Reaction-Diffusion Systems Consisting of Single-Electron Oscillators

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This paper proposes a single-electron device that is analogous to the reaction-diffusion system, which is a chemical complex system producing various dynamic phenomena in the natural world. This electrical reaction-diffusion device consists of a two-dimensional array of single-electron nonlinear oscillators that are combined with one another through diffusive coupling. Computer simulation revealed that the device produces animated spatiotemporal patterns of node voltages, e.g., a rotating spiral pattern similar to that of a colony of cellular slime molds and a dividing-and-multiplying pattern that reminds us of cell division. A method of fabricating actual devices by using self-organized crystal growth technology is also described.

Keywords: single-electron device, unconventional electronics, reaction-diffusion

INTRODUCTION

A promising area of research in nanoelectronics is the development of electrical systems that imitate the dynamics of life. To proceed toward this goal, we propose a single-electron device that imitates the behavior of the reaction-diffusion system, which is a chemical complex system producing dynamic, self-organizing phenomena in the natural world. Constructing an electrical analog of reaction-diffusion systems would enables us to generate

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artificial biodynamics on a LSI chip and develop bioinspired information-processing systems.

A reaction-diffusion system (RD system) is a chemical system where chemical reactions and material diffusion coexist in a nonequilibrium state. It produces orderly spatiotemporal patterns of chemical concentration called *dissipative structures*. There are various RD systems in nature, and a variety of dynamic, self-organizing natural phenomena can be explained using the concept of a dissipative structure. Not a few biophysicists have the opinion that life itself is a dissipative structure produced by the natural world, a large RD system in itself.

In this paper, we propose constructing an electrical analog of RD systems, i.e., an electrical RD system consisting of single-electron circuits. The first of the following sections (Section 2) outlines RD systems. The next section (Section 3) proposes a method of constructing electrical RD systems. An RD system can be considered an aggregate of chemical nonlinear oscillators interacting with one another, so we can construct electrical RD systems by using electrical oscillators instead of chemical ones (Section 3.1). We used, as the electrical oscillator, a single-electron circuit that produces nonlinear oscillation caused by the Coulomb blockade phenomenon (Sections 3.2 and 3.3). The action of diffusion in RD systems can be imitated by capacitive coupling between the oscillators (Section 3.4). By arranging coupled oscillators into a network, we can construct a single-electron RD system (Section 3.5) and describes its operation (Section 4). We show through computer simulation that the system produces electrical dissipative structures, or animated spatiotemporal patterns of node potential in the circuit, which is a characteristic similar to that in chemical RD systems. Section 5 concludes with the design of actual RD systems and discusses their fabrication.

REACTION-DIFFUSION SYSTEMS AS THE STAGE OF BIODYNAMICS

Bioscience—the attempt to describe biological phenomena in terms of physics and chemistry—has developed considerably in recent years, and molecular biology, particularly with DNA-related fields, has become the mainstream in this area. That is, there has been an attempt to obtain a deeper understanding of biological phenomena by researching the components that make up living organisms. At the same time, this approach by itself cannot provide a complete picture of life, i.e., its animated, dynamical nature
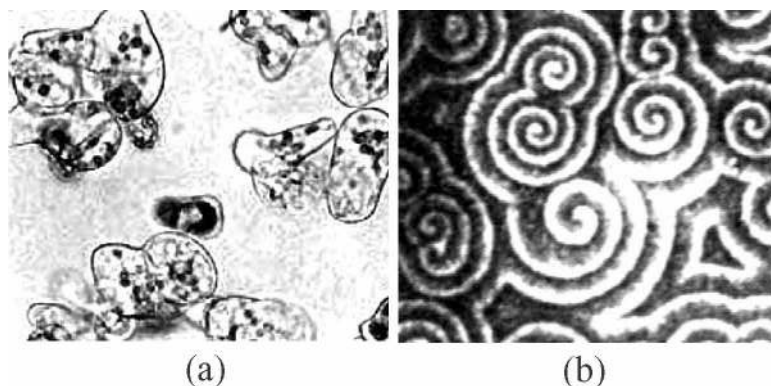
FIGURE 1
Examples of animated behavior in biological phenomena: (a) division and multiplication of asparagus cells [1], and (b) spiral pattern drawn when individual cells of cellular slime molds aggregate into a slug [2].

reflected by growth, differentiation, and morphogenesis. A "living thing" is different from a "thing" because it creates a temporal and spatial order on its own (Fig. 1). To understand this essence of life, another approach is needed and, because of this, nonlinear science on RD systems has come to be studied.

The RD system is a chemical complex system in a nonequilibrium, open state where chemical reactions and material diffusion coexist. In such a system, many elementary reactions proceed with the participation of various chemical substances, influencing one another through the synthesis and resolution of the substances. As a result, an RD system exhibits high-order nonlinear behavior and produces various dynamic phenomena unpredictable from an equilibrium state. A particular feature of the RD system is its generation of a dissipative structure. In other words, the concentrations of individual chemical substances in RD systems are not uniform either spatially or temporally but form an orderly spatiotemporal structure on a macroscopic level. This is called a dissipative structure. As system parameters change, varied dissipative structures appear as spatiotemporal patterns of chemical concentration. This behavior of RD systems can provide important clues to understanding animated, dynamical natural phenomena, in particular biodynamics. Because RD systems create orderly spatiotemporal patterns on their own, we can expect that they can be used to explain at least some aspects of dynamical nature of life. In the field of biophysics, there are not a few researchers with the idea that life is a dissipative structure arising in the RD system we call earth.

