Non-Boussinesq Convection at Low Prandtl Numbers: Hexagons and Spiral Defect Chaos

Santiago Madruga and Hermann Riecke

1Department of Engineering Sciences and Applied Mathematics, Northwestern University, Evanston, IL 60208, USA

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We study the stability and dynamics of non-Boussinesq convection in pure gases (CO\textsubscript{2} and SF\textsubscript{6}) with Prandtl numbers near \(Pr \simeq 1\) and in a H\textsubscript{2}-Xe mixture with \(Pr = 0.17\). Focusing on the strongly nonlinear regime we employ Galerkin stability analyses and direct numerical simulations of the Navier-Stokes equations. For \(Pr \simeq 1\) and intermediate non-Boussinesq effects we find reentrance of stable hexagons as the Rayleigh number is increased. For stronger non-Boussinesq effects the hexagons do not exhibit any amplitude instability to rolls. Seemingly, this result contradicts the experimentally observed transition from hexagons to rolls. We resolve this discrepancy by including the effect of the lateral walls. Non-Boussinesq effects modify the spiral defect chaos observed for larger Rayleigh numbers. For convection in SF\textsubscript{6} we find that non-Boussinesq effects strongly increase the number of small, compact convection cells and with it enhance the cellular character of the patterns. In H\textsubscript{2}-Xe, closer to threshold, we find instead an enhanced tendency toward roll-like structures. In both cases the number of spirals and of target-like components is reduced. We quantify these effects using recently developed diagnostics of the geometric properties of the patterns.

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I. INTRODUCTION

Rayleigh-Bénard convection of a horizontal fluid layer heated from below is a paradigmatic system for the study of complex spatial and spatio-temporal patterns. Usually, the system parameters are chosen such that variations of the fluid properties across the layer are very small. In the theoretical description this allows the Oberbeck-Boussinesq (OB) approximation, in which the temperature dependence is neglected in all fluid properties except for the density term that is responsible for the buoyancy. For large temperature differences across the layer the variation of the fluid properties with the temperature become significant. These non-Oberbeck-Boussinesq (NB) effects break the up-down symmetry that is characteristic of the OB approximation and, thus, allow a resonant triad interaction among three Fourier modes whose wavevectors form a hexagonal planform. Due to the resonant interaction the primary bifurcation to the hexagons is transcritical, and hexagons are preferred over rolls in the immediate vicinity of onset.

Within the framework of the leading-order weakly nonlinear analysis, hexagons typically become unstable to rolls further above threshold, where the amplitudes are larger and the resonant-triad interaction loses significance compared to interactions involving four modes. This scenario of a transition from hexagons to rolls has been confirmed in quite a number of experimental investigations and quite commonly it has been assumed that hexagon patterns in NB convection are confined to the regime close to onset.

Two convection experiments using SF\textsubscript{6} as the working fluid have shown, however, that even in the strongly nonlinear regime stable hexagon patterns can be observed. Under OB conditions hexagons were found at relatively high Rayleigh numbers, \(\epsilon \equiv (R - R_c)/R_c \approx 3.5\). Due to the mid-plane symmetry of OB convection hexagons with up-flow in the center coexist stably with down-flow hexagons in this regime. In experiments using SF\textsubscript{6} under NB conditions the hexagons that appear at threshold were replaced by rolls for somewhat larger heating and then reappeared in the strongly nonlinear regime near \(\epsilon = \mathcal{O}(1)\). The restabilization was attributed to the large compressibility of SF\textsubscript{6} near its critical point. The hexagons that regain stability were termed reentrant hexagons.

Recent numerical computations have demonstrated that hexagons can restabilize in NB convection even if the fluid is incompressible. For instance, in water hexagons that are unstable at \(\epsilon = (R - R_c)/R_c = 0.15\) can already restabilize at \(\epsilon \approx 0.2\). The origin of this reentrance was traced back to the existence of stable hexagons in OB convection at large Rayleigh numbers, and the dependence of the NB effects on the Rayleigh number.

At low Prandtl numbers \((Pr \simeq 1)\) and further above onset OB convection exhibits a new state: spiral defect chaos (SDC). It was first found experimentally and then investigated theoretically using simple models as well as simulations of the full fluid equations. This fascinating state of spatio-temporal chaos is characterized by rotating spirals with varying numbers of arms and of different size, which appear and disappear irregularly and interact with each other and with other defects. SDC arises from the roll
state at a threshold that can be as low as \( \epsilon = 0.1 \) in the limit of small \( Pr \). It is driven by large-scale flows that are induced by roll curvature and have a strength that is proportional to \( Pr^{-1} \).

So far, strongly non-linear NB convection has been studied mostly for large Prandtl numbers [16, 24, 25], but little is known for small ( \( Pr \approx 1 \)) or very small Prandtl numbers ( \( Pr \ll 1 \)). In particular, whether reentrant hexagons exist at large \( \epsilon \) in the presence of the large-scale flows that arise at low \( Pr \), and how NB effects impact spiral defect chaos are interesting questions, which we address in this paper.

Here we study NB convection in gases with small Prandtl numbers. Specifically, we consider parameters corresponding to convection in CO\(_2\) and SF\(_6\) ( \( Pr \approx 0.8 \)) and in a H\(_2\)-Xe mixture ( \( Pr = 0.17 \)). We show that reentrant hexagons are possible in convection in CO\(_2\). For spiral defect chaos in SF\(_6\) we find that NB effects promote small convection cells (‘bubbles’). In H\(_2\)-Xe, closer to threshold, roll-like structures dominate. In both cases the NB effects reduce the spiral character of the pattern. We quantify the impact of the NB effects on spiral defect chaos using geometric diagnostics that we have proposed recently [26].

The paper is organized as follows. In Sec. II we briefly review the basic equations, emphasizing how our computations focus on weakly non-Boussinesq effects, but strongly nonlinear convection. In Sec. III we present the results for the linear stability of hexagons and rolls in CO\(_2\) for a range of parameters accessible experimentally. To compare with experiments, we study the influence of different lateral boundary conditions on the transition from hexagons to rolls in Sec. IV. In Sec. V we discuss spiral defect chaos in SF\(_6\) under NB conditions. The stability of hexagons and spiral defect chaos in fluids with very low Prandtl number ( \( Pr = 0.17 \)) is studied in a mixture of H\(_2\) and Xe in Sec. VI. Finally, conclusions are drawn in Sec. VII.

