The ferromagnetism intrinsically origins from the spin-independent Coulomb interaction and the Pauli exclusion principle (PEP), as initially proposed by Heisenberg [1]. The Hubbard model [2], which includes both two elements with on-site electron-electron ($e-e$) interaction $U$, is regarded as the minimal model for ferromagnetic (FM) states. Unfortunately, it has not been well addressed whether the Hubbard model has a general FM phase, except under some special conditions[3–6]. The Hartree-Fock approximation once predicted an itinerant Stoner-like FM phase [7], but we now know that the mean-field theory deduces incorrect results and the FM region has been overestimated [3]. Besides the Hubbard model, the Anderson (multi)-impurity model [8] may act as another minimal model for magnetic phase in a bottom-up fashion, with the advantage of implementation simplicity in quantum dots (QDs). For example, the antiferromagnetic (AFM) correlation $J_{AF}$ due to nearest-neighbour electron hopping or tunneling $t$ ($J_{AF} \sim 4t^2/U$) has been well understood experimentally in series-coupled double QDs (SDQDs) [see Fig.1(a)] [9]. Theoretically, $J_{AF}$ is responsible for the AFM ground state at half filling in the Hubbard model, while it induces the spin singlet competing with the Kondo singlet at temperature $T < T_K$ ($T_K$ being the Kondo temperature) in the Anderson two-impurity model [9,13].

Does there exist a FM phase in the Anderson two-impurity model or in SDQDs? That issue may help to understand Heisenberg’s original idea and to determine the FM phase in various strongly correlated models. Please be noted that the sign-indefinite Ruderman-Kittel-Kasuya-Yosida (RKKY) magnetic order, whose implementation must through a third mediated dot in experiments[14], is not our concern here. What we are seeking is a stable FM phase strong enough to compete with the AFM one in SDQDs, which has not been explicitly determined yet in the phase diagrams of SDQDs [9] and other two-impurity systems [14].

The FM phase in SDQDs also has great application potential in solid-state quantum computing. QDs-based spin qubit is one of the most possible physical realization of scalable qubit put forward so far, which has been extensively studied in last two decades [16,17] since its original proposal in SDQDs [18]. It has the advantages of fast operation and long coherence times but the disadvantage of seriously dependence on magnetic fields. The technical difficulties caused by magnetic fields are transparent: (i) The localized oscillating magnetic fields required in qubit or quantum gate manipulation are very hard to realize in practice; (ii) The Zeeman energy is an inefficient way to control spin states; and (iii) The magnetic fields are incompatible with present large-scale integrated circuit. If a stable FM phase in SDQDs does exist, these difficulties may be overcome by possible magnetic-field-free manipulations.

In the present work, by nonperturbatively solving the nonequilibrium Anderson two-impurity model, we will firstly verify no FM phase in the range of parameters investigated under the equilibrium condition in SDQDs. Then, we will report a robust FM phase under nonequilibrium conditions at finite bias and detuning energy, which are strong enough to suppress the AFM phase in the strongly correlated limit ($t \ll U$). We will demonstrate that the FM exchange interaction origins from the passive parallel spin arrangement caused by the PEP during the electrons transport [see Fig.1(b)]. The FM phase is the effect of PEP on magnetic properties, beyond the current collapse in DQDs, another effect of PEP called Pauli spin blockade [19–21]. At large $t$, the AFM phase keeps stable, which defines a tunnel-barrier control of spin states through the FM–AFM transition in SDQDs, similar to the initial proposal in Ref. [15] but no magnetic field (or auxiliary FM-dots) needed any more.

The SDQDs we study here can be described by the
nonequilibrium Anderson two-impurity model. The total Hamiltonian reads

$$H_{\text{total}} = H_s + H_{\text{res}} + H_{\text{sys-res}},$$

where the isolated QD part is

$$H_s = \sum_{\alpha, \delta} \epsilon_{i, s} \hat{c}_{i, s}^\dagger \hat{c}_{i, s} + \frac{U}{2} \sum_{i, s} \hat{n}_{i, s} \hat{n}_{i, s} + t \sum_{\alpha, \delta} (\hat{c}_{i, s}^\dagger \hat{c}_{2, s} + \text{h.c.}),$$

