Letter

Hopf bifurcation in addition-shattering kinetics

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In aggregation-fragmentation processes, a steady state is usually reached. This indicates the existence of an attractive fixed point in the underlying infinite system of coupled ordinary differential equations. The next simplest possibility is an asymptotically periodic motion. Never-ending oscillations have not been rigorously established so far, although oscillations have been recently numerically detected in a few systems. For a class of addition-shattering processes, we provide convincing numerical evidence for never-ending oscillations in a certain region $\mathcal{U}$ of the parameter space. The processes which we investigate admit a fixed point that becomes unstable when parameters belong to $\mathcal{U}$ and never-ending oscillations effectively emerge through a Hopf bifurcation.

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Two complementary processes, aggregation and fragmentation, are widespread in nature [1–10]. Mathematically, a well-mixed system undergoing aggregation and fragmentation is described by equations

$$\frac{dc_k}{dt} = \frac{1}{2}\sum_{i+j=k} K_{ij}\, c_i\, c_j - c_k \sum_{j\geqslant 1} K_{kj}\, c_j + \sum_{j\geqslant 1} F_{kj} c_{j+k} - \frac{1}{2} c_k \sum_{i+j=k} F_{ij}. \qquad (1)$$

Here $c_k(t)$ denotes the density of clusters composed of k monomers, $K_{ij} = K_{ji} \geqslant 0$ is the rate of aggregation

$$[i] \oplus [j] \xrightarrow{K_{ij}} [i+j], \qquad (2)$$

and $F_{ij} = F_{ji} \geqslant 0$ is the rate of binary fragmentation

$$[i+j] \xrightarrow{F_{ij}} [i] + [j]. \qquad (3)$$

The system (1) of infinitely many nonlinear ordinary differential equations (ODEs) is analytically intractable apart from a few special cases. The long-time behavior is easier to probe. If the mass distribution becomes stationary, then one may guess the stationary distribution by equating the rate of the aggregation process $[i] \oplus [j] \to [i+j]$ to that of the reverse fragmentation process $[i+j] \to [i] + [j]$. The detailed balance condition, $K_{ij}\, c_i\, c_j = F_{ij}\, c_{i+j}$, however, is an overdetermined system that does not possess a solution [11] apart from a few exceptional cases [12].

More rich stationary states have been found in some systems amenable to analysis, e.g., in addition to a stationary distribution of finite clusters an infinite cluster comprising a finite fraction of mass of the entire systems is sometimes formed (see Refs. [13–17]). Some aggregation-fragmentation processes are characterized by unlimited growth, namely the typical cluster mass diverges in the long-time limit. Nonthermodynamic behaviors and nonequilibrium phase transitions have been also observed [11,18]. These complicated behaviors reflect the peculiarities arising in infinitely many ODEs.

Persistent oscillations have been numerically observed in Ref. [19] for some open aggregating systems driven by input at small masses and sink at large masses. Oscillations could be caused by the drive, however. In closed systems, never-ending oscillations have been numerically detected in a class of processes with collision-controlled fragmentation where each fragmentation event leads to complete shattering of colliding clusters into monomers:

$$[i] \oplus [j] \xrightarrow{S_{ij}} \underbrace{[1] + \cdots + [1]}_{i+j}. \qquad (4)$$

Since the binary collision can lead to aggregation or shattering, the reaction rates that differ only by an amplitude, $S_{ij} = \lambda K_{ij}$, have been explored [20–25]. For the family of rates $K_{ij} = (i/j)^a + (j/i)^a$, never-ending oscillations have been detected [21,22] for $\frac{1}{2} < a \leqslant 1$ and $0 < \lambda \leqslant \lambda_c(a)$.

