Oscillation and Synchronization in the Combustion of Candles

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We investigate a simple experimental system using candles; stable combustion is seen when a single candle burns, while oscillatory combustion is seen when three candles burn together. If we consider a set of three candles as a component oscillator, two oscillators, that is, two sets of three candles, can couple with each other, resulting in both in-phase and antiphase synchronization depending on the distance between the two sets. The mathematical model indicates that the oscillatory combustion in a set of three candles is induced by a lack of oxygen around the burning point. Furthermore, we suggest that thermal radiation may be an essential factor of the synchronization.

1. Introduction

In nature, our eyes are attracted to phenomena that show oscillation due to their broken temporal symmetry yet with high degree of regularity. Some of them even show coherent behavior such as synchronized oscillation and/or propagation of waves. It has been known that a nonlinear differential equation is necessary to understand these behaviors, and such a research field is classified as nonlinear science. Limit-cycle oscillation and synchronization are two of the most important concepts in nonlinear science, and they have been extensively studied both theoretically and experimentally.[1−10] Recently, as one of the simple systems of a nonlinear oscillator, the oscillatory combustion of a set of candles was reported,[11] although the mechanism has not yet been clarified. In this study, we experimentally analyzed the characteristics of the oscillation of candle flames. Different from the known combustion systems that were reported to show wave propagation,[12,13] it was found that the two oscillators composed of a set of candles can couple with each other in a synchronous manner in the present system. Both in- and antiphase synchronization can be observed depending on the distance between the two oscillators. Previously, studies on the behavior of combustion have been widely performed from the viewpoint of engineering, such as for the synthesis of chemical compounds or for the design of engines for rockets and airplanes.[14,15] These studies mainly sought to control combustion to avoid explosions or instability. Thus, these studies were confined to linear stability analyses or detailed numerical simulations. In this article, in contrast, we describe the mechanism of characteristic phenomena such as oscillation and synchronization in the combustion of candles.

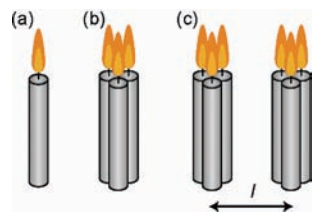

Figure 1. Schematic illustration of the system. (a) A single candle burns constantly. (b) Three candles burn in an oscillatory manner. (c) Two sets of three candles exhibit in- or antisynchronization depending on the distance between them, *l*.

2. Experiments and Results: Oscillation and Synchronization

All experiments were performed at room temperature under the condition without external air flow. We used a cylindrical candle made of paraffin (Cando Ltd., Japan) with a diameter and height of 6.0 and 50 mm, respectively. Images were recorded under a dark background using a high-speed video camera (Silicon Video 9M001, Epix, U.S.A.) at 250 fps. A schematic representation of the experimental setup is shown in Figure 1. The experimental features are represented by the brightness, which was defined by integrating the brightness in an adequate area on every frame of the movie.

When a burning candle was placed alone, the shape of the flame was stable, as shown in Figure 2a. In contrast, when three candles were placed close together, the shape of the flames exhibited oscillation, as shown in Figure 2b. The frequency of

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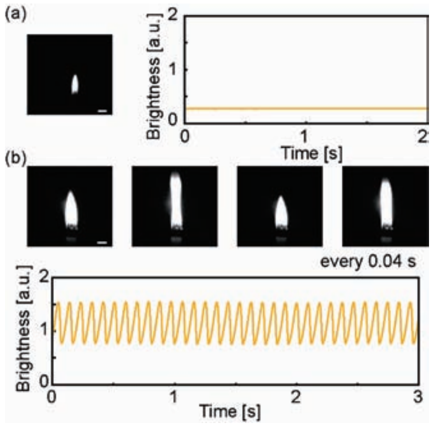

Figure 2. Experimental results on the combustion of a single candle (a) and a set of three candles (b). Snapshots every 0.04 s and the time course of light intensity obtained from images taken with a high-speed camera at 200 fps. Scale bars are 10 mm.

oscillation did not change within the experimental observations, whereas the height of the candle was slightly decreased due to the consumption of paraffin by combustion. When oscillation was disturbed by external factors such as air flow, oscillation was modulated or stopped for a short time but restarted after the disturbance was removed. Thus, the combustion of these three candles can be considered to be a self-sustaining oscillation, that is, a limit-cycle oscillation.

