Two important features of mesoscopic Aharonov-Bohm (A-B) electronic interferometers are analyzed: decoherence due to coupling with other degrees of freedom and the coupled transport of charge and heat. We first review the principles of decoherence of electronic interference. We then analyze the thermoelectric transport in a ring threaded by such a flux, with a molecular bridge on one of its arms. The charge carriers may also interact inelastically with the molecular vibrations. This nano-system is connected to three terminals; two of them are electric and thermal, held at slightly different chemical potentials and temperatures, and the third is purely thermal. For example, a phonon bath thermalizing the molecular vibrations. When this third terminal is held at a temperature different from those of the electronic reservoirs, both an electrical and a heat current are, in general, generated between the latter. Likewise, a voltage and/or temperature difference between the electronic terminals leads to thermal current between the thermal and electronic terminals. The transport coefficients governing these conversions (due to energy exchange between the electrons and the vibrations) and their dependences on the A-B flux are analyzed. Finally, the decoherence due to these inelastic events is discussed.

1. Introduction

Interference, resulting from the superposition of different amplitudes, is a basic attribute of Quantum Mechanics. Akira Tonomura Sensei has made decisive lasting contributions to the study of these effects for electron beams.\textsuperscript{1} Here we shall review the analogous Physics in mesoscopic solid-state systems.\textsuperscript{2} Here, small sizes and/or low temperatures are necessary in order to keep electrons coherent. Clearly, the understanding of the processes of electronic decoherence is crucial here. We shall consider an Aharonov-Bohm (A-B) interference experiment on a ring-type structure. This proves to be a convenient way to observe interference patterns in such samples,
providing an experimentally straightforward way of shifting the interference pattern. This experiment involves two electron wave packets, $\ell(x)$ and $r(x)$ ($\ell, r$ stand for left, right), crossing the ring along its two opposite sides and interacting with an environment, represented by a (usually thermal) bath. We assume that the two wave-packets follow classical paths, $x_\ell(t), x_r(t)$ along the arms of the ring. The interference is examined after each of the two wave packets had traversed half of the ring’s circumference. The combined wave function is then

$$\psi = \ell(x) + r(x).$$

The mixed terms, $2\text{Re}[\ell(x) \times r^*(x)]$, constitute the interference contribution, clearly sensitive to a phase shift (e.g. AB) introduced between the two partial waves $\ell(x)$ and $r(x)$. Interaction with the environment can reduce the strength of the interference. This process is called “decoherence” and it is of great relevance in this paper.

We will review the effects of interference, first on the purely electronic transport. Then, on coupled electric and thermal transport. When changes in the environment state, caused by the electrons, occur, decoherence may follow. These changes will be modeled by the effect of a local vibrational mode (“vibronic coupling”), which is in turn coupled to a reservoir that is restoring equilibrium to the local vibration.

Thermoelectric effects in bulk conductors usually necessitate breaking of particle-hole symmetry, which can be substantial and controllable in mesoscopic structures. As a result there is currently much interest in investigations of thermoelectric phenomena in nanoscale devices at low temperatures.

Very recently, the paramount importance of another symmetry breaking has been pointed out. It has been proposed that the thermal efficiency of a thermoelectric heat engine, or refrigerator, can be significantly enhanced once time-reversal symmetry is broken (by, e.g., a magnetic flux). The Onsager symmetry of the thermopower coefficients is accordingly modified.

The coupling of the charge carriers to vibrational modes of the molecule should play a significant role in thermoelectric transport through molecular bridges. The thermopower coefficient was proposed as a tool to monitor the excitation spectrum of a molecule forming the junction between two leads, and to determine both the location of Fermi level of the charge carriers and their charge.

Theoretically, when the coupling to the vibrational modes is ignored, the transport coefficients are determined by the usual energy-dependent transmission coefficient, replacing the conductivity. Even when the corrections to the thermoelectric transport due to the coupling to the vibrational modes are small, their study is of interest because of fundamental questions related to the symmetries of the conventional transport coefficients, and since they give rise to additional coefficients connecting the heat transport in-between the electrons and the vibrational modes. Recently, we have analyzed these phenomena in a molecular bridge. In particular
we have considered the case where the molecule is (relatively) strongly coupled to a heat bath of its own, which maintains its temperature different from those of the source and sink of the charge carriers. Namely, we have assumed that the relaxation time due to the coupling of the molecule to its own heat bath, $\tau_V$, is short on the scale of the coupling of the molecular vibrations to the charge carriers. This coupling, $\gamma$ is our small parameter. $\hbar/\tau_V$ may still be very small on all other physical scales, such as $\hbar\omega_0$, where $\omega_0$ is the frequency of the vibration, or the molecular (electronic) level width. The phonon bath may be realized by an electronically insulating substrate or a piece of such material touching the junction, each held at a temperature $T_V$. Experimental realizations of three-terminal setups have been discussed.\textsuperscript{12–14}

![Fig. 1. A mixed, thermal-electronic, three-terminal system, as explained in the text, threaded by a magnetic flux $\Phi$.]

