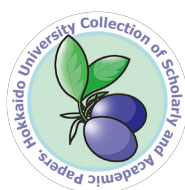




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Shot-Noise-Induced Random Telegraph Noise in Shuttle Current

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Random telegraph noise in the electric current produced by shot noise is predicted for an array of movable colloid particles by Monte Carlo and molecular dynamics calculations. The electron transport is attributed to the shuttle mechanism where moving colloid particles carry charges. The colloid-particle motion induced by the source-drain voltage shows periodic and/or quasiperiodic vibrations, and the current value depends on the vibration modes. Shot noise that is uncorrelated with the colloid-particle motion causes transitions between the periodic and quasiperiodic vibration modes, resulting in random switching between the current levels corresponding to the vibration modes.

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Dynamic motion of nanoscale structures, single electron tunneling, and Coulomb blockade effects play key roles in electron transport in nanoelectromechanical systems (NEMS) [1]. The electron tunneling modulated by the motion of nanostructures provides transport properties inherent to the new systems. For example, a single C_{60} molecule transistor exhibits a conduction gap expanding in proportion to the gate voltage [2], that is substantially different from transport properties predicted by the orthodox theory for single electron transistors with fixed quantum dots [3]. The relevant electron transport properties arise from the shuttle mechanism where charges are carried by the C_{60} molecule movement driven by the source-drain voltage [4,5].

Another example is the random telegraph noise (RTN) of the current in a one-dimensional array of moveable metal nanoparticles [6]. Colloid particles and their arrays have been proposed for use as building units for Coulomb blockade devices [7,8], and the electron transport properties of these devices have attracted much attention. In a previous paper [6], I made numerical investigations of the electron transport properties in colloid-particle arrays, in which the shuttle mechanism is responsible for the electron transport. However, in contrast to the case for single molecules, the dynamic motion of colloid-particle arrays is complicated even for an array of two particles. The two-particle array supports two vibrational modes. Whether one or two modes are expressed is a strong function of the source-drain voltage. For a certain region of applied voltage, in which both modes can be excited, the vibrations randomly switch from one of the modes to the other and vice versa. Because the current has a different value for each mode, the transitions result in random switching between the two current levels, or RTN in the current.

RTN in the current is observed in many systems such as small metallic conductors, ultrasmall tunnel junctions, metal-oxide-semiconductor field-effect transistors (MOSFETs), and so on [9]. The RTN was found to be produced by the common mechanisms of the transition of a

single electron or a single defect [9]. On the other hand, the colloid-particle arrays mentioned above cannot be described by such mechanisms yielding RTN. Rather, the RTN in the colloid-particle arrays is known to be somehow associated with the dynamics of the colloid particles [6], although the switching mechanism for the vibration modes is not well understood. In this Letter, we elucidate the switching mechanism for the dynamics of colloid-particle arrays, and demonstrate that this is a new type of mechanism for the RTN in current, appearing for the first time in NEMS.

An array of colloid particles can be modeled by metal spheres linked to others and electrodes by springs representing the ligands, as shown in the inset of Fig. 1. Metal-core motion may be treated classically because of large mass M of the colloid particles. Limiting the motion to the

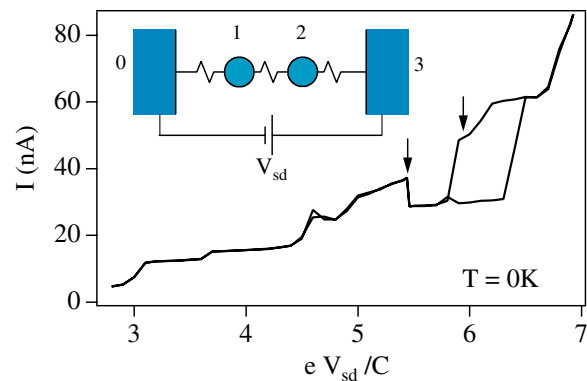


FIG. 1 (color online). I - V characteristics of the two-linked gold nanoparticles (Au_{55}) at $T = 0$ K for $R_T = 10^{10} \Omega$ [8]. The arrows indicate V_{sd} for the dip in current, $\frac{eV_{sd}}{C} = 5.5$, and RTN in the current, $\frac{eV_{sd}}{C} = 6.0$, respectively. The inset shows the model of the linked two-colloid particle array. Relevant parameters for Au_{55} clusters are $r = 0.7$ nm, $d = 2.1$ nm [7], $M = 1.8 \times 10^{-23}$ kg, $\epsilon^* = 3$ [8], $\lambda = 0.05$ nm [6], and $K = 0.05$ Nm $^{-1}$ [6]. The damping parameter is $\gamma = M\omega/3$, where ω is a characteristic frequency given by $\omega = \sqrt{K/M}$.

