Dear Prof. Glotzer,

Please find attached a manuscript titled "*Shape Matters in Magnetic-field Assisted Assembly of Colloidal Ellipsoids"* co-authored by Antara Pal, Carlo Andrea De Filippo, Thiago H. Ito, Md. Arif Kamal, Andrei V. Petukhov, Cristiano De Michele and Peter Schurtenberger. We would like to re-submit this manuscript for publication as a regular article in ACS Nano.

We have submitted this work last year to ACS Nano (ref no.-nn-2020-004642, Title: 'Magnetic-field Assisted Assembly of Colloidal Ellipsoids'). Unfortunately, the manuscript was rejected based on the referee reports, which recommended to corroborate our experimental findings with simulations. Instead of choosing the quick solution and opting for the suggested manuscript transfer to a more specialized journal, we instead decided to follow the constructive criticisms/suggestions provided by the referees. Therefore, we have decided to team up with our collaborators from Rome and gone ahead with a systematic simulation study to uncover the origin of the novel experimental observations reported in the previous version of the manuscript.

The current version of the article now also includes the simulation results, which not only corroborate our experimental findings, but also contribute significantly towards putting the results/claims of our manuscript on a firm footing. In retrospect, we are extremely happy about this decision, as it has not only provided a theoretical basis for our phenomenological observations, but moreover allowed us to uncover the unexpected dramatic effect of small shape imperfections on field-induced self-assembly that will have to be considered in future attempts to compare computer simulations of anisotropic particles with "real-life" systems.

Our detailed MC simulation along with an additional careful particle shape analysis has led us to the intriguing conclusion that subtle variations in the actual particle shape can lead to a much greater diversity in the accessible range of self-assembled phases when compared to the known phase behaviour of simple geometrical shapes. When attempting to design optimized building blocks for directed self-assembly of new high-density phases, a focus on global shape parameters can thus be misleading and result in missed opportunities. We believe these new results to be of significant importance for the readership of ACS Nano and would thus like to re-submit the current version of the manuscript to ACS Nano.

We believe that our manuscript is suitable for publication in ACS Nano for the following reasons:

1. We have demonstrated the occurrence of a counter-intuitive phenomenon where under the influence of an external magnetic field, 'prolate-shaped' colloidal particles self-assemble into a series of 'oblate' liquid crystalline phases. This unusual behaviour stems from the magnetic properties of these particles where their magnetic dipole moments lie in a plane perpendicular to their long axes, making them align with their short axes parallel to an external magnetic field. As a result, one observes oblate self-assembled liquid crystalline phases from prolate-shaped building blocks.
2. Detailed characterization of the concentration dependence of the self-assembled structures for different aspect ratios (ρ) reveals that for smaller ρ, an increase in concentration results in a series of four different anisotropic phases: para-nematic, nematic, smectic and an oriented glass. Out of these four phases, the *smectic* phase is particularly noteworthy as it has neither been predicted nor been reported before for ellipsoidal colloids.
3. In our quest for uncovering the origin of this smectic phase, we have serendipitously stumbled upon a very important yet oft neglected/overlooked aspect when considering field-directed self-assembly of anisotropic colloids – the details of the *actual shape* of the particles. Our study

clearly demonstrates that the final self-assembled structure is extremely sensitive not only to the global shape anisotropy such as aspect ratio, but also to subtle systematic deviations from the ideal shape. Small variations from the ideal shape of the building blocks must be considered because they can give rise to a much greater diversity in the accessible range of self-assembled phases in a real experimental scenario that aims to manipulate self-assembled structures by external fields.

ACS Nano is an interdisciplinary journal that attracts a broad readership from various disciplines, like Material Scientist, Chemists, Physicists, Chemical Engineers, Nanotechnologist to name but a few. We sincerely feel that publishing our results here will engage their attention, and we firmly believe that our study is of considerable interest to a diverse scientific community.

We thank you in advance for your time and your consideration of our resubmission.

Sincerely yours,

Antara Pal and Peter Schurtenberger