Coherent control of double-dot molecules using Aharonov-Bohm magnetic flux

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Bonding and antibonding states of artificial molecules have been realized in experiments by directly coupling two quantum dots. Without a direct coupling between two nearby quantum dots, here we show that under a very unusual condition (i.e., a large asymmetrical couplings to the leads at a large bias) continuous coherence control of double-dot charge states can be achieved by changing the flux through a double-quantum-dot Aharonov-Bohm (AB) interferometer. Using magnetic flux to control double-dot molecular-state coherence is very robust against charge noise. We explicitly present the flux-dependent real-time processes of molecular-state formation. In contrast with the transport current, which has a 2π period, the quantum state of the double-quantum-dot molecule has a 4π period in the AB flux.

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I. INTRODUCTION

It is important to tailor quantum states, especially, to control the coherent phase between two superposition states. In the past decades, artificial atoms and molecules in solid-state systems, such as superconducting Josephson junctions, and semiconductor quantum dots (QDs) have provided novel platforms for exploring such quantum-coherent effects. Due to the tunability of various electronic couplings, double-quantum-dot (DQD) systems, which are archetypes of artificial molecules, have attracted considerable attention because they can serve as qubit systems for quantum information processing. The two DQD charge states with one electron residing in either of the two dots are defined as the computational basis of the DQD charge qubit. Coherence arising from superpositions between such charge qubit states, called DQD molecular states in this paper, is of primary interest. It can be achieved by directly coupling the two quantum dots. The tunability of such direct interdot coupling has been experimentally demonstrated and the charge coherence has also been observed in various experiments. However, the tunnel coupling between the charge states is implemented via voltage gates, which is susceptible to charge noises.

In this paper, we show that for an uncoupled DQD in an Aharonov-Bohm (AB) interferometer, by solely tuning the AB flux, the coherent control of the DQD charge states can be realized. Controlling the charge coherence via magnetic fields could circumvent charge noise. Magnetic fluxes have been utilized to investigate the coherence of electron transport through a single QD in AB interferometers. Combining an interdot tunnel coupling with a magnetic flux has also been studied theoretically and experimentally. In particular, controlling the DQD molecular states through AB phases has been of recent experimental interest. Besides the charge noise induced from voltage gates, coupling the DQD to electrodes also causes decoherence. Although the tunnelings to the electrodes may be turned off to avoid the electron-reservoirs-induced decoherence, such tunnelings are indispensable for the AB effect. Thus, controlling the charge coherence of the uncoupled DQD through the AB flux is very robust against charge noises but it still faces the challenge of the electron-reservoir-induced decoherence.

II. MODEL SYSTEM AND ITS EXACT SOLUTION

To focus on the influence of the AB flux on the quantum state of the artificial molecule we consider only polarized noninteracting electrons. The total Hamiltonian of the system is conventionally given by $H = H_L + H_E + H_T$, in which $H_L = \sum_{\alpha} \epsilon_{\alpha} \eta_{\alpha}^{\dagger} \eta_{\alpha}$ describes an uncoupled DQD and $H_E = \sum_{\alpha \beta} \epsilon_{\alpha \beta} \eta_{\alpha}^{\dagger} \eta_{\beta}$ is the Hamiltonian for the leads with $\alpha = L(R)$.
and fluctuations induced by the tunnel coupling to the quantum dots in an Aharonov-Bohm interferometer.

FIG. 1. (Color online) Schematic diagram of a pair of uncoupled quantum dots in an Aharonov-Bohm interferometer.

The tunneling amplitudes harbor the applied magnetic flux $\Phi$ via $V_{jL} = \tilde{V}_{jL} e^{-i\phi_{jL}}$ and $V_{jR} = \tilde{V}_{jR} e^{i\phi_{jR}}$ with the relation $\phi_{jL} - \phi_{2L} + \phi_{1R} - \phi_{2R} = \phi \equiv 2\pi \Phi/\Phi_0$, where $\phi = 2\pi \Phi/\Phi_0$ and $\Phi_0 = h/e$ is the flux quantum. The line widths induced by tunneling are then given by $\Gamma_{ij} = 2\pi |V_{jL}|^2 \delta_{ij}$, where $\delta_{ij}$ is the density of states in the lead $i$. The DQD molecular states described in terms of the reduced density matrix are governed by the following exact master equation:

$$\frac{d}{dt} \rho(t) = -i[H_0, \rho(t)] + \sum_{i \alpha} \left[ L_{ia}^+(t) + L_{ia}^-(t) \right] \rho(t),$$

