Clusters of ionization clumps in weakly ionized annular rf discharges

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The self-organized localized structures in a medium pressure annular rf glow discharge are investigated. Clusters with different number of ionization clumps are observed through inversed bifurcation processes after the onset of the periodic crystal-like structure. The interaction between clusters is long range repulsive, and short range attractive before reaching the center highly repulsive core. The scattering and the formation of the stable and metastable larger clusters through the collision between two clusters are demonstrated. [S1063-651X(98)15902-0]

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Localized patterns can be self-organized in many nonlinear dissipative extended systems including fluids, optical, and chemical systems under continuous input of energy to overcome dissipation [1–9]. Their structures, formation processes, motions, and interaction behaviors have long been interesting subjects. The stable structures with single pulse and clusters of pulses have been observed. Between the two single pulses, various interactions such as repulsion, mutual penetration, annihilation, survival of one pulse, etc., upon their collisions have been demonstrated [7–9]. However, the interaction between the two multipulse clusters has been much less well addressed. The glow discharge plasma is a typical nonequilibrium dissipative system exhibiting rich universal nonlinear spatiotemporal phenomena [10–17]. Localized structures are also expected in different discharge systems. For example, the formation of the single pulse current filament structures and their repulsive interaction in a dc discharge system, and the formation of the two-dimensional (2D) multipulse clusters in a 200 kHz high pressure discharge system have been reported [14]. In this work, using a weakly ionized annular rf glow discharge system with rotational symmetry, we demonstrate the formation of traveling clusters with different number of ionization clumps through a cascade of bifurcations from the periodic state. We further investigate their generic dynamical behavior, especially concentrating on their interactions such as scattering with and without pulse exchange process, recombination, and dissociation.

The glow discharge can be generated at pressures from a few mTorr to a few tens of Torr by applying an electric field between two electrodes [10,11]. The ionization and recombination are similar to the reaction processes in chemical systems. The diffusion and space charged field induced transport further provide spatial coupling. The interplay among the above processes leads to the ionization instability [11–15]. Although it is more complicated than the typical chemical reaction-diffusion or fluid systems, it also exhibits many universal nonlinear spatiotemporal behaviors [11–17]. In our recent work, a periodic ionization pattern was observed and their transient bifurcation processes were studied in a rf discharge in the annular gap between two concentric cylindrical electrodes at a few hundred mTorr pressure [17]. The system is driven radially and has rotational and reflection symmetries. Basically, by increasing pressure, a periodic ionization pattern with different wave numbers (number of periods in one circle) along the azimuthal direction can be formed through a cascade of subcritical bifurcations associated with Eckhouse instability. The pattern can rotate clockwise or counterclockwise after the onset through a spontaneous parity breaking process or pinned by the surface defect. The previous preliminary studies also showed that, through the inverse bifurcation, the system can be switched from the periodic pattern to the single-pulse state with localized but separated ionization clumps. In this study, we further concentrate on the multisoliton behavior and their interactions in the fine-tuned inverse bifurcation processes. It is found that, by slowly decreasing system pressure after the onset of the first traveling periodic pattern, the system first bifurcates to a periodic rocking mode and then to the states with traveling clusters. Depending on system pressure, each cluster consists of a few ionization clumps. The intercluster interaction is long range repulsive and short range attractive before reaching the center repulsive core. By using magnetic tweezers, the recombination of two clusters to larger stable and metastable clusters, and the scattering processes with and without exchanging clumps are demonstrated.

The experiment is carried out in an rf cylindrical discharge system similar to that reported elsewhere [17]. It consists of a hollow stainless steel outer electrode with a 9-cm diameter and capacitively coupled to a 14-MHz rf power system, and a grounded center electrode with a 6-cm diameter. Argon at a few hundred mTorr pressure is used for the discharge. An annular groove with 3.5-mm height and 8.5-mm width is cut along the top edge of the center electrode (Fig. 1) for the easier generation (due to the hollow

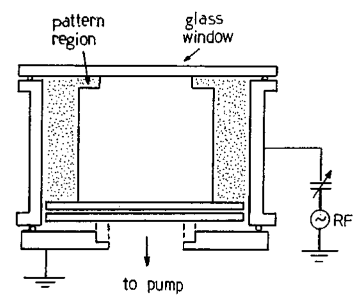

FIG. 1. Sketch of the side view of the cylindrical discharge system.