We organize this paper as follows. Section 2 discusses the principles of decoherence. Our theoretical model is presented in section 3 and the A-B oscillations of the transmission of the interferometer and their decoherence are discussed in section 4. After a short review of thermoelectricity in section 5, we give the main results of the analysis including the magnetic-flux dependence of the three-terminal transport coefficients Section 6 presents these coefficients of the three-terminal junction in the linear-response regime and verifies their Onsager symmetries. Section 7 is devoted to a discussion of the results in simple examples. Section 8 summarizes the article.

2. Principles of decoherence

Interference effects are strongly affected by the coupling of the interfering particle to its environment, e.g., to a heat bath. The way such a coupling modifies quantum phenomena has been studied for a long time, both theoretically,\textsuperscript{15,16} and experimentally.\textsuperscript{17} Some of the effect of the coupling to the environment may be described by the “phase-breaking” time, $\tau_\phi$, which is the characteristic time for the interfering particle to stay phase coherent as explained below.

Our discussion will be based on the work of Stern \textit{et al.}\textsuperscript{18} Two points of view
have been used to describe how the interaction of a quantum system with its environment might suppress quantum interference. The first regards the environment as measuring the path of the interfering particle. When the environment has all the information on that path, no interference is seen. The second description considers the phase uncertainty induced on the interfering particle by the interaction with the environment. The two descriptions were proven to be equivalent. Here we will review the first point of view mentioned above, using the example of an A-B interference experiment on a ring. The electron (whose coordinate is $x$) and the environment (whose wave functions for the two paths are denoted by $\chi$ and $\eta$) interact during the traversal of the ring. As a result, the combined wave function of Eq. 1 becomes

$$\psi = [\ell(x) + r(x)] \otimes \chi_0(\eta)$$

(2)

At time $\tau_0$ (measured with respect to the start of the experiment), when the interference is examined, the wave function is, in general,

$$\psi(\tau_0) = l(x, \tau_0) \otimes \chi_\ell(\eta, \tau_0) + r(x, \tau_0) \otimes \chi_r(\eta, \tau_0)$$

(3)

and the interference term is (see below),

$$2 \text{Re} \left[ \ell^*(x, \tau_0) r(x, \tau_0) \int d\eta \chi_\ell^*(\eta, \tau_0) \chi_r(\eta, \tau_0) \right].$$

(4)

Without the coupling to the environment, the interference term would have been just Eq. 1. So, the effect of the interaction is to multiply the interference term by the scalar product $\int d\eta \chi_\ell^*(\eta, \tau_0) \chi_r(\eta, \tau_0)$. The first way to understand the dephasing is clear from this expression, i.e. the reduction in the interference due to the response of the environment to the interfering waves. When the two states of the environment become orthogonal, the final state of the environment identifies the path the electron took. Quantum interference, which is the result of an uncertainty in this path, is then lost. The phase breaking time, $\tau_\phi$, is the time at which the two interfering partial waves shift the environment into states to become orthogonal to each other, i.e., when the environment has the full information on the electron path.

As seen from the above discussion, the phase uncertainty remains constant when the interfering wave does not interact with the environment. Thus, if a trace is left by a partial wave on its environment, this trace cannot be wiped out after the interaction is over. This statement can be proven also from the point of view of the change the interfering wave induces in its environment. This proof follows simply from unitarity. The scalar product of two states evolving under the same Hamiltonian does not change in time. The only way to change it is by another interaction of the electron with the same environment (see the discussion at the end of this section). The interference will be retrieved only if the orthogonality is transferred from the environment wave function to the electronic wave functions which are not traced on in the experiment.
The above discussion was concerned with the phase $\phi = \phi_r$, accumulated by the right-hand path only. The left-hand path accumulates similarly a phase $\phi_l$ from the interaction with the environment. The interference pattern is governed by the relative phase $\phi_r - \phi_l$, and it is the uncertainty in that phase which determines the loss of quantum interference. This uncertainty is always smaller than, or equal to, the sum of uncertainties in the two partial waves' phases.

Often the same environment interacts with the two interfering waves. A typical example is the interaction of an interfering electron with the electromagnetic fluctuations in vacuum. In this case, if the two waves follow parallel paths with equal velocities, their dipole radiation, despite the energy it transfers to the field, does not dephase the interference. This radiation makes each of the partial waves’ phases uncertain, but does not alter the relative phase. Another well-known example is that of “coherent inelastic neutron scattering” in crystals. This process follows from the coherent addition of the amplitudes for the processes in which the neutron exchanges the same phonon with all scatterers in the crystal.

