

## Friedel Sum Rule for an Interacting Multiorbital Quantum Dot

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A generalized Friedel sum rule is derived for a quantum dot with internal orbital and spin degrees of freedom. The result is valid when all many-body correlations are taken into account and it links the phase shift of the scattered electron to the displacement of its *spectral* density into the dot.

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The Friedel sum rule (FSR) is one of the few exact results of solid state physics [1], with a vast range of applications in the fields of scattering theory, magnetic and nonmagnetic impurities in metals [1], Kondo effect [2], and, very recently, coherent transport through quantum dots (QDs) [3] and molecules [4]. This powerful relation connects the charge density  $\varrho(\omega)$  displaced by an impurity or a nano-object, acting as a scattering center in a conductor, with the variation of the phase shift  $\delta_F$  of the scattered wave with respect to the energy  $\hbar\omega$ :

$$\frac{1}{\hbar} \frac{d\delta_F(\omega)}{d\omega} = \frac{\pi}{e} \varrho(\omega). \quad (1)$$

The Friedel phase  $\delta_F$  of Eq. (1), which holds for single-channel elastic scattering only, is linked to the eigenvalue of the scattering matrix  $S$  through the identity  $S = e^{2i\delta_F}$ . In the many-channel case,  $\delta_F$  appearing on the left-hand side of (1) is replaced by  $\text{Tr} \ln S / 2\pi i$  [5].

Recently, Lee [6] and Taniguchi and Büttiker [7] showed the relevance of the FSR in measurements of the transmission phase acquired by an electron passing through a QD embedded in the arm of an Aharonov-Bohm interferometer [8]. These experiments [8,9] allow for both directly measuring the phase shift and arbitrarily controlling the Fermi energy (or, equivalently, the plunger gate voltage of the QD), namely, the two quantities appearing on both sides of (1). Even if the identification of  $\delta_F$  with the transmission phase is unjustified in generic situations [6,7], Aharonov-Bohm interferometry nevertheless paves the way to the direct experimental test of the FSR for a QD whose internal structure, charge, spin, and correlation can be externally controlled. Indeed, the “nonuniversal” behavior of the QD transmission phase in the regime of very few electrons ( $N < 10$ ) suggests that the QD orbital and spin degrees of freedom may play a major role [9].

In real QDs used in interference experiments, single-particle levels have a small energy separation ( $\sim 0.5$  meV), if compared to characteristic Coulomb energies ( $\sim 1-3$  meV) [9], and therefore many of them should be included in any reliable model for electron correlation [10,11]. The FSR is generally believed to hold even in the presence of electron-electron interaction [6]. This was rigorously demonstrated only in two cases: (i) electrons in the metal form a Fermi liquid and the impurity has no

internal degrees of freedom [5]; (ii) the interaction is limited to a localized impurity orbital (Anderson model [12] and its specific extensions to open atomic shells [2,12]). This Letter shows that the FSR must be reformulated in the experimentally relevant case of a multilevel interacting dot. A generalized statement holds, with the spectral density  $\mathcal{N}(\omega)$  of the scattered electron accumulated in the QD replacing the noninteracting density of states  $\varrho(\omega)/e$  appearing in (1). We discuss the relevance of this result for the qualitative understanding of a few puzzling features of the experiment of Ref. [9].

We consider a generic system whose Hamiltonian  $H$  is made of three terms separately describing the multiorbital interacting quantum dot,  $H_{\text{dot}}$ , the conduction electrons in the leads,  $H_{\text{lead}}$ , and the hopping term between dot and leads,  $H_{\text{mix}}$ :

$$H = H_{\text{dot}} + H_{\text{lead}} + H_{\text{mix}}. \quad (2)$$

The conduction electrons, in typical experimental setups, can freely move in a two-dimensional heterostructure [13], according to the Hamiltonian  $H_{\text{lead}} = \sum_{k\sigma} \varepsilon_k n_{k\sigma}$ , where  $n_{k\sigma} = c_{k\sigma}^\dagger c_{k\sigma}$  and  $c_{k\sigma}^\dagger$  creates an electron into the Bloch state of crystal momentum  $k$ , spin  $\sigma$ , and energy  $\varepsilon_k$ . The QD Hamiltonian  $H_{\text{dot}}$  includes both the single-particle and the interacting part:

$$H_{\text{dot}} = \sum_{\alpha i \sigma} \varepsilon_{\alpha i} n_{\alpha i \sigma} + H_{\text{int}}. \quad (3)$$

