Universal exact expression for adiabatic pumping in terms of non-equilibrium steady states

Naoko Nakagawa
College of Science, Ibaraki University, Mito, Ibaraki 310-8512, Japan
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We develop a unified viewpoint on pumping and nonequilibrium thermodynamics. We show that the pumping current generated through an adiabatic mechanical operation in equilibrium can be expressed in terms of the stationary distribution of the corresponding driven nonequilibrium system. We also show that the total transfer in pumping can be estimated by the work imported to the driven counterpart. These findings lead us to a unified viewpoint for pumping and nonequilibrium thermodynamics as well as serve as a useful guide in the design of efficient pumps.

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For centuries, heat pumping has been considered an important topic. The Carnot engine showed the direct relation between mechanical work and pumping of heat. Pumping induced by electric current known as the Peltier effect, was explained by the linear response theory as an example of the reciprocal relation. In molecular scales, the possibility of realizing heat pumps with thermal ratchets is related to geometric asymmetry for the Hamiltonian $H^\alpha$. For equilibrium processes $(\varepsilon = 0)$, we use $\langle f \rangle_{\rho_{eq}}$ and $\langle f \rangle_{\rho_{st}}$ instead of $\langle f \rangle_0$ and $\langle f \rangle_{\rho_{st}}$, respectively.

The time evolution of the system is governed by the deterministic dynamics according to the Hamiltonian $H^\alpha(\Gamma)$ and the stochastic Markovian dynamics owing to the external bath coupling. One operates the system mechanically by varying the parameters $\alpha$. The protocol for this operation is denoted as $\tilde{\alpha} := (\alpha(t))_{t \in [0, \tau]}$. We restrict ourselves to a quasi-static operation such that the system is always maintained in a steady state. When discussing the time evolution of $\Gamma$, we denote its value at time $t$ by $\Gamma(t)$ and its path in the whole time interval $[0, \tau]$ by $\hat{\Gamma} = (\Gamma(t))_{t \in [0, \tau]}$.

In order to theoretically analyze pumping problems, we also study a system driven by a certain driving field $\varepsilon$. We assume that the system reaches a unique nonequilibrium steady state (NESS) when we fix $\varepsilon$ and $\alpha$ for a sufficiently long time. The transition probability associated with the path $\hat{\Gamma}$ is denoted by $T_{\tilde{\alpha}, \varepsilon}[\hat{\Gamma}]$ in a protocol $\tilde{\alpha}$ under the driving $\varepsilon$. The probability distribution in the unique NESS is denoted by $\rho_{\tilde{\alpha}, \varepsilon}^\mathrm{eq}[\hat{\Gamma}]$, with which we define

$$\psi_{\tilde{\alpha}}^\varepsilon(\Gamma) := -\log \rho_{\tilde{\alpha}, \varepsilon}^\mathrm{eq}(\Gamma). \quad (1)$$

The canonical distribution $\rho_{eq}^\alpha(\Gamma)$ corresponds to $\rho_{\tilde{\alpha}, 0}^\mathrm{st}(\hat{\Gamma})$, and we use $\psi_{\tilde{\alpha}}$ instead of $\psi_0$.

For any function $f(\hat{\Gamma})$ of a path, we define its average in the protocol $\tilde{\alpha}$ as

$$\langle f \rangle_{\tilde{\alpha}} := \int \mathcal{D}\hat{\Gamma} \rho_{\tilde{\alpha}, \varepsilon}^\mathrm{st}(\Gamma(0)) T_{\tilde{\alpha}, \varepsilon}[\hat{\Gamma}] f(\hat{\Gamma}), \quad (2)$$

where $\mathcal{D}\hat{\Gamma}(\cdots)$ denotes the integral over all the possible paths $\hat{\Gamma}$. For any function $f(\Gamma)$ of a state, we define its average in the steady state as

$$\langle f \rangle_\rho := \int d\Gamma \rho_{\tilde{\alpha}, \varepsilon}^\mathrm{st}(\Gamma) f(\Gamma). \quad (3)$$

For equilibrium processes $(\varepsilon = 0)$, we use $\langle f \rangle_{\rho_{eq}}$ and $\langle f \rangle_{\rho_{st}}$ instead of $\langle f \rangle_0$ and $\langle f \rangle_{\rho_{st}}$, respectively.