where $L_{ia}^\pm(t)$ are the superoperators describing the dissipations and fluctuations induced by the tunnel coupling to the electrodes (for details see Ref. 26). Denoting the state of the empty DQD by $|0\rangle$, one electron on the first and the second dots by $|1\rangle$ and $|2\rangle$, respectively, and the state of both dots occupied by $|3\rangle$, the density matrix $\rho(t)$ can be generally expressed as

$$\rho(t) = \begin{pmatrix} \rho_{00}(t) & 0 & 0 & 0 \\ 0 & \rho_{11}(t) & \rho_{12}(t) & 0 \\ 0 & \rho_{21}(t) & \rho_{22}(t) & 0 \\ 0 & 0 & 0 & \rho_{33}(t) \end{pmatrix},$$

where $\rho_{ij} = \langle i | \rho | j \rangle$ with $i, j = 0, 1, 2, 3$. The QD molecular state, featured as one electron in the QD shared between the two charge states of the DQD molecule, is embedded in the central $2 \times 2$ block matrix of Eq. (2).

To see how molecular states in this QD are formed in time, we solve the master equation (1) with the initial preparation of the empty QD, namely, $\rho_{00}(0) = 1$ and $\rho_{ij}(0) = 0$, for all $i \neq 0$, $j \neq 0$. The exact solution from the master equation for each matrix element gives

$$\begin{align*}
\rho_{11}(t) &= v_1(t) - \det \psi(t), \\
\rho_{22}(t) &= v_2(t) - \det \psi(t), \\
\rho_{12}(t) &= v_1(t), \\
\rho_{21}(t) &= v_2(t), \\
\rho_{00}(t) &= \det[I - \psi(t)], \\
\rho_{33}(t) &= \det \psi(t),
\end{align*}$$

with $I$ being an identity matrix and

$$\psi(t) = \int \frac{d\omega}{2\pi} \omega(t, \omega) \sum_a f_a(\omega) \Gamma_a \left( \frac{1}{e^{\pm\phi/2} - 1} \right) u(t, \omega)$$

is a $2 \times 2$ hermitian matrix, where $f_a(\omega)$ is the Fermi distribution function of the reservoirs, the upper (lower) sign is for $\alpha = L$ ($R$), and $u(t, \omega) = \int d\tau e^{i\omega(t-\tau)} u(\tau)$ with

$$u(\tau) = \exp \left[ \left( \frac{1}{e^{iE_1 + \Gamma} - 1} \right) + \left( \frac{1}{e^{iE_2 + \Gamma} - 1} \right) \right] r.$$

Here we have defined $\Gamma_\alpha(\phi) = \Gamma_\alpha \cos(\phi/2) + i \Delta \sin(\phi/2)$ with $\Gamma_\alpha = \Gamma_L + \Gamma_R$ and $\Delta = \Gamma_L - \Gamma_R$. The functions $u(t)$ and $\psi(t)$ are indeed the spectral and correlation Green functions in the Schwinger-Keldysh nonequilibrium Green function theory.\(^{34}\) The AB flux $\phi$, the coupling asymmetry $\delta \Gamma$, the nonequidensity $\delta E = E_1 - E_2$, and the nonequilibrium dynamics from the electron tunnelings all influence the consequent quantum states of the QDQ molecule.

On the other hand, we can rewrite the QDQ molecular state [i.e., the central block matrix of Eq. (2)] as

$$\rho_{00}(t) = \frac{1}{2} \left[ 1 + r(t) \cdot \sigma \right] - \frac{1}{2} \left[ \rho_{00}(t) + \rho_{33}(t) \right] I,$$

where $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ consists of the Pauli matrices and $r(t) = (r_x, r_y, r_z)$ with

$$r_x = 2\text{Re} \rho_{21}(t), \quad r_y = 2\text{Im} \rho_{21}(t), \quad r_z = 2[\rho_{11}(t) - \rho_{22}(t)],$$

being the polarization vector for the molecular states. So the coherence dynamics of QDQ molecular state formations, described by the off-diagonal matrix element $\rho_{12}$ of the reduced density matrix, can also be visualized through the motion of the polarization vector with the Bloch sphere. Also, the leakage out of the one-electron state space can be easily seen from the term proportional to the probability of the empty and the doubly occupied states, $\rho_{00}(t) + \rho_{33}(t)$.