As an extension of our study on a single oscillator (a set of three candles), we observed the nature of the coupling of two oscillators, that is, two sets of three candles. When two sets of three candles were placed at a certain distance l, the flames exhibited synchronization depending on l, as shown in Figure 3. When the distance was smaller, that is, $l \leq 30$ mm, the two oscillators exhibited in-phase synchronization, as shown in Figure 3a. The figure shows snapshots of the flames and the time courses of the brightness of the two flames. When the distance was larger, that is, $30 \leq l \leq 48$ mm, the two oscillators exhibited antiphase synchronization, as shown in Figure 3b. When the distance was even larger, that is, $l \geq 46$ mm, oscillations proceeded independently. In the intermediate regions around $l \sim 30$ and 47 mm, one of the two modes was selected, and the system sometimes fluctuated between these modes. The relationship between the mode of synchronization and the distance of the two oscillators is shown in Figure 3c. The frequencies of oscillation that showed antiphase synchronization were greater than those that showed in-phase synchronization.

3. Experiments and Results: Observation of Flow Profile

In order to make clear the mechanism of synchronization, we measured the flow profile of air around candle oscillators using a Mach−Zehnder interferometer. The optical system is shown schematically in Figure 4a. In this system, differences in the refractive index can be measured through interference with a laser beam. Thus, in the measurement with a Mach−Zehnder interferometer, we can detect isothermal lines.

Figure 4b-1 and b-2 shows the results for a single candle and a set of three candles. For one candle, the isothermal line did not move very much, which reflects the constant burning. In contrast, for a set of three candles, the isothermal lines exhibited oscillation synchronously with the oscillation of the shape of the flames. Thus, we can guess that the air flow around the candles changes with the shape of the flames.

Figure 4b-3 and b-4 shows the isothermal lines for two sets of candle oscillators, that is, two sets of three candles. The

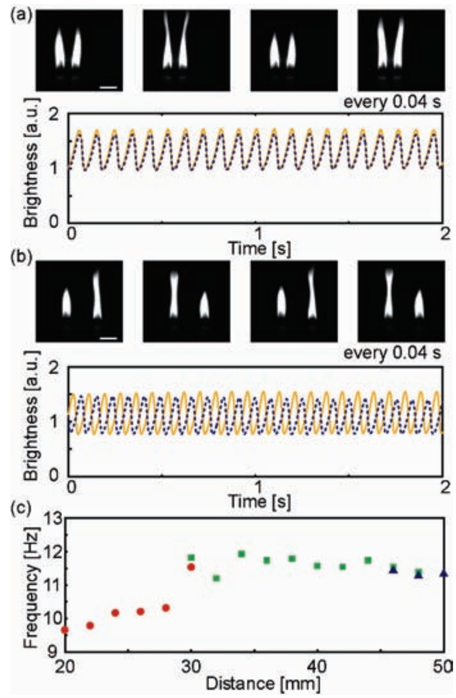

Figure 3. Experimental results on the synchronization of two sets of three candles. (a,b) Snapshots every 0.04 s and the time course of the light intensity when the distance between the two sets is (a) 20 and (b) 40 mm. Scale bars are 20 mm. (c) Phase diagram of the mode of synchronization and the frequency versus the distance between the two sets of three candles. The frequency was achieved by averaging five experiments. The red circle, green square, and blue triangle correspond to in-phase synchronization, antiphase synchronization, and desynchronization, respectively.

distance between the two oscillators was 20 and 40 mm, which correspond to in- and antiphase synchronization, respectively. For in-phase synchronization, (Figure 4b-3) the profile of the isothermal line was symmetric with regard to the center of the two oscillators, whereas the profile was asymmetric for antiphase synchronization (Figure 4b-4). Noteworthy enough, the isothermal lines in the central part did not move very much in both cases. From these measurement, we can confirm that the air flow around the candle is laminar and that the lateral air flow does not exist even when synchronous oscillation of the flames is exhibited.

