

## Persistent Currents in One Dimension: The Counterpart of Leggett's Theorem

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We discuss the sign of the persistent current of  $N$  electrons in one dimensional rings. Using a topology argument, we establish lower bounds for the free energy in the presence of arbitrary electron-electron interactions and external potentials. Those bounds are the counterparts of upper bounds derived by Leggett. Rings with odd (even) numbers of polarized electrons are always diamagnetic (paramagnetic). We show that unpolarized electrons with  $N$  being a multiple of four exhibit either paramagnetic behavior or a superconductorlike current-phase relation.

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A persistent current [1] flows at low temperatures in small conducting rings when they are threaded by a magnetic flux  $\phi$ . This current is a thermodynamic effect which is deeply connected to the presence of quantum coherence. Its magnitude can be expressed as  $I = -\partial F/\partial \phi$  in terms of the free energy  $F$ . Persistent currents have been the focus of intensive theoretical activity (see for example [2–7]). Nevertheless, the understanding of the experimentally measured currents in metallic [8–11] and semiconductor [12,13] rings remains incomplete. In particular, the large discrepancy between observed currents in diffusive rings and the theory for noninteracting electrons points to the importance of interactions.

The persistent current itself generates a magnetic field which is detected in the experiments. Besides the magnitude of the current, the sign of this magnetic response is of particular interest. In the absence of interactions, mesoscopic sample-to-sample fluctuations between paramagnetic ( $F(\phi) < F(0)$ ) and diamagnetic ( $F(\phi) > F(0)$ ) behavior are expected. Taking into account repulsive (attractive) interactions leads to the prediction of a paramagnetic (diamagnetic) average response in ensembles of diffusive rings. However, only diamagnetic signals have so far been observed in experiments.

A very general theoretical result in this domain is the theorem by Leggett [14] which states that the sign of the zero-temperature persistent current for spinless fermions (fully polarized electrons) in one-dimensional (1D) rings is given by the parity of the particle number. This result appears in the form of upper bounds for the ground-state energy. Leggett's theorem holds for arbitrary potential landscape and electron-electron interactions.

In this Letter, we establish general lower bounds of the free energy that can be seen as the counterparts of Leggett's upper bounds. Our result is valid at any temperature and also for unpolarized electrons. Scenarios allowed

by those bounds are studied using the density matrix renormalization group (DMRG) method [15].

We start with the Hamiltonian

$$H = \sum_{i=1}^N \frac{1}{2m} [p_i - eA(r_i)]^2 + V(r_i) + \sum_{i<j} U(r_i - r_j)$$

describing  $N$  spinless electrons in a 1D ring of size  $L$ , where  $p_i$  is the momentum of electron  $i$ ,  $A(r)$  the vector potential corresponding to a magnetic flux  $\phi$ ,  $V(r)$  an external potential, and  $U(r)$  the electron-electron interaction. A gauge transformation allows for the replacement of the vector potential by flux-dependent boundary conditions

$$\Psi(L, r_2, \dots, r_N) = e^{i\Phi} \Psi(0, r_2, \dots, r_N)$$

with  $\Phi = 2\pi\phi e/h$  for the antisymmetric wave function  $\Psi$ .

Leggett's argument [14] is based on the ansatz

$$\Psi_v(r_1, \dots, r_N) = e^{i\theta(r_1, \dots, r_N)} \Psi_0$$

for the variational ground state  $\Psi_v$  at  $\Phi \neq 0$  in terms of the exact ground-state wave function  $\Psi_0$  at  $\Phi = 0$ . The energy corresponding to this ansatz is

$$E_v = E(\Phi = 0) + \frac{N\hbar^2}{2m} \int d^N r |\partial_{r_1} \theta|^2 |\Psi_0|^2,$$

where  $E(\Phi)$  is the exact ground-state energy. The phase  $\theta$  is a symmetric function of its arguments and should go from 0 to  $\Phi$  as  $r_1$  goes around the ring from 0 to  $L$ . In this loop,  $r_1$  crosses the coordinates of the other  $N - 1$  particles. At each crossing the sign of  $\Psi_0$  changes. Since  $\Psi_0$  must be back to its initial value at the end of the loop, for even  $N$  there must be at least *one more* sign change. The function  $\theta$  can be chosen such that its gradient is concentrated around this additional point where  $\Psi_0$  vanishes, yielding  $E_v = E(0)$ . This variational energy provides an upper bound  $E(\Phi) \leq E(0)$  for the ground-state energy and the system is paramagnetic. A similar argument leads to

$E(\Phi) \leq E(\pi)$  for odd  $N$ . In the sequel, we show that these upper bounds can be complemented by corresponding lower bounds.

