A diffusion-induced transition in the phase separation of binary fluid mixtures subjected to a temperature ramp

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Abstract – Demixing of binary fluids under slow temperature ramps shows repeated waves of nucleation which arise as a consequence of the competition between generation of supersaturation by the temperature ramp and relaxation of supersaturation by diffusive transport and flow. Here, we use an advection-reaction-diffusion model to study the oscillations in the weak- and strong-diffusion regime. There is a sharp transition between the two regimes, which can only be understood based on the spatial distribution of the composition, rather than in terms of the average composition. Our results shed light on the parameter drift and secondary features observed in phase separating fluids subjected to a temperature ramp, and they bear intriguing communalities with macroscopic oscillations due to synchronization of life cycles in ageing populations.

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Introduction. – Liquid-liquid phase separation occurs whenever an isotropic binary mixture is transferred into a bi-phasic region where its isotropic state is no longer stable, and the mixture decomposes into two equilibriums phases. In classical approaches, phase separation was studied after sudden temperature quenches, in constant temperature conditions ([1] and references therein). Recent research explores parallels to systems showing pattern formation [2], and addresses the influence of hydrodynamic effects [3] on phase separation. On the other hand, motivated by many industrial [4] and natural [5–7] applications in which temperature is not constant, many works focus on phase separation subjected to sustained temperature variations [8–15]. In particular, demixing of fluid mixtures subjected to slow temperature ramps continues to provide challenges for fundamental research [5,10–13] since it shows a rich variety of unexpected non-equilibrium phenomena. For instance, rather than continuously following the gradual change in temperature, the phase separation can exhibit consecutive bursts of droplet nucleation and rapid growth alternating with so-called quiescent periods where there are few droplets. In the latter periods supersaturation is slowly building up, until it is released in another burst.

The strength of diffusive transport plays a critical role in the alternation of the bursts and quiescent periods of phase separation because the diffusive mass flux onto the droplets competes with the buildup of supersaturation by the change of temperature [11,15]. The importance of understanding its role has been emphasised by recent experimental work [12] that reported strong changes of the time lag between consecutive bursts. It was speculated that this might be related to considerable changes in the diffusion coefficient, $D$, that rises monotonously [16] as the system departs from the critical point.

Here, we aim at a better understanding of the consequences of this parameter drift. To this end we investigate systematically the behaviour of the oscillations in different (weak- and strong-) diffusion regimes. Upon increasing the diffusion coefficient we find a clearly defined transition in the features of the oscillations—in particular in their period.

The model. – At any given temperature, $T$, a pure substance can dissolve at most a concentration $\phi_0(T)$ of another, minority component, and upon a change of temperature the surplus material will be removed by nucleation of droplets. Subsequently, the droplets coalesce in the background flow $\mathbf{u}$ of the continuous phase. Surprisingly, in response to a slow continuous temperature change, repeated waves of nucleation and subsequent coagulation
Its decay rate evolution can be based on the composition, towards droplets. A convenient description \[11–15\] of this arises \[9,10\]. The period of these waves was related \[11\] to \(\zeta_t\)Benczik and Jürgen Vollmer other cells we account for the changes of the normalized \(\sigma\). This choice reflects i) that for \(\sigma_{th}\) spinodal decomposition causes instantaneous phase separation \[13,15\], and ii) that above a threshold value, \(\sigma_{th}\), the mixture is so close to equilibrium that practically no nucleation events occur. Our results do not depend on the particular choice of \(a(\sigma)\) as long as these criteria are met.

The droplets are advected by the background flow \(\mathbf{u}\) as passive tracers which can merge when they approach each other. To follow the evolution of the composition and the droplets we use a numerical model with a prescribed flow.

Numerical implementation. – We consider the coarse-grained composition field \(\sigma(i,j)\) defined on an \(N \times N\) lattice in a square of size \(L\) with periodic boundaries. There is no supersaturation in cells containing droplets, i.e., their composition is fixed to \(\sigma_0 = 1\). In all other cells we account for the changes of the normalized composition due to the temperature ramp by applying a “reaction” step after each time increment \(\tau\),

\[
\sigma(i,j) \rightarrow \sigma(i,j) - \xi \tau \sigma(i,j).
\]

This step is followed by a diffusion step according to the transformation (see \[17\]),

\[
\sigma(i,j) \rightarrow \sigma(i,j) + D \tau \cdot D(i,j),
\]

where \(D(i,j) = -\sigma(i,j) + [\sigma(i+1,j) + \sigma(i-1,j) + \sigma(i,j+1) + \sigma(i,j-1)]/4\) is the discrete Laplace operator, and \(D\) the lattice diffusion coefficient. In the continuum limit \((\tau \rightarrow 0, \varepsilon = L/N \rightarrow 0)\) this \(D\) corresponds to a diffusion coefficient \(D = D\varepsilon^2/4\).

