Hexagonal superlattice state in dielectric barrier discharge

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We report on the observation of a hexagonal superlattice state (HSS) in dielectric barrier discharge in air/argon near atmospheric pressure. It bifurcates directly from the hexagonal state by increasing the applied voltage. The correlation measurements indicate that the HSS is an interleaving of three different transient sublattices. The spatial power spectrum demonstrates that the HSS has two separate wave vectors. The big wave vector and the small wave vector belong to the harmonic mode and the subharmonic mode, respectively, and they obey the triad resonant interaction $\vec{q}_1^{\,s}+\vec{q}_2^{\,s}=\vec{K}_c^{\,h}$.

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Dielectric barrier discharge (DBD, also referred to as the silent discharge) is well known for its industrial applications such as ozone generation and plasma display panels [1]. In recent years, the DBD system has become an attractive pattern formation system since various types of patterns have been observed [2–5]. Depending on the product of gas pressure and gas gap width, the discharge operates in different regimes. When the product is at several tens of Torr cm, the discharge consists of many filaments. These filaments can self-organize into various types of regular patterns for different parameters. In past years, some simple patterns like the square pattern, hexagon pattern, and spiral pattern were studied [4–6]. Recently we have observed some different types of superlattice patterns such as a square superlattice pattern [7], grid state [to be published elsewhere], and a hexagonal superlattice state (HSS). In this paper, we will focus on the spatiotemporal dynamics of the HSS.

The HSS patterns are commonly observed in many systems, such as reaction-diffusion, Faraday, and nonlinear optical systems [8–12]. In a reaction-diffusion system, Berenstein *et al.* observed the HSS using illumination through the inverted hexagonal mask [8]. Kudrolli *et al.* observed the instantaneous HSS in a two-frequency forcing Faraday system [9]. In a nonlinear optical system, the HSS was obtained by using a Fourier spatial filter consisting of lenses in the feedback loop [10]. In a ferrofluid system, the HSS was observed by using a vertical oscillation that is driven by a sinusoidal magnetic field on colloidal suspension of magnetic power. The instantaneous transition from the hexagon to the HSS is so rapid that the process in detail cannot be observed [13]. Theoretically, Silber presented a symmetry-based approach to analyze the bifurcation problems from the standing hexagon to the HSS pattern, which is one of the possible solution branches [14].

In this letter, we will report the observation of the HSS in dielectric barrier discharge by using single driving frequency. The spatiotemporal dynamics of the HSS, which is a secondary bifurcation of the hexagon state (HS), is studied.

The experimental setup has been described in previous papers [4–7]. The following is a brief description. Two cylindrical containers, with diameter of 75 mm, sealed with 1.5 mm thick glass plates, are filled with water. A metallic ring immerses in each of the containers and is connected to a power supply. Thus, the water acts as a liquid electrode. The glass plates serve as dielectric layers. A glass frame with the thickness of 1.5 mm was placed between the two parallel glass plates, serving as the lateral boundary. A sinusoidal ac voltage is applied to the electrodes. All of the apparatuses are enclosed in a big container filled with the mixture of argon (60% ± 5%) and air (40% ± 5%). The pressure p can be changed from 60 to 70 kPa. The amplitude of the applied voltage is measured by a high-voltage probe (Tektronix P6015A 1000X). The light signals emitted from two individual discharge filaments are detected by two lensaperture-photomultiplier tubes (PMTs, RCA 7265) and recorded by an oscilloscope (Tektronix TDS3054B, 500 MHz) simultaneously. Thus, the correlation between two discharge filaments can be studied.

In our gas discharge system, the supply voltage serves as an experimental bifurcation parameter. The HSS bifurcates from the hexagonal state (HS) at the critical point. Fig. 1 shows the time-averaged pictures of the HS and the HSS with their corresponding spatial power spectra.

With a gradual increase in the applied voltage, the wave number of the HS increases smoothly through the generation of new discharge filaments. Some filaments at the edge of the discharge area become longer and then each of those splits up to two filaments when the voltage is increased. The new generated filaments separate until all filaments reach a new balance position. However, the symmetry of the HS is not broken until $U_c = 4.5$ kV at which the system enters into the critical region. The domain of the HSS appears if the voltage increases further.