(1)

where $\hat{c}_{i, s}^\dagger$ ($\hat{c}_{i, s}$) is the operator that creates (annihilates) an $s$-spin ($s = \uparrow, \downarrow$) electron with energy $\epsilon_{i, s}$ in the dot $i$ ($i = 1, 2$). $\hat{n}_{i, s} = \hat{c}_{i, s}^\dagger \hat{c}_{i, s}$ corresponds to the $s$-spin electron number operator of dot $i$. As mentioned above, $U (U = U_1 = U_2)$ is the on-dot Coulomb interaction between $s$- and $\uparrow$-spin electrons ($s$ being the opposite spin of $s$), and $t$ is the interdot coupling strength.

The Hamiltonians of reservoirs are

$$H_{\text{res}} = \sum_{\alpha, \delta} \epsilon_{\alpha, \delta} \hat{c}_{\alpha, \delta}^\dagger \hat{c}_{\alpha, \delta},$$

(2)

where $\hat{c}_{\alpha, \delta}^\dagger$ ($\hat{c}_{\alpha, \delta}$) denotes the creation (annihilation) operator of an electron in the $s$-spin state in the $\alpha$-reservoir with wave vector $k$. We set the Fermi energy $E_F = \mu_{\text{L}} = \mu_{\text{R}} = 0$ at equilibrium and $\mu_{\text{L}}/e = -\mu_{\text{R}}/e = V/2$ at nonequilibrium. The system-reservoir coupling is

$$H_{\text{sys-res}} = \sum_{\alpha, \delta, \alpha'} \Gamma_{\alpha, \alpha'} \hat{c}_{\alpha, \delta}^\dagger \hat{c}_{\alpha', \delta} + \text{h.c.}.$$

The hybridization function is assumed to be a Lorentzian form

$$J_{\alpha, \delta}(\omega) = \frac{\pi}{2} \sum_{\alpha'} \Gamma_{\alpha, \alpha'} \delta(\omega - \epsilon_{\alpha', \delta}).$$

We adopt the hierarchical equations of motion (HEOM) approach to numerically solve the nonequilibrium Anderson two-impurity model in a non-perturbative fashion. The HEOM can achieve the same level of accuracy as the latest high-level numerical renormalization group (NRG) for both static and dynamical quantities under equilibrium conditions. Under nonequilibrium conditions, the HEOM has many advantages above other approaches in the prediction of dynamical properties. The HEOM formalism is in principle exact and applicable to arbitrary electronic systems, including Coulomb interactions, under the influence of arbitrary time-dependent bias voltage and external fields. The details of the HEOM formalism and the derivation of physical quantities are supplied in Refs. 22, 13 and 30.

The parameters in our calculations are chosen as follows: the on-dot $e - e$ interaction $U = 2.0 \text{ meV}$; the singly occupied energy level $\epsilon_{1, \uparrow} = \epsilon_{1, \downarrow} = -1.0 \text{ meV} + \Delta$ and $\epsilon_{2, \uparrow} = \epsilon_{2, \downarrow} = -1.0 \text{ meV} - \Delta$, where the detuning energy $2\Delta$ can be finely regulated by gate voltages in experiments; the temperature $T = 0.1 \text{ meV}$ unless otherwise noted; the effective bandwidth of the reservoirs $W_L = W_R = W = 4.0 \text{ meV}$ and the reservoir-dot coupling strength $\Gamma_L = \Gamma_R = \Gamma = 0.1 \text{ meV}$. The inter-dot coupling $t$, bias of voltage $V$ and detuning energy $\Delta$ are three main variables in our calculations.

In order to figure out whether there exists a FM state, we calculate the spin-spin correlation function between QD1 and 2:

$$C_{12} \equiv \langle \hat{S}_1 \cdot \hat{S}_2 \rangle - \langle \hat{S}_1 \rangle \cdot \langle \hat{S}_2 \rangle,$$

where $\hat{S}_i$ is the quantum spin operator at dot $i$.

