Testing the violation of the Clausius inequality in nanoscale electric circuits

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The Clausius inequality, one of the classical formulations of the second law, was recently found to be violated in the quantum regime. Here this result is formulated in the context of a mesoscopic or nanoscale linear RLC circuit interacting with a thermal bath. Previous experiments in this and related fields are analyzed, and the possibilities of experimental detection of the violation are pointed out. It is discussed that recent experiments reached the range of temperatures where the effect should be visible and that a part of the proposal has already been confirmed.

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I. INTRODUCTION

The application of thermodynamics to electric circuits has a long and remarkably fruitful history.[1–6] In the late 1920's, by applying general principles of thermodynamics—in particular, the second law—Nyquist[1] deduced the spectrum of the fluctuation force acting in an equilibrium electric circuit. This result was much later confirmed by microscopic approaches[5,7–10] and became known as the Nyquist spectrum. Nearly 20 years later Brillouin[2] applied the second law to analyze a circuit containing rectifying elements. Another formulation of the second law—the Clausius inequality—was considered in the context of electric circuits by Landauer.[3] The equilibrium thermodynamics of linear and nonlinear circuits was thoroughly analyzed by Stratonovich.[5] Further research in this field was stimulated by two facts: first, by the technical importance of circuits in electronics and, second, by their feasibility, which allows one to create experimental conditions close to those in theory.[6]

In view of these successful applications of thermodynamics, one naturally expects that electrical circuits can play also a complementary role by acting as experimental and theoretical laboratories for testing new ideas and results in statistical thermodynamics itself. The present paper makes such an attempt in the context of our recent discussion of the applicability of the second law to quantum systems coupled to thermal baths.[11,12] The general philosophy of the approach is that thermodynamic relations are not introduced axiomatically or phenomenologically, but should be derived from first principles: namely, the laws of quantum mechanics. For linear systems—e.g., a set of harmonically bounded Brownian particles interacting with a quantum thermal bath—this program can be carried out exactly. As the main result we were able to check some formulations of the second law, whose validity in the classical domain was confirmed several times via analogous approaches.[3–5] One of these formulations—the Clausius inequality—appeared to be broken in the low-temperature quantum regime. Here we reformulate this result for a quantum linear RLC circuit.[7,8,13] Our purpose is rather straightforward: we explain that the above violation can be detected experimentally in low-temperature mesoscopic circuits. To this end we analyze some known experimental results and show that several important parts of our proposal were already realized in experiment.

Our plan for the present paper is the following: In Sec. II we will briefly describe the quantum RLC circuit coupled with a thermal bath. We continue with an explanation of the Clausius inequality in Sec. III. In the following section we analyze some experimental results, and in their context we make quantitative estimates for our effect. Our conclusions are presented in the last section.

II. RLC CIRCUIT AND ITS HEISENBERG-LANGEVIN EQUATIONS

A. Classical RLC circuit

The classical scheme of the simplest RLC circuit is well known. It consist of capacity C, inductance L, and resistance R. The loss of voltage across the resistance is given by the Ohm law IR, where $I=dQ/dt$ is the current and Q stands for the charge. The capacitor enters the total voltage as Q/C. Finally, the inductive element induces a magnetic field with the flux $\Phi=LI$, which in turn contributes to the voltage (Faraday's law). Altogether, one finally obtains

$$\dot{Q}=\frac{\Phi}{L},\quad \dot{\Phi}=-\frac{Q}{C}-\frac{R}{L}\Phi. \tag{2.1}$$

The first equation is just the definition of the current, and the second one expresses the fact that the total voltage in the closed circuit is zero. Apart from the term connected with the resistance, Eqs. (2.1) can be viewed as the canonical equation of motion generated by the Hamiltonian

$$H_S=\frac{\Phi^2}{2L}+\frac{Q^2}{2C}, \tag{2.2}$$

where Q and Φ are the canonical coordinate and momentum. Within the language of Brownian motion the contribution of the resistance in Eq. (2.1) corresponds to the Ohmic friction with the damping coefficient R. In the same context C corresponds to the inverse strength of the external harmonic potential, and L corresponds to the mass of the Brownian particle.

