Synchronization and Chaos Induced by Resonant Tunneling in GaAs/AlAs Superlattices

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A semiconductor superlattice represents an ideal one-dimensional nonlinear dynamical system with a large number of degrees of freedom. The effective nonlinear coupling originates from sequential resonant tunneling between adjacent wells. We have observed spontaneous chaotic and periodic current oscillations in a doped GaAs/AlAs superlattice by changing only the applied bias. When the system is driven with an incommensurate sinusoidal voltage for a fixed bias, transitions between synchronization and chaos are observed via pattern forming bifurcations. A driving signal of sufficiently large amplitude can suppress the occurrence of chaos and produce a synchronized oscillation mode with a subharmonic of the driving frequency. [S0031-9007(96)01318-X]

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Chaotic behavior of nonlinear systems with many degrees of freedom is a subject of great current interest [1-6]. In particular, the influence of disorder on the chaotic behavior has been reported very recently [6]. Some authors have performed theoretical investigations of chaotic behavior induced by a purely quantum mechanical process such as resonant tunneling [7,8]. Recently, theoretical and experimental investigations on the quantum signatures of classical chaos in single particle systems have been reported, where the effect of classical chaotic motion on the quantum mechanical energy spectrum of quantum wells in high magnetic fields has been investigated [9]. Criteria for stability in systems with negative differential resistance have also attracted recent attention [10]. Although many theoretical investigations have dealt with a system with a large number of degrees of freedom, there are only a few reports available of experimental studies.

Semiconductor superlattices are known to represent a nonlinear system exhibiting the formation of electric field domains [11–16]. The nonlinearity originates from negative differential conductance induced by sequential resonant tunneling. Under domain formation the I-V characteristic clearly shows multistability [17], which is a typical property of a nonlinear system. Recently, spontaneous self-oscillations of the current were observed in a biased GaAs/AlAs superlattice (SL) [18]. Sequential resonant tunneling between adjacent wells leads to a complete loss of the phase memory of the electrons after completion of the sequential tunneling process. Therefore, the system can be described by two dynamical variables (electron density and electric field) and a quasi-Fermi level for each quantum well. The chaotic behavior of oscillating electric field domains in semiconductor superlattices was investigated theoretically by Bulashenko and Bonilla [19]. However, it was assumed that all wells of the superlattice are identical so that the transport can be described by an effective drift velocity, which is the same for all wells. In this description, the number of degrees of freedom is small, although the studied system contains many wells.

In this Letter, we demonstrate that such a biased, weakly coupled GaAs/AlAs superlattice exhibits spontaneous periodic and chaotic current oscillations as well as transitions between synchronization and chaos via pattern forming bifurcations when driven with an incommensurate sinusoidal modulation voltage. In contrast to the results in Ref. [19], the observed pattern is much richer in structure leading to the conclusion that this system represents a model system for a one-dimensional chain of nonlinear oscillators with a large number of degrees of freedom. At sufficiently large driving amplitudes, chaos is suppressed and a synchronized oscillation mode with a subharmonic of the driving frequency is observed.

The investigated sample consists of a 40-period, weakly coupled GaAs/AlAs superlattice with 9.0-nm GaAs wells and 4.0-nm AlAs barriers with each GaAs well being Si doped at 3.0×10^{17} cm⁻³ within the central 5.0 nm. Further details of the sample structure and characterization are given in Refs. [17,18]. All experimental data have been recorded in a He-flow cryostat using high-frequency coaxial cables with a bandwidth of 20 GHz. The time-averaged current voltage characteristics are recorded by a Keithley SMU 236 using a time constant, which is orders of magnitude larger than the period of the oscillations. The current oscillations are detected with a Tektronix CSA 803 sampling oscilloscope and an Advantest R3361 spectrum analyzer. The driving voltage is provided by a Wavetek 81 function generator.