The behavior of RD systems, or the spatiotemporal patterns of chemical concentration, can be expressed by the reaction-diffusion equation, a partial differential equation with chemical concentrations as variables:

$$\frac{\partial \boldsymbol{u}}{\partial t} = \boldsymbol{f}(\boldsymbol{u}) + D\Delta \boldsymbol{u} \quad (\boldsymbol{u} = (u_1, u_2, u_3, ...)),$$

(1)

where t is time, $\boldsymbol{u}$ is the vector of chemical concentrations, u_i is the concentration of the ith substance, and D is the diagonal matrix of diffusion coefficients. Nonlinear function $\boldsymbol{f}(\boldsymbol{u})$ is the reaction term that represents the reaction kinetics of the system. Spatial derivative $D\Delta \boldsymbol{u}$ is the diffusion term that represents the change of $\boldsymbol{u}$ due to the diffusion of the substance. A greater number of variables results in more complex dynamics and a more complicated dissipative structure. A simple reaction-diffusion system with few variables, however, will still exhibit dynamics similar to biological activity. Figure 2 shows examples of two-variable systems. Note the similarity of these examples with those in Fig. 1 (see Note).

Note:

The similarity between Figs. 1(a) and 2(a) is not meant to imply that the model in Fig. 2(a) can directly explain the mechanism for cell division. The study of nonlinear science on RD systems is still at an early stage—it has not reached the point where it can explain the essence of life. Nevertheless, as an approach to representing and understanding biological phenomena, the nonlinear science of RD systems has proven to be quite effective.

CONSTRUCTING ELECTRICAL ANALOG OF
REACTION-DIFFUSION SYSTEMS

RD systems as an aggregate of coupled oscillators

An RD system can be considered an aggregate of coupled chemical oscillators, or a chemical cellular automaton, as described in Fig. 3. Each oscillator represents the local reaction of chemical substances and generates nonlinear dynamics $du/dt = f(u)$ that corresponds to reaction kinetics in Eq. (1). The oscillator interacts with its neighbors through nonlocal diffusion of substances; this corresponds to the diffusion term in Eq. (1) and produces dynamics $du/dt = D\Delta u$. Because of diffusion, all oscillators correlate with one another to generate synchronization and entrainment. Consequently, the
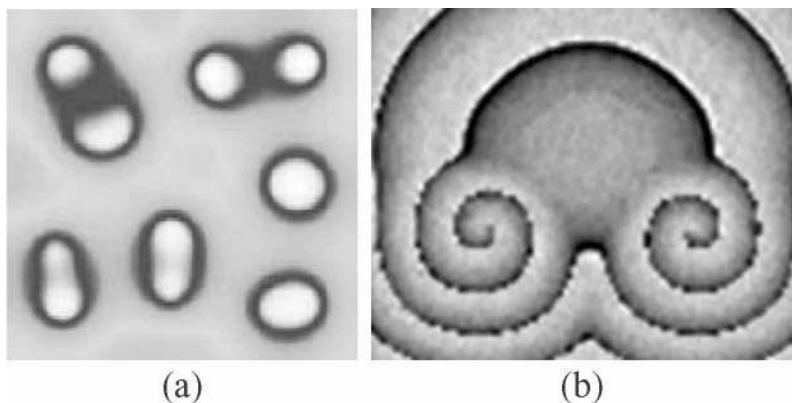
FIGURE 2
Examples of dynamics in reaction-diffusion systems; chemical concentrations are expressed by shading in two-dimensional systems. (a) Self-duplicating pattern by Gray-Scott reaction-diffusion system, and (b) spiral pattern by Brusselator reaction-diffusion system.
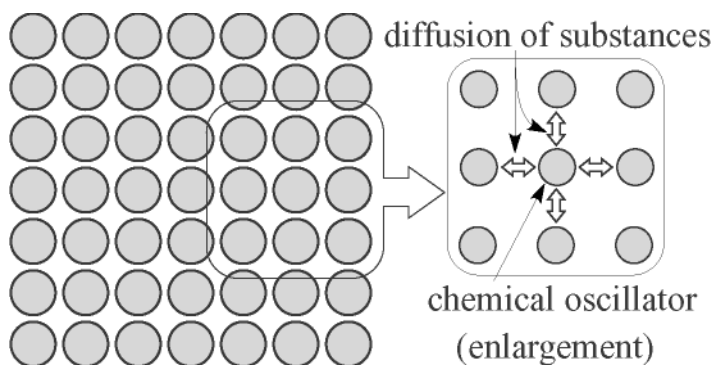