In order to label the QD orbitals, we here introduce two indices,  $\alpha = 1, \dots, N_{\text{class}}$  and  $i = 1, \dots, N_\alpha$ , respectively. The former index,  $\alpha$ , labels the  $N_{\text{class}}$  irreducible representations of the QD point-symmetry group, while the latter,  $i$ , enumerates the truncated set of  $N_\alpha$  orbitals considered, belonging to the same  $\alpha$  representation. We assume that QD orbitals form also a basis for representing the symmetry of the whole dot + leads system [14]. Typical symmetry groups of realistic devices range from  $D_{\infty h}$  to  $C_{2v}$ , going from circular [10] to elliptic [15] dots, respectively [16]. In Eq. (3),  $n_{\alpha i \sigma} = c_{\alpha i \sigma}^\dagger c_{\alpha i \sigma}$ ,  $c_{\alpha i \sigma}^\dagger$  creates an electron with spin  $\sigma$  in the orbital  $(\alpha, i)$  of energy  $\varepsilon_{\alpha i}$ , and  $H_{\text{int}}$  includes the full intradot Coulomb interaction. Note that  $H_{\text{int}}$  does not commute with  $n_{\alpha i \sigma}$  except if there is either one level only,  $N_{\text{class}} = 1$ ,  $N_\alpha = 1$  (nondegenerate Anderson model), or the Coulomb interaction takes an oversimplified form.

Here the full inclusion of all Coulomb matrix elements in  $H_{\text{int}}$  turns out to be crucial in the following. Finally, the tunneling term  $H_{\text{mix}}$  allows for electron hopping between delocalized Bloch states  $\mathbf{k}$  and confined orbitals  $(\alpha, i)$  via the matrix elements  $V_{k\alpha i}$ :

$$H_{\text{mix}} = \sum_{k\alpha i\sigma} (V_{k\alpha i} c_{k\sigma}^\dagger c_{\alpha i\sigma} + \text{c.c.}) \quad (4)$$

As a first step, we introduce the zero-temperature exact retarded Green function:

$$i\mathcal{G}_{XX'}(t) = \vartheta(t) \langle c_X(t) c_{X'}^\dagger(0) + c_{X'}^\dagger(0) c_X(t) \rangle, \quad (5)$$

where either  $X = \alpha i\sigma$  or  $X = \mathbf{k}\sigma$ , and  $\langle \cdot \cdot \cdot \rangle$  is the average on the interacting ground state  $|\Psi_0\rangle$  of the whole dot + leads system in the Heisenberg representation. We work with the analytic continuation of the Fourier transform of (5) in the complex plane of the energy,  $\mathcal{G}_{XX'}(z)$ .

In the following, we neglect spin-flip scattering processes [12], and therefore we may drop spin indices of  $\mathcal{G}$ . Moreover, off-diagonal Green functions of type  $\mathcal{G}_{\alpha i\beta j}(z)$  must vanish due to symmetry [but not  $\mathcal{G}_{\alpha i\alpha j}(z)$ , henceforth indicated as  $\mathcal{G}_{ij}^\alpha(z)$ ]. We first consider the non-interacting case (then  $H_{\text{int}} = 0$  and  $G$  is printed in *italic*), where explicit solutions are readily obtained using the Green function's equations of motions [14,17]:

$$\hbar^{-1} G_{kk}(z) = \frac{1}{z - \varepsilon_k} + \sum_{\alpha ij} \frac{V_{k\alpha i} V_{\alpha jk}}{(z - \varepsilon_k)^2} G_{ij}^\alpha(z), \quad (6)$$

and

$$\sum_m [(z - \varepsilon_{\alpha i}) \delta_{im} - \Delta_{im}^\alpha(z)] G_{mj}^\alpha(z) = \hbar \delta_{ij}, \quad (7)$$

with  $\Delta_{ij}^\alpha(z)$  being the self-energy due to the dot-lead interaction:  $\Delta_{ij}^\alpha(z) = \sum_k V_{\alpha ik} V_{k\alpha j} (z - \varepsilon_k)^{-1}$ . In the noninteracting case the diagonal QD Green function assumes the familiar form  $\hbar^{-1} G_{ii}^\alpha(z) = [z - \varepsilon_{\alpha i} - \Delta_{ii}^\alpha(z)]^{-1}$ , where  $\text{Im}[\Delta_{ii}^\alpha(z)]$  is the virtual level width and  $\text{Re}[\Delta_{ii}^\alpha(z)]$  renormalizes the single-particle level  $\varepsilon_{\alpha i}$ . Equation (7) is modified to take into account electron-electron interaction by introducing the intradot proper self-energy matrix  $\Sigma_{ij}^\alpha(z)$ :

