Thermoelectric generators as self-oscillating heat engines

Robert Alicki

Institute of Theoretical Physics and Astrophysics, University of Gdańsk, 80–952 Gdańsk, Poland

E-mail: fizra@ug.edu.pl

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Abstract

In a previous paper [1] a model of a solar cell was proposed in which the non-periodic source of energy—photon flux—drives the collective periodic motion of electrons in a form of plasma oscillation. Subsequently, plasma oscillations are rectified by the p–n junction diode into dc (work). This approach makes a solar cell similar to standard macroscopic heat motors or turbines which always contain two heat baths, the working medium and the periodically moving piston or rotor. Here, a very similar model is proposed in order to describe the operation principles of thermoelectric generators based either on bimetallic or semiconductor p–n junctions. Again plasma oscillation corresponds to a piston and sunlight is replaced by a hot bath. The mathematical formalism is based on the Markovian master equations which can be derived in a rigorous way from the underlying Hamiltonian models and are consistent with the laws of thermodynamics.

Keywords: thermoelectric generators, quantum heat engines, quantum master equations

1. Introduction

While macroscopic heat motors or turbines always contain periodically moving elements such as pistons, flywheels and rotors [2, 3], photovoltaic devices, thermoelectric generators and biological engines driven either by sunlight (photosynthesis) or chemical energy seem, according to standard wisdom, to avoid any self-oscillation mechanisms. On the other hand extensive theoretical studies of quantum heat engines within the formalism of quantum open systems [4–9] show that the process of work extraction must involve a work reservoir, being a system of a single degree of freedom and executing an oscillatory motion which in the semi-classical limit can be replaced by periodic external driving. The presence of such a system is even necessary to properly define the notion of work as a deterministic form of energy. It
suggests that for the examples mentioned above a certain ‘hidden’ self-oscillation mechanism of work generation must also be present. Indeed, as already observed the standard picture of current generation as caused by the emerging electric field in a junction cannot be correct because a dc current cannot be driven in a closed circuit by a purely electrical potential difference [10]. In a previous paper [1] it was shown that the consistent description of work generation by a solar cell can be formulated, introducing a work reservoir in the form of THz plasma oscillations.

The aim of the present contribution is to show that a very similar model can explain the operation principles of thermoelectric generators. The basic difference between semiconductor and bimetallic devices concerns the frequency of plasma oscillations. For doped semiconductors plasma frequencies are in the THz domain (low infrared) [11, 12] and can be treated as slow while for metals they correspond to the ultraviolet region (PHz = 10^{15} Hz) and therefore are too fast to be effectively coupled to the thermal motion of a lattice. However, it is argued in the appendix, using a hydrodynamic model of electron gas, that under reasonable assumptions there should exist THz modes of plasma oscillations in bimetallic contacts.

2. A heat engine with a slow piston

Consider a simplified version of the generic quantum heat engine model introduced in [4], successfully used, e.g. in [5], and recently in [1] in the context of solar cells. The system, corresponding to the ‘working medium’ interacts weakly with two heat baths at different temperatures. The macroscopic piston is described by the external driving V(t) added to the free Hamiltonian H_0 of the system.

For slowly varying V(t) (in comparison to fast internal dynamics) the irreversible evolution of the system’s time-dependent reduced density matrix \( \rho(t) \) satisfies the following Markovian master equation (MME)

\[
\frac{d}{dt}\rho(t) = -i[H(t), \rho(t)] + \mathcal{L}(t)\rho(t),
\]

where \( H(t) = H_0 + V(t) \) is the total Hamiltonian of the system and \( \mathcal{L}(t) \) describes the action of the baths on the system (\( \hbar = \kappa_0 = 1 \)).

