## Chaotic Behavior of Instability Due to Unipolar Ion Injection in a Dielectric Liquid

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The chaotic regime encountered in electrohydrodynamic instability of an insulating liquid subjected to space-charge-limited unipolar injection has been investigated experimentally. Two types of behavior of the power spectra of the intensity fluctuations have been characterized: an exponential decay obtained when viscous terms are dominant and a powerlaw decay obtained when inertial terms are dominant. Emphasis is put on the quasiuniversality of these behaviors and this is discussed in relation to the phenomenon of smallscale intermittency.

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In the past few years a remarkable amount of progress has been made both experimentally<sup>1</sup> and theoretically<sup>2</sup> in understanding the different sequences of hydrodynamic instabilities leading to chaotic motions and in relating them to the properties of dynamical systems with a few degrees of freedom. But so far rather little attention has been paid to the frequency dependence of the broadband noise power spectra in the chaotic regime with the exception of an  $f^{-4}$  law<sup>3</sup> mentioned for the Rayleigh-Bénard problem. One may ask if there are common trends or some universal behavior in the chaotic regime.

This question is examined here in experiments on electrohydrodynamic (EHD) instability due to space-charge-limited unipolar injection which bears some analogy with the Rayleigh-Bénard (RB) problem. Let us recall briefly the main features (see Atten and Lacroix<sup>4</sup> and Lacroix, Atten, and Hopfinger<sup>5</sup>). Ions are injected by one electrode in a plane layer of an insulating liquid subjected to a dc voltage U; the bulk Coulomb force  $q\vec{E}$  exerted by the electric field  $\vec{E}$  on the space charge of density q plays a destabilizing role. The transport mechanism without liquid motion (ions drift with velocity  $K\vec{E}$ ; K is the mobility) is different from that in the RB case (heat diffusion) and this explains the occurrence of two instability criteria, one linear and the other nonlinear, associated with an hysteresis loop. As U is raised beyond the critical voltage  $U_c$ , one observes first a viscous-dominated convection regime (z component of the rms velocity  $w' \propto U^2/d$  -balance of viscous term and Coulomb force). At high enough voltages, we have an inertially dominated regime  $(w' \propto U/d)$ . The transition between them is progressive and the voltage  $U_T$  for which inertial and viscous effects are of the same order is determined on I-V plots

(see Fig. 5 of Ref. 5). In our experimental conditions,  $U_c \simeq 50$  V and  $U_T \sim 1000$  V.

We have previously studied the transition to chaotic motion in this system by measuring the fluctuations of the very weak intensity of the total electric current. This has been done for large<sup>6</sup> aspect ratios  $\Gamma = D/2d$  (D is the diameter of the cylindrical cavity and d the distance between electrodes), as well as in the case  $\Gamma \simeq 1$ , where only one convective cell<sup>7</sup> would be expected. Independently of  $\Gamma$ , as U passes the critical voltage  $U_c$ , the liquid passes directly from rest to time-dependent motion. For small  $\Gamma$  the frequency power spectra of current fluctuations are discrete; they consist of one fundamental peak  $f_1$ , its harmonics, and its subharmonics. By slightly increasing U one obtains biperiodic motion and then continuous spectra. For large  $\Gamma$  the spectra are always continuous but exhibit an enlarged peak corresponding to the same fundamental oscillation as for small  $\Gamma$ . The  $f_1$  variation with U has been studied and it appears to vary in proportion to w'.<sup>6,7</sup>

Let us consider the chaotic regime. In conditions where viscous effects are dominant  $(U < U_r)$ the power spectra exhibit a well-defined exponential decay  $\exp(-f/f_c)$  in the frequency range between  $f_1$  and the further limit where the power P reaches the instrumental noise level. This is for large<sup>6</sup> as well as for small  $\Gamma$  values.<sup>8</sup> By varying both the depth d of the cell (0.25 to 2 mm) and the aspect ratio (from 0.83 to 13) we found practically no difference in the power spectra between small and large  $\Gamma$  values (Fig. 1). Furthermore, the characteristic frequency  $f_c$  of the exponential part of the spectra varies with U as the oscillatory frequency  $f_1$  (Fig. 2). Their ratio is close to 1:  $f_c/f_1 \simeq 0.7$  (see inset). Hence we can speak of a quasiuniversal shape of the spec-



FIG. 1. Power spectra of intensity fluctuations in the viscous-dominated chaotic regime. (In all figures P is in units  $10^{-24} \text{ A}^2/\text{Hz}$ .)

tra as revealed by the curves of Fig. 1 (the frequency has been normalized with  $f_c$ ).

