Interfacial Instability and Pattern Formation

Takao Ohta

Department of Physics, The University of Tokyo, Tokyo 113-0033, Japan, and Toyota Physical and Chemical Research Institute, Nagakute, Aichi 480-1192, Japan E-mail address: ohta@daisy.phys.s.u-tokyo.ac.jp

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1. Introduction

Physics of pattern formation is to study mainly the origin of structures and dynamics emerged in non-equilibrium open systems which are defined as a system subjected to steady heat current or material flows so that the state of the system is far from equilibrium. For example, a fluid confined between two parallel horizontal plates and heated below causes convective flows when the temperature difference between the two plates exceeds a certain threshold. This phenomenon is known as the Rayleigh-Benard convection. Roll pattern of this convection, grid pattern in electro-hydrodynamic convection of liquid crystals, concentric wave and spiral wave in chemically reacting systems, and needle-like and dendritic crystal growth are the examples of non-equilibrium patterns. In fact, Physics out of equilibrium has been developed towards understanding these experiments for these four decades (Cross and Greenside, 2009).

2. Dynamics of Interface

When two different states (such as solid and liquid) coexist, a spatial structure is constituted in a homogeneous system. An interface is a boundary separating the areas occupied by these two states. Since the location of a flat interface is arbitrary in an extended system having a translational invariance, interface deformation with long wave length is a relevant relaxation mode which governs the slow dynamics of the system. This is one of the reasons why we are concerned with the interface dynamics as a powerful method for pattern formation problems. Not only the time scale but also the spatial scale associated with an interface should be separated enough from other degrees of freedom such as the velocity field in the convection and the concentration fields in chemical reactions. That is, the interface is infinitesimally thin and should be regarded as a geometrical boundary without any internal structures. A systematic theory to reduce the degrees of freedom by relying on this kind of scale difference has been developed mathematically (Nishiura, 2003; Pismen, 2006).

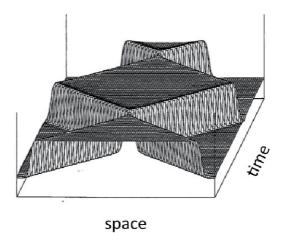


Fig. 1. Elastic-like collisions of interfaces in a reaction-diffusion system (Ohta and Kiyose, 1996). Because of the Neumann boundary condition, a collision with the interface at the mirror position also occurs at the system boundaries.

Instabilities of a flat interface enrich nonequilibrium spatial patterns. One of the most well known examples is the Mullins-Sekerka instability in crystal growth. An interface separating solid and liquid tends to be flat to diminish the interfacial energy. However, latent heat is produced in a growing crystal surrounded by a supercooled liquid. If a bump of solid is formed, the diffusion of the latent heat is more efficient in the bump region so that the local temperature decreases there and crystallization is accelerated. When this effect dominates the increase of the interfacial energy due to the bump, a flat interface becomes unstable. A mathematically equivalent interfacial instability is the one at a boundary between two fluids in two parallel glass plates where less viscous fluid pushes more viscous fluid. It is also possible that the Marangoni effect in which the interfacial energy depends on local concentration and/or local temperature causes an interfacial instability. In fact, a hexagonal convective pattern in Benard convention, which appears for a free upper boundary is due to the Marangoni effect.

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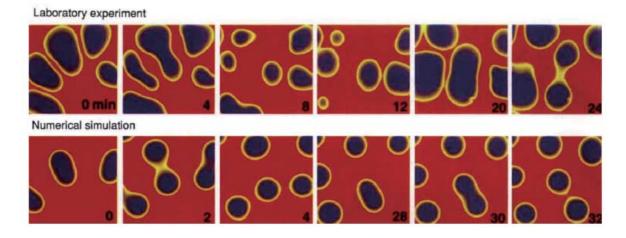


Fig. 2. Self-replication of localized domains (Lee *et al.*, 1994). Upper column: snapshots of domains obtained by experiments of the ferrocyanide-iodate-sulphite reaction. Lower column: snapshots of domains obtained by computer simulations of the reaction-diffusion system. The location of interfaces is indicated by the grey lines. This figure is reproduced with permission from Nature Publishing Group.