The last example also demonstrates that an exchange of energy is not a sufficient condition for dephasing. It is also not a necessary condition for dephasing. What is important is that the two partial waves flip the environment to orthogonal states. It does not matter in principle that these states are degenerate. Simple examples were given by Stern et al. Thus, it must be emphasized that, for example, long-wave excitations (phonons, photons) cannot dephase the interference. But that is not because of their low energy but rather because they do not influence the relative phase of the paths.

We emphasize that dephasing may occur by coupling to a discrete or a continuous environment. In the former case the interfering particle is more likely to “reabsorb” the excitation and “reset” the phase. In the latter case, the excitation may move away to infinity and the loss of phase can then be broadly regarded as irreversible. The latter case is that of an effective “bath”.

We point out that in special cases it is possible, even in the continuum case, to have a finite probability to reabsorb the created excitation and thus retain coherence. This happens, for example, in a quantum-interference model due to Holstein for the Hall effect in insulators (see also Refs. and ). This model deals with phonon-induced hopping between two localized states with different energies, $E_1$ and $E_2$, which necessitates the (real) absorption of a phonon with energy $E_2 - E_1$ for $E_2 - E_1 > 0$. It focuses on transitions which occur via an intermediate localized state with yet another energy, $E_3$. We take, for definiteness $E_3 > E_1$. That intermediate state transition also involves the virtual absorption of a phonon with frequency $\omega_q$, which is then re-emitted with the transition from 3 to 2. This higher-order process has a perturbation theory energy-denominator which at “resonance” ($\hbar \omega_q = E_3 - E_1$)
produces a term \(i\delta(\hbar\omega_q - E_3 + E_1)\). The integration over the continuous \(\omega_q\) then yields a term with a \(\pi/2\) phase shift in the transition amplitude between 1 and 2.

References \(^{20}\) and \(^{22}\) analyzed the effects of a superposition (or interference) of the above amplitude with the direct one between 1 and 2, possibly including a phase shift \(\phi = 2\pi\Phi/\Phi_0\), (where \(\Phi_0 = \hbar c/e\) is the flux quantum) between the direct and the compound transition paths, due e.g. to a magnetic flux \(\Phi\) enclosed between them.

Note that these two interfering paths involve for \(E_2 - E_1 > 0\) the absorption of the same phonon and they therefore stay coherent (see discussion above). Interestingly, it turns out that although this process creates a \(\sin(\phi)\) term in some transition probabilities, it retains the basic Onsager symmetry in \(\Phi\) of the conductivity,

\[
\sigma_{xx}(\Phi) = \sigma_{xx}(-\Phi).
\]

and also the “non diagonal” Hall conductivity, \(\sigma_{xy}\),

\[
\sigma_{xy}(\Phi) = \sigma_{yx}(-\Phi).
\]

Since a \(\sin(\phi)\) dependence is allowed here by symmetry, there is no reason why it should not exist. Such a dependence is generated, in fact, by the Holstein process.\(^{20,22}\)

This is the basic reason why the Holstein process furnishes an explanation for the Hall effect in (localized) insulators.

The adaptation of the Holstein idea to transitions between two leads with continuous energy spectra is actually simpler than the original model, in which an additional phonon is needed to conserve energy. Here an elastic, purely electronic, transition can be made from a full state on one side to an empty state on the other. This transition can again be accomplished via a superposition of the direct transition with a compound one through the molecule, involving again a “resonant” virtual phonon. Clearly, here this effect is of order \(\gamma^2\) (see below, \(\gamma\) is the electron-phonon coupling). The calculations to that order were done in Ref. \(^{23}\) Again, there is no \(\sin(\phi)\) term in either the electrical or the thermal conductivity. It exists, however, in other “non diagonal” transport coefficients, such as the thermopowers,\(^{23}\) as will be discussed in section 7.