### II. BASIC EQUATIONS

The basic equations that we use for the description of NB convection have been discussed in detail previously [16, 24]. We therefore give here only a brief summary. We consider a horizontal fluid layer of thickness \( d \), density \( \rho \), kinematic viscosity \( \nu \), heat conductivity \( \lambda \), thermal diffusivity \( \kappa \), and specific heat \( c_p \). The system is heated from below (at temperature \( T_1 \)) and cooled from above (at temperature \( T_2 < T_1 \)).

To render the governing equations and boundary conditions dimensionless we choose the length \( d \), the time \( d^2/\kappa_0 \), the velocity \( \kappa_0/d \), the pressure \( \rho_0 \nu_0 \kappa_0 / d^2 \), and the temperature \( T_e = \nu_0 \kappa_0 / a \rho_0 g d^3 \) as the respective scales. The subscriptsed quantities refer to the respective values at the middle of the fluid layer in the conductive state. The non-dimensionalization gives rise to two dimensionless quantities: the Prandtl number \( Pr = \nu / \kappa_0 \), and the Rayleigh number \( R = \alpha_0 \Delta T g d^3 / \nu \kappa_0 \). Furthermore, we write the equations in terms of the dimensionless momentum density \( \bar{v}_i = \rho \bar{u}_i / \rho_0 \kappa_0 \) instead of the velocities \( \bar{u}_i \).

The dimensionless form of the temperature \( \bar{T} = T / T_e \), heat conductivity \( \bar{\lambda} = \lambda / \lambda_0 \), density \( \bar{\rho} = \rho / \rho_0 \), kinematic viscosity \( \bar{\nu} = \nu / \nu_0 \), and specific heat \( \bar{c}_p = c_p / c_{p0} \) will be used in the ensuing equations and the hats dropped for clarity. In dimensionless form the equations for the momentum, mass conservation and heat are then given, respectively, by

\[
\frac{1}{Pr} \left( \partial_t \bar{v}_i + \nu \bar{v}_j \partial_j \left( \frac{\bar{u}_i}{\rho} \right) \right) = -\partial_i p \\
+ \delta_3 \left( 1 + \gamma_1 (-2z + \frac{\Theta}{R}) \right) \Theta \\
+ \partial_j \left[ \nu \bar{v}_i \left( \partial_i \frac{\bar{u}_j}{\rho} + \partial_j \frac{\bar{v}_i}{\rho} \right) \right] \\
\partial_j \bar{v}_j = 0, \\
\partial_t \Theta + \frac{\bar{v}_j}{\rho} \partial_j \Theta = \frac{1}{\rho c_p} \partial_j (\lambda \partial_j \Theta) - \gamma_3 \partial_z \Theta - \frac{R \bar{v}_z}{\rho} (1 + \gamma_3 z). \tag{3}
\]

with the dimensionless boundary conditions

\[
\bar{v}(x, y, z, t) = \Theta(x, y, z, t) = 0 \text{ at } z = \pm \frac{1}{2}. \tag{4}
\]

Here \( \Theta \) is the deviation of the temperature field from the basic conductive profile. Summation over repeated indices is assumed.

We consider the NB effects to be weak and retain in a Taylor expansion of all material properties only the leading-order temperature dependence beyond the OB approximation. For the density this implies also a quadratic term with coefficient \( \gamma_1 \). It contributes, however, only to the buoyancy term in (2); in all other expressions it would constitute only a quadratic correction to the leading-order NB effect. Thus, the remaining temperature dependence of the fluid parameters \( \rho, \nu, \lambda, \) and \( c_p \) in (2) is taken to be linear

\[
\rho(\Theta) = 1 - \gamma_0 (-z + \frac{\Theta}{R}), \\
\nu(\Theta) = 1 + \gamma_2 (-z + \frac{\Theta}{R}), \\
\lambda(\Theta) = 1 + \gamma_3 (-z + \frac{\Theta}{R}), \\
c_p(\Theta) = 1 + \gamma_4 (-z + \frac{\Theta}{R}). \tag{5-8}
\]

The coefficients \( \gamma_i \) give the difference of the respective fluid properties across the layer. They depend therefore linearly on the Rayleigh number,

\[
\gamma_i(\Delta T) = \gamma_i^c \left( \frac{R}{R_e} \right) = \gamma_i^c (1 + \epsilon), \tag{9}
\]
where $\gamma_n^c$ is the value of $\gamma$ at the onset of convection and $\epsilon \equiv (R - R_c(\gamma_n^c))/R_c(\gamma_n^c)$ is the reduced Rayleigh number.

In analogy to (2), we further omit NB terms that contain cubic nonlinearities in the amplitudes $v_i$ or $\Theta$, as they arise from the expansion of the advection terms $v_i \partial_j (v_j/p)$ and $(v_j/p) \partial_j \Theta$ when the temperature-dependence of the density is taken into account. Since we will be considering Rayleigh numbers up to twice the critical value, which implies enhanced NB effects, these approximations may lead to quantitative differences compared to the fully NB system, even though the temperature-dependence of the material properties themselves is in most situations well described by the linear (or quadratic in the case of the density) approximation.

To quantify the overall strength of the NB-effects we use Busse’s NB parameter $Q$, which is given by

$$Q = \sum_{i=0}^{4} \gamma_i^c \mathcal{P}_i,$$  \hspace{1cm} (10)

where the quantities $\mathcal{P}_i$ are linear functions of $Pr^{-1}$. The NB parameter $Q$ quantifies the breaking of the up-down symmetry, which renders at most one of the two types of hexagons stable. Gases have a positive value of $Q$ and exhibit hexagons with down-flow in the center (g-hexagons), whereas liquids have negative $Q$ and show hexagons with up-flow (l-hexagons).