FIGURE 3
Simplified model of RD systems, consisting of many chemical oscillators. Each oscillator has variables corresponding to chemical concentrations $u_1, u_2, u_3, ...$ in Eq . (1) and interacts with its neighbors through diffusion of substances.

system as a whole produces orderly dissipative structures on a macroscopic level. The size of each oscillator, or the size of the local space in which chemical concentrations are roughly uniform, depends on the diffusion coefficients and reaction velocities in the system. It is several micrometers in diameter in many liquid RD systems; therefore, even a tiny RD system in a test tube contains millions of oscillators.
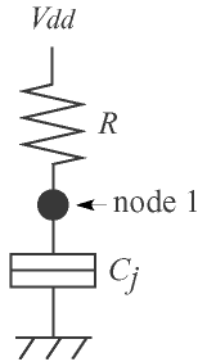
FIGURE 4
Single-electron oscillator (a SET cell) consisting of tunneling junction C_j, high resistance R connected at node 1, and positive bias voltage *Vdd*.

An electrical analog of RD systems can be created by using electrical oscillation circuits instead of chemical oscillators and coupling these circuits with one another in a way that imitates diffusion. Variables are the electrical potential of nodes in the oscillation circuits in this electrical RD system. The system will produce electrical dissipative structures, i.e., orderly spatiotemporal patterns of node potentials, under appropriate conditions.

Our purpose is to construct an electrical RD system in the form of an LSI chip. The key to building such a system is to integrate a large number of oscillation circuits on a chip with coupling subcircuits. A large arrangement of oscillators (e.g., 1000 x 1000 or more) is needed to generate complex, varied dissipative structures as observed in chemical RD systems. To achieve such large scale integration, we propose using single-electron circuits as the oscillators. A single-electron circuit can generate nonlinear oscillation through a simple circuit structure, so it can effectively be used in producing small oscillators for electrical RD systems.

Nonlinear oscillator using a single-electron circuit

We used a single-electron circuit (Fig. 4) as an electrical oscillator. It consists of tunneling junction C_j and high resistance R connected in series at node 1 and biased by positive voltage *Vdd*. This circuit is an elementary component of single-electron circuits known as the SET cell (see [3] for detailed explanation). A SET cell only has a single variable, voltage V_1 of node 1, but it can be oscillatory or excitatory in operation—which is
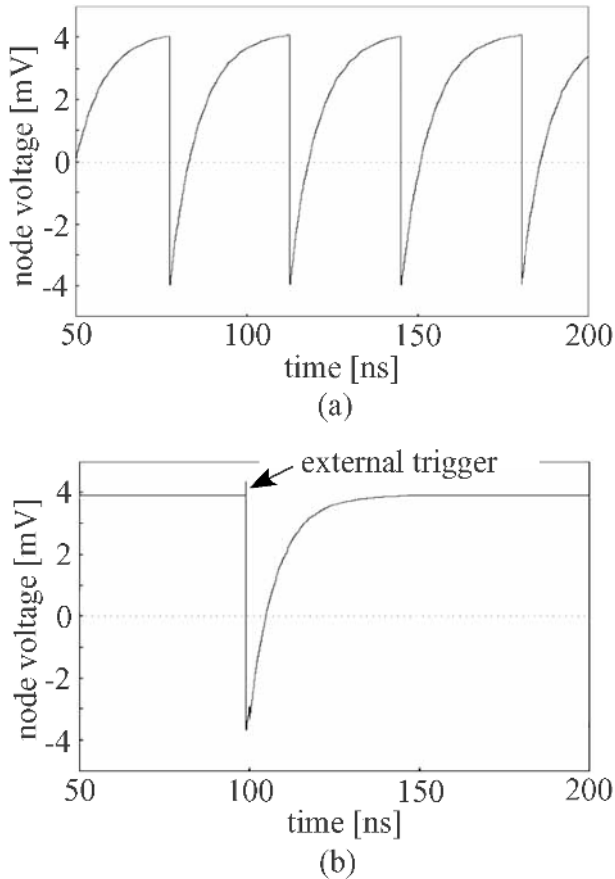
FIGURE 5
Operation of the oscillator. Waveforms of node voltage are shown for (a) self-induced oscillation and (b) monostable oscillation, simulated with following set of parameters: tunneling junction capacitance C_j = 20 aF, tunneling junction conductance = 1 μS, high resistance R = 400 MΩ, and zero temperature. Bias voltage Vdd = 4.2 mV for self-induced oscillation, and Vdd = 3.8 mV for monostable oscillation.

indispensable in creating RD systems—because the node voltage can produce a discontinuous change because of electron tunneling. (In continuous-variable systems such as chemical reaction systems, two or more variables are needed for oscillatory and excitatory operations.)

