Dynamics of electrons in a double quantum dot biased by an ac voltage: Nonlinear effect of Coulomb interaction

Zhi-Yong Zhang[1] and Shi-Jie Xiong[1,2]
[1]*National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China*
[2]*China Center of Advanced Science and Technology (World Laboratory), P.O. Box 8730, Beijing 100080, China*
(Received 2 July 1997; revised manuscript received 5 November 1997)

We investigate the dynamics of electrons in a double quantum dot biased by an ac voltage. For a closed system with two electrons, the exact diagonalization is performed on a many-body basis with the help of the time-evolution operator. We find three singlet states which show different dependences of quasienergies on the strength of Coulomb interaction U. The amplitude, as well as the nonlinear features, of the oscillation of the occupation number in a dot depends on the state and the value of U. [S1063-651X(98)03603-4]

PACS number(s): 41.20.−q, 41.90.+e, 73.50.Pz, 73.20.Dx

Coulomb interaction plays an important role in the electronic properties of mesoscopic systems. By neglecting the many-body interaction, electrons always fill up the lowest single-electron energy levels of the system, and the intensity of the tunneling current is determined by the relative offset between the Fermi energy of the leads and the levels of the system. If the interaction of the electrons is strong enough, single-electron states are no longer a good description of the system, and the electrons occupy a many-body state as a whole. The number of electrons in the dots constantly changes during the tunneling process, and, consequently, the many-body state occupied by the electrons also varies. This many-body effect in the tunneling processes was investigated via the mean-field approximation (MFA) for the double-barrier structure [1,2] and the superlattices (SL's) [3–8]. Originating from the nonlinear nature of the dependence of the system energy on the electronic charging, the bistability and multistability were found in the *I-V* characteristics [3,4,8]. This nonlinear effect was used to explain the formation of the high-field domain [3,4,8] and the self-sustained oscillating current in SL's at low doping density [5]. Chaos was also predicted theoretically and then confirmed experimentally in these systems [6,7].

The quantum dot presents another excellent type of system to study the effect of the Coulomb interaction [9,10]. Since the electron number in an isolated quantum dot is smaller than that in the large area quantum well, the MFA treatment for the Coulomb interaction becomes poor because of the strong fluctuations. On the other hand, if the applied voltage is not large enough to excite the system to the state of $N+1$ electrons, which has much higher energy owing to the strong interaction, then none of electrons can tunnel through the dot. This phenomenon is known as the Coulomb blockade [11,12]. Nevertheless, the Coulomb blockade can still be lifted in special environment due to the quantum tunneling effect [12]. Based on the same reason, single electron tunneling can be realized by modulating the applied voltage.

At the same time, photon-assisted tunneling (PAT) in the quantum dot was also studied [13–16]. In this case, owing to the formation of the virtual states, electronic tunneling can be obtained in the Coulomb blockade regime. Because of the existence of ac bias voltage, PAT is a nonequilibrium process in nature. Recently, Stafford and Wingreen [17] and Stoof and Nazarov [18,19] investigated resonant PAT through a double quantum dot with Rabi spatial oscillation.

Here we investigate the dynamics of electrons in a closed double quantum dot biased by an ac voltage. We construct a many-body basis for two electrons, and perform the exact diagonalization with the help of the time-evolution operator [20]. For different values of the on-site Coulomb interaction U, quasienergies are calculated, and the oscillations of the electron number on each dot are obtained. We find three triplet states and three singlet states. The triplet states are trivial ones with zero quasienergy and constant occupation number 1 on each dot due to the Pauli principle. The three singlets belong to two categories. The first category has two states, for which with U increased the amplitude of oscillations of the occupation number diminishes and the nonlinearity grows. The second category has only one state for which the amplitude increases first and then decreases by increasing U. These results may shed light on the nonlinear behavior of a double-dot system exposed to the radiation.