3. The model

In our analysis, the molecular bridge is represented by a single localized electronic level, standing for the lowest available orbital of the molecule; when an electron resides on the level, it interacts (linearly) with an Einstein oscillator. Our analysis does not include electronic interactions, but focuses on the electron-vibron ones. Thus, the Hamiltonian of the molecular bridge, which includes the coupling with
the vibrations, reads
\[ H_M = \epsilon_0 c_0^{\dagger} c_0 + \omega_0(b^{\dagger} b + \frac{1}{2}) + \gamma (b + b^{\dagger}) c_0^{\dagger} c_0, \tag{7} \]
where \( \epsilon_0 \) is the energy of the localized level, \( \omega_0 \) is the frequency of the harmonic oscillator representing the vibrations, and \( \gamma \) is its coupling to the electrons. The Hamiltonian describing the tunneling between the molecule and the leads is
\[ H_{\text{coup}} = \sum_k (V_k c_k^{\dagger} c_0 + H_c) + \sum_p (V_p c_p^{\dagger} c_0 + H_c) \tag{8} \]
[using \( k(p) \) for the left (right) lead]. The leads’ Hamiltonian is
\[ H_{\text{lead}} = H_L + H_R + H_{LR}. \tag{9} \]
Here
\[ H_{L(R)} = \sum_{k(p)} \epsilon_{k(p)} c_{k(p)}^{\dagger} c_{k(p)} \tag{10} \]
is the Hamiltonian of each of the leads and
\[ H_{LR} = \sum_{kp} V_{kp} e^{i\phi} c_{k}^{\dagger} c_{p} + H_c \tag{11} \]
describes the direct coupling between the two leads (pictorially shown as the lower arm of the ring in Fig. ??). The A-B magnetic flux is included in the phase factor \( \exp[i\phi] \), see Eq. (??). Since we use units in which \( \hbar = 1 \), the flux quantum is \( 2\pi c/e \).

Thus, our model Hamiltonian is
\[ H = H_{\text{lead}} + H_M + H_{\text{coup}}, \tag{12} \]
where the operators \( c_0^{\dagger}, c_k^{\dagger}, \) and \( c_p^{\dagger} (c_0, c_k, \) and \( c_p) \) create (annihilate) an electron on the level, on the left lead, and on the right lead, respectively, while \( b^{\dagger} (b) \) creates (annihilates) an excitation of frequency \( \omega_0 \) on the molecule.

In the spirit of the Landauer approach,\(^2\) the various reservoirs (which are assumed in the simplest case to be large enough to stay in equilibrium in spite of the small currents that flow from/to them) and which supply charge and energy to the leads, are described by equilibrium distributions with given chemical potentials and temperatures. This keeps the populations on each lead of the excitations pertaining to a specific reservoir to be in equilibrium as well. The electronic reservoirs on the left and on the right of the bridge are characterized by the electronic distributions \( f_L \) and \( f_R \),
\[ f_{L(R)}(\omega) = \left( 1 + \exp[\beta_{L(R)}(\omega - \mu_{L(R)})] \right)^{-1}, \tag{13} \]
determined by the respective Fermi functions, with $\beta_{L(R)} = 1/k_B T_{L(R)}$. The phonon reservoir, which determines the vibration population on the bridge, is characterized by the Bose-Einstein distribution,

$$N = \left(\exp[\beta_V \omega_0] - 1\right)^{-1},$$

with $\beta_V = 1/k_B T_V$.

4. A-B oscillations in the transmission of the interferometer and their decoherence

We start by considering the interferometer having just a resonance level without vibrational coupling on one arm and a direct transmission on the other. The interacting with the vibrations is later included.

4.1. A simple interferometer: no vibronic coupling

The transport through the “direct” arm of the interferometer (the one not carrying the dot) is characterized by the dimensionless coupling

$$\lambda(\omega) = (2\pi)^2 \sum_{k,p} |V_{kp}|^2 \delta(\omega - \epsilon_k) \delta(\omega - \epsilon_p),$$

such that the bare transmission (reflection) amplitude through that branch is $t_o$ ($r_o$),

$$t_o^2(\omega) = \frac{4\lambda(\omega)}{1 + \lambda(\omega)^2}, \quad r_o^2 = 1 - t_o^2.$$}

The transport through the arm carrying the quantum dot is characterized by

$$\Gamma_{L(R)}(\omega) = \pi \sum_{k(p)} |V_{k(p)}|^2 \delta(\omega - \epsilon_{k(p)}).$$

Below we denote the total width of the level $\epsilon_0$ of the quantum dot by $\Gamma(\omega)$, with

$$\Gamma(\omega) = \frac{\Gamma_L(\omega) + \Gamma_R(\omega)}{1 + \lambda(\omega)},$$

and the asymmetry of the coupling of the dot to the leads by $\alpha(\omega)$,

$$\alpha^2(\omega) = \frac{4\Gamma_L(\omega) \Gamma_R(\omega)}{[\Gamma_L(\omega) + \Gamma_R(\omega)]^2}.$$
In Ref. 23 it was found that the current, $I$, and the (electronic) energy current, $I_E$, across the interferometer are determined by the same transmission function,

$$I = -e \int \frac{d\omega}{2\pi} [f_R(\omega) - f_L(\omega)] T(\omega) ,$$

$$I_E(\omega) = \int \frac{\omega d\omega}{2\pi} [f_R(\omega) - f_L(\omega)] T(\omega) ,$$