$$\sum_m [(z - \varepsilon_{\alpha i}) \delta_{im} - \Sigma_{im}^\alpha(z) - \Delta_{im}^\alpha(z)] \mathcal{G}_{mj}^\alpha(z) = \hbar \delta_{ij}. \quad (8)$$

Henceforth we focus on the fully interacting system, and we consider the case of elastic scattering when only a single QD level  $(\bar{\alpha}, \bar{i})$  is coupled to the leads. Indeed, this is a reasonable scenario for the electrostatic potential barriers separating dot and leads in many experimental setups [9], where matrix elements  $V_{k\alpha i}$  strongly depend on both energies  $\varepsilon_k$  and  $\varepsilon_{\alpha i}$ , respectively. We then set  $V_{k\alpha i} = 0$  if  $(\alpha, i) \neq (\bar{\alpha}, \bar{i})$  and  $V_{k\bar{\alpha}\bar{i}} \neq 0$  with  $\varepsilon_{\bar{\alpha}\bar{i}} - \varepsilon_{\text{cut}} \leq \varepsilon_k \leq \varepsilon_{\bar{\alpha}\bar{i}} + \varepsilon_{\text{cut}}$ , where  $\varepsilon_{\text{cut}}$  is a suitable cutoff. Note that Coulomb correlation is included in full and the intradot self-energy has off-diagonal matrix elements,  $\Sigma_{ij}^\alpha(z) \neq 0$ ,  $\alpha = 1, \dots, N_{\text{class}}$ , and  $i, j = 1, \dots, N_\alpha$ . This scenario is generic enough to correctly describe many experimental

situations, except the case of degeneracies between QD levels  $\varepsilon_{\bar{\alpha}\bar{i}}$  and  $\varepsilon_{\beta j}$ .

We proceed in close analogy with Langreth [12] and calculate the charge  $\mathcal{N}_{\bar{\alpha}\bar{i}}$  (in units of  $e$ ) displaced at the dot level  $(\bar{\alpha}, \bar{i})$  as the difference between the charge at equilibrium in the presence and in the absence of the dot, respectively:

$$\mathcal{N}_{\bar{\alpha}\bar{i}} = -\frac{1}{\hbar\pi} \text{Im} \int_{-\infty}^{\mu+i\eta} dz \left\{ \left[ \sum_k \mathcal{G}_{kk}(z) + \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(z) \right] - \sum_k G_{kk}^{\text{free}}(z) \right\}, \quad (9)$$

where  $\eta$  is a positive infinitesimal quantity,  $\mu$  is the equilibrium chemical potential [18], and  $\hbar^{-1} G_{kk}^{\text{free}}(z) = (z - \varepsilon_k)^{-1}$  is the propagator of a free traveling wave in the absence of the QD. By using Eqs. (9) and (6) where  $G$ 's are replaced with  $\mathcal{G}$ 's, one obtains the following expression for  $\mathcal{N}_{\bar{\alpha}\bar{i}}$ :

$$\mathcal{N}_{\bar{\alpha}\bar{i}} = -\frac{1}{\hbar\pi} \text{Im} \int_{-\infty}^{\mu+i\eta} dz \left[ 1 - \frac{\partial \Delta_{\bar{i}\bar{i}}^{\bar{\alpha}}(z)}{\partial z} \right] \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(z). \quad (10)$$

We now use the identity

$$\text{Im} \int_{-\infty}^{\mu+i\eta} dz \frac{\partial \Sigma_{\bar{i}\bar{i}}^{\bar{\alpha}}(z)}{\partial z} \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(z) = 0, \quad (11)$$

which is a natural generalization of the Luttinger relation of Fermi liquids [19], and it has been already applied by Langreth [12] to the single-level case. Combining Eqs. (10) and (11) gives

$$\mathcal{N}_{\bar{\alpha}\bar{i}} = \frac{1}{\pi} \text{Im} \int_{-\infty}^{\mu+i\eta} dz \frac{\partial}{\partial z} \ln \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(z). \quad (12)$$