The periodic driving executed by a large semi-classical oscillator commutes with the system Hamiltonian and reads

\[
V(t) = g \sin(\Omega t)M, \quad [H_0, M] = 0,
\]

where \( g \ll 1 \) is a small amplitude of oscillations. The dissipative generator \( \mathcal{L}(t) \) obtained by the weak coupling limit procedure is a function of the magnitude of perturbation and can be written as

\[
\mathcal{L}(t) \equiv \mathcal{L}[\xi(t)], \quad \xi(t) = g \sin \Omega t.
\]

For all \( \xi \) the generators \( \mathcal{L}[\xi] \) possess the stationary states \( \tilde{\rho}[\xi] \) satisfying the identities

\[
\mathcal{L}[\xi] \tilde{\rho}[\xi] = 0, \quad \mathcal{L}'[\xi] \tilde{\rho}[\xi] = -\mathcal{L}[\xi] \tilde{\rho}'[\xi]
\]

where \( \mathcal{L}'[\xi] \equiv \frac{d}{d\xi} \mathcal{L}[\xi], \quad \tilde{\rho}'[\xi] \equiv \frac{d}{d\xi} \tilde{\rho}[\xi].
\]

2.1. Formula for power

The power \( P(t) \) provided by the engine and the net heat current \( J(t) \) supplied by the baths are defined as [4]
\[ P(t) = -\text{Tr}\left( \rho(t) \frac{dH(t)}{dt} \right), \quad J(t) = \text{Tr}\left( H(t) \frac{d\rho(t)}{dt} \right). \] (5)

Those definitions are the only ones which are consistent with the first and second law of thermodynamics and the time-dependence of the Hamiltonian is necessary to define work. The stationary average power output of the engine reads

\[ \bar{P} = -g\Omega \lim_{t_0 \to \infty} \frac{1}{t_0} \int_{t_0}^{\infty} \text{Tr}(\rho(t)M) \cos \Omega t \, dt. \] (6)

Expanding the average power with respect to \( g \) one obtains the second order approximation [1] (compare also [5])

\[ \bar{P} = -\frac{1}{2}g^2 \text{Tr}\left( \rho'[0] \frac{\Omega^2}{\Omega^2 + L^*[0]}L^*[0]M \right). \] (7)

where \( L^*[0] \) is the Heisenberg picture counterpart of the Schrödinger picture generator \( L[0] \).

To derive the final expression one assumes that, although the modulation is slow with respect to the intrinsic motion of the system, its frequency is much higher than the relaxation rate of the observable \( M \) hidden in \( L^*[0] \). Then, we have

\[ \bar{P} = -\frac{1}{2}g^2 \text{Tr}(\rho'[0]L^*[0]M). \] (8)

The lowest order formula (8) is still consistent with thermodynamics [1] and is the basic one for the further analysis of thermoelectric generators. Note that the stationary output power is proportional to the square of the amplitude of piston oscillations. This amplitude is a free parameter which is determined by the energy flux from the hot bath and the load attached to the oscillator (e.g. the resistance of the external electric circuit).

3. Model of a thermoelectric generator

The model of a thermoelectric generator is illustrated in figure 1. The working medium is an electron gas distributed in two boxes \( A \) and \( B \), corresponding to different materials, metals or doped semiconductors, connected by a junction. The electrostatic potential jump through the junction denoted by \( E_g \) is typically of the order of few eV. We assume that in the case of metals the density of electrons in box \( A \) is lower than in \( B \) which implies that the potential energy of the electron in box \( A \) is higher than in box \( B \). For semiconductors box \( A \) is a p-type and box \( B \) an n-type doped material. The hot bath interacts with the electrons in the interface region influencing their transport through the junction while the cold bath at the ambient temperature cools down the electrons in the bulk. In the junction region an interface between two different materials is formed with the local concentration of charges and built-in electrostatic potential. One assumes the existence of collective charge oscillations localized at the junction with typical frequencies in the THz domain. For semiconductor devices such oscillations are experimentally confirmed [11, 12], and interpreted as plasma oscillations with the plasma frequency

\[ \omega_p = \sqrt{\frac{e^2}{m_n \varepsilon_0}}, \] (9)

where \( n \) is the density of charge carriers and \( m_n \) their effective mass. The bimetallic junction is more complicated as the bulk and surface plasma oscillations in metals possess frequencies in the PHz domain (\( 10^{15} \)Hz) and hence cannot be effectively excited by thermal phonons. In
the appendix the mechanism of slow coherent charge oscillations is proposed which is related to the particular physical structure of metal–metal junctions.

