

Aggregation of finite size particles with variable mobility

D. D. Holm, Los Alamos National Laboratory*

V. Putkaradze, Colorado State University

Summary

A new model equation has been derived for dynamics of aggregation of finite-size particles. Two differences from standard Debye-Hückel (1923) and Keller-Segel (1970) models are:

- (1) the mobility of particles depends on the configuration of their neighbors and*
- (2) linear diffusion acts on locally-averaged particle density, rather than pointwise.*

The evolution of collapsed states in these models reduces exactly to finite-dimensional dynamics of interacting particle clumps. Simulations show these collapsed (clumped) states emerge from smooth initial conditions, even in one spatial dimension. Extensions to 2D and 3D have also been made, but are not discussed here.

In the Applied Math Research program effort at Los Alamos, researchers have derived and analyzed a new fundamental theory for the length-scale competition between a short-range stochastic process and a long range attraction. This is of course a non-Markovian process, which has seen a lot of recent interest in the physics literature. Such a process may be thought of as an ensemble of random walkers which are attracted to each other. This is also related to the classic Poisson-Smoluchowsky model of nucleation and planetary formation. The new idea for physics is the startling effect of the corrected Fokker-Planck flux introduced in this new model to allow coalescence, or quenching, of the stochastic process due to the long-range attraction. Thus, the long-range force controls and eventually quenches the small-scale stochastic process.

Precisely this control is needed in many new technological applications, such as directed assembly in nanoscience. In addition to this

quenching result, we discovered that the dynamics and steady states of these quenched states may be solved *exactly* as the solutions of simple ordinary differential equations. The mathematical derivation of the multiscale equations for competition between short- and long-range forces, as well as the analysis of their evolution and the interactions among their singular solutions are presented in [1].

In this approach, the dynamics of the quenched states may be found as weak solutions of a broad class of new equations arising in a familiar context. These startling new effects are being realized physically in recent new directed assembly processes for nanoscience [2]. These corrections allow for strong coalescence (collapse to delta functions), in the presence of a long-range attraction, even in one dimension (which is a new result), thereby producing the control needed for a new route to extreme nanoscale patterning and functionality. These ideas and

* Computer and Computational Science, Continuum Modeling Group CCS-2
(505) 667-6398 dholm@lanl.gov

results will be guides whenever competition between length-scales occurs in future applied physics contexts, such as bioscience and nanoscience.

Figure 1 shows that the singular solutions emerge spontaneously in a numerical simulation of the new equations. Hence, they are essential in understanding its dynamics.

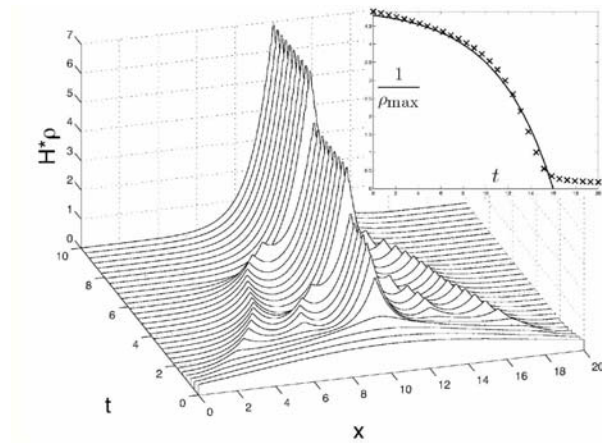


Figure 1: Numerical simulation demonstrates the emergence of particle clumps, showing formation of density peaks in a simulation of the initial value problem starting from smooth initial conditions for density. The vertical coordinate represents the density integrated against a kernel H , which remains finite even when the density forms delta-functions. This result shows the development of singular solutions in finite time, as our theory predicted.

When the mobility saturates at a maximum average density, the analytical stationary solution of our equations corresponds to states with all particles being closely packed at that average density, as emerges in Figure 2. This Figure also demonstrates that the analytical stationary solution is stable, since smooth initial conditions are seen to converge to this solution.

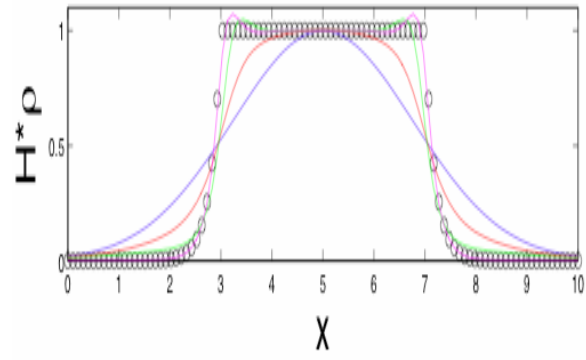


Figure 2: Evolution of a Gaussian initial condition for density with saturating mobility. The solution quickly forms a plateau of maximal possible density. Different colors denote different times in the evolution; the stationary solution is drawn with black ellipses.

References:

- [1] Holm, D. D. and V. Putkaradze, Aggregation of finite size particles with variable mobility. *Phys. Rev. Lett.*, **95** 226106 (2005).
- [2] Mertens, K., V. Putkaradze, D. Xia and S. Brueck, Theory and experiment of nanoparticle self-assembly, *J. Applied Physics*, **98**, 034309 (2005).

Acknowledgements:

Los Alamos National Laboratory Report LA-UR-06-4803. We are grateful for partial support under the auspices of the National Nuclear Security Administration of the U.S. Department of Energy at Los Alamos National Laboratory under Contract No. DE-AC52-06NA25396 from NNSA's Laboratory Directed Research and Development Program and from the DOE Office of Science Advanced Scientific Computing Research (ASCR) Program in Applied Mathematics Research.

For further information on this subject contact:

Dr. Anil Deane, Program Manager
 Mathematical, Information, and Computational
 Sciences Division
 Office of Advanced Scientific Computing Research
 Phone: 301-903-1465
 deane@er.doe.gov