Figure 1(a) shows the frequency spectra of spontaneous current oscillations as a function of the bias for voltages between 6.670 and 8.222 V. The current oscillations ex-

hibit no damping when measured in real time. The logarithm of the amplitude of the current oscillation is indicated on a gray scale. The oscillations occur in a plateau of the time-averaged I-V characteristic, which is shown in the inset of Fig. 1(b). This plateau originates from electric-field domain formation due to sequential resonant tunneling between different conduction subbands in adjacent wells. With increasing bias, several chaotic windows are observed. Between the chaotic windows the oscillations are periodic except for the voltage regime between 6.694 and 6.728 V, where no oscillations appear. The evolution process of the current oscillation with bias exhibits a small hysteresis. The transitions from the chaotic to the periodic windows occur over a very small voltage range, which is on the order of mV. However, the transitions from periodic to chaotic window usually take place over a much wider voltage range through a random-enhancing process, which contains several chaotic and periodic win-



FIG. 1. (a) Frequency spectra of spontaneous current oscillations vs applied voltage at 30 K. The current power spectra are shown as a density plot on a logarithmic scale, where darker areas correspond to larger amplitudes. (b) Time-averaged I-Vcharacteristic at 30 K with the inset showing the whole plateau.

dows. Surprisingly no quasiperiodic oscillatory modes are observed.

The structures in the current plateau shown in Fig. 1(b) are quite different from the regular spikes in the I-V characteristic, which are induced by the motion of the domain boundary under static electric-field domain formation [11-14]. The chaotic windows in Fig. 1(a) are correlated with the appearance of regions with large negative differential conductivity (NDC) in the I-V characteristics in Fig. 1(b), while the periodic windows occur for positive differential conductivity (PDC) or small NDC. The transitions from the chaotic to the periodic windows take place over a very narrow voltage range, over which the average current decreases sharply. Note that the shape of the peaks between 6.8 and 7.6 V are to some extent self-similar in the same way as the chaotic windows in Fig. 1(a). This self-similarity could indicate universality for the transitions from synchronization to chaos.

From the time-averaged I-V characteristics, we can obtain some important information about this spatially distributed dynamical system. In theoretical investigations, some authors use averaged equations in order to analyze the dynamics of an oscillator system with N degrees of freedom [20,21]. For our SL system, a change in the dc voltage can result in a different coupling between the degrees of freedom. PDC regions characterize attractive coupling between the degrees of freedom, i.e., the SL system behaves as a self-synchronized unit. Therefore, we observe the spontaneous periodic current oscillation in the PDC regions [2,22]. However, for NDC regions the coupling is repulsive. For large NDC, the coupling becomes strongly repulsive. With increasing repulsion, the synchronized oscillations between the degrees of freedom become more and more destabilized [2], and finally this SL system enters a chaotic or even turbulent state for large NDC.

In order to investigate the transition from synchronization to chaos for an external driving voltage, the dc bias was fixed at 8.35 V and temperature was set to 4.2 K, where periodic current oscillations are observed with a fundamental frequency f_0 of 23.14 MHz. In order to vary the nonlinear coupling, the amplitude of the driving voltage was changed between 0 and 0.1 V, and the response of the superlattice was studied for different driving frequencies. In Fig. 2 two examples are displayed showing the evolution of the current amplitude spectra as a function of the amplitude of driving signal for $f_d = 50$ [Fig. 2(a)] and 37.5 MHz [Fig. 2(b)]. For each amplitude step, the logarithm of the amplitude of the current oscillation is shown on a gray scale. For small driving amplitudes at 50 MHz, the driven SL system enters frequency-locked states with a rational winding number f_0/f_d of 6/13, which is close to the frequency ratio of 0.463. Outside the first frequency-locked region, each frequency branch splits into two or three branches, and the frequency spacing continuously increases with increasing driving amplitude. When the branches cross, they are locked to new periodic states. In this regime,



FIG. 2. Frequency bifurcation diagrams for a driving frequency of 50.0 (a) and 37.5 MHz (b) at 4.2 K. The logarithmic current power spectra are shown as a density plot vs the driving voltage. Darker areas correspond to larger amplitudes. The winding numbers are indicated above each graph.

the SL system alternately enters quasiperiodic and periodic states with rational winding number n/(2n + 1) for $n = 7, 8, 9, \ldots$, respectively. The periodic windows, however, are very narrow. This alternate transformation becomes more frequent with increasing driving amplitude before the SL system finally enters the chaotic state.