We consider the partition function  $Z = \text{Tr}_A e^{-\beta H}$  where  $\text{Tr}_A$  is the trace over antisymmetrized many-body states. One can divide the inverse temperature  $\beta$  in  $K$  increments  $\Delta\tau = \beta/K$  and insert  $1 = \int dR |R\rangle\langle R|$  at each step, where  $|R\rangle = |r_1, r_2, \dots, r_N\rangle$  denotes the list of positions of all particles. The result reads

$$Z = \sum_P \sum_{R_1, \dots, R_K} |P| [e^{-\Delta\tau H}]_{R_1 R_2} \times [e^{-\Delta\tau H}]_{R_2 R_3} \dots [e^{-\Delta\tau H}]_{R_{K-1} R_K} [e^{-\Delta\tau H}]_{R_K P(R_1)},$$

where  $P$  is a permutation of the  $N$  particles and  $|P| = \pm 1$  its signature. This path integral formula can be formally rewritten in continuous form as

$$Z = \sum_P |P| \int \mathcal{D}R(\tau) e^{-S[R(\tau)]}, \quad (1)$$

where  $S[R(\tau)]$  is the action of the path  $R(\tau)$  which satisfies  $R(\beta) = P[R(0)]$ . Examples for such paths are shown in Fig. 1. The effect of a magnetic flux on the path integral is a phase  $\pm\Phi$  each time a particle crosses the boundary, yielding

$$Z(\Phi) = \sum_P |P| \int \mathcal{D}R(\tau) e^{i\Phi m[R(\tau)]} e^{-S[R(\tau)]}, \quad (2)$$

where the winding number  $m[R(\tau)]$  counts the total number of boundary crossings contained in  $R(\tau)$  (+1 from left to right and  $-1$  in the opposite direction). With  $F =$

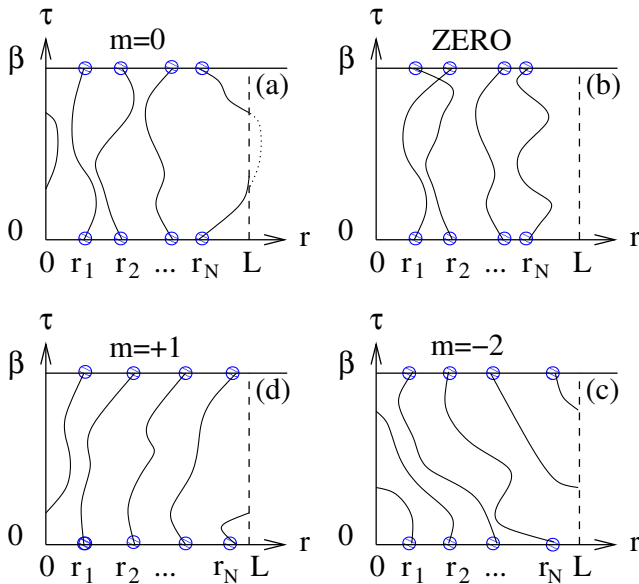


FIG. 1 (color online). Four different paths  $R(\tau)$  of the path integral (1). The winding numbers  $m$  are equal to (a) 0, (b) 0, (c)  $-2$  and (d)  $+1$ . Path (b) does not contribute in 1D rings because two particle lines cross, which is forbidden by antisymmetry.

$-\log Z/\beta$ , Eq. (2) leads to  $\partial^2 F/\partial\Phi^2|_{\Phi=0} = \langle m^2[R] \rangle/\beta$  which is widely used to determine the superfluid fraction in bosonic systems, in particular, in the context of quantum Monte Carlo simulations [16]. Anyway, Eq. (2) can be expressed as

$$Z(\Phi) = \sum_{m=-\infty}^{+\infty} Z_m e^{i\Phi m}$$

with the partial partition functions  $Z_m$  for a given  $m$ .