The SET cell operates as a nonlinear oscillator at the low temperatures at which the Coulomb-blockade effect occurs. It is oscillatory (astable) if Vdd > $e/(2C_j)$ (e is elementary charge) and produces nonlinear oscillation in

voltage at node 1 (Fig. 5(a)). The node voltage gradually increases as junction capacitance C_j is charged through resistance R, then drops discontinuously because of electron tunneling through the junction, again gradually increasing to repeat the same cycles. In contrast, the oscillator is excitatory (monostable) if $Vdd < e/(2C_j)$ and produces single-pulse operation excited by an external trigger (Fig. 5(b)). (In simulation, we used a modified Monte Carlo method. Kuwamura and his colleagues [4] have given details of this method. Also see Appendix in [5].) In constructing electrical RD systems, we will use oscillatory oscillators and excitatory ones, or both.

The oscillator exhibits discontinuous, probabilistic kinetics resulting from electron tunneling. The kinetics is given in the form of

$$\frac{dV_1}{dt} = \frac{Vdd - V_1}{RC_j} - \frac{e}{C_j} \cdot \delta(V_1 - \frac{e}{2C_j} - \Delta V) ,$$

(2)

where δ–function $\delta(...)$ represents a discontinuous change in node voltage caused by electron tunneling. Probabilistic operation arises from the stochastic nature of tunneling; i.e., a time lag (a waiting time) exists between when junction voltage exceeds tunneling threshold $e/(2C_j)$ and when tunneling actually occurs. This effect is represented by delay term ΔV in the equation. Because the value of ΔV has probabilistic fluctuations in every tunneling event and cannot be expressed in analytical form, so we have to use Monte Carlo simulation to study the behavior of the oscillator.

Single-electron oscillator with a multiple tunneling junction

In fabricating actual oscillators, a high resistance of hundreds of megaohms or more is not easy to implement on an LSI chip. A better way is to use a multiple tunneling junction, i.e., a series of many tunneling junctions, instead of high resistance (Fig. 6(a)). This structure also enables oscillatory and excitatory operations to be obtained because sequential electron tunneling through a multiple tunneling junction has a similar effect to current flowing at high resistance (Fig. 6(b)). In the following sections, however, we will use the high-resistance SET cell (Fig. 4) to construct electrical RD systems because less computing time is required in simulating RD operation. We can expect that the knowledge obtained from high-resistance RD systems will be able to be applied to RD systems consisting of multiple-junction oscillators.
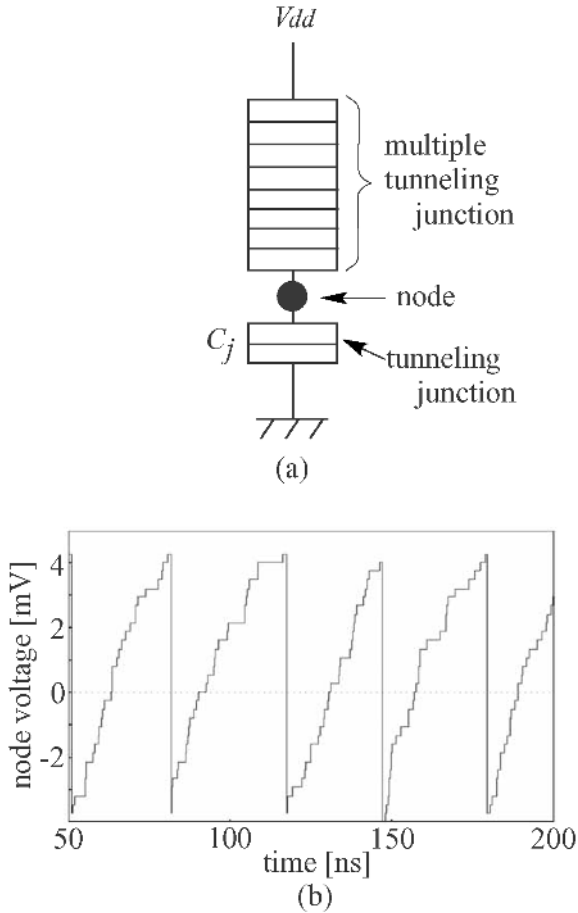
(a)

(b)

FIGURE 6
Single-electron oscillator with a multiple tunneling junction: (a) circuit configuration, and (b) simulated self-induced oscillation. Parameters are: capacitance of single tunneling junction $C_j = 10$ aF, conductance of the single tunneling junction = 1 μS, 30 tunneling junctions in the multiple tunneling junction, capacitance and conductance of a tunneling junction in the multiple tunneling junction are 300 aF and 50 nS, bias voltage $Vdd = 8.6$ mV, and zero temperature.

Diffusive coupling of oscillators

To construct RD systems, oscillators have to be connected with one another so that they will interact through "diffusive" coupling to generate synchronization and entrainment. To do this, we propose connecting the oscillators by means of intermediary oscillation cells and coupling capacitors.