We focus in this paper on the stability properties of patterns in the strongly nonlinear regime. They are determined by a Galerkin method (e.g. [27]). We use a Fourier expansion on a hexagonal lattice in the lateral directions. The Fourier wave vectors $\mathbf{q}$ are constructed as linear combinations of the hexagon basis vectors $\mathbf{b}_1 = q(1,0)$ and $\mathbf{b}_2 = q(1/2, \sqrt{3}/2)$ with $\mathbf{q} = m\mathbf{b}_1 + n\mathbf{b}_2$ where the integers $m$ and $n$ are in the range $|m\mathbf{b}_1 + n\mathbf{b}_2| \leq n_q q$. The largest wavenumber is then $n_q q$ and the number of Fourier modes retained is given by $1 + 6 \sum_{j=1}^{n_q}$ typically we use $n_q = 3$. The top and bottom boundary conditions are satisfied by using appropriate combinations of trigonometric and Chandrasekhar functions in $z$ ([2, 28]). In most of the computations we use $n_z = 6$ modes for each field in Eq. (2,2,3). The linear analysis yields the critical Rayleigh number $R_c$ as well as the critical wavenumber $q_c$. Both depend on the NB-coefficients $\gamma_n^c$ which in turn depend on $R_c$. Thus, in principle one obtains an implicit equation for the $\gamma_n^c$. The shift in the critical Rayleigh number away from the classical value $R_c = 1708$ due to the NB-effects is, however, quite small (less than 1 percent) and therefore the resulting change in the $\gamma_n^c$ is negligible. In this paper we therefore choose the $\gamma_n^c$ corresponding to $R_c = 1708$.

To investigate the nonlinear hexagon solutions, we start with the standard weakly nonlinear analysis to determine the coefficients of the three coupled amplitude equations for the modes making up the hexagon pattern. To obtain the fully nonlinear solutions requires the solution of a set of nonlinear algebraic equations for the expansion coefficients with respect to the Galerkin modes.

This is achieved with a Newton solver for which the weakly nonlinear solutions serve as convenient starting solutions. In the Galerkin code amplitude instabilities are tested by linear perturbations of the expansion coefficients. In addition, modulational instabilities are considered, which involve the introduction of Floquet multipliers $\exp(is \cdot (x, y))$ in the Fourier ansatz for the linear perturbations of the Galerkin solutions.

We also study the temporal evolution of the system. For that we employ a Fourier spectral code on a rectangular grid $(i dq_x \cdot j dq_y)$, $i, j = 1...N$, with $dq_x/dq_y = \sqrt{3}/2$ to accommodate perfect hexagonal patterns. The same vertical modes are used as in the Galerkin stability code [22]. To solve for the time dependence a fully implicit scheme is used for the linear terms, whereas the nonlinear parts are treated explicitly (second order Adams-Bashforth method). The time step is typically taken to be $t_v/500$, where $t_v$ is the vertical diffusion time. We have tested that the stability regimes obtained from the Galerkin analysis are consistent with the direct numerical simulations. Both codes employed in this paper have been kindly provided by W. Pesch [22, 24].

### III. REENTRANT HEXAGONS IN CO$_2$

In this paper we investigate specific scenarios for convection in gases that should be experimentally realizable. We focus in this section on CO$_2$ in a conventional range for the layer thickness, pressure, and temperature. Table II provides the NB coefficients and the $Q$-value at the onset of convection for a representative range of the mean temperature $T_0$ in a layer of thickness $d = 0.08$ cm.

| $T_0$ | $\Delta T_0$ | $Pr$ | $\gamma_0^c$ | $\gamma_1^c$ | $\gamma_2^c$ | $\gamma_3^c$ | $\gamma_4^c$ | $Q$ |
|-------|-------------|------|--------------|--------------|--------------|--------------|--------------|-----|
| 29    | 0.43        | 0.87 | 0.0486       | -0.0669      | 0.0794       | 0.0246       | -0.0254      | 1.199 |
| 40    | 15.52       | 0.84 | 0.0686       | -0.0883      | 0.1132       | 0.0598       | -0.0185      | 1.724 |
| 60    | 23.80       | 0.82 | 0.0931       | -0.1148      | 0.1566       | 0.0919       | -0.0074      | 2.430 |

#### A. Amplitude instabilities

In our analysis we first concentrate on spatially periodic solutions with their wavenumber being fixed at the critical wavenumber and discuss their domains of existence and their stability with respect to amplitude instabilities, which do not change the wavenumber. For three different cells with thicknesses $d = 0.07, 0.08,$ and 0.09 cm, respectively, we consider the range $0 < \epsilon < 1$ at a pressure of $P = 300$ psi.
The results of the stability analysis for hexagons and rolls are shown in Fig. 1. The hexagons are linearly stable for very small $\epsilon$. For a given layer thickness and not too high mean temperature $T_0$ the hexagons become unstable as the control parameter is increased. The hexagon patterns then undergo a second steady bifurcation as the control parameter is increased further and become stable again. Such restabilized hexagons have been termed 

reentrant hexagons $^{14,15,16}$. As the mean temperature is increased or the layer thickness is decreased the critical heating and with it the NB effects increase. This shifts the point of reentrance to lower $\epsilon$ and the lower stability limit to higher $\epsilon$, decreasing the $\epsilon$-range over which the hexagons are unstable, until the two limits merge at a temperature $T_m$. For $T_0 > T_m$ the hexagons are amplitude-stable over the whole range of $\epsilon$ considered ($0 \leq \epsilon \leq 1$).

We have also computed the stability of rolls with respect to amplitude perturbations. The corresponding stability limits are indicated in Fig. 1 by thin lines. Rolls are stable above these lines. As the NB effects become stronger the stabilization of rolls is shifted to larger $\epsilon$. In contrast to the hexagons, the rolls do not undergo a second bifurcation within the parameter regime investigated and remain amplitude-stable beyond $\epsilon = 1.0$. For strong NB effects one has therefore a large range of parameters over which the competing rolls and hexagons are both linearly amplitude-stable.

The amplitude-stability limits of the hexagons and rolls depend on their wavenumber. This is illustrated for the hexagons in Fig. 2 for a mean temperature of $T_0 = 40^\circ C$. The instability region with respect to amplitude perturbations forms a bubble-like closed curve, inside of which the hexagons are unstable with respect to amplitude perturbations.

It is worth mentioning that the stability limits for hexagons in CO$_2$ are quite similar to those of NB convection in water, except that in CO$_2$ the NB effects increase rather than decrease with increasing mean temperature $^{16}$.