In 1D rings, most of the permutations  $P$  do not contribute to the path integral since they would correspond to paths where two particles cross at some point. Those paths [see Fig. 1(b)] have zero contribution due to antisymmetry. As a result, only paths with *cyclic permutations* contribute to the path integral. There is only one single cyclic permutation  $P_m$  per winding number  $m$  (see Fig. 1), whose signature determines the sign of the corresponding  $Z_m$ . Since the signature of a cyclic permutation of  $N$  particles with winding number  $m$  is given by  $|P_m| = (-1)^{m(N-m)}$ , we have

$$Z_m = (-1)^{m(N-m)} |Z_m|.$$

If  $N$  is odd, then  $m(N-m)$  is even and  $Z_m$  is positive for all  $m$ . Since  $Z_m = Z_{-m}$ , we have

$$Z(\Phi) = Z_0 + 2 \sum_{m=1}^{\infty} Z_m \cos(m\Phi) \leq Z_0 + 2 \sum_{m=1}^{\infty} |Z_m| = Z(0)$$

so that  $F(\Phi) \geq F(0)$ , and the system is diamagnetic. When  $N$  is even,  $m(N-m)$  has the same parity as  $m$  and  $Z_m = (-1)^m |Z_m|$ , leading to

$$Z(\Phi) \leq Z_0 + 2 \sum_{m=1}^{\infty} |Z_m| = Z(\pi)$$

such that  $F(\Phi) \geq F(\pi)$ . At zero temperature, the above inequalities together with those obtained by Leggett read

$$E(0) \leq E(\Phi) \leq E(\pi) \quad \text{for } N \text{ odd}, \quad (3)$$

$$E(\pi) \leq E(\Phi) \leq E(0) \quad \text{for } N \text{ even}. \quad (4)$$

The previous results can be partially extended to unpolarized 1D systems with  $N_\uparrow$  up and  $N_\downarrow$  down electrons under a spin conserving Hamiltonian. Introducing winding numbers  $m_\uparrow$  and  $m_\downarrow$  for the two species, we have

$$Z(\Phi) = \sum_{m_\uparrow, m_\downarrow} Z_{m_\uparrow, m_\downarrow} e^{i\Phi(m_\uparrow + m_\downarrow)}.$$

Using similar arguments as above, we find  $Z_{m_\uparrow, m_\downarrow} = (-1)^{|P_{m_\uparrow}| + |P_{m_\downarrow}|} |Z_{m_\uparrow, m_\downarrow}|$  and conclude that

$$F(0) \leq F(\Phi) \quad \text{for } N_\uparrow \text{ and } N_\downarrow \text{ odd}, \quad (5)$$

$$F(\pi) \leq F(\Phi) \quad \text{for } N_\uparrow \text{ and } N_\downarrow \text{ even}. \quad (6)$$

Those lower bounds are valid at arbitrary temperature. Equations (3) and (4) for the polarized case and (5) and (6) for unpolarized electrons are the central results of this work.

We now consider rings with a number of electrons  $N = N_\uparrow + N_\downarrow = 4n$  being a multiple of four, and characterize the flux dependence of the free energy using the curvatures

$$C_{0(\pi)} = \partial^2 F / \partial \Phi^2 |_{\Phi=0(\pi)}$$

at  $\Phi = 0$  and  $\pi$ . In the unpolarized case ( $N_\uparrow = N_\downarrow = 2n$ ) Eq. (6) applies, the free energy assumes a minimum at  $\Phi = \pi$ , and  $C_\pi$  is positive. The curvature at zero flux  $C_0$  is not constrained such that the two scenarios illustrated in Fig. 2 remain. (i) If  $C_0$  is negative, the system is paramagnetic and lowers its energy in the presence of a magnetic flux. (ii) If  $C_0 > 0$ , the system is diamagnetic. In this case the usual sinusoidal current-phase relation ( $F \propto -\cos\Phi$ ) is prohibited by  $C_\pi > 0$ . There is a strong contribution of the second harmonic in  $\Phi$  and it exists at least one flux value  $0 < \Phi_c < \pi$  where the free energy is maximum and the persistent current vanishes. Without interaction, the spin channels are independent and one finds case (i) by applying Leggett's inequality (4) to each of them. Case (ii) is characteristic, for instance, of superconducting fluctuations induced by a weak attractive interaction (in a superconductor,  $F \propto -\cos(2\Phi)$  and  $\Phi_c = \pi/2$ ).

The overall conclusion is that a simple diamagnetic response is prohibited. The system must either be in a paramagnetic state or have a superconductorlike current-phase relation [17].