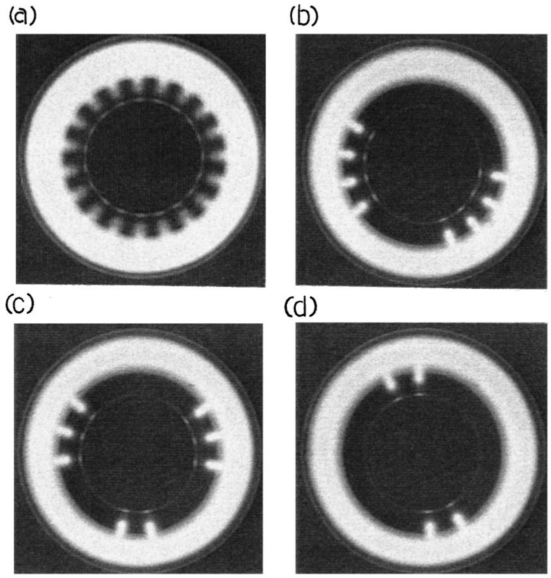

FIG. 2. The CCD pictures of the plasma emission from the top of the chamber at different pressures. (a) The periodic pattern at 1020 mTorr (the dashed bright circle is due to the reflected emission from the inner lower corner of the groove). (b) The 4-4 cluster state at 910 mTorr. (c) The 3-3-2 cluster state at 426 mTorr. (d) The 2-2 cluster state at 419 mTorr.

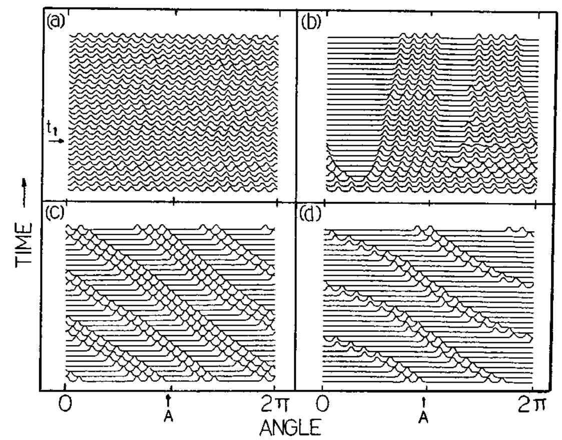

FIG. 3. The space-time diagrams of the traveling pattern at 14 W rf power (arbitrary unit for the pattern amplitude). (a) The periodic traveling pattern with varying direction as pressure linearly decreases from 1020 to 1012 mTorr. (b) The bifurcation to the 4-4 cluster state as the pressure linearly decreases from 918 to 910 mTorr. (c) The traveling 3-3-2 cluster state at 426 mTorr constant pressure. (d) The traveling 2-2 cluster state at 419 mTorr constant pressure. The rotation is modulated by the defect at point A. The time step $\Delta t = 0.1$ s for (a) and (b) and 1/3 s for (c) and (d).

cathode confinement effect) and observation of the ionization pattern. The groove size is chosen to limit the pattern as a quasi-1D pattern. The pattern becomes 2D when the groove is too wide. Unlike the Al center electrode in the last experiment [17], the center electrode is made of stainless steel, which is more defect free (e.g., avoiding the surface scratching and oxidation which easily pin the clumps). At low rf power (or low pressure), an azimuthally uniform discharge can be sustained in the gap between the two electrodes. If the increasing pressure (or power) passes a certain threshold, an ionization instability with periodic bright and dark regions can be formed in the groove [e.g., Fig. 2(a)] through a subcritical bifurcation. The emission intensity distribution over the horizontal plane (integrated along the axial direction) is monitored by a video camera 2.5 m above the system through the top glass window and is digitally processed. Since the emission of the glow is from the electron-impact excited atoms, the bright regions correspond to the regions with high ionization degree and electron density. Similar to the previous experiment, the periodic pattern can travel either clockwise or counterclockwise as the symmetry is spontaneously broken by the fluctuation at bifurcation.

