frequency detuning. Finally, the emission spectrum (vibrational intensities) must be independent of the wavelength of the transfer laser. All of these properties are in direct contrast to what is actually observed.

It is possible to make an estimate of the cross section observed experimentally for the sum of processes (1a) and (1b). We calculate that 2% of the $A^1\Pi$ state is transferred to the $B^1\Sigma^+$ state via this laser-assisted collisional process at 8×10^9 W/cm. This corresponds to a total cross section of $\sigma_{a+b}=5\times10^{-16}$ cm^2 compared to our estimate of $\sigma_{a+b}=20\times10^{-16}$ cm^2. Agreement is reasonable considering the total uncertainties in the input parameters.

This work shows the important role that laser-assisted collisional phenomena can play in laser spectroscopy studies at high laser intensities. As the first observation of switched collisions to achieve inelastic energy transfer in a molecular system, it is an important step in developing and understanding models for laser field effects in molecular dynamics, and provides the basis for exploring dressed-state chemistry in other areas such as photofragmentation and even reactive scattering. Finally, the large cross sections achieved in these experiments establish the feasibility of future applications, as for example exciting vacuum ultraviolet lasers.

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Transition to Chaotic Behavior via a Reproducible Sequence of Period-Doubling Bifurcations

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We present the results of measurements of the vertical and horizontal temperature gradients in a Rayleigh-Benard cell. By an appropriate preparation of the initial state, the system can be brought into a single-frequency oscillatory regime. Further stepwise increase in the imposed temperature gradient makes the system go through a reproducible sequence of period-doubling bifurcations up to $f_1/16$. The Feigenbaum δ and μ universal numbers are determined.

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A recent theory by Feigenbaum[1,2] suggests that nonlinear systems which can be led into chaotic behavior via a sequence of period-doubling bifurcations will exhibit universal behavior. As the stress parameter λ is increased, the bifurcation points λ_n occur in such a way that the ratio

$\delta_n = (\lambda_{n+1} - \lambda_n)/(\lambda_{n+2} - \lambda_{n+1})$ will approach the universal number $\delta = 4.669\ldots$. Furthermore, an appropriately defined ratio of the amplitudes of the Fourier components of neighboring subharmonics will also approach a universal number $\mu = 6.574\ldots$.

Experimental evidence supporting the applicability of the Feigenbaum picture to real physical systems is not very rich. Libchaber and Maurer[3-5] have reported spectra obtained in a low-Prandtl-number Rayleigh-Benard system showing remarkable agreement with the regular subharmonic amplitude decrease predicted by the theory. Because of the lack of resolution in R/R_c, the number δ could only be estimated. Gollub and Benson[6] have reported on the observation of period-doubling bifurcations in an intermediate-Prandtl-number Rayleigh-Benard system. Again the spectra shown exhibit the regular decrease in subharmonic amplitudes.

In this Letter we report on results obtained in a low-aspect-ratio Rayleigh-Benard cell filled with water. We have found a technique for the preparation of the initial state of the system which leads to a very reproducible sequence of period-doubling bifurcations up to $f_1/16$. We determine the first three terms in the δ_n sequence. Also we determine the value of the number μ for a few spectra close to bifurcation points. The overall experimental evidence is in favor of the Feigenbaum picture.

The inner volume of the convection cell is 25 mm wide, 15 mm long, and 7.9 mm high. The lateral boundaries are four 5-mm-thick glass plates. They are glued between two aluminum blocks, whose temperature difference is controlled to within 2 mK ($\Delta T_{\rm chaos}$ is close to 7 K). Measurements of the vertical and horizontal temperature gradients averaged along a horizontal line parallel to the short side, roughly at midheight, and close (3 mm) to the sidewall, are obtained by a laser-beam-deflection technique. Deflections along both axes are determined simultaneously to an accuracy of a few microradians with the aid of a two-axis solid-state quadrant sensor (typical angular oscillations are close to 1 mrad). The two signals can be plotted one against the other on an XY recorder (pen recorder or digital oscilloscope). Alternatively, each signal can be fed to a fast-Fourier-transform analyzer for spectral analysis.

An important point we must make is related to the manipulations we perform in order to prepare the initial state of the system. If the temperature difference is increased in small steps starting from zero, and observations are made under stationary conditions at each fixed temperature difference, we notice that the system has a quite complicated behavior and the route to chaos does not seem to be uniquely defined. We have not attempted to collect data under these conditions and therefore we cannot make any meaningful comparison with the results obtained by other authors[6] under similar conditions. We have found, however, that if we suddenly apply a large temperature difference (larger than $\Delta T_{\rm chaos}$), and then we rapidly come back to smaller ΔT values, the system starts oscillating very regularly. If the temperature difference is then changed in small steps, the system can be brought into dynamical states which are very reproducible. Each run (which lasts typically two weeks) consists therefore of one initial quenching operation followed by a sequence of small variations of the temperature difference. Each measurement is performed at fixed ΔT (ΔT is our stress parameter λ). Data collected over three runs show very good consistency, and the details of the initial quenching operations seem to have little influence. It appears that the quenching technique brings the system into a phase-space basin from which a Feigenbaum-type route to chaos is accessible.

