Berry-Phase-Induced Heat Pumping and Its Impact on the Fluctuation Theorem

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Applying adiabatic, cyclic two-parameter modulations we investigate quantum heat transfer across an anharmonic molecular junction contacted with two heat baths. We demonstrate that the pumped heat typically exhibits a Berry-phase effect in providing an additional geometric contribution to heat flux. Remarkably, a robust fractional quantized geometric phonon response is identified as well. The presence of this geometric phase contribution in turn causes a breakdown of the fluctuation theorem of the Gallavotti-Cohen type for quantum heat transfer. This can be restored only if (i) the geometric phase contribution vanishes and if (ii) the cyclic protocol preserves the detailed balance symmetry.

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Understanding and controlling of heat transfer due to phonons occurring in low dimensional nanoscale systems is both of prime and practical importance [1]. Pioneering experimental works carried out recently, such as nanotube thermal rectifier [2], nanotube phonon waveguide [3] has spawn *phononics*, i.e., the science and engineering of phonons [1], as an emerging new scientific discipline where heat flow can be manipulated as flexibly as electronic current. Although the nonlinear (anharmonic) interaction has been demonstrated as a crucial component [4,5] in various functional thermal devices, the heat control has heretofore typically been achieved by applying a temperature bias, for which in accordance with the second law of thermodynamics—heat flows from "hot" to "cold" spontaneously.

Recent studies show that spontaneous, rare fluctuations of anomalous heat transfer may occur [6], thus being seemingly in apparent violation with the second law. Clearly, however, no violation of the second law occurs on average. The typical measure of such violations is the (small) probability for such anomalous events as they emerge from a heat exchange fluctuation theorem (FT) [6–9]. The FT for (nonequilibrium) entropy production [10,11] and heat flux [7,8] describes that the distribution, $P_\tau(Q)$, of the heat Q transferred from the left (L) bath at temperature T_L to the right (R) bath at T_R over a long time interval τ, obeys the relation: $\lim_{\tau\to\infty}\tau^{-1}\ln[P_\tau(Q)/P_\tau(-Q)] = Q(\beta_R - \beta_L)/\tau$, where $\beta_{L,R} = 1/k_B T_{L,R}$. This FT thus shows explicitly that heat can transfer spontaneously from cold to hot with finite, although typically with very small probability. In particular, Ref. [8] demonstrates this FT in the quantum case for heat transfer across a quantum harmonic chain coupled with thermal reservoirs. A particular challenge that arises is then whether this quantum Gallavotti-Cohen type FT remains valid also in the nonlinear quantum regime beyond the quantum harmonic chain limit, and, more generally, whether such a heat-flux FT still can be formulated in presence of cyclic time-dependent manipulations of certain control parameters.

In the context of time-dependent manipulations various molecular heat pumps have been proposed to efficiently control heat flux against thermal gradients at the nanoscale. In all those cases the system is driven far away from equilibrium by use of an external modulation imposed on system parameters. For example, a molecular model with modulated energy levels, has been found to operate as a heat pump [12]. Likewise, a spin system leading to the heat pumping has been studied with Ref. [13]. Other schemes investigated pumping of heat in electronic nanoscale devices by applying time-periodic laser fields [14]. Moreover, Brownian heat motors fueled by oscillating temperatures have recently been devised as well [15,16]. Given such time-dependent manipulations one may therefore scrutinize whether the physics of a nonvanishing geometric phase does impact the transfer of heat under external

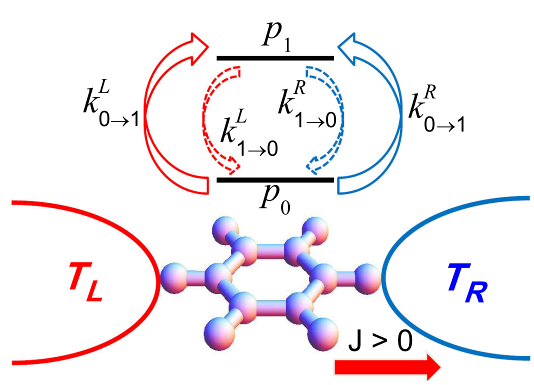

FIG. 1 (color online). A schematic representation of the anharmonic molecular junction. Quantum heat transfer is generated via a dynamics of excitation and relaxation of the local single mode. The heat flux J from the center to the right bath is defined as positive.

modulations. If so, what is its impact on the existence of a heat-flux FT?