B. Side-Band Instabilities

In systems with sufficiently large aspect ratio side-band instabilities can be the most relevant instabilities. Using the Galerkin method, we have studied the stability of the hexagons with respect to long- and short-wave side-band perturbations for $T_0 = 40^\circ C$. The results are shown in Fig. 2. We find that over the whole range $0 \leq \epsilon \leq 1$ the only relevant side-band perturbations are long-wave and steady, as is the case in the weakly nonlinear regime. The same is true also in water with higher Prandtl number $^{16}$. In this parameter regime the stability region consists of two disconnected domains, reflecting the reentrant nature of the hexagons. The stability domain near onset is very small and closes up as the amplitude stability limit is reached. In the reentrant regime the stable domain opens up again in an analogous fashion when the amplitude-stability limit is crossed. Note that the stability boundaries are leaning toward lower wavenumbers. Thus, stable reentrant hexagon patterns are expected to
have wavenumbers below $q_c$. This is related to the fact that in the OB case hexagons can be stable for large $\epsilon$, but they are side-band stable only for small wavenumbers\(^3\).

As the mean temperature is increased the bubble of the amplitude instability shrinks and eventually disappears, as shown in Fig. 4 for a cell with thickness $d = 0.052\, \text{cm}$ and mean temperature $T_0 = 27^\circ\text{C}$. As before, the relevant side-band instabilities are long-wave all along the stability limits and with increasing $\epsilon$ the wavenumber range over which the hexagons are stable shifts towards smaller wavenumbers. For these parameters the region of side-band-stable hexagons reaches without interruption from the strongly nonlinear regime all the way down to threshold. For yet stronger NB effects the range of stable wavenumbers widens.

IV. COMPARISON WITH EXPERIMENTS IN CO\(_2\)

Bodenschatz et al.\(^{12}\) carried out a set of experiments on convection in CO\(_2\) in a cylindrical cell with aspect ratio $\Gamma \sim 172$, thickness $d = 0.052\, \text{cm}$, and pressure $P = 300\, \text{psi}$. Under these conditions NB effects are relevant. In the experiments a weakly hysteretic transition from hexagons to rolls was found near $\epsilon = 0.1$. Noting that this transition point was below the amplitude instability of hexagons to rolls as predicted by weakly nonlinear theory, the authors interpreted their results in terms of the heterogeneous nucleation of rolls by the sidewalls. They found that for small $\epsilon$ the concentric rolls induced by the sidewall heating remained confined to the immediate vicinity of the sidewalls; however, as $\epsilon$ was increased the rolls invaded the hexagons and filled the whole cell, inducing a transition from hexagons to rolls.

A comparison of the experimental findings with the stability results shown in Fig. 4 shows that indeed the transition cannot be due to an amplitude instability of the hexagons. In fact, in this regime the NB effects are so strong that, in contrast to the predictions of the weakly nonlinear theory, the hexagons do not undergo an amplitude instability at all. To clarify the influence of the sidewalls and to assess the significance of the side-band instabilities for the transition from hexagons to rolls, we perform direct simulations of the Navier-Stokes equations for two different sets of boundary conditions

i) periodic boundary conditions,

ii) concentric rolls as boundary conditions. In our computations this type of boundary condition is generated by a suitably patterned heating in the interior of the fluid. In the experiments concentric rolls near the side walls were generated by a side-wall heating\(^{12}\).

i) according to Fig. 4 for $\epsilon = 0.3$ hexagons with wavenumbers larger than $q > 2.98$, which includes the critical wavenumber, are unstable with respect to side-band instabilities. To test whether these side-band instabilities trigger a transition to rolls we perform numerical simulations with periodic boundary conditions and hexagons as initial conditions. Fig. 4 presents some snapshots of the ensuing temporal evolution in a cell of size $L = 8 \cdot 2 \pi / q_c = 16.11$ using $128 \times 128$ Fourier modes. More precisely, to allow perfect hexagons in a rectangular container the container cannot be square. In our simulations we use $L_x = L$ and $L_y = \sqrt{3}L/2$. The sideband instability of the initially almost perfect hexagon pattern (cf. Fig. 4a)) induces a shearing of the pattern (cf. Fig. 4b)). At the same time a few penta-hepta defects arise and some hexagonal convection cells tend to connect with each other forming short roll-like structures. However, as time evolves the system becomes progressively more ordered again and eventually after losing a number of convection cells a defect-free hexagon pattern with a smaller wavenumber is established (cf. Fig. 4c) at $t \approx 60 \tau_o$).

Thus, while roll-like features appear for this value of $\epsilon$, with periodic boundary conditions no transition to rolls occurs and the system relaxes to a new ordered hexagon pattern. Only for yet larger values of $\epsilon$ the roll-like structures that arise in the intermediate, disordered state take over and lead to a transition to rolls induced by the side-band instabilities.

ii) clearly, the simulations for periodic boundary conditions do not match the experimental results described above, where a transition from hexagons to rolls occurs.

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**Figure 3:** Stability regions for hexagons in CO\(_2\) with respect to side-band perturbations for layer thickness $d = 0.052\, \text{cm}$, pressure $P = 300\, \text{psi}$, mean temperature $T_0 = 27^\circ\text{C}$, and Prandtl $Pr = 0.86$. The NB coefficients are $\gamma_0^2 = 0.2053$, $\gamma_1^2 = -0.2877$, $\gamma_2^2 = 0.3267$, $\gamma_3^2 = 0.1152$, and $\gamma_4^2 = -0.086$. The hexagons are stable with respect to side-band perturbations in the region inside the solid lines and unstable outside. The dashed line corresponds to the neutral curve.
already for $\epsilon \gtrsim 0.11$. To address this disagreement we take into account the fact that in the experiments the side walls promote the nucleation of rolls [12].

Strictly speaking, the code we are using does not allow non-periodic boundary conditions. To mimic the experimentally used cylindrical cell we employ a step ramp in $\epsilon$ that reduces $\epsilon$ to values well below $\epsilon = 0$ outside a circle of radius $r = 0.45L$ with $L = 16 \cdot 2\pi/q_r = 32.22$ [22]. To induce concentric rolls near the sidewalls we introduce for $r > 0.45L$ an additional heating in the interior of the fluid in the form of concentric rings. Using hexagonal initial conditions in the bulk, this leads to an initial state as shown in Fig. 5a.

