Mass-dependence of self-diffusion coefficients in disparate-mass binary fluid mixtures

I. Binas and I. Mryglod

Institute for Condensed Matter Physics of the National Academy of Sciences of Ukraine, 1 Svientsitskii Str., 79011 Lviv, Ukraine,
E–mail: idyachuk@icmp.lviv.ua

Binary fluids with strong asymmetry in masses, sizes or charges of particles, forming a mixture present a good example of the system with complicated multiscale dynamics. As the result of asymmetry special features of such mixtures are revealed: fast sound, anomalous diffusion, dynamic arrest and cage effect, crossover to Brownian limit, etc. Some of these features can be observed already on the behaviour of self-diffusion. In particular, $D_2/D_1 \sim (m_1/m_2)^\kappa$, where D_2 and D_1 are the self-diffusion coefficients of heavy and light particles correspondingly, m_2 and m_1 are masses. The coefficient κ is equal to 0.5 in the kinetic theory, it varies from 0.06 to 0.1 according to simulation results, and should be equal to 0 in Brownian limit because of mass independence.

Self-diffusion coefficients were calculated within memory function formalism, using the systematic subsequence of approximations for the relaxation times of velocity autocorrelation function. In the limit $\mu \ll 1$ we obtain:

$$D_i = \frac{k_B T}{\sqrt{m_1}} \cdot A^{(i)}(1 + B_1^{(i)}\mu + B_2^{(i)}\mu^2 + \ldots),$$

where μ is the ratio of masses of light and heavy particles; $A^{(i)}, B_1^{(i)}, B_2^{(i)}$ are some coefficients which are expressed via static correlation functions and do not depend on the masses. The obtained expression has a correct Brownian limit. We developed the hierarchy of approximations for $A^{(i)}, B_1^{(i)}$ and $B_2^{(i)}$ that tends to exact result from above and below when the order of approximations increases. The results are planned to be tested in molecular dynamics.