In order to illustrate the behavior of the curvatures  $C_0$  and  $C_\pi$ , we consider 1D tight-binding rings of  $M$  sites, described by the Hamiltonian

$$H = \sum_{\sigma} \sum_{j=1}^M \{ -t(c_{j+1,\sigma}^\dagger c_{j,\sigma} + \text{H.c.}) + V_{j,\sigma} n_{j,\sigma} \} + \sum_{j=1}^M (U_0 n_{j,\uparrow} n_{j,\downarrow} + U_1 n_{j+1} n_j). \quad (7)$$

Here,  $c_{j,\sigma}^\dagger$  creates an electron with spin  $\sigma = \{\uparrow, \downarrow\}$  on site  $j$ , the random on-site energies  $V_{j,\sigma}$  are drawn independently from the interval  $[-W/2, W/2]$ , and the kinetic energy scale is given by the hopping amplitude  $t$ . We include Hubbard on-site and nearest-neighbor interactions of strength  $U_0$  and  $U_1$ , respectively. The density operators are defined as  $n_{j,\sigma} = c_{j,\sigma}^\dagger c_{j,\sigma}$  and  $n_j = n_{j,\uparrow} + n_{j,\downarrow}$ , and the

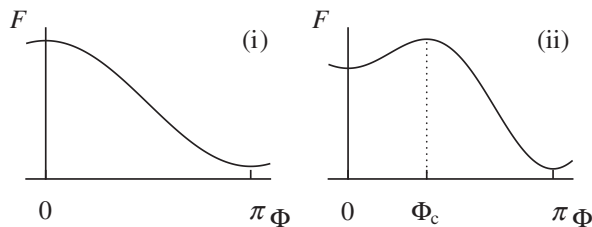


FIG. 2. Two possible flux-dependences of the free energy  $F$  for  $N = 4n$  with even  $N_\uparrow = N_\downarrow$ , corresponding to paramagnetic (i) and diamagnetic (ii) behavior.

boundary condition  $c_{M+1,\sigma} = e^{i\Phi} c_{1,\sigma}$  accounts for a magnetic flux threading the ring.

We use the DMRG algorithm [15] adapted to disordered systems [18] to calculate the ground-state energies and the zero-temperature curvatures [19] for  $\Phi = 0$  and  $\Phi = \pi$ , fully taking into account the many-body correlations. For the largest systems, 750 states per block are kept in the DMRG iterations. All of the numerical results should, and do, satisfy the relations (5) and (6).

In Fig. 3, we show the effect of an attractive interaction on the curvature  $C_0$ , for single realizations of disordered quarter-filled Hubbard rings of sizes up to  $M = 2N = 32$  sites with  $W = t$  and  $U_1 = 0$ . For even  $N_\uparrow = N_\downarrow$  ( $N = 4n$ ), an attractive interaction reverses the sign of  $C_0$ . Hence the interactions induce a transition from scenario (i) to (ii) of Fig. 2 and drive the system from paramagnetic towards “superconducting.” In contrast, the sign of  $C_0$  for odd  $N_\uparrow = N_\downarrow$  ( $N = 4n + 2$ ) remains positive for all values of  $U_0$ , as dictated by (5).

A molecular realization of the  $N = 4n$  case is cyclooctatetraene ( $C_8H_8$ , see the inset of Fig. 4), which consists of a ring of eight carbon atoms with eight  $\pi$  electrons. In Fig. 4, we plot the curvatures  $C_0$  and  $C_\pi$  as a function of the nearest-neighbor interaction strength  $U_1$  for the model Hamiltonian (7) with  $M = 8$  and  $W = 0$ , using the parameters  $t = 2.64$  eV and  $U_0 = 8.9$  eV given in Ref. [20] for cyclooctatetraene (COT). Depending on the strength of  $U_1$  (and the neglected longer range parts of the interaction), the system can undergo a transition [21] from a paramagnetic spin-density-wave ( $U_1 \leq 4.6$  eV and  $C_0 < 0$ ) to a diamagnetic charge-density-wave ( $U_1 \geq 4.6$  eV and  $C_0 > 0$ ) ground state.