Fig. 5a,b shows two snapshots at $t = t_v$ and $t = 158 t_v$ demonstrating how the rolls induced by the side walls invade the carefully prepared hexagonal state in the bulk already for $\epsilon = 0.2$. This is well below the $\epsilon$-value for which with periodic boundary conditions the hexagons persisted even through the side-band instability. The final steady state consists of concentric rolls as observed in the experiments (cf. Fig. 5 in [12]). For lower $\epsilon$, however, the experimentally observed final state consists of hexagons in the bulk of the system surrounded by wall-induced concentric rolls (cf. Fig. 4 in [12]). We find this also in our numerical simulations, as shown in Fig. 5(c-d). There the forcing of rolls is identical to that in Fig. 5(a-b) but $\epsilon = 0.05$. Starting with random initial conditions, the forcing gives rise to a ring contained in the square integration domain. At the beginning of the simulation (Fig. 5c)) the rolls created by the forcing invade the interior of this small system. However, as time progresses the rolls pull out of the central region of the cell, and the final steady state (for $t \gtrsim 165 t_v$) consists of stable hexagons surrounded by a couple of concentric rolls in addition to those induced by the forcing.

Thus, our simulations suggest that the experimentally observed transition from hexagons to rolls is neither due to amplitude instabilities nor to side-band instabilities. Rather, there is a large range of parameters in which hexagons and rolls are both linearly stable and the final state is selected by one type of pattern invading the other. The transition to rolls at these low values of $\epsilon$ is made possible by the boundaries, which provide a seed for the nucleation of rolls. We expect that by applying a forcing that is confined to the region near the boundaries and that replaces the rolls by hexagons the transition to rolls could be shifted to substantially larger values of $\epsilon$. Such a forcing could be achieved by a patterned heating of the interior of the fluid [31] or possibly by a suitable geometric patterning of the bottom plate [32].

V. NB SPIRAL DEFECT CHAOS IN SF$_6$

A fascinating state observed in convection at low Prandtl numbers is spiral defect chaos. It is characterized by self-sustained chaotic dynamics of rotating spirals, as well as dislocations and disclinations, and arises in fluids with $Pr \lesssim 1$ [17, 19, 20, 21, 22] in a parameter range where straight rolls are linearly stable. Spiral defect chaos has so far predominantly been investigated under OB conditions, in which up-flows and down-flows are equivalent.

As mentioned before, NB effects break the up-down
symmetry and different flow structures may be predominantly associated with up-flow and down-flow, respectively. Moreover, in the absence of the OB symmetry a resonant triad interaction is allowed. If it is strong enough it leads to the formation of hexagons. For weaker interaction one may still expect an enhancement of cellular rather than roll-like structures.

To investigate the impact of NB effects on spiral defect chaos we consider convection in a layer of SF$_6$. This gas has been used previously in experimental convection studies under usual laboratory conditions and near the thermodynamical critical point. In Fig. 6(a) we present the stability diagram for hexagons and rolls with respect to amplitude perturbations in a layer of SF$_6$ of thickness $d = 0.0542$ cm, pressure $P = 140$ psi, and a range of temperatures that is experimentally accessible. Hexagons are amplitude-stable to the right of the solid line and rolls above the dashed line. As in the case of CO$_2$, the NB effects increase with increasing mean temperature $T_0$ above a certain value of $T_0$ hexagons are linearly amplitude-stable over the whole range of $\epsilon$ investigated. Here we focus on relatively strong NB effects. We therefore show in Fig. 6(b) the stability limits with respect to side-band perturbations for a relatively large mean temperature, $T_0 = 80^\circ$C. As in the case of CO$_2$, the wavenumber range over which the hexagons are stable is leaning towards smaller wavenumbers. Overall, amplitude and side-band stability limits are qualitatively similar to those of convection in CO$_2$ (cf. Fig. 1).

The snapshots depicted in Fig. 7 show, as expected, that due to the NB effects down-flow convection cells, which are white in Fig. 7, outnumber cells with up-flow (black). Moreover, in this regime the NB effects enhance the overall cellular rather than roll-like character of SDC. This manifests itself in the appearance of numerous small down-flow convection cells (white ‘bubbles’) and in the appearance of quite noticeable bulges on the NB convection rolls. To quantify these and other differences we analyse a long sequence of snapshots with a recently introduced geometric approach. It is based on the contour lines corresponding to the intensity half-way between the minimal and maximal intensity of all snapshots in a given run. The contour lines corresponding to the temperature field of snapshots Fig. 7(a,b) are shown in Fig. 7(c,d). In the following we present various statistics of these contour lines.

The most striking difference between the OB and the NB case is the asymmetry that is induced by the NB effects between ‘black’ and ‘white’ components, i.e. between closed contours that enclose up- and down-flow regions, respectively. To quantify this asymmetry we measure the number of white and black components. These two topological measures correspond, respectively, to the Betti numbers of order 1 and 2 of the pattern defined by the white components. Fig. 8 shows these two quantities as a function of time in the OB and the NB case. As expected, in the OB case the number of black and white components is essentially the same at all times,

![Stability regions of hexagons and rolls in a layer of SF$_6$ for $T_0 = 80^\circ$C and $\epsilon = 1.4$ in a convective cell of thickness $d = 0.0542$ cm and horizontal size $L = 16 \cdot 2 \pi / q_o = 32.22$. Periodic boundary conditions are used with 128 x 128 Fourier modes and 6 vertical modes. Both states are obtained after an integration time of 160 $C_c$, starting from random initial conditions. While in Fig. 6, all NB are retained, in Fig. 7 the same values are used for $Pr$ and $\epsilon$, but all NB parameters $\gamma_o^C$ are set to 0, i.e. the system is treated as if it was Boussinesq.

![Figure 6: Stability regions of hexagons and rolls in a layer of SF$_6$ of thickness $d = 0.0542$ cm, pressure $P = 140$ psi, and Prandtl number $Pr = 0.8$.](image)
Figure 7: Direct numerical simulation of equations (2-4) for SF$_6$ in a cell of thickness $d = 0.0542$ cm, pressure $P = 140$ psi, mean temperature $T_0 = 80^\circ C$, Prandtl number $Pr = 0.8$, and control parameter $\epsilon = 1.4$. The cell has size $L = 16 \cdot 2 \pi / q_c = 32.22$ with periodic boundary conditions. Starting from random initial conditions both snapshots are taken at $t = 160 t_v$. Left panels for OB conditions ($\gamma_i = 0, i = 0..4$, left), right panels for NB conditions appropriate for $T_0 = 80^\circ C$ (cf. Fig6b). Bottom panels give the corresponding contour lines used for the pattern diagnostics.

whereas in the NB case the white components significantly outnumber the black ones. The ratio of white to black components is therefore a sensitive indicator for the significance of NB effects. Fig8 also illustrates how much the number of components fluctuates during these runs. Recently, the two Betti numbers have also been measured based on patterns obtained in experiments on SDC in convection in CO$_2$. Scanning $\epsilon$ in very small steps the authors report steps in the Betti numbers indicative of transitions between different chaotic, disordered states [36].