Figure 2 shows different pattern images by decreasing system pressure after the onset of the periodic pattern at 14 W rf power. Figures 3(a) and 3(b) shows the spatiotemporal evolution of the discharge intensity along a circle crossing the pattern as the pressure is linearly ramped down. First the traveling pattern runs into a rocking mode periodically alternating its traveling direction [after t_1 in Fig. 3(a), from 1020 to 1012 mTorr]. The mode number (the total number of oscillations in a circle) of the periodic pattern decreases as pressure further decreases. Finally, some clumps disappear and the periodic pattern dissociates into two large clusters of clumps [Fig. 3(b), 918 to 910 mTorr]. Figure 2(b) shows the corresponding image of the two clusters. Further decreasing pressure can form clusters with a smaller number of clumps through the similar dissociation process. For example, Figs. 2(c) and 2(d) show the states with 2-3-3 (i.e., with 2, 3 and 3 pulses in each cluster, respectively), and 2-2 cluster structures at 426 and 419 mTorr, respectively. The 1-1 structure with 180° separation has also been observed (not shown). The clusters can be stably sustained under the fixed control parameters. The half-width of the clumps in Figs. 2(c) and 2(d) is 3.8 mm.

Similar to the particlelike behavior of single ionization clumps found in our previous study [17], the clusters interact through long range repulsive force. The small clusters tend to keep the largest mutual separation. For example, the clusters in Figs. 2(c) and 2(d) keep 120° and 180° separations, respectively. The clusters also spontaneously rotate clockwise or counterclockwise. The rotation speed increases as pressure decreases. The rotation of the 2-3-3 state and the 2-2 state at constant pressures are shown in Figs. 3(c) and 3(d), respectively. The electrode defect at point A modulates the motion of clusters. The defect is caused by some localized surface magnetic impurity, which generates about 1.5 G local field on the electrode surface. The slowing down of the cluster around point A also slows down the motion of the neighboring clusters. The modulation becomes more serious as the intercluster distance increases. This manifests that the intercluster repulsion decreases with increasing separation, which makes the system more compressible.

We also conduct some interesting collision experiments between clusters by fixing the position of one cluster. For example, one cluster can be trapped by putting the tip of a magnetized screw driver on the glass window surface right over point A [Fig. 4(a)]. The second spontaneously rotating cluster is either bounced by the pinned cluster and alternates its moving direction [around t_1 in Fig. 4(a)] or pushes the pinned cluster out of the trap [around t_2 in Fig. 4(a)]. It is accelerated and falls into the trap as the pinned one leaves the trap.

Figure 4(b) shows another interesting scattering process.

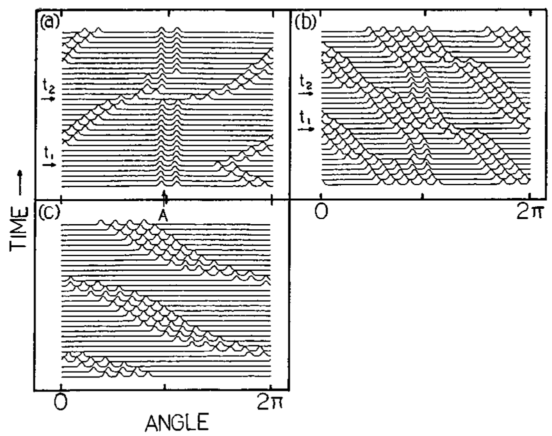

FIG. 4. The space-time diagrams of the collision processes between clusters at 14 W rf power. (a) The scattering without clump exchange ($P=419$ mTorr; $\Delta t=0.5$ s). (b) The collision with one clump exchange through the formation of a metastable large cluster ($P=426$ mTorr; $\Delta t=0.2$ s). A tweezer is put at point A to pin one cluster in (a) and (b). (c) The evolution of the 4 clump cluster after removing the magnetic tweezers, which force the fusion between the two 2-clump clusters ($P=426$ mTorr; $\Delta t=0.5$ s).