Beyond the critical point, some new filaments appear near some filaments at the edge of the discharge region. They are confined by the original filaments forming the pairs of filaments. Each pair of filaments, which is called the big spot later, rotates in random direction with a rapid velocity, looking like a hollow ring. The six small spots around a big spot stay stationary and form a cell of the HSS with the big spot. Generally, a HSS domain appears near the boundary of the discharge region at first and propagates via the front toward the other side of boundary gradually until this state dominates the entire plate as the applied voltage is increased. The HSS will bifurcate to the chaos state when the applied volt-

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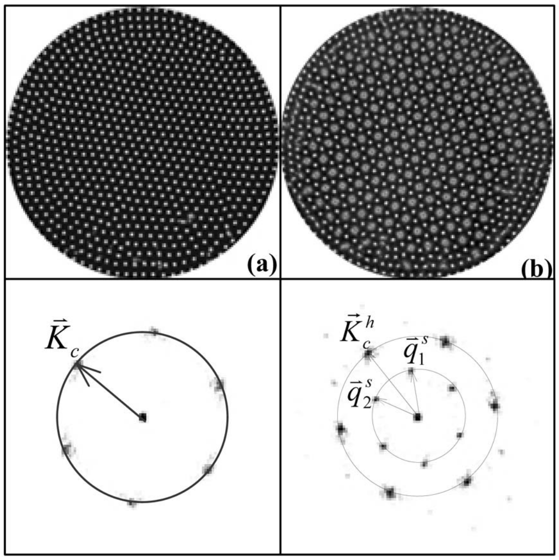

FIG. 1. Images of the time-averaged states (the top row) obtained in the barrier discharge in air/Ar mixture by increasing the applied voltage with their corresponding spatial power spectra (the bottom row). (a) The HS, $U_c=4.5$ kV and (b) the HSS, $U=5.1$ kV. The other parameters are the gas pressure $p=70$ kPa, driven frequency $f=58$ kHz, and the gas gap $d=1.5$ mm.

age is increased further. The inversion progress can occur when the applied voltage is decreased.

From Fig. 1, it can be clearly seen that the critical wave number K_c^h of the HSS is less than the K_c of the HS, and six additional components q in the inner ring appear in the HSS. The triad cross coupling between different modes results in the relative 30°-orientation between the waves $(\vec{K}_c^h, \vec{q}^s)$. The magnitude of the vector $q_1^s = q_2^s$ is approximation $K_c^h/\sqrt{3}$. The spatial power spectrum demonstrates that this pattern formation can be explained by a triad resonant interaction $\vec{q}_1^s + \vec{q}_2^s = \vec{K}_c^h$.

Compared with that in other systems, the HSS in our experiments can exist for a long time without any modulation, which is useful for us to study the characters of the HSS. In order to study the spatiotemporal behavior of the HSS, the correlations between the light signals of two different filaments are measured and shown in Fig. 2. Figure 2(a) shows that each big spot in the HSS has two discharge pulses per half cycle, and the light emissions of two big spots selected randomly have the same temporal parallelism structures. Figure 2(b) shows that each small spot has a single discharge pulse per half cycle and all small spots discharge synchronously. The same interval between two pulses in consecutive half cycles shows that the small spots display a harmonic response with respect to the driven period of light emission. (Note: The driven period is the half cycle of applied voltage because the light emission is independent of the sign of voltage.) Figure 2(c) reveals that the discharges of the small spots fire at the mid-time of the two pulses of the big spots. If we let the light from the part of the big spot go into the

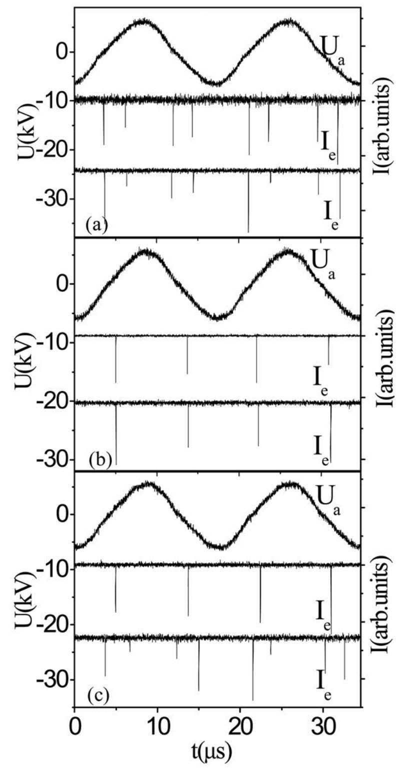

FIG. 2. The waveforms of applied voltage U_a (top) and the light emissions I_e (middle and bottom) from two big spots in the HSS in(a), two small spots in (b), and a small spot and a big spot in (c), respectively.

PMT, it turns out that only one pulse appears per half cycle, as shown in Fig. 3, which demonstrates that the big spot of the HSS is composed of two filaments appearing at almost the same position but different times. This is different from the big spot in the square superlattice pattern, which ignites twice in each half cycle of the applied voltage [7]. In addition, it can be seen in Fig. 3 that the intervals of two discharge pulses in consecutive half cycles are alternating between a long one and a short one, displaying a subharmonic response. Therefore, the HSS is an interleaving of three sublattices appearing at different times in each half cycle and the HSS is a mixed-mode oscillation.

