

# Simple scenario for glass transition phenomena based on liquid-liquid transition framework

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We propose a simple scenario for glass transition phenomena. There are two new aspects in it: (1) Liquid-liquid (*l-l*) transition with a critical point is assumed and the entropy accompanying it is discussed. (2) Time concept is introduced to this transition entropy.

In the traditional framework [1], the slow dynamics of glass forming liquids has attributed to supercooling and the configurational entropy has been discussed. But it propounds some critical problems including so-called Kauzmann paradox [2]. In our scenario, we seek the origin of the slow dynamics in critical fluctuations of *l-l* transition and discuss the transition entropy accompanying it. It enables us to avoid the Kauzmann paradox and to obtain a simple interpretation to the fragility index of glass forming liquids: It is a parameter indicating how close the pressure of the liquid to the critical pressure of *l-l* transition. This can explain the pressure dependence of the fragility observed in liquid glycerol [3] and is consistent with Angell's 'big picture' [4]. On the other hand, it has been concluded that the entropy effect accompanying *l-l* transition substantially slows down the ultrasonic sound velocity at MHz frequency [5]; however, our recent study [6] on liquid water and liquid Te mixtures (with Se or Ge) [7], which are expected to undergo *l-l* transition, revealed that the sound velocity at THz obtained by inelastic x-ray scattering measurements is insensitive to *l-l* transition. These results can be simply interpreted if we introduce time concept to this transition entropy: If the frequency is high enough compared to the inverse of the characteristic time of the system, the transition entropy is frozen. Similar concept has been discussed among theorists to overcome the broken ergodicity [8]. In this scenario, the abrupt change of the specific heat observed at the glass transition is no more than the time dependence of transition entropy. That is, the glass transition is not a thermodynamic phase transition at all, but is regarded as a part of *l-l* transition.

Our scenario can give many results with a simple manner as will be presented at the conference. We look forward to fruitful discussion with all of you.

- [1] G. E. Gibson et al, J. Am. Chem. Soc. **45**, 93 (1923)
- [2] W. Kauzmann, Chem. Rev. **43**, 219 (1948)
- [3] R. L. Cook et al, J. Chem. Phys. **100**, 5178 (1994)
- [4] C. A. Angell, MRS Bull. **33**, 1 (2008)
- [5] Y. Tsuchiya et al, J. Phys. C: Solid State Phys. **15**, L687 (1982)
- [6] Y. Kajihara, Rev. High Press. Sci. Tech. (in Japanese) **26**, 288 (2016)
- [7] Y. Kajihara et al, J. Phys.: Condens. Matter **20**, 494244 (2008) and in preparation
- [8] J. C. Mauro et al, J. Chem. Phys. **126**, 184511 (2007)