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Comment

Comment on "Loss-Free Excitonic Quantum Battery"

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Quantum batteries have primarily been modeled as an ensemble of isolated systems that store energy and from which work can be extracted by applying unitary transformations.[1] Only recently have investigations begun in the direction of *dissipative* quantum batteries[2−4] wherein the charge of the battery is protected against a dissipative environment.

In ref 2, a novel model for a quantum battery is proposed on the basis of a degenerated quantum system with a topological symmetry. The working substance consists of a *p*-benzene ring that is initially prepared in a dark-state, $\rho_{\mathrm{dark}} = |\mathrm{DS}\rangle\langle\mathrm{DS}|$ with $|\mathrm{DS}\rangle = 1/2(|5\rangle + |6\rangle - |2\rangle - |3\rangle)$, considered as the "charged" state of the battery. The geometric symmetries of the *p*-benzene are translated to open system symmetries[5] since the environment selectively acts on sites 1 and 4 independently. The authors claim that after breaking the system's symmetry, by the addition of an external probe, the energy in the system is redistributed, and it is transferred to site 4, where it can be harnessed by connecting a sink. The system dynamics are calculated in the single-excitation picture by a mixed quantum-classical method known as the deterministic evolution of coordinates with initial decoupled equations (DECIDE).[6] The authors present only the system dynamics but do not discuss the energetics (e.g., ergotropy), which is essential to understand a quantum battery's proper functioning. Moreover, the simulations do not show how exactly the energy can be extracted using a sink in this complex set up. We point out that the capacity of a system of providing excitations is not enough to be considered a battery. There are examples of passive states (e.g., states whose ergotropy is zero and therefore they cannot provide any useful work)[1] that present population imbalance in site representation.

We do not dispute their approach or their result on the single-excitation populations. We argue that observing only the single-excitation populations is insufficient to know the energetics. Specifically, since an infinite probe is attached to break the symmetries, it not only redistributes the energies as the authors suggest but also exchanges energy with the system. Moreover, the accumulation of a single-excitation population at site 4 does not ensure that this site has more energy (from a many-body perspective) that can be extracted to do useful work. Thus, despite their single-excitation populations being correct, the *p*-benzene's energetics do not behave as a battery.

In order to capture the energetics we have modeled the same system in the many-body fully quantum picture ($\hbar = 1$) using the Markovian Redfield equation,[7]

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\mathrm{i}[H, \rho] + \sum_{k=1,4,p} \mathcal{R}_k[\rho] + \mathcal{L}_{\mathrm{sink}}[\rho] \tag{1}$$

where the dissipator $\mathcal{R}_k$ is given by

$$\mathcal{R}_k[\rho] = \int_0^\infty \mathrm{d}\tau\, [S_k, \rho S_k(-\tau)] C_k(\tau) + \mathrm{Hc} \tag{2}$$

Above $S_k(\tau) = \mathrm{e}^{\mathrm{i}H\tau} S_k \mathrm{e}^{-iH\tau}$ is the freely evolving system operator that connects to the bath, $C_k(\tau) = \mathrm{Tr}[B_k B_k(\tau) \mathrm{e}^{-\beta_k H_B^k}/Z_B]$ is the correlation function of the bath with Z_B being the partition function of the bath at inverse temperature β_k and $B_k(\tau) = \mathrm{e}^{\mathrm{i}H_B^k\tau} B_k \mathrm{e}^{-iH_B^k\tau}$. Here, $H_B{}^k$ is the Hamiltonian of the kth bath, H_B^p is the probe Hamiltonian, $S_k = \sigma_k^+\sigma_k^-$ for $k = 1, 4$ and $S_p = \sqrt{\chi}(\sigma_2^+\sigma_2^- + \sigma_3^+\sigma_3^-)$. These operators are the generalization of the ones used in ref 2 to the multiple-excitation general case. We want also to point out that the baths and the probe preserve the total number of excitations of the system as they commute with the total number operator defined as $N = \sum_{i=1}^6 \sigma_i^+\sigma_i^-$. Thus, in the many-body representation even with a probe we obtain seven steady states each corresponding to the number of particles in the system ranging from 0 to 6. In this comment we will choose $\beta_1 = \beta_2 = [k_B T]^{-1} = \beta_p = [k_B T_p]^{-1}$ similar to ref 2. We also attach a sink $\mathcal{L}_{\mathrm{sink}}[\rho] = \Gamma\left(\sigma_4^-\rho\sigma_4^+ - \frac{1}{2}\{\sigma_4^+\sigma_4^-, \rho\}\right)$ in order to extract energy in the discharging phase which breaks the particle-number conserving symmetry giving a single unique steady state. The many-body system Hamiltonian reads

$$H = \sum_{i=1,\ldots,6} \epsilon_i \sigma_i^+\sigma_i^- + t \sum_{\langle i,j\rangle} (\sigma_i^+\sigma_j^- + \mathrm{Hc}) \tag{3}$$

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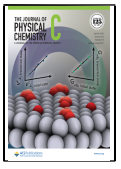