4. Mathematical Modeling

Here, we discuss the mechanism of the coupling between the two oscillators. The changes in temperature should interact through a process of heat transfer. Generally, there are three possible forms of heat transfer between two oscillators, thermal diffusion, convection, and radiation. Historically thermal diffusion and convection were thought to be the essence of instability in combustion systems. However, the radiation effect seems to be the key for synchronization of candle flames by the following consideration of order estimation. First, we consider whether diffusion can play an essential role in synchronization. The typical distance between two oscillators is ∼10 mm, and the diffusion constant of a normal gas is around 10 mm^2 s^{-1}; thus, the typical time scale for thermal diffusion for a distance of 10 mm is estimated to be10 s. Since the typical time scale of the present oscillation is ∼0.1 s, thermal diffusion cannot play an essential role. As for convection, the results in Figure 4 suggest that interaction through air flow does not seem to cause synchronization. It is likely that convection serves only

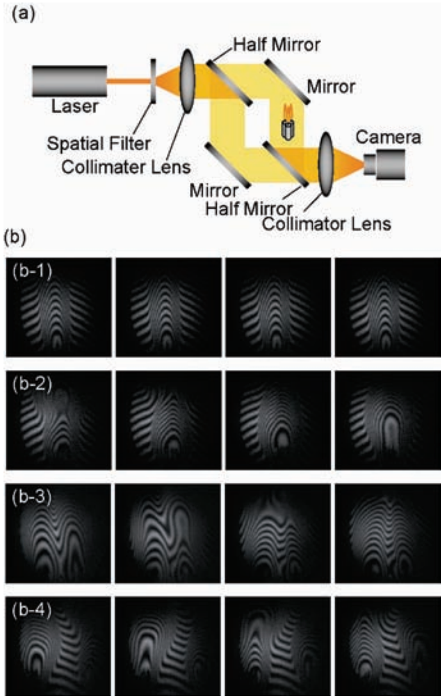

Figure 4. (a) Schematic representation of the Mach–Zehnder interferometer to measure the flow of air around candle oscillators. (b) Air flow profile measured by the Mach–Zehnder interferometer. Flow profiles around (b-1) a single candle and (b-2) a set of three candles are shown. (b-3) When the two oscillators exhibit in-phase synchronization. The distance between the two oscillators is 20 mm. (b-4) When the two oscillators exhibit antiphase synchronization. The distance between the two oscillators is 40 mm. The snapshots were taken every 0.025 s in (b-1)–(b-4).

to decrease the temperature by taking heat away from the burning area. Regarding radiation, the temporal change in the amplitude of radiation and that of the brightness of the flames were measured simultaneously in experiments when the three candles showed oscillatory burning (data not shown). The results show that radiation coupling can be a factor of interaction and can induce in- and antiphase synchronization between two sets of three candles. Thus, the consideration of radiation coupling is necessary to understand the candle flame oscillation.

Adopting the coupling through radiation, we discuss the mechanisms of oscillation and synchronization through a mathematical model. Before we discuss the synchronization through the radiation, we construct a model for a set of three candles, that is, a single oscillator. Experimentally, a set of three candles can exhibit oscillatory combustion, while a single candle exhibits stable combustion. In our model, the difference in the number of candles is regarded as a difference in the supply rate of the paraffin, that is, fuel. Here, the chemical reaction and temperature change in the flames are taken into consideration. We assume that (i) the paraffin in a candle becomes gaseous and reacts with oxygen in the air phase, (ii) paraffin is present in a sufficient amount and the gas of the paraffin is supplied at a constant rate, and (iii) oxygen is supplied from the bulk air far from the candle and heat is taken away by convection but not by diffusion. On the basis of these assumptions, we construct the following equations

$$C\frac{\mathrm{d}T}{\mathrm{d}t} = \omega_1\left[h(T_0 - T) + \beta a n \exp\left(-\frac{E}{RT}\right)\right] - \sigma T^4 \quad (1)$$