B. Quantum RLC circuit

Equation (2.2) makes obvious that for $R=0$ one can quantize the model regarding Φ and Q as the corresponding operators:

$$[Q,\Phi]=i\hbar. \tag{2.3}$$

Then Eqs. (2.1) are just the Heisenberg equations of the problem. A quantum description of electrical circuits became necessary at the beginning of 1980s with the appearance of gravitational-wave-measuring setups and Josephson junctions. These devices operate at low temperatures and are very susceptible to their environment, so that both the circuit and its photonic thermal bath have to be described quantum mechanically. Since then the problem of quantization for the electrical circuit was considered in numerous contributions (see, e.g., Refs. 7, 8, and 13) with special emphasis on the dissipative aspects of the problem. More recently interest in this subject was renewed in the context of low-temperature mesoscopic circuits.[16,17] Though within the classical approach the resistivity can be introduced phenomenologically, this is impossible for the quantum case, in particular because it will violate the Heisenberg relation. The cause is that even if Eq. (2.3) is valid at the initial moment, a non-Hamiltonian dynamics does not conserve it in time. Thus, the dissipative quantum situation should be investigated starting from a more fundamental level i.e., by explicitly describing the thermal bath. The strategy here is exactly the same as when studying the dynamics of open quantum systems in general:[9] One models the resistance as an open chain of linear LC circuits (thermal bath) attached to the studied circuit and then applies the standard canonical quantization scheme to the whole closed Hamiltonian system. In a second step one traces out the bath, since only the degrees of freedom of the initial circuit are considered to be observable. Since the bath consists of harmonic oscillators, this procedure can be realized explicitly. Omitting technicalities which can be found in Refs. 7–9 and 11, we will write down the final quantum Langevin equations

$$\dot{Q}=\frac{\Phi}{L}, \tag{2.4}$$

$$\dot{\Phi}=-\frac{Q}{C}-R\Gamma\int_0^t ds e^{-\Gamma(t-s)}\dot{Q}(s)+\eta(t)-R\Gamma e^{-\Gamma t}Q(0), \tag{2.5}$$

where Γ is the maximal frequency of the bath and where $\eta(t)$ is the quantum Gaussian noise (random emf) with the Nyquist spectrum:

$$\begin{aligned}K(t-t')&=\frac{1}{2}\langle\eta(t)\,\eta(t')+\eta(t')\,\eta(t)\rangle\\&=\frac{\hbar R}{\pi}\int_0^\infty d\omega\frac{\omega\coth(\frac{1}{2}\beta\hbar\omega)}{1+(\omega/\Gamma)^2}\\&\quad\times\cos\omega(t-t'),\end{aligned} \tag{2.6}$$

where $\beta=1/k_BT$ and we use units in which Boltzmann's constant $k_B=1$.

If Γ is much larger than other frequencies of the problem (this is the most typical situation), then for $t>0$ one can get the Langevin equation (2.5) in a more standard form

$$\dot{\Phi}=-\frac{Q(t)}{C}-RI(t)+\eta(t). \tag{2.7}$$

In the classical limit (large T) the spectrum (2.6) would become the Nyquist white noise spectrum

$$K(t-t')=RT\,\Gamma e^{-\Gamma|t-t'|}\approx 2RT\delta(t-t'), \tag{2.8}$$

but that regime will not be of our concern. Though in the classical situation the noise can be omitted at $T=0$, for the quantum case the presence of a resistivity without the corresponding noise is excluded.

Generally, one should keep the parameter Γ in Eq. (2.6) for the noise correlation function, since otherwise some divergences will occur. However, provided that Γ is large a concrete form of the cutoff function (here taken to be Lorentzian) is not essential.[9,11]

C. Stationary state of the circuit

Equation (2.7) is linear and can be solved exactly. We will not repeat the derivation of this solution, since it was thoroughly investigated in Refs. 9 and 11. Starting from any initial state the circuits relaxes to its stationary state, where Φ and Q are independent random Gaussian quantities with zero averages, $\langle\Phi\rangle=\langle Q\rangle=0$, and have the following dispersions:[7]

$$\langle\Phi^2\rangle=\int\frac{d\omega}{2\pi}\frac{\omega^2k(\omega)}{(1+\omega^2/\Gamma^2)[(\omega^2-\omega_0^2)^2+\omega^2R^2/L^2]},$$

$$\langle Q^2\rangle=\int\frac{d\omega}{2\pi}\frac{k(\omega)}{(\omega^2-\omega_0^2)^2L^2+\omega^2R^2},$$

$$k(\omega)=\hbar R\omega\coth\frac{\hbar\omega}{2T}, \tag{2.9}$$

where $\omega_0=1/\sqrt{LC}$ is the frequency of the free circuit. Statistically these variables are independent, which is expressed by the relation $\langle Q\Phi+\Phi Q\rangle=0$. Explicit formulas expressing $\langle Q^2\rangle$ and $\langle\Phi^2\rangle$ in terms of digamma functions are given in Refs. 11 and 12.