(20)

where the Fermi functions in the leads, $f_{L,R}$ are given in Eq. (13). The function $T(\omega)$ is

$$T(\omega) = t_0^2 \left( 1 - \Gamma \text{Im} G^a + \frac{\Gamma^2}{4} [1 - \alpha^2 \cos^2 \Phi] |G^a|^2 \right) + t_o r_o \Gamma \alpha \cos \Phi \text{Re} G^a + \frac{\Gamma^2 \alpha^2}{4} |G^a|^2 .$$

(21)

Here we have omitted the explicit $\omega-$dependence of (some of) the functions for brevity. The first term in this expression, $t_0^2$, yields the conductance in the absence of the ring arm carrying the bridge (see Fig. ??), while the last term yields the conductance of that arm alone. The other three terms in Eq. (21) result mainly from various interference processes (see below).

The advanced Green function of the dot is

$$G^a(\omega) = \frac{1}{\omega - \bar{\epsilon}(\omega) - i\Gamma(\omega)/2} ,$$

(22)

where $\bar{\epsilon}(\omega)$ is the resonance energy shifted by the coupling to the other parts of the interferometer,

$$\bar{\epsilon}(\omega) = \epsilon_0 + \frac{1}{1 + \lambda(\omega)} [\lambda(\omega) \Gamma_L(\omega) \Gamma_R(\omega)]^{1/2} \cos \phi .$$

(23)

This result describes well many interferometer experiments. It satisfies the Onsager-related symmetry of the conductance, which follows from the property that $T(\omega)$ is even in the flux $\Phi$.

We consider for simplicity the symmetric case, $\alpha^2(\omega) = 1$, which will be assumed in the rest of this section; all $V$’s are taken to be real (time-reversal symmetry at zero flux). We now turn to the explanation of the three terms in Eq. (21) which are not direct transmissions through one of the arms. The second term in the brackets is just a correction to the direct transmission $t_0$ due to attempting to go through the bridge (and instead being reflected) either before or after the direct transition. The third term is the “weak localization” correction to the transmission, due to the increase of the total reflection via the two time-reversed paths which encircle the whole ring clockwise and anti-clockwise. The term before the last, having the product of $t_0$ and (real part of) the transmission amplitude through the bridge, $\Gamma \text{Re} G^a$, resembles the mixed term in the good old two-wave interference. There is however a very
important small difference: the latter should have the real part of $\Gamma G^a e^{i\phi}$, while our expression has $\cos \phi \text{Re} G^a$. It thus has a pure $\cos \phi$ dependence on the flux as it should. This is so, because we consider a closed interferometer which conserves particles. This differs from the ordinary two-wave interference, in which particles are lost and therefore, a $\sin(\phi)$ dependence is also generated in the transmission (“no phase rigidity”, see Ref. 26). Technically, this is due to the fact that here the Green’s function of the dot, which is determined by paths starting at and returning to it, is dressed by paths (or “diagrams”) in which the electron experiences the whole interferometer. Physically, the point is that the fact that electrons can enter and exit the interferometer from/to the two leads, does not break particle conservation for the scattering matrix of the interferometer. Therefore, the Onsager symmetry, for which particle conservation, or “unitarity”, is essential, holds.

4.2. Interferometer with vibronic coupling

The technical calculations of the transport coefficients with the electron-vibron coupling $\gamma$ have been done in Ref. 23. They are rather complicated, and so far it has only been feasible to do them to order $\gamma^2$, which is sufficient to see the effects of vibronic excitation/deexcitation. The full results to this order are given in that reference and there is no point to repeat them here. Suffice it to say that, for the electrical conductance, indeed the occurrence of a vibronic change of state does eliminate, in the term resulting from the interaction with vibrations, the $\cos \phi$ term in the conductance, as expected due to decoherence. It was however found that a $\cos^2 \phi$ (or $\cos 2\phi$) dependence survives. This dependence resembles the weak-localization-type flux dependence, as in the third term of Eq. (21) discussed above. This in fact results from a novel generalization of the usual weak-localization process, in which the two time-reversed paths change the vibrational state in the same fashion. Thus, there is no decoherence of this special process! The situation with the thermal and thermoelectric coefficients is much more complicated and interesting. It will be reviewed in the rest of this paper.

