Nonequilibrium orbital magnetization of strongly localized electrons

Y. Galperin\textsuperscript{(a)} and O. Entin-Wohlman\textsuperscript{(b)}

\textsuperscript{(a)}Department of Physics, University of Oslo, P. O. Box 1048 Blindern, 0316 Oslo, Norway and A. F. Physico-Technical Institute, 194021 St. Petersburg, Russia

\textsuperscript{(b)}School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel

(Received November 3, 2018)

The magnetic response of strongly localized electrons to a time-dependent vector potential is considered. The orbital magnetic moment of the system, away from steady-state conditions, is obtained. The expression involves the tunneling and phonon-assisted hopping currents between localized states. The frequency and temperature dependence of the orbital magnetization is analyzed as function of the admittances connecting localized levels. It is shown that quantum interference of the localized wave functions contributes to the moment a term which follows adiabatically the time-dependent perturbation.

I. INTRODUCTION

The explanation of the orbital magnetic moment at thermal equilibrium in terms of “ring currents” has been known for more than half a century. The most studied configuration is that of a small mesoscopic structure penetrated by a constant magnetic flux, in which the electrons move ballistically or diffusively. Recently, the problem of the ring currents and the associated magnetic moment have been addressed in the case of strong disorder. In this regime the electronic states are localized and their overlaps are small. It has been shown that since the coupling of the electrons to the thermal bath enables resonant processes, there appears a current flowing in opposite sense to the one due to tunneling, which exists in the absence of the electron-phonon interaction. This counter-current has been related to the dc Hall effect in insulators.

The study of the orbital magnetic response to a time-dependent flux is of interest by itself and also because it is relevant to experiments designed to detect persistent currents in mesoscopic rings. Nonequilibrium magnetization may be used to measure specific electronic relaxation rates. The magnetic moment in response to a time-dependent flux has been discussed so far in the context of weak disorder.

In this paper we investigate the response of strongly localized electrons to a time-dependent flux, which sets up a magnetic moment in the system. We obtain the nonequilibrium orbital magnetization and study it when the electrons are coupled to the thermal bath.

The calculation is based on the Kubo approach that yields, for strongly localized electrons, the current between two localized states in response to the time-dependent flux. The proper definition of the orbital magnetization away from steady-state conditions, in terms of these currents, is a somewhat delicate point. We devote the next section to a discussion of this issue. In Sec. III we obtain the current that flows between two localized states. Section IV contains the analysis of the characteristic frequencies that dominate the nonequilibrium magnetization of an isolated group of three localized states. (This is the smallest cluster which can have an orbital moment.) Section V contains concluding comments.

The formalism we develop allows for the calculation of the ac conductance of strongly localized electrons coupled to a thermal bath. We derive in the Appendix quantum interference corrections to the ac hopping conductance and show that they yield nonlocal contributions to the ‘bond’ admittances.

II. NONEQUILIBRIUM MAGNETIZATION OF LOCALIZED ELECTRONS

Our aim is to obtain an expression for the orbital magnetic moment of localized electrons, at nonequilibrium conditions. We will show that the moment can be given in terms of the currents between the localized states. When these currents flow in response to a constant magnetic field, i.e., at equilibrium, this is a straightforward task. In that case the equilibrium value of the orbital magnetization, \( \bar{M}_{\text{eq}} \), is just a manifestation of the Aharonov-Bohm effect and can be obtained from the thermodynamic potential. The result reads

\[
\bar{M}_{\text{eq}} = -\frac{1}{2e} \sum_{\langle j \ell \rangle} I_{\ell j} |\vec{R}_\ell \times \vec{R}_j|,
\]

in which \( I_{\ell j} \) is the current between the state localized at \( \vec{R}_\ell \) and the state localized at \( \vec{R}_j \), and \( \langle j \ell \rangle \) indicates a sum over pairs. At equilibrium, current conservation implies that the ‘bond’ currents \( I_{\ell j} \) satisfy \( \sum_{\ell} I_{\ell j} = 0 \), which in turn yields that \( \bar{M}_{\text{eq}} \) is independent of the choice of the coordinate origin. At nonequilibrium one has

\[
\dot{\rho}_j = \sum_{\ell} I_{\ell j},
\]
where \( \dot{\rho}_j \) is the time derivative of the electron occupation on the \( j \)-th localized state. The contribution of the dielectric currents resulting from the nonequilibrium occupations has to be included in the orbital magnetic moment. For an homogeneous continuous system, this is achieved by introducing a single vector function that encompasses both the (spatial-dependent) charge and the current densities. In the present case, the analogous quantity is

\[
\mathcal{J}_{\ell j} = I_{\ell j} + \frac{1}{N}(\dot{\rho}_\ell - \dot{\rho}_j),
\]

where \( N \) is the number of localized states. This is so because the generalized currents \( \mathcal{J}_{\ell j} \) satisfy

\[
\sum_{\ell \neq j} \mathcal{J}_{\ell j} = 0
\]

