An Analytical Study of Hamiltonian Chaos in Nonsteady Finite-Amplitude Electroconvection

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The extent of chaos and the suppressory role of Coulomb repulsion in the chaotic dynamics associated with nonsteady finite-amplitude electroconvection is revisited in the context of dynamical systems. Specifically, it is theoretically demonstrated that the threshold value of the charge density taming the heteroclinic chaos associated with laminar chaotic mixing depends on the sinusoidal perturbation’s frequency of the fluid velocity field, which is at variance with the fixed threshold value previously reported [Chacón et al., 1994]. Additionally, the consideration of time-periodic multiharmonic perturbations reveals the great complexity of the chaotic mixing scenario.

Keywords: Electroconvection; Hamiltonian chaos; Melnikov’s method.

Among the most fascinating long-lasting problems in electrohydrodynamics, that of electroconvection in a liquid layer subjected to unipolar injection has attracted great interest due to its theoretical and practical relevance [Atten & Moreau, 1972; Castellanos & Atten, 1987]. The fluid’s response to the injection is highly complex because it depends strongly on the trapping and detrapping of ions in a region bounded by a separatrix curve that is associated with a steady but unstable state. Indeed, some experimental results pointed to the existence of temporal chaos in the problem [Malraison & Atten, 1982; Atten et al., 1980]. This was followed by the theoretical prediction and numerical confirmation of laminar chaotic mixing of charge in the case of finite-amplitude electroconvection with a sinusoidal time-periodic dependence in the velocity field when the injection of charge is sufficiently weak for Coulomb repulsion to be neglected [Pérez & Castellanos, 1991]. The effect of weak but non-negligible injection of charge on the Hamiltonian chaos induced by the time-periodic velocity field was considered in [Chacón et al., 1994], in which a fixed, frequency-independent, threshold value of the charge density needed to suppress chaos was reported.

In this present work, it is theoretically shown that such a charge-induced suppression scenario is highly sensitive to the strength of the heteroclinic chaos associated with the perturbed separatrix, which in its turn depends on the spectral properties of the temporal fluctuations of the velocity field. Specifically, the Hamiltonian arising from the electroconvection problem is used as a model to study the relevance of nonharmonic periodic fluctuations on the extent of the chaotic mixing and its charge-induced suppression.

The present electroconvection problem considers a dielectric liquid of permittivity $\epsilon$, confined between two parallel metallic plates a distance $d$ apart with an applied voltage difference $V$ between them. Injection takes place from one of the plane...
electrodes, where the charge density is assumed to be constant and of value \( q_0 \). Ions are removed from the system once they reach the opposite electrode, the collector. When equations are put in dimensionless form taking as units \( d \) for distances, \( V/d \) for the electric field, \( dV/d^2 \) for the charge density, and \( K_\infty V/d \) for the velocity \( (K_\infty \) is the ion mobility), the charge conservation equation is written

\[
\frac{\partial q}{\partial t} + \mathbf{u} \cdot \nabla q + q^2 = 0,
\]  

(1)

where \( \mathbf{u} \) is the liquid velocity, \( \mathbf{E} \) the electric field, and \( q \) the charge density. The charge density is related to the electric field through Poisson’s equation

\[
\nabla \cdot \mathbf{E} = q.
\]

(2)

The boundary conditions for Eqs. (1) and (2) are \( \int_0^d \mathbf{E} \cdot d\mathbf{r} = 1 \) and \( q = C \) at \( z = 0 \), where the parameter \( C = \frac{q_0 d^2}{\epsilon V} \) is a measure of the injected charge. The coordinate system is chosen such that the injecting electrode is at \( z = 0 \) while the collector is at \( z = 1 \). Note that the term \( q^2 \) in Eq. (1) represents the decrease of charge in a parcel of fluid due to the Coulomb repulsion between ions. Also, it is assumed that the fluid motion is two-dimensional and in the form of a self-similar roll: \( \mathbf{u} = A \mathbf{u}_0 \), with \( A \) being the velocity amplitude and \( \mathbf{u}_0 = (A, 0, 0) \). In particular, the stream function \( \Psi(x, z) \equiv \frac{1}{\sqrt{2\pi}}[1 - \cos(2\pi z)]\sin(\frac{\pi x}{L}) \) is considered in the following [Castellanos & Atten, 1987; Pérez & Castellanos, 1991; Chacón et al., 1994].