We assume that the current at time $t$ is the function of $\Gamma$, $J(\Gamma; t)$, i.e. it depends only on the system’s path but not on the system’s environment. Because the probability of the path depends on the environment and the
applied protocol, the average $\langle J \rangle^\alpha$ in turn depends on them. The total transfer in the whole time interval is given by

$$\Xi(\hat{\Gamma}) = \int_0^T dt J(\hat{\Gamma}; t).$$

(4)

In the context of a pump, $\Xi(\hat{\Gamma})$ is the “total pumping” in a single execution of the protocol.

**Pumping current and its conjugate driving:** For a heat pump carrying energy from one place to the other, $J(\hat{\Gamma}; t)$ is the heat current between the two places and $\Xi(\hat{\Gamma})$ is the total transferred heat.

It is crucial for us to observe that the mean heat current can be produced not only by the mechanical operation for pumping but also by imposing a difference in the temperatures at the two places. In the latter case, the mean current flows spontaneously along the natural direction, satisfying the second law of thermodynamics.

The difference of the inverse temperatures is often called thermodynamic force corresponding to the heat current. In this Letter, we call it the “conjugate driving” corresponding to the heat current.

We refine the above situation as follows: In order to study the heat pumping in the system in contact with two separate isothermal heat baths indexed by $k$ ($k = 1, 2$), for which the inverse temperature is denoted by $\beta$, we also study its counterpart with the conjugate driving, i.e. the same system, for which the inverse temperatures $\beta_1, \beta_2$ of the baths are different. We choose $\beta_k$ so as to satisfy $\beta = (\beta_1 + \beta_2)/2$.

Letting $J_k(\hat{\Gamma}; t)$ be the heat current from the $k$-th heat bath to the system at time $t$ in the path $\hat{\Gamma}$, the heat current from one heat bath to the other is formulated as

$$J(\hat{\Gamma}; t) = \frac{J_1(\hat{\Gamma}; t) - J_2(\hat{\Gamma}; t)}{2},$$

(5)

for both the pumping system and its driven counterpart. By computing the average, we have $\langle J \rangle = \langle J_1 \rangle = -\langle J_2 \rangle$ under steady driving or any cyclic protocol. The conjugate driving, i.e. the thermodynamic force corresponding to the heat current is

$$\varepsilon = \beta_2 - \beta_1.$$  

(6)

The entropy production owing to the heat current is $\varepsilon J(\hat{\Gamma}; t) = (\beta_2 - \beta_1)J(\hat{\Gamma}; t)$.

For stochastic pumps represented using flashing ratchet models, we consider a particle in a potential with a periodic boundary condition in a certain coordinate $x$. When applying a cyclic operation to the potential, the system may have a non-vanishing circulation in its microstate, and this may be observed as directed mean current $\langle J \rangle^\alpha_{eq}$ of the particle, where

$$J(\hat{\Gamma}; t) = \dot{x}(t).$$

(7)

We notice that $J(\hat{\Gamma}; t)$ is determined by the system’s microstate and not by the operation.

The driven counterpart is the same system in which the particle is pulled by a constant non-conservative force $f$ along the coordinate $x$. The conjugate driving is

$$\varepsilon = \beta f,$$  

(8)

and the entropy production is $\beta f J(\hat{\Gamma}; t)$, where $J(\hat{\Gamma}; t)$ for the driven system is the same as Eq. (7).

Even though we present our claims for closed system setup in this Letter, they can also be extended to include open systems with particle baths by modifying the setup, as discussed in Sec. 5 of [15]. For such open systems, we can consider particle pumping between two particle baths, where the particle current is defined parallel to Eq. (5). Here, the conjugate driving corresponds to $\varepsilon = \beta(\mu_2 - \mu_1)$, where $\mu_k$ is the chemical potential for $k$-th particle bath.