Fig8 shows that the total number of components (closed contours) is considerably larger in the NB case than in the OB case. This is presented in more quantitative detail in Fig9 which gives the mean value of the total number of components, i.e. the sum of black and white components, as a function of $\epsilon$ for OB as well as NB conditions. In the NB case the total number of components is up to four times larger than in the OB case. We attribute this difference to the resonant triad interaction that is made possible by the breaking of the OB symmetry and which tends to enhance cellular rather than filamentary roll-like structures.

To characterize the components better and to distinguish cellular and roll-like structures we introduce the 'compactness' $C$ of components

$$C = 4\pi \frac{A}{P^2}$$

(11)

Here $A$ is the area inside a closed contour and $P$ its perimeter. With the normalization used in (11) compact, cellular structures are charactereized by $C \lesssim 1$, whereas filamentary, roll-like structures have $C \ll 1$.

Fig. 10 shows the mean number of closed contours per snapshot for a given compactness $C$ for the NB and the OB simulation at $\epsilon = 1.4$ over the duration $t = 360 t_v$. As expected, in the NB case the number of white components is much larger than that of black components,
The relative distribution among components with different compactness is more clearly visible in the relative white, filamentary contours with small compactness. NB case, which also shows an increase in the number of total number of components is noticeably larger in the whereas in the OB case both are about the same. The total number of components is noticeably larger in the NB case, which also shows an increase in the number of white, filamentary contours with small compactness.

The relative distribution among components with different compactness is more clearly visible in the relative frequency of contours as a function of the compactness, which is shown in Fig. 11. More precisely, its data result from running averages over adjacent 10 points of the corresponding data shown in Fig. 10, which are then normalized. The normalized data show that the increase in the relative frequency of white filamentary components ($C \ll 1$) is essentially the same in the NB case and in the OB case. However, only very few black filamentary components arise in the NB case. For $C \sim 1$, we observe a shoulder of the main peak which is extended significantly towards smaller contourlength. We associate this peak with the appearance of target-like structures, i.e. with a second contourline encircling a smaller compact, almost circular contourline. In the NB case this second peak is barely visible. Instead, the shoulder of the main peak is extended significantly towards smaller contourlength. It signifies the appearance of compact objects that are smaller than a typical wavelength, which we associate with the small ‘bubbles’ that are easily recognized in the snapshot of the NB case Fig. 7b. Thus, the comparable relative frequency of small components shown in Fig. 11 has a different origin in the OB and the NB case. Whereas in the NB case it is mostly due to small bubbles, it seems to originate from target-like structures in the OB case.

In the logarithmic scaling used in Fig. 12 a straight ridge arises in the distribution function for large contourlengths. It is characteristic for filamentary structures with a typical width, which corresponds here to half a wavelength $\lambda$ of the convection rolls,

$$C \sim 4\pi \lambda P \propto P^{-1}.$$  \hspace{1cm} (12)

In the OB case one can discern deviations from this scaling. They are confined to larger rather than smaller compactness values for a given contourlength, indicating that the long rolls can be wider but not narrower than a certain thickness. In the NB case these deviations are much smaller.

To identify spiral components in the pattern directly we also measure the winding number of the components. It is defined via the angle $\theta$ by which the (spiral) arm of a pattern component is rotated from its tip to its end at the vertex at which it merges with the rest of the component. In cases in which a component has no vertices we split it into two arms at the location of minimal curvature. The winding number is then defined as $|W| = \theta/2\pi$. To assess the impact of the NB effects on the spiral character of the pattern we measure the number ofspirals in each snapshot and show the resulting histogram over the whole run in Fig. 13. We use three different thresholds $W_{\text{min}}$ for the identification of spirals, $W_{\text{min}} = 1$, $W_{\text{min}} = 1/2$, and $W_{\text{min}} = 1/4$. As Fig. 13 shows, the number of small spirals with $|W| \geq 1/4$ is quite similar in the OB and the NB case. However, larger spirals with $|W| \geq 1/2$ or even $|W| \geq 1$ are much more rare in the NB case; in fact, for the system size $L = 16 \cdot 2\pi/q_c = 32.22$ that we have used in these simulations there was at most one spiral with $|W| \geq 1$ at any given time.

The reduced spiral character of NB spiral defect chaos is also quite apparent in Fig. 14 which shows the cor-
Figure 12: Distribution function for contour length and compactness for OB (a) and NB conditions (b) $Pr = 0.8$ and $\epsilon = 1.4$.

Figure 13: Distribution function for the number of spirals in spiral defect chaos under OB and NB conditions for three values of the threshold, $W_{\text{min}} = 1/4$ (circles), $W_{\text{min}} = 1/2$ (squares), and $W_{\text{min}} = 1$ (diamonds).

Figure 14: Correlation between the arclength and the winding number of spiral arms under OB (black squares) and NB conditions (red circles).

The relation between the winding number and the arclength of the spiral arm. More precisely, each dot marks the occurrence of one spiral arm in a snapshot. In the OB case one can see quite clearly a maximal winding number for any given arclength, which is consistent with an Archimedean shape of the spiral [26][47]. In the NB case, however, only components with very small contourlength reach the Archimedean limit and most components have winding numbers that are much smaller, i.e. the components are quite straight.

VI. HEXAGONS AND SPIRAL DEFECT CHAOS AT VERY LOW PRANDTL NUMBERS. H$_2$-XE MIXTURES

As mentioned above, the restabilization of NB hexagons at larger Rayleigh number is related to the existence of stable OB hexagons at large Rayleigh numbers. The wavenumber range over which the OB hexagons are stable shrinks with decreasing Prandtl number and for $Pr < 1.2$ the OB hexagons are side-band unstable at all wavenumbers [30]. However, as seen in the case of CO$_2$ and SF$_6$, NB hexagons can be side-band stable at large $\epsilon$ even below $Pr = 1.2$ due to the additional stabilizing effect of the resonant triad interaction. It is therefore of interest to investigate whether the NB effects can be sufficient to stabilize strongly nonlinear hexagons even for Prandtl numbers significantly below $Pr = 1$.