longitudinal displacement of the cores relevant to electron transport, the Newtonian equation of motion is as follows:

$$M\ddot{x}_i = F(x_i - x_{i-1}) - F(x_{i+1} - x_i) - \gamma\dot{x}_i - \frac{en_i}{\varepsilon^*L}V_{sd} - (\delta_{i,1} - \delta_{i,2})\frac{e^2n_1n_2}{4\pi\varepsilon^*\varepsilon_0(x_1 - x_2)^2}, \quad (1)$$

for $i = 1$ and 2 , where x_1 and x_2 are the positions of the cores, and n_1 and n_2 are the number of excess electrons on the cores with respect to the electrically neutral state. L is the distance between the electrodes. V_{sd} is the source-drain voltage, and ε^* is the effective dielectric constant of a ligand. The elasticity of ligands stems from the folded structure of the atomic bonds, analogous to the behavior of rubber. Then the following empirical formula [10] is assumed for F appropriate for rubberlike materials: $F(x) = -K[x - 2r - \alpha(x - 2r)^{-2}]/3$, where K is a spring constant, $\alpha = (d - 2r)^3$, d is the equilibrium distance between the particles or between the electrodes and the adjacent cores, and r is the core radius. The third term of the right-hand side of Eq. (1) is a damping term due to energy dissipation such as that due to the internal friction of the ligand.

The change in the electron numbers n_i is due to electron tunneling among colloid particles and electrodes. Taking account of the exponential increase of the tunnel resistance with increasing distance [4] and limiting the tunneling to that between nearest neighbors, the tunneling rate Γ_{ij} for the tunneling from position i to j is given at $T = 0$ K, from the golden rule, by

$$\Gamma_{ij} = \frac{\exp(-\delta u_{ij}/\lambda)}{e^2R_T} \Delta\mathcal{F}_{ij} \Theta(\Delta\mathcal{F}_{ij}) \quad \delta_{i,j\pm 1}, \quad (2)$$

where R_T is the tunnel resistance at the equilibrium distance d , δu_{ij} is the relative displacement given by $\delta u_{ij} = |x_i - x_j| - d$, $\Delta\mathcal{F}_{ij}$ is the change in electrostatic energy of the system due to the tunneling. λ is the decay parameter of the electron wave function in the barrier region, and $\Theta(x)$ is the Heaviside step function. The electrostatic energy is given by

$$\mathcal{F} = -eV_L m_1 - eV_R m_2 + \sum_{i=1}^2 \left(\frac{e^2 n_i^2}{2C} + en_i x_i \frac{V_{sd}}{\varepsilon^*L} \right) + \frac{e^2 n_1 n_2}{4\pi\varepsilon^*\varepsilon_0 |x_1 - x_2|}, \quad (3)$$

where constant terms and the charge induced by the difference of the work functions are neglected [11,12]. $m_{1(2)}$ is the number of electrons that have entered from the left (right) electrode to the core 1 (2). The self-capacitance of the cores C is assumed to be independent of the motion, and is approximated by $C = 4\pi\varepsilon^*\varepsilon_0 r$.

This model was applied to gold nanocluster (Au_{55}) arrays [6], which have often been used in experimental studies [8,13,14]. The current-voltage (I - V) characteristics

were obtained by means of molecular dynamics for the colloid-particle motion as well as by a Monte Carlo method for electron tunneling [11]. Figure 1 shows the characteristic behavior of the I - V curves at $T = 0$ K for an array of two Au_{55} nanoclusters: the staircase structure at low V_{sd} , a dip in current at $eV_{sd}/C = 5.5$, and hysteresis between $eV_{sd}/C = 6$ and 6.5 . RTN in the current appears in the narrow voltage region $eV_{sd}/C \approx 6$ marked by an arrow in Fig. 1 near the lower end of the hysteresis I - V curves, which are shown in Fig. 2.