For simplicity, we suppose there is just one energy level on each dot. Thus, the Hamiltonian of the system is

$$H(t)=\sum_{\sigma,k=1,2}\epsilon_k(t)C_{k\sigma}^{\dagger}C_{k\sigma}+\tilde{T}\sum_{\sigma}\left(C_{1\sigma}^{\dagger}C_{2\sigma}+C_{2\sigma}^{\dagger}C_{1\sigma}\right)+U\sum_{k}n_{k\uparrow}n_{k\downarrow}+W\sum_{\sigma_1,\sigma_2}n_{1\sigma_1}n_{2\sigma_2}. \tag{1}$$

Here $C_{k\sigma}^{\dagger}$ ($C_{k\sigma}$) creates (destroys) an electron with spin σ on the kth dot. When biased by an ac voltage, the energy levels on the dots become $\epsilon_k(t)=(-1)^{k+1}(\Delta+\tilde{\epsilon}\cos\omega t)/2$, where Δ is the detuning between the two dots, $\tilde{\epsilon}$ the amplitude of the ac voltage, and ω the driving frequency with period T. $\tilde{T}$ describes the transition between the two dots, and U and W are the on-site and interdot Coulomb interaction, respectively.

To construct a many-body basis, we suppose there are two electrons in the system; then the Hamiltonian can be written as a 6×6 matrix:

$$\hat{H}(t)=\begin{pmatrix} W & & & & & & \\ & W & & & \tilde{T} & & \tilde{T} \\ & & W & & \tilde{T} & & \tilde{T} \\ & & & W & & & \\ & \tilde{T} & \tilde{T} & & U+\Delta+\tilde{\epsilon}\cos\omega t & & \\ & \tilde{T} & \tilde{T} & & & U-\Delta-\tilde{\epsilon}\cos\omega t & \end{pmatrix}, \tag{2}$$

with $|11\rangle$, $|1\,\overline{1}\rangle$, $|\overline{1}\,1\rangle$, $|\overline{1}\,\overline{1}\rangle$, $|20\rangle$ and $|02\rangle$ as the six basis vectors, where $|mn\rangle$ denotes the state of m electrons on the first dot and n electrons on the second dot, and the values 1 and $\overline{1}$ indicate the up-spin and down-spin, respectively. When the ac voltage is *not* applied, the eigenenergies can be calculated from the equation

$$(W-E)^3[(W-E)(U+\Delta-E)(U-\Delta-E)-4\tilde{T}^2(U-E)] =0.$$

Generally, we have three triplet states and three singlet states. $(1\ 0\ 0\ 0\ 0\ 0)^T$, $(0\ 0\ 0\ 1\ 0\ 0)^T$ and $(0\ 1\ -1\ 0\ 0\ 0)^T$ (neglecting the normalization coefficient) form the triplet subspace. The time-dependent terms do not mix the triplet states with the singlet ones. In triplet subspace the electron number on each dot is invariably one. These states remain triplet when time-dependent terms are added. On the other hand, the eigenstates in the singlet subspace are changed by the time-dependent terms. Thus in what follows we will pay attention to the singlet subspace.

Since $\hat{H}(t)$ is a periodic function of time, the eigenfunction is of the Bloch form $e^{-iEt}u(t)$, with E the quasienergy and $u(t+T)=u(t)$ (here we have set $\hbar=1$). Introducing the time-evolution matrix $\hat{U}(t,t')$ [17,20], we have that, on the basis of the above Bloch form, $\hat{U}(T,0)$ is diagonal with eigenvalue e^{-iET}. Thus using the initial condition $\hat{U}(0,0)=\hat{I}$, we can integrate the equation

$$i\frac{\partial}{\partial t}\hat{U}(t,0)=\hat{H}(t)\hat{U}(t,0), \tag{3}$$

and diagonalize $\hat{U}(T,0)$ to obtain the quasienergies E_α and the Floquet states $u_\alpha(t)$ in the whole period. Then we can calculate the variation of the electron number on the kth dot when the system is on the αth Floquet state:

$$\langle u_\alpha(t)|\sum_\sigma n_{k\sigma}|u_\alpha(t)\rangle. \tag{4}$$

For the triplet states the quasienergy is zero, and the occupation number on each dot is always one. For the singlet states, we present the quasienergies versus U with $W=\Delta=0$ in Fig. 1. Owing to the time periodicity the quasienergies are restricted within the first Brillouin zone $[0,\omega)$. There are three bands of quasienergies indicated with triangles, open squares, and closed squares. At $U=0$, which is not shown in the figure due to the logarithmic axis, the corresponding quasienergies are 0.5041, 0.4958, and 0. When U is increased from zero, the curves all grow up. If the quasienergy reaches the upper boundary of the zone, ω, it jumps to the lower boundary 0, and the curve is broken (note that $\omega \bmod \omega=0$). When U is small the triangle and open-square curves are almost degenerate; their first breaking point is near $U=0.7$, and after that they are separated. The second breaking point of the open-square curve and the first breaking point of the triangle curve are both near $U=2$. When U is large the curve of triangles approaches the upper boundary, while the curves of open and closed squares approach the lower one. In Figs. 2 and 3 we plot the oscillations of the occupation number of the first dot over one driving period for the states which are in the triangle and open-square curves and marked by $+$ and $\times$ symbols in Fig. 1, respectively. In these figures, it can be seen that if $U=0$ the electrons undergo the Rabi oscillation around the average value 1 with amplitude 1, corresponding to the one-electron picture. When U is increased, the oscillations change in two ways: First, the amplitude of the oscillation diminishes since the on-site Coulomb repulsion prevents the two electrons occupying the same dot simultaneously. Second, the oscillation pattern deviates from the standard sinusoidal shape and the nonlinearity appears. In Fig. 4, the same figure is pre-

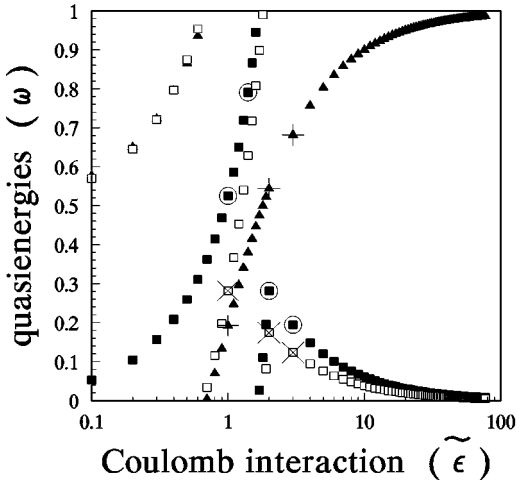

FIG. 1. The variation of quasienergies vs U for the singlet states in a close system. $\tilde{\epsilon}$, the unit of U, and ω, the unit of the quasienergies, are the amplitude and frequency of the applied ac field, respectively. $\tilde{T}=\frac{1}{2}$.

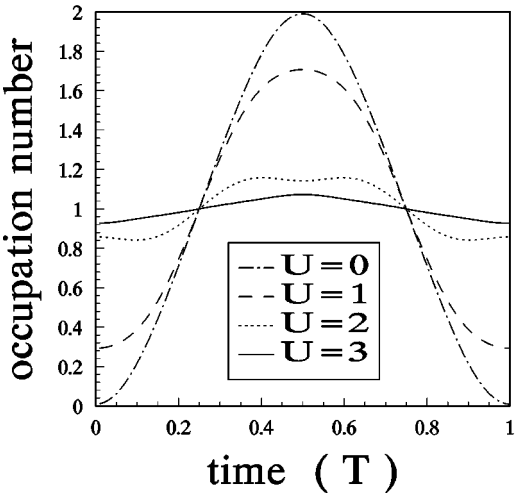

FIG. 2. The oscillation of the occupation number in the first dot within one driving period for the states marked by + symbols in Fig. 1, and the state with $U=0$ and $E=0.5041$.