In Fig. 1(c)–(e), we depict the phase diagram at bias $V = 0.5 \text{ mV}$ and (e) $V = 1.0 \text{ mV}$.
cluded. However, the condition for that equation (t ≪ U and D ≪ U/2) makes J_{AF} < 0 impossible, even under nonequilibrium conditions. Thus, the following FM phase can not result from this mechanism. As shown in Fig. 3(c), with increasing t, C_{12} positively increases, and finally an AFM QD-molecule forms in the large t limit [31], as an analogue of hydrogen molecule.

When a positive bias applied, as shown in Fig. 1(d) and (e), our results reveal a FM phase appearing in the region of 0 < t ≪ U and 0.2U < D < 0.7U. In view of the phase changes from Fig. 1(d) to (e), the FM phase can be seen as growing from the AFM background at finite bias. The FM–AFM phase boundary (where C_{12} changing its sign) seems quite smooth with no abrupt phase transition occurring, instead, a continuous crossover behaviour is clearly visible. With increasing bias, the area of FM phase is enlarged and the strength of exchange interaction enhanced, as C_{12} positively increases. In the strongly correlated limit (0 < t ≪ U), the FM phase can only suppress the AFM one and dominate the phase diagram at finite V and D, as shown in Fig. 1(e). However, the AFM molecular state will survive at large t and very small D, which respectively determine the right and bottom boundary of FM phase. If D is too large to destroy the single occupation of any dot, C_{12} will decrease to zero rapidly, which determines the upper boundary. The left boundary is naturally at t ∼ 0. As a comprehensive result, the FM phase forms a closed irregular circle area in the phase diagram, as shown in Fig. 1(d) and (e).

In order to better understand the details of the AFM–FM transition, we theoretically lift the spin degeneracy in QD1 by applying a local magnetic field B_1, with its direction parallel to ↓-spins. B_1 is chosen to be strong enough to push ε_{1↑} much higher than μ_{L} but let ε_{1↓} = −1.0mV + Δ, which can be achieved by simultaneously adjusting the gate voltage on QD1. By fixing t = 0.2 meV and D = 0.75 meV, we calculate both static and dynamical quantities as functions of V and summarize the results in Fig. 2, where Fig. 2(a) depicts some typical static quantities (ε_{1↑} and C_{12}) and Fig. 2(b)–(e) show the spectral functions [A_{1↑}(ω), A_{2↑}(ω) and A_{2↓}(ω)] at V = 0, 0.14 (V_c, AFM–FM phase crossover point), 0.2, 0.5, 1.0 mV, respectively. As a starting point, the AFM phase at V = 0 is clearly shown in Fig. 2(a), where the magnetic moments m_1 ≡ n_{1↑} − n_{1↓} ≈ −n_{1↓} < 0 and m_2 ≡ n_{2↑} − n_{2↓} > 0. Accordingly, the degeneracy of A_{2↑}(ω) and A_{2↓}(ω) is lifted due to the AFM exchange interaction J_{AF}, as shown in Fig. 2(b), where the singly-occupation transition peak of A_{2↑}(ω) is higher than that of A_{2↓}(ω).

Under nonequilibrium conditions, ↓-spin electrons irreversibly flow from L- to R-reservoir through interdot tunneling. During the transport process, the PEP affects both electrical [19,21] and magnetic properties, of which the latter is our focus here. In Fig. 2(a), the continuous crossover from AFM to FM phase is shown in detail. With increasing V, n_{2↓} gradually decreases while n_{2↑} increases, thus m_{2} positively decreases. At V ∼ 0.14 mV, n_{2↑} = n_{2↓} ⇒ m_{2} ∼ 0. As a consequence, C_{12} ∼ 0, which defines an AFM–FM phase crossover point, V_c, as shown in Fig. 2(a). By checking the spectral functions, we find the singly-occupation transition peak of A_{2↑}(ω) almost overlaps with that of A_{2↓}(ω) at V = V_c with a little splitting [see Fig. 2(c)]. With further increasing V at V > V_c, m_{2} becomes to negatively increase and C_{12} positively increase, as shown in Fig. 2(a), thus the FM phase is gradually enhanced. At V ∼ 0.9 mV, both m_{2} and C_{12} reach their saturation values of 0.9 and 0.21, re-
spectively. The continuous increase of $C_{12}$ with a smooth sign change indicates the competition between AFM and FM phases is far from intense.