The disorder present in the circuit is characterized by the occupied phase-space volume

$$\Sigma=\frac{\Delta\Phi\Delta Q}{\hbar}\equiv\sqrt{\frac{\langle\Phi^2\rangle\langle Q^2\rangle}{\hbar^2}}. \tag{2.10}$$

The lower bound $\Sigma=\frac{1}{2}$ follows from the Heisenberg relation $\Delta\Phi\Delta Q\geqslant\frac{1}{2}\hbar$. It means that the charge and flux fluctuate close to their average values.

It is important to notice that in general the dispersions are not equal to their Gibbsian values

$$\langle\Phi^2\rangle_{\rm G}=\tfrac{1}{2}L\hbar\omega_0\tanh\tfrac{1}{2}\beta\hbar\omega_0,$$

$$\langle Q^2\rangle_{\rm G}=\tfrac{1}{2}C\hbar\omega_0\tanh\tfrac{1}{2}\beta\hbar\omega_0, \tag{2.11}$$

which are obtained by assuming a Gibbs distribution for the circuit, valid for a weak coupling with the bath—i.e., when taking $R\to 0$. That is why $\langle \Phi^2\rangle_G$ and $\langle Q^2\rangle_G$ do not contain the resistance R anymore, in contrast to the general expressions for $\langle \Phi^2\rangle$ and $\langle Q^2\rangle$, presented above.

It is natural to identify the average energy stored in the circuit with

$$U\equiv\langle H_S\rangle=\frac{\langle \Phi^2\rangle}{2L}+\frac{\langle Q^2\rangle}{2C}. \tag{2.12}$$

There is a general argument why the dispersions $\langle \Phi^2\rangle$ and $\langle Q^2\rangle$ are not equal to their Gibbsian values.[11,12] For $T\to 0$ the Gibbs distribution predicts that the circuit is in the ground state of its Hamiltonian H_S. Indeed, it can be checked that when the values (2.11) are inserted into Eq. (2.12), one gets $U=\frac{1}{2}\hbar\omega_0$, just the exact ground-state energy of the free (i.e., $R=0$) circuit. In quantum mechanics two interacting systems are typically not in pure states, even though the overall state of the total system may be pure. This is the intriguing property of quantum entanglement. Thus, we should not expect that a quantum circuit interacting non-weakly with its low-temperature bath will be found in a pure state. The approximate equalities $\langle \Phi^2\rangle\approx\langle \Phi^2\rangle_G$, $\langle Q^2\rangle\approx\langle Q^2\rangle_G$ are valid only for two particular cases: the weak-coupling situation, where in Eqs. (2.9) one takes $R\to 0$, and the classical case $\hbar/T\to 0$, where the temperature of the bath is so high that all signs of the quantum effects disappear. In both these situations the entanglement is very weak.

III. CLAUSIUS INEQUALITY

Let one of the parameters of the circuit (e.g., the inductivity L) be varied by an external source from L to $L+dL$, in a certain time interval. The variation is assumed to be very slow, so that at any moment the distributions of the flux and the charge are still given by Eqs. (2.9) with the instantaneous inductance $L=L(t)$. The variation itself is a accompanied by the work done by the external source. A part of that work is stored in the circuit, and the rest is transferred to the bath as heat. The energy budget of the variation is given by the first law

$$\frac{dU}{dL}=\frac{đW}{dL}+\frac{đQ}{dL},\quad \frac{đW}{dL}=\left\langle\frac{\partial H_S}{\partial L}\right\rangle=-\frac{\langle \Phi^2\rangle}{2L^2}, \tag{3.1}$$

where dU is the change of the energy stored in the circuit, $đW$ is the work done by external source on the system, and the difference between them, the heat $đQ$, is the energy that goes from the bath to the system.[10,11,14,15]