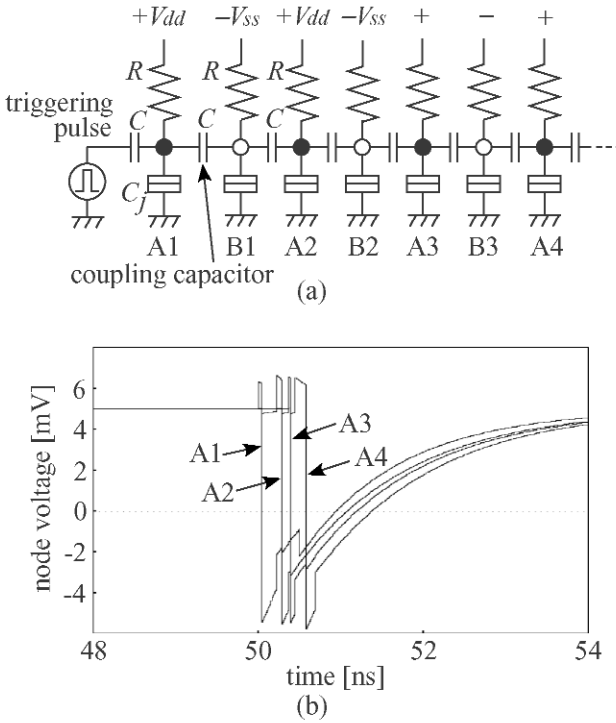Figure 7(a) illustrates the method of connection with a one-dimensional

FIGURE 7

Diffusive connection of oscillators. (a) One-dimensional chain of oscillators (A1, A2, ...) with intermediary cells (B1, B2, ...) and coupling capacitors C. For study of the transmission of tunneling, a triggering pulse generator is connected to the left end. (b) Transmission of tunneling through the chain of excitatory oscillators. The waveform of node voltage is plotted for each oscillator. A triggering pulse was applied to the leftmost oscillator, and tunneling started at the oscillator to transmit along the chain with delay. Jumps in curves A1-A4 result from electron tunneling in oscillators A1-A4. Simulated with a set of parameters: $C_j = 10$ aF, $C = 2$ aF, $R = 77$ MΩ, tunneling junction conductance $= 5$ μS, $Vdd = 5$ mV, $-Vss = -5$ mV, and zero temperature.

chain of oscillators. The oscillators (SET cells denoted by A1, A2, ..., with their nodes represented by closed circles) are connected with their neighboring oscillators through intermediary oscillation cells (SET cells denoted by B1, B2, ..., with their nodes represented by open circles) and coupling capacitors C. We use an *excitatory* SET cell biased with a *negative* voltage $-Vss$ as the intermediary oscillation cell.

When electron tunneling occurs in an oscillator in this structure, the node voltage of the oscillator changes from positive to negative, and this induces, through coupling capacitor C, electron tunneling in an adjacent intermediary

cell. The induced tunneling changes the node voltage of the intermediary cell from negative to positive, and this induces electron tunneling in an adjacent oscillator. In this way, electron tunneling is transmitted from one oscillator to another along the oscillator chain. There is a time lag between two tunneling events in two neighboring oscillators as if these oscillators interacted through diffusion. This phenomenon is not diffusion itself and cannot be expressed in the form $D\Delta u$ in Eq. (1) but can be used as a substitute for diffusion.

The transmission of tunneling with delay is illustrated in Fig. 7(b) with simulated results for a chain of excitatory oscillators with intermediary cells. Electron tunneling was induced in the leftmost oscillator by a triggering pulse, and it was transmitted to the right along the chain with delay. In other words, an excitation wave of tunneling traveled to the right along the chain. Its delay in traveling from one oscillator to a neighbor has probabilistic fluctuations because of the stochastic nature of tunneling, but this is not a problem for applications to RD systems.

Electrical RD system with single-electron oscillators

An electrical RD system can be constructed by connecting oscillators into a network by means of intermediary cells and coupling capacitors (Fig. 8). Each oscillator is connected to its neighboring 4 oscillators by means of 4 intermediary cells and coupling capacitors. This is a two-dimensional RD system, and in the next section, we will study its behavior through computer simulation. A three-dimensional RD system can also be constructed in a similar way by arranging oscillators into a cubic structure and connecting each oscillator with its 6 neighboring oscillators by means of 6 intermediary cells and coupling capacitors. However, we will not take this up here because three-dimensional systems take an enormous amount of computing time to simulate their operation.

Spatiotemporal dynamics produced by the single-electron RD system

In the RD system we proposed, the node voltage of each oscillator changes temporally as the oscillators operate through mutual interactions. Consequently, a two-dimensional spatiotemporal pattern of the node voltages is produced on the RD system. Since this voltage pattern corresponds to the dissipative structure in chemical RD systems, we call it an electrical dissipative structure.

