**Dear Karl,**

Please find enclosed in the present submission our manuscript entitled “A novel route to crystallization via viscoelastic phase separation”, that we would like you to consider for publication in Nature as a Letter.

Our Letter represents the first particle-level experimental study of the dynamical interplay between crystallization and phase separation. We find that this crystal growth can be described by the Wegener-Bergeron-Findeisen mechanism. The process occurs naturally in mixed phase clouds, between 0 degree C and -38 degree C, where water vapour, supercooled water and ice crystals coexist, and is responsible for the growth of ice crystals in clouds and rain formation. But we argue that this mechanism of crystal growth is much more common than previously thought, and show that it can be accessed directly in a Soft Matter model system. We study a mixture of poly(methyl methacrylate) colloids with non-adsorbing polymer (polystyrene) with confocal microscopy experiments, that give us access to the full kinetic process with particle resolution. We have built a novel experimental setup that allows the evolution of the system to be observed directly from the very early stages and in absence of spurious fluid flows. This allows us to directly study the dynamics of phase separation and of crystal growth at single-particle resolutions, and for the first time characterize the Bergeron process at a microscopic level.

Understanding the process of crystal growth in mixed-phase systems is of fundamental importance, and has several technological applications. As an example, understanding water crystallization in clouds is recognized as one of the main challenges in the modeling of Earth's radiation budget and climate. Moreover, our study addresses the formation of crystal networks, which are observed in many systems, including magma, biominerals and foods, where the origin and formation of the networks still lacks fundamental understanding. The difficulties arise both from our poor understanding of small-scale microphysical effects, and to the difficulty of experimental investigations. In our colloidal system, both difficulties are overcome due to the colloid's accessible length and time scales.

Our manuscript thus represents a fundamental investigation on one of the most important crystal-growth mechanisms, which will resonate with the broad science community. Moreover, it highlights a very deep connection between the disciplines of Soft Matter and Climate Modeling, and we thus believe that will have broad appeal to the readership of Nature. We hope that you will consider it for publication.

For your convenience, we provide here a list of possible Referees who are widely considered to be experts in the field.

Prof. Daniel Bonn, University of Amsterdam, [D.Bonn@uva.nl](mailto:D.Bonn@uva.nl)

Prof. Daan Frenkel, University of Cambridge, df246@cam.ac.uk

Prof. Henk Lekkerkerker, University of Utrecht, H.N.W.Lekkerkerker@uu.nl

Prof. Francesco Sciortino, University of Rome ``La Sapienza'',

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Prof. Michael Solomon, University of Michigan, mjsolo@umich.edu

Prof. David Weitz, Harvard University, weitz­@seas.harvard.edu

Warm thanks to you for your kind attention.

**Sincerely yours,**

**Hideyo Tsurusawa, John Russo, Mathieu Leocmach, and Hajime Tanaka**