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Glassy Dynamics Altered by a Free Surface[1]

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Studies of polymer dynamics in thin films showed that a highly mobile region exists at the free surface of most if not all polymers. In this talk, I shall review some of these observations, with highlights given to the recent findings that chain flexibility and connectivity may on occasions be necessary for the free surface to exercise its influence. Afterward, I shall ponder on how the influence of the free surface may penetrate as far as several polymer radii of gyration into the inner region, as found both in experiments and simulations. Near the glass transition temperature, our MD simulations showed that the dynamics consist mainly of string-like particle hopping motions, as found by others. Importantly, as the temperature decreases, the hopping motions become increasingly repetitive and back-and-forth, contributing no structural relaxations. We propose that structural relaxations are then brought about by pair-interactions between strings. Near the free surface, however, similar repetitive hopping motions are only observed sufficiently far removed from the free surface. We propose that the free surface induces a penetrating surface mobile region by breaking the memory in the particle dynamics. A possible mechanism based on string interactions will be discussed.

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