We analyze a slightly simpler class of processes and provide much stronger evidence for never-ending oscillations. We consider systems in which each aggregation event involves at least one monomer:

$$[1] \oplus [s] \xrightarrow{A_s} [1+s]. \qquad (5)$$

This naturally occurs if only monomers are mobile as it happens, e.g., in monolayer growth [26,27]. The shattering is assumed to be spontaneous,

$$[s] \xrightarrow{B_s} \underbrace{[1] + \cdots + [1]}_{s}, \qquad (6)$$

as opposed to the collision-induced shattering (4). The governing equations read

$$\frac{dc_s}{dt} = c_1[A_{s-1}c_{s-1} - A_s c_s] - B_s c_s, \quad s \geqslant 2, \tag{7a}$$

$$\frac{dc_1}{dt} = \sum_{s\geqslant 2}^{\infty} sB_s c_s - 2A_1 c_1^2 - c_1 \sum_{s\geqslant 2} A_s c_s. \tag{7b}$$

The system is closed, so the mass density is conserved:

$$M = \sum_{s=1}^{\infty} sc_s(t) \equiv \text{const}. \tag{8}$$

There is no natural relation between spontaneous shattering rates B_s and collision-controlled addition rates A_s. For instance, pure addition processes with rates $A_s = s^a$ have been investigated in Refs. [28,29].

We take $A_s = s$ (cf. Ref. [30]) and recast (7a)–(7b) into

$$\frac{dc_s}{dt} = c_1[(s-1)c_{s-1} - sc_s] - B_s c_s, \quad s \geqslant 2, \tag{9a}$$

$$\frac{dc_1}{dt} = \sum_{s\geqslant 2}^{\infty} sB_s c_s - c_1^2 - Mc_1. \tag{9b}$$

Without loss of generality, one can set $M = 1$. This can be achieved by rescaling

$$c_s \mapsto Mc_s, \quad B_s \mapsto MB_s, \quad t \mapsto M^{-1}t.$$

Suppose the system falls into a steady state. Equation (9a) gives $c_s = c_{s-1}(s-1)/(s + B_s/c_1)$, leading to

$$\frac{c_s}{c_1} = \prod_{j=2}^{s} \frac{j-1}{j + B_j/c_1}. \tag{10}$$

The mass density

$$1 = \sum_{s\geqslant 1} sc_s = c_1 \sum_{s\geqslant 1} s \prod_{j=2}^{s} \frac{j-1}{j + B_j/c_1} \tag{11}$$

implicitly determines c_1. The right-hand side in Eq. (11) increases monotonically with c_1, so there is at most one steady-state solution.

The above results are rather formal, so we specialize them to a class of models with algebraic break-up rates

$$B_s = Bs^\beta, \quad \beta \geqslant 0. \tag{12}$$

When $\beta = 0$, the rate equation (9b) is closed

$$\frac{dc_1}{dt} = B(1 - c_1) - c_1^2 - c_1. \tag{13}$$

The stationary density of monomers is therefore

$$c_1 = \frac{b - 1 - B}{2}, \quad b \equiv \sqrt{B^2 + 6B + 1}, \tag{14}$$

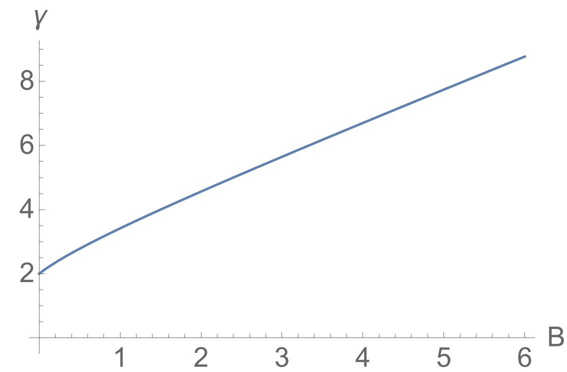

FIG. 1. The plot of the exponent γ given by (16).

while (10) simplifies to

$$\frac{c_s}{c_1} = \frac{\Gamma(s)\Gamma(2 + B/c_1)}{\Gamma(s + 1 + B/c_1)}. \tag{15}$$

This mass distribution has an algebraic tail, $c_s \propto s^{-\gamma}$ for $s \gg 1$, with

$$\gamma = 1 + \frac{B}{c_1} = \frac{b - 1 + B}{b - 1 - B}. \tag{16}$$

The exponent γ is an increasing function of the amplitude B. Starting from $\gamma = 2$ for $B = 0$, the exponent $\gamma(B)$ grows asymptotically as $B + 1$ for $B \gg 1$; see Fig. 1.