The actual planform of the instability is rather unclear. Crude whole-field shadowgraph observations indicate that the temperature-gradient oscillations are generated by propagating features which regularly pass under the exploring beam. It seems unlikely that we are dealing with oscillations of a simple spatial structure like in the case of the oscillatory instability studied by Libchaber and Maurer.[3,4] In Fig. 1, we report a sequence of temperature-gradient orbits observed at different values of ΔT (R/R_c values are reported, where R is the Rayleigh number and R_c is the value at threshold for convection). Splittings leading to the appearance of $f_1/8$ can

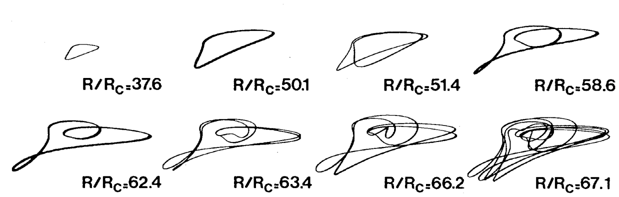

FIG. 1. Plots of vertical temperature gradients vs horizontal temperature gradients for different values of R/R_c.

be easily identified in the sequence. We point out that each orbit has been retraced at least fifteen times. Also, orbits obtained at the same R/R_c but on different days are virtually superimposable, and this justifies our previous comments on the general reproducibility of the data.

We must point out, however, that orbits can appear in a rather different form. Immediately after a bifurcation, orbits split into two closely lying replicas that eventually become more separated and distorted as λ is further increased. By recording the orbit traces over extended periods of time, we noticed that the separation of the newly split orbits did not remain constant. Indeed, the orbits execute a very slow oscillatory motion passing back and forth through each other. The orbits never quite retrace themselves, thus indicating that the oscillatory motion occurs at an incommensurate frequency. This effect is quite noticeable after the λ_3 and λ_4 bifurcations. Somewhere in between λ_3 and λ_4 the oscillations disappear, and the orbits resume their stable form as indicated at the end of the sequence in Fig. 1. Beyond λ_4, however, we have never been able to observe stable sixteenfold orbits.

The above observation is essential in order to understand some features of the spectra shown in Fig. 2. The spectra refer to the signals of the horizontal temperature gradient recorded after the λ_2, λ_3, and λ_4 bifurcations. The position of the fundamental frequency f_1 is indicated by the arrow. At $R/R_c=62.6$ the $f_1/4$ is already present, and all the frequency components are in the form of sharp peaks. At $R/R_c=66.2$ the emerging $f_1/8$ appears in the form of a finely divided doublet, with separation close to $f_1/38$. Notice that all the other features are sharp, as expected as a consequence of the orbit oscillatory behavior (orbit oscillations introduce an almost 100% modulation on the emerging subharmonic and its odd multiples). At $R/R_c=67.4$ the emerging subharmonic $f_1/16$ is in reality an even more widely spaced doublet with separation close to $f_1/19$ (twice that of the previous bifurcations). Lack of graphical resolution prevents close examination of the detail, but expanded scale plots show the splitting in a very unambiguous way. Notice that at this stage the splitting is barely larger than the frequency of the new subharmonic.

At this stage, we can attempt to estimate the Feigenbaum universal number μ. Unfortunately, the interpolation and averaging procedure necessary to construct $S(i)$ according to the original Feigenbaum scheme is fairly complex and difficult to use in analyzing our data. (It is best applicable when a large number of bifurcations has occurred.) We have, therefore, taken for $S(i)$ the geometric average of the odd multiples of the 2^ith subharmonic, and we define $\mu_{n,i}$ as the ratio $S(i)/S(i+1)$ evaluated immediately after the λ_n bifurcations.[7] If the average is taken up to $2f_1$, we obtain $\mu_{3,1}=4.1$, $\mu_{4,1}=3.8$, and $\mu_{4,2}=3.8$. An average up to $4f_1$ yields $\mu_{3,1}=4.0$, $\mu_{4,1}=4.2$, and $\mu_{4,2}=3.6$. These numbers are appreciably lower than the value reported in the introduction but in slightly better agreement with the value $\mu\sim 5.0$ estimated by Feigenbaum[8] when one takes for $S(i)$ the geometric average. We can also compare