Since the asymptotic form of the Green function as  $z \rightarrow -\infty$  is  $\mathcal{G}_{ij}^\alpha(z) \approx \hbar \delta_{ij}/z$ , Eq. (12) may be cast into the form

$$\mathcal{N}_{\bar{\alpha}\bar{i}} = \frac{1}{\pi} \text{Im} [\ln \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(\mu + i\eta) - i\pi], \quad (13)$$

where we have chosen the cut along the positive real axis. To connect the displaced charge  $\mathcal{N}_{\bar{\alpha}\bar{i}}$  to the Friedel phase, we observe that the  $T$  scattering matrix of the single channel  $\mathbf{k}$  is  $\mathcal{T}_{kk}(z) = |V_{k\bar{\alpha}\bar{i}}|^2 \hbar^{-1} \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(z)$  (cf. Sec. 5.2 of Ref. [2]); therefore, the phase on the energy shell  $\delta_F(\mu/\hbar)$  is

$$\delta_F(\mu/\hbar) = \arg \mathcal{T}_{kk}(\mu + i\eta) = \text{Im} \ln \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}}(\mu + i\eta) - \pi, \quad (14)$$

where we pick the branch of the logarithm as before and add the reference constant  $-\pi$ , so that  $0 \leq \delta_F \leq \pi$ . Comparison of (13) and (14) gives the desired result:

$$\delta_F(\mu/\hbar) = \pi \mathcal{N}_{\bar{\alpha}\bar{i}}. \quad (15)$$

The exact sum rule (15) states that the Friedel phase shift, due to scattering by the dot, is proportional to the net charge accumulated at the dot level  $(\bar{\alpha}, \bar{i})$  with respect to the free system (i.e., without a dot). In the case of a single level ( $N_{\text{class}} = 1$ ,  $N_\alpha = 1$ ), Eq. (15) is equivalent to the result of Langreth for the Anderson model [12]. In the

many-level case, Eq. (15) is a nontrivial generalization of previous theories (see, e.g., [2,12]), which can be summarized as follows: The sum of phase shifts due to consecutive filling of many levels is fixed by the *total* charge which can be placed into the dot, ruled by  $\mu$ . If the dot levels, possibly broadened due to hybridization with continuum states, lie well below  $\mu$  and  $T > T_K$ , where  $T_K$  is the Kondo temperature, then the afore mentioned charge is an integer quantity fixed by orbital degeneracy. According to (15), however, this simple picture is generally incorrect, as the following conceptual tunneling experiment illustrates.

Think of varying continuously the chemical potential  $\mu$  across an energy window centered at the resonant value  $\hbar\omega_{\text{res}}$ , which is implicitly given by the real part of a certain pole of  $\mathcal{G}_{\bar{\alpha}\bar{i}}^{\bar{\alpha}}(z)$ ,  $\hbar\omega_{\text{res}} = \varepsilon_{\bar{\alpha}\bar{i}} + \text{Re}[\sum_{\bar{i}\bar{i}}^{\bar{\alpha}}(\hbar\omega_{\text{res}}) + \Delta_{\bar{i}\bar{i}}^{\bar{\alpha}}(\hbar\omega_{\text{res}})]$ . For the sake of clarity we here focus on the Coulomb blockade regime only, despite the fact that sum rule (15) applies to the Kondo regime as well. The level at  $\hbar\omega_{\text{res}}$  is located between two blockaded regions with  $N$  and  $N+1$  electrons in the dot, respectively. The transport window is chosen to be large enough to fully contain the width of the QD level, given by  $\varepsilon_{\text{cut}}$ , but narrower than the spacing between neighboring resonant levels. Therefore, as  $\mu$  is swept upward across the QD level, the dot is charged by exactly one electron, in addition to those already localized. Nevertheless, the variation of the spectral density  $\Delta\mathcal{N}_{\bar{\alpha}\bar{i}}$  of Eq. (15) across the energy window is generally less than 1, and consequently the phase shift increment of the outgoing wave spreading out from the dot at the top of the energy window is less than  $\pi$ .