Next, we nucleate droplets in cells that are far from equilibrium \((\sigma < \sigma_{th})\) with a probability \(a(\sigma)\) given by eq. (2). Upon nucleation the composition is reset to \(\sigma_0 = 1\) in the cell where nucleation took place, as well as in the eight neighboring cells.

The background flow \(\mathbf{u}\) in eq. (1) mixes the composition field \(\sigma(i,j)\) and the droplets. In our model this is achieved by an incompressible, periodically alternating shear flow \[18\] that produces chaotic trajectories \[19\],

\[
\mathbf{u}(i,j,t) = \frac{AL}{T} \sin \frac{2\pi j}{N} \hat{e}_x, \quad \text{for } t \bmod T < \frac{T}{2},
\]

\[
\mathbf{u}(i,j,t) = \frac{AL}{T} \sin \frac{2\pi i}{N} \hat{e}_y, \quad \text{for } \frac{T}{2} < t \bmod T,
\]

where \(\hat{e}_x\) and \(\hat{e}_y\) are the unit vectors in the \(x\)- and \(y\)-direction, respectively. The period \(T\) of the flow and its strength \(A\) are characteristic quantities characterizing the correlation time and mixing strength of the flow (cf. \[18\]).

Finally, we take into account the merging of droplets: whenever two droplets approach each other to within a distance \(r_0\), the two droplets are replaced by a new droplet placed in the centre of mass of the two droplets.

Motivated by experiments \[12\] where the droplet distance is \(\varepsilon \approx 100 \mu m\) and the system size is \(L \approx 1 cm\), we choose \(N = L/\varepsilon = 100\). We measure length in units of \(L\), time in units of \(T\), and perform simulations with \(T = T/16\) on a lattice with \(N \times N = 10^5\) sites. In addition we note that in the absence of droplets, neither nucleation nor coagulation occurs above the threshold \(\sigma_{th}\). Thus, all initially uniform configurations \(\sigma(i,j,0) > \sigma_{th}\) lead to the same evolution up to a trivial time shift accounting for the initial decay towards \(\sigma_{th}\). Consequently, as initial condition we choose a uniform composition field \(\sigma(i,j,0) = 0.667\) without droplets.

Oscillations in the weak- and strong-diffusion regimes. – To explore how the increasing diffusion coefficient modifies the oscillations of the droplet density we fix the flow rate, the decay rate of the composition and the droplet interaction radius to \(A = 0.8\), \(\xi = 0.04\), and \(r_0 = \varepsilon\), respectively, and study the oscillations for diffusion coefficients ranging from \(D = 0.05\) to \(D = 0.55\).

The oscillations are qualitatively different for low, intermediate and high values of the diffusion coefficient. In the weak-diffusion regime, fig. 1(a), the period of the oscillations increases with increasing \(D\), while their amplitude decreases faster than in the diffusionless case. As the diffusion coefficient increases further, this tendency stops at a
Fig. 1: (Colour on-line) Evolution of the droplet density $n_d$ for $A = 0.8$ and $\xi = 0.04$ in the (a) weak- ($D = 0.05$, red line; $D = 0.1$, green line), (b) intermediate- ($D = 0.14$, 0.15 and 0.16 in decreasing order of the amplitudes), and (c) strong- ($D = 0.25$, 0.35, 0.45, and 0.55 in decreasing order of the periods) diffusion regimes. All simulations were started with a uniform initial composition field $\sigma_0 = 0.667$. The black line represents the diffusionless case. Note the appearance of secondary oscillations in panel (b).

Fig. 2: (Colour on-line) Diffusion induced transition in the oscillatory behaviour of the phase separating mixture. For $\xi = 0.04$ and $A = 0.8$ the transition occurs in the highlighted region: $0.1 < D < 0.2$, determined as the region where secondary oscillations are observable. In the transition region: (a) the oscillations period reaches its maximal value; (b) the decay rate $\gamma_{\text{amplit}}$ of the oscillation amplitude reaches its maximum; and (c) the decay rate $\gamma_{\sigma}$ of the average composition $\sigma_{av}$ decreases suddenly from $\gamma_{\sigma} \approx 0.04$ to $\gamma_{\sigma} \approx 0.03$.

certain value of $D$. For intermediate values of $D$ (fig. 1(b)) secondary oscillations appear. Subsequently, the period of the oscillations becomes smaller again (fig. 1(c)), and the decay rate of the amplitude decreases with increasing $D$.