In this Letter, we shall answer these above mentioned objectives by studying quantum heat transport across an anharmonic molecular junction model. We start with a system consisting of a molecular junction coupled to two thermal baths [12], as illustrated in Fig. 1. The total Hamiltonian $H_{\rm tot}$ is composed of the following contributions: $H_{\rm tot} = H_S + H_B^L + H_B^R + V_{SB}^L + V_{SB}^R$: system Hamiltonian $H_S = \sum_{n=0}^{N-1} E_n|n\rangle\langle n|$, with $E_n = n\hbar\omega_0$, where we assume that heat transport is dominated by a single mode and thus consider a two-level system ($N = 2$) to simulate the strong nonlinearity [17]. If $N \to \infty$, the system reduces to the quantum harmonic case. The two thermal baths are represented by sets of independent harmonic modes, i.e., $H_B^\nu = \sum_k \hbar\omega_k b_{k,\nu}^\dagger b_{k,\nu}$, with $\nu = L, R$, where $b_{k,\nu}^\dagger$, $b_{k,\nu}$ are the bosonic creation and annihilation operators associated with the phonon mode k of bath ν. The system-bath interactions is taken to be bilinear, i.e., $V_{SB}^\nu = B_\nu \sum_{n=1}^{N-1} \sqrt{n}\,|n\rangle\langle n-1| + \text{c.c.}$, $B_\nu = \sum_k \gamma_{k,\nu}(b_{k,\nu}^\dagger + b_{k,\nu})$, where the system-bath interaction is characterized by the phonon spectral function $\Gamma_\nu(\omega) = 2\pi\sum_k \gamma_{k,\nu}^2 \delta(\omega - \omega_k)$. In the following, we use wide-band limit $\Gamma_\nu(\omega) = \Gamma_\nu$. As shown with Ref. [17], in the limit of fast dephasing and using the Redfield approximation for weak system-bath coupling, the underlying dynamics can be modeled as follows:

$$\dot{p}_1(t) = -p_1(t)(k_{1\to0}^L + k_{1\to0}^R) + p_0(t)(k_{0\to1}^L + k_{0\to1}^R). \quad (1)$$

Here, $p_n (n = 0, 1)$ denotes the probability of the molecule to occupy the state $|n\rangle$, satisfying $p_0(t) + p_1(t) = 1$. The activation and relaxation rates read

$$k_{0\to1}^\nu = \Gamma_\nu N_\nu(\omega_0), \qquad k_{1\to0}^\nu = \Gamma_\nu[N_\nu(\omega_0) + 1], \quad (2)$$

where $N_\nu(\omega_0) = [e^{\beta_\nu\hbar\omega_0} - 1]^{-1}$ is the Bose-Einstein occupation probability. Finally, the steady-state heat flux at the right contact (being equal to the heat flux at the left contact) is expressed as

$$J = \hbar\omega_0[p_1^s k_{1\to0}^R - p_0^s k_{0\to1}^R], \quad (3)$$

where the superscript s means the steady state. The first term denotes the energy flux going from the molecule into the bath R while the second term provides the opposite heat flux from the bath R back into the system.

Geometric Berry-phase-induced heat pumping.—For heat pump operation, the molecular junction connected to the two reservoirs is subjected to cyclic parameter modulations. This could be realized by imposing a modulation on either of the following parameters: $\omega_0(t)$, $\Gamma_L(t)$, $\Gamma_R(t)$, $T_L(t)$, $T_R(t)$. Throughout the following, the modulations acting on such system parameters are assumed to be slow, i.e., we employ adiabatic modulations. Let the period of modulation be $\mathcal{T}_p = 2\pi/\Omega$. The typical frequency for a carbon-carbon bond is 1.4×10^{14} s^{-1} [18]. Γ_ν is around 10^{15} s^{-1}, according to the measurement with alkane molecular junction [19]. The relaxation time for fast thermalization usually is on the order of a few fs or ps. Thus, the modulation time scale must obey $2\pi/\Omega \gg 1$ ps. In this way, the assumption of adiabatic modulation is valid whenever the driving frequency $\Omega \ll 1$ THz.