Because the tunneling rate increases exponentially with decreasing distance between colloid particles or between the electrodes and colloid particles, most of the tunneling events take place near the electrodes or between the proximal colloid particles, in correlation with the motion. Charges are, as a result, dominantly carried by moving colloid particles between the electrodes, in a similar way to that for the single nanoparticle case [4]. Hence, the current value depends on how often the colloid particles approach the other particle or the electrodes, that can be specified from a knowledge of the vibration classes that the array can execute. The two-nanoparticle array supports two kinds of vibration classes; a quasiperiodic vibration mode and a periodic one. Figure 3 shows representative cases for their trajectories as well as their power spectra just below and above the dip in current at $eV_{sd}/C = 5.5$. The trajectories and the electrodes are offset for legibility, so that they cross when a nanoparticle collides with the other one or the electrodes. Figure 3(a) shows a typical power spectrum of the quasiperiodic vibrations, exhibiting a structure containing four conspicuous peaks, together with the corresponding trajectories of the colloid particles. On the other hand, the spectrum in Fig. 3(b) exhibits three sharp peaks, two of which are the harmonics of the peak at the lowest frequency f_a , characteristic of the periodic vibrations of the nanoparticles.

The RTN is caused by the transitions between the quasiperiodic and periodic vibrations. We see that the transitions are triggered off by electron tunneling events which

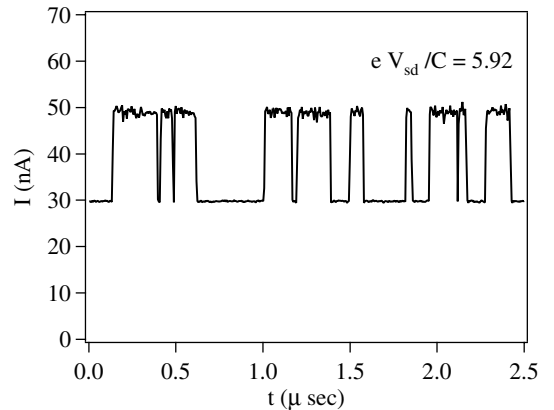


FIG. 2. RTN in the current at $eV_{sd}/C = 5.92$ for $R_T = 10^{10} \Omega$.

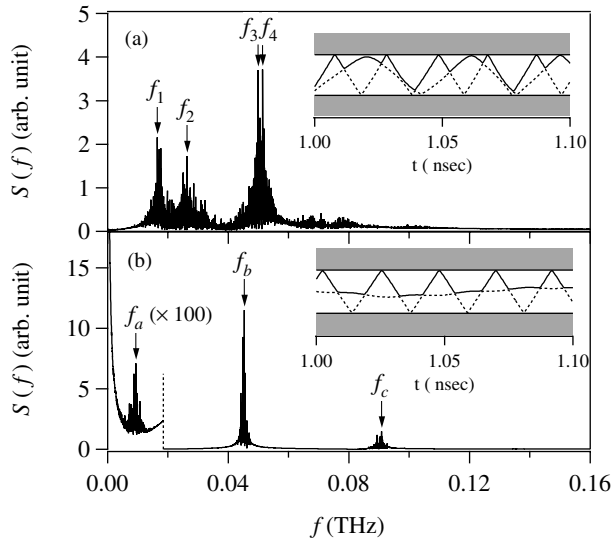


FIG. 3. Power spectra of the nanoparticle vibrations together with their trajectories for (a) $\frac{eV_{sd}}{C} = 2.70$ and (b) $\frac{eV_{sd}}{C} = 2.80$ with $R_T = 10^{10} \Omega$. The frequencies f_i denote those of the spectral peaks. The trajectories are denoted by the solid and dashed lines, and the shaded regions denote the electrodes. The trajectories are offset for legibility.

are uncorrelated with the colloid-particle motion since these classes of vibration are stable as long as the tunneling events occur in correlation with the colloid-particle motion. Figure 4 shows that such a tunneling event causes a transition from the periodic vibrations to the quasiperiodic ones. Each symbol ($\cdot$) indicates the relative displacement