The electrons in the two boxes $A$ and $B$ are described by two sets of annihilation and creation operators $a_k, a_k^\dagger$ and $b_l, b_l^\dagger$, respectively, subject to canonical anticommutation relations. The electrons are treated as non-interacting fermions moving in a self-consistent potential with the unperturbed Hamiltonian

$$H_0 = \sum_k E_a(k) a_k^\dagger a_k + \sum_l E_b(l) b_l^\dagger b_l.$$  \hfill (10)

In a junction a non-homogeneous free carrier distribution created in a self-consistent built-in potential can be perturbed producing collective plasma oscillations with the frequency $\Omega$. These oscillations periodically modulate the Hamiltonian (10) but the detailed mechanism is slightly different for semiconductor and bimetallic junctions (see the appendix for the derivations). In both cases the associated time-dependent perturbation which should be added to the electronic Hamiltonian (10), is proportional to the difference of electron densities in boxes $B$ and $A$ and has a mean-field form ($\xi(t) = g \sin \Omega t$)
Here, $\xi$ is a small dimensionless parameter describing the magnitude of deformation, $V_A$, $V_B$ are the volumes of the boxes, $V_J$ is the effective volume of the junction region and $E_g$ is the relevant energy scale chosen here to be the potential jump across the junction. Remember that for the semiconductor p–n junction box $A$ corresponds to the `p' part and $B$ to the 'n' part. In this case $-a_k^\dagger a_k$ should be rather interpreted as the number operator of holes minus an irrelevant constant.

The interaction with two independent heat baths is described by the sum of two terms: $\mathcal{L}_c$ for the cold bath at the ambient temperature $T$ and $\mathcal{L}_h$ for the hot bath at the temperature $T_1 > T$. The coupling to the cold bath thermalizes electrons in both boxes independently and hence preserves separately both electron numbers $\sum_k a_k^\dagger a_k$ and $\sum_k b_k^\dagger b_k$. It means that $\mathcal{L}_c^0M = 0$ and $\mathcal{L}_h^0$ disappears from formula (8) for stationary power output. Note that in the discussed idealized model, heat transport leading to a temperature gradient is neglected, which corresponds to the infinite heat conductivity of the bulk.

3.1. Hot bath generator and stationary state

The generator $\mathcal{L}_h$ describes thermally induced electron transitions from one box to another across the potential barrier and can be written in the following form

$$\mathcal{L}_h[0] = \sum_{kl} \mathcal{L}_{kl}^{(b)}$$

where

$$\mathcal{L}_{kl}^{(b)} = \gamma_{kl} \left( a_k^\dagger b_1^\dagger \rho b_k a_k - \frac{1}{2} \{ b_1^\dagger a_k^\dagger a_k b_1^\dagger, \rho \} \right) + e^{-E_k/T} \gamma_{kl} \left( a_k^\dagger b_1^\dagger \rho b_k a_k - \frac{1}{2} \{ b_1^\dagger a_k^\dagger a_k b_1^\dagger, \rho \} \right).$$

The single term (13) describes the following processes:

(a) electron transfer from box $A$ to $B$ accompanied by a positive energy release to the hot bath equal to

$$E_g(k) - E_g(l) \simeq E_g,$$

(b) the inverse process of electron transfer form the lower energy state in $B$ to the higher energy one in $A$ with the probability suppressed by the Boltzmann factor.

Condition (14) follows from the approximate energy conservation valid for the tunneling process through a barrier of height $E_g$ assisted by thermal fluctuations of the hot bath. This also means that the relaxation rates $\gamma_{kl}$ are essentially different from zero only if (14) is satisfied. They are proportional to the spectral density of the hot bath at $\omega_{kl} = E_g(k) - E_g(l) \simeq E_g$.

Although the generator $\mathcal{L}_h^0$ is absent in the formula for power the thermal relaxation of electrons in a given box is the fastest process and hence determines the form of the stationary state $\bar{\rho}$. Within a reasonable approximation one can assume that the stationary state of the electronic systems with the total Hamiltonian $H_0 + \xi M$ is a product of grand canonical ensembles for electrons in both boxes with the same temperature $T$ of the device and different electro chemical potentials $\mu_a$ and $\mu_b$, respectively. The associated density matrix has the form
The electro chemical potentials \( \mu_a, \mu_b \) are determined by the number of carriers and hence by density of electrons in both boxes. The difference of the electro chemical potentials is related to the measured voltage \( \Phi \) between \( A \) and \( B \)

\[
\mu_a - \mu_b = e \Phi.
\] (16)