Of prime interest is the heat flux from the molecule into the bath R during the long time span τ. This is achieved upon introducing the characteristic function for the phonon counting field χ, i.e., [20,21]

$$\mathcal{Z}_\tau(\chi) = \sum_{q=-\infty}^{\infty} P_\tau(q)e^{iq\chi} = \mathbf{1}^\dagger \hat{T}[e^{-\int_0^\tau \mathcal{H}(\chi,t)dt}]\mathbf{p}(0), \quad (4)$$

$$\mathcal{H}(\chi, t) \doteq \begin{bmatrix} k_{0\to1}^L + k_{0\to1}^R & -k_{1\to0}^L - k_{1\to0}^R e^{i\chi} \\ -k_{0\to1}^L - k_{0\to1}^R e^{-i\chi} & k_{1\to0}^L + k_{1\to0}^R \end{bmatrix}, \quad (5)$$

where $P_\tau(q)$ is the probability distribution of having heat $Q = q\hbar\omega_0$ transferred from the molecule into the bath R during time $\tau \to \infty$. Here, $1^\dagger = [1, 1]$, $\hat{T}$ denotes the time-ordering operator, and $\mathbf{p}(0) = [p_0(0), p_1(0)]^T$ are the initial occupation probabilities. Then, the cumulant generating function is obtained as $\mathcal{G}(\chi) \equiv \lim_{\tau\to\infty}\tau^{-1}\ln\mathcal{Z}_\tau(\chi)$, which generates the heat current via the relation $J = \hbar\omega_0\partial\mathcal{G}(\chi)/\partial(i\chi)|_{\chi=0}$. Denote by $\lambda_0(\chi, t)$ the instantaneous eigenvalue of $\mathcal{H}(\chi, t)$ with the smallest real part and $|\psi_0(\chi, t)\rangle$ ($\langle\varphi_0(\chi, t)|$) the corresponding normalized right (left) eigenvector. The cumulant generating function takes on the following form, being composed of two parts [21,22], namely,

$$\mathcal{Z}_\tau(\chi) \sim e^{\tau\mathcal{G}} = e^{\tau(\mathcal{G}_{\rm dyn} + \mathcal{G}_{\rm geom})}, \quad (6)$$

$$\mathcal{G}_{\rm dyn} = -\mathcal{T}_p^{-1}\int_0^{\mathcal{T}_p} dt\lambda_0(\chi, t), \quad (7)$$

$$\mathcal{G}_{\rm geom} = -\mathcal{T}_p^{-1}\int_0^{\mathcal{T}_p} dt\langle\varphi_0|\partial_t|\psi_0\rangle. \quad (8)$$

The first contribution $\mathcal{G}_{\rm dyn}$ presents the temporal average and defines the dynamic heat transfer. This is the only term which survives in the static limit. The second, geometric part $\mathcal{G}_{\rm geom}$ presents an additional contribution caused by the adiabatic cyclic evolution. As we shall see it is this part which possesses a nontrivial geometric interpretation. Let us rewrite $\mathcal{G}_{\rm geom}$ as a line integral over the closed contour $\mathcal{R}$ in the parameter space $\mathbf{u}$:

$$\mathcal{G}_{\rm geom} = -\mathcal{T}_p^{-1}\oint_{\mathcal{R}} d\mathbf{u}\cdot\mathcal{A}_{\mathbf{u}}, \quad (9)$$

with $\mathcal{A}_{\mathbf{u}}(\chi) = \langle\varphi_0(\mathbf{u})|\partial_{\mathbf{u}}|\psi_0(\mathbf{u})\rangle$. Thus, this is an analog of a Berry phase [23], which does not contain time t explicitly and only depends on the geometry of the modulation contour in the parameter space $\mathbf{u}$. In the case of two parameters being modulated, say u_1, u_2, using Stokes theorem, we find

$$\mathcal{G}_{\text{geom}} = -\mathcal{T}_p^{-1} \iint_{\mathcal{S}_\mathcal{R}} du_1 du_2 \mathcal{F}_{u_1 u_2}, \tag{10}$$

where $\mathcal{S}_\mathcal{R}$ is the integral area enclosed by the contour $\mathcal{R}$.