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \omega_2\left[k(n_0 - n) - a n \exp\left(-\frac{E}{RT}\right)\right] \quad (2)$$

where $T(t)$ is the temperature of the flame and $n(t)$ is the concentration of oxygen in the air. C, R, E, T_0, n_0, h, k, β, and σ are positive constants that correspond to the specific heat, gas constant, active energy, external temperature, external oxygen concentration, heat flow by convection, supply rate of oxygen by convection, heat production by combustion per unit volume of paraffin, and the Stefan–Boltzmann constant, respectively, and ω_1 and ω_2 are characteristic time constants for the changes in temperature and in the oxygen concentration. If we suppose that the temperature increases rapidly by a slight reaction, we obtain $0 < \omega_2 \ll \omega_1$. The first, second, and third terms on the right side in eq 1 mean the transport of heat by flow, heat production by combustion, and energy loss by radiation, respectively. It is noted that, by the Stefan–Boltzmann law, the total energy radiation from a blackbody is proportional to the fourth power of temperature.[16] The first and second terms on the right side of eq 2 mean the transport of oxygen by the flow and consumption of the oxygen by combustion. The important parameter here is a, which corresponds to the supply rate of the fuel.

For numerical calculation, nondimensionalization of eqs 1 and 2 was made by introducing the following variables

$$u = c\frac{T - T_0}{T_0} \qquad v = \frac{n}{n_0} \qquad \tau = \omega_2 k t$$

$$c = \frac{E}{RT_0} \qquad \varepsilon = \frac{Ck\omega_2}{h\omega_1} \qquad a_{\mathrm{u}} = \frac{\beta c n_0 a}{T_0 h}\exp(-c)$$

$$a_{\mathrm{v}} = \frac{a}{k}\exp(-c) \qquad \sigma_0 = \frac{\sigma c T_0^{\,3}}{C\omega_2 k}$$

Then, the following nondimensionalized equations were obtained

$$\frac{\mathrm{d}u}{\mathrm{d}t} = \frac{1}{\varepsilon}\left[-u + a_{\mathrm{u}} v \exp\left(\frac{u}{1 + u/c}\right)\right] - \sigma_0\left(1 + \frac{u}{c}\right)^4 \quad (3)$$

$$\frac{\mathrm{d}v}{\mathrm{d}t} = 1 - v - a_{\mathrm{v}} v \exp\left(\frac{u}{1 + u/c}\right) \quad (4)$$

The numerical results with the nondimensionalized equations derived from eqs 3 and 4 are shown in Figure 5a and b. When the parameter a is small, which corresponds to single-candle burning, the temperature $T(t)$ does not change but rather maintains a constant value, as in Figure 5a. When a is larger, which corresponds to three-candle burning, the time series of the temperature $T(t)$ exhibits oscillatory behavior, as in Figure 5b.

On the basis of this mathematical model for one oscillator, we consider coupling between two oscillators. Here, the temperature and oxygen concentration for oscillator 1 are $T_1(t)$ and $n_1(t)$, and those for oscillator 2 are $T_2(t)$ and $n_2(t)$. We introduce the following coupling terms and perform numerical calculations