**The expression for total pumping:** Our first result is the expression for the total pumping produced in equilibrium operations, the derivation for which is given later.

For the protocol $\hat{\alpha}$ applied to an equilibrium system, the total pumping is

$$\langle \Xi \rangle^\alpha_{eq} = \int \alpha \cdot \left( \nabla_{\alpha} \psi^{eq}_{\alpha} \partial_{\varepsilon} \psi^{eq}_{\alpha} \right)_{\varepsilon=0}^\alpha,$$

(9)

$$= \int \alpha \cdot \left( \nabla_{\alpha} \partial_{\varepsilon} \psi^{eq}_{\alpha} \right)_{\varepsilon=0}^\alpha,$$

(10)

where $\int_\alpha d\alpha \cdots$ is the line integral along the protocol $\alpha$ in the parameter space of $\alpha$. It is remarkable that the total pumping is directly related to the steady probability distribution $\rho^{st}_{\alpha,\varepsilon}(\Gamma)$ for the driving counterpart, $\rho^{eq}_{\alpha,\varepsilon}(\Gamma)$ depends on the type of the conjugate driving $\varepsilon$, as does the equilibrium pumping.

The expression (9) indicates that the pumping is efficient when $\nabla_{\alpha} \psi^{eq}_{\alpha}(\Gamma)$ is parallel to $\partial_{\varepsilon} \psi^{eq}_{\alpha}(\Gamma)$ in the phase space of $\Gamma$. In other words, it is efficient when the operation $\hat{\alpha}$ well mimics the nonequilibrium driving. It is worth noting that the kernels of (9) and (10) correspond to the off-diagonal components of the Fisher information matrix because $\nabla_{\alpha} \partial_{\varepsilon} \psi^{eq}_{\alpha} = \partial_{\varepsilon} \psi^{eq}_{\alpha} |_{\rho^{eq}}$ for $\varepsilon \to 0$.

In cyclic protocols $\hat{\alpha}_{cyc}$, we can apply the Stokes’ theorem to $\int_\alpha d\alpha \cdot \nabla_{\alpha} \psi^{eq}_{\alpha}(\Gamma) \nabla_{\alpha} \psi^{\alpha}_{\alpha}$ in the r.h.s. of (10). Therefore,

$$\langle \Xi \rangle^\alpha_{cyc} = \int_S dS \langle J_\alpha \rangle_{\rho^{eq}},$$

(11)

where $S$ is the region in the parameter space enclosed by the closed line of $\hat{\alpha}_{cyc}$. We call $\langle J_\alpha \rangle_{\rho^{eq}}$ the “pumping density”. When the number of the parameters is two, i.e., $\alpha = (\alpha_1, \alpha_2)$, the pumping density is

$$J_\alpha(\Gamma) = \partial_{\varepsilon} [\partial_{\alpha_1} \psi^{eq}_{\alpha_1}(\Gamma) \partial_{\alpha_2} \psi^{eq}_{\alpha_2}(\Gamma) - \partial_{\alpha_1} \psi^{eq}_{\alpha_1}(\Gamma) \partial_{\alpha_2} \psi^{eq}_{\alpha_2}(\Gamma)]_{\varepsilon=0}.$$

(12)
Various studies relating pumping to a geometric effect or the Berry phase report similar result to \cite{12}, which are derived from the Master equation or the cumulant generating function in cyclic operations in equilibrium. We emphasize that the key point of our formula \cite{12} is the use of the probability distribution $\rho^{st}_{\alpha, \varepsilon}$ for the driven counterpart.