Although the frequency bifurcations of this driven system are complicated, all the peaks in the spectra as shown in Fig. 2(a) are linear combinations of the two basic frequencies f_0 and f_d . The scenario of quasiperiodic and frequency-locked behavior is associated with a characteristic self-similar emergence of high-order mixing frequencies [23]. The spatial synchronization remains until the chaotic state is reached. Furthermore, at the onset of the chaotic window in Fig. 2(a), the spectra consist of broad peaks with a weaker noisy background, i.e., the chaos is synchronized. A further increase of the driving amplitude enhances the noisy background and results in a randomization of the current oscillations. Actually, the chaotic state attains a higher complexity, in which the SL has entered chaos with the breakdown of spatial coherence [1].

Other driving frequencies can produce much more complicated patterns. Figure 2(b) shows the frequency bifurcation diagram of this SL system for a driving frequency adjusted to the golden mean (1.618) times the fundamental frequency. A pattern forming bifurcation sequence is observed with increasing driving amplitude. Several distinct windows appear with different behavior. When the driving amplitude is smaller than 0.028 V, the transition to chaos is ascribed to a quasiperiodic route with two independent frequencies f_0 and f_d . In this window, the chaos remains synchronized, since now the driven SL system acts as a forced single anharmonic oscillator. As the driving amplitude is increased to 0.030 V, the chaotic behavior disappears and the system enters a frequency-locked state with a rational winding number of n/(2n - 1) with n = 3, which again is close to the frequency ratio of 0.617. The driving signal seems to have effectively suppressed the chaotic signal [3]. This state remains until the driving amplitude reaches 0.031 V. Then, each peak splits into many peaks, and the frequency spacing between these new peaks increases with increasing driving amplitude. In contrast to the situation at low driving amplitudes (< 0.028 V), the closely spaced peaks will lock together to form a new frequency-locked state. The widths of the frequency-locked windows are determined by the strength of the locked peaks. The crossings of the main peaks are responsible for the alternating formation of a series of frequency-locked windows, whose corresponding rational winding numbers are n/(2n-1)for $n = 4, 5, 6, 7, 8, \ldots$ While this kind of bifurcation is similar to the one in Fig. 2(a), there are more complicated bifurcations between the frequency-locked windows in Fig. 2(b), which exhibit self-similarity.

The bifurcation sequence between the main frequencylocked windows as shown in Fig. 2(b) is indeed of high complexity. The new frequency-locked states, which evolve from coupled crossings of the frequency branches, are unstable and always superimposed by intermittent chaos, which originates from the burst instabilities of spatial synchronization. These so-called frequency-locked states are actually laminar periodic oscillations. The frequency-locked peaks are then divided into many peaks, which alternately produce new frequency-locked states (actually laminar periodicity) [1]. During these bifurcation sequences, the evolution tendency of the main peaks remains almost unchanged, even though they also interact with other peaks. This kind of cascade bifurcation sequence has resulted in the complicated mosaic structure as shown in Fig. 2(b). The theoretical investigation of such a system did not reveal such a pattern formation with the sequence of winding numbers given above [19]. There are several possible explanations for this discrepancy. First, the periodicity of the experimental superlattice might by disturbed by fluctuations of doping and/or

well and barrier thickness. This would clearly add additional degrees of freedom to the experimental system. Second, the drift velocity curve assumed for the calculations in Ref. [19] was assumed the same for all periods. This curve might change from period to period due to differences in doping, layer thickness, and interface quality, which again would result in an increase of the number of degrees of freedom.

Finally, we will discuss the driving amplitude range above 0.08 V. In Fig. 3 the power spectra for three different driving amplitudes are shown. At 0.0805 V, the system exhibits a spectrum with broadened peaks indicating that the system has entered the chaotic state, but with some residual spatial stability. For 0.0810 V, the spectrum has evolved to broadband noise. The spatial and temporal motion has become more randomized and unstable; i.e., the SL system has now entered the chaotic state, which may be of spatiotemporal nature. However, the chaotic window is narrow. As the driving amplitude is further increased to 0.0830 V, some chaotic modes are softened, but the distribution of the softened modes in the spectra is randomized in time. Figure 3 only displays snapshots of the spectra. The enhanced driving amplitude will produce more softened modes until the formation of a new, stable frequency-locked state with a subharmonic and harmonics of the driving frequency occurs.