sented for the states marked by circles in the closed-square curve of Fig. 1. We recall that at $U=0$, the quasienergy of this curve is zero, and is degenerate with the triplet states. This corresponds to a constant occupation number on one dot and zero amplitude of oscillations. When U increases the degeneracy is removed, and as a result the oscillation amplitude increases, as opposed to the states corresponding to the triangle and open-square curves of Fig. 1. If U is further increased and becomes larger than 2, the breaking point for this curve, the amplitude decreases with increasing U, the same behavior as in Figs. 2 and 3. Remarkably, the maximum oscillation corresponds to the breaking point, $U=2$, and the nonlinearity becomes larger by decreasing U in the range $U<2$.

The different features of these states may lead to bistability or multistability in the PAT process of the double-dot system. At $U=0$ there are two types of states: one has a constant occupation number on each dot, and the other shows a Rabi oscillation. The former includes three triplet

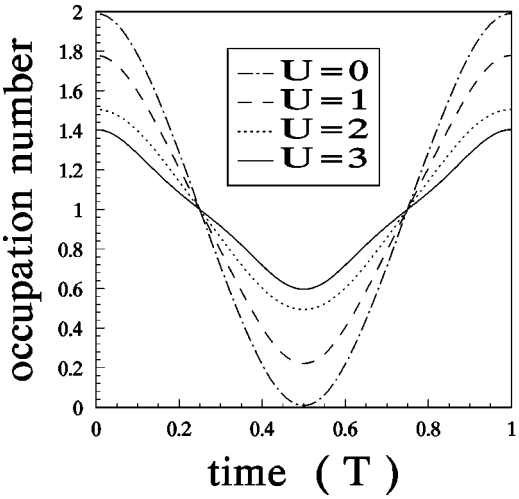

FIG. 3. The same as in Fig. 2 but for the states marked by $\times$ symbols in Fig. 1 and the state with $U=0$ and $E=0.4958$.

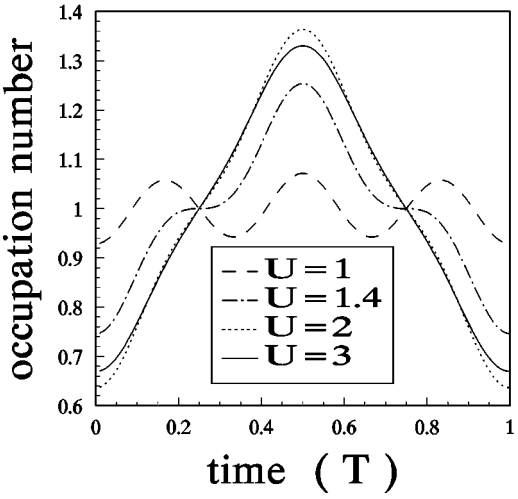

FIG. 4. The same as in Fig. 2, but for the states marked by open circles in Fig. 1.

states and one singlet state with degenerate quasienergies, and the latter corresponds to the other two singlet states. By increasing U the triplet states remain unchanged but the singlet states show complicated oscillations with different U dependences of the amplitude and nonlinearity. This provides a possible origin for the multiple nonlinear behavior in tunneling processes.

In the above four figures, we do not consider the interdot Coulomb interaction W and the detuning Δ. This corresponds to the case where the two dots are separated far enough and they are identical. When the two dots are close to each other, W cannot be omitted. Note that W is small compared to U in ordinary situations, and its effect is the opposite of that of U, so the essential features of the quasienergies and the oscillations in the occupation number are not changed. When Δ is included, the change is that the average of the occupation number is no longer 1, because the spatial symmetry is broken by the detuning.