### 3.2. Power and efficiency

One can insert all elements computed in the previous section into the expression for power \( (8) \). Then one uses the properties of the quasi-free (fermionic Gaussian) stationary state \( (15) \) which allow the reduction of the averages of even products of annihilation and creation fermionic operators into sums of products of the only non-vanishing two-point correlations

\[
\langle a_k a_k \rangle_0 = \delta_{kk} f_a (k), \quad \langle a_k a_k \rangle_0 = \delta_{kk} (1 - f_a (k)),
\]

\[
\langle b_l b_l \rangle_0 = \delta_{ll} f_b (l), \quad \langle b_l b_l \rangle_0 = \delta_{ll} (1 - f_b (l)).
\]

Here \( \langle \cdots \rangle_0 \) denotes the quantum average with respect to the state \( \tilde{\rho}[0] \) given by \( (15) \), and \( f_a (k) \) and \( f_b (l) \) are the Fermi–Dirac statistical distribution functions

\[
f_a (k) = \frac{1}{e^{\beta E_a (k) - \mu_a} + 1}, \quad f_b (l) = \frac{1}{e^{\beta E_b (k) - \mu_b} + 1},
\] (17)

with \( \beta = 1/T \). Inserting the elements defined above into the general formula \( (8) \) one obtains the leading order contribution to power in the following form

\[
\tilde{P} = \frac{g^2 E_a^2 V_f^2}{T} (V_A + V_B)(\bar{n}_b - \bar{n}_a)
\]

\[
\times \frac{1}{V_A V_B} \sum_{kl} \gamma_{kl} (e^{-E_f / T} [1 - f_a (k)] f_b (l) - [1 - f_a (l)] f_b (k)),
\] (18)

where \( \bar{n}_a = \frac{1}{V_A} \langle \sum_k a_k a_k \rangle_0, \bar{n}_b = \frac{1}{V_B} \langle \sum_l b_l b_l \rangle_0 \) are electron densities. Using \( (16) \) one can rewrite \( (18) \) as

\[
\tilde{P} = \frac{g^2 E_a^2 V_f^2}{T} (V_A + V_B)(\bar{n}_b - \bar{n}_a) \Gamma
\]

\[
\times \left\{ \exp \left\{ \frac{1}{T} \left( 1 - \frac{T}{T_1} \right) E_f - e \Phi \right\} - 1 \right\},
\] (19)

where \( \Gamma = \frac{1}{V_A V_B} \sum_{kl} \gamma_{kl} (1 - f_a (l)) f_b (k) \geq 0 \) is finite in the thermodynamic limit.

The condition for work generation in the discussed model of idealized thermoelectric device reads \( (\Delta T = T_1 - T) \)

\[
e \Phi < e \Phi_0 = E_f \left( 1 - \frac{T}{T_1} \right) = \frac{E_f T}{T_1} \Delta T
\] (20)
The inequality in (20) implies that $\phi_0$ can be interpreted as an \textit{open-circuit voltage} of the device and $\frac{E_g}{k_B T}$ as the \textit{relative Seebeck coefficient}. By closing the external circuit one reduces the voltage and hence the output power is strictly positive driving collective charge oscillations (positive feedback). The charge oscillations are subsequently rectified by the diode mechanism of the junction producing a dc.

The presence of the Carnot factor $1 - T/T_1$ suggests also the interpretation of equation (20) in terms of thermodynamic efficiency. Indeed, the transport of a single electron from box B to A through the junction requires at least $E_g$ of thermal energy extracted from the hot bath. Then, part of energy $E_g$ is transformed into useful work, equal at most to $e\phi_0$ per single electron flowing in the external circuit.

For real systems the efficiency is much smaller than the Carnot bound because of the finite heat conductivity and damping of plasma oscillations. Similarly, for a typical $T_1 \sim 500$ K, the relative Seebeck coefficient given by (20) is of the order of mV K$^{-1}$, which is comparable to the highest values obtained for some semiconductors. For metals, the neglected irreversible transport processes reduce the Seebeck coefficient by two or three orders of magnitude.