Finally, Eq. (1) is equivalent to

\[
\begin{align*}
\frac{dx}{dt} &= \partial \Psi \overline{\partial z} + E_x, \\
\frac{dz}{dt} &= -A \partial \Psi \overline{\partial z} + E_z, \\
\frac{dq}{dt} &= -q^2.
\end{align*}
\]

(3)

Remarkably, the problem allows a Hamiltonian description when Coulomb repulsion is neglected in Eq. (3):

\[
\begin{align*}
\frac{dx}{dt} &= \partial H \overline{\partial z}, \\
\frac{dz}{dt} &= -\partial H \overline{\partial x},
\end{align*}
\]

(4)

where \( H(x, z) \equiv -x + A \Psi(x, z) \) is the Hamiltonian. Figure 1 shows some typical trajectories, including

![Figure 1. Phase portrait of the unperturbed Hamiltonian](image)

the separatrix, for a constant amplitude \( A > 1 \). Next, one considers perturbations of the Hamiltonian \( H(x, z) \) to take into account temporal fluctuations of the velocity amplitude in the form of a sinusoidal perturbation [Pérez & Castellanos, 1991], as well as a weak but non-negligible injection of charge in the inner region of the unperturbed separatix [Chacón et al., 1994] (see Fig. 1). For \( A \sim 1 \), and after rescaling variables, \( x \rightarrow x/L, z \rightarrow z - 1/2, t \rightarrow \pi^2 t, A \rightarrow (A - 1)/\pi^2 \), one obtains the dynamics equations

\[
\begin{align*}
\frac{dx}{dt} &= -2xz + Ca_x, \\
\frac{dz}{dt} &= -A + \frac{x^2}{2} + z^2 - \epsilon \sin(\Omega t) + C(b + \epsilon z),
\end{align*}
\]

(5)

where \( \epsilon \ll 1 \), and the parameter \( C \) \((C \ll 1)\) is a measure of the injected charge in the inner region: \( q_{\text{inj}} = \nabla \cdot \delta \mathbf{E} = C(1 + \epsilon) \), with \( \delta \mathbf{E}_x = Ca_x, \delta \mathbf{E}_z = C(b + \epsilon z) \), and where the coefficients \( a, b, \epsilon \) are assumed constant in a first approximation [see Pérez & Castellanos, 1991; Chacón et al., 1994 for additional details]. The application of Melnikov’s method [Melnikov, 1963; Guckenheimer & Holmes, 1983] to Eq. (5) yields the Melnikov function [Chacón et al., 1994].
Hamiltonian Chaos in Nonsteady Finite-Amplitude Electroconvection

\[ M(t_0) = \frac{\sqrt{6\pi\Omega}}{2} \text{sech} \left( \frac{\pi\Omega}{4\sqrt{A}} \right) \cos(\Omega t_0) \]

\[ -\frac{\sqrt{2\pi}}{2} q_{\text{inner}}. \]  

(6)

It is clear from Eq. (6) that heteroclinic chaos can be suppressed when \( q_{\text{inner}} \) is greater than a certain value. One can expect that this suppression threshold value should depend on the extent of the heteroclinic tangle associated with the perturbed separatrix when \( C = 0 \), i.e. on the width of the subsequent chaotic separatrix layer. Since the appearance of the heteroclinic tangle is the basic mechanism allowing the mixing of charge between the inner and outer regions of the unperturbed separatrix, one can obtain an estimate of the charge in the inner region by using lobe (turnstile) dynamics [Rom-Kedar et al., 1990; Wiggins, 1992]. A rough estimate can be obtained by limiting the analysis to primary lobes, i.e. capturing behavior over only the timescale of one period \( T \equiv 2\pi/\Omega \). Note that this may well be enough to obtain an upper estimate since the charge will decrease with time due to Coulomb repulsion [Eq. (3)]. Thus, one assumes

\[ q_{\text{inner}} \simeq C \frac{S_4}{S_3}, \]  

