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The liquid-liquid transition and its order parameter

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我々は Triphenyl Phosphite(TPP) および normal butanol という分子性液体において液体・液体相転移を発見した。そのキネティクスを顕微鏡、熱量測定を用いた調べた結果、核形成・成長型 (NG-type) 相転移とスピノーダル分解型 (SD-type) 相転移の2種類の相転移過程があることがわかった。スピノーダル分解型相転移を詳細に調べた結果、液体・液体相転移を支配する秩序変数は非保存系であることがわかった。そこで、Photon Factory(PF) において放射光X線回折実験を行い、液体・液体相転移中における液体のミクロスコピックな変化を調べた。その結果、液体・液体相転移の秩序変数は局所安定構造の数密度であることが示唆された。

Even a single-component liquid may have more than one kind of isotropic liquid state. The transition between the different liquid states is called a " liquid-liquid phase transition (LLT)" and this is discovered by atomic liquids such as carbon, Si, P[1]. Although the existence of LLT has become more and more convincing, the nature of the transition is still largely unknown. This situation mainly comes from the experimental difficulties. The transition is located at high temperature and high pressure for atomic liquids or is hidden by crystallization. Recently, we found convincing experimental evidence for the existence of LLT in a molecular liquid, Triphenyl Phosphite(TPP) and n-butanol[2, 3, 4]. Both nucleation-growth-type (NG-type) and spinodal-decomposition-type (SD-type) phase transformations were directly observed with optical microscopy. From the image obtained with phase-contrast microscopy, we can calculate the structure factor $F(q)$. In the early stage, the peak intensity $F(q_p) \propto \exp(\Gamma t)$ (Γ is a constant), whereas the peak wavenumber q_p is constant with time. This is the linear growth regime of phase transformation, which is known as the Cahn's linear regime for SD-type phase separation[5]. In the intermediate stage, the temporal decrease of q_p is well described by the power law $q_p \sim t^{-0.5}$. This exponent is characteristic of the late-stage coarsening of SD-type ordering of a nonconserved order parameter.

We believe that this nonconserved order parameter is the number density of locally favored structures (LFSs) [3, 6]. LFSs exist in many liquids, for example, tetrahedra structures in water and icosahedra structures in metallic liquids. LLT is the transformation from liquid I including

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a few of LFSs to a lot of LFSs liquid.

Thus, we investigate microscopic change during LLT to confirm our idea with synchrotron X-ray scattering. This work was done at BL-15A, Photon Factory (PF), KEK, and Tsukuba. We measured the scattering factor I(q) of TPP every 10 minutes.

We found that I(q) changes during LLT and this change comes from the increase of the number density of LFSs. We report that the nonconserved order parameter governing LLT is the number density of LFSs.

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