A variety of electrical dissipative structures are produced from different sets of system parameters. To understand the behavior of an electrical RD
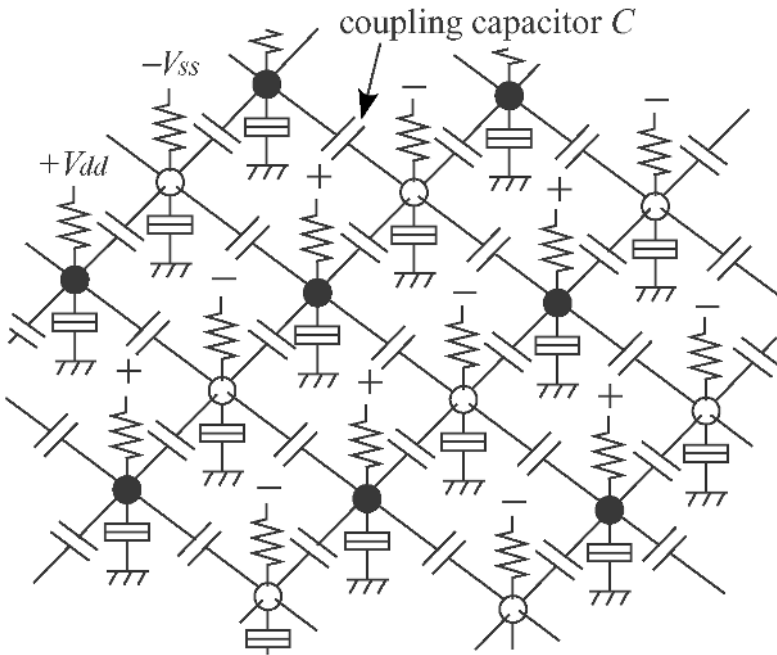
FIGURE 8
Two-dimensional RD system consisting of the network of single-electron oscillators. Each oscillator (closed-circle node) is connected with 4 neighboring oscillators by means of 4 intermediary cells (open-circle nodes) and coupling capacitors.

system entirely, we need to draw a phase diagram for the system, i.e., a diagram that depicts—in the multidimensional space of system parameters—what kind of dissipative structure will appear for each set of parameter values. However, we cannot draw a phase diagram for our RD system without a long numerical computer simulation because its reaction-diffusion kinetics cannot be expressed in analytical form. We are currently carrying out computer simulations on the phase diagram but have yet to complete it. Instead, we will show a few examples of electrical dissipative structures simulated with a few sample sets of parameter values.

Although a single-electron RD system differs greatly from chemical RD systems in terms of reaction-diffusion kinetics, it can produce dissipative structures similar to those of chemical RD systems. Here, we will discuss three examples, i.e., an expanding circular pattern, a rotating spiral pattern similar to Fig. 2(b), and a dividing-and-multiplying pattern with some
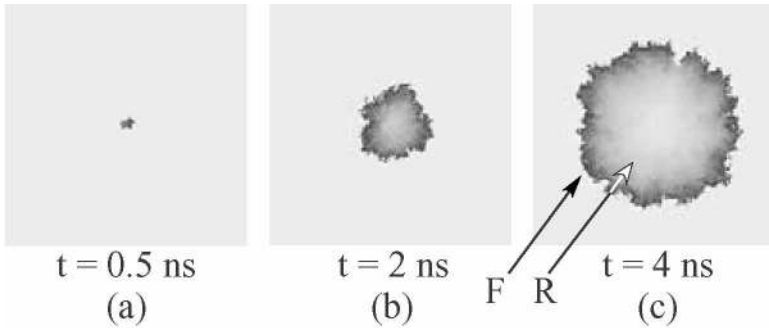
FIGURE 9
Expanding circular pattern in the single-electron RD system. Snapshots for three time steps. Simulated with parameters: tunneling junction capacitance C_j = 1 aF, tunneling junction conductance = 1 μS, high resistance R = 137.5 MΩ, coupling capacitance C = 1 aF, bias voltage Vdd = 16.5 mV, $-Vss = -16.5$ mV, and zero temperature

resemblance to Fig. 2(a). The following will have the results simulated for a RD system consisting of 201 x 201 excitatory oscillators and 200 x 200 intermediary cells.

(a) Expanding circular pattern

A single-electron RD system consisting of excitatory oscillators is in a stable uniform state as it stands. Once a triggering signal is applied to an oscillator in the system, an excitation wave of tunneling starts at the oscillator and propagates in all directions to form an expanding circular pattern. This can be seen in Fig. 9; the node voltage of each oscillator is represented by a gray scale: the light shading means high voltage, and the dark means low voltage. The front F of the wave is the region where tunneling just occurred and, therefore, the node voltage of the oscillators is at the lowest negative value. The front line is uneven or irregular because the velocity of the traveling wave fluctuated in each direction throughout the process because of the stochastic waiting time of tunneling.