### III. COHERENT PHASES CONTROLLED BY THE AB FLUX

The coherence between the two charge states of the QDQ molecule is characterized by the off-diagonal element $\rho_{21}$. To develop a clear picture of the coherence of the QDQ molecular state, let us first look at the off-diagonal matrix element $\rho_{12}$ in Eq. (3) in the steady-state limit ($t \gg \Gamma^{-1}$) at zero temperature. The general solution is

$$\rho_{21} = \frac{1}{2} [r_x + ir_y]$$

$$\times \left[ \tan^{-1} \left( \frac{eV}{2\Gamma_+}(\phi) \right) + \tan^{-1} \left( -\frac{eV}{2\Gamma_-}(\phi) \right) \right]$$

$$\times \left[ \frac{\delta \Gamma \cos \phi + i \Delta \sin \phi}{2\Gamma \cos \phi} \right]$$

$$\times \left[ \frac{1}{\tan \left( \frac{eV}{2\Gamma_+}(\phi) \right) - \tan^{-1} \left( -\frac{eV}{2\Gamma_-}(\phi) \right) \right]$$

$$\times \left[ \frac{i}{\Gamma} \left( \Gamma^2 - \delta \Gamma^2 \sin \phi - \frac{\Delta \Gamma}{2} \sin \phi \right) \right],$$

where $\Gamma_\pm = \Gamma_\alpha \pm \delta \Gamma$.
The coherence between the charge states in the AB flux was also included. However, with the larger asymmetry coherence amplitude the state of the DQD molecule is 4 to the molecular basis Eq. (9). By setting the phase is continually driven by the AB flux, as seen from the often-used condition of degeneracy $\delta E = 0$ and symmetric coupling $\delta \Gamma = 0$. Such decoherence-induced localization of the coherent phase hinders the manipulation of the coherent phase of molecular states. Remarkably, when the DQD couples largely asymmetrical to the left and the right leads ($\delta \Gamma \neq 0$), we find that the coherent phase $\varphi$ can be continuously tuned by AB fluxes.

To achieve typical molecular states, the DQD is set at degeneracy ($\delta E = 0$). Then the second term in Eq. (8) vanishes. Equations (3) and (8) show that the formation of molecular states is essentially determined by the applied bias and the coupling asymmetry to the source and the drain. The basic states is essentially determined by the applied bias and the electron states abruptly. Therefore the phase of $\varphi = \pm \pi/2$ (i.e., the phase localization shown in previous work). In the case that $\varphi = 0$ (or $\varphi = 2\pi$), the state $|\pm\rangle$ becomes decoupled from the electron reservoirs, and electrons can only occupy the opposite molecular state $|\mp\rangle$. The corresponding phase of $\rho_{z1}$ is then 0 (or $\pi$). On the other hand, when $\varphi = 0$, the occupation of the antisymmetric state $|\mp\rangle = (|1\rangle - |2\rangle)/\sqrt{2}$ becomes a constant of motion. Turning on the flux $\varphi \neq 0$ breaks such symmetry and consequently changes the electron states abruptly. Therefore $\rho_{z1}$ changes abruptly across $\phi = 0$ as indicated in Eq. (9).

**IV. REAL-TIME PROCESSES OF MOLECULAR-STATE FORMATIONS**

The full information of the quantum state of the DQD molecule at finite temperature is depicted by the time-dependent reduced density matrix. In Fig. 3, we plot the evolution of the full reduced density matrix of the DQD molecule. Initially, the DQD is prepared in an empty state, $\rho_{00}(0) = 1$ as shown by Figs. 3(b1), 3(c1), and 3(d1) and $r(t) = 0$ given in Fig. 3(a1) (where the length of the red strip is zero). After injecting electrons from the left and the right reservoirs, $\rho_{00}$ decreases [see Figs. 3(b1) to 3(b3)] while the electron occupation and coherence increase with time [see Figs. 3(b1) to 3(b3), Figs. 3(c1) to 3(c3), and also Figs. 3(d1) to 3(d3)]. The coherent phase $\varphi$ between the charge states has been fixed shortly after the electron injection [see Figs. 3(a1) to 3(a4) and also Figs. 3(d2) to 3(d4)]. Then $|r(t)|$ grows in time with fixed $\varphi$, and finally a stable molecular state, $\rho \approx |\psi\rangle\langle\psi|$, where $|\psi\rangle = (|1\rangle + e^{-i\phi/2}|2\rangle)/\sqrt{2}$, is reached in a short time.
FIG. 3. (Color online) Typical process for forming molecular states. The dashed black line in (a1) to (a4) is the trajectory taken by \( r(t) \) from \( t = 0 \) in (a1), starting from the origin, to \( t = 3\Gamma^{-1} \) in (a4), where it almost touches the surface of the sphere. The red strip in each plot is the trajectory up to the corresponding time points, as shown above the spheres. From the trajectory, we see the coherent phase \( \phi \) [which is the angle made by \( r(t) \) with \( r_x \) axis] has been fixed after the electron is injected into the DQD. Plots (b1) to (b4) display the real part of the reduced density matrix of the DQD system, while the imaginary part is plotted in (c1) through (c4). The coherent phase \( \phi \) between the two charge states is better visualized through the vector plots (d1) to (d4). Every arrow represents an element of the reduced density matrix \( \rho_{ij} \), whose horizontal projection stands for the real part and the vertical projection stands for the imaginary part. The AB flux here is \( \phi = -\pi/2 \).