As the voltage is increased beyond  $U_T$  the spectral shape loses this quasiuniversal characteristic. Clearly the variation of P vs f departs from the exponential law above some frequency [Figs. 3(a) and 3(b)]. A log-log plot [Figs. 3(c) and 3(d)] indicates a tendency toward a power-law decrease in the high-frequency part. A systematic study showed the following features: (a) The fundamental oscillation is always present and the variation of the peak frequency  $f_1$  has the asymptotic law  $f_1 \propto U/d^2$ . (b) The frequency interval where the decay is exponential has a width which



FIG. 2. Variation with applied voltage  $U \text{ of } f_c d^2$  for various aspect ratios. Inset: ratio  $f_c / f_1$  vs U.

decreases as the voltage is increased, but as far as the characteristic frequency  $f_c$  can be evaluated the relation  $f_c/f_1 \simeq 0.7$  always holds (Fig. 2). (c) For high enough frequency, P tends to the asymptotic negative power law:  $P \propto f^{-\alpha}$ . It is difficult to determine accurately the exponent  $\alpha$  because of its high value which limits the frequency interval where the power law can be observed. We obtained  $\alpha = 7 \pm 1$ . Such a power law characterizing the inertially dominated regime could be related to the Kolomogorov law  $(E \propto k^{-5/3})$  by modeling the influence of the small eddies of wave number k and characteristic frequency  $f \sim k^{2/3}$  on the total current.

We have characterized experimentally two different behaviors (the exponential and power laws) of chaotic-regime power spectra. This distinction is not so clear in other physical systems. In Taylor-Couette instability, spectra with exponential decay can be found,<sup>9</sup> but it seems that this is for conditions where inertial effects cannot be neglected. In the Rayleigh-Bénard problem one has to distinguish between small and large  $\Gamma$  values. For small  $\Gamma$  and high Prandtl number exponential decay has been found<sup>10</sup>; for small  $\Gamma$  in experiments with water where inertial effects could be dominant Gollub<sup>11</sup> has found in the highfrequency part of some chaotic spectra a negative power law with an exponent consistent with 4. For large  $\Gamma$  the interaction between different convective rolls described by the phenomenon of phase turbulence<sup>12</sup> has to be taken into account. An  $f^{-4}$  law has been obtained in liquids of both low<sup>13</sup> and high<sup>14</sup> Prandtl number (i.e., for both inertially dominated and viscous-dominated convection) and this for  $R_a$  just above  $(R_a)_c$  (before any oscillatory instability had begun). Let us



FIG. 3. Power spectra of intensity fluctuations in the inertially dominated regime for two different applied voltages (semilog and double-log plots of same spectra).



FIG. 4. High-pass filtering of intensity fluctuations for  $\Gamma = 1$ . (a) Viscous-dominated regime: U = 200 V;  $f_1 \approx 8$  Hz; bandpass filtering between 30 and 100 Hz. (b) Inertially dominated regime:  $U \approx 5000$  V;  $f_1 \approx 100$  Hz; bandpass filtering between 300 and 500 Hz.

note that in our EHD experiments, where no difference with variable  $\Gamma$  has been found, the strong basic oscillatory mechanism in each convective cell presumably prevents any phase turbulence from becoming established. Thus, apart from the contrast between the viscous-dominated regime and the inertially dominated regime, the basic oscillatory mechanism plays an important role in the form that the chaotic motion takes.

The contrast between exponential and power-law spectra is discussed by Greenside *et al.*<sup>15</sup> in their attempt to model Ahlers's experiments: The authors relate the  $f^{-4}$  law to stochastic properties of the system and the exponential law to deterministic models with a small number of degrees of freedom. This argument could be compatible with our results if we admit that the stochasticity arises from the "intrinsic noise" induced by the nonlinear term  $(V \cdot \text{grad})V$ . Our experimental result of exponential decay in the viscous regime can be discussed in the light of theoretical work by Frisch and Morf.<sup>16</sup> In order to model small-scale intermittency in fully developed turbulence, they have analyzed the highfrequency behavior of a nonlinear Langevin equation. They found that bursts of intermittency are related to an exponentially decaying power spectrum. To check this relation in our physical situation we have high-pass filtered our signal of global intensity fluctuation in the chaotic regimes. In the viscous regime, i.e., at relatively small applied voltage, we notice well-separated intermittent bursts in the filtered signal [Fig. 4(a)]. Conversely in the inertial regime we cannot distinguish any bursts [Fig. 4(b)]. This relation between bursts of intermittency and exponential decay has also been obtained numerically by Manneville.<sup>17</sup>