As a summary, we studied from a strict many-body scheme, the influence of Coulomb interaction on the quasienergies and Rabi oscillations for a closed double-dot system with two electrons. With the help of the time-evolution operator, the quasienergies and eigenvectors are obtained by exact diagonalization of a 6×6 Hamiltonian. The system has three triplet states and three singlet states. For the triplet states the occupation number on each dot is trivially 1, and the quasienergy is zero. For the singlet states the electrons undergo spatial oscillations. For two of the singlet states the amplitude of the oscillations diminishes, and the oscillating curve deviates from the standard sinusoid shape when the interaction strength U increases. For the third one the quasienergy is degenerate with the triplet states, the occupation number per dot is constant when $U=0$, and the amplitude of the oscillations of the occupation number first increases and then decreases as U increases. Possible consequences of these states are multistability and nonlinearity in the PAT process of the system.

This work was supported by the National Natural Science Foundation of China.

[1] V. J. Goldman, D. C. Tsui, and J. E. Cunningham, Phys. Rev. Lett. **58**, 1256 (1987).
[2] J. Iñarrea and G. Platero, Europhys. Lett. **33**, 477 (1996).
[3] H. T. Grahn, R. J. Haug, W. Müller, and K. Ploog, Phys. Rev. Lett. **67**, 1618 (1991).
[4] A. Wacher, M. Moscoso, M. Kindelan, and L. L. Bonilla, Phys. Rev. B **55**, 2466 (1997).
[5] J. Kastrup, R. Hey, K. Ploog, H. T. Grahn, L. L. Bonilla, M. Kindelan, M. Moscoso, A. Wacker, and J. Galán, Phys. Rev. B **55**, 2476 (1997).
[6] O. M. Bulashenko and L. L. Bollina, Phys. Rev. B **52**, 7849 (1995).
[7] Y. Zhang, J. Kastrup, R. Klann, K. H. Ploog, and H. T. Grahn, Phys. Rev. Lett. **77**, 3001 (1996).
[8] R. Aguado, G. Platero, M. Moscoso, and L. L. Bonilla, Phys. Rev. B **55**, R16 053 (1997).
[9] O. Agam, Ned S. Wingreen, Boris L. Altshuler, D. C. Ralph, and M. Tinkham, Phys. Rev. Lett. **78**, 1956 (1997).
[10] Ya. M. Blanter, A. D. Mirlin, and B. A. Muzykantskii, Phys. Rev. Lett. **78**, 2449 (1997).
[11] D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids*, edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (North-Holland, Amsterdam, 1991).
[12] L. I. Glazman and K. A. Matveev, Zh. Éksp. Teor. Fiz. **98**, 1834 (1990) [Sov. Phys. JETP **71**, 1031 (1990)].
[13] L. P. Kouwenhoven, S. Jauhar, J. Orenstein, P. L. McEuen, Y. Nagamune, J. Motohisa, and H. Sakaki, Phys. Rev. Lett. **73**, 3443 (1994).
[14] C. Bruder and H. Schoeller, Phys. Rev. Lett. **72**, 1076 (1994).
[15] A. Johnson, L. P. Kouwenhoven, W. de Jong, N. C. van der Vaart, C. J. P. M. Harmans, and C. T. Foxon, Phys. Rev. Lett. **69**, 1592 (1992).
[16] T. H. Oosterkamp, L. P. Kouwenhoven, A. E. A. Koolen, N. C. van der Vaart, and C. J. P. M. Harmans, Phys. Rev. Lett. **78**, 1536 (1997).
[17] C. A. Stafford and Ned S. Wingreen, Phys. Rev. Lett. **76**, 1916 (1996).
[18] Yu. V. Nazarov, Physica B **189**, 57 (1993).
[19] T. H. Stoof and Yu. V. Nazarov, Phys. Rev. B **53**, 1050 (1996).
[20] M. Holthaus and D. Hone, Phys. Rev. B **47**, 6499 (1993).