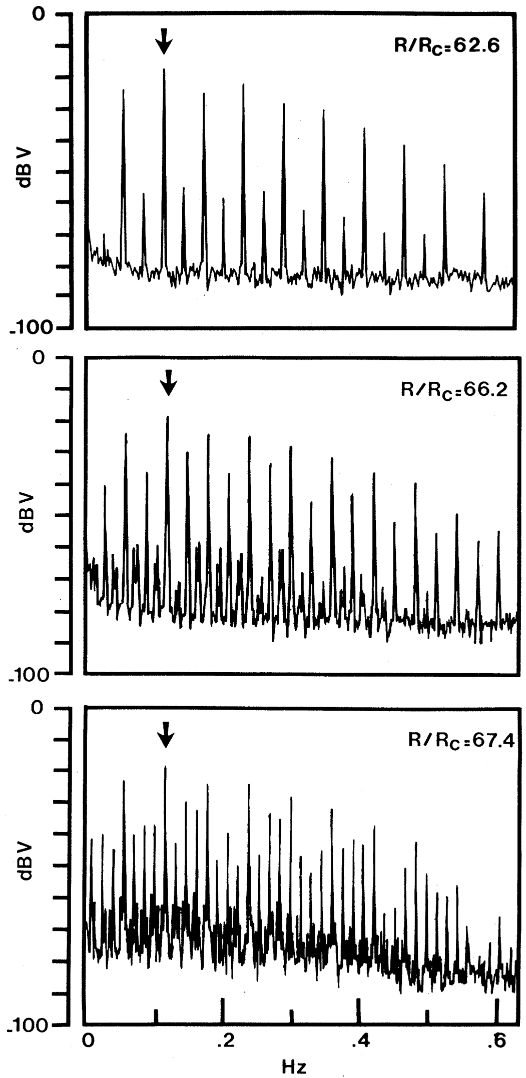

FIG. 2. Horizontal temperature-gradient spectra obtained close to λ_2, λ_3, and λ_4. The arrow indicates the position of f_1.

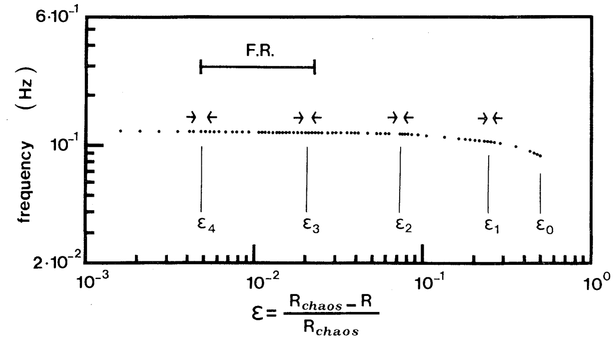

FIG. 3. Log-log plot of f_1 as a function of $\epsilon = (R_{\text{chaos}} - R)/R_{\text{chaos}}$. The data have been obtained over three runs. The location of bifurcations is reported together with error bars indicating the ϵ ranges outside which the presence (or absence) of a new subharmonic could be unambiguously assigned.

our results with the prediction $\mu = 4.58$ put forward by Nauenberg and Rudnick[9] who take for $S(i)$ the rms integrated spectrum over all odd subharmonic multiples. When analyzed in this way, our data give $\mu_{3,1} = 3.3$, $\mu_{4,1} = 3.0$, and $\mu_{4,2} = 4.0$ (averaged up to $4f_1$). Experimental results seem invariably smaller than the theoretical predictions.

Since the locations of the first four bifurcations are known, we can calculate the first three values of the δ_n sequence. They are $\delta_0 = 1.35$, $\delta_1 = 3.16$, and $\delta_2 = 3.53$. Estimates for δ_n can also be obtained by presenting the data in a different way. From the location of the bifurcations we can estimate R_{chaos}. We report in Fig. 3 the behavior of f_1 as a function of $\epsilon = (R_{\text{chaos}} - R)/R_{\text{chaos}}$, and we indicate the location of the bifurcations on this scale. Since the plot is logarithmic, the spacing of the bifurcations should approach a constant length (this is a consequence of the fact that the λ_n are described by a geometric sequence). In Fig. 3 we also report the length of the Feigenbaum ratio (F.R.) for comparison. The actual values thus determined are $\delta_0 = 2$, $\delta_1 = 3.3$, $\delta_2 = 3.6$, and $\delta_3 = 4.3$ and typical error bars can be estimated from the figure.

Beyond the last bifurcation we have indications that the system is deviating from the Feigenbaum picture. It is tempting to say that the crossover between the last subharmonic frequency and orbit oscillation frequency is heralding the premature termination of the sequence.

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