Prandtl numbers well below $Pr = 1$ can be reached by using a mixture of a heavy and a light gas. An experimentally investigated case is a mixture of H$_2$ and Xe [37]. With a mole fraction of $\chi = 0.4$, one can reach Prandtl numbers as small as $Pr = 0.17$. The Lewis number of such a mixture is close to one [19, 38]. Therefore such mixtures are expected to behave essentially like a pure fluid with the same Prandtl number [36].

We investigate the stability of hexagon and roll con-
vection in a H$_2$-Xe mixture with mole fraction $\chi = 0.4$ at a pressure of 300 psi and a layer thickness of $d = 0.1$ cm. With respect to amplitude instabilities the stability diagram is very similar to that of convection in CO$_2$ and SF$_6$ with hexagons becoming reentrant at $\epsilon$-values as low as $\epsilon = 0.14$ for $T_0 = 20^\circ C$.

Focusing on strong NB effects we perform a detailed stability analysis with respect to sideband perturbations at a mean temperature of $T_0 = 80^\circ C$ using the same layer thickness of $d = 0.1$ cm. For these low Prandtl numbers the numerical resolution has to be increased to obtain sufficiently well resolved flows. While for the stability analyses of hexagons in CO$_2$ and SF$_6$ it is sufficient to use $n_z = 6$ and $n_q = 3$ in the Galerkin expansion, for the H$_2$-Xe mixture with $P_r \simeq 0.17$ at least $n_q = 5$ and $n_z = 6$ are required. Fig. [13] depicts the resulting stability diagram. It shows that the region of sideband stable hexagons is not contiguous but consists of the usual region immediately above threshold and an additional, disconnected region at larger Rayleigh numbers. We could not follow the stability limits to smaller values of $q$ than shown in Fig [14] due to numerical convergence problems. Presumably, these arise due to bifurcations involving additional, resonant wavevectors somewhat similar to the resonances studied in Taylor vortex flow [10, 11]. The fact that the region of stability is disconnected is remarkable since the region of amplitude-stability (not shown) is actually contiguous. This is in contrast to the behavior found in CO$_2$ and SF$_6$ where the two side-band stable regions become connected when the bubble-like region of amplitude instability disappears (cf. Fig 13). The comparison of the stability limits with those of CO$_2$ and of SF$_6$ (Fig 13) shows further that the maximal wavenumber $q$ at which the hexagons are stable with respect to side-band perturbations decreases with decreasing Prandtl number and as a result the over-all stability region shrinks as well.

The stability analysis of the H$_2$-Xe mixture also reveals an oscillatory instability of the hexagon patterns at $\epsilon \sim 1$. Within the $\epsilon$-range investigated, no such oscillatory instability was found at the larger Prandtl numbers relevant for CO$_2$ and SF$_6$. Unfortunately, it turns out that before the onset of the oscillatory instability the hexagons already become unstable to a side-band instability at half the hexagon wavelength, which will always preempt the oscillatory instability. For the rolls we also find an oscillatory instability. It is presumably related to the well-known oscillatory instability of Boussinesq rolls [12].

For not too small $\epsilon$ generic initial conditions will not lead to hexagonal patterns but rather to spiral defect chaos. A typical snapshot of a pattern in this state is shown in Fig [14] for $\epsilon = 0.3$. Compared to the patterns obtained in NB convection in SF$_6$ (cf. Fig 1b) the patterns in H$_2$-Xe are less cellular and do not show a large number of small bubbles. To quantify these and other characteristics of the patterns we again apply the geometric diagnostics introduced earlier [20].

In Fig 17 we show the normalized distribution functions for the compactness of white and black components.

![Figure 15: Stability limits with respect to sideband perturbations in a mixture of H$_2$–Xe with molar fraction $\chi = 0.4$, thickness $d = 0.1$ cm, pressure $P = 300$ psi, mean temperature $T_0 = 80^\circ C$, and Prandtl number $P_r = 0.17$. The NB coefficients are $\gamma_0 = 0.5535$, $\gamma_1 = -0.6421$, $\gamma_2 = 0.9224$, $\gamma_3 = 0.3647$, $\gamma_4 = -0.0712$ resulting in $Q = 13.8$. Hexagons are stable in the regions enclosed by the solid lines and unstable outside. The dashed line represents the neutral curve. The results for H$_2$-Xe are obtained with $n_z = 6$ and $n_q = 5$.](image)

![Figure 16: Typical pattern for convection in H$_2$-Xe at $\epsilon = 0.3$.](image)
compactness $C$ is significantly reduced compared to the case of SF$_6$; in fact, while in SF$_6$ the maximum of the distribution function is close to $C = 1$, in H$_2$-Xe the absolute maximum is at $C \approx 0.1$, which corresponds to filamentary structures.

The lack of small bubbles is demonstrated in more detail in the joint distribution function for the contour length and the compactness, which is shown in Fig. 18. Note that the view is rotated compared to Fig. 12. The distribution function is lacking the broad shoulder seen in NB convection in SF$_6$ (see Fig. 12b). Instead, the decay of the distribution function towards long filamentary contours is quite slow.

To assess the spiral character of NB spiral defect chaos at these low Prandtl numbers we show in Fig. 19 the distribution function for the absolute value $|W|$ of the winding number for NB convection in H$_2$-Xe (at $\epsilon = 0.3$ and $Pr = 0.17$) as well as for Boussinesq and non-Boussinesq convection in SF$_6$ (at $\epsilon = 1.4$ and $Pr = 0.8$). As had been noted previously in the Boussinesq case [26, 43] the distribution function is roughly consistent with exponential behavior. In the NB case the exponential decays substantially faster than in the Boussinesq case and spirals with winding numbers above $|W| = 1$ are rare. In the Boussinesq case we had found that the decay rate depends mostly on the Prandtl number, but only very little on $\epsilon$ [26]. Unfortunately, we do not have enough non-Boussinesq data to investigate such trends in the $\epsilon$- and $Pr$-dependence. However, it is worth noting that in the two non-Boussinesq cases shown in Fig. 19 the decay rates are essentially the same despite their substantial difference in Prandtl number and both decays are much faster than that in the Boussinesq case. Thus, possibly the impact of NB effects dominates the dependence on the Prandtl number.