Note that due to possible leakage, see Fig. 3(b3) where \( \rho_{00} \) has a small finite value, the DQD is not in a perfect pure state. But the situation can be optimized by changing the bias and the coupling asymmetry, as shown by Eq. (9).

To better understand the role played by the AB flux, we show the time evolutions of \( \rho_{21} \) in Fig. 4 under various values of \( \phi \). Figures 4(a1) and 4(a2) show the process of coherence generation for a strong asymmetric coupling \((\delta / \Gamma = 0.91\Gamma)\). The rate of approaching steady-coherent-molecular states is only weakly dependent on the flux. The stable molecular states are soon reached after a few \( \Gamma^{-1} \). This is totally different from the symmetric coupling \((\delta / \Gamma = 0)\), as shown by the authors of Refs. 33 and 35, where \( \text{Re} \rho_{21} \) displays the severe flux-dependent decays due to the decoherence induced by the large electron transport in the symmetric coupling. Therefore, the larger coupling asymmetry can strongly suppress the decoherence from the electron transitions, and makes the coherence control of the QDQ molecule feasible.

FIG. 4. (Color online) The full coherence time evolutions of \( \rho_{21} \), (a1) and (a2), in the large asymmetrical coupling \((\delta / \Gamma = 0.91\Gamma)\), compared with the phase localization, (b1) and (b2), in the symmetrical couplings \((\delta / \Gamma = 0)\), the letter was shown in our previous work (Ref. 33), as well as in Ref. 35.

V. DISCUSSIONS

The general solution shows that the quantum state of the DQD molecule has a period of \( 4\pi \) in the AB flux. It is an
intrinsic property of this pseudospin system, independent of the coupling geometry and the bias configurations. Besides, we can calculate the tunneling current to reservoir $\alpha = L, R$ within the same framework: $I_\alpha(t) = \epsilon \sum_\omega \text{tr}_i \{ \mathcal{L}^{\alpha}_\omega(t) \rho(t) \}$. The steady-state transport current $I = \frac{1}{2}(I_L - I_R)$ has been calculated

$$ I(\phi) = \int \frac{d\omega}{2\pi} \left[ f_L(\omega) - f_R(\omega) \right] T(\omega, \phi), \quad (11) $$

where the transmission coefficient is given by

$$ T(\omega, \phi) = \frac{(1 - \delta \Gamma^2) \left[ \frac{\omega^2 \cos^2 \frac{\phi}{2} + \frac{1}{2} \delta E \sin^2 \frac{\phi}{2}}{\omega^2 + \Gamma_1^2(\phi)} \right]}{[\omega^2 + \Gamma_1^2(\phi)] [\omega^2 + \Gamma_2^2(\phi)]}. \quad (12) $$

By taking $\delta \Gamma = 0$ it reproduces the result of the authors of Ref. 28. Equation (12) clearly shows that the transport current has a period in the AB flux of $2\pi$. This $2\pi$ period, as a feature for the coherence of transport, is well known and has been observed in experiments. A nontrivial character of the quantum state of the DQD molecule, requires further experimental investigation. Note that although the coherent phase of the off-diagonal density matrix element is gauge dependent, the AB flux dependence of the coherence phase and its periodicity are both independent of the gauge choice.

In summary, we have demonstrated the unusual conditions for the coherence control of DQD artificial molecules using AB fluxes. We have analyzed the AB flux-dependent coherence controlling through the exact solution of the master equation. When a large bias is applied with a strong asymmetry in couplings to the source and the drain, coherent control by the AB flux can be achieved. The decoherence induced by the electron tunnelings can be efficiently suppressed. We also find that the period of the quantum state of the DQD molecule in the AB flux is $4\pi$, in contrast with the $2\pi$ periodicity of the transport current. The revelation of the underlying quantum coherence of the molecular states is thus beyond the usual transport measurement. The verifications of these molecular states would rely on a suitable quantum-state-tomography protocol for further investigations. We hope that this theory for DQD molecules could inspire new experiments on coherence control of molecular states via AB fluxes, and become also useful for the quantum emulation of artificial molecular processes.

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36 When $\phi = 0$, the amplitude enhancement of the coherence is determined by the sign of $\delta \Gamma$. When $\mu_L > \mu_R$, the asymmetry of $\Gamma_L > \Gamma_R$ is preferred for larger $\rho_{21}$.