which follows from the requirement of charge conservation (\( \sum_j \dot{\rho}_j = 0 \)). Note that the requirement (4) corresponds to a current density with zero divergence in the case of a homogeneous continuous system. As a result, the nonequilibrium orbital magnetic moment, \( \vec{M} \), is given by

\[
\vec{M} = -\frac{1}{2c} \sum_{\ell \neq j} \mathcal{J}_{\ell j} [\vec{R}_\ell \times \vec{R}_j]
\]

and is also independent of the choice of the coordinate origin. It takes a particularly convenient form when the relative coordinates \( \vec{r}_\ell \)

\[
\vec{r}_\ell = \vec{R}_\ell - \vec{R}, \quad \vec{R} = \frac{1}{N} \sum_j \vec{R}_j,
\]

are introduced (\( \vec{R} \) is the coordinate of the center of gravity). Then, using Eqs. (3) and (4) we obtain

\[
\vec{M} = -\frac{1}{2c} \sum_{\ell \neq j} I_{\ell j} [\vec{r}_\ell \times \vec{r}_j].
\]

In this expression the radius-vectors \( \vec{R}_\ell \) appearing in (8) are replaced by the relative coordinates \( \vec{r}_\ell \). It is seen that for the choice (8) the displacement currents disappear from the expression for \( \vec{M} \).

**III. THE LINEAR RESPONSE EXPRESSION FOR THE CURRENT**

The properties of strongly localized electrons interacting with a phonon bath are most conveniently discussed using the Hamiltonian introduced by Holstein. In that description the electron site density is coupled to the ion displacements. (The term ‘site’ denotes a localized state.) The Holstein Hamiltonian, in the presence of a vector potential, can be written in the form

\[
\mathcal{H} = \sum_\ell \epsilon_\ell c_\ell^\dagger c_\ell + \sum_q \omega_q b_q^\dagger b_q + \sum_{\ell j} J_{\ell j} Q_{\ell j} e^{i\phi_{\ell j}} c_\ell^\dagger c_j, \tag{8}
\]

in which \( Q_{\ell j} \) results from the electron-phonon interaction

\[
Q_{\ell j} = \exp \left[ -\sum_q \frac{\omega_q}{\omega_q} (b_q - b_q^\dagger) \right], \quad v_q^{\ell j} = v_q^\ell - v_q^j. \tag{9}
\]

Here \( c_\ell \) and \( c_j^\dagger \) are the annihilation and creation operators of an electron in the state localized on site \( \ell \) and \( \epsilon_\ell \) are the on-site energies, assumed to be randomly distributed (energies are measured from the Fermi level). The phonon operators are denoted \( b_q \) and \( b_q^\dagger \), and \( \omega_q \) is the phonon energy. In Eq. (9), \( J_{\ell j} \) is the overlap of two electronic wave functions localized on sites \( j \) and \( \ell \), and \( v_q^\ell \) is the matrix element for the electron-phonon interaction on site \( \ell \). The strong localization regime is characterized by \( |J_{\ell j}| \ll |\epsilon_\ell - \epsilon_j| \). The transfer of an electron between two sites, as described by the last term of \( \mathcal{H} \), can be accomplished by tunneling, or can be accompanied by the exchange of one or more phonons with the thermal bath (as is evident by expanding \( Q_{\ell j} \) in terms of \( v_q^{\ell j} \)).

In this description the vector potential applied to the system appears as a phase factor, \( e^{i\phi_{\ell j}} \), in which \( \phi_{\ell j} \) is the time-dependent flux (in units of the flux quantum \( \hbar/2e \)) contained in the area \( [\vec{R}_\ell \times \vec{R}_j]/2 \), where \( \vec{R}_\ell \) is the radius vector to the site \( \ell \). The effect of this flux is treated as a perturbation. Accordingly, we re-write the Hamiltonian (9) in the form

\[
\mathcal{H} = \mathcal{H}_e + \mathcal{H}_{ph} + \mathcal{H}_1 + \mathcal{H}_{per}, \tag{10}
\]

in which

\[
\mathcal{H}_e = \sum_\ell \epsilon_\ell c_\ell^\dagger c_\ell, \quad \mathcal{H}_{ph} = \sum_q \omega_q b_q^\dagger b_q,
\]

\[
\mathcal{H}_1 = \sum_{\ell j} J_{\ell j} Q_{\ell j} e^{i\phi_{\ell j}} c_\ell^\dagger c_j, \tag{11}
\]

refer to the unperturbed system, and

\[
\mathcal{H}_{per} = \sum_{\ell j} J_{\ell j} Q_{\ell j} e^{i\phi_{\ell j}}(t)c_\ell^\dagger(t)c_j(t). \tag{12}
\]