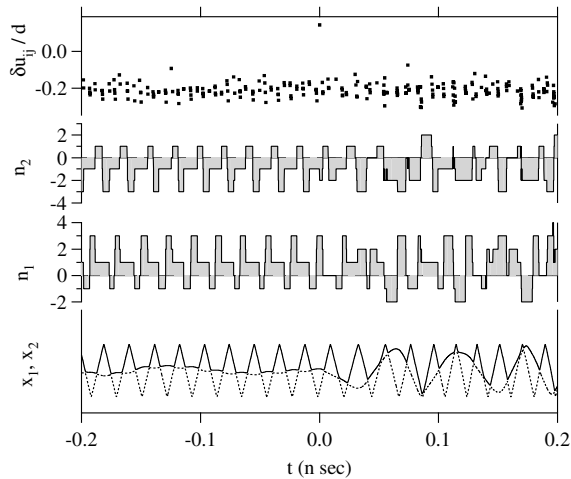


FIG. 4. A transition from periodic to quasiperiodic vibrations. The symbols ($\cdot$) denote a relative displacement divided by the equilibrium distance, $\delta u_{ij}/d$, between the colloid particles or between the colloid particle and electrodes when electron tunneling occurs between them. (top) The tunneling event with positive $\delta u_{ij}/d$ at $t = 0$ is found to cause changes from periodic to quasiperiodic behavior both for the electron numbers n_1 and n_2 (middle) and for the trajectories (bottom).

$\delta u_{ij}/d$ in Eq. (2) for a tunneling event between i and j . The average $\langle \delta u_{ij}/d \rangle$ is -0.21 and the standard deviation is 0.03 , demonstrating the correlation between the electron tunneling and the colloid-particle motion. Hence the electron tunneling event with positive $\delta u_{ij}/d$ at $t = 0$ in Fig. 4 is obviously uncorrelated with the colloid-particle motion, which is referred to as a *shot noise*, hereafter. The shot noise causes irregular changes in the electrostatic force acting on the colloid particles via the change in the n_i s, making the periodic vibrations unstable. This leads to the transition from the periodic trajectories into the quasiperiodic vibrations. Because the current value for the quasiperiodic vibrations is larger than that for the periodic ones, the transition of the vibration modes gives rise to the jump in the current, which appears in the entire voltage region for the hysteresis I - V curves.

The shot noise also induces the transitions from the quasiperiodic vibrations to periodic ones. Figure 5 exhibits a transition from the quasiperiodic vibrations to periodic ones after an electron tunneling event at $t = 0$. The transitions are, however, found only in the narrow voltage region near the lower end of the hysteresis I - V curves, in contrast to the former transitions. Before discussing the difference, one should pay attention to the mechanism for the dip in current at $eV_{sd}/C = 5.5$, which is caused by the transition from quasiperiodic vibrations to periodic ones.

In general, quasiperiodic vibrations are characterized by a set of elemental spectral peaks with incommensurable frequencies, which are f_1 and f_2 in Fig. 3(a), where f_3 and f_4 are the harmonics of f_1 and f_2 , i.e., $f_3 = 3f_1$ and $f_4 = 2f_2$. Frequencies at other subordinate peaks are given by

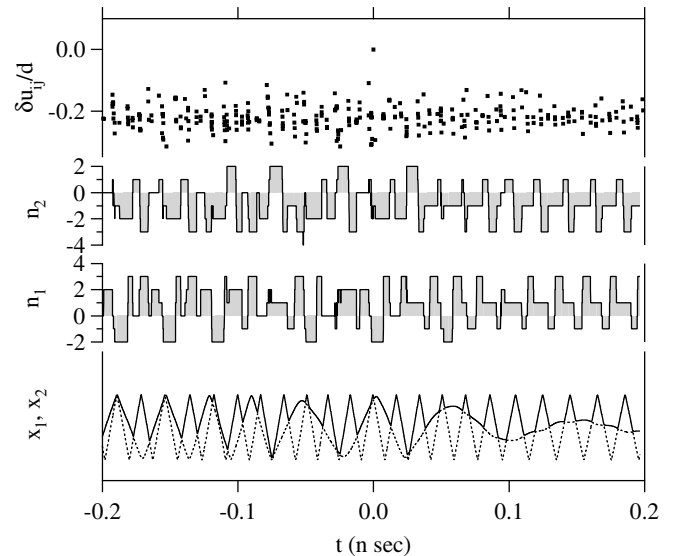


FIG. 5. A transition from quasiperiodic to periodic vibrations. The symbols ($\cdot$) denote $\delta u_{ij}/d$ (top). The tunneling event with almost zero $\delta u_{ij}/d$ at $t = 0$ gives rise to a change from aperiodic to periodic behavior for the number of electrons n_1 and n_2 (middle) and for the trajectories (bottom).