We emphasize that (14) is a stable fixed point for the monomer density. If $c_1(0) = 1$, then the explicit expression for the monomer density reads

$$c_1(t) = c_1 + \frac{b(1 - c_1)^2}{2\, e^{bt} - (1 - c_1)^2} \tag{17}$$

with $c_1 \equiv c_1(\infty)$ and b given by (14). The remaining equations (9a) can be rewritten as

$$\frac{dn_s}{d\tau} = (s-1)n_{s-1} - sn_s, \quad s \geqslant 2 \tag{18}$$

with $n_s(\tau) = e^{Bt}\, c_s(t)$ and $\tau = \int_0^t dt'\, c_1(t')$. These equations with already known $n_1(\tau) = e^{Bt}\, c_1(t)$, where $c_1(t)$ is given by (17), can be solved recurrently from which one verifies the stability of the fixed point (14) and (15).

For the break-up rates (12) with $\beta > 0$, we extract from (10) the asymptotic behaviors

$$\frac{c_s}{c_1} \propto \begin{cases} (c_1/B)^s (s!)^{-(\beta-1)} & \beta > 1 \\ s^{-1} \exp[-s^\beta B/\beta c_1] & 0 < \beta < 1 \end{cases} \tag{19}$$

for $s \gg 1$. A qualitative change happens at $\beta = 1$ where one obtains more precise results:

$$\frac{c_s}{c_1} = s^{-1}(1 + B/c_1)^{1-s}, \quad c_1 = \frac{\sqrt{B^2 + 4B} - B}{2}. \tag{20}$$

The stability of the steady state is difficult for theoretical analysis when $\beta > 0$. Owing to mass conservation, the sets of equal-mass size distributions are invariant for (9a) and (9b) and each of them can be considered as phase space (when we talk about the birth of limit cycles we always confine the system to distributions of fixed mass). We write $c_s(t) = c_s +$

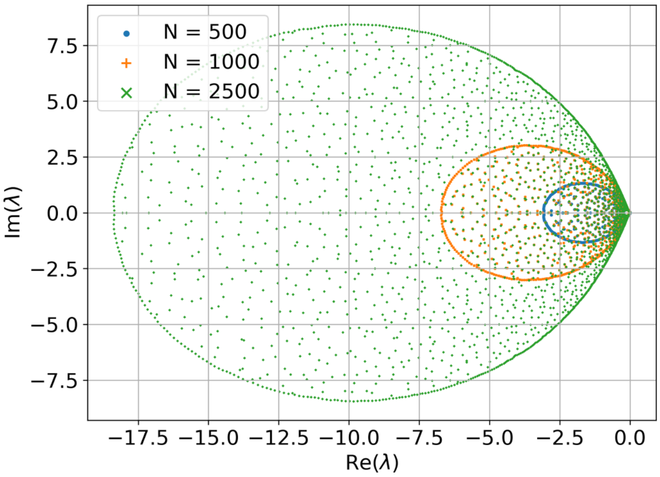

FIG. 2. The eigenvalues of the linearized truncated system of N equations with $\beta = 2$, $B = 10^{-7}$, and $M = 1$. The eigenvalues have negative real parts and concentrate close to zero, with possible exceptional pairs of eigenvalues that have positive real parts. Such pairs appear in the region $\mathcal{U}$ of the parameter space, and they are responsible for oscillations.

