can be expressed as the ideality of the structure (Ryu, et al., 2020). Furthermore, temperature also affects this balance. Local density fluctuations due to the atomic-level pressure fluctuations reduces the coherence of the density wave. This argument quantitatively explains the Curie-Weiss law, **Eq. 2** (Egami and Ryu, 2021).

The idea of geometric frustration is based on the hard-sphere (HS) packing argument (Nelson, 1983). In the DRP-HS model dynamics is controlled by jamming, because no potential energy is involved. To keep the density constant external pressure is applied, which plays the role of the attractive potential. It is interesting to note, however, if we use the pseudopotential, $\phi_{\mathrm{HS}}(q)$ has a minimum at $q_{1,\mathrm{HS}}$ as shown in **Figure 7**, with $D_{\mathrm{HS}}q_{1,\mathrm{HS}} = 5.76$, where $D_{\mathrm{HS}} = 2R_{\mathrm{HS}}$ and R_{HS} is the hard-sphere radius. This value is not far from the value of $D_{\mathrm{HS}}Q_{1,\mathrm{HS}} = 6.9$ for the hard-sphere model, where $Q_{1,\mathrm{HS}}$ is the Q_1 for hard-spheres (Lange, et al., 2009). It could also be related to the long-range oscillation found recently (Rissone et al., 2021), but its value, $D_{\mathrm{HS}}Ql = 7.5$, is considerably higher. If the pseudopotential concept applies even for the hard-sphere mode, it means that the mere action of exclusion on a gas under pressure may induce a density wave state. The height of the pseudopotential is irrelevant as long as it is substantially larger than k_BT. The $\phi_{\mathrm{HS}}(q)$ has a long oscillating tail, and it influences the structure through the high Q part of $S(Q)$. However, the strong minimum of $\phi_{\mathrm{HS}}(q)$ has a dominant effect on the first peak of $S(Q)$, producing the density wave and the MRO even in the hard-sphere model.

Thus, the idea of the minimum of $\phi(q)$ producing the density wave and the MRO appears to work widely, independent of details of the interatomic potential, possibly including even the hard-sphere model. As will be described elsewhere the dynamics of the density wave, through the amplitudons and phasons, is directly related to atomic dynamics. Thus, the stiffness of the density wave controls the cooperative atomic dynamics, affecting many physical properties of liquid. To what extent the same argument applies to covalently bonded network glass needs to be examined.

CONCLUSION

To characterize the atomic structure of liquid and glass, it is customary to start with focusing on the local structural units made of several atoms, and to consider adding them up to form the global structure. This *bottom-up* approach is so prevalent that its wisdom has not even been questioned. However, glasses are not formed in such a manner. In the process of glass formation through the glass transition all atoms equally contribute to glass formation during cooling through cooperative processes. To capture this many-body aspect of global glass formation, we propose a new holistic concept, made of dual *bottom-up* and *top-down* approaches. In the top-down approach we consider an assembly of many atoms in a high-density gas state and apply interatomic potentials to all atoms at once. The interatomic potential in reciprocal space causes collective density wave instability, which tries to create the ideal glass state. The final structure is determined through the competition and compromise of frustration between these two, local and global, driving forces. The balance between the two driving forces can be quantified in terms of the coherence length of the MRO, ξ_s, and the ideality of the liquid structure (Ryu et al., 2020). This approach explains the distinct natures of short-range order and medium-range order, and strong temperature dependence of various properties of liquid.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

This work was conceived and authored by TE with assistance by CR. The data in this work were generated by CR. Both authors contributed to the article and approved the submitted version.

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