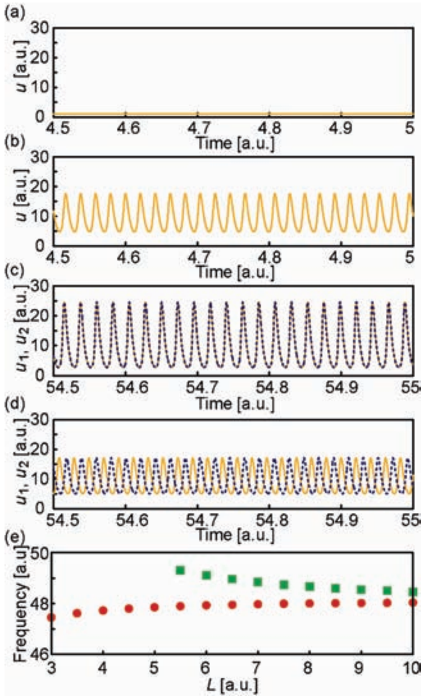

Figure 5. Numerical results. (a) For one candle. When a is smaller, the time series of temperature maintains a constant value, which reflects constant combustion. Here, $a = 0.5$. (b) For three candles. When a is larger, the time series exhibits oscillatory behavior. Here, $a = 37$. In (a) and (b), $\alpha_u/\alpha = \alpha_v/\alpha = 0.1$. (c) For in-phase synchronization of two sets of three candles. When the distance between the two oscillators L is smaller, the two oscillators exhibit in-phase synchronization. Here, $\mu/L^2 = 0.5$. (d) For antiphase synchronization. When L is larger, they exhibit antiphase synchronization. Here, $\mu/L^2 = 0.01$. (e) Frequency dependency on L, the distance between two oscillators, which corresponds to Figure 3c. We set $\mu = 1$. The red circle and green square correspond to in-phase synchronization and antiphase synchronization, respectively. It is noted that the range of initial conditions for in-phase synchronization is much smaller than that for antiphase synchronization when $L \geq 5.5$. The numerical calculation was performed using nondimensional equations (eqs 3 and 4 for a single oscillator and eqs 7 and 8 for coupled oscillators). The parameters are $\varepsilon = 10^{-3}$, $a_u = a_v = 3.7$, $c = 5$, and $\sigma_0 = 1$. The frequencies are (b) 48.1, (c) 44.4, and (d) 48.5 in the arbitrary unit, respectively.

$$C\frac{dT_i}{dt} = \omega_1\left[h(T_0 - T_i) + \beta a n_i \exp\left(-\frac{E}{RT_i}\right)\right] - \sigma\left(\frac{\mu}{L^2}T_j^4 - T_i^4\right) \quad (5)$$

$$\frac{dn_i}{dt} = \omega_2\left[k(n_0 - n_i) - a n_i \exp\left(-\frac{E}{RT_i}\right)\right] \quad (6)$$

for $i, j = 1, 2, (i \neq j)$, where μ is a constant that corresponds to the light absorption rate in gas. Radiation coupling is expressed as the term $\sigma(\mu/L^2)(T_j^4 - T_i^4)$.

Equations 5 and 6 were also nondimensionalized just like eqs 1 and 2 for the numerical calculation by introducing u_i and v_i, which are defined as

$$u_i = c\frac{T_i - T_0}{T_0} \qquad v_i = \frac{n_i}{n_0}$$

for $i = 1, 2$. The nondimenisonalized equations are

$$\frac{du_i}{dt} = \frac{1}{\varepsilon}\left[-u_i + a_u v_i \exp\left(\frac{u_i}{1 + u_i/c}\right)\right] - \sigma_0\left(1 + \frac{u_i}{c}\right)^4 + \sigma_0\mu_0\left(1 + \frac{u_j}{c}\right)^4 \quad (7)$$

$$\frac{dv_i}{dt} = 1 - v_i - a_v v_i \exp\left(\frac{u_i}{1 + u_i/c}\right) \quad (8)$$

for $i, j = 1, 2, (i \neq j)$, where $\mu_0 = \mu/L^2$.

With eqs 7 and 8, we calculated the time series of T_1 and T_2; the results, as shown in Figure 5c and d for different L (the distance between the two oscillators), reproduce the experimental results shown in Figure 3. Detailed analysis of the numerical results shows that the two stable synchronization modes coexist and that the ratio between the range of the initial values for in-phase synchronization and that for antiphase synchronization changes depending on L (data not shown). This feature is consistent with the experimental observation of the coexistence of the two modes around the intermediate region. The frequency dependence on the modes of synchronization, as shown in Figure 3c, is also reproduced, that is, the frequencies of oscillation that show antiphase synchronization are greater than those that show in-phase synchronization (Figure 5e).

5. Discussion

On the basis of the experimental and theoretical results, the mechanism of the oscillatory combustion of a set of three candles and the synchronous behavior in a coupled oscillator system can be discussed. For a single oscillator, oscillation is induced because of a limited oxygen supply. When one candle burns, sufficient oxygen is supplied. In contrast, when three candles are located close to each other, the supply of oxygen is insufficient to maintain a constant combustion compared with the supply rate of fuel, which results in the oscillatory behavior in combustion. Oscillatory combustion is also seen for the single flame of another type of candle, for example, a Japanese traditional candle with a thicker wick. This also supports the notion that oscillatory combustion is induced depending on the supply rate of paraffin.