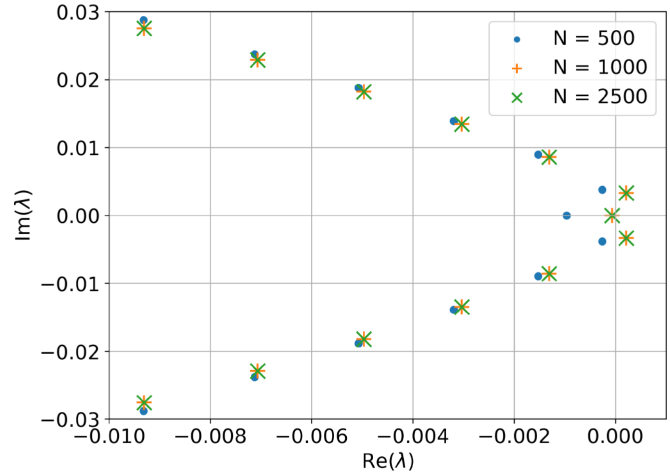

FIG. 3. The eigenvalues near $\lambda = 0$, the same parameters as in Fig. 2. The pair of eigenvalues with a positive real part that causes oscillations is visible and is present when N is sufficiently large. For $N = 500$ the truncated steady state of the infinite system loses its mass rapidly (22) and is not an approximate steady state of the truncated system, and hence the corresponding eigenvalues have no physical meaning.

$x_s(t)$ with $\sum_{s\geqslant 1} s x_s(t) = 0$ due to mass conservation, linearize Eqs. (9a) and (9b) and get

$$\frac{dx_s}{dt} = c_1[(s-1)x_{s-1} - s x_s] + B_s\left[\frac{c_s}{c_1}x_1 - x_s\right] \tag{21a}$$

for $s \geqslant 2$ and

$$\frac{dx_1}{dt} = -(2c_1 + 1)x_1 + \sum_{s\geqslant 2} s B_s x_s. \tag{21b}$$

The eigenvalues of this infinite system determine stability of the steady state (10). To probe them numerically, we truncate our original infinite system (7a) and (7b) by keeping the first N equations. For this finite system, mass is no longer conserved (8); instead, it decays:

$$\frac{dM_N}{dt} = \frac{d}{dt}\left(\sum_{s=1}^{N} s c_s\right) = -N(N+1)c_1 c_N. \tag{22}$$

As a result, the finite system has a single steady state, the zero one. However, given the asymptotic behavior (19) of the original steady state (10), we can choose N to make dM_N/dt arbitrarily small (below machine precision) when evaluated on the first N components of (10). In that case $(c_s)_{s=1}^N$ is, numerically, a steady state and the eigenvalues of the truncated Eqs. (21a) and (21b) do provide insight into its stability. When N is not sufficiently large, $(c_s)_{s=1}^N$ cannot be considered a steady state and the eigenvalues lose physical meaning.

In Figs. 2 and 3, we show the eigenvalues (computed with standard LAPACK routines [31]) for fixed physical parameters and different values of N: The eigenvalues have negative real parts with maybe a few exceptional complex conjugate pairs with positive real parts. A single pair is visible in Fig. 3 for $N = 1000$ and $N = 2500$ but not for $N = 500$. Turning to (22), we obtain

$$\frac{dM_N}{dt} \approx \begin{cases} -3.5\times 10^{-4} & N = 500 \\ -6.5\times 10^{-8} & N = 1000\,. \\ -2.2\times 10^{-35} & N = 2500 \end{cases} \tag{23}$$

Hence the results for $N = 500$ are physically meaningless, while those for $N = 1000$ and $N = 2500$ coincide near $\lambda = 0$ and represent the actual behavior.