To understand it, note that the total number of scattering states of the whole dot + leads system, in the energy window considered above, must be exactly equal to the sum of both free traveling waves in the leads and confined states in the dots, when the two subsystems are decoupled ( $H_{\text{mix}} = 0$ ). Therefore, in this case  $\Delta\mathcal{N}_{\bar{\alpha}\bar{i}}$  may be calculated by simply integrating the spectral density of the isolated dot:

$$\begin{aligned} \Delta\mathcal{N}_{\bar{\alpha}\bar{i}} &= -\frac{1}{\hbar\pi} \text{Im} \int_{\hbar\omega_{\text{res}} - \varepsilon_{\text{cut}} + i\eta}^{\hbar\omega_{\text{res}} + \varepsilon_{\text{cut}} + i\eta} dz \mathcal{G}_{\bar{i}\bar{i}}^{\bar{\alpha}\text{free}}(z) \\ &= |\langle \Psi_0^{N+1} | c_{\bar{\alpha}\bar{i}}^\dagger | \Psi_0^N \rangle|^2, \end{aligned} \quad (16)$$

where  $|\Psi_0^N\rangle$  is the exact interacting ground state of the isolated dot with  $N$  electrons. The following is clear: (i)  $\Delta\mathcal{N}_{\bar{\alpha}\bar{i}}$  is a positive quantity, which can considerably deviate from the unit charge that could fill in the level  $(\bar{\alpha}, \bar{i})$ , due to the correlation between electrons localized in the dot.  $\Delta\mathcal{N}_{\bar{\alpha}\bar{i}}$  can even be zero due to the spin blockade; namely, the difference between total spins of  $|\Psi_0^{N+1}\rangle$  and  $|\Psi_0^N\rangle$  is not equal to  $\pm 1/2$ . (ii) The missing weight is recovered by integration on the whole spectrum. (iii) Equation (16) provides the basis for exact numerical evaluation of the phase shift, by computing the spectral density of a

confined system, e.g., by means of the full configuration interaction method [11,20].

The discrepancy between the total and the spectral charge density of Eq. (15) can be neglected only if intradot correlation effects are absent; namely, the ground state of the isolated dot is a single Slater determinant. In such a case, the matrix element appearing in Eq. (16) is either one or zero, depending if level  $(\bar{\alpha}, \bar{i})$  is filled in or not, respectively, in the  $N \rightarrow N+1$  tunneling event. This occurs in the degenerate Anderson model [2,12,17,21], where  $H_{\text{int}}$  assumes a Hartree-Fock-like form diagonal in the  $(\alpha, i)$  indices. While such a mean-field model is satisfying for magnetic impurities in a bulk metal or many-electron dots, it breaks down for large dots with very few electrons, like those of Ref. [9], where correlation effects may dominate [10], driving the ground state of the isolated dot even towards the Wigner crystallization regime [11].

The idea behind (15) is general, as we prove in the following case of arbitrary coupling between leads and QD levels ( $V_{k\alpha i} \neq 0 \quad \forall k, \alpha, i$ ). We start from the exact relation between Friedel phase and delay time  $\tau_{\text{delay}}$  of a traveling wave packet when trapped in the dot [22]:

$$2 \frac{d\delta_{\text{F}}(\omega)}{d\omega} = \tau_{\text{delay}}(\omega), \quad (17)$$

where  $\hbar\omega$  is the average energy of the wave packet. We assume that Eq. (17), proved for independent particles [23], holds even in the presence of correlation since  $H_{\text{int}} \neq 0$  only in the region  $\Omega$  occupied by the dot, while in the outer space the packet moves freely. The delay  $\tau_{\text{delay}}$  can be calculated as the difference between the (“dwell”) times spent by the packet in  $\Omega$  in the presence and in the absence of the dot, respectively [22,24].

By extending the approach of Iannaccone [25], we write the wave packet  $|\Phi\rangle$  as a superposition of the interacting incoming scattering states  $|\Psi_k\rangle$ ,  $|\Phi\rangle = \sum_k \alpha(\mathbf{k}) |\Psi_k\rangle$ , where  $\sum_k |\alpha(\mathbf{k})|^2 = 1$  so that  $|\Phi\rangle$  is normalized to unity. The exact stationary interacting state,  $|\Psi_k\rangle$ , satisfies  $H|\Psi_k\rangle = (E_0 + \varepsilon_k)|\Psi_k\rangle$ , with  $E_0$  being the energy of the ground state without the extra electron to be scattered, and  $H|\Psi_0\rangle = E_0|\Psi_0\rangle$ . The probability amplitude for finding the scattered electron at position  $\mathbf{r}$  is obtained by projecting  $|\Phi\rangle$  on an eigenstate of position of the extra electron [20],  $\Psi^\dagger(\mathbf{r}, t)|\Psi_0\rangle$ , where we introduce the field operator  $\Psi^\dagger(\mathbf{r}, t)$ , creating an electron in  $\mathbf{r}$ . The field  $\Psi(\mathbf{r}, t)$  may be decomposed onto a mixed basis of Bloch free waves  $\phi_k(\mathbf{r})$  and confined QD orbitals  $\phi_{\alpha i}(\mathbf{r})$ :  $\Psi(\mathbf{r}, t) = \sum_k \phi_k(\mathbf{r})c_k + \sum_{\alpha i} \phi_{\alpha i}(\mathbf{r})c_{\alpha i}$  [26]. We therefore define the mean dwell time in  $\Omega$  associated with the wave packet  $|\Phi\rangle$  as