Figure 4 is a schematic diagram of the spatiotemporal evolution of the HSS in one half cycle of the applied voltage. At first, part of the big spots sublattice (denoted by ○) appears, whose light signal corresponds to the first spike in the bottom curve in Fig. 2(c). Then the small spots (denoted by

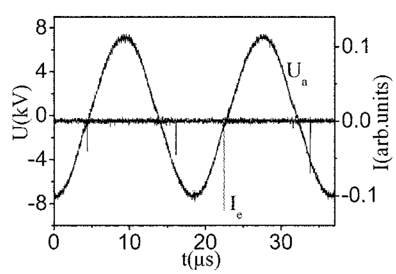

FIG. 3. The light signal of part of the big spot in the HSS.

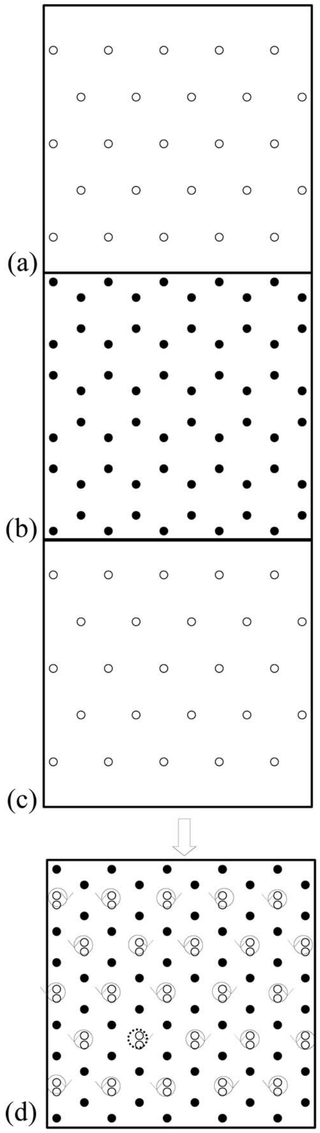

FIG. 4. The schematic diagram of the spatiotemporal evolution of the HSS in a positive half cycle of the applied voltage. (a) The spatial distribution of the part of the big spots (denoted by ○) whose light signals correspond to the first spike in light emission in Fig. 2(c). (b) The spatial distribution of the small spots (denoted by ●) whose light signals correspond to the middle curve in Fig. 2(c). (c) The spatial distribution of the other part of the big spots (also denoted by ○) whose light signals correspond to the second spike in light emission in Fig. 2(c). (d) The time integral of the (a), (b), and (c) results in the HSS. The dashed circles with arrows represent that the pairs of filaments composing the big spots rotate with random directions.

●) sublattice ignites, whose light signal corresponds to the middle curve in Fig. 2(c). At last the other part of the big spots sublattice (also denoted by ○) appears, whose light signal corresponds to the second spike in the bottom curve in Fig. 2(c). Figure 4(d) gives the HSS resulting from the time integral of Figs. 2(a), 2(b), and 2(c). It is worth pointing out that the two sublattices corresponding to the first stage and the third stage almost overlap, looking like the big spots sublattice. In Fig. 4(d), the dashed circles with arrows represent that the pairs of the filaments rotate in random directions.

In the DBD system, the temporal response of the wave vector can be facilely obtained by measuring the temporal behavior of the different filaments. Let us look back to the spatial power spectrum of the HSS in Fig. 1(b). The big wave vector ($\vec{K}_c^h$) belongs to a harmonic as it corresponds to the spatial frequency of the small spots sublattice, while the small wave vector ($\vec{q}^{\,s}$) belongs to a subharmonic as it corresponds to the spatial frequency of the big spots sublattice. Therefore, the different parity modes can be excited simultaneously by only one driven frequency in dielectric barrier discharge. However, in a Faraday system, multiple driven frequencies are needed generally for exciting the different parity modes simultaneously.

As is well known, the parity of the wave vector plays a key role in the triad resonance. Two modes of like parity can couple quadratically to a harmonic mode but not a subharmonic mode [15]. The triad resonance $\vec{q}_1^{\,s}+\vec{q}_2^{\,s}=\vec{K}_c^h$ in Fig. 1(b) results in that the critical wave vector K_c^h must belong to the harmonic mode. Therefore, the temporal response of the small spots sublattice, whose spatial frequency corresponds to the critical wave vector, must be harmonic no matter what the parity of the big spots sublattice is.

In conclusion, the spatiotemporal dynamics of the HSS in DBD is studied by the measurements of correlation between filaments. The HSS is an interleaving of three different transient sublattices. The spatial power spectrum demonstrates that the HSS has two separate wave vectors with a wavelength ratio of $\sqrt{3}$. The harmonic and subharmonic modes obey the triad resonant interaction $\vec{q}_1^{\,s}+\vec{q}_2^{\,s}=\vec{K}_c^h$.

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