In addition to our work on chaotic behavior we also noticed that for small aspect ratio, when discrete sharp peaks are present in the spectra the ratio of the amplitude of the nth harmonic to



FIG. 5. Power spectrum obtained in the cell d = 0.3 mm,  $\Gamma = 0.83$ , for U = 130 V. Full lines characterize the exponential decay for (1) the sharp peaks and (2) the broadband noise.

that of the (n-1)th is often roughly constant, thus revealing an exponential decay to which we can associate another characteristic frequency  $f_c'$ (Fig. 5). The ratio  $f_c'/f_1$  seems to be half the ratio  $f_c/f_1$  for continuous chaotic spectra, clearly seen in Fig. 5 just above the onset of the chaotic regime when a series of peaks are present with broadband noise. A similar observation about  $f_c$ ' and  $f_c$  can be derived from Fig. 1 of Gollub, Benson, and Steinmann.<sup>18</sup> This exponential decay of both continuous and discrete parts of the spectra could be a rather general phenomenon and, as suggested by Pomeau,<sup>19</sup> related to the stability properties of the phasespace trajectories as reflected by the Floquet or Lyapunov exponents.

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<sup>&</sup>lt;sup>1</sup>For the Rayleigh-Bénard case, see J. P. Gollub and S. V. Benson, J. Fluid Mech. <u>100</u>, 449 (1980); G. Ahlers, in *Systems Far from Equilibrium*, edited by L. Garrido, Lecture Notes in Physics Vol. 132 (Springer-Verlag, New York, 1980), p. 143; P. Berge and M. Dubois, *ibid.*, p. 381; A. Libchaber and J. Maurer, in *Nonlinear Phenomena at Phase Transition and Instabilities*, edited by T. Riste (Plenum, New York, 1981), Vol. B77, pp. 259-286.

<sup>&</sup>lt;sup>2</sup>For an extensive panorama of present ideas on roads to turbulence, see J. P. Eckmann, Rev. Mod. Phys. <u>53</u>, 643 (1981), and references therein.

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<sup>13</sup>G. Ahlers and R. P. Behringer, Phys. Rev. Lett. <u>40</u>, 712 (1978), and Ref. 3. See also Fig. 4 of A. Libchaber and J. Maurer, J. Phys. (Paris), Lett. <u>39</u>, L369 (1978).

<sup>14</sup>P. Berge, in *Dynamical Critical Phenomena and Related Topics*, edited by C. P. Enz (Springer-Verlag, New York, 1979), p. 288. Figure 11 is compatible with

an  $f^{-4}$  power-law spectrum.

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## Nonlinear Pattern Formation near the Onset of Rayleigh-Bénard Convection

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It is shown that many of the experimentally observed features of pattern formation in Rayleigh-Bénard convection near onset can be understood in terms of a two-dimensional relaxational equation. In particular, it is shown that disordered roll patterns follow a complicated dynamics that can require up to a hundred horizontal diffusion times to reach equilibrium.

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In attempting to understand the essential ways in which nonlinear nonequilibrium systems become turbulent, considerable experimental and theoretical effort has been devoted to one of the simplest systems, Rayleigh-Bénard convection.<sup>1</sup> While a fairly satisfactory understanding has emerged for small-aspect-ratio cells,<sup>2</sup> no such understanding currently exists for the onset of turbulence in large-aspect-ratio cells, whose large lateral dimensions (compared with the depth of the fluid) allow the excitation of longwavelength modes and the presence of defects. An experiment in a large cylindrical cell shows chaotic behavior at the onset of convection<sup>3</sup> while a recent experiment in a large rectangular cell<sup>4</sup> suggests that the fluid becomes clearly time dependent only a finite distance above onset. Theory predicts that parallel rolls within a certain band of wave numbers are stable but makes no prediction of the possible time dependence of curved or disordered rolls.

In this Letter, we use a two-dimensional equation<sup>5</sup> to study numerically the evolution and formation of curved roll patterns (corresponding to three-dimensional flow in the fluid) in large rectangular cells just above the onset of convection. Our model accurately reproduces the physics of the Boussinesq equations<sup>1</sup> sufficiently close to onset and for sufficiently large-aspect-ratio cells. Unlike most experiments to date, we determine *both* the Nusselt number and the velocity field as functions of time, permitting a more quantitative



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