Magnetic moments in biased quantum circuits

Michele Cini,[1,2] Enrico Perfetto,[1] and Gianluca Stefanucci[1,2,3]
[1]*Dipartimento di Fisica, Università di Roma Tor Vergata, Via della Ricerca Scientifica 1, I-00133 Rome, Italy*
[2]*Laboratori Nazionali di Frascati, Istituto Nazionale di Fisica Nucleare, Via E. Fermi 40, 00044 Frascati, Italy*
[3]*European Theoretical Spectroscopy Facility (ETSF)*
(Received 23 November 2009; revised manuscript received 16 February 2010; published 9 April 2010)

We consider a quantum ring connected to leads and the current which is excited by biasing the circuit in the *absence* of external magnetic field. The magnetic moment $\mathcal{M}_{\rm ring}$ that arises in this way depends on the current distribution inside the ring. We perform a thought experiment in which $\mathcal{M}_{\rm ring}$ is determined by measuring the torque due to an infinitesimally small *probe* magnetic field. This leads to a definition $\mathcal{M}_{\rm ring}$, which is given by the derivative of the grand-canonical energy of the quantum ring with respect to an external magnetic flux in the zero flux limit. We develop the many-body formalism by Green's-function techniques and carry on illustrative model calculations. The resulting theory predicts that at small bias the current in the ring is always *laminar*, that is, the magnetic moment vanishes in linear-response theory. The approach most naturally lends itself to include induction effects by a self-consistent procedure.

DOI: 10.1103/PhysRevB.81.165202 PACS number(s): 73.23.Ad, 72.10.Bg

I. INTRODUCTION

There is widespread interest in systems containing mesoscopic and nanoscopic rings where quantum coherence effects are important. The persistent currents[1] in mesoscopic metallic rings[2] have been investigated in depth. Recent progress in connecting aromatic molecules to metallic leads brought the ringlike topology into the nanoworld.[3] The quantum behavior of electrons and fundamental paradigms such as Aharonov-Bohm oscillations,[4,5] quantum interference pattern of the total current,[6–8] and of the ring bond currents[9–12] have received considerable attention.

Many state-of-the-art computations of ring magnetic moments produced by the current excited by the bias have already been reported by several authors.[7,13–16] The established quantum transport theory[17–22] yields the currents flowing on each bond as quantum averages of the current operators. This paper deals with the current distribution inside multiply connected circuits excited by the bias and their magnetic effects but we focus on the physical interpretation of those results and on the relation to an actual magnetic measurement performed on the ring. Indeed we argue that different experimental setups will measure markedly different quantities; therefore, we aim to an operational definition suitable to nanorings of molecular dimensions, when one must consider the possibility that the measurement perturb the system.

Below for the sake of definiteness we specialize the discussion to circuits containing one tight-binding ring connected to biased leads. The continuum counterpart deserves a similar discussion. We consider a system which consists of a left (L) and right (R) biased leads connected to a polygonal ring with N sites, see Fig. 1, described by the Hamiltonian[23]

$$\hat{H} = \hat{H}_{\rm ring} + \hat{H}_{\rm L} + \hat{H}_{\rm R} + \hat{H}_{T} + \hat{H}_{\rm bias}. \tag{1}$$

Here, for the quantum ring we use the tight-binding Hamiltonian

$$\hat{H}_{\rm ring} = \sum_{m>n=1}^{N} h_{mn} c_m^\dagger c_n + \text{H.c.} \tag{2}$$

with hopping integrals $h_{mn}=0$ if m and n are not nearest neighbors. Further, in Eq. (1) $\hat{H}_\alpha$ represents the unperturbed α=L,R lead while $\hat{H}_T$ stands for the tunneling Hamiltonian connecting the leads to the ring. The occupation of the system in equilibrium is fixed by the chemical potential μ. The system is driven out of equilibrium by the bias term

$$\hat{H}_{\rm bias} = U_{\rm L}\hat{N}_{\rm L} + U_{\rm R}\hat{N}_{\rm R} \tag{3}$$

with $\hat{N}_\alpha$ the total number operator for electrons in lead α. Eventually it reaches a steady state which is[17] a Slater determinant of current-carrying eigenstates of Eq. (1) with left-going/right-going states populated up to the electrochemical potential $\mu+U_{\rm R/L}$ of the right/left lead. This interplay of equilibrium and out-of-equilibrium terms is also familiar in the partition-free time-dependent formulation[19] where the contribution of the current-carrying states is weighted by the Fermi functions which assign the occupations before the external potential is applied.

It is intuitively clear that the current pattern must be a superposition of a circulating current which produces the ring moment and a laminar one which relates to the interaction of the bias with the external circuit. How to calculate the magnetic moment and hence the circulating current is the main contribution of this work.

The classical definition of the ring magnetic moment $\mathcal{M}_{\rm ring}$ is[24]

$$\mathcal{M}_{\rm ring} = \frac{1}{2N}\sum_{m>n=1}^{N} I_{mn}(\vec{r}_m \wedge \vec{r}_n), \tag{4}$$

where $\vec{r}_m$ is the position of the m site of the ring, R is the radius of the ring, and I_{mn} is the current flowing along the m-n bond. For example, in the interesting special case when the ring is a regular N-sided polygon, we may write,

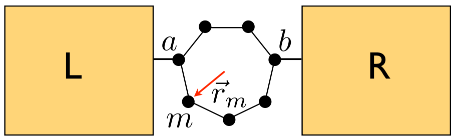

FIG. 1. (Color online) Sketch of a ring with $N=7$ sites in contact with a L and R lead. The site m of the ring is positioned in $\vec{r}_m$. The ring is connected to lead L via site a and to lead R via site b.

$$\mathcal{M}_{\text{ring}} = \frac{R^2}{2}\sin\frac{2\pi}{N}\sum_{m>n=1}^{N} I_{mn} = \frac{S}{N}\sum_{m>n=1}^{N} I_{mn}, \tag{5}$$

where $S=\frac{1}{2}R^2N\sin\frac{2\pi}{N}$ is the area of the ring. In order to evaluate $\mathcal{M}_{\text{ring}}$ we need to know the currents I_{mn} in the internal wires. In quantum mechanics[18,23]

$$\hat{J}_{mn} = -i(h_{mn}c_m^\dagger c_n - h_{nm}c_n^\dagger c_m) \tag{6}$$

is the electron current operator between sites m and n connected by a bond with hopping integral h_{mn}. Such interpretation naturally stems from the continuity equation

$$\frac{d}{dt}\hat{n}_m = \sum_n \hat{J}_{mn} \tag{7}$$

in which the rate of change in the density $\hat{n}_m$ on site m is seen as the sum of the currents flowing from site m to all connected sites n. (In a similar way one obtains the familiar formula for the current density in a continuum system.) We shall call $\hat{J}_{mn}$ the *bond current* operator since it depends on the operators straddling a bond.

For an isolated ring, in the absence of a bias, a current can be excited by a threading magnetic flux; all the sides of the polygon have a common value $I_{mn}\equiv I$, and the classical result (5) we reduces to the elementary formula $\mathcal{M}^{\text{isolated}}=SI$. In state-of-the-art calculations for wired rings[7,13–16] the magnetic moment is calculated by setting in Eq. (4) $I_{mn}=J_{mn}\equiv\langle\hat{J}_{mn}\rangle$, where the average is taken over the many-body current carrying states. Next, we show that such a prescription is questionable.

A. Need for a proper definition of the vortex current

In classical circuits the continuity equation is encoded in Kirchhoff's law, which determines the current together with Ohm's law. Indeed, since the electric field is irrotational, if the bonds have equal conductance Ohm's law requires that the current be irrotational as well as divergenceless; in any case the current is completely specified. On the other hand, in a macroscopic ring connected to leads, one can measure the current in each wire by using an amperometer or by exploring the magnetic field around each branch of the circuit and performing the line integral. To sum up, the current in each wire can be independently measured and is determined by Kirchhoff's and Ohm's laws.

This does not apply to ballistic transport in the quantum regime. For circuits containing loops the continuity equation alone cannot uniquely determine the currents I_{mn}. The current in Eq. (6) solves Eq. (7) but it is not the unique solution. One is free to add a vortex field $J_{mn}\to J_{mn}+V$ for $m>n$ and therefore there is no reason that the physical current flowing through the bond m-n is the same as the expectation value of $\hat{J}_{mn}$. In the continuum case this corresponds to the possibility of adding a vortex field $\vec{V}$, with $\vec{\nabla}\cdot\vec{V}=0$, to the current density. Moreover the current cannot be taken to be defined at a given bond in quantum systems where one cannot tell if the electron goes through the upper or lower branch of the ring.

B. Peierls prescription and symmetric phases

In the tight-binding description an external magnetic flux ϕ piercing the ring is included using the Peierls prescription, i.e.,

$$h_{mn} \to h_{mn}e^{(2\pi i/\phi_0)\int_{\vec{R}_m}^{\vec{R}_n}\vec{A}d\vec{r}} = h_{mn}e^{i(\alpha_{mn}/c)}, \tag{8}$$

where $\phi_0=\frac{hc}{e}$ is the flux quantum, which becomes $\phi_0=2\pi c$, where c is the speed of light, in atomic units $\hbar=1$, $e=1$; the line integral of the vector potential $\vec{A}$ goes from site m to site n; the phases are related to the flux ϕ threaded by the ring by the relation

$$\sum_{m>n=1}^{N}\alpha_{mn} = \oint \vec{A}d\vec{r} = \phi. \tag{9}$$

For an isolated ring, one can regain the classical result $\mathcal{M}^{\text{isolated}}=SI$ by setting

$$\mathcal{M}^{\text{isolated}} = cS\frac{d}{d\phi}\langle\hat{H}_{\text{ring}}\rangle\bigg|_{\phi=0} \tag{10}$$

and using the Hellmann-Feynman theorem; this gives $\mathcal{M}^{\text{isolated}}=SI$ with $I=J\equiv\langle\hat{J}_{mn}\rangle$. Let us try to extend this procedure to the wired rings. The formula in Eq. (4) with $I_{mn}=J_{mn}$ can be obtained as the derivative of the total energy of the system with respect to ϕ at $\phi=0$ provided that we adopt the symmetric phase choice $\alpha_{mn}=\phi/N$ for $m>n$. Indeed, the Hellmann-Feynman theorem now yields,

$$\begin{aligned}\tilde{\mathcal{M}}_{\text{ring}} &\equiv cS\frac{d}{d\phi}\langle\hat{H}\rangle\bigg|_{\phi=0} \\ &= cS\sum_{m>n=1}^{N}\left\langle\frac{d}{d\phi}h_{mn}e^{i(\phi/Nc)}c_m^\dagger c_n + \text{H.c.}\right\rangle\bigg|_{\phi=0} \\ &= \frac{S}{N}\sum_{m>n=1}^{N} J_{mn},\end{aligned} \tag{11}$$

and $S=\frac{1}{2}R^2N\sin\frac{2\pi}{N}$ is the ring area. This agrees with Eq. (5).

Should we feel reassured after this confirmation of the classical formula? The answer is definitely no since this is an *ad hoc* derivation based on contrived assumptions. The magnetic moment $\mathcal{M}_{\text{ring}}$ should only depend on the magnetic flux ϕ or from the flux derivative, not on the specific choice of the phase factors α_{mn}. We illustrate this point by considering for simplicity the triangular ring ($N=3$) with sites a, b, and c connected to one-dimensional leads through sites a and b, see Fig. 2. The formula (11) with $\alpha_{ab}=\phi$ and $\alpha_{bc}=\alpha_{ca}=0$ yields $\tilde{\mathcal{M}}_{\text{ring}}=SJ_{ab}$. On the other hand, the choice $\alpha_{ab}=\alpha_{bc}$

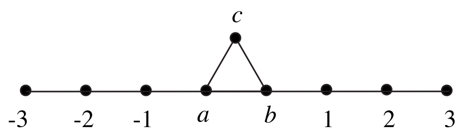

FIG. 2. Sketch of a ring with three sites in contact with two leads. The sites of the ring are labeled with Roman letters.

=0 and $\alpha_{ca}=\phi$ corresponds to the same flux ϕ but yields $\tilde{\mathcal{M}}_{\text{ring}}=SJ_{ca}$. Different choices give different results, which disagree from the result in Eq. (11) corresponding to the symmetric phase choice. In Sec. II C below, we show that the different choices are indeed not gauge equivalent for an infinite system because there is a subtle point about representing the external circuit by infinite wires.

It is evident that the above difficulties constitute the warning light of a physical problem. The solution is physical and we must ask how the magnetic moment could be measured.

II. THOUGHT EXPERIMENTS

The approach introduced originally by Stern and Gerlach to measure magnetic moments uses a weak external *probe* field and a mechanical force measurement. They measured the force acting on the Ag atoms by means of the deflection of the spin-polarized atomic beams. In the case of a wired ring we could think about a direct mechanical experiment performed by an atomic force microscope. We develop a *gedanken* experiment as an idealized version of an actual experiment that one should make in order to give an operational meaning to $\mathcal{M}_{\text{ring}}$. Let us now better specify which force (or torque) one should measure.

While the system is biased, and the current flows, we switch a very weak probe magnetic field of strength B at an angle θ with respect to the normal of the ring. The field must be weak because we want to know $\mathcal{M}_{\text{ring}}$ caused by the current, without the modification that a large field could produce. The probe field produces forces on the ring, and on the whole circuit, that one can measure.

A. Torque acting on the circuit

We denote by ϕ the magnetic flux through the ring and by $|\Psi(\phi)\rangle$ the current carrying state of the system after all transient effects have disappeared. The state $|\Psi(\phi)\rangle$ is therefore an eigenstate of the Hamiltonian in the presence of the magnetic flux which we call $\hat{H}(\phi)$.

The total energy of the infinite system diverges but one could consider

$$E(\phi)=\langle\Psi(\phi)|\hat{H}(\phi)|\Psi(\phi)\rangle-\langle\Psi(0)|\hat{H}(0)|\Psi(0)\rangle, \quad (12)$$

which may be interpreted as the total energy change induced by the field.

The flux depends on the direction of the field and the angular derivatives of $E(\phi)$ define a mechanical torque acting on the system. Let us assume for convenience that the field couples exclusively to the bonds within the ring and therefore the magnetic perturbation is localized. This can be arranged if H_{ring} depends on the field but the rest of H is field independent. Even so, the field in the ring changes the wave function everywhere in the circuit and hence $\langle\Psi(\phi)|\hat{H}(\phi)-\hat{H}_{\text{ring}}(\phi)|\Psi(\phi)\rangle$ does depend on the flux. Physically, this means that the field-free external circuit experiences a torque. If the whole circuit acts as a rigid body and we measure a torque acting on it, we are actually measuring the magnetic moment of the whole system, not the one of the ring. The use of the total energy to calculate the torque experienced by the ring is therefore ambiguous. To measure $\mathcal{M}_{\text{ring}}$ we must arrange a different experiment.

B. Local measurement

We define $\mathcal{M}_{\text{ring}}$ for the connected ring by a magnetic measurement to be performed *in situ* on the ring itself. This means that we perturb the system by inserting a small flux through the ring and measure the resulting torque on the ring itself, e.g., by an atomic force microscope. Although ours is a thought experiment, it is also a suggestion of a real one since the recent developments of technology can make our proposal a practical one. We shall see that the Hamiltonian contains enough information to compute $\mathcal{M}_{\text{ring}}$ since the coupling to a probe field (which is eventually set to zero) via the Peierls prescription encodes all the necessary information. A local experiment requires a measurement of the forces acting on the ring and the result can be interpreted in terms of the ring magnetic moment $\mathcal{M}_{\text{ring}}$. The ring energy is

$$E_{\text{ring}}(\phi)=\langle\Psi(\phi)|\hat{H}_{\text{ring}}(\phi)-\mu\hat{N}_{\text{ring}}|\Psi(\phi)\rangle, \quad (13)$$

where $\hat{N}_{\text{ring}}$ is the number operator of the ring and μ is the equilibrium chemical potential as discussed in Sec. I. Thus one gets the magnetic moment $\mathcal{M}_{\text{ring}}$ directly by measuring

$$E_{\text{ring}}(\phi)-E_{\text{ring}}(0)=-\mathcal{M}_{\text{ring}}B\cos\theta=-\mathcal{M}_{\text{ring}}\frac{\phi}{S}, \quad (14)$$

where S is the ring surface. For isolated rings, $\hat{N}_{\text{ring}}$ is a conserved operator, $\langle\Psi(\phi)|\hat{N}_{\text{ring}}|\Psi(\phi)\rangle$ is flux independent, and therefore one can safely discard the $\mu\hat{N}_{\text{ring}}$ term. For connected rings, the term which references the energy to the chemical potential is needed to ensure the invariance of the theory under a uniform energy shift. Otherwise any shift in the energy origin $\hat{H}_{\text{ring}}\to\hat{H}_{\text{ring}}+\Delta E\hat{N}_{\text{ring}}$ would give a flux-dependent contribution proportional to $\langle\Psi(\phi)|\hat{N}_{\text{ring}}|\Psi(\phi)\rangle$ and hence it would change the magnetic moment, which is absurd.

Note that $\langle\Psi(\phi)|\hat{N}_{\text{ring}}|\Psi(\phi)\rangle$ depends on ϕ, and therefore Eqs. (13) and (14) predict that the term proportional to μ contributes to $\mathcal{M}_{\text{ring}}$. Physically, this must be expected because the insertion of a flux can attract charge of either sign in the wired ring and this effect must be accounted for.

Since forces are easier to measure than energies, we propose measuring $\mathcal{M}_{\text{ring}}$ as

$$\mathcal{M}_{\rm ring} = - S\frac{dE_{\rm ring}}{d\phi}\bigg|_{\phi=0}. \quad (15)$$

We have stressed that no magnetic field is needed since the current is excited by the bias, however in the presence of flux ϕ the obvious extension is

$$\mathcal{M}_{\rm ring} = - S\frac{dE_{\rm ring}}{d\phi}\bigg|_{\phi}. \quad (16)$$

The magnetic moment is due to a current

$$I_{\rm ring} = -\frac{c}{S}\mathcal{M}_{\rm ring} \quad (17)$$

that will be referred to below as the circulating current. Physically one expects that at least the flux due to self-induction should be introduced, and since this flux is proportional to the current in the ring, flux and current should be found self-consistently. However we defer this development to future work. For isolated rings, one can take $\mu=0$ and get back the formula (10) which represents the persistent current response of an isolated ring by an external magnetic field.

C. Ring local gauge invariance

There is another strong reason why the local definition in Eq. (15) using $H_{\rm ring}$ is suitable for the ring moment while the global definition (with H instead of $H_{\rm ring}$) is not. Both definitions are expressed in terms of observables and are obviously gauge invariant. The magnetic moment $\mathcal{M}_{\rm ring}$ in Eq. (15), however, enjoys a more subtle invariance, that allows us to choose the Peierls phases within the ring as we please provided Eq. (9) is fulfilled, without keeping into account what happens at large distances, in the rest of the system. It is a stronger condition than the standard gauge invariance.

Consider the patterns in Fig. 3. In a1, a2, and a3 the ring is connected to wires with open-boundary condition, in b1, b2, and b3 the external circuit is considered. An arrow along the m-n bond means that $\int_{\vec{R}_m}^{\vec{R}_n}\vec{A}d\vec{r}=\phi$, no arrow means that the hopping integral is real; ai and bi have the same choice of phases. Two configurations are equivalent (i.e., connected by a gauge transformation) if the flux threaded in any closed path is the same. We shall use the symbol $\equiv$ to denote this equivalence. By inspection, $a1\equiv a2\equiv a3$ and $b2\equiv b3$ but the external circuit is threaded by a flux ϕ in b1 and by a flux 0 in b2 and b3. Physically, one expects that the ring has a well-defined magnetic moment but in the case of a ring connected to leads there are different gauge-inequivalent ways to insert the flux. The different choices imply threading flux in the external circuit or not, and yield different total energies in Eq. (12), and hence different magnetic moments. It is necessary to know the geometry of the field and circuit in order to evaluate the total energy given in Eq. (12). This confirms our interpretation that Eq. (12) does not bring unambiguous information to extract the ring magnetic moment.

On the contrary the invariance of Eq. (13) follows from the fact that the operator $\hat{H}_{\rm ring}(\phi)-\mu\hat{N}_{\rm ring}$ is a local operator and hence its average only depends on the projection onto the ring of the single particle states $\{\Psi_k\}$ forming the Slater

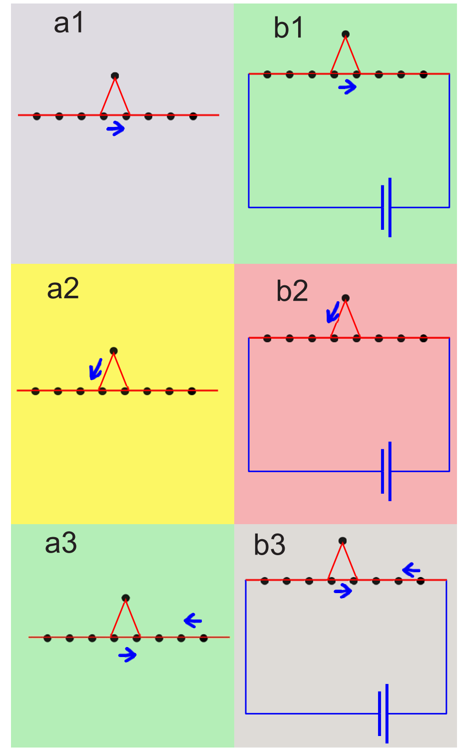

FIG. 3. (Color online) Flux patterns for a ring inserted in a wire with open boundary conditions (a1, a2, and a3) and in a closed circuit (b1, b2, and b3). The arrows mark the bonds where the hopping integral is complex (see text).

determinant $|\Psi(\phi)\rangle$. The effects of any flux concatenated with the circuit is removed in this way. For example, the single-electron wave functions which make up the many-body wave functions undergo a phase change in the trip around the external circuit in b1 and not in b2; however the solution at a given energy can be taken to be locally the same in the ring region, despite the change in a distant bond.

D. Magnetic moment of the ring

Equation (15) for $\mathcal{M}_{\rm ring}$ with $E_{\rm ring}$ in Eq. (13) is the central result of this work. It does not suffer from any of the problems of the standard approach. The ring current, which is not uniquely determined by the continuity equation, is now fixed by the dynamics. It represents the forces acting on the ring acting as a rigid body hinged on the circuit and is independent of all interactions of the field far from the ring. Below we develop a many-body theory by Green's-function technique to calculate $\mathcal{M}_{\rm ring}$ in Eq. (15) and explore its physical contents.

III. GREEN'S-FUNCTION APPROACH

In order to compute the magnetic moment in Eq. (15) for the system described by Hamiltonian in Eq. (1) we need to calculate the expectation value of $E_{\rm ring}$ over the stationary state of the current carrying system in the presence of a flux ϕ. The lesser Green's function $G^<(t,t')$ which describes such stationary nonequilibrium state depends on the time difference $t-t'$. In terms of its Fourier transform $G^<(\omega)$,

$$E_{\rm ring}(\phi) = -i\int \frac{d\omega}{2\pi}{\rm Tr}_{\rm ring}[(h-\mu)G^<(\omega)], \qquad (18)$$

where h has elements $h_{mn}e^{i\alpha_{mn}/c}$ and the trace is taken over the sites of the ring. It is convenient to obtain $G^<(\omega)$ by an embedding technique with $\Sigma^< = \Sigma^<_{\rm L} + \Sigma^<_{\rm R}$ the lesser embedding self-energies of the left and right leads. Thus,

$$G^<(\omega) = G^{\rm R}(\omega)[\Sigma^<_{\rm L}(\omega) + \Sigma^<_{\rm R}(\omega)]G^{\rm A}(\omega). \qquad (19)$$

The retarded/advanced Green's function projected onto the ring is $G^{\rm R/A}(\omega) = [\omega - h - \Sigma^{\rm R/A}(\omega)]^{-1}$. Using the fluctuation-dissipation theorem for lead $\alpha = {\rm L},{\rm R}$ one obtains for the lesser embedding self-energy $\Sigma^<_\alpha(\omega) = -2if(\omega - U_\alpha)\,{\rm Im}[\Sigma^{\rm R}_\alpha(\omega)]$, where $f(\omega)$ is the Fermi distribution function at chemical potential μ. The retarded and advanced components are related as $\Sigma^{\rm R}_\alpha = [\Sigma^{\rm A}_\alpha]^\dagger$ with $\alpha = {\rm L},{\rm R}$. Substituting Eq. (19) in Eq. (18) and taking the flux derivative at $\phi = 0$ we obtain for the magnetic moment

$$\mathcal{M}_{\rm ring} = iS\int \frac{d\omega}{2\pi}{\rm Tr}_{\rm ring}\left[\frac{dh}{d\phi}G^{\rm R}\Sigma^< G^{\rm A} + (h-\mu)G^{\rm R}\right.$$
$$\left.\times\left(\frac{dh}{d\phi}G^{\rm R}\Sigma^< + \Sigma^< G^{\rm A}\frac{dh}{d\phi}\right)G^{\rm A}\right]_{\phi=0}. \qquad (20)$$

As already noted, the first term in the square brackets yields a linear combination of the bond currents, combination that depends on the Peierls phase configuration. The independence of $\mathcal{M}_{\rm ring}$ from the phase choice is restored by adding $(h-\mu)\frac{dG^<(\omega)}{d\phi}$ which is explicitly given by the second term in Eq. (20). We wish to emphasize that for $\phi = 0$ the magnetic moment has no diamagnetic contribution.

IV. RESULTS AND DISCUSSION

We consider one-dimensional tight-binding leads as in Fig. 2 with nearest-neighbor hopping $t_{\rm lead}$ described by the Hamiltonian

$$\hat{H}_{\rm L} = t_{\rm lead}\sum_{j=-\infty}^{-1}(d^\dagger_j d_{j-1} + {\rm H.c.}),$$

$$\hat{H}_{\rm R} = t_{\rm lead}\sum_{j=1}^{\infty}(d^\dagger_j d_{j+1} + {\rm H.c.}), \qquad (21)$$

connected to the triangular ring via the tunneling term

$$\hat{H}_T = t_{\rm lead}(d^\dagger_{-1}c_a + d^\dagger_1 c_b) + {\rm H.c.} \qquad (22)$$

The embedding self-energies have only one nonzero matrix element, namely, $[\Sigma^{\rm R}_{\rm L}(\omega)]_{ij} = \delta_{ia}\delta_{ja}\sigma(\omega - U_{\rm L})$ and $[\Sigma^{\rm R}_{\rm R}(\omega)]_{ij} = \delta_{ib}\delta_{jb}\sigma(\omega - U_{\rm R})$, where $U_{\rm L} = -U$ and $U_{\rm R} = U$. The function $\sigma(\omega)$ can be easily calculated and reads

$$\sigma(\omega) = \frac{1}{2}\left[(\omega + i\eta) - \frac{(\omega + i\eta) + 2t_{\rm lead}}{\sqrt{1 + \frac{4t_{\rm lead}}{(\omega + i\eta) - 2t_{\rm lead}}}}\right]. \qquad (23)$$

Here and below all energies and currents are measured in units of $t_{\rm lead}$. In Fig. 4 we display the magnetic moment

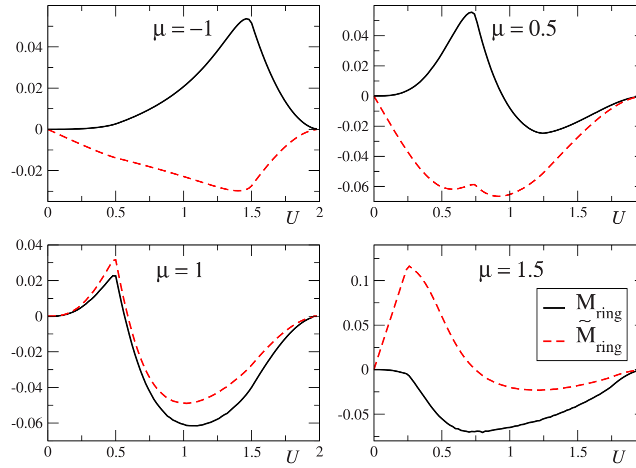

FIG. 4. (Color online) Plot of $\mathcal{M}_{\rm ring}$ (black solid line) and $\tilde{\mathcal{M}}_{\rm ring} = \frac{1}{3}(J_{ab} + J_{bc} + J_{ca})$ (red, dashed line) as obtained in Eq. (5) with $I_{mn} = J_{mn}$. All magnetic moments are in units of $St_{\rm lead}/c$.

$\mathcal{M}_{\rm ring}$ of Eq. (20) as a function of the bias U for different values of the chemical potential $\mu = -1.5, 0, -1.0, 0.5, 1.0$. We compare the results to the magnetic moment $\tilde{\mathcal{M}}_{\rm ring}$ obtained by the bond current according to Eq. (5) evaluated at $I_{mn} = J_{mn}$. The bond currents are computed using a Landauer-type formula.[12] All currents vanish for bias $U = 2$ as the right continuum is lifted by 2 while the left continuum is lowered by the same amount. Since the bandwidth is 4 the bias $U = 2$ represents the minimum value of U for which there is no more overlap between the left and right continua. The maximum of $\mathcal{M}_{\rm ring}$ versus bias U is seen in Fig. 4 to shift to lower U with increasing μ. For $\mu > 0$ the maximum of $\mathcal{M}_{\rm ring}$ and of $\tilde{\mathcal{M}}_{\rm ring}$ occurs at the same value of U. For $\mu < 0$ the maximum of $\mathcal{M}_{\rm ring}$ occurs at the minimum of the bond currents, Fig. 4.

By numerical inspection we have observed that the derivative of $\mathcal{M}_{\rm ring}$ as function of U at zero bias is always vanishing. It is interesting to dwell on the meaning of this finding. On physical grounds, a circulating current should be localized and should not change the average number of electrons $\hat{N}_\alpha$ in lead $\alpha = {\rm L}/{\rm R}$. As a consequence the first-order response in $\mathcal{M}_{\rm ring}(t)$ to the bias should vanish for $t \to \infty$. According to the Kubo formula the response function is given by the average over the unbiased ground state of the commutator between the operator of the *cause* at time 0 and the one of the *effect* at time t. In the Hamiltonian in Eq. (1) the cause is the bias operator $\hat{H}_{\rm bias}$ while the effect (in the Heisenberg picture with respect to the unbiased Hamiltonian) is $\tilde{\mathcal{M}}_{\rm ring}(t)$. To first order this means that

$$\int_{-\infty}^{t}[\hat{\mathcal{M}}_{\rm ring}(\tau), U_{\rm L}\hat{N}_{\rm L} + U_{\rm R}\hat{N}_{\rm R}]d\tau = 0, \qquad (24)$$

where $\hat{\mathcal{M}}_{\rm ring}$ is the operator of the magnetic moment and $\hat{N}_\alpha(t)$ is in the Heisenberg picture with respect to the unbiased Hamiltonian. This can be true for any $U_{\rm L}, U_{\rm R}$ only if

$$\int_{-\infty}^{t} [\hat{\mathcal{M}}_{\rm ring}(\tau),\hat{N}_{\rm L}]d\tau = 0 \tag{25}$$

and

$$\int_{-\infty}^{t} [\hat{\mathcal{M}}_{\rm ring}(\tau),\hat{N}_{\rm R}]d\tau = 0. \tag{26}$$

The above relations imply that the circulating current does not carry charge from one wire to the other and should be at least quadratic in the applied bias. By contrast, the standard theory of Eq. (4) with $I_{mn}=J_{mn}$ usually predicts in general a linear response. On the other hand, the laminar current is not coupled to the local magnetic field.

We have checked numerically that at small U $\tilde{\mathcal{M}}_{\rm ring}$ is generally linear in U, while $\mathcal{M}_{\rm ring} \propto U^3$, see Fig. 3. The absence of the quadratic term in Fig. 3 is due to the high symmetry of the system, in which we have chosen $U_{\rm L}=-U_{\rm R}=-U$. Indeed letting the system lie on the xy plane, with the wires along the x axis, the reflection $(x,y)\to(-x,y)$ is equivalent to $U\to -U$ and hence both $\mathcal{M}_{\rm ring}$ and $\tilde{\mathcal{M}}_{\rm ring}$ are odd function of U. We have numerically verified that for $U_{\rm L} \neq -U_{\rm R}$ the magnetic moment has a quadratic contribution at small bias.

We note that there are special values of μ (e.g., $\mu=1$) at which the estimate $\tilde{\mathcal{M}}_{\rm ring}$ starts with vanishing U derivative at small U.[12] In this case $I_{\rm ring}$ coincides with the bond current along the *c*-*a* bond *for all* values of U. Since $I_{\rm ring}$ yields the circulating current, we conclude that all the laminar current goes along the *a*-*b* bond.

In a similar way one can compute $\mathcal{M}_{\rm ring}$ for rings with N sites, arms of different length and different hopping as well as on-site energy parameters. We have verified that $\mathcal{M}_{\rm ring}=0$ for symmetrically connected rings, as physically expected.

V. CONCLUSIONS

The definition of the magnetic moment of a quantum ring connected to bias leads must comply with the way it is measured. One can measure the magnetic field produced by the ring, e.g., by means of a superconducting quantum interference device (SQUID), or the response of the ring to an external field. However the SQUID is at least a mesoscopic object, and has not been used so far to measure the magnetic moment of molecular-sized objects; in addition the external circuit also produces a magnetic field. We argue that a local mechanical measurement (of a force or a torque) by an atomic force microscope would be ideally suitable for a nanoscopic ring. By thought experiments we propose $\mathcal{M}_{\rm ring}$ in Eq. (14) which depends on the interaction energy with an external field and does not involve the bond currents (as we call the averages $\langle \hat{J}_{mn}\rangle$ of the current operator with m,n in an internal bond of the ring). Explicit calculations show that $\mathcal{M}_{\rm ring}$ is, in general, quite unlike a linear combination of the bond currents, except for special situations when the conductance of the ring vanishes.

Remarkably we have found that the circulating current generating the magnetic moment has an at least quadratic rather than linear response at small bias. Such circulating current is localized inside the ring and does not contribute to the conduction while the linear-response current is always laminar and contributes to the overall Lorentz force acting on the circuit.

Since the effects that we predict are qualitatively important, we hope that they can be directly compared with experiment by measuring the ring magnetic moments. We point out that for more complex systems with several rings the individual magnetic moment of each ring makes sense if one can thread the field through and measure the mechanical force on a single ring. Otherwise one should model the experiment according to the detailed way it is performed. However, more complex systems are outside the scope of the present paper.

Finally we expect that the present theory and its extensions to circuits with several loops should pave the way to include induction and self-induction effects in quantum transport theory. In the extended theory, even in the absence of an external magnetic field one will need to include a flux $\phi=LI_{\rm ring}$, where L is the self-induction coefficient, by a self-consistent procedure.

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