5. Generalities on thermoelectric transport

The electronic part of thermoelectric linear-transport problem is fully characterized for the two-terminal situation by

$$\begin{pmatrix} I_e \\ I_Q \end{pmatrix} = \begin{pmatrix} G & L_1 \\ L_1 & K^0_e \end{pmatrix} \begin{pmatrix} \delta \mu/e \\ \delta T/T \end{pmatrix}, \quad (24)$$

where $I_e$ is the charge current and $I_Q$ the heat current, $\delta T = T_l - T_r$ and $\delta \mu/e \equiv V$ is the voltage between the left and right terminals. The $2 \times 2$ matrix contains the
regular conductance $G$, the bare electronic thermal conductance, $K_0$ and the (off-diagonal) thermoelectric coefficients $L_1$. That the latter two are equal is the celebrated Onsager relation (valid for time-reversal symmetric systems; with a magnetic field, $B$, if one of them is taken at $B$, it is equal to the other one at $-B$). We remind the reader that since the (Seebeck) thermopower is the voltage developed due to a temperature difference at zero electrical current, $S = L_1/G$. All currents and transport coefficients in Eq. (24) are given in 1D for noninteracting electrons in terms of integrals involving the energy-dependent elastic transmission, $T(E)$ and the Fermi function, $f(E)$, at the common chemical potential $\mu$ and temperature $T$, $G = I_0; L_1 = I_1; K_0 = I_2$, where

$$I_n = \frac{2e}{h} \int_{-\infty}^{\infty} dE T(E) f(E)[1 - f(E)] (E - \mu)^n / (k_B T).$$

We recall that since the heat conductance is defined as the heat current due to a unit temperature difference, at vanishing electrical current, the electronic heat conductance is given by

$$K_e = K_0^e - GS^2 = I_2 - I_1^2 / I_0.$$

In a thermoelectric energy conversion device, the performance is governed by the well-known dimensionless “figure of merit”, $zT$,

$$zT = \frac{L^2}{KG} = \frac{GS^2}{K},$$

where $K = K_e + K_{ph}$ is the full thermal conductance. Here $K_{ph}$ is the phonon (+ any other neutral mobile excitation of the solid) thermal conductance. The efficiency is a fraction, $g(zT)$, of the, maximal allowed, Carnot efficiency. Large $zT$’s yield better performance, with $g(zT)$ rising monotonically from 0 for $zT \to 0$ to 1 for $zT \to \infty$.

An important remark here is that the determinant of the transport matrix in Eq. (24) is nonnegative. This implies that the figure of merit based on the bare conductance $K_0^e = K_0^e + K_{ph}$, $zT$ would be limited by unity. It is the “renormalization” of $K$ as in Eq. (26), which allows larger $zT$ values and opens the way to thermoelectric applications!

Based on the above, Mahan and Sofo,\textsuperscript{10} in an important development, suggested that regarding $T(E)f(E)[1 - f(E)]/G$ as a (positive, normalized) weight function, then $S = < E - \mu >$ and $K_e = < (E - \mu)^2 > - < E - \mu >^2$. One can then make $K_e$ very small by having a very narrow transmission band away from the Fermi level. This is needed in order to have a finite $< E - \mu >$, not relying only on the, usually small, asymmetry of $T(E)$ near $E_F$, to break electron-hole symmetry. This implies that $zT$ would be limited only by $K_{ph}$. Tricks for reducing $K_{ph}$ by acoustic and geometrical mismatch, have been suggested.\textsuperscript{28}
As an example we take an impenetrable barrier of height $W >> k_B T$, whose transmission we take as $\mathcal{T}(E) \simeq \Theta(E - W)$. We measure all energies from the common chemical potential $\mu$. Under these conditions, $f(E)$ to the Boltzmann distribution, leading to $f(E)[1 - f(E)] \simeq \exp[-E/(k_B T)]$. Hence,

$$<E> = e L_1 / G = W + k_B T,$$

$$<E^2> - <E>^2 = (k_B T)^2,$$

(28)

with the charge and heat electron current given by Eqs. (20). To reiterate, the formulae above say that $S$ is the average energy transferred by an electron, divided by $e$, while $K_0^2 / G$ is the average of the square of that energy, divided by $e^2$. Therefore $K_e / G$ is proportional to the variance of that energy. The latter obviously vanishes for a very narrow transmission band. In the high barrier case that band is a few $k_B T$ above $W$. Thus, it is not surprising that $K_e$ is of the order of $(k_B T/W)^2$.