In NB convection in H$_2$-Xe, the distribution function for the number of spirals is qualitatively similar to that in SF$_6$, which is shown in Fig. 13 above. In particular, there are almost no spirals with a winding number $|W| > 1$. Similarly, the correlation between the arclength and the winding number of spirals reveals that in NB convection in H$_2$-Xe there is no significant trend towards Archimedean spirals.

VII. CONCLUSION

In this paper we have studied non-Boussinesq convection in gases (CO$_2$, SF$_6$, H$_2$-Xe) with Prandtl numbers ranging from $Pr \sim 1$ down to $Pr = 0.17$ in experimentally relevant parameter regimes. We have complemented a Galerkin stability analysis of hexagon patterns with direct numerical simulations of the fluid equations to study transitions between different hexagon patterns and to quantify the impact of non-Boussinesq effects on spiral defect chaos.

We find that the reentrance of hexagons that we have identified previously in non-Boussinesq convection in wa-
ner \cite{16} also occurs at low Prandtl numbers. As was the case at large Prandtl numbers, compressibility is not necessary for reentrance. Since, in addition, the range of wavenumbers for which the reentrant hexagons are stable differs significantly from that of the reentrant hexagons observed in experiments on convection in SF$_6$ near the thermodynamic critical point \cite{17}, the mechanisms underlying the two types of restabilization of the hexagons is most likely different. Reflecting the fact that in gases the non-Boussinesq effects increase with increasing temperature the reentrance is shifted to lower values of the Rayleigh number when the mean temperature is increased, opposite to the behavior in water \cite{16}. In convection in water the reentrant hexagons are stable only for wavenumbers below the critical wavenumber. This trend becomes more pronounced with decreasing Prandtl number. In fact, for the gas mixture with $Pr = 0.17$ the wavenumber at the stability limit of the hexagons decreases so rapidly with increasing Rayleigh number that the range in Rayleigh number over which the hexagons are stable becomes quite small.

The comparison of our stability results with experiments on the transition between hexagons and rolls in CO$_2$ \cite{12} shows that this transition is not due to an amplitude or a side-band instability. As a matter of fact, for the parameters of the experimental system the hexagons do not undergo any linear amplitude instability to rolls, contrary to the prediction of the weakly nonlinear theory \cite{12}. We have performed detailed numerical simulations with various lateral boundary conditions and confirm that the transition is the result of the heterogeneous nucleation of rolls at the side walls of the container, which then invade the whole system if the Rayleigh number is sufficiently high. Our simulations suggest that hexagons could be stabilized well beyond the experimentally observed transition point if the influence of the lateral walls can be reduced by applying a spatially patterned forcing that drives hexagons at the wall. Such a forcing can be achieved by localized heating \cite{31} or by geometric patterning of the bottom plate \cite{32}. Of course, the wavenumber of the forced hexagons would have to be adjusted to lie in the stable range.

We have also investigated the stability of hexagons in H$_2$-Xe mixtures with very small Prandtl number ($Pr = 0.17$). There also stable reentrant hexagons are possible, but they are restricted to a small range in wavenumber ($q < q_c$) and Rayleigh number. Since for such small Prandtl numbers Boussinesq hexagons are always side-band unstable \cite{30}, the stability of the hexagons is a result of the non-Boussinesq effects.

A fascinating state that is characteristic for convection at low Prandtl numbers is spiral defect chaos \cite{17}. We have studied the influence of NB effects on spiral defect chaos in a set-up corresponding to convection in SF$_6$ with a Prandtl number of $Pr = 0.8$ and in H$_2$-Xe with a Prandtl number of $Pr = 0.17$. To quantify the differences between Boussinesq and non-Boussinesq spiral defect chaos we have employed a recently suggested set of geometric diagnostics of the patterns \cite{29}.

As expected, in SF$_6$ and in H$_2$-Xe the non-Boussinesq effects break the equivalence of up- and down-flows and, consequently, the mean number of pattern components (closed contours) corresponding to down-flow differs significantly from that of the up-flow components. More interesting is our finding that in SF$_6$ the total number of components (Betti number \cite{35}) is more than twice as large in the non-Boussinesq case than in the Boussinesq case for otherwise equal parameters. We attribute this enhancement of cellular rather than roll-like structures to the resonant triad interaction that is introduced by the non-Boussinesq effects.

Another striking difference between Boussinesq and non-Boussinesq spiral defect chaos in SF$_6$ becomes apparent in the joint distribution function for the contourlength of the components and their compactness. While the Boussinesq case exhibits a strong signature of target-like structures, the non-Boussinesq case displays instead a marked increase in the number of small compact components (‘bubbles’). This trend towards a more cellular structure is also apparent in the analysis of spirals. The number of spirals with a winding number above $1/2$ is much smaller in the non-Boussinesq than in the Boussinesq case. In the correlations between the winding number and the arclength of the components this is reflected by the missing of large Archimedean spirals in the non-Boussinesq case.

Interestingly, we do not find many small bubbles in our simulations of H$_2$-Xe at $Pr = 0.17$. In fact, the maximum of the distribution function for the compactness of pattern components is shifted away from $C \lesssim 1$ to $C \sim 0.1$, i.e. to filamentary structures. We do not know whether this implies that the tendency towards small bubbles is maximal at moderately small Prandtl numbers, i.e. for $Pr \sim 1$, or whether it is due to the fact that for computational reasons the simulations in H$_2$-Xe were performed at a lower Rayleigh number ($\epsilon = 0.3$) than in SF$_6$ ($\epsilon = 1.4$).

Due to computational limitations we have not been able to investigate the transition from hexagons to spiral defect chaos at very small Prandtl numbers. Since for very small Prandtl numbers there are essentially no large spirals, the transition from hexagons to spiral defect chaos may follow a different path than that observed experimentally in CO$_2$, where large spirals were reported to arise in the transition from hexagons to rolls \cite{12}. We surmise that the geometric diagnostics that we have employed to characterize the well developed spiral defect chaos would provide also valuable insight into this transition. Since the diagnostics require a substantial amount of data the transition would be best investigated experimentally.

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[37] These values were obtained with a code kindly provided by G. Ahlers.
[38] In the experimental setup the top temperature is held constant at $T = 12.84^\circ C$, and therefore the mean temperature changes as $\epsilon$ is increased. In our computations, however, we keep $T_0$ fixed. Since the transition occurs quite close to threshold this is a good approximation to the experimental procedure.
[39] Note that detailed analyses of large spirals show deviations from the Archimedean shape due to the dislocations that accompany finite spirals [40].