4. Conclusions

The aim of this paper is to show that the idea of ‘hidden self-oscillations’ proposed in [1] for solar cells can be extended to thermoelectric generators. It is illustrated by an idealized model in which heat conduction is neglected. The ‘moving piston’ is again represented by collective charge oscillations with frequencies in the THz domain. While such oscillations are observed in p–n junctions, for bimetallic ones a plausible mechanism of their generation is proposed in the appendix. The challenging open question is to apply similar ideas to the following systems: fuels cells and ‘biological engines’ based on organic molecules. In the first case thermal baths are replaced by chemical ones and plasma oscillations in electrolytes, predicted e.g. in [14], could serve as a piston. For the second case, including organic photovoltaic systems, proton pumps and photosynthesis, the important role of coherent molecular oscillations in energy and charge transfer has been observed (see e.g. [15]).

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Appendix

In this appendix the arguments for the existence of slow plasma oscillation modes in bimetallic junctions are presented and the origin of the diagonal modulation Hamiltonian is explained for both semiconductor and bimetallic devices. Here, $\hbar$ is put explicitly in the formulas.

A.1. Slow charge oscillations in bimetallic junctions

In order to find collective oscillating modes of electron gas in a bimetallic junction one can use the one-dimensional hydrodynamic model. The basic variables are: the electron density
Here $m_\ast$ is the effective mass of an electron and $n_0 = n_0(x)$ is the time-independent density of background positive ions (jellium model). The pressure $p(x, t)$ is not an independent variable but is given by the standard formula for the degenerated electron gas
\begin{equation}
  p = \frac{(3\pi^2)^{2/3}}{2m_\ast} n_0^{5/3} \equiv \alpha n^{5/3}.
\end{equation}

The stationary solution is given by $v = 0$ and the density $\hat{n}(x)$ which up to a certain smoothing essentially follows the background density $n_0(x)$. Consider a small perturbation of the stationary electron gas distribution in a form of well-localized wave packet, i.e. $n(x, t) = \hat{n}(x) + \delta n(x, t)$. Inserting also $\psi(x, t) = \tilde{\psi}(x) + \delta \psi(x, t)$ into equations (21)–(23) and assuming that the stationary density is a slowly varying function of $x$ in comparison with the variation of the wave packet $\delta n$, one obtains a set of linearized equations
\begin{equation}
  m_\ast \partial_t v + \frac{5}{3} \frac{\alpha}{\hat{n}^{1/3}} \partial_x \delta n - e \frac{\partial}{\partial x} \delta \psi = 0,
\end{equation}
\begin{equation}
  \frac{\partial}{\partial t} \delta n + \hat{n} \frac{\partial}{\partial x} \delta \psi = 0,
\end{equation}
\begin{equation}
  \frac{\partial^2}{\partial x^2} \delta \psi - \frac{e}{\epsilon_0} \delta n = 0.
\end{equation}

Taking the derivative of equation (25) with respect to $x$, then inserting relations (26), (27) (omitting the derivatives of $\hat{n}$) one obtains the following one-dimensional Klein–Gordon-like equation
\begin{equation}
  \left[ \frac{\partial^2}{\partial t^2} - c_p^2 \frac{\partial^2}{\partial x^2} + \omega_p^2 \right] \delta n = 0
\end{equation}
for the density perturbation $\delta n$. Here, the maximal velocity $c_p$ is given by
\begin{equation}
  c_p^2 = \frac{5}{3} \frac{(3\pi^2)^{2/3} \hbar^2}{2m_\ast^2 \hat{n}^{2/3}} \approx \frac{5}{6} \frac{\hbar^2}{v_F^2},
\end{equation}
where $v_F$ is the Fermi velocity and $\omega_p$ is given by (9).

For an inhomogeneous material such as a bimetallic junction $c_p$ and $\omega_p$ are assumed to be slowly varying functions of $x$. Using the analogy to the relativistic Klein–Gordon equation one concludes that the center of the localized perturbation $\delta n$ moves like a fictitious one-dimensional particle with the ‘relativistic’ Hamiltonian
\begin{equation}
  H(x, p) = \sqrt{c_p^2(x) p^2 + \hbar^2 \omega_p^2(x)}.
\end{equation}
The approximate Hamiltonian equations in the ‘non-relativistic regime’ (small $p$) read
\begin{align}
\dot{x} &= \frac{p}{M}, \quad M = \frac{\hbar \omega_p}{e^2} \\
\dot{p} &= -\frac{\partial}{\partial x} U(x), \quad U(x) = \hbar \omega_p(x).
\end{align}