In conclusion, we have observed synchronization and temporal chaos without an external driving voltage in a semiconductor superlattice. In the synchronized state, the SL exhibits a large internal multiplicity. A small driving signal can destroy this multiplicity and create a series of new oscillation modes, which results in a complex pattern forming bifurcations. When the driving amplitude



FIG. 3. Snapshots of the current power spectra for a driving frequency of 37.5 MHz with different driving amplitudes as indicated at 4.2 K.

becomes very large, the driving, linear oscillator dominates, and the coupled oscillator system is locked to periodic states with the driving frequency, its harmonics, and a subharmonic.

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- J. Crutchfield and K. Kaneko, in *Directions in Chaos*, edited by Hao Bai-lin (World Scientific, Singapore, 1987), Vol. 1, p. 272.
- [2] Y. Kuramoto, *Chemical Oscillations, Waves and Turbulence* (Springer-Verlag, New York, 1984).
- [3] H. Gang and Q. Zhilin, Phys. Rev. Lett. **72**, 68 (1994); D. Auerbach, *ibid*. 72, 1184 (1994); I. Aranson, H. Levine, and L. Tsimring, *ibid*. **72**, 2561 (1994).
- [4] J. M. Flesselles, V. Croquette, and S. Jucquois, Phys. Rev. Lett. 72, 2871 (1994).
- [5] V.I. Nekorkin and V.A. Makarov, Phys. Rev. Lett. 74, 4819 (1995).
- [6] Y. Braiman, J.F. Lindner, and W.L. Ditto, Nature (London) 378, 465 (1995).
- [7] G. Jona-Lasinio, C. Presilla, and F. Capasso, Phys. Rev. Lett. 68, 2269 (1992).
- [8] Y. Ashkenazy et al., Phys. Rev. Lett. 75, 1070 (1995).
- [9] T. M. Fromhold *et al.*, Phys. Rev. Lett. **72**, 2608 (1994);
 T. M. Fromhold *et al.*, *ibid*. **75**, 1142 (1995);
 G. Müller *et al.*, *ibid*. **75**, 2975 (1995);
 P. B. Wilkinson *et al.*, Nature (London) **380**, 608 (1996).
- [10] A. Wacker and E. Schöll, J. Appl. Phys. 78, 7352 (1995).
- [11] L. Esaki and L. L. Chang, Phys. Rev. Lett. 33, 495 (1974).
- [12] K. K. Choi et al., Phys. Rev. B 35, 4172 (1987).
- [13] H.T. Grahn et al., Phys. Rev. Lett. 67, 1618 (1991).
- [14] Y. Zhang et al., Appl. Phys. Lett. 65, 1148 (1994).
- [15] S. H. Kwok et al., Phys. Rev. B 51, 9943 (1995).
- [16] B.J. Keay et al., Phys. Rev. Lett. 75, 4098 (1995).
- [17] J. Kastrup et al., Appl. Phys. Lett. 65, 1808 (1994).
- [18] H. T. Grahn *et al.*, Jpn. J. Appl. Phys. **34**, 4526 (1995); J. Kastrup *et al.*, Phys. Rev. B **52**, 13761 (1995).
- [19] O. M. Bulashenko and L. L. Bonilla, Phys. Rev. B 52, 7849 (1995); O. M. Bulashenko, M. J. García, and L. L. Bonilla, *ibid.* 53, 10008 (1996).
- [20] S. Watanabe and S. H. Strogatz, Physica (Amsterdam) 74, 197 (1994).
- [21] K. Wiesenfeld, P. Colet, and S. H. Strogatz, Phys. Rev. Lett. 76, 404 (1996).
- [22] L. M. Pecora and T. L. Carroll, Phys. Rev. Lett. 64, 821 (1990); H. G. Winful and L. Rahman, Phys. Rev. Lett. 65, 1575 (1990).
- [23] U. Rau, J. Peinke, J. Parisi, R. P. Huebener, and E. Schöll, Phys. Lett. A **124**, 335 (1987).