(7)

where \( S_4 \) is the area of any primary lobe while \( S_3 = \sqrt{\pi \varepsilon A} / 2 \) is the area of the inner region of the unperturbed separatrix. (Note that there is a typo in the expression for \( S_4 \) in [Chacón et al., 1994].) Also, the area of all the primary lobes is the same because the associated mapping is area-preserving when \( C = 0 \). The area of the primary lobe when \( C = 0 \) is given by

\[ S_i = \frac{t_{n+1} - t_n}{t_{n+1}} |M(t)| dt = \frac{\sqrt{6\pi\Omega}}{2} \text{sech} \left( \frac{\pi\Omega}{4\sqrt{A}} \right), \]  

(8)

where \( t_n \) and \( t_{n+1} \) are two consecutive zeros of the Melnikov function. After substituting Eqs. (7) and (8) into Eq. (6), one finally obtains

\[ M(t_0) = \frac{\sqrt{6\pi\Omega\varepsilon}}{2} \text{sech} \left( \frac{\pi\Omega}{4\sqrt{A}} \right) \left[ \cos(\Omega t_0) - 2C \frac{\Omega}{\Omega} \right], \]  

(9)

which has no zeros if

\[ C > C_{th}(\Omega) = \frac{\Omega}{2}. \]  

(10)

The condition \( C_{th} = 1/2 \) was deduced instead in [Chacón et al., 1994], because of an erroneous calculation of \( S_l \) (cf. Eq. (12) in [Chacón et al., 1994]), providing thus a fixed threshold value of the injected charge which is independent of the driving frequency. On the contrary, Eq. (10) provides a sufficient condition for the disappearance of the heteroclinic tangle that takes into account the strength of the Hamiltonian chaos existing when \( C = 0 \), and hence the value of the driving frequency, as physically expected. Indeed, for negligible injection \( (C \approx 0) \), the width \( \Delta \), of the chaotic separatrix layer is found to be [Pérez & Castellanos, 1991]

\[ \Delta = \Delta(\Omega, A) \equiv \frac{\sqrt{6\pi\Omega}}{4\sqrt{A}} \text{sech} \left( \frac{\pi\Omega}{4\sqrt{A}} \right), \]  

(11)

which presents a single maximum at \( \Omega = \Omega_{\text{max}} \approx 1.52 \sqrt{A} \) and the limiting behaviors \( \lim_{\Omega \to 0} \Delta = 0 \). Since the effective expressions of Eq. (5) are only valid for small values of \( A \) (recall the aforementioned rescaling), this constraint limits the range of validity of the condition represented by Eq. (10) to values of \( C \) up to \( \sim 0.76 \sqrt{A} = \Omega_{\text{max}}/2 \). For frequencies higher than \( \Omega_{\text{max}} \), the driving period may become so small that the analysis of primary lobes no longer captures the essential physics.

It is worth mentioning that broadband spectra of the velocity amplitude have been experimentally observed for strong injection \( (C \gtrsim 1) \) injections [Vázquez et al., 2006]. It is thus expected that, in a general situation, the temporal fluctuations of the velocity amplitude may present non-negligible additional harmonics beyond the main one, even in the simplest case of a periodic excitation. In the following, the role of multiharmonic excitations on the chaotic mixing scenario is discussed by considering the \( T \)-periodic excitation

\[ f(t; T, m) \equiv \sin \left( \frac{4Kt}{T}; m \right) \]  