The appearance of secondary oscillations in the intermediate-diffusion regime is accompanied by noticeable changes (cf. fig. 2) of (a) the oscillations period, (b) the decay rate of the oscillation amplitude, and (c) the decay of the composition.

We quantify the decay of the amplitude by following how the difference between the maximum droplet density and its average value $\Delta n_d \equiv n_{d, \text{max}} - n_{d, \text{av}}$ evolves in time. For each diffusion coefficient, we observed an exponential decay of the amplitude with rate $\gamma_{\text{amplit}}(D)$,

$$\Delta n_d \sim \exp[-\gamma_{\text{amplit}}(D) \cdot t]$$

(6)

As a function of the diffusion coefficient $D$ (fig. 2(b)), this decay rate has an extremum localized at the end of the diffusion range in which secondary oscillations are present. The fastest decay of the amplitude corresponds to that value of $D$ at which secondary oscillations disappear from the system. For higher diffusion coefficients, the exponent $\gamma_{\text{amplit}}$ decreases slightly. Then it seems to saturate.

The most abrupt change can be observed in the behaviour of the average composition in the system. We observe that in the quiescent period of each oscillation the average composition decreases exponentially as

$$\sigma_{av} \sim \exp[-\gamma_{\sigma}(D) \cdot t].$$

(7)

In the absence of diffusion onto droplets this decay is determined by the local decay, $\sigma(x) \sim \exp(-\xi \cdot t)$, of the composition in individual cells, such that $\gamma_{\sigma} = \xi$. When droplets are present in the system the diffusion of supersaturation into the droplets slows down the decay. For small values of the diffusion coefficient the magnitude of this effect is small, and the average composition still decays with a rate $\gamma_{\sigma} \approx \xi$. However, when the diffusivity is increased $\gamma_{\sigma}$ drops abruptly to a much lower value when the secondary oscillation appear (fig. 2(c)).

Hence, the region where secondary oscillations occur represents a transition regime between the weak- and strong-diffusion regimes, where the oscillation period takes its maximal value $D_{tr}$. To determine the dependence of $D_{tr}$ on the parameters of the system, we also run numerical simulations for different decay rates $\xi$, and different flow rates $A$. The position of the transition point is not affected significantly by the flow. However, it is very sensitive to the decay rate $\xi$: the transition occurs at $D_{tr} \approx 0.16$ for $\xi = 0.04$, at $D_{tr} \approx 0.08$ for $\xi = 0.02$, and at $D_{tr} \approx 0.04$ for
\( \xi = 0.01 \). This is compatible with a linear scaling

\[ D_{tr} \simeq 4\xi. \tag{8} \]

The diffusion coefficient \( D \) and the decay rate \( \xi \) define the length scale \( \Lambda_{tr} = \sqrt{D/\xi} \) at which the effects of the temperature ramp and the effects of diffusion are comparable. Diffusion can efficiently relax the supersaturation accumulated by the continuous temperature ramp up to a length scale of order \( \Lambda \). In the simulations, we find for the transition from the weak to the strong diffusion

\[ \Lambda_{tr} = \sqrt{\frac{D_{tr}}{\xi}} = \sqrt{\frac{D_{tr} \varepsilon^2}{4 \xi}} = \varepsilon. \tag{9} \]

Hence, the transition occurs when the range in which diffusion can relax the accumulated supersaturation amounts to the droplet length scale \( \varepsilon \). In accordance with experimental results (e.g., fig. 3(a) in [11]) it does not depend on the flow rate \( A \).

Distributions of supersaturation. – In the intermediate- and strong-diffusion regimes nucleation waves are initiated by a small fraction of high-supersaturation “spots” that reach the nucleation threshold, while the average composition \( \sigma_{av} \) stays above \( \sigma_{th} \) (see fig. 3, left panels). This calls for a description that deals with the full distribution of the composition rather than only with the average composition. In this section we focus therefore on the evolution of the probability distribution functions (pdf-s) of the composition in the different diffusion regimes. We obtain the pdf-s by counting the fraction of lattice cells with composition falling in different bins of size \( \delta \sigma = 1/200 \). The results are presented in fig. 3 for different time instances in the weak- (upper panels), intermediate- (middle panels), and strong- (bottom panels) diffusion regimes, respectively.