After the excitation wave passed through, the node voltage of each oscillator gradually increased to return to its initial value, the positive bias voltage. This is indicated in the figure by the light shading on the rear R of the wave. If a triggering signal is applied repeatedly to one oscillator, a concentric circular wave—called a *target pattern* in chemical RD systems—will be generated.
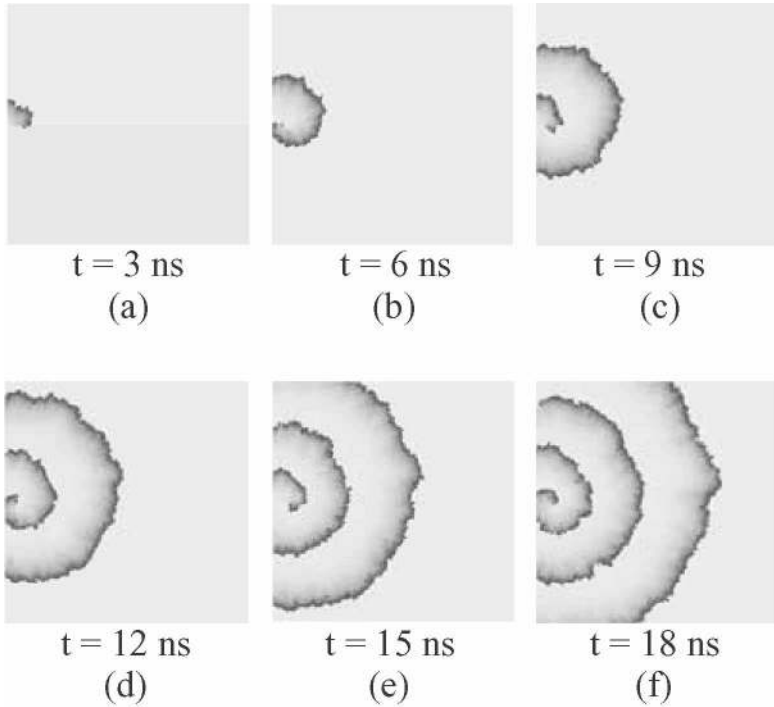
FIGURE 10
Rotating spiral pattern. Snapshots for six time steps. Simulated with the same parameters as used for Fig. 9.

(b) Rotating spiral pattern

This pattern appears when an expanding circular wave is chipped by external disturbance, thereby making an endpoint to appear in the wave front. With this endpoint acting as a center, the wave begins to curl itself to form a rotating spiral pattern (Fig. 10). The principle of curling is similar to that in chemical RD systems.

In this example, a triggering signal was applied to the middle oscillator on the left of the RD system. When an excitation wave started and expanded a little, the lower half of the wave was chipped by resetting the node voltage of oscillators to zero (Fig. 10(a)). After that, the RD system was left to operate freely, and a rotating spiral pattern of node voltages automatically generated as can be seen in Figs. 10(b)-10(f).
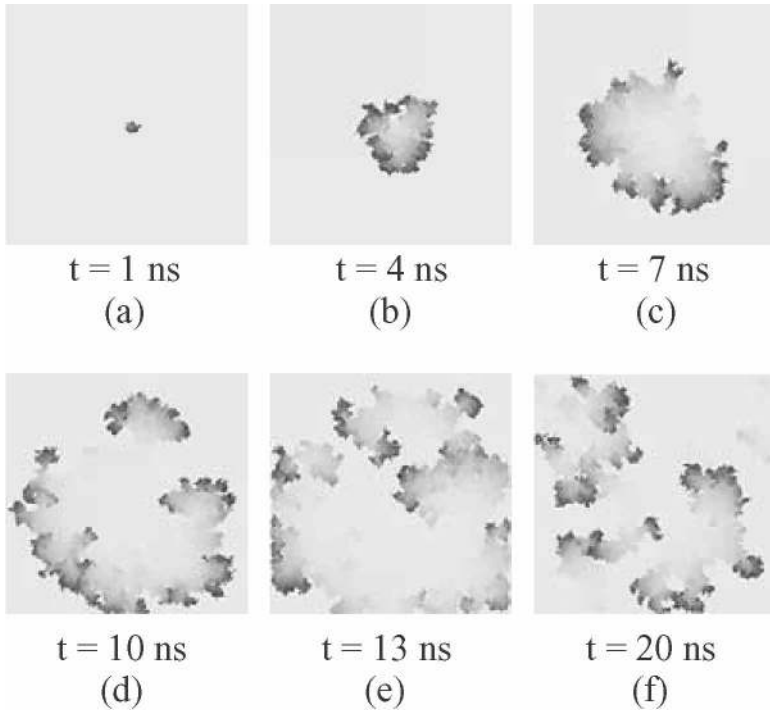
FIGURE 11
Dividing-and-multiplying pattern. Snapshots for six time steps. Simulated with the same parameters as used for Fig. 9 except that $R = 150.5$ MΩ, $Vdd = 15.8$ mV, and $-Vss = -15.8$ mV.

(c) Dividing-and-multiplying pattern

This pattern appears when the coupling between oscillators is weak (i.e., small coupling capacitance or low bias voltage). When this happens, electron tunneling in an oscillator cannot be transmitted to all four adjacent intermediary cells; e.g., tunneling can be transmitted to the right and left cells but not to the upper and lower cells. As a result, an expanding node-voltage pattern splits into pieces, and each piece again expands to split again. This produces dividing-and-multiplying patterns (Fig. 11). The principle of division is different from that in chemical RD systems, but the behavior of created patterns is somewhat similar. In a way, we may consider that there are electrical microbes consisting of negative voltages living on the RD system, eating positive charges on nodes as food, and propagating so that they will spread all over the system.

multiple tunneling junction

$-V_{SS}$

$+V_{dd}$

bias electrodes

insulating layers

nanodots

tunneling junction

arms

substrate (conductive)

(a)

500 nm

20.0kV X45.0K 662nm

(b)

arm (n-GaAs)