(12)

instead of \( \sin(\Omega t) \) in Eq. (5), where \( \sin(\ldots; m) \) is the Jacobian elliptic function of parameter \( m \in [0, 1] \), and \( K = K(m) \) is the complete integral of the first kind [Armitage & Eberlin, 2006]. When \( m = 0 \), one has \( f(t; T, m = 0) = \sin(\Omega t) \) (i.e. one recovers the
R. Chacón

case of a harmonic excitation). In the other limit,

\[ f(t; T, m = 1) = \frac{1}{\pi} \sum_{n=0}^{\infty} \sin \left( \frac{(2n+1)2\pi t}{T} \right) \quad (13) \]

(i.e. one recovers the square-wave function of period \( T \)). The effect of renormalization of the elliptic sine argument is clear: with \( T \) constant, solely the excitation shape is varied by increasing the elliptic parameter \( m \) from 0 to 1, and there is a smooth transition from a sine function to a square-wave [see Fig. 2(a)]. For \( C = 0 \), it was found numerically that the response of the system of Eq. (5) to a sinusoidal excitation differs qualitatively and quantitatively from the response to a square-wave of the same period and amplitude [Pérez et al., 1996]. Here, it will be shown theoretically that such differences also appear in the charge-induced suppression scenario \((C > 0)\). Using the Fourier expansion of \( \sin(\ldots; m) \) [Armitage & Eberlin, 2006], and after some simple algebraic manipulations, the Melnikov function may be written as

\[ M(b_0) = \frac{\sqrt{6\pi t}}{2} \sum_{n=0}^{\infty} c_n(m) b_n(\Omega, A) \times \left\{ \cos[(2n+1)\Omega t] - \frac{2C}{(2n+1)^2} \right\} \quad (14) \]

where the coefficients are given by

\[ c_n(m) \equiv \frac{\pi}{K(m)\sqrt{m}} \left[ \pi(2n+1)K(1-m) \right] \frac{\text{csch} \left( (2n+1)\pi \right)}{4\sqrt{A}}. \quad (15) \]

For negligible injection \((C = 0)\), the width of the chaotic separatrix layer is now

\[ \Delta = \Delta(\Omega, A, m) \equiv \sqrt{\frac{9\pi t}{4}} \sum_{n=0}^{\infty} c_n(m) b_n(\Omega, A), \quad (16) \]

which, for \( A \) and \( m \) constant, exhibits a single maximum as a function of \( \Omega \) at \( \Omega_{\text{max}} = \Omega_{\text{max}}(A, m) \) such that \( \Omega_{\text{max}} \) decreases as \( m \) is increased from 0 [see Fig. 2(b)]. For a fixed period \( T \), one sees that the width \( \Delta \) increases as \( m \) is increased from 0, i.e. as the perturbation changes from a sine function to a square-wave. Numerical simulations confirm that the chaotic layer exhibits an increasing width as a function of the elliptic parameter \( m \). Figure 3 provides an illustrative sequence for four increasing values of \( m \) from 0 (sinusoidal perturbation) to \( 1 - 10^{-6} \) (nearly square-wave). One sees that this effect is greater the closer the perturbation is to a square-wave, i.e. when the presence of successive harmonics is significantly increased, which is a direct consequence of the nonlinear dependence of the function \( K(m) \) on the elliptic parameter \( m \) [Abramowitz & Stegun, 1972].

In the presence of injection \((C > 0)\), it is straightforward to demonstrate from Eqs. (14) and (15) that a heteroclinic bifurcation is frustrated (and hence the heteroclinic tangle no longer exists).
Fig. 3. Trajectories in the stroboscopic (for \( t = nT \) with \( n = 0, \ldots, 300 \)) Poincaré section for \( \varepsilon = 0.05, A = 0.01, C = 0, \Omega = 0.4 \), and four values of the elliptic parameter: \( m = 0, 0.95, 0.99, \) and \( 1 - 10^{-6} \). The same initial conditions were used in all the versions.

