Comment on "Cooling by Heating: Refrigeration Powered by Photons"

In a recent Letter, Cleuren *et. al.* [1] proposed a model of a refrigerator composed of two metallic leads connected to two coupled quantum dots and powered by (solar) photons. In their analysis the refrigerator can cool one of the leads to arbitrarily low temperature, $T_r \rightarrow 0$, with the cooling flux $\dot{Q}_r \propto T_r$. We comment that this model strongly violates the dynamical version of the third law of thermodynamics. Furthermore, under more realistic assumptions concerning transitions between dot levels mediated by an electromagnetic field, we show that their model will not operate as a refrigerator.

There are seemingly two independent formulations of the third law. The first, known as the Nernst heat theorem, implies that the entropy flow from any substance at absolute zero is zero. Consider a system coupled simultaneously to a few heat baths with the aim to cool one of these baths to zero temperature. The entropy flow from this bath, given by $-\frac{\dot{Q}_k}{T_k}$, satisfies the Nerst theorem if the heat current $\dot{Q}_k$ flowing from the bath to the system scales like $\propto T_k^{\alpha}$ with $\alpha > 1$.

The second formulation of the third law is a dynamical one, known as the unattainability principle: No refrigerator can cool a system to absolute zero temperature at finite time. The dynamics of the cooling process is governed by the equation

$$\dot{Q}_k(T_k(t)) = -c_V(T_k(t))\frac{dT_k(t)}{dt}, \qquad (1)$$

where c_V is heat capacity of the bath. Putting $\dot{Q}_k \propto T_k^{\alpha}$ and $c_V \propto T^{\delta}$, $\delta \geq 0$, we can quantify this formulation by evaluating the characteristic exponent ζ of the cooling process,

$$\frac{dT(t)}{dt} \propto -T^{\zeta}, \qquad T \rightarrow 0, \qquad \zeta = \alpha - \delta. \qquad (2)$$

Namely, for $\zeta < 1$ the bath is cooled to zero temperature in a finite time. This formulation is more restrictive than the Nernst heat theorem and imposes limitations on the spectral density and the dispersion law of the heat bath [2].

The model of the refrigerator presented in Ref. [1] strongly violates the unattainability principle. For an electron reservoir at low temperatures, heat capacity is $c_V \propto T$. The heat current of the refrigerator of Ref. [1] is $\dot{Q}_r \propto T_r$, therefore, one obtains $\zeta = 0$, and zero temperature is reached at finite time.

Finding the flaw in the analysis of Ref. [1] is not a trivial task. A possible explanation emerges from the assumption made in Ref. [1] that transitions between lower and higher levels within the individual dots are negligible. However, photon- assisted tunneling between dots produces a rather weak tunneling current [3], while quenching transition rates in the individual dots are at least comparable and hence cannot be neglected.

A modified master equation that includes quenching transitions can be constructed for a five-level system: $\dot{\vec{p}} = M \cdot \vec{p}$, where $\vec{p} = (p_0, p_{ld}, p_{rd}, p_{lu}, p_{ru})^T$. Here, p_0 is the probability of finding no electron in the double dots and p_{ij} is the probability of finding one electron in the corresponding energy level, with l for left, r for right, d for down, and u for up. The 5×5 matrix M contains all transition rates, including quenching transition within the individual dots. Using this modified model, we can show analytically that, under the technical assumption that strictly positive quenching rates are equal for both dots, the condition for cooling ($\dot{Q}_r > 0$) and the condition of zero net electric current cannot be simultaneously satisfied at the stationary state. On the other hand, a crucial condition for this device to operate as a refrigerator [1] is that there is no net electric charging of the baths (leads). Otherwise, the electric current flowing through the device must be compensated by an external flow of electrons from the hot to the cold bath which would annihilate the cooling effect.

In conclusion, transitions in the individual dots, which are always present in real systems, cannot be neglected when treating electron transport in the double-dot systems. The dynamical form of the third law is a strong tool for testing designs of such nanodevices acting as refrigerators. Quantum models of refrigerators powered by heat (absorption refrigerators), which do not violate the third law, were studied in Refs. [2,4,5].

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