$$\mathcal{F}_{u_1 u_2} = \langle \partial_{u_1} \varphi_0 | \partial_{u_2} \psi_0 \rangle - \langle \partial_{u_2} \varphi_0 | \partial_{u_1} \psi_0 \rangle \tag{11}$$

is an analog of the gauge invariant Berry curvature [23].

Let us next specify the case that the bath temperatures $T_L(t)$, $T_R(t)$ are subjected to adiabatic modulations. Then Eq. (11) yields the Berry curvature in temperature space, reading

$$\mathcal{F}_{T_L T_R}(\chi) = -C_L C_R \frac{2i \sin(\chi) \Gamma_L \Gamma_R (\Gamma_L + \Gamma_R)}{(\sqrt{K^2 + 4D})^3}, \tag{12}$$

where $C_\nu = k_B \beta_\nu^2 \hbar\omega_0 e^{\beta_\nu \hbar\omega_0} N_\nu^2$, $K = \Gamma_L(1 + 2N_L) + \Gamma_R(1 + 2N_R)$, $D = \Gamma_L \Gamma_R N_L N_R (e^{\beta_R \hbar\omega_0} e_{+\chi} + e^{\beta_L \hbar\omega_0} e_{-\chi})$ with $e_{\pm\chi} \equiv e^{\pm i\chi} - 1$. Upon substituting this Berry curvature into Eq. (10), the total heat flux emerges as

$$J_{\text{tot}} = \hbar\omega_0 \frac{\partial[\mathcal{G}_{\text{dyn}}(\chi) + \mathcal{G}_{\text{geom}}(\chi)]}{\partial(i\chi)} \bigg|_{\chi=0} = J_{\text{dyn}} + J_{\text{geom}},$$

$$J_{\text{dyn}} = \frac{\hbar\omega_0}{\mathcal{T}_p} \int_0^{\mathcal{T}_p} dt \frac{\Gamma_L \Gamma_R (N_L - N_R)}{K}, \tag{13}$$

$$J_{\text{geom}} = \frac{\hbar\omega_0}{\mathcal{T}_p} \iint_{\mathcal{S}_{\mathcal{R}_1}} dT_L dT_R \frac{-\partial \mathcal{F}_{T_L T_R}(\chi)}{\partial(i\chi)} \bigg|_{\chi=0}, \tag{14}$$

where

$$-\frac{\partial \mathcal{F}_{T_L T_R}(\chi)}{\partial(i\chi)} \bigg|_{\chi=0} = \frac{2 C_L C_R \Gamma_L \Gamma_R (\Gamma_L + \Gamma_R)}{K^3}. \tag{15}$$

The dynamic part J_{dyn} just coincides with the temporal average of the heat flux obtained from $J \equiv J(t)$ in Eq. (3). The geometric part J_{geom} is the additional heat flux that results from the nontrivial Berry-phase effect. The ratio of this geometric heat flux and the dynamic one is typically about Ω/Γ_ν. To avoid that J_{geom} is masked by J_{dyn}, we choose a symmetric molecular junction with $\Gamma_L = \Gamma_R$, and modulate $T_L(t)$, $T_R(t)$ as indicated by the circle contour in Fig. 2(a). Then one finds that $J_{\text{dyn}} \equiv 0$ and $J_{\text{geom}} \neq 0$, see Fig. 2(b), so that the Berry-phase-induced J_{geom} dominates the heat transport. This is the case for which the geometric phase effect on heat transport is distinctly experimentally detectable. As a main finding we have that the Berry-phase effect acts as a heat pump, providing an additional heat flux across the molecular junction even though on average no thermal bias acts and the system is symmetric. Note also, distinct from the irreversible heat flux J_{dyn}, J_{geom} is time reversible, i.e., under the time-reversed modulation ($t \to -t$) the Berry-phase induced heat flux just reverses sign.