$f = |mf_1 + nf_2|$, where m and n are integers. The ratio f_1/f_2 varies, depending on V_{sd} . They become commensurate, i.e., $f_1 = \frac{2}{3}f_2$, at $eV_{sd}/C = 5.5$, and the peak frequencies, as a result, focus onto a new elemental frequency given by $\frac{1}{3}f_2$ and its harmonics. These new peak frequencies agree well with the frequencies $f_a (= \frac{1}{3}f_2)$, $f_b (= 5f_a)$, and $f_c (= 2f_b)$ of the peaks for the periodic vibrations in Fig. 3(b). Thus, the transition associated with the dip in current is caused by frequency locking [15] of colloid-particle vibrations, in which the shot noise is not concerned.

The commensurate relation among the spectral peaks $f_1 = \frac{2}{3}f_2$ is considered to be maintained during the voltage region from the dip in current to the lower end of the hysteresis I - V curves. However, the commensurate relation does not hold in the voltage region for the hysteretic I - V curves, that becomes a bottleneck for the transitions from the quasiperiodic vibrations to periodic ones owing to the frequency locking. In spite of this, we can consider that the frequency locking is responsible for the RTN in the current. The shot noise disturbs the nanoparticle vibrations, as discussed above, and accordingly the power spectrum fluctuates. Because of the spectral fluctuations, the commensurability is expected to hold for a transient period near the lower end of the hysteresis I - V curves, where the commensurate relation is expected to be approximately satisfied. This commensurability will lead to the transitions from quasiperiodic vibrations to periodic ones. This interpolation agrees with the results of the simulations that the transitions from the quasiperiodic vibrations to periodic ones are actually found only in the limited voltage region. Hence we may conclude that the shot noise causes the transitions from the quasiperiodic vibrations to the periodic ones and also the associated current drops, by inducing frequency locking of colloid-particle vibrations.

Finally, I shall discuss the role of temperature in the RTN. At finite temperature, thermally excited electrons can tunnel in the reverse direction with respect to the bias, with a rate that increases with temperature. The tunnel events disturb the vibrations, which leads to quicker switching of the modes with increase of temperature, and eventually to the vanishing of the periodic vibrations above a certain temperature. The simulations actually show the disappearance of RTN due to the suppression of the periodic vibration mode at electron temperatures higher than 200 K. Here I replaced $\Theta(\Delta\mathcal{F})$ in Eq. (2) by $(1 - e^{-\Delta\mathcal{F}/k_B T})^{-1}$ whereas the thermal fluctuations in the trajectories are not taken into account, and assumed weak coupling to the heat baths. Thus, thermal effects on the RTN are found to be significant.

In conclusion, I have demonstrated, by means of numerical simulations, that shot noise produces RTN in the shuttle current in metal nanoparticle arrays, which appears for the first time in the NEMS of nanoparticle arrays. The distinct feature of the system is that the nanoparticles obey

a set of equations of motion containing Coulomb-interaction terms with different electron numbers. The dynamics is described by a patchwork of orbits connected via the electron tunneling, showing a trajectory winding the *attractors* representing periodic or quasiperiodic vibrations expressed in the language of dynamical systems. The shot noise changes the basins of attraction of the attractors. Here there arise basic questions about the formation and stability of the attractors relevant for describing the typical rate of switching events responsible for the RTN. However, there have been very few reports on such dynamical systems with external inputs based on probability [16,17], and even then the dynamic behavior is not well understood from the theoretical viewpoint. Further theoretical study is necessary to elucidate more general cases, and the present work is expected to stimulate this. Finally, one-dimensional chains of nanometal particles have been actually fabricated by means of templates made of DNA [18] or by the soft lithography technique [8]. Experimental confirmation of RTN is anticipated in such systems.

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