The screw driver is put at point A to lock the position of the two-clump cluster of the 3-3-2 state. The spontaneous rotation makes the 3-clump cluster coming from right recombine with the 2-clump cluster in the trap and form an intermediate larger cluster with 5 clumps at t_1. The lifetime of the 5-clump cluster is about 2 sec and it quickly dissociates into 2 clusters at t_2. After the dissociation, the 2 clumps on the right are locked and the 3 clumps on the left travel together as a cluster to the left after the dissociation. Namely, this is similar to the collision between two molecules with one particle exchange through the formation of a larger metastable molecule.

Not all the states of the two recombined clusters have particle exchange and short lifetime. For example, two magnetized screw drivers are used as tweezers to push the two repulsive clusters of a 2-2 state to fuse into a larger cluster with 4 clumps. The tweezer is removed right after recombination. Similar to the 2-2 state in Fig. 4(a), the clusters are repulsive at a short distance. However, once they are forced to overcome the repulsive barrier, they form a stable state. Figure 4(c) shows the rotation of the stable 4-clump cluster after removing the tweezers. Again their rotational motion is modulated by the defect at point A.

The possible mechanism for the formation of ionization clumps was briefly mentioned in our previous works [15–17]. The rf electric field is the main source feeding energy transversely to this open dissipative system. The electron impact ionization process (the rate increases with electron density and energy) and the recombination process (the rate is proportional to the square of the electron density) are the local source and sink for enhancing and suppressing electron density fluctuation, respectively. If we use a 1D model (along the azimuthal direction) for this system, the transverse transport of charged particles to the surrounding wall due to diffusion and electric field can also be treated as a local sink. Azimuthally, the ambipolar diffusion provides spatial coupling and tends to spread out the highly localized sharp structure.

Although the system looks quite complicated, generically, the state of the system is determined by complicated reaction-diffusion-type partial differential equations similar to chemical systems [9,15]. The periodic and localized structures are generated under the interplay between the above processes. For example, if we start from a state with a local region with slightly higher electron density than the background, the electron density will grow due the positive feedback provided by the electron impact ionization process. Under the rf electric field, the electron loss to the chamber wall is much higher than that of ions, which makes the plasma float positively with respect to the electrode [10]. The increasing local plasma density increases space charge and in turn increases the local plasma potential. The electrons can thereby gain higher energy, which further enhances the local ionization rate and electron density until the profile is steep enough and the amplitude is large enough to cause enough diffusion and nonlinear recombination losses. Note that the ionization rate is a highly nonlinear function of the electron energy, and the diffusion rate strongly depends on the profile of the structure. The amplitude, scale, and shape of the steady state clump, cluster, or the periodic structure are determined by the balance between the local growth and loss rates. A recent theoretical study also demonstrates the possibility of the formation of periodic and soliton structure of this system [18].

The mean free paths for electron impact ionization and electron-neutral momentum transfer collisions both decrease with increasing pressure [10]. At low pressure, they are larger or comparable to the groove width. Electrons are hardly generated through ionization and hardly confined through collisions with neutral Ar in the groove. Therefore, no discharge can be sustained in the groove until the pressure is high enough to balance the electron generation and loss to the wall. The onset pressure increases with the decreasing groove height. In the reverse bifurcation process by decreasing pressure after onset, the pattern in the groove switches to the cluster structure instead of completely turning off due to the hysteresis effect of this nonlinear system. The half-width of the clump also increases with the increasing mean free paths as the pressure decreases. The groove height sets up a lower bound for the pressure over which all the clusters or clumps diminish.