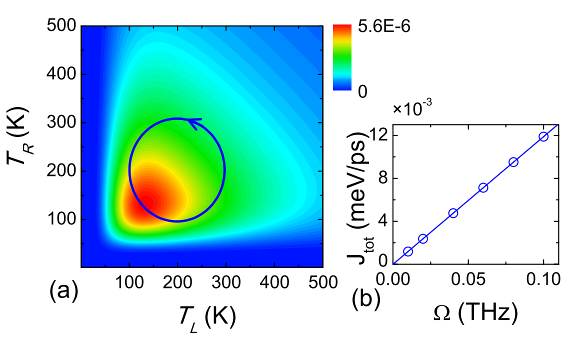

FIG. 2 (color online). (a) The contour map of $-\partial \mathcal{F}_{T_L T_R}(\chi)/\partial(i\chi)|_{\chi=0}$, for $\Gamma_L = \Gamma_R$ and $\hbar\omega_0 = 25$ meV. The (blue) circle with an arrow denotes the path of two-parameter temperature modulations: $T_L(t) = 200 + 100\cos(\Omega t + \pi/4)$, $T_R(t) = 200 + 100\sin(\Omega t + \pi/4)$. The integral area $\mathcal{S}_\mathcal{R}$ is within the circle. (b) Pure Berry-phase induced heat current: $J_{\text{tot}} = J_{\text{geom}}$ ($J_{\text{dyn}} = 0$). The straight line is the analytical result from Eq. (14), while the open circles give the simulation results by integrating Eq. (1).

Fractional quantization of phonon response.—Remarkably, we find a fractional quantized phonon response for large temperature driving: the integral in Eq. (14) can be rewritten as $\int_0^\infty \int_0^\infty dN_L dN_R 2\Gamma_L \Gamma_R (\Gamma_L + \Gamma_R)/K^3 = 1/4$, yielding [22]

$$J_{\text{geom}} = \tfrac{1}{4}\hbar\omega_0/\mathcal{T}_p. \tag{16}$$

This 1/4 fractional quantized geometric phonon response is robust since it does not depend on the specific values of $\hbar\omega_0$, Γ_L, Γ_R. It means that the geometric phase effect caused by two bath temperature modulations is able to pump maximally on average one phonon $\hbar\omega_0$ per four cycles.

Impact on heat-flux fluctuation theorem.—Besides the dynamic $\mathcal{G}_{\text{dyn}}$, $\mathcal{G}_{\text{geom}}$ will generally not only contribute additionally to the average heat transfer but also impact the higher moments of the heat current (such as the phonon counting statistics) and other heat transport characteristics as well. In the following, we study its impact on the heat-flux fluctuation theorem. Before doing so, let us address first the static situation with $\mathcal{G}_{\text{geom}}(\chi) \equiv 0$, yielding

$$\mathcal{G}(\chi) = \mathcal{G}_{\text{dyn}}(\chi) = -\lambda_0(\chi) = \frac{-K + \sqrt{K^2 + 4D}}{2}. \tag{17}$$

Then, λ_0 obeys the Gallavotti-Cohen (GC) symmetry [24] [and alike for $\mathcal{G}_{\text{dyn}}(\chi)$, $\mathcal{G}(\chi)$ and $\mathcal{Z}_\tau(\chi)$], reading

$$\lambda_0(\chi) = \lambda_0(-\chi + i\beta^*), \tag{18}$$

where $\beta^* = \ln[(k^L_{0\to1} k^R_{1\to0})/(k^R_{0\to1} k^L_{1\to0})]$. In virtue of Eq. (2), yielding the detailed balance relation $k^\nu_{0\to1} = k^\nu_{1\to0} e^{-\beta_\nu \hbar\omega_0}$, we find that $\beta^* = \hbar\omega_0(\beta_R - \beta_L)$. Via an inverse Fourier transform of Eq. (4), this GC symmetry results in the *quantum* FT of heat transport for an *anharmonic* molecular junctions, reading with $Q = q\hbar\omega_0$:

$$\lim_{\tau\to\infty} \frac{1}{\tau} \ln\left[\frac{P_\tau(Q)}{P_\tau(-Q)}\right] = Q(\beta_R - \beta_L)/\tau, \tag{19}$$

which precisely coincides (without any correction) with the result for the quantum harmonic chain [8]. This FT gives the probability of observing spontaneous "second law violation": Assume $T_L < T_R$, i.e. $\beta^* < 0$; the upper bound to observe the violation for spontaneous heat transfer from (left) cool to (right) hot is estimated as $\int_c^\infty dq P_\tau(q) = \int_c^\infty dq P_\tau(-q) e^{q\beta^*} \leq e^{c\beta^*}$. It indicates that in absence of external modulations, the probability of at least c phonons (or net energy $c\hbar\omega_0$) transporting against the thermal bias is nonvanishing detectable, although decaying exponentially.

For the time-modulated system the GC symmetry ceases to hold when $\mathcal{G}_{\rm geom}(\chi) \neq 0$. For example, in the case of cyclic temperature modulations $T_L(t)$ and $T_R(t)$, the Berry curvature $\mathcal{F}_{T_L T_R}(\chi)$ contains the factor $\sin(\chi)$, which explicitly breaks the GC symmetry of $\mathcal{G}_{\rm geom}(\chi)$, and alike for $\mathcal{G}(\chi)$ and $\mathcal{Z}_\tau(\chi)$. Thus, the FT Eq. (19) becomes *violated* as a consequence of a geometric phase induced breakdown of GC symmetry. Moreover, even for parameter modulations yielding $\mathcal{G}_{\rm geom}(\chi) = 0$, and with time-dependent $\beta^* \to \beta^*(t)$, the GC symmetry for $\mathcal{G}_{\rm dyn}(\chi) = -\mathcal{T}_p^{-1} \int_0^{\mathcal{T}_p} dt \lambda_0(\chi, t)$ generally cannot be recovered, despite $\lambda_0(\chi, t) = \lambda_0(-\chi + i\beta^*(t), t)$.

Interestingly, we find that for time modulations of the system-bath couplings $\Gamma_L(t)$, $\Gamma_R(t)$ the detailed balance relation $k^\nu_{0\to1}/k^\nu_{1\to0} = e^{-\beta_\nu \hbar\omega_0}$ remains intact, thus providing a vanishing Berry curvature $\mathcal{F}_{\Gamma_L\Gamma_R}(\chi) \equiv 0$. Meanwhile, with the resulting time-independent $\beta^*(t) = \beta^*$, one finds that the GC symmetry of $\mathcal{G}_{\rm dyn}(\chi) = -\mathcal{T}_p^{-1} \int_0^{\mathcal{T}_p} dt \lambda_0(\chi, t)$ still holds. Consequently, we obtain a vanishing Berry-phase induced heat pumping and, surprisingly as well, also no violation of the FT, no matter how $\Gamma_L(t)$ and $\Gamma_R(t)$ are modulated.

In summary, through investigating heat transport across an anharmonic molecular junction by applying cyclic two-parameter modulations, we find that the system generally undergoes, apart from dynamic pumping, also a Berry-phase-induced heat pumping. This geometric contribution exhibits a robust fractional quantized phonon response. Furthermore, the quantum FT for heat transport in presence of a static temperature bias holds true in the anharmonic case as well. The presence of the geometric phase, however, violates the heat-flux FT. Only in situations of vanishing Berry curvature and restoration of detailed balance symmetry can the validity of the FT be recovered.

Although our present work did focus on the adiabatic regime, it likely can be extended to the case of a non-adiabatic geometric phase [25], and maybe also for non-cyclic modulation schemes in the spirit of [26]. Because the geometric phase has profound effects on material properties [23] we hope that our present findings do invigorate others to undertake related studies aimed at uncovering intriguing novel geometric phase induced thermal effects (such as thermoelectricity) which will enrich further the discipline of *phononics*.

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