6. Interference effects and their decoherence in thermoelectric transport

The linear-transport full set of thermoelectric coefficients\(^{23}\) is given by

$$\begin{bmatrix} I \\ I_Q \\ -\dot{E}_V \end{bmatrix} = \mathcal{M} \begin{bmatrix} \Delta \mu / e \\ \Delta T / T \\ \Delta T / T \end{bmatrix},$$

(29)

where the matrix of the transport coefficients, $\mathcal{M}$, is

$$\mathcal{M} = \begin{bmatrix} G(\Phi) & K(\Phi) & X^V(\Phi) \\ K(-\Phi) & K_2(\Phi) & \widetilde{X}^V(\Phi) \\ X^V(-\Phi) & \widetilde{X}^V(-\Phi) & C^V(\Phi) \end{bmatrix}.$$ 

(30)

This matrix satisfies the Onsager-Casimir relations. All three diagonal entries are even functions of the flux $\Phi$. The off-diagonal entries of $\mathcal{M}$ consist each of a term even in the flux, and another one, odd in it, obeying altogether $\mathcal{M}_{ij}(\Phi) = \mathcal{M}_{ji}(-\Phi)$.

The flux dependence of the transport coefficients is very interesting. We have found (see Ref. 23 for details) that there are three types of flux dependencies hiding in those six coefficients. First, there is the one caused by interference. The interference processes modify the self energy of the Green functions pertaining to the A-B ring, in particular the broadening of the electronic resonance level due to the coupling with the leads. The interference leads to terms involving $\cos \phi$. Secondly, there is the flux dependence which appears in the form of $\cos 2\phi$ (or alternatively, $\sin^2 \phi$). This reflects the contributions of time-reversed paths. These two dependencies are even in the flux. They yield the full flux dependence of the diagonal entries of the matrix $\mathcal{M}$, Eq. (30), and the even (in the flux) parts of the off-diagonal elements. Finally,
there is the odd dependence in the flux, that appears as \( \sin \phi \). This dependence characterizes the odd parts of the off-diagonal entries of \( \mathcal{M} \). These terms necessitate the coupling of the electrons to the vibrational modes. They arise from Holstein-type processes, briefly discussed toward the end of section ??.

7. Examples and discussion

As is clear from the results the previous section and from Refs. 11 and 23, the interesting effect induced exclusively by the coupling of the electrons to the vibrational modes is the possibility to create an electric current, or an electronic heat current, by applying a temperature difference \( \Delta T_V \) on the phonon bath thermalizing this mode. These new thermoelectric phenomena are specified by the two coefficients \( X^V \) and \( \tilde{X}^V \), see Eq. (30), respectively. All other transport coefficients related to the electronic currents are mainly due to the transport of the electrons between the electronic terminals, with slight modifications from the (small) coupling to the vibrations. We therefore confine the main discussion in this section to the coefficients \( X^V \) and \( \tilde{X}^V \). To make a closer connection with possible experiments, we introduce the (dimensionless) coefficients

\[
S^V = e \beta \frac{X^V}{G},
\]

and

\[
\tilde{S}^V = \frac{\tilde{X}^V}{K_2}.
\]

The first gives the potential drop across the molecular bridge created by \( \Delta T_V \) when the temperature drop there, \( \Delta T \), vanishes, and the second yields the temperature difference created by \( \Delta T_V \) when \( \Delta \mu = 0 \) [or the inverse processes, see Eqs. (29) and (30)]. For both the conductance, \( G \), and the thermal conductance, \( K_2 \), we use below their leading terms, resulting from the coupling to the leads alone.

As mentioned above, the transport coefficients of our three-terminal junction obey the Onsager-Casimir relations. They do it though in a somewhat unique way: the “off-diagonal” elements are related to one another by the reversal of the magnetic field. However, they are not a purely odd functions of it. A special situation arises when the molecule is connected symmetrically to the two leads. In that case, the asymmetry parameter \( \alpha = 1 \) [see Eq. (19)]. Then, the two coefficients, \( X^V \) and \( \tilde{X}^V \), are odd functions of the flux.

\[
X^V(\Phi) = e \omega_0 \sin \phi \int \frac{d\omega}{\pi} \pi^V(\omega, \Phi) \left( t_0(\omega_-) - t_0(\omega_+) \right),
\]
and
\[
\tilde{X}^V(\Phi) = \omega_0 \sin \phi \int \frac{d\omega}{\pi} T^V(\omega, \Phi) \\
\times \left( t_o(\omega_-)(\omega_- - \mu) - t_o(\omega_+)(\omega_+ - \mu) \right).
\]

(34)

Here, \( \omega_\pm = \omega \pm \omega_0/2 \), and all other quantities are given in in Eqs. (30), (43), and (54) of Ref. 23. In other words, the thermoelectric processes described by \( X^V(\Phi) \) and \( \tilde{X}^V(\Phi) \) require a certain symmetry-breaking. In the absence of the magnetic field, that is supplied by the spatial asymmetry of the junction; in the presence of a flux, those processes appear also for a junction symmetrically coupled to the leads, provided that the couplings to the leads depend on the energy.