The unstable pairs are present in a certain region in the parameter space

$$\mathcal{U} = \{(\beta, B)\,|\, \beta > 1,\ 0 < B < B_{\text{crit}}(\beta)\}. \tag{24}$$

The steady state loses stability via Hopf bifurcation when B crosses the critical value $B_{\text{crit}}(\beta)$ and enters $\mathcal{U}$. This leads to the birth of a stable limit cycle. The imaginary part of the critical eigenvalue decreases monotonically, ignorant to the bifurcation, as B decreases (Fig. 4). The real part changes its

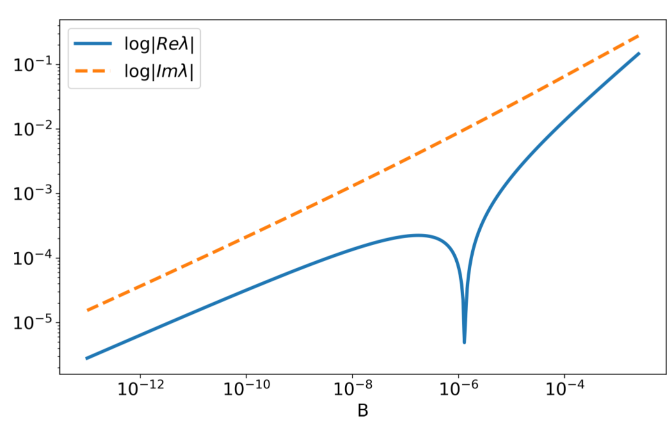

FIG. 4. Real and imaginary parts of the eigenvalue that crosses the imaginary axis as B varies with $\beta = 2$ and $M = 1$ fixed. The cusp in the plot of $\log|\text{Re}\lambda|$ corresponds to Hopf bifurcation: The critical eigenvalue changes its sign, and a limit cycle is born.

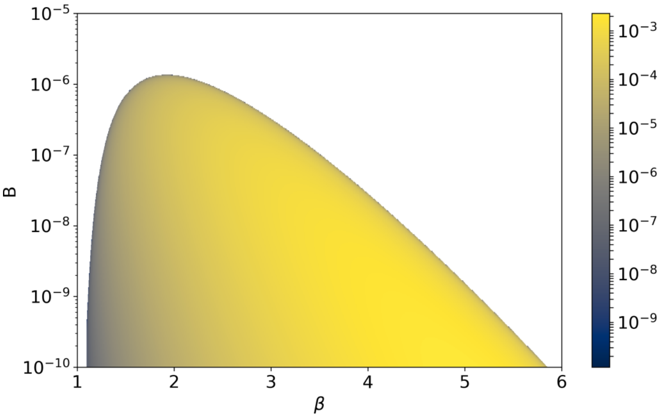

FIG. 5. The region $\mathcal{U}$ of the (β, B) plane where unstable eigenvalues exist and oscillations are born for $M = 1$.

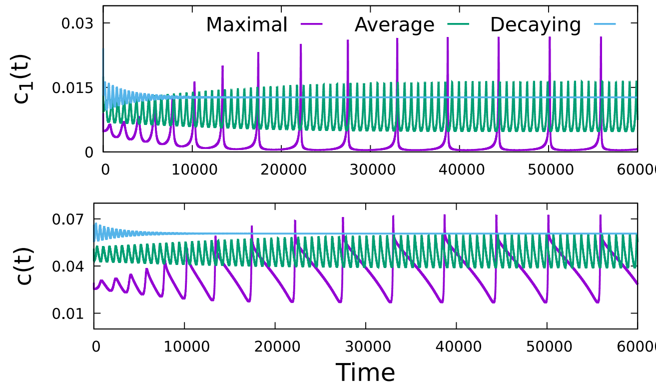

FIG. 6. Oscillatory regimes for monomers $c_1(t)$ and total density $c(t) = \sum_{k=0}^{\infty} c_k$ for different values of B with $\beta = 2$ and unit mass density. The oscillations decay when $B = 3.1622776602 \times 10^{-6}$ (Decay), persist when $B = 1.2195704602 \times 10^{-6}$ (Average), and have the largest possible (for $\beta = 2$ and unit mass) amplitude when $B = 1.668100537 \times 10^{-7}$ (Maximal). In all three cases we used initial conditions as in (25).

behavior once the eigenvalue becomes unstable: $\mathrm{Re}(\lambda)$ keeps growing for a short while before reaching its maximum value and then decays monotonically.