If

\[
C > C_{th}(\Omega, A, m)
= \frac{\Omega}{2} \sum_{n=0}^{\infty} c_n(m) b_n(\Omega, A)
\]

(17)

A plot of the difference \( \Delta C_{th} \equiv C_{th}(\Omega, A, m) - C_{th}(\Omega, A, m = 0) \) for \( A \) constant is shown in Fig. 4(a). For a fixed period \( T \), one sees that the suppressory threshold value \( C_{th} \) increases as \( m \) is increased from 0, i.e. an ever greater charge density is needed to suppress heteroclinic chaos as the perturbation changes from a sine function to a square-wave. Again, this effect is greater the closer the perturbation is to a square-wave, i.e. when the presence of successive harmonics is significantly increased. Also, for a fixed elliptic parameter \( m \), the difference \( \Delta C_{th} \) exhibits a single maximum as a function of \( \Omega \) at \( \Omega'_{\text{max}} = \Omega_{\text{max}}(A, m) \) such that \( \Omega'_{\text{max}} \) decreases as \( m \) is increased from 0 [see Fig. 4(b)].

Fig. 4. (a) Difference \( \Delta C_{th} \equiv C_{th}(\Omega, A = 0.01, m) - C_{th}(\Omega, A = 0.01, m = 0) \) [Eq. (17)] versus \( m \) and \( \Omega \). (b) Difference \( \Delta C_{th} \equiv C_{th}(\Omega, A = 0.01, m) - C_{th}(\Omega, A = 0.01, m = 0) \) versus \( \Omega \) for three values of the elliptic parameter: \( m = 0.9 \) (thin line), \( m = 0.9999 \) (medium line), and \( m = 1 - 10^{-4} \) (thick line).
Fig. 5. Trajectories in the stroboscopic (for \( t = nT \) with \( n = 0, \ldots, 100 \)) Poincaré section for \( \varepsilon = 0.05, A = 0.01, C = 0.02, \delta = 0, a = c = 0.01, \Omega = 0.4 \), and four values of the elliptic parameter: \( m = 0, 0.95, 0.99, 1 - 10^{-6} \). The same initial conditions were used in all the versions.

which is coherent with the aforementioned behavior of the width \( \Delta \). From the comparison of Eqs. (16) and (17), one may expect \( \Omega_{\text{max}}(A, m) \) and \( \Omega'_{\text{max}}(A, m) \) to be correlated. Similarly to the limiting case of a sinusoidal perturbation, one must expect the range of validity of condition (17) to be limited to values of \( C \) up to \( \sim C_{\text{th}}(\Omega'_{\text{max}}, A, m) \). It has been shown that, even in the absence of the heteroclinic tangle, the fluid velocity field is unsteady, and exhibits rather complex time-dependence in the presence of injection [Chicón et al., 1997]. Numerical simulations confirm indeed this observation, so that one will find it difficult to distinguish the suppressory effect of the injected charge on the chaotic layer. Figure 5 provides an illustrative sequence for \( C > 0 \) and the remaining parameters as in Fig. 3. Further numerical investigations of this particular point are beyond the scope of the present theoretical study.

To conclude, the present theoretical study has shown that both the extent of chaos and the suppressory effect of charge carriers in the laminar chaotic mixing associated with finite-amplitude electroconvection are rather complex phenomena in which the coupling between spatial charge distribution and time-dependent velocity field depends crucially on the spectral properties of temporal fluctuations of the velocity field, even in the simplest case of assuming them to be strictly periodic. Specifically, the width of the separatrix chaotic layer and the suppressory threshold value of the charge density were found to depend not only on the main harmonic (characteristic frequency) but also on the remaining harmonics (wave form) of the periodic fluctuations. A recent spectral analysis [Traoré & Pérez, 2012] of the temporal evolution of the maximum fluid velocity for the case of strong injection indicated that, for both discrete and broadband spectra, the characteristic frequency is proportional to the mean fluid velocity. Thus, while the present results provide some insight into the origin, extent, and disappearance of heteroclinic chaos in electroconvection due to unipolar injection, further analysis beyond the perturbative regime should take detailed spectral information into account.

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Hamiltonian Chaos in Nonsteady Finite-Amplitude Electroconvection