Thermodynamics imposes a general relation between the heat received by the circuit and the change of its phase-space volume $d\Sigma$. This statement was proposed by Clausius in the last part of the 19th century and became established as one of the formulations of the second law.[10,15] There are several levels of mathematical rigor by which the Clausius formulation can be presented.[10,11,14,15] For our present purposes it will be enough to use the simplest version:[11,12] If the circuit receives from the bath a positive amount of heat $đQ>0$, then its phase-space volume is increased: $d\Sigma>0$. On the other hand, if the circuit is subjected to the squeezing of its phase-space volume $d\Sigma<0$, then it has to release heat to the bath: $đQ<0$. In formulas it reads

$$đQ>0 \Rightarrow d\Sigma>0,$$

$$d\Sigma<0 \Rightarrow đQ<0. \tag{3.2}$$

In the classical domain everybody had a chance to observe the validity of the Clausius formulation when looking at a squeezed substance which heats its environment (e.g., a working pomp) or at a heated substance which tends to increase its volume (e.g., boiling water). For the reader who is familiar with the formal structure of thermodynamics we mention that the Clausius formulation can be presented as the Clausius inequality $đQ\leqslant TdS$, where S is the entropy of the system For our circuit the so-called von Neumann entropy reads[11]

$$S=(\Sigma+\tfrac{1}{2})\ln(\Sigma+\tfrac{1}{2})-(\Sigma-\tfrac{1}{2})\ln(\Sigma-\tfrac{1}{2}), \tag{3.3}$$

which is a well-behaved function, since as we discussed, the variable Σ is larger than or equal to $\frac{1}{2}$. It starts at $S(\frac{1}{2})=0$, increases monotonically, and behaves for large Σ as $S=\ln\Sigma+1+\mathcal{O}(1/\Sigma)$.

Equations (3.2) follow from assuming $đQ\leqslant T\,dS$ upon noticing $dS\propto +d\Sigma$.[11,12] In particular, for $T=0$ this inequality produces another version of the Clausius formulation: No heat can be extracted from a zero-temperature thermal bath. The remaining inequality $đQ(T=0)\leqslant 0$ says that heat can only be dumped into the bath.

As can be checked directly, if the dispersions of the flux and charge have their Gibbsian values (2.11), the Clausius statement is valid. This fact has received a special attention in the context of electrical circuits.[3,5] More generally, any statistical system which in its stationary state is described by Gibbs distribution has to satisfy the Clausius formulation.[10,11,14,15] So it is interesting to ask what will happen with the Clausius formulation if the temperature of the bath will be low enough—i.e., in the quantum situation. Notice that the physical relevance of this question is exactly the same as in the classical situation, since it is expected that thermodynamical relations should not change upon lowering the temperature. As we argued above, the dispersions $\langle \Phi^2\rangle$, $\langle Q^2\rangle$ are in general not Gibbsian, and the Clausius inequality need not be satisfied. Moreover, as was shown in Refs. 11 and 12 it can be violated in the quantum regime. Here we will present these results in the context of RLC circuits.

First of all, we notice that there is a general result $đQ/dL\geqslant 0$ valid in all ranges of the parameters.[12] To see the violation of the Clausius formulation we show that one can have $d\Sigma/dL\leqslant 0$. We consider low temperatures i.e., the quantum frequency $T/\hbar$ is comparable with at least one of the other frequencies ω_0, $1/(CR)$, and R/L involved in the problem. Depending on the value of the quality factor $\omega_0 L/R$ one can obtain from Eqs. (2.9) two extreme cases[12]

$$\frac{d\Sigma^2}{dL} = -\frac{R}{4L^2\omega_0}\ln\left(\frac{\Gamma}{\omega_0}\right), \quad \text{for} \quad \frac{\omega_0 L}{R} \gg 1, \tag{3.4}$$

$$\frac{d\Sigma^2}{dL} = -\frac{1}{\pi^2 L}\ln\left(\frac{\Gamma L^2}{CR^3}\right), \quad \text{for} \quad \frac{\omega_0 L}{R} \ll 1. \tag{3.5}$$

Recall that Γ is assumed to be much larger than any other frequency, so that both logarithms are positive, implying that in both cases $d\Sigma/dL$ is negative. The first case is realized in case of high quality (weak damping); it is then natural that $d\Sigma$ is proportional to the small inverse quality, since for $R=0$, Σ is just equal to $\frac{1}{2}$ (recall that the temperature is low) and, thus, does not vary with L. It is seen also that, apart from a small prefactor, $d\Sigma$ is multiplied by the logarithm of a large number. The second equation describes the low-quality situation, and here $d\Sigma$ is just proportional to the logarithm of a large number. This makes the situation especially interesting, since $Ld\Sigma^2/dL$ is at least of order unity. For both above cases the change of heat is given by[12]

$$\frac{đ\mathcal{Q}}{dL} = \frac{\hbar R}{2\pi L^2} > 0. \tag{3.6}$$

Two things have to be noted with this formula: it does not depend on Γ, not even through a logarithm, and its ratio to the ground-state energy $\sim\hbar\omega_0$ of the circuit just produces the quality factor $\hbar\omega_0/\Delta\mathcal{Q} \sim L\omega_0/R$, where $\Delta\mathcal{Q} \sim L đ\mathcal{Q}/dL$. So this zero-temperature heat is potentially observable for low-quality circuits. Notice that the very existence of the positive zero-temperature heat contradicts the Clausius inequality.