The uniform initial composition field \( \sigma_{0} = 0.667 \) corresponds in the pdf-s to a \( \delta \)-peak that moves to the left due to the temperature ramp. When it crosses the nucleation threshold \( \sigma_{th} = \frac{2}{3} \), it gives rise to a nucleation wave. The cells in which nucleation took place move up to \( \sigma = 1 \). In the panels (1a), (2a), and (3a) of fig. 3 we show the distributions at the end of the first nucleation wave. Regardless of the diffusion regime the behaviour of the \( \sigma \)-field is similar in all the three cases: after several cells nucleate droplets the distribution becomes bimodal. There is a big number of cells being already around the equilibrium value \( \sigma_{0} = 1 \), while a small number of cells remain slightly above the threshold and form a second peak. This small peak is subjected to two opposing forces. Diffusion (red arrows (ii) in fig. 3) tends to move the peak to the right, i.e., into the direction of the average composition that is already close to equilibrium. The second force is that of the temperature ramp (black arrows (i) in fig. 3). It favours nucleation, i.e., it drives the peak further to the left in the direction of the nucleation threshold. The competition between the effects of diffusion and temperature ramp determines the fate of the small peak. Accordingly, the character of the oscillations differs in the three diffusion regimes.

In the weak-diffusion regime the effects of diffusion are small ((ii)<(i) in fig. 3, panel (1a)). Thus, the temperature ramp will dominate, and the peak will slowly cross the threshold line \( \sigma_{th} \) and nucleate droplets. This delayed nucleation results in a long tail of the main
peak, modifying its Gaussian shape (fig. 3, panel (1b)). The following oscillations (fig. 3, panels (1c) and (1d)) will take place according to the same scenario. The slowing down is more and more efficient as the diffusion coefficient increases. Consequently, the duration of the nucleation wave increases, explaining the increase of the oscillation period in the weak-diffusion regime. The slow decay of the oscillations is due to the broadening of the peak.

In the intermediate-diffusion range the effects of the temperature drift and of diffusion balance each other ((ii)≈(i) in fig. 3, panel (2a)). The small peak is arrested from nucleation and stopped in the close vicinity of the nucleation threshold without actually reaching it. Meanwhile, the continuous ramp of temperature moves the big peak to lower levels such that the effect of diffusion, (ii), becomes smaller. Eventually, the small peak manages to cross the nucleation level. This happens after the first nucleation wave was finished and gives rise to a separate small wave of nucleation. In this way, a secondary oscillation of the droplet density (as shown in fig. 1(b)) occurs in the system. After this secondary nucleation wave a distinct small peak forms close to $\sigma = 1$ which does not attach to the big peak. Both peaks maintain their Gaussian character while travelling downward (fig. 3, panel (2b)), maintaining the bimodal distribution of the composition. In the next wave of nucleation, the bigger peak breaks again in two distinct peaks, forming a trimodal distribution. The number of peaks increases by one at each subsequent principal or secondary nucleation wave. Thus, the distribution converges rapidly to a flat distribution in which the standard deviations of the individual peaks are larger than the distances between them. Subsequently, phase separation proceeds continuously rather than in distinguishable oscillations. This explains the rapid increase of $\gamma_{\text{amplit}}$ upon increasing the diffusion coefficient $D$.

In the strong-diffusion regime the effects of diffusion overcome the effects of the temperature ramp ((ii)> (i) in fig. 3, panel (3a)). The small peak is not only arrested from nucleation, but it moves upward towards the big peak. The big peak in turn, travels downward in the direction of the threshold because of the intense diffusive exchange. Soon the two peaks merge (fig. 3, panel (3b)) and form again one single peak with an (almost) Gaussian shape (fig. 3, panel (3c)). The next nucleation waves follow the same scenario (fig. 3, panel (3d)). In this regime, there is always a certain number of cells in which the supersaturation is released by diffusive transport. For high values of $D$, the downward travel of the big peak is accelerated by the rapid diffusive exchange with the small peak. It hence moves faster than in the weak-diffusion regime where this motion was caused by the temperature ramp alone. As a result the period of the oscillations decreases.

When starting the simulations with a random initial condition instead of a uniform $\sigma$-field this picture does not change because diffusion tends to synchronize the composition. In the strong-diffusion regime the synchronization of lattice cells is so fast that the character (period, amplitude) of the oscillations is hardly affected by the initial condition. Even in the weak-diffusion regime the oscillations are still clearly observable. Their period remains almost the same, but their amplitude is strongly reduced as compared to the case of the uniform initial condition.