Fundamentally, finite bias injects ↓-spin electrons from L-reservoir into QD1, followed by interdot tunneling to QD2. In the next step, the PEP prohibits the double occupation of two ↓-spin electrons, and electrons can only flow out through off-resonance cotunneling or many-body tunneling into R-reservoir, both of which produce small current. As shown in Fig.1(b), for electrons in QD2, increasing $V$ and/or $\Delta$ will enhance their inflowing probability and meanwhile decrease their off-resonance outflowing probability. When the former becomes much larger than the latter at $V > V_2$ and $\Delta > 0.2U$, ↓-spin electrons will accumulate within QD2, which induces a positive to negative sign change of $m_2$. As a consequence, the exchange of $S_1$ and $S_2$ produces a FM order characterized by $C_{12} > 0$. The spectral functions shown in Fig.2(d) at $V = 0.2$ mV verifies this FM correlation (although still weak), where the singly-occupation transition peak of $A_{21}(\omega)$ becomes lower than $A_{22}(\omega)$.

With further increasing $V$, the FM exchange interaction becomes stronger. In spectral functions, this trend is represented by the gradually increasing of the singly-occupation transition peak of $A_{22}(\omega)$ and decreasing of that of $A_{21}(\omega)$ [see Fig.2(e)]. At $V > 0.9$ mV, the former reaches its maximum value and the latter almost disappears, as shown in Fig.2(f). By summarizing Fig.2(a)-(f), one can see that the FM phase in SDQDs origins from the passive parallel spin arrangement caused by the PEP during the electrons transport in the presence of $e-e$ interactions. That mechanism is universal, which should play roles in other strongly correlated models including the Hubbard model.

We are now on the position to elucidate the temperature effect, especially the low temperature properties of the FM phase. In what follows, we recover the spin degeneracy in QD1 and fix $V = 1.0$ mV, $t = 0.2$ meV and $\Delta = 0.75$ meV. The dependence of the inverse of magnetic susceptibility $1/\chi$ on temperature $T$ is depicted in Fig.3(a), which shows an unambiguous Curie-Weiss behaviour at high temperature, $\chi = C/(T - T_c)$, with a fitted Curie point $T_c \sim 0.15$ meV ($\sim 1.75$ K). We also find an upward deviation at very low temperature $T < 0.02$ meV, resulting from the Kondo screening of the FM phase at $T < T_K$. Under equilibrium conditions, this kind of $S = 1$ Kondo screening induces a 'singular Fermi liquid state' [33, 35]. Here, some nonequilibrium Kondo features are expected.

The present HEOM approach can not directly determine $T_K$ as NRG does, but it can easily obtain spectral functions and current at sufficient low temperature to elucidate nonequilibrium Kondo characteristics. The HEOM results of $A_{1s}(\omega)$ at current-voltage ($I-V$) curve at $T = 0.01$ meV are respectively shown in Fig.3(c) and (d), where the $I-V$ curve at $T = 0.1$ meV ($T > T_K$) is also shown for comparison. As shown in Fig.3(b), one small Kondo peak is developed at $\omega = \mu_L$ in $A_{1s}(\omega)$, and another developed at $\omega = \mu_R$ in $A_{2s}(\omega)$. It can be seen as the DQD extension of the bias-induced Kondo peak splitting in single QDs [36]. Although the Kondo peaks in $A_{1s}(\omega)$ seem not high in Fig.3(b), their effects are quite significant on both of the magnetic and transport properties. For the latter, the nonequilibrium Kondo resonance assists the electrons transport, which is characterized by the low-temperature current enhancement shown in Fig.3(c), when the FM phase dominates at $V > 0.25$ mV.

In summary, we have theoretically reported a robust ferromagnetic phase under nonequilibrium conditions in series-coupled double quantum dots by nonperturbatively solving the Anderson two-impurity model. The ferromagnetic exchange interaction origins from the passive parallel spin arrangement caused by the Pauli exclusion principle during the electrons transport. The ferromagnetic phase can conduces to understand the Heisenberg’s initial idea of ferromagnetic order. In addition, it also predicts a convenient way to internally control spin states without magnetic field.

The support from the NSFC (Grant No. 11374363) and Research Funds of Renmin University of China...
(No. 11XNJ026) is gratefully appreciated.

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