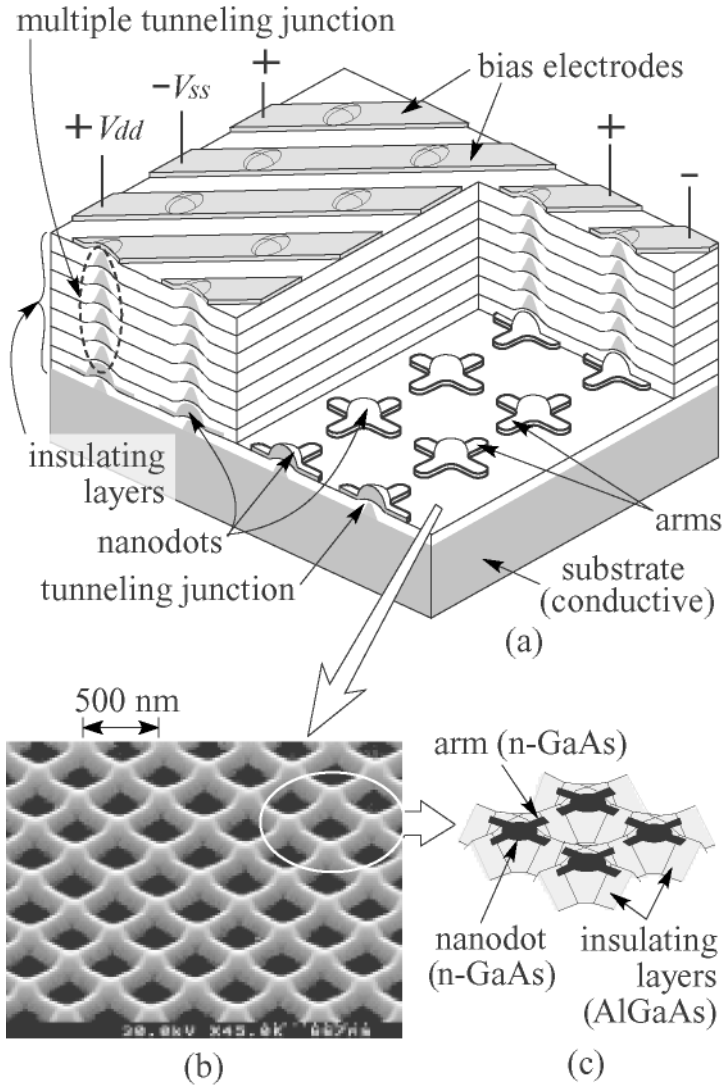nanodot (n-GaAs)

insulating layers (AlGaAs)

(c)

FIGURE 12
Device structure for the single-electron RD system. (a) Three-dimensional and cross-sectional schematics; (b) SEM photograph of a two-dimensional array of GaAs nanodots with coupling arms and tunneling junctions; (c) schematic diagram of the nanodots with coupling arms.

TOWARD ACTUAL RD DEVICES

The unit element in our RD system is a single-electron oscillator coupled with four neighbors. The multiple-tunneling-junction oscillator (Fig. 6) is preferable for this element because it can be made without high resistance, which is difficult to implement on an LSI chip. Arranging such oscillators into a two-dimensional array produces an RD system, so our next task is to fabricate many identical oscillators on a substrate. Figures 12(a) shows the three-dimensional and cross-sectional schematics for the structure of the device. Each oscillator consists of a conductive nanodot (minute dot) with four coupling arms, and there is a tunneling junction between the nanodot and the conductive substrate beneath it. Many series-connected junctions run between the nanodot and a positive-bias or a negative-bias electrode. Capacitive coupling between neighboring oscillators can be achieved by laying their coupling arms close to each other.

The key in this construction is to prepare a large arrangement of nanodots with coupling arms and tunneling junctions. We previously proposed and demonstrated a process technology that could be used to fabricate the RD-system structure [6]. This technology uses self-organized crystal growth achieved by selective-area metalorganic vapor-phase epitaxy (SA-MOVPE), and it can be used to fabricate GaAs nanodots with arms and tunneling junctions on a GaAs substrate by making use of the dependence of the crystal-growth rate on crystal orientation (for detailed explanation, see Refs. [7] and [8]). With this technology, a nanodot with four coupling arms can be formed automatically in a self-organizing manner. This technology can also be used to automatically create the structure for multiple tunneling junctions on nanodots simply by repeating the growth of an n-type GaAs layer and an insulating AlGaAs layer. Using such a process, we succeeded in forming GaAs nanodots with their arms and tunneling junctions beneath them in the form of a two-dimensional array on a substrate (Figs. 12(b) and 12(c)), though our technology is not yet perfect and we have yet to fabricate a complete device. We are now developing an improved process technology to form GaAs nanodots with arms and multiple tunneling junctions, arranged regularly with a smaller pitch of 100 nm or less (corresponding to 10^{10} oscillators/cm^2). With the improved process technology, we will be able to integrate coupled single-electron oscillators on a chip and proceed from there to develop reaction-diffusion LSIs.

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