When the two leads connected to the electronic reservoirs are identical (making the molecular bridge symmetric) the transmission amplitude of the direct bond between the two leads, \( t_o \) [see Eq. (16)], is an even function of \( \omega \). The transmission function \( T^V(\omega, \Phi) \) given in Ref. 23 is not entirely even or odd in \( \omega \), and therefore \textit{a priori} the integrals which give \( X^V(\Phi) \) and \( \tilde{X}^V(\Phi) \) do not vanish. However, the asymmetry in the \( \omega \)-dependence of the integrand (which results from the \( \omega \)-dependence of the Green function) is not significant. As a result, \( S^V \) is extremely small, while \( \tilde{S}^V \) is not (because of the extra \( \omega \) factor in the integrand), see Fig. ???. These plots are computed using \( \Gamma(\omega) = \Gamma^0 \sqrt{1 - (\omega/W)^2} \), and \( \lambda(\omega) = \lambda^0 \{1 - (\omega/W)^2\} \), where \( W \) is half the bandwidth, and all energies are measured in units of \( (1/\beta = k_B T) \) (we have set \( \Gamma^0 = \lambda^0 = 1 \) and \( W = 50 \)).

![Fig. 2](color online.) The transport coefficients \( S^V \) and \( \tilde{S}^V \) as functions of the flux (measured in units of the flux quantum) and \( \beta \omega_0 \), for a symmetric bridge.

The relative magnitudes of \( S^V \) and \( \tilde{S}^V \) are significantly changed when the molecule is coupled \textit{asymmetrically} to the leads. Let us assume that the left reservoir is represented by an electron band, such that the partial width it causes to the
resonant level is given by

$$\Gamma_L(\omega) = \Gamma_0^L \sqrt{\frac{\omega - \omega_c}{\omega_v - \omega_c}} ,$$

(35)

while the right reservoir is modeled by a hole band, with

$$\Gamma_R(\omega) = \Gamma_0^R \sqrt{\frac{\omega_v - \omega}{\omega_v - \omega_c}} .$$

(36)

The corresponding quantity pertaining to the lower arm of the ring in Fig. ?? is

$$\lambda(\omega) = \lambda^0 \sqrt{\frac{\omega_v - \omega}{\omega_v - \omega_c} \frac{\omega - \omega_c}{\omega_v - \omega_c}} .$$

(37)

Here, $\omega_c$ is the bottom of the conductance band (on the left side of the junction), while $\omega_v$ is the top of the hole band (on the right one). The energy integration determining the various transport coefficients is therefore limited to the region $\omega_c \leq \omega \leq \omega_v$. (For convenience, we normalize the $\Gamma$’s by the full band width, $\omega_v - \omega_c$.) Note that the density of states is increasing (decreasing) with energy in the electron (hole) lead. Exemplifying results in such a case are shown in Fig. ??, computed with $\Gamma_0^L = \Gamma_0^R = \lambda^0 = 1$ [see Eqs. (35), (36), and (37)], and $\omega_c = -\omega_v = 100$, all in units of $k_B T$.

![Fig. 3. (color online.) Same as Fig. 4, for an asymmetric bridge.](image)

Remarkably enough, the coefficient $\tilde{S}^V$, which measures the ability of the device to turn a temperature difference on the phonon bath thermalizing the molecular vibrations into a temperature difference across the molecule is not very sensitive to the details of the model, apparently because the term proportional to $\sin \phi$ in Eq. (66) of Ref. 23 is the dominant one. This means that by reversing the direction of the magnetic field, one reverses the sign of the temperature difference or alternatively,
the sign of the electronic heat current. On the other hand, the coefficient $S^V$, which sets the scale of the capability to turn a temperature difference across that phonon bath into an electric current, is far more sensitive to the details of the model (as expressed e.g. in our choice for the density of states on the leads) and is less affected by the magnetic field.

8. Conclusions

We reviewed the interference effects of mesoscopic-scale solid-state A-B interferometers, with special emphasis on how they are reduced/eliminated by decoherence. Coupling to local vibrations, to lowest significant order, as well as to a heat bath, was introduced. The latter makes the problem much richer. We first discussed just the electronic transport, with and without the vibronic coupling. Then, we reviewed the electric and thermal transport in an interferometer with the above-mentioned couplings. In particular, we have found that the thermoelectric transport coefficients through a vibrating molecular junction, placed on an Aharonov-Bohm interferometer, have an interesting dependence on the magnetic flux. The coefficients which relate the temperature difference between the phonon and electron reservoirs to the charge and heat currents carried by the electrons, which exist only due to the electron-vibron coupling, can be enhanced by the magnetic flux. One of them is also very sensitive to the spatial asymmetry.

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