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Global synchronization in two-dimensional lattices of discrete Belousov–Zhabotinsky oscillators

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Abstract

Macroscopic synchronization was investigated experimentally in the Belousov–Zhabotinsky reaction with a two-dimensional lattice arrangement of coupled chemical oscillators. The natural frequency of each oscillator was distributed randomly and the spacing distance d between beads was varied, where the coupling strength between two oscillators was controlled by changing d. At small d beyond a certain critical value, the oscillators synchronized spontaneously with each other. Global synchronization in the lattice was described as a function of d. The critical coupling strength for global synchronization depends on the total number of oscillators. When noisy light was applied uniformly to whole oscillators, the order of synchronization was maximized at optimal noise intensity: noise synchronization was observed.

Keywords: Coupled oscillators; Entrainment; Noise synchronization; Belousov–Zhabotinsky reaction

1. Introduction

The phenomenon of synchronization in a large number of coupled nonlinear oscillators has been studied in physical [1–12], chemical [13,14], and biological systems [15,16]. Populations of coupled oscillators may synchronize spontaneously to a common frequency in spite of differences in their natural frequencies. The onset of macroscopic synchronization in populations has been investigated using a simple model based on a phase description of individual oscillators [2,3]. Theoretical and numerical results predict a transition, at a certain coupling constant, from an individual oscillation to a coherent one. In particular, the transition to global synchronization in a spatially distributed oscillator system (SDOS) has attracted wide attention [4–

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10] because the formation and mergence of synchronization clusters with interesting dynamics can be observed.

Experimental studies of spatially distributed and coupled oscillator systems have been performed extensively using the Belousov–Zhabotinsky (BZ) reaction. For example, spatiotemporal patterns, e.g., target and spiral patterns, were observed in a spatially continuous BZ system [17–19]. Their dynamic features have already been explained well [2]. The BZ reaction in SDOS has also been studied using cation-exchange beads, which absorbed the catalyst ions [20–26]. When the beads are immersed in the catalyst-free BZ solution with proper concentrations, the BZ reaction occurs only on the surface of individual beads. Thereupon, individual beads become localized oscillators that exhibit a stable limit-cycle oscillation. In general, the SDOS may have four different characteristic scales: the distance between the nearest-neighbor oscillators, the coupling length via diffusion, the characteristic length of the generated pattern field, and the system size [27]. In the bead oscillator system, there is one more characteristic scale: the radius of the oscillator. Depending on relative magnitudes among these scales, local, global and non-local couplings are possible. Therefore, the bead oscillator system may also show a variety of dynamics such as global and non-local coupling systems, as suggested by Kuramoto [28–30]. By careful choices of the concentration for chemicals and scales in the bead oscillator system, a non-locality of the coupling could be realized. Very recently, there are reports that noise enhances the synchronization in coupled bead oscillators at an optimal noise intensity [25,26]. Therefore, noise may also play an important role in influencing interactions and characteristic scales. However, that role has not been clarified experimentally yet in a large number of BZ oscillators.

This study investigated global synchronization in SDOS experimentally using cation-exchange beads absorbing catalyst ions and the catalyst-free BZ solution as the discrete chemical oscillators. They were distributed spatially in two-dimensional lattice arrangements with spacing d. Coupling among the oscillators via mass diffusion of chemicals can be varied by changing the spacing d. We used a photo-sensitive catalyst for the BZ reaction to control the reaction using light illumination. The present study addresses the onset of global synchronization with and without external noise in lattice oscillators as a function of d. We also perform numerical simulation using a modified Oregonator model.

2. Experimental setup

Chemicals and their concentrations were 0.83 M H_2SO_4, 0.28 M $KBrO_3$, 0.06 M KBr, and 0.11 M $CH_2(COOH)_2$ at room temperature of $24 \pm 1^{\circ}$C. To absorb the catalyst into cation-exchange beads (50 W × 4; Muromachi Technos Co. Ltd.), beads with diameter a (0.9 ± 0.01 mm) were immersed in a ruthenium(II)–bipyridyl complex ($Ru(bpy)_3^{2+}$) solution controlled to the desired concentration of 2.3×10^{-5} mol/g for more than 6 h with stirring. We prepared glass boards with regular holes (0.6 ± 0.01 mm diameter) in a lattice arrangement with various lattice constants l_0 ($l_0 - a = d$) to obtain a regular lattice arrangement of beads. A bead was fixed at each hole on the board, which was then immersed into the BZ solution (20 ml) in a Petri dish. The difference of the intrinsic period of oscillation for each bead was mainly attributable to the concentration fluctuation in beads absorbing the catalyst. Oscillation was observed using a CCD camera, images of which were recorded for later analysis on a videotape and the hard disk of a computer. The time sequence of light intensity from each oscillator was obtained by averaging the gray level of the central region on the bead surface, corresponding to a grid of 8×8 pixels. A halogen lamp with intensity that was controlled by a computer was prepared to apply photic noises. Noisy light was illuminated though a 460 nm band-pass filter.

The period distribution of isolated oscillators (total: 223) was described by a unimodal distribution with an average period $T_0 = 256$ s and a standard deviation $\sigma = 7.5$ s. Analyses for each oscillator were performed from 5000 to 20 000 s after initiation of the reaction to discard the transient process to a steady state oscillation. Temporal variation of the amplitude and period of each oscillator over time were less than 10% and 5%, respectively, during the time interval.

Computer simulation was performed using the fourth-order Runge–Kutta method with the time step $\Delta t = 0.01$ s. Statistical quantities were calculated from a run of 2×10^6 time steps after discarding the initial 5×10^5 time steps as transient.