**Discussion and conclusions.** – The understanding of oscillatory demixing of binary fluids under slow temperature ramps has recently improved considerably due to precision experiments [10,12], and theoretical descriptions [11,13–15]. Nevertheless, the parameter dependence of the oscillations period is still an open question. Secondary effects appear even in well-controlled experiments and shield the principal behaviour of the system. For instance, a) typically the oscillation period decreases during an experimental run [10–12] b) bimodal droplet size distributions occur [6,20], and c) for compositions very close to the critical point also secondary oscillations can appear [21]. Our model system allows us now to identify mechanisms that provide a straightforward explanation of these secondary effects. To make the comparison we observe that in typical experimental settings, where oscillatory demixing due to slow temperature ramps has been studied, one has $D > 10^{-11} \text{m}^2/\text{s}$ and $\xi < 10^{-3} \text{s}^{-1}$ such that the resulting diffusion scale, $\Lambda = (D/\xi)^{1/2} \approx 10^{-4} \text{cm}$ is just agreeing with the droplet spacing. In the terminology of the present paper typical data therefore amount to strong diffusion.

a) Our results, fig. 2(a), show that in the strong-diffusion regime the period of the oscillations decreases considerably when the diffusion coefficient is increased. This has indeed been observed in experiments where the diffusion coefficient increases as the system further departs from the critical point (fig. 10 in [10]; fig. 3 in [11]; fig. 15 in [12]).

b) Nucleation arises at those locations (cells), where the maximum supersaturation first reaches the nucleation threshold, and the maximum supersaturation is typically vastly different from its average value (fig. 3). In particular, droplets may be formed and start to grow at different time instances during an oscillation, which can then lead to bimodal droplet size distributions as they have been observed in [6] (fig. 3) and [20] (fig. 5.5).

c) Secondary oscillations have been observed in some experiments where the system is very close to the critical point such that the diffusion coefficient is exceedingly small [21]. In our model they arise in the crossover regime, where the length scale in which diffusion can relax the accumulated supersaturation matches the droplet scale (fig. 1(b)). It will be a challenge for forthcoming experiments to explore whether this regime can be approached systematically by running faster temperature ramps in systems with a very small diffusion coefficient.

We thus conclude that exploring secondary effects which arise from the interplay of diffusion and the non-trivial
distribution of supersaturation provides an important step to a better qualitative understanding of the dynamics of phase separation under slow temperature ramps. In order to arrive also at a quantitative description, the effects of droplet sedimentation will be added in forthcoming work. Here we conclude with pointing out that due to the general formulation of the model our present results sheds light on a range of other problems, too.

State-of-the-art models for droplets undergoing irreversible coagulation in the presence of a net volume flux onto droplets without [9,11] and with [22] removal of large particles adopt a Smoluchowski approximation for the collision kernel and calculate the flux based on a mean-field approximation for the supersaturation in the system. The cluster size distribution in these models performs oscillations that are analogous to the ones presented here, and it was pointed out [22] that this would be relevant for a description of gravitational clustering in astrophysics and of the differential sedimentation of falling water droplets in clouds. Our present results complement these studies by an analysis of the role of the spatial distribution of the supersaturation in such systems. Doing so we provided an explicit mechanism that can generate binary size distributions of the supersaturation. Forthcoming work will explore how these non-trivial features of the distributions act back on the droplet growth which is presently modeled based on only the mean-field averages. In such a setting, where also sedimentation of droplets can be taken into account, it will become possible to also quantitatively compare model predictions to experimental data of, e.g., [12].

Another intriguing set of questions arises from an interpretation of our model in terms of ageing populations: consider each fluid element (lattice cell) as an individual entity following its own life cycle. The supersaturation, \(1 - \sigma\), measures its age. It increases from zero (at equilibrium) to the value between \(1 - \sigma_{15}\) and \(1 - \sigma_{50}\) when the cell dies, and gives birth to several new entities with initial age (supersaturation) zero, and subsequently, these offspring follow the same life cycle. Such a dynamics has been encountered in a study of phytoplankton species [23], where the cells age with a constant maturation velocity until they die by cell division, giving birth to a finite number of offsprings. In both cases the cells cannot sense their mutual phase difference. Interaction is rather mediated by a mean field, namely by the droplet density in our model, and the nutrient concentration in [23].

Both the numerical and experimental findings of [23] are analogous to the results presented here: macroscopic oscillations at the population level, bimodal size distributions ([23], fig. 2), and relaxation oscillations ([23], fig. 3), including secondary oscillations ([23], fig. 1) were observed in the dynamics of the population. This connection seems to suggest that macroscopic oscillations are universal features that typically occur in growing populations following internal life cycles.

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