**Equilibrium pumping and work in driven counterpart:** Our second result is the relation of equilibrium pumping to the mechanical work in NESS. We apply an adiabatic operation to both the equilibrium system and its driven counterpart. We arrive at an approximated equality between the quantities of these two distinct systems as,

$$\langle \Xi \rangle_{eq}^{\varepsilon = \lambda} = -\beta \langle W \rangle_{\varepsilon}^{\lambda} + O(\varepsilon).$$

(13)

Here, the work in the adiabatic operations in NESS is given by $\langle W \rangle_{\varepsilon}^{\lambda} = \int d\alpha \cdot \langle \nabla_{\alpha} H \rangle_{\rho^{eq}_{\alpha, \varepsilon}}$. Eq. (13) is a direct consequence of the thermodynamic relation for a process connecting two NESSs,

$$\langle W \rangle_{\varepsilon}^{\lambda} = \Delta F - \frac{\varepsilon}{\beta} \ln \langle \Xi \rangle_{eq}^{\lambda} + O(\varepsilon^2),$$

(14)

which we validate later. $\Delta F$ is the difference of the equilibrium free energy. The work relation (14) is consistent with the extended Clausius equality in \cite{16, 17}.

For general operations with a finite speed, the total pumping is related to work in the nonequilibrium counterpart as

$$\langle \Xi \rangle_{eq}^{\varepsilon = \lambda} = -\beta \langle W \rangle_{\varepsilon}^{\lambda} + O(\varepsilon),$$

(15)

where $\lambda$ is the reverse protocol of $\lambda$, i.e. $\lambda = (\lambda - t)_{t \in [0, t]}$. The relation (15) follows from an extended Jarzynski equality to NESS \cite{18}.

**Numerical demonstration of the pumping density:** We construct pumping density maps for a model system, using which we can design a desired pump. As we will show, the system can work as a heat pump as well as stochastic pump, where the efficiency of the protocol depends on the type of current one wants to pump.

As the model system, we take a one-dimensional Markov jump model of three states ($x = 1, 2$ and $3$) with a periodic boundary condition identifying $x = 3$ with $x = 0$. In order to design the rate constants for the jump, we assume virtual energy barriers at every midpoint of the neighboring two states. We set the energies of the three states as $v_1$, $v_2$ and $v_3$, and the energies of the barriers as $u_{12}$, $u_{23}$ and $u_{31}$, respectively. Then, we express the jump rates $R_{xy}$ from $x$ to $y$ as $R_{xy} = e^{-\beta(u_{xy} - v_x)}$. We assume the parameters for the operation as $\alpha = (v_2, u_{23})$.

First, we show the pumping density when the system works as a heat pump. For this purpose, we assume the system is in contact with two heat baths: The one (say $\beta_1$) is in the region $1 < x < 2.5$ and the other (say $\beta_2$) is in $2.5 < x < 4(= 1)$. The rate matrix for the conjugate driving $\varepsilon = \beta_2 - \beta_1$ is expressed as

$$R^c = \begin{pmatrix}
-\lambda_1^c & R_{12} e^{\varepsilon(u_{12} - v_1)} & R_{13} e^{\varepsilon(u_{13} - v_1)} \\
R_{21} e^{\varepsilon(u_{12} - v_2)} & -\lambda_2^c & R_{23} e^{\varepsilon(u_{23} - v_2)} \\
R_{31} e^{\varepsilon(u_{13} - v_3)} & R_{32} e^{\varepsilon(u_{23} - v_3)} & -\lambda_3^c
\end{pmatrix},$$

(16)

where $\lambda_x^c = \sum_{y \neq x} R_{y|x}^c$. We numerically calculate the probability distribution $\rho^{eq}_{\alpha, \varepsilon}(x)$ for various values of $\alpha$. Figure 1(a) shows the pumping density resulting from the set of $\rho^{eq}_{\alpha, 0}(x)$ and $\rho^{st}_{\alpha, \varepsilon}(x)$.