It should be mentioned that there is a widespread argument against a positive zero-temperature heat, stating that since at $T=0$ the bath is in its ground state, it cannot provide energy to the circuit. This is clearly incorrect, because if the circuit and bath do interact, the bath by itself cannot be in its ground state. It is always in a mixed state, and this is the property of quantum entanglement. Changing a parameter of the junction can lead to a transfer of zero-point energy from the bath to the junction, and this should be identified with heat, since it is arising from the unobservable bath modes.

IV. EXPERIMENTAL RESULTS

In the present section we will briefly discuss the possibilities of experimental detection of the violation of the Clausius formulation. In general, one needs to observe $\langle\Phi^2\rangle$ and $\langle Q^2\rangle$ for several different values of the inductivity L. These are sufficient to recover the corresponding changes of energy, the phase-space volume, and the work according to formulas (2.12), (2.10), and (3.1), respectively. In the second step one can check the consistency of the results by observing directly the work done by the external source, as can be done using an additional control circuit.[18] The observed work is then subtracted from the total energy to get the heat and to confirm $đ\mathcal{Q}(T\to 0)\neq 0$ and $đ\mathcal{Q}/dL>0$. Altogether, the main challenge of the experimental observation is in observation of the variances.

We are not aware of experiments which measure both $\langle\Phi^2\rangle$ and $\langle Q^2\rangle$ directly. However, there are several experiments which report indirect observations of the variances in different regimes. In Ref. 16 the authors considered mesoscopic electrical circuits in the context of single-charge tunneling. The used circuits had thickness of the order of 10 nm and width of the order of 1 μm. The observations allowed indirect determination of $\langle Q^2\rangle$. With the subsequent improvment made in Ref. 17, the correspondence with the theoretical expression (2.9) is perfect. The observations were done with $C=4.5$ fF, $L=4.5$ nH, and for R in the range 10^1—10^3 kΩ, which corresponds with the quality factor varying from 10^{-1} to 10^{-3}. To avoid thermal noises, the circuits were cooled down to 20 mK. At such a low temperature quantum effects are really dominating, since the quantum frequency $T/\hbar\sim 10^8$ s^{-1} is comparable with the system's characteristic frequencies $\omega_0\sim 10^9-10^{10}$ s^{-1}, $R/L \sim 10^8$ s^{-1}, and $1/(RC)\sim 10^9$ s^{-1}.

Let us now estimate the outcome of our effect with the above parameters. Taking $R=10^3$ kΩ one gets, from Eq. (3.6), $\Delta\mathcal{Q}\sim L\, đ\mathcal{Q}/dL \sim 10^{-19}$ J$\sim$1 eV, an observable effect. On the other hand, restoring Boltzmann's constant, the right-hand side of the Clausius inequality $k_B T\Delta S\sim k_B T$ takes a much smaller value, since for $T=20$ mK one has $k_B T \sim 10^{-25}$ J$\sim 10^{-6}$ eV. Thus to verify the violation of the Clausius inequality it suffices to take the sign of ΔL as positive, which brings a positive $\Delta\mathcal{Q}$.

V. CONCLUSION

The present paper discusses the Clausius inequality, one of the formulations of the second law, in the context of equilibrium RLC circuits. Following Refs. 11 and 12 it is confirmed that this inequality is broken if the bath temperature is low enough—namely, if the characteristic quantum time scale $\hbar/T$ is comparable with other relevant times of the circuit. The result can be briefly summarized as follows: localization of the system—i.e., decrease of its entropy or phase-space volume, can be connected with absorption of heat from the bath. This is in a sharp contrast with the classical experience, where localization occurs with emission of heat. We provide a simple and sufficiently general formula (3.6), which describes the effect at low temperatures.

One of our main purposes was to compare our result with recent experiments done on nanoscale low-temperature circuits.[16,17] This comparison led us to conclude that an experimental verification of the Clausius inequality breaking is fully within the reach of modern experiments. It is, therefore, hoped that the present paper will stimulate further experimentation on the issue whether nonthermodynamic energy flows occur in nature.

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