3. Recent Theories

In 1951, the theory was introduced for spiral growth of crystal surface where a dislocation core is a nucleation center of steps (Burton et al., 1951). This is one of the earliest studies of non-equilibrium phenomena where interface motions are involved. Later, in the second half of 1970s, the interface dynamics was formulated for domain growth in order-disorder transitions and phase separations in alloys (see Nishiura (2003)). As a most recent research in this direction, theories of molecular membranes have been introduced to describe mesoscopic structural formation and its kinetics in soft matter (Taniguchi et al., 2011). Pattern selection in crystal growth is one of the examples where the interface dynamics was mostly successful. The shape and the growth velocity of a tip of dendritic crystal are determined uniquely in experiments when the external condition such as the temperature and the degree of super-saturation is fixed. The mechanism of tip growth was, however, unknown theoretically for a long time. In the second half of 1980s, it was clarified that small anisotropy of the surface tension acts as a manner of singular perturbation (Pecle, 1988) to select the shape and the velocity at the tip.

4. Computer Simulations

The interaction between interfaces plays a fundamental role to the dynamics of ordered structures far from equilibrium. Since a non-equilibrium system is a dissipative dynamical system, it was believed for many years that a pair of interfaces (or waves) annihilate upon collision as observed experimentally in concentration waves of Belousov-Zhabotinsky reaction. However, recent studies by computer simulations of reaction-diffusion equations and complex Ginzburg-Landau equations have revealed that the dynamics of interfaces and spatially localized structures (called pulses) can be complex much more than expected. There are many findings that interfaces and pulses in one and two dimensions behave, upon collision, as an elastic object or preserve their shape like a soliton in integrable conserved systems (Petrov et al., 1994; Krischer and Mikhailov, 1994). Figure 1 shows a space-time evolution

of a concentration obtained by numerical simulations of a reaction-diffusion system. It is evident that a pair of interfaces undergoes repeatedly elastic-like collisions. However, it has been clarified by an interface dynamics that a supercritical bifurcation from a motionless state to a propagating state (either to the right or to the left depending on the initial conditions) of an interface, in other words, breaking of mirror symmetry is essential for this collision dynamics (Ohta and Kiyose, 1996).

Another interesting dynamic pattern formation is selfreplication of a pulse. One pulse splits into two pulses and after growing to a certain size, they replicate again spontaneously. This phenomenon was discovered in computer simulations of reaction-diffusion equations (Petrov *et al.*, 1994) and later, a similar domain splitting was found in experiments of the ferrocyanide-iodate-sulphite reaction (Lee *et al.*, 1994). In Fig. 2, the self-replication of domains obtained by laboratory experiments is compared with that by numerical simulations in two dimensions.

The soliton-like behavior and self-replication of pulses indicate that one has to take into account generally the internal degrees of freedom of a pulse as well as the position and the velocity of the center of mass. To formulate the interface dynamics including these relevant variables is one of the important future problems.

It is mentioned here that, after the Japanese version was published, an important progress has been made for the interface dynamics of the Kardar-Parisi-Zhang equation both theoretically and experimentally (Takeuchi and Sano, 2010).

References

- Burton, W. K., Cabrera, N. and Frank, F. C. (1951) The growth of crystals and the equilibrium structure of their surfaces, *Phil. Trans. Roy. Soc.*, A243, 299–358.
- Cross, M. and Greenside, H. (2009) Pattern Formation and Dynamics in Nonequilibrium Systems, Cambridge.
- Krischer, K. and Mikhailov, A. (1994) Bifurcation to traveling sopts in reaction-diffusion systems, *Phys. Rev. Lett.*, 73, 3165.
- Lee, K.-J., McCormick, W. D., Pearson, J. E. and Swinney, H. L. (1994) Experimental observation of self-replicating spots in a reactiondiffusion system, *Nature*, **369**, 215–218.

Nishiura, Y. (2003) Far-From Equilibrium Dynamics, AMS.

- Ohta, T. and Kiyose, J. (1996) Collision of domain boundaries in a reaction-diffusion system, J. Phys Soc. Jpn., 65, 1967–1970.
- Pecle, P. (ed) (1988) Dynamics of Curved Fronts, Academic Press.
- Petrov, V., Scott, S. K. and Showalter, K. (1994) Excitability, wave reflection and wave splitting in a cubic autocatalysis reaction-diffusion system, *Philos. Trans. R. Soc. London*, Ser. A347, 631.
- Pismen, L. M. (2006) Patterns and Interfaces in Dissipative Dynamics, Springer.
- Takeuchi, K. A. and Sano, M. (2010) Universal fluctuations of growing interfaces: evidence in turbulent liquid crystals, *Phys. Rev. Lett.*, 104, 230601.
- Taniguchi, T., Yanagisawa, M. and Imai, M. (2011) J. Phys. Cond. Matt., 23, 284103.