Second, we map the pumping density when the same system works as a stochastic pump. For the conjugate driving, we consider a uniform non-conservative force $f$ in the direction of $x$, i.e. $\varepsilon = \beta f$. The rate constants $R_{y|x}^c$ is

$$R^c = \begin{pmatrix}
-\lambda_1^c & R_{12} e^{\varepsilon} & R_{13} e^{\varepsilon} \\
R_{21} e^{\varepsilon} & -\lambda_2^c & R_{23} e^{\varepsilon} \\
R_{31} e^{\varepsilon} & R_{32} e^{\varepsilon} & -\lambda_3^c
\end{pmatrix}. $$

(17)

The pumping density is shown in Fig. 1(b). Comparing Fig. 1(b) with Fig. 1(a), we confirm that efficient pumping depends on the type of current to be pumped even though the same system acts as a heat pumps as well as a stochastic pump.

**Discussions:** We have developed a unified viewpoint on pumping and nonequilibrium thermodynamics by introducing a driven counterpart to pumping. With our
unified viewpoint one can not only rederive various pumping results such as \([9, 12]\) and \([13]\), but also design new pumping based on one’s knowledges on NESS. The information on the stationary distribution or the Fisher information matrix in NESS provides the estimate of the total pumping for the designed protocol. Once we construct pumping density map in the parameter space, we can design efficient pumps. When the information on the stationary distribution is not accessible, we can use \([13]\) for which a better designed protocol shows a steeper slope of work as a function of \(\varepsilon\). When the designed protocol is a finite speed, we can use \([15]\) for an estimate of the total pumping.

We comment that the geometric effects of pumping in \([12]\) and in \([10, 14]\) have the same origin as the geometric effect of excess heat reported in \([19]\). This is because the relation \([14]\) is a version of an extended Clausius relation, in which the excess heat equals the entropy difference in the precision of \(O(\varepsilon)\) \([16]\).

**Derivation of \([9]\):** We reported in \([15]\) that the probability distribution of NESS under the steady driving field \(\varepsilon\) has a linear response representation, 
\[
\rho^{\text{eq}}_{\alpha,\varepsilon}(\Gamma) = \rho^{\text{eq}}_{\alpha}(\Gamma) \exp \left[ -\varepsilon \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} \right] + O(\varepsilon^2),
\]
where a conditioned expectation is defined as
\[
\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} = \int d\Gamma’ \delta(\Gamma(0) - \Gamma’)/\mathcal{T}^{\text{eq}}_{\alpha}(\Gamma’) \Xi(\Gamma’),
\]
with a fixed initial state \(\Gamma\). The notation \((\alpha)\) represents the protocol in which the parameters are kept constant at \(\alpha\) and \(\mathcal{T}^{\text{eq}}_{\alpha} = \mathcal{T}_{\alpha,0}\). The conditioned average \(\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}}\) gives the total transfer observed in the relaxation process from the state \(\Gamma\). There is no transfer on average in equilibrium, i.e.,
\[
\int d\Gamma \rho^{\text{eq}}_{\alpha}(\Gamma) \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} = 0.
\]

We first concentrate on the protocol of an infinitesimal stepwise change from \(\alpha\) to \(\alpha’ = \alpha + \Delta \alpha\). Even though we do not observe any current before the stepwise change, we may observe it in the relaxation process after the stepwise change. Noting that \(\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}}\) is the total transfer in the relaxation from the state \(\Gamma\), the total transfer after the stepwise change is
\[
\langle \Xi \rangle_{\varepsilon} = \int d\Gamma’ \rho^{\text{eq}}_{\alpha}(\Gamma) \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}},
\]
\[
= -\int d\Gamma’ (\rho^{\text{eq}}_{\alpha’}(\Gamma) - \rho^{\text{eq}}_{\alpha}(\Gamma)) \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}},
\]
where we subtract \([20]\) from the first line of \([21]\) in order to obtain the expression in the second line. If \(\rho^{\text{eq}}_{\alpha}(\Gamma) = \rho^{\text{eq}}_{\alpha’}(\Gamma) + O(\Delta \alpha)\) and \(\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} = \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} + O(\Delta \alpha)\), then
\[
\langle \Xi \rangle_{\varepsilon} = -\Delta \alpha \cdot \int d\Gamma’ (\nabla_{\alpha} \rho^{\text{eq}}_{\alpha}(\Gamma)) \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}}
\]
with an error of \(O(\Delta \alpha^2)\).