Figures 5(a) and 5(b) show the structures in the moving frame and the homoclinic trajectories of the 2- and 3-clump clusters in the phase space reconstructed by $I(\phi)$ versus $I(\phi+\Delta\phi)$ plots for the 2- and 3-clump clusters, respectively, where I is the emission intensity and ϕ is the azimuthal angle in the moving frame. The periodic pattern has a limit cycle trajectory (not shown). Unlike the pulse structures in other dissipative systems [2,7–9], our localized structures cannot interpenetrate or annihilate upon collision and their height is constant under a fixed control parameter. Their overall shapes have reflection symmetry within our noise limit, even when they are traveling pulses. Around the similar pressure, the emission intensity profiles of a single isolated clump and the clumps in the 2-clump and 3-clump clusters are about the same except the tail part. The inter-clump separation is the same for the 2-clump and 3-clump clusters. Namely, under a fixed operating condition, the nonlinear partial differential equations of the system support

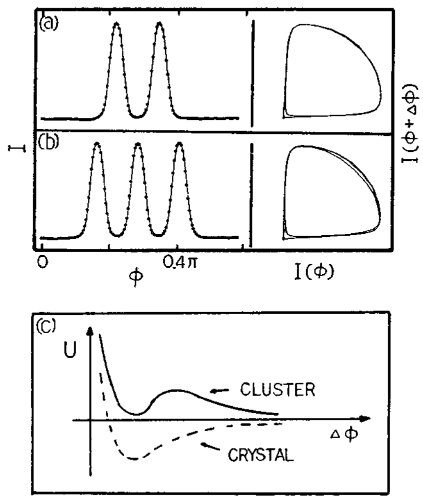

FIG. 5. (a),(b) Left column: the plasma emission intensity profiles (ϕ is the azimuthal angle) of the 2-clump cluster and the 3-clump clusters [averaged over 30 pictures in the defect-free region from the 3-3-2 sate shown in Fig. 3(a)] in the moving frame. Right column: their corresponding phase portraits using $I(\phi)$ vs $I(\phi+\Delta\phi)$ plots with $\Delta\phi=5.625°$. (c) The sketch of the effective pair interaction potentials for the cluster (low pressure, e.g., 420 mTorr) and periodic crystal (high pressure, e.g., 1020 mTorr) states.

multistable cluster structures with different numbers of clumps. The complicated nonlinear processes (ionization, recombination, ambipolar diffusion, etc.) do not favor the overlapping of the clump tails and make the single clumps and clusters behave as quasiparticles with long-range interactions, although their interactions are really unlike the direct Coulomb-type interactions between the real charged particles.

We can qualitatively sketch their effective interaction potential for those quasiparticles in the form shown in Fig. 5(c) according to our above observation. It has a long range repulsive tail, a short range attractive well, and a harder center repulsive core. Whether the pair can form a stable or a metastable state depends on the depth of the attractive well and the height of the barrier. Their values are functions of the system control parameters such as pressure or rf power. If the attractive well is too shallow, a small fluctuation can kick the particle over the barrier, i.e., dissociate the metastable bound state and keep the pair at the largest possible separation. On the other hand, if the barrier keeps decreasing and the attractive well deepens (e.g., by increasing pressure), the effective interacting potential will eventually be attractive except the center repulsive core. The system favors the state with one-dimensional crystal-like periodic structure as observed at higher pressure. It is also interesting to point out that the bifurcation from a periodic traveling mode to a rocking mode with alternating traveling directions [Fig. 3(a)] is a consequence of the slight symmetry breaking due to the presence of defect. The observation generically agrees with the theoretical prediction of a similar system [19].

In conclusion, we have investigated the localized structure of ionization instability in a medium pressure steady state rf discharge system. The important observations are listed as follows: (1) Decreasing pressure from a state with a periodic traveling pattern can turn the system into a periodic rocking mode under slight system asymmetry. (2) At low pressure, the fission into stable clusters is associated with the disappearance of some ionization clumps. (3) The amplitude, shape, and interclump distance for different clusters are the same under a fixed operating condition regardless of the number of clumps in the cluster. The clusters can be treated as systems with bonded quasiparticles, which can travel spontaneously. (4) The scattering and fusion experiments show that the intercluster interaction is repulsive at long range and attractive at short range before reaching the center hard core. The interaction can be controlled by system parameters.

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