Dear Editor(s),

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 submit this manuscript for publication as a regular article in *Advanced Materials.*

To understand and mimic nature’s ability to create higher order structures with novel materials properties, research efforts in contemporary soft matter science has resulted in an increasing shift from the study of the self-assembly of isotropic building blocks to anisotropic ones. The effects of shape on the self-organisation of soft matter systems are believed to be the key to understand, design and control the formation of a next generation of smart materials. Over the last couple of decades, colloidal science has emerged as one of the key players in this quest. Particularly promising here is an approach where the design of anisotropic particles with tuneable functionality and interaction potential is combined with external fields to direct self-assembly, thus creating new routes towards the fabrication of advanced materials.

In our continuing efforts to develop novel routes towards the design and fabrication of advanced materials through field directed self-assembly of anisotropic colloids, we have investigated the self-assembly of *prolate-shaped* ellipsoidal colloids under the influence of an external magnetic field. Our investigations using small-angle x-ray scattering demonstrate that in the presence of an external field, despite their ‘prolate’ shape, these particles self-assemble into a series of ‘oblate’ liquid crystalline phases. Detailed MC simulation along with particle shape analysis 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 that our manuscript is suitable for publication in Adv. Mat. 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. To the best of our knowledge, present articles is the first one to report the formation of oblate self-assembled phases by prolate 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.

Advanced Materials 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 submission.

Sincerely yours,

Antara Pal and Peter Schurtenberger