Figure 5 shows the transition curve $B_{\rm crit}(\beta)$ in the parameter space. In particular, it shows that there is a singularity at $\beta = 1$, whose existence is connected with the qualitative changes in the steady state (19) and (20). In our numerical experiments, we exploit the structure of the Jacobian and use the inverse power method [32] with absolute numerical tolerance of 10^{-10} to find and track unstable eigenvalues (see Supplemental Material [33] for details). Note that while we tracked only the first complex conjugate pair that crosses the imaginary axis, more pairs can appear in $\mathcal{U}$ as the parameters change.

We carried out numerical integrations to study the oscillatory solutions of the system truncated to N equations, with N sufficiently large to ensure that mass conservation holds on each iteration with machine precision. (The size N used in numerical integrations was larger than that used to compute the eigenvalues because the oscillatory waveform has a longer tail than the steady state.) This makes the finite system numerically indistinguishable from the infinite one. The results are presented in Fig. 6 for multiple values of B with $\beta = 2$. The initial condition was taken as a perturbation of the steady state (10)–(19) that preserves its mass density:

$$\tilde{c}_1 = c_1 + 1.8c_2, \quad \tilde{c}_2 = 0.1c_2, \quad \tilde{c}_s = c_s. \tag{25}$$

Comparing Figs. 4 and 6 we see that the oscillations die out when B is above the critical value $B_{\rm crit}(2)$ and persist when B is below it. As B continues to decrease, the amplitude of the oscillations at first grows, reaches its maximum, and starts decaying to zero. The frequency of the oscillations decreases monotonically with B.

In our numerical experiments, we have observed that Eqs. (9a) and (9b) cease to have unstable eigenvalues for parameters from the region (24) when N is not big enough (see Fig. 3). The "big enough" grows as B tends to zero or β tends to 1, together with the effective length of the stationary distribution.

At another extreme, one can consider Eqs. (7a) and (7b) and set $A_s = 0$ for $s \geqslant 3$. Finding limit cycles is difficult even for such simple systems of two coupled ODEs. Several tools allow one to rule out the limit cycles or prove their existence [34–36]. In our case, the application of the Dulac criterion shows the absence of limit cycles independently of the rates (see Supplemental Material [33]). Recent results on Hopf bifurcation in a finite Becker-Döring exchange model also show that the number of ODEs in such finite systems has to be sufficiently large to obtain oscillatory solutions [37,38]. This perhaps explains why despite years of searching, the oscillatory solutions have not been observed.

To summarize, we have found oscillatory solutions in the realm of addition-shattering models (9a) and (9b) with algebraic break-up rates (12). These solutions are born through the Hopf bifurcation mechanism: The steady states exist whenever $\beta \geqslant 0$ but become unstable for parameters from (24) and give birth to never-ending oscillations via Hopf bifurcation. Oscillatory solutions in other models have been detected recently [21,22,37]. For instance, Hopf bifurcation has been found in a finite Becker-Döring system with constant kinetic coefficients [37,38]. Our infinite system with algebraically growing rates also exhibits oscillatory solutions, at least the numerical evidence is very convincing.

In a class of addition-shattering processes that we investigated, persistent oscillations occur in a small region of the phase space; the same holds for the model studied in Refs. [21,22]. This rarity is similar to the empirical evidence with limit cycles in planar systems with quadratic polynomials: The rule of thumb is that a "generic" planar system has no limit cycles (see Ref. [39]). The same seemingly holds for aggregation-fragmentation systems. Limit cycles are very rare [40–45], and systems with more than one limit cycle are currently unknown. Another avenue for future work is to seek oscillations in systems with standard binary fragmentation. Among the biggest challenges is providing rigorous proof of persistent oscillations in an infinite system and finding chaos.

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