To advocate the existence of slow modes in bimetallic junctions one can propose the qualitative but plausible shape of the effective potential $U(x)$ which interpolates between two bulk values characteristic for both metals (see figure 2). The well in the middle can be explained by the presence of a large number of defects in the transition region between two different lattices. These defects trap a certain number of electrons reducing the density of free electrons and hence also the local value of $\hat{n}(x) \sim \sqrt{\rho}(x)$. As the jump of the effective potential $\Delta U = \hbar (\omega_p^{(B)} - \omega_p^{(A)})$ is of the order of few eV one can estimate the depth of the well as $\Delta E = 0.1 - 1$ eV (still higher than the typical value of $T_v \approx 0.05$ eV) and a reasonable value for its width as $L = 100$ nm (comparable to the typical roughness of well polished metallic surfaces). Using the harmonic well approximation $U(x) \simeq \frac{1}{2} M \Omega^2 x^2$ one can estimate the oscillation frequency of the plasmonic wave packet confined in the well by
\begin{equation}
\Omega = \sqrt{\frac{2\Delta E}{ML^2}}.
\end{equation}

Figure 2. Bimetallic junction. The shape of the effective potential for the plasmonic wave packet. It is instructive to compare the proposed shape of $U(x)$, proportional to $\sqrt{\rho}(x)$, with the very similar shape of electron number density obtained numerically in [13] for the case of an Al–Mg contact. In figure 1 of [13] the ideal plane surfaces are separated by a small distance of 0.3 nm. This gap produces physical effects similar to the real metal–metal interface.
Taking typical values $\omega_p \simeq 10^{15}\text{ s}^{-1}$ and $v_F \simeq 10^6\text{ m s}^{-1}$, one obtains $M \simeq 10^{-30}\text{ kg}$ and hence, finally $\Omega \simeq 1\text{ THz}$—the value comparable to that for plasma oscillations in semiconductor p–n junctions.

A.2. Effective modulation Hamiltonians

The aim of this section is to justify the choice of the diagonal modulation as a generic consequence of the weak coupling between the system and the driving oscillations.

Consider first the case of slow and weak perturbation described by the Hamiltonian $H_0 + \lambda V(t)$ which is applicable to p–n junctions. For a fixed $t$ one can apply the lowest order of the standard perturbation theory for approximate eigenvectors and eigenenergies

$$\phi_j \simeq \phi_j^{(0)},$$

$$E_j \simeq E_j^{(0)} + \lambda \langle \phi_j^{(0)}|V|\phi_j^{(0)} \rangle.$$  \hspace{1cm} (34)

Within this approximation one can use the effective time-dependent Hamiltonian with diagonal perturbation

$$H(t) = H_0 + \lambda \sum_j \langle \phi_j(0)|V(t)|\phi_j^{(0)} \rangle \langle \phi_j^{(0)}|\phi_j(0) \rangle.$$  \hspace{1cm} (35)

For the case of a bimetallic junction one should take into account the fact that the source of perturbation is a ‘wave packet’ with fast ‘internal’ charge oscillations at the frequency $\omega_p$ and slow oscillatory motion of its envelope. Hence, the resulting time-dependent Hamiltonian possesses the following structure

$$H(t) = H_0 + \lambda \int [f(t) \cos \omega_p t] V,$$  \hspace{1cm} (36)

where $f(t)$ describes the slowly varying envelope. In this case the terms linear in $\lambda$ are practically averaged out. Using the second order of the standard perturbation theory

$$\phi_j \simeq \phi_j^{(0)} + \lambda \sum_{k,j} \langle \phi_k^{(0)}|V|\phi_j^{(0)} \rangle \phi_k^{(0)}.$$  \hspace{1cm} (37)

and averaging over fast oscillations one obtains the following effective Hamiltonian

$$H(t) = H_0 + \frac{1}{2} \lambda^2 \int [f(t) \cos \omega_p t]^2 V,$$  \hspace{1cm} (38)

and hence much weaker than for the semiconductor devices. However, this is compensated by the much higher electron densities in metals.

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