As the next step, we refer to the representation \([18]\), where \(\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}}\) is related to \(\rho^{\text{eq}}_{\alpha,\varepsilon}\). Therefore, it is apparent that
\[
\langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} = \partial_{\varepsilon} \psi^\alpha_{\varepsilon}(\Gamma^*)|_{\varepsilon=0}.
\]

Substituting \([23]\) into \([22]\), we have
\[
\langle \Xi \rangle_{\varepsilon} = -\Delta \alpha \cdot \int d\Gamma' (\nabla_{\alpha} \rho^{\text{eq}}_{\alpha}(\Gamma')) \partial_{\varepsilon} \psi^\alpha_{\varepsilon}(\Gamma')|_{\varepsilon=0},
\]
\[
= \Delta \alpha \cdot \int d\Gamma' \rho^{\text{eq}}_{\alpha}(\Gamma') \nabla_{\alpha} \psi^\alpha_{\varepsilon}(\Gamma') \partial_{\varepsilon} \psi^\alpha_{\varepsilon}(\Gamma')|_{\varepsilon=0},
\]
where the negligible error term of \(O(\Delta \alpha^2)\) is ignored. We used \(\rho^{\text{eq}}_{\alpha}(\Gamma) = \rho^{\text{eq}}_{\alpha}(\Gamma^*)\) to have the first line of \([24]\).

Finally, we note that any adiabatic protocol is the accumulation of infinitesimal steps. We need to extend \([24]\) to the line integral along the protocol \(\alpha\), as is expressed in \([9]\). In order to arrive at expression \([10]\), we use an identity,
\[
\int d\Gamma' \rho(\Gamma) \partial_{\varepsilon} \psi(\Gamma) = \int d\Gamma' \rho(\Gamma) \partial_{\varepsilon} \psi(\Gamma),
\]
which is derived from integration by parts and the conservation law \(\int \rho(\Gamma) d\Gamma = 1\).

**Derivation of \([14]\):** We again begin from an infinitesimal stepwise change. The mechanical work is written as
\[
\langle W \rangle_{\varepsilon} = \Delta \alpha \cdot \int d\Gamma' \rho^{\alpha}(\Gamma') (\nabla_{\alpha} H^\alpha(\Gamma') + O(\Delta \alpha^2)).
\]

We substitute the representation \([18]\) and expand it in \(\varepsilon\) to obtain
\[
\langle W \rangle_{\varepsilon} = \Delta \alpha \cdot \left( \langle \nabla_{\alpha} H^\alpha \rangle_{\varepsilon} - \varepsilon \langle \nabla_{\alpha} H^\alpha(\Gamma) \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}} \rangle_{\rho^{\alpha}} \right) + O(\varepsilon^2).
\]

The kernel of \([22]\) in the r.h.s. is transformed as
\[
\langle \nabla_{\alpha} \rho^{\alpha}(\Gamma) \langle \Xi \rangle \rangle_{\Gamma,\varepsilon=\text{eq}} = \beta \rho^{\alpha}(\Gamma) \langle \nabla_{\alpha} F - \nabla_{\alpha} H^\alpha(\Gamma) \rangle \langle \Xi \rangle_{\Gamma,\varepsilon=\text{eq}},
\]
and therefore,
\[
\langle \Xi \rangle_{\varepsilon} = \beta \Delta \alpha \cdot \langle \nabla_{\alpha} H^\alpha(\Gamma) \langle \Xi \rangle \rangle_{\Gamma,\varepsilon=\text{eq}} + O(\Delta \alpha^2),
\]
where we applied \([20]\) to delete the term with \(\nabla_{\alpha} F\). Substituting \([20]\) and \(\langle \nabla_{\alpha} H^\alpha \rangle_{\rho^{\alpha}} = \nabla_{\alpha} F\) into \([27]\), we arrive at the work relation \([14]\) for the step protocols. By generalizing this to the accumulation of infinitesimal steps, we arrive at the final formula \([14]\).

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