For two sets of three candles, the main factor of interaction appears to be radiation. The order estimation suggests that diffusion coupling is not a factor due to the longer distance between the component oscillators compared to the characteristic length of thermal diffusion. Convection does not cause synchronization directly but plays a role in the release of heat from the burning area and in the supply of oxygen to maintain the burning state. Heat transfer with radiation is known to be proportional to the fourth power of temperature.[16] It should be noted that the variable T corresponds to the average value of temperature in the area near the flame. Thus, strictly speaking, the radiation term is not the fourth power of T but an increasing function of T, like σT^n $(1 < n \leq 4)$. We have confirmed that the similar results are achieved in the case that $n = 2$ and 3. In such cases, the range of the initial values for antiphase synchronization becomes smaller compared with that when $n = 4$. Nonlinear coupling with the fourth power of a variable, radiation coupling, is unique and may present interesting problems from the mathematical viewpoint since there have been few studies on coupled oscillators with a nonlinear coupling term. Furthermore, it is notable that both in-phase and antiphase synchronization and desynchronization can be observed with changes in only one parameter, that is, the distance between two oscillators, while in most coupled nonlinear oscillators, we have to change the manner of coupling to realize both in-phase and antiphase synchronization.

6. Summary

We constructed a simple experimental system on the combustion of candles, which showed limit-cycle oscillation and synchronization. A set of three candles exhibits oscillatory behavior, while a single candle exhibits constant combustion. We considered a set of three candles to be an oscillator and examined the coupling of two oscillators. If the distance between the two oscillators is small enough, they exhibit in-phase oscillation. When the distance is larger, they exhibit antiphase oscillation, and when it is much larger, they do not synchronize at all. We made mathematical models for a single oscillator and coupled oscillators and reproduced the characteristic behaviors of the candles. With this numerical calculation, we found that a lack of oxygen is a key factor in the oscillatory behavior of a set of three candles. The results also suggested that the synchronization of two sets of three candles is induced by coupling through thermal radiation.

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References and Notes

(1) Winfree, A. T. *The Geometry of Biological Time*; Springer: New York, 1980.

(2) Zaikin, A. N.; Zhabotinsky, A. M. *Nature* **1970**, *225*, 535.

(3) Jakubith, S.; Rotermund, H. H.; Engel, W.; von Oertzen, A.; Ertl, G. *Phys. Rev. Lett.* **1990**, *65*, 3013.

(4) Sakurai, T.; Mihaliuk, E.; Chirila, F.; Showalter, K. *Science* **2002**, *296*, 2009.

(5) Nicolis, G.; Prigogine, I. *Self-organization in Nonequilibrium Systems*; Wiley & Sons: New York, 1977.

(6) Kuramoto, Y. *Chemical Oscillations, Waves, and Turbulence*; Springer: Berlin, Germany, 1984.

(7) Pikovsky, A.; Maistrenko, Y. *Synchronization: Theory and Application*; Kluwer Academic: London, 2003.

(8) Strogatz, S. H. *Sync: How Order Emerges from Chaos in the Universe, Nature, and Daily Life*; Theia: New York, 2003.

(9) Kohira, M. I.; Magome, N.; Kitahata, H.; Yoshikawa, K. *Am. J. Phys.* **2007**, *75*, 893.

(10) Yoshimoto, M.; Yoshikawa, K.; Mori, Y.; Hanazaki, I. *Chem. Phys. Lett.* **1992**, *189*, 18.

(11) Ishida, T.; Harada, S. *Kagaku to Kyoiku* **1999**, *47*, 716.

(12) Scott, S. K.; Showalter, K. *J. Phys. Chem.* **1992**, *96*, 8702.

(13) Nagayama, M.; Ikeda, T.; Ishiwata, T.; Tamura, N.; Ohyanagi, M. *J. Mater. Synth. Process.* **2001**, *9*, 153.

(14) Yang, V.; Anderson, W. E. *Liquid rocket engine combustion instability*; American Institute of Aeronautics and Astronautics: Washington, DC, 1995.

(15) Lang, W.; Poinsot, T.; Candel, S. *Combust. Flame* **1987**, *70*, 281.

(16) Landau, L. D.; Lifshitz, E. M. *Statistical Physics*; Pergamon: Oxford, U.K., 1968.

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