**Dear Bart,**

Please find enclosed in the present submission our manuscript entitled “A novel physical mechanism for the spontaneous formation of porous crystals via viscoelastic phase separation”, that we would like you to consider for publication in *Nature Physics* 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 include the Wegener-Bergeron-Findeisen mechanism. This is known as a major process of rain droplet formation, but its microscopic mechanism remains elusive. 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. Thus, we have succeeded in characterizing the Bergeron process at a microscopic level and revealing the microscopic mechanism of this important process for the first time. We note that understanding water crystallization in clouds is recognized as one of the main challenges in the modelling of Earth's radiation budget and climate.

Furthermore, understanding the process of crystal growth in mixed-phase systems is not only of fundamental importance, but also has many technological applications. The most novel and important point of our paper is the discovery of a general, and potentially widely applicable, method of forming porous crystals (of crystal gels) in a single step. This can be seen clearly in our Figure 2, which compares the usual outcome of colloidal gelation with the new crystal-gel phase. As can be seen by comparing the two panels, crystal-gels are characterized by a new morphology, which is intermediate between the gel state and the crystalline state. These properties come from a novel physical mechanism via the gas phase, i.e., evaporation of the liquid network and de-sublimation of the gas phase, which has so far been completely overlooked. This mechanism allows crystals to outgrow from the percolated liquid network and cover the surface of network by well-defined crystal planes. The last point is particularly important in catalytic applications of noble metal porous crystals. We believe that our finding leads to a conceptual advance in our physical understanding of dynamical coupling between crystallization and phase separation.

The crystal-gel phase has properties that are intermediate between the gel phase and the crystal phase, which makes it extremely interesting for catalytic, optical, super-capacitor, sensing and filtration applications. Now there is a high demand for the formation of crystalline nanoporous structures, and our mechanism provides a novel method to spontaneously form such structures with one step. Besides applications, porous crystals are found in natural phenomena such as magma, biominerals and foods, where its formation mechanism is still being debated, especially due to the inability to follow the kinetics of crystal formation in these systems.

Just to summarize what we believe are the accomplishments of our research:  
  
- novel one-step process of spontaneous porous crystal formation

- first single-particle study of crystal-gel formation

- novel physical mechanism of crystal-gel formation

- suggestion for a general mechanism for crystal-gel formation, which relies only on the

kinetics of the slow-component during viscoelastic phase separation

As explained above, our manuscript thus represents a fundamental investigation on one of the most important crystal-growth mechanisms, which will resonate with the broad Physics community. 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. Walter Kob, University Montpellier, walter.kob@univ-montp2.fr

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

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

We would also like to avoid the following reviewers since there may be a possibility of conflict of interest.

Prof. Wilson Poon, Edinburgh University

Prof. Paul Bartlett, Bristol University

Warm thanks to you for your kind attention.

**Sincerely yours,**

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