The presence of paramagnetism or orbital ferromagnetism [22] in COT is still a matter of debate [20,23]. A ferromagnetic instability can occur in small paramagnetic rings provided their inductance is large enough. Then, a magnetic field fluctuation generates a current which reinforces the magnetic field. For small flux, the persistent

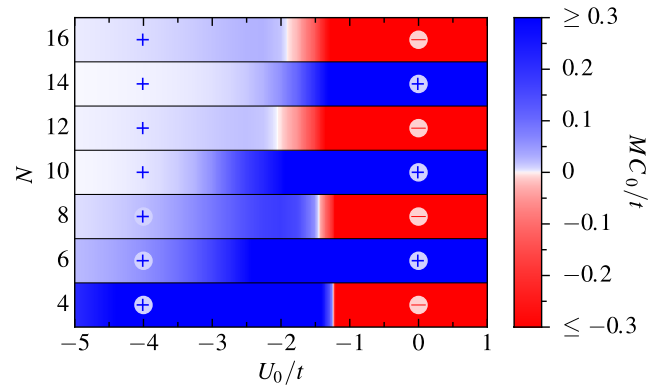


FIG. 3 (color online). Curvatures  $C_0$  at zero temperature for disordered quarter-filled Hubbard rings of different sizes  $M = 2N$ , as a function of  $U_0$ , at  $U_1 = 0$  and  $N_\uparrow = N_\downarrow$ , for one disorder realization with  $W = t$ .

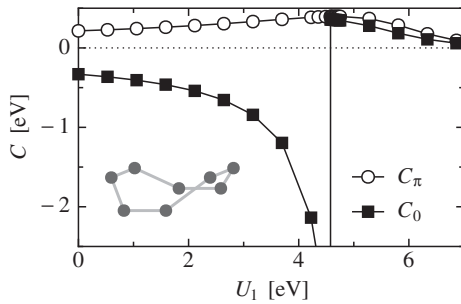


FIG. 4. Curvatures  $C_0$  and  $C_\pi$  as a function of  $U_1$  for an unpolarized clean ring with  $M = 8$  at zero temperature, for the parameters of COT ( $N = 8$ ,  $t = 2.64$  eV,  $U_0 = 8.9$  eV). Inset: Sketch of the C atom configuration in COT.

current is  $I \approx C_0 e \Phi / \hbar$  and the magnetic flux generated by this current is  $\Phi \approx \mu_0 e L I / \hbar$  ( $L \approx 1$  nm is the typical circumference of the molecule and  $\mu_0 L$  is its typical inductance). The instability occurs when the magnetic field generated current amplifies field fluctuations, i.e., when  $C_0 < 0$  and  $X = \mu_0 |C_0| e^2 L / \hbar^2 > 1$ . In our model, the most pronounced negative curvature  $C_0 \approx -5$  eV occurs in the spin-density-wave regime close to the transition at  $U_1 \approx 4.6$  eV. We therefore have  $X \lesssim 3 \times 10^{-3}$ , far from the ferromagnetic instability. Moreover, Jahn-Teller distortions in COT have been predicted [23] which would reduce the curvatures.

An alternative to small molecules are rings made in semiconductor heterostructures. Important progress has been realized in the fabrication of such systems and 1D rings with a single conduction channel could be produced. Estimates show that one still has  $X \ll 1$ . Therefore, no orbital ferromagnetism is to be expected, reminiscent with the result for large 1D Luttinger liquid rings [24].

However, as illustrated by the results shown in Fig. 3, we predict that changing the number of electrons by two (with a back gate for instance) leads to a change of either the sign of the magnetic response (paramagnetism) or the periodicity of the response to a magnetic field (“superconductor-like”). This could allow us to detect small attractive interactions in those systems, whose presence is suggested by the fact that so far only diamagnetic responses have been observed in multichannel rings.

In conclusion, we have established general relations for the sign of persistent currents in 1D rings that are the counterpart of Leggett’s theorem for spinless fermions. Both theorems depend on the topology of the system and do not hold in higher dimensions or in the presence of a side stub [25]. Our theorem is valid at arbitrary temperature and also with spin. For spinless fermions, it implies that interactions and disorder cannot affect the sign of the persistent current. In particular, the possibility discussed by Leggett of having maxima of the energy at integer and half integer flux quantum, corresponding to a paramagnetic signal in an assembly of rings, is ruled out by (3) and (4). For electrons with spin, when Leggett’s theorem does not

apply, only the lower bounds (5) and (6) for the free energy remain. This allows for a superconductorlike current-phase relation when attractive electron-electron interactions change the sign of the unconstrained curvature. The phenomenological Hückel rule that cyclic molecules with  $4n + 2$  electrons like benzene are aromatic while those with  $4n$  electrons are not is put on firm ground by our theorem.

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