$$\int_{-\infty}^{\infty} dt \int_{\Omega} d\mathbf{r} |\langle \Psi_0 | \Psi(\mathbf{r}, t) | \Phi \rangle|^2. \quad (18)$$

The integral (18) converges since the probability of finding the scattered electron in  $\Omega$  vanishes for time approaching  $\pm\infty$ . After decomposing the wave packet on the basis

made of  $|\Psi_k\rangle$ 's and performing the time integration in (18), the delay time is obtained as a sum of stationary state contributions [25], which are separately written as

$$\tau_{\text{delay}}(\omega) = 2\pi\hbar\delta(\hbar\omega - \varepsilon_k) \times \int_{\Omega} d\mathbf{r} [|\langle\Psi_0|\Psi(\mathbf{r})|\Psi_k\rangle|^2 - |\phi_k(\mathbf{r})|^2]. \quad (19)$$

We extend the range of integration in (19) to the whole space, since the contribution outside  $\Omega$  is null, and by combining (17) and (19) and the orbital spectral representation of  $\Psi(\mathbf{r})$ , we obtain the desired result

$$\frac{1}{\hbar} \frac{d\delta_F(\omega)}{d\omega} = \pi\mathcal{N}(\omega), \quad (20)$$

where  $\mathcal{N}(\omega)$  is the total spectral density [cf. (9)]:

$$\mathcal{N}(\omega) = -\frac{1}{\hbar\pi} \text{Im} \left\{ \sum_{\alpha i} \mathcal{G}_{ii}^{\alpha}(\omega + i\eta) + \sum_k [\mathcal{G}_{kk}(\omega + i\eta) - G_{kk}^{\text{free}}(\omega + i\eta)] \right\}. \quad (21)$$

Equation (20) is the natural generalization of (15), namely, a generalized FSR where the phase shift variation as a function of energy is proportional to the variation of the total displaced spectral density  $\mathcal{N}$ . Again, all considerations of the previous example [Eq. (16)] apply; i.e.,  $\Delta\mathcal{N}$  does not need to be one between two consecutive Coulomb blockade regions. The result (20) is generic and independent of the nature of the coupling between dot and leads.

We are now able to focus on the experiment of Ref. [9]. The transmission phase shift of an electron tunneling into a QD is determined together with the integer charge filling in the dot, which is in the Coulomb blockade regime. It turns out that the phase increment between specific neighboring conductance valleys, as a function of the plunger voltage, is a fraction of  $\pi$  ( $\sim 0$  between  $N = 3$  and  $N = 4$  and  $\sim 3\pi/4$  between  $N = 6$  and  $N = 7$  blockaded regions). Moreover, for these specific voltage ranges, the phase variation is continuous and smooth. Under such circumstances,  $\delta_F$  and the transmission phases are expected to coincide [6,7]. According to previous theories based on the FSR [6,7], one would predict a unit increment of  $\pi$  in all cases, unless some additional charge unpredictably accumulates outside the dot [27]. Our theory suggests an alternative natural explanation: i.e., fractional (or even zero) increments of  $\delta_F$  originate from strong electron correlation (or spin blockade). This interpretation is supported by the estimated low value of electron density. In fact, as the density diminishes, electrons in the dot are expected to crystallize [11], which affects  $\delta_F$  in a density-dependent manner. Extensive numerical simulations will be reported elsewhere.

In conclusion, we derived an exact generalized Friedel sum rule for an interacting multilevel nano-object. The

variation of the Friedel phase through Coulomb blockade regions for values which are fractions of  $\pi$  is the fingerprint of electron correlation.

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