The current flowing in response to the time-dependent flux can be obtained by using the expression

\[
\dot{I}_{\ell j} = 2e\Im J_{\ell j} Q_{\ell j} e^{i\phi_{\ell j}} c_\ell^\dagger c_j \tag{13}
\]

for the current operator. Then linear response theory yields that the net current between sites \( j \) and \( \ell \) is

\[
I_{\ell j}(t) = \int_{-\infty}^{t} dt_1 \left\langle \left[ \mathcal{H}_{per}(t_1), 2\Im J_{\ell j} Q_{\ell j}(t)c_\ell^\dagger(t)c_j(t) \right] \right\rangle + \phi_{\ell j}(t)(2\Re J_{\ell j} Q_{\ell j}(t)c_\ell^\dagger(t)c_j(t)). \tag{14}
\]
In this expression the time dependences of the operators, as well as the thermal averages (indicated by the angular brackets) are determined by the Hamiltonian $\mathcal{H}_e + \mathcal{H}_{ph} + \mathcal{H}_t$, Eqs. (11).

We now derive an explicit expression for the current, valid in the strong localization regime. As this regime is characterized by the small parameter $|J_{j\ell}|/|\epsilon_j - \epsilon_{\ell}|$, we expand $I_{j\ell}$ in powers of the wave-function overlaps. Clearly, [cf. Eqs. (2) and (3)] the expansion starts at order $J^2$. Quantum interference effects appear first in order $J^3$, which allows for the interference of the direct amplitude $j \rightarrow \ell$ with the indirect one, via a third site $k$.

$$I_{j\ell}^{(2)}(\omega) = e|J_{j\ell}|^2 \int_{-\infty}^{t} dt_1 \phi_{j\ell}(t_1) \left( n_j(1-n_\ell)23e^{i\epsilon_j(t-t_1)}\langle Q_{j\ell}(t_1)Q_{j\ell}(t) \rangle - n_\ell(1-n_j)23e^{i\epsilon_\ell(t-t_1)}\langle Q_{j\ell}(t)Q_{j\ell}(t_1) \rangle \right)$$

$$- e|J_{j\ell}|^2 \phi_{j\ell}(t) \int_{-\infty}^{t} dt_1 \left( n_j(1-n_\ell)23e^{i\epsilon_j(t-t_1)}\langle Q_{j\ell}(t_1)Q_{j\ell}(t) \rangle - n_\ell(1-n_j)23e^{i\epsilon_\ell(t-t_1)}\langle Q_{j\ell}(t)Q_{j\ell}(t_1) \rangle \right),$$

(15)

where we have used $\phi_{j\ell} = -\phi_{\ell j}$. (The superscript (2) indicates a result to order $J^2$.) Here $n_j$ denotes the equilibrium electron occupation of site $j$, $n_j = (e^{\beta\epsilon_j} + 1)^{-1}$ and $\epsilon_{j\ell} = \epsilon_{\ell} - \epsilon_j$.

The time-dependent correlators of the phonon operators $Q_{j\ell}$ are calculated with respect to the free phonon Hamiltonian, $\mathcal{H}_{ph}$. One finds

$$\langle Q_{j\ell}(t)Q_{j\ell}(t_1) \rangle = e^{2g(t-t_1)-2g(0)},$$

$$g(t) = \sum_\mathbf{q} \frac{|v_\mathbf{q}|^2}{\omega_\mathbf{q}} \left[ (1+N_\mathbf{q})e^{-i\omega_\mathbf{q}t} + N_\mathbf{q}e^{i\omega_\mathbf{q}t} \right],$$

(16)

where $N_\mathbf{q} = (e^{\beta\omega_\mathbf{q}} - 1)^{-1}$ is the Bose function. Here we have taken $v_\mathbf{q}^2\omega_\mathbf{q}^2 \sim \delta_{jk}|v_\mathbf{q}|^2$, which is based on $v_{\ell j}^k = v_\mathbf{q}\exp(i\mathbf{q} \cdot \mathbf{R}_\ell)$. We now insert Eq. (14) into Eq. (2) and put

$$\phi_{j\ell}(t) = \bar{\phi}_{j\ell} e^{i\omega t},$$

(17)

where $\bar{\phi}_{j\ell}$ is the amplitude of the ac flux $\phi_{j\ell}$. It then follows that

$$I_{j\ell}^{(2)}(t) = I_{j\ell}^{(2)}(\omega) e^{i\omega t},$$

(18)

with the amplitude $I_{j\ell}^{(2)}$ given by

$$I_{j\ell}^{(2)}(\omega) = e\bar{\phi}_{j\ell}|J_{j\ell}|^2 \int_{0}^{\infty} dt \left( 1 - e^{-i\omega t} \right) \times \left[ n_j(1-n_\ell)23e^{i\epsilon_j(t+2g(t)-2g(0))} - n_\ell(1-n_j)23e^{i\epsilon_\ell(t+2g(t)-2g(0))} \right].$$

(19)

One notes that $I_{j\ell}^{(2)}(\omega)$ vanishes as the frequency tends to zero. In other words, to order $J^2$ the current is driven by the time dependence of the flux. This is not the case in higher orders in the overlaps. We show in the Appendix that due to quantum interference of the electron wave functions (which appear first to order $J^3$), the bond currents and consequently the orbital magnetic moment have a part that follows adiabatically the external flux.

One can now define an effective admittance $z_{j\ell}^{(2)}$ for the $j\ell$ bond

$$z_{j\ell}^{(2)}(\omega) = \frac{e\bar{\phi}_{j\ell}|J_{j\ell}|^2}{\omega}. $$

(20)

For weak electron-phonon coupling, the real part of the admittance becomes

$$\Re z_{j\ell}^{(2)} = e^2|J_{j\ell}|^2 n_j(1-n_\ell)\gamma_{j\ell}(\omega),$$

(21)

with

$$\gamma_{j\ell}(\omega) = \frac{1}{2\omega} \left[ (1-e^{-\beta\omega}) \int_{-\infty}^{\infty} dt e^{i(\epsilon_{j\ell}+\omega)t}(1+g(t)) \right. + (e^{\beta\omega} - 1) \left. \int_{-\infty}^{\infty} dt e^{i(\epsilon_{j\ell}-\omega)t}(1+g(t)) \right],$$

(22)

(It is assumed that the localized levels are not accidentally degenerate.) It is seen that two types of transitions between the levels $\ell$ and $j$ contribute to $\Re z_{j\ell}^{(2)}$, resonant transitions and non-resonant, phonon-assisted ones that exist due to the coupling with the thermal bath. At low frequencies such that $\beta\omega \ll 1$ we have

$$\Re z_{j\ell}^{(2)} = e^2|J_{j\ell}|^2 n_j(1-n_\ell)\pi \left[ \delta(\epsilon_{j\ell} + \omega) + \delta(\epsilon_{j\ell} - \omega) \right] + G_{j\ell},$$

(23)
with $G_{jl}$ being the dc bond conductance. At low temperatures such that $\beta|\epsilon| \gg 1$, it becomes

$$G_{jl} = e^2\beta|J_{jl}|^2 \frac{\alpha D(|\epsilon_{jl}|)}{\epsilon_{jl}^2} e^{-\frac{2}{\beta}|\epsilon_{jl}| + |\epsilon| + |\epsilon_{jl}|}. \quad (24)$$

Here $v$ is the interaction matrix element for the electron-phonon coupling, and $D$ is the phonon density of states. The resonant transitions, described by the first term in (24), require that the probability to find a resonating pair of sites will not vanish. In a bulk system this mechanism may therefore be expected to contribute. The full quantum-mechanical analysis of an isolated pair of sites, valid for arbitrary frequencies, is given e.g. in Ref. [14].

The imaginary part of $z_j^{(2)}$ is associated with the dielectric response of the $jl$ bond, and is out-of-phase with the real term. In the absence of the coupling to the thermal bath it becomes

$$\Im z_j^{(2)}(\omega) = e^2|J_{jl}|^2 \frac{n_j - n_l}{\epsilon_{jl}} \frac{\omega}{\varepsilon_{jl}^2 - \omega^2}. \quad (25)$$

The coupling with the phonons introduces additional contributions to the dielectric response which are ignored here for simplicity.

IV. ANALYSIS OF THE MAGNETIC MOMENT

In exploiting the Kubo formula, one should interpret the driving force as an effective one, which arises from the externally applied field and the fields induced in the system. This means that the time derivative of the flux should incorporate the changes in the local chemical potentials. Using the relative coordinates introduced in (3) one thus has

$$i\omega \delta_{jl} = i\frac{e}{2c} \vec{H} \cdot (\vec{r}_j \times \vec{r}_l)$$

$$-i\omega \frac{e}{2c} (\vec{H} \times \vec{R}) \cdot (\vec{r}_j - \vec{r}_l) + \delta \mu_j - \delta \mu_l, \quad (26)$$

in which $\delta \mu_j$ is the change of the chemical potential at site $j$. The first term results from the time dependence of the magnetic field $\vec{H}$. In fact, $(e/2c)\vec{H} \cdot (\vec{r}_j \times \vec{r}_l)$ is just the magnetic flux (divided by the flux quantum $\phi_0$) acquired from the magnetic field between sites $j$ and $l$. The second term can be interpreted as arising from an electric field, $\vec{E} = i\omega (\vec{H} \times \vec{R})/2c$. Accordingly, the last three terms in (26) represent the local electro-chemical potential difference.

To determine the local chemical potentials, and consequently the bond currents, we need to solve the set of equations (3), where

$$\dot{\rho}_j = i\omega e(n_j - n_l)\delta \mu_j,$$  

and $J_{lj}$ is given by Eq. (20). (We omit the superscript (2) for brevity.) Thus we have

$$i\omega C_j \delta \mu_j = \sum_{\ell} \zeta_{j\ell} \left( i\omega \frac{e}{2c} \vec{H} \cdot (\vec{r}_j \times \vec{r}_\ell) - i\omega \frac{e}{2c} (\vec{H} \times \vec{R}) \cdot (\vec{r}_j - \vec{r}_\ell) + \delta \mu_j - \delta \mu_\ell \right), \quad (28)$$

where

$$C_j = e^2\beta n_j (1 - n_j) \quad (29)$$

is the capacitance connected to the $j$th site. As the calculation is restricted to terms linear in the magnetic field $\vec{H}$ the second term on the right-hand side of (28) can be omitted since it vanishes upon averaging over the site locations. It follows that the local chemical potentials and consequently the currents are determined by the magnetic fluxes $\varphi_{jl}$

$$\varphi_{jl} = \frac{e}{2c} \vec{H} \cdot (\vec{r}_j \times \vec{r}_l). \quad (30)$$

The magnetic moment, Eq. (7), is given in terms of the bond currents. To find those, we use Eqs. (3) to write (cf. Eq. (28))

$$\delta \mu_j = \frac{e}{i\omega C_j} \sum_{\ell} I_{j\ell}. \quad (31)$$

We then obtain

$$I_{\ell_1,\ell_2} = \frac{z_{\ell_1,\ell_2}}{e} \left( i\omega \varphi_{\ell_1,\ell_2} \right.$$  

$$+ \frac{e}{i\omega C_{\ell_1}} \sum_{j} I_{j\ell_1} - \frac{e}{i\omega C_{\ell_2}} \sum_{j} I_{j\ell_2} \right) \quad (32)$$

This can be rearranged to yield

$$I_{\ell_1,\ell_2} \left( 1 + \frac{z_{\ell_1,\ell_2}}{e} \frac{1}{C_{\ell_1}} \right) + \frac{z_{\ell_1,\ell_2}}{i\omega C_{\ell_1}} \sum_{j \neq \ell_1} I_{j\ell_2}$$

$$+ \frac{z_{\ell_1,\ell_2}}{i\omega C_{\ell_2}} \sum_{j \neq \ell_2} I_{j\ell_1} = \frac{i\omega}{e} \varphi_{\ell_1,\ell_2} z_{\ell_1,\ell_2}. \quad (33)$$

Equation (33) represents an array of $L \times L$ equations for the bond currents, where $L$ is the number of bonds in the system. We shall not attempt to present the formal general solution for this array. Instead, we will obtain the limiting behaviors for the frequency dependence of $M$, and exemplify them by considering the smallest possible cluster of sites, a triangle, which possesses a magnetic moment. (For a two-site cluster the orbital magnetization vanishes because $[\vec{r}_1 \times \vec{r}_2] = 0$ by construction.)

Inspection of Eqs. (33) reveals the appearance of the characteristic frequency $\omega_{0l}^j$, given by

$$\omega_{0l}^j = \frac{z_{j\ell}}{C_j(\ell)}. \quad (34)$$
When the external frequency $\omega$ is much higher than all $\omega_0^\ell$'s, the local chemical potentials are ineffective in determining the bond currents. In that case

$$I_{tj} \sim \frac{i\omega}{c} \varphi_{tj} z_{tj}. \quad (35)$$

On the other hand, when $\omega$ is less than all $\omega_0^\ell$'s, the local chemical potentials dominate the bond currents. In that situation [cf. Eq. (31)] the current in the $j\ell$ bond "feels" the fluxes on all other bonds, and the solution of Eqs. (33) is much more complex. For example, for a three-site cluster we find

$$I_{12} = I_{23} = I_{31} = \frac{i\omega \varphi}{e} \left( \frac{1}{z_{12}} + \frac{1}{z_{23}} + \frac{1}{z_{31}} \right)^{-1}, \quad (36)$$

where $\varphi$ is the total flux enclosed by the triangle

$$\varphi = \varphi_{12} + \varphi_{23} + \varphi_{31}. \quad (37)$$

That is, $i\omega \varphi/c$ is the e.m.f. applied to the cluster. Recalling that for a triangle $\vec{r}_1 \times \vec{r}_2 = \vec{r}_2 \times \vec{r}_3 = \vec{r}_3 \times \vec{r}_1 = 2\vec{S}/3$, where $\vec{S}$ is the (vectorial) area of the three-site group, we find that the magnetic moment can be written in the form

$$M_{\text{triangle}} = \frac{i\omega}{c^2} H S^2 z_{j\ell}, \quad (38)$$

where $z_{j\ell}$ stands for the largest admittance of the three in the high frequency case, and for the smallest one at very low frequencies. Note, however, that for a triangle, the numerical coefficient in the high frequency limit includes a geometrical factor ($\varphi_{12}/3\varphi$) which diminishes the ratio $M/\omega$.

Let us now focus on the frequency dependence of the admittance itself. We consider the cases where $\omega \ll |\epsilon_{j\ell}|$ and $\omega \gg |\epsilon_{j\ell}|$. One can then ignore the resonant contribution to $\Re z_{j\ell}$. For $\omega \ll |\epsilon_{j\ell}|$ we have

$$z_{j\ell} = G_{j\ell}(1 + \frac{\omega}{\omega_1^\ell}), \quad (39)$$

with $\omega_1^\ell$ given by

$$\omega_1^\ell \sim \frac{\nu^2 D \beta_{j\ell}}{n_j - n_\ell} e^{-\frac{\beta}{2}(|\epsilon_j| + |\epsilon_\ell| + |\epsilon_{j\ell}|)} \sim e^{-\beta |\epsilon_{j\ell}|}. \quad (40)$$

In the other extreme limit, where $\omega \gg |\epsilon_{j\ell}|$, the imaginary part of the admittance becomes inversely proportional to $\omega$. When the electron-phonon coupling is weak, $z_{j\ell}$ in this regime is dominated by the imaginary part, leading to

$$z_{j\ell} \sim iZ_{j\ell}/\omega, \quad Z_{j\ell} = e^2 |J_{j\ell}|^2 (n_\ell - n_j)/\epsilon_{j\ell}. \quad (41)$$

Collecting the above results, we obtain the following pattern for the behavior of $M_{\text{triangle}}$ as function of the temperature and the external frequency. Starting at very low frequencies, such that $\omega < |\epsilon_{j\ell}|$ and $\omega < \omega_0^\ell$, the admittance is mainly given by the bond dc conductance, so that

$$M_{\text{triangle}} \sim \frac{i\omega}{c^2} HS^2 G_{j\ell}. \quad (42)$$

The magnetic moment decays exponentially as $T \to 0$. As the frequency increases such that $\omega > \omega_0^\ell$, then $z_{j\ell} \sim iG_{j\ell}\omega/\omega_0^\ell$ and

$$M_{\text{triangle}} \sim \frac{\omega^2}{c^2} HS^2 G_{j\ell}/\omega_0^\ell. \quad (43)$$

The leading frequency dependence is modified as compared to the very low $\omega$ regime, but the moment is still exponentially decaying with the decrease of the temperature [see Eqs. (24) and (40)]. Finally, in the high frequency limit such that $\omega \gg |\epsilon_{j\ell}|$, the moment becomes almost independent of the temperature and the frequency

$$M_{\text{triangle}} \sim Z_{j\ell} HS^2 /c^2. \quad (44)$$

For frequencies in the intermediate regime the behavior is rather complicate, and depends upon the detailed relationships between the frequency, temperature, $|\epsilon_{j\ell}|$, and the characteristic frequencies $\omega_0^\ell$ and $\omega_1^\ell$.

**V. CONCLUDING REMARKS**

The orbital magnetization of localized electrons in response to the temporal variation of a vector potential has been studied. We have obtained an expression for the magnetic moment in terms of the tunneling and phonon-assisted hopping currents between localized states. These currents have been derived from the Kubo formula.

The mere existence of an orbital magnetic moment in the strongly localized system requires a group of at least three states. We have found that the magnetization of this small cluster is proportional to the relevant dominant admittance between sites (the smallest one for small frequencies, $\omega \ll \omega_0^\ell$, and the largest one in the opposite case). Consequently, $M_{\text{triangle}}$ is proportional to $|J|^2$, where $J$ is the overlap integral of two wave functions. The *equilibrium* magnetic moment of a triangle, on the other hand, is of order $J^3$. This is because $M_{\text{eq}}$ results from quantum interference processes among the three sites.

The magnetization of a bulk system involves summation over all bonds [see Eq. (3)]. Its full calculation requires the knowledge of the distribution function of the site energies and the localized site locations. This may lead to a temperature dependence different from the exponential one characterizing the small cluster. Consider for example the case of nearest-neighbor hopping at low enough frequencies. Then the typical energies involved are within a region of width $\sim \beta^{-1}$ around the Fermi level. Consequently, for a smooth distribution of the site energies, within an energy band of width $W$, the
the sum is proportional to $n \propto (\beta W)^{-3}$. The typical spatial separation between sites is $\propto \xi L_\beta$, with $\xi$ being the localization length and $L_\beta \propto \ln |\beta J_0|$. ($J_0$ is the bare overlap integral between two localized levels.) It follows that the area of a typical triangle is of order $(\xi L_\beta)^2$. Collecting these estimates and using Eq. (32) we obtain that the bulk magnetic moment per unit volume is

$$M_{\text{bulk}} \propto \frac{i\omega(\beta c)}{e} H v^2 D(\beta^{-1}) \beta (\frac{n_i}{\beta W})^3 (\xi L_\beta)^3 \xi^2.$$  \hspace{1cm} (45)

Here $n_i$ is the concentration of localized levels. The bulk moment is proportional to the frequency and depends on the temperature $T$ as $T^2 D(T) \ln^3 (J_0/T)$. Modifications, due e.g. to the temperature dependence of the relevant area in the case of variable-range hopping, are possible. Finally we remark that in this work electronic correlations were ignored. These have to be taken into account in the calculation of the bond currents. We hope to pursue this issue in a future article.

---

**ACKNOWLEDGMENTS**

We thank Y. Imry and I. Goldhirsh for helpful discussions. Support by the German-Israel Fund (GIF), and the Fund for Basic Research administered by the Israel Academy of Sciences and Humanities is acknowledged. We gratefully acknowledge partial support from the joint Israel-Norway Program for Cultural Exchange.

---

**APPENDIX A: QUANTUM INTERFERENCE EFFECTS IN THE CURRENT**

As is mentioned above, quantum interference between the direct amplitude $j \to \ell$ and the indirect one, via a third site $k$, appears first in order $J^3$. It is therefore necessary to derive the corrections to the current to third order in the overlaps. We denote this quantity by $I_{jl}^{(3)}$. Returning to Eq. (33) we find

$$I_{jl}^{(3)}(t) = -e \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \left\langle \left[ \mathcal{H}_1(t_2), \mathcal{H}_{\text{perc}}(t_1) \right] 2 \Re J_{lj} Q_{lj}(t)c_{lj}^\dagger(t)c_j(t) \right\rangle$$

$$- \left\langle \mathcal{H}_{\text{perc}}(t_1), \left[ \mathcal{H}_1(t_2), 2 \Re J_{lj} Q_{lj}(t)c_{lj}^\dagger(t)c_j(t) \right] \right\rangle$$

$$- \phi_{lj}(t) \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \left\langle 2 \Re J_{lj} Q_{lj}(t)c_{lj}^\dagger(t)c_j(t) + 2 \Re J_{lj} Q_{lj}(t)c_{lj}^\dagger(t)c_j(t) \right\rangle$$

$$+ \left\langle \mathcal{H}_1(t_1) 2 \Re J_{lj} Q_{lj}(t)c_{lj}^\dagger(t)c_j(t) \right\rangle \right\rangle. \hspace{1cm} (A1)$$

The calculation of the terms appearing in Eq. (A1) is straightforward, though rather cumbersome. It yields for the ac amplitude of $I_{jl}^{(3)}$, $I_{jl}^{(3)}$, the expression

$$I_{jl}^{(3)}(\omega) = 2e \sum_k \int_{0}^{\infty} d\tau_1 \int_{0}^{\infty} d\tau_2 \left\{ F_{k,\ell,k}(\tau_1, \tau_2) \left[ \phi_{lj} + \bar{\phi}_{lj} e^{-i\omega \tau_1} + \bar{\phi}_{jk} e^{-i\omega \tau_2} \right] n_k n_j n_\ell - F_{k,\ell,k}(\tau_1, \tau_2) \right\}$$

$$+ \left\{ \phi_{lj} + \bar{\phi}_{lj} e^{-i\omega \tau_1} + \bar{\phi}_{k\ell} e^{-i\omega \tau_2} \right\} n_k n_j n_\ell - F_{k,\ell,k}(\tau_1, \tau_2) \left[ \phi_{lj} + \bar{\phi}_{lj} e^{-i\omega \tau_1} + \bar{\phi}_{k\ell} e^{-i\omega \tau_2} \right] n_k n_j n_\ell$$

$$+ F_{k,\ell,k}(\tau_1, \tau_2) \left[ \phi_{lj} + \bar{\phi}_{lj} e^{-i\omega \tau_1} + \bar{\phi}_{k\ell} e^{-i\omega \tau_2} \right] n_k n_j n_\ell + F_{k,\ell,k}(\tau_1, \tau_2) \left[ \phi_{lj} + \bar{\phi}_{lj} e^{-i\omega \tau_1} + \bar{\phi}_{k\ell} e^{-i\omega \tau_2} \right] n_k n_j n_\ell \right\}. \hspace{1cm} (A2)$$

where $n_j = 1 - n_j$, and

$$F_{k,\ell,k}(\tau_1, \tau_2) = \Re \left\{ J_{kj} J_{jk} e^{i \xi_{ij} \tau_1 + i \xi_{kj} \tau_2 + g(\tau_1 + \tau_2) + g(\tau_1) + g(\tau_2) - 3g(0)} \right\}. \hspace{1cm} (A3)$$

Contrary to the behavior of $I_{jl}^{(2)}(\omega)$, $I_{jl}^{(3)}(\omega = 0)$ is finite. Setting $\omega = 0$ in Eq. (A2) one finds that each $k-$term in the sum is proportional to

$$\tilde{\phi}_{lj} = \phi_{lj} + \bar{\phi}_{lj} + \bar{\phi}_{k\ell}, \hspace{1cm} (A4)$$
that is, to the total flux enclosed in the triangle $\ell jk$. The zero frequency part of the current is just the persistent current flowing in response to a constant magnetic flux. Its properties in the strong localization regime have been studied in great detail elsewhere.\cite{Buttiker85}

Denoting

$$\bar{I}_{j\ell}^{(3)}(\omega = 0) = I_{pc},$$

we derive from (A2) $\bar{I}_{j\ell}^{(3)}(\omega) - I_{pc}$ for weak electron-phonon interaction, and in the small $\omega$ limit. The result is

$$\bar{I}_{j\ell}^{(3)}(\omega) - I_{pc} = i\omega e \sum_k J_{jk}I_{k\ell}J_{\ell j} \\{ \bar{\phi}_{k\ell} \left[ \frac{1}{\epsilon_{k\ell}} + \frac{1}{\epsilon_{j\ell}} \right] \beta n_k \bar{n}_{\ell j} \gamma_{k\ell} + \left( \frac{1}{\epsilon_{jk}} + \frac{1}{\epsilon_{k\ell}} \right) \beta n_{\ell j} \bar{n}_{jk} \gamma_{jk} \right] + \bar{\phi}_{jk} \left[ \frac{1}{\epsilon_{jk}} \beta n_j \bar{n}_{\ell j} \gamma_{jk} \right] + \frac{1}{\epsilon_{jk}} \beta n_{\ell j} \bar{n}_{jk} \gamma_{jk} \right\}.$$

A remarkable observation is that $\bar{I}_{j\ell}^{(3)}(\omega)$, which is part of the current in the $j\ell$ bond, is driven by the magnetic phases on the bonds $k\ell$ and $jk$, but not by the phase on the bond $j\ell$ itself. This is a manifestation of interference effects. The interference affects other electronic properties of the system. One example is the conductivity. The formalism presented here can be used to find that quantity. In that case the electric field $\vec{E}$ is related to the fluxes by

$$i\omega \bar{\phi}_{j\ell} \rightarrow e\vec{E} \cdot \vec{R}_{j\ell}.$$  

Then the ring current $I_{pc}$ vanish by virtue of (A4). Using Eq. (A7) one finds that Eqs. (20) and (A6) give the conductivity of the $j\ell$-bond. In particular, Eqs. (20) and (A4) yield the well-known result for the bond conductivity to second order in the overlaps, while Eq. (A6) gives the nonlocal corrections to it, which result from interference.\cite{Efros85}

Namely, the current in the $j\ell$-bond is affected by the voltage drops on the $k\ell$ and the $jk$ bonds.

---

1. L. Pauling, J. Chem. Phys. 4, 673 (1936); F. London, J. Phys. Radium, 8, 397 (1937).
2. M. Büttiker, Y. Imry, and R. Landauer, Phys. Lett. A96, 365 (1985); H. F. Cheung, Y. Gefen, E. K. Riedel, and W. H. Shih, Phys. Rev. B37, 6050 (1988); H. Bouchiat and G. Montambaux, J. Phys. (Paris) 50, 2696 (1989).
3. O. Entin-Wohlman, Y. Imry, A. G. Aronov, and Y. Levinson, Phys. Rev. B51, 11584 (1995).
4. T. Holstein, Phys. Rev. 124, 1329 (1961).
5. O. Entin-Wohlman, A. G. Aronov, Y. Levinson, and Y. Imry, Phys. Rev. Lett. 75, 4094 (1995).
6. W. Kohn, Phys. Rev. 133, A171 (1964).
7. R. Landauer and M. Büttiker, Phys. Rev. Lett. 54, 2049 (1985).
8. L. P. Levy, G. Dolan, J. Dunsmuir, and H. Bouchiat, Phys. Rev. Lett. 64, 2074 (1990); V. Chandrasekhar, R. A. Webb, U. J. Brady, M. B. Keta, W. J. Gallager, and A. Klein, Phys. Rev. Lett. 67, 3578 (1991); D. Mailly, C. Chapelier, and A. Benoit, Phys. Rev. Lett. 70, 2020 (1993).
9. Triveldi and D. A. Browne, Phys. Rev. B38, 9581 (1988).
10. L. Swann, E. N. Bogachek, Yu. M. Galperin, M. Jonson, and R. I. Shekhter, Phys. Rev. Lett. 73, 162 (1994).
11. W. K. H. Panovsky and M. Phillips, Classical Electricity and Magnetism, y Addison-Wesley, Reading, Massachusetts, 1962.
12. T. Holstein, Annals od Phys. (NY) 8, 343 (1959).
13. V. Ambegaokar, B. I. Halperin, and J. S. Langer, Phys. Rev. B4, 2612 (1971).
14. S. V. Maleev, Zh. Eksp. Teor. Fiz. 79, 1995 (1980); 84, 260 (1983) [Sov. Phys. JETP 52, 1008 (1980); 57, 149 (1983)].
15. A. L. Efros and B. I. Shklovskii, in: Electron-Electron Interactions in Disordered Systems, edited by A. L. Efros and M. Pollak (Elsevier Science Publishers B.V., 1985), p. 409.