Collective disentanglement of entangled polymers

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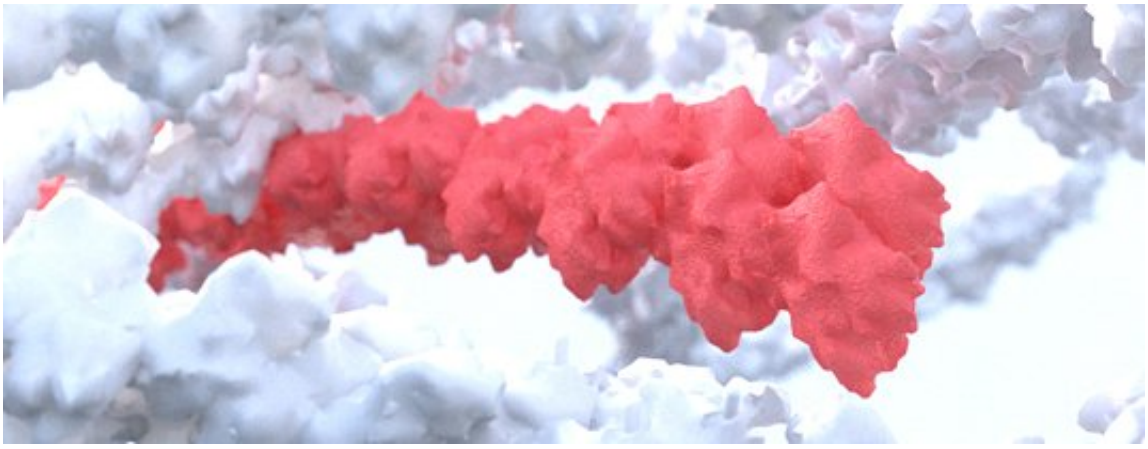

An actin filament in motion. Credit: C. Hohmann, NIM

LMU researchers have disproven the conventional theory used to explain the dynamics of polymer solutions. They show that for biopolymers collective effects facilitate chain mobility, which is reminiscent of the behavior of glass-like materials.

What do silk threads, plastics and DNA have in common? They are all made up of polymeric constituents and are representative of what scientists call 'soft matter'. Unlike hard condensed matter, these materials are intrinsically flexible ('soft'), but can nevertheless form stable structures under ambient conditions. Hence, soft matter cannot be unequivocally classified as either solid or liquid, because its material

properties are very sensitive to environmental parameters. – Changes in temperature can have a marked effect on their susceptibility to deformation, for instance. Indeed, their structural dynamics is the key to their behavior and the major determinant of their functions and applications. In a new study, LMU physicists Professor Erwin Frey and Dr. Philipp Lang have refuted the conventional theory hitherto used to account for the rheological characteristics (i.e. their response to external mechanical stresses) of polymer solutions, and developed an alternative model. The new study appears in the online journal *Nature Communications*.

Frey and Lang focused on the dynamics of semi-flexible to stiff polymers – the group to which biopolymers including DNA, or actin filaments and microtubules that are a major component of the cytoskeleton belong. All polymers are made up of repeating subunits that are linked together to form long macromolecular chains. In solution, these macromolecules are intricately entangled with each other, like the fibers in clumps of fluff. In the 1970s, a model was developed to describe their dynamics. In this reptation model, each polymer molecule is viewed as being confined within a flexible tube through which it moves in an undulatory manner, like the proverbial snake in the grass (hence the name). The walls of these tubes are themselves defined by all the other polymer molecules in the medium. In this way, the model captures how the mobility of each individual macromolecule is restricted by the spatial distribution of all the others. In this picture, the only way to disentangle such nest of vipers is to pull individual fibers out of their confining tubes, because motion orthogonal to the tube walls is not possible.

"Our extensive computer simulations, however, suggest a very different type of polymer dynamics for biopolymers," says Frey. "We do not observe convoluted motions of individual polymers. Instead, we find relatively rapid, collective reorganization of the tubes, which results in

the disentanglement of polymer chains." According to the authors, the dynamics resembles that of glass-like materials. This kind of behavior is not based on the independent motions of individual polymer molecules, but derives from interactions between polymers on much larger scales. This leads to collective motion of all the polymer chains in a local neighborhood, such that the whole jumble begins to sort itself out, tangled chains are unraveled, the dense ball loosens up, and new paths through the maze are created.

"We have developed a new theoretical concept that can account for the collective dynamics and reproduces the results obtained in our simulations," says Frey. "Our findings will fundamentally change current assumptions concerning the relevance of collective effects not only in biopolymer solutions but potentially also in other soft-matter systems." The theoretical predictions made by the new model should be quite easy to test experimentally in systems involving either biopolymers or carbon nanotubes. Indeed, Lang and Frey have already shown that the data derived from their simulations are in complete agreement with results of a study on carbon nanotubes published by a Dutch-American research group.

More information: Philipp Lang et al. Disentangling entanglements in biopolymer solutions, *Nature Communications* (2018). DOI: 10.1038/s41467-018-02837-5

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