Transient Two-Ion Chaos

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Transient order → chaos transitions of two Ba^+ ions are observed where the duration of the chaotic state increases smoothly with an increasing Mathieu control parameter q. The transient regime precedes the transition point q_c for stationary chaos, which is well defined and reproducible. The perturbative step that initiates chaos takes place on a time scale of 1 min, far longer than previous estimates, and is due to a single H_2-Ba^+ collision (H_2 pressure 5×10^{-11} Torr). In contrast to random collision events, chaos can be initiated deterministically by a new pulse technique. These observations resolve earlier anomalies and challenge computer simulations.

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The recent observation of deterministic chaos of two trapped Ba^+ ions[1] opens the way for understanding chaos in a simple physical system, an atomic two-body system. In these experiments, the ions are stored at the center of a Paul trap of radius $\bar{r}_0$ having an electric quadrupole potential

$$V=[V_{dc}+V_{ac}\cos(\Omega t)][z^2-(x^2+y^2)/2]/\bar{r}_0^2$$

with a static and an oscillatory component of rf frequency Ω. The ion-trap system is dissipative and thus is characterized by attractors[2] due to near-resonant laser light that cools the ions by imparting one momentum kick ($\hbar\mathbf{k}$) each excitation. In the ordered state, the two nonrotating ions execute only small harmonic oscillations (limit-cycle attractor), but in the chaotic state large erratic orbits appear (strange attractor). An order → chaos transition occurs when (1) the initial conditions are sufficiently energetic, the result of a perturbation, and (2) the control parameter q, which is proportional to the rf voltage, reaches a critical value. Recent calculations[3] of the Lyapunov exponents reveal that the route to two-ion chaos, which differs from previous scenarios such as period doubling, arises from ion-ion collisions that *turn on* the Coulomb interaction, the nonlinearity responsible for chaos.

Nevertheless, important aspects of the problem have remained elusive. For example, the perturbative mechanism that initiates chaos has never been tested, whether that be energy fluctuations arising from spontaneous emission during the laser-cooling process, infrequent collisions of the ions with the residual background gas at ultrahigh vacuum, or the weak noise of the rf source. In addition, the transition points are not always reproducible, but can be plagued by a randomness that is not understood. Thus, the Munich group[4-6] has reported that for Mg^+ ions order → chaos transitions are not reproducible whereas the reverse process chaos → order is. Based on these findings, they come to the surprising conclusion that order → chaos transitions simply do not occur while the reverse process does. Furthermore, their results conflict with our earlier observation[1] of a two-ion Ba^+ order → chaos transition point at $q_c\simeq0.86$.

In this Letter, we report the observation of transient two-ion chaos and identify the origin of the initiation step, topics that have not been addressed and are responsible for the above anomalies. We find that *transient chaos* is a precursor of *stationary chaos*, merging into it with increasing q at a well-defined and reproducible point. Since the time scale of transient chaos and the initiation step can each be as long as a minute or more, it now becomes clear that the randomness encountered arises when the time scale of the measurement is too short. Furthermore, we show that two-ion chaos can be launched deterministically using a simple pulse technique that bypasses the random initiation step. The observation of these long time scales provides a guide for future theoretical studies as well as a challenge for numerical simulations.

The experimental arrangement is an improved version of Ref. 1. Two Ba^+_{138} ions are stored in an electric quadrupole trap of radius $\bar{r}_0=0.25$ cm and are driven at an rf frequency of $\Omega/2\pi=3.55$ MHz, the background pressure being 5×10^{-11} Torr of H_2 as observed on a mass spectrometer. The ions are detected by scattered laser light at 55° to the trap z axis and at a wavelength of 493.4 nm (blue), the $6^2P_{1/2}\to6^2S_{1/2}$ transition, using a Surface Science imaging photon detector that records the spatial positions and the count rate, typically 1500 s^{-1}/ion with a 50-s^{-1} background. This cw laser is frequency locked to a Te_2 line that is 680 MHz below the Ba^+_{138} line center and then is phase modulated to produce a tunable sideband (0 to ±1 GHz) with a frequency stability of 0.5 MHz. Typically, this laser was tuned 40–80 MHz below the Ba^+_{138} line center. A collinear cw laser at 649.9 nm (red) excites the $5^2D_{3/2}\to6^2P_{1/2}$ transition to avoid loss to the $5^2D_{3/2}$ state, and similarly is locked to a reference cavity and phase modulated for tunability (stability 0.5 MHz). When the red laser is pulsed (6 nsec at a 25-MHz repetition rate), the three-level Ba^+ system behaves as a two-level system where the ions are cooled by the blue laser. Each laser beam is focused to a 75-μm beam waist at trap center, the blue and red cw laser powers being 20 μW and 1 mW, respectively.

Passage to chaos can be initiated by applying a dc voltage pulse across the trap, corresponding to the a term of the modified Mathieu equation[7] of motion

$$\ddot{R}_i+\Gamma\dot{R}_i+[a_i+2q_i\cos(2\tau)-\alpha/R^3]R_i=0\,. \qquad (1)$$

Here, R is the ion-ion distance, i is a Cartesian component, time $\tau=\Omega t/2$ and the potential $q\equiv q_3=4eV_{\rm ac}/m\Omega^2\bar{r}_0^2$ are dimensionless, Γ is a laser damping parameter, $\alpha=e^2/m\Omega^2$ is the ion-ion Coulomb coupling parameter, $q_1=q_2=-q_3/2$, and $a_i=2q_iV_{\rm dc}/V_{\rm ac}$ which is nonzero only during the pulse.

A dc pulse results in the ions receiving a kick in the Coulomb energy. Solutions[3] of Eq. (1) show that $\alpha/R^3>q^2/2$ is a necessary condition for chaos, which is equivalent to the two ions acquiring a Coulomb energy of $E_C=2\alpha/R>(2q\alpha)^{2/3}$. For $q=0.86$, $E_C>9.2$, which can be achieved according to numerical solutions of Eq. (1) with a pulse of amplitude $a=\pm0.2$ (±78 V) and duration $\Delta\tau=2.2$ (180 nsec). Entry to the chaotic state occurs when the symmetry of the ordered state is broken by the two ions exchanging position.[1] For this to occur, the potential surface $V=2\alpha/R+q^2(z^2/2+r^2/8)$ of the secular motion contains a saddle point at $(r,z)=(0,(2\alpha/q^2)^{1/3})$, a barrier that must be surmounted. The route is through the z motion which grows as $z(\tau)=z(0)e^{\lambda_z\tau}$, where $z(0)$ is the initial value of the ordered state at the end of the pulse. Numerical solutions of Eq. (1) for the above pulse reveal that the Floquet exponent $\lambda_z=0.12$. Once the z amplitude is of order 10, the r-z motions couple through the Coulomb interaction, allowing passage over the radial barrier. Numerical solutions for $q\geq0.86$ show that pulse preparation leads to chaos even for initial values as small as $z(0)=10^{-30}$. Since amplitude fluctuations arising from spontaneous emission are of order $\delta z=0.03$, pulse preparation assures entry to the chaotic state when q is sufficiently large.

The initial clue for transient chaos emerged from observations of the kind shown in Fig. 1 where the two-ion scattering rate is recorded as a function of time, with $q=0.81$ and all experimental parameters *fixed*. A transient bistability occurs where the two ions are either in the ordered state with a large scattering rate or in a disordered state with a low scattering rate. Figure 1 also reveals a random jumping back and forth between the two states and is unexpected. We find that the average time between jumps out of the ordered state is a measure of the perturbation that induces the order→chaos transition whereas the average time in the disordered state is a fundamental characteristic of the nonlinear dynamics, transient chaos, which is q dependent.

In Fig. 2, transients are induced by applying a sequence of dc pulses (80 V, 250 nsec). Pulses are applied at 20-s intervals in Fig. 2(a) where $q=0.75$ and the duration of each transient is $\sim$1 s, the resolution limit of the counter. Apparently, the phase of the ion motion is important because the amplitude of the transient varies, and in some cases is not discernible. We see in Fig. 2(b) that when $q=0.845$, the duration of the transient increases considerably, and in Fig. 2(c) where $q=0.87$, a single pulse tips the ions into the chaotic state where they remain for the duration of the measurement, in this case 3 min or 2 orders of magnitude longer than for $q\leq0.83$. Figure 3 summarizes these results for four in-

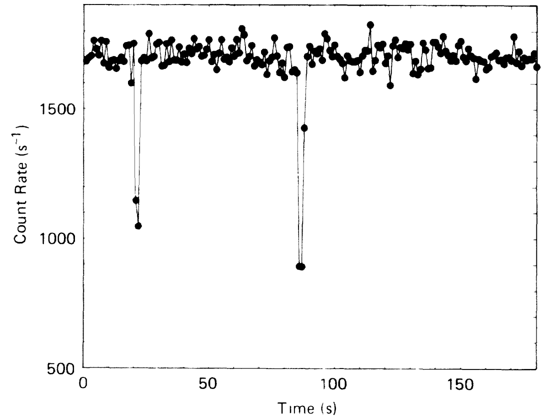

FIG. 1. Optical scattering rate of two Ba^+ ions vs time showing two H_2-Ba^+ collision-induced transients where $q=0.81$, $P_{H_2}=6.4\times10^{-11}$ Torr, and the time between collisions is 65 s. A dot represents 1 s.

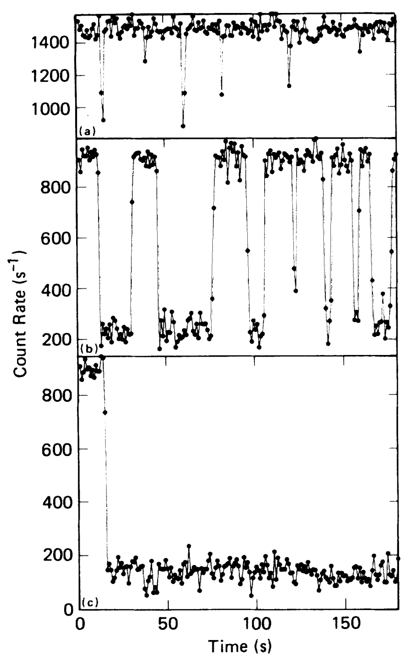

FIG. 2. Optical scattering rate of two Ba^+ ions vs time as a function of q. (a) Pulse-induced transients with $q=0.75$ and $P_{H_2}=6.4\times10^{-11}$ Torr. (b) Pulse-induced transient chaos with $q=0.845$ and $P_{H_2}=8.0\times10^{-11}$ Torr. (c) Pulse-induced stationary chaos with $q=0.87$ and $P_{H_2}=8.0\times10^{-11}$ Torr.

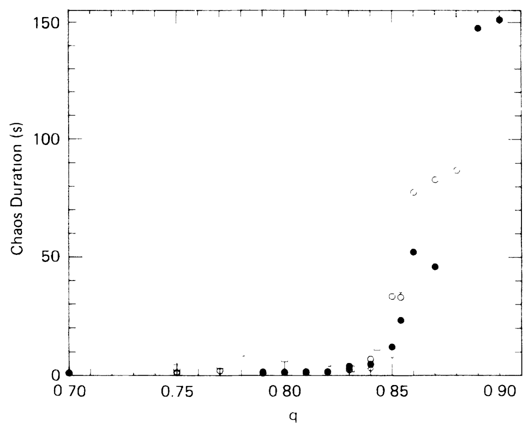

FIG. 3. Average time T in the chaotic state vs q. ●, collision-induced chaos with $P_{H_2}=1.2\times10^{-10}$ Torr; ○, pulse-induced chaos with $P_{H_2}=1.3\times10^{-10}$ Torr; □, pulse-induced chaos with $P_{H_2}=8.0\times10^{-11}$ Torr; ◇, pulse-induced chaos with $P_{H_2}=6.4\times10^{-11}$ Torr. Each point is an average of several observations, typically five or more.

dependent sets of data, three corresponding to pulse-induced and one to spontaneously induced transients. It is apparent that the average period T in the disordered state increases smoothly with q, the most precipitous change occurring at $q_c=0.855$ which we designate as the transition point to stationary chaos. We therefore label these transients as transient chaos. In the region $0.855 \le q < 0.91$, stationary chaos appears to persist up to the two-ion center-of-mass Mathieu instability of $q=0.91$ at which point the chaotic cloud is no longer stable. This behavior is observed whether the initiation step is a dc pulse or a spontaneous perturbation, processes we consider below.

Stationary chaos implies that the two ions remain in the chaotic state indefinitely, which in a rigorous sense cannot be tested due to the finite duration of the measurement. Therefore, there exists an inescapable uncertainty as to whether the two-ion system is in a true stationary state or whether it displays transient chaos on an extraordinarily long time scale. In the region $0.855 \le q < 0.91$, we find that the ions are usually in the chaotic state for the duration of the 3-min measurement, but infrequently for some trajectories they revert back to the ordered state. For the purpose of this Letter, we define *stationary chaos* to mean that on the average the ions reside in the chaotic state most of the observation time and for most trajectories. It is interesting that the same ambiguity arises in computer simulations.[3]

Numerical solutions of Eq. (1) for $q=0.86$ and $\Gamma=0.0005$ indicate that an order → chaos transition requires a perturbative energy kick of $\delta\dot{z}^2=0.28$ (reduced units) or a temperature rise of $\delta T=0.6$ K. A more realistic calculation, which gives the same result,[8] replaces Γ by spontaneous-emission fluctuations where Bloch-like equations are coupled to the modified Mathieu equation, Eq. (1). At equilibrium these fluctuations contribute an average energy $\delta\dot{z}^2=2\times10^{-3}$ for $q=0.86$, a laser detuning of $\Delta=10$, and a Rabi frequency $\chi=8.9$, all in reduced units. This energy is about 2 orders magnitude smaller than the 0.28 required for triggering chaos. Hence, contrary to our initial discussion,[1] spontaneous-emission fluctuations do not initiate two-ion chaos. A second possibility of rf noise emanating from the rf amplifier is excluded because increasing this noise level by 4 orders of magnitude above the Johnson noise limit does not affect the results.

Finally, H_2-Ba^+ elastic collisions, although infrequent at an H_2 pressure of 5×10^{-11} Torr, are sufficiently energetic to effect chaos. Assuming an interaction potential of the form $V=-\alpha e^2/r^4$, where α is the hydrogen-molecule polarizability, we numerically determine the scattering angle[9,10]

$$\theta=\pi-\frac{2\sqrt{2}}{1+[1-(b_0/b)^4]^{1/2}}F(\pi/2|\alpha) \tag{2}$$

in terms of the impact parameters for capture (b_0) and scattering (b) and the complete elliptic integral of the first kind $F(\pi/2|\alpha)^7$. Here,

$$\sin^2\alpha=\frac{1-[1-(b_0/b)^4]^{1/2}}{1+[1-(b_0/b)^4]^{1/2}}$$

and $b_0^2=2e\sqrt{\alpha/E_1}$, with E_1 the room-temperature kinetic energy of the incident H_2. The scattering angle in turn is determined by $\delta E/E_{\max}=\sin^2(\theta/2)$, where δE is the collisional energy transfer, which must satisfy the minimum requirement for chaos, and $E_{\max}\simeq4(m_{H_2}/m_{Ba^+})E_1$. Thus, the maximum scattering cross section resulting in chaos is $\sigma_{\max}=278$ Å^2, whereas the minimum value is limited by the capture cross section $\sigma_c=\pi b_0^2=188$ Å^2. Ignoring this small variation, we take $\sigma_{\max}$ to calculate the average elastic-scattering rate $\gamma=2n\sigma_{\max}v$, the factor of 2 accounting for the two Ba^+ ions, with n and v the H_2 density and average velocity. These calculations are compared to average experimental collision rates γ in Fig. 4 where the H_2 pressure varies by a factor of 50. The agreement is remarkably good, and supports the idea that the spontaneously initiated path to chaos is via collisions. Thus, at the low H_2 pressure of 6.4×10^{-11} Torr, a sufficiently energetic collision occurs on average once every 80 s, consistent with Fig. 1. It is interesting that a *single* perturbative event, either a collision or a dc voltage pulse, is sufficient to start the two ions on their path to chaos.

Consider now the earlier experiments[1,4-6] where the transition points were observed by sweeping the rf voltage rapidly. The observation of an order → chaos transition then depends on the probability of a sufficiently energetic collision occurring during the sweep. Thus, on some occasions when no collisions occur, a transition will not be seen [Fig. 6(b) of Ref. 5]. Moreover, the transitions observed were most often transient rather than sta-

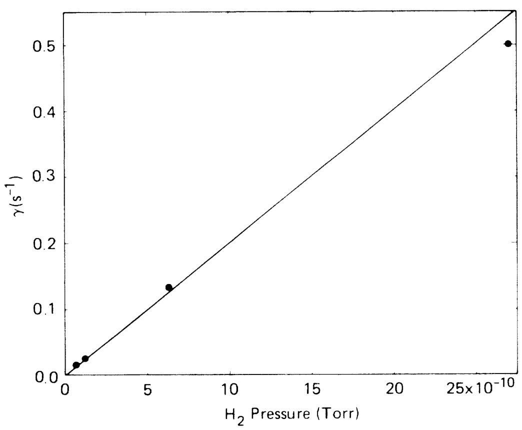

FIG. 4. Average rate γ of H_2-Ba^+ collisions vs H_2 pressure. ——, theory; ●, experiment.

tionary chaos, explaining the large variation in the transition points.[4-6] On the other hand, in this Letter the transition to stationary chaos begins at a well-defined value of $q_c = 0.855$, in agreement with our preliminary result.[1] The conclusion[4-6] that such a transition will not occur until the two-ion center-of-mass Mathieu instability is reached at $q = 0.91$ is clearly erroneous.

Numerical simulations[1,3] of Eq. (1) with fixed damping indicate an order $\rightarrow$ stationary chaos transition at $q_c = 0.86$ and transient chaos at lower q values. However, the integration times have not exceeded 2×10^6 (reduced units) or 0.2 s, which falls far short of the several minute time scales encountered experimentally. When Eq. (1) is modified to include variable damping and spontaneous-emission fluctuations,[8] the algorithm becomes even slower due in part to the greatly reduced damping time in the chaotic state. In addition, frequency locking is highly evident in the calculations, but thus far there is no evidence experimentally.

Finally, recent studies[11,12] of dissipative dynamical systems show that chaotic transients can replace a chaotic attractor when a control parameter exceeds a critical value, $q > q_c$. The average transient lifetime then scales as $T \sim |q - q_c|^{-\gamma}$, where γ is a critical exponent. It follows that these predictions also apply for the inverse process, the subject of this Letter, where chaotic transients precede a chaotic attractor that is suddenly created as the parameter increases. From the data of Fig. 3, we find that the above scaling is obeyed with $\gamma = 1.1$. Whether γ itself can be predicted awaits further study. Thus, this prediction[11] corroborates our findings that the stationary two-ion order $\rightarrow$ chaos transition occurs at the unique value q_c and is preceded by transient chaos.

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[1]J. Hoffnagle, R. G. DeVoe, L. Reyna, and R. G. Brewer, Phys. Rev. Lett. **61**, 255 (1988).

[2]P. Berge, Y. Pomeau, and C. Vidal, *Order Within Chaos* (Wiley, New York, 1984).

[3]R. G. Brewer, J. Hoffnagle, R. G. DeVoe, L. Reyna, and W. Henshaw, Nature (London) **344**, 305 (1990).

[4]F. Diedrich, E. Peik, J. M. Chen, W. Quint, and H. Walther, Phys. Rev. Lett. **59**, 2931 (1987); R. Blumel, J. M. Chen, E. Peik, W. Quint, W. Schleich, Y. R. Shen, and H. Walther, Nature (London) **334**, 309 (1988).

[5]R. Blumel, C. Kappler, W. Quint, and H. Walther, Phys. Rev. A **40**, 808 (1989).

[6]R. Blumel, E. Peik, W. Quint, and H. Walther, in *Quantum Optics V*, edited by J. D. Harvey and D. F. Walls (Springer-Verlag, Berlin, 1989), p. 152.

[7]M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (U.S. GPO, Washington, DC, 1968), p. 722.

[8]L. Reyna, J. Hoffnagle, and R. G. Brewer (to be published).

[9]L. D. Landau and E. M. Lifshitz, *Mechanics* (Pergamon, New York, 1978), p. 41.

[10]J. L. Synge, in *Encyclopedia of Physics*, edited by S. Flugge (Springer-Verlag, Berlin, 1960), Vol. 3, Pt. 1, pp. 1-225.

[11]C. Grebogi, E. Ott, and J. A. Yorke, Phys. Rev. Lett. **57**, 1284 (1986).

[12]W. L. Ditto *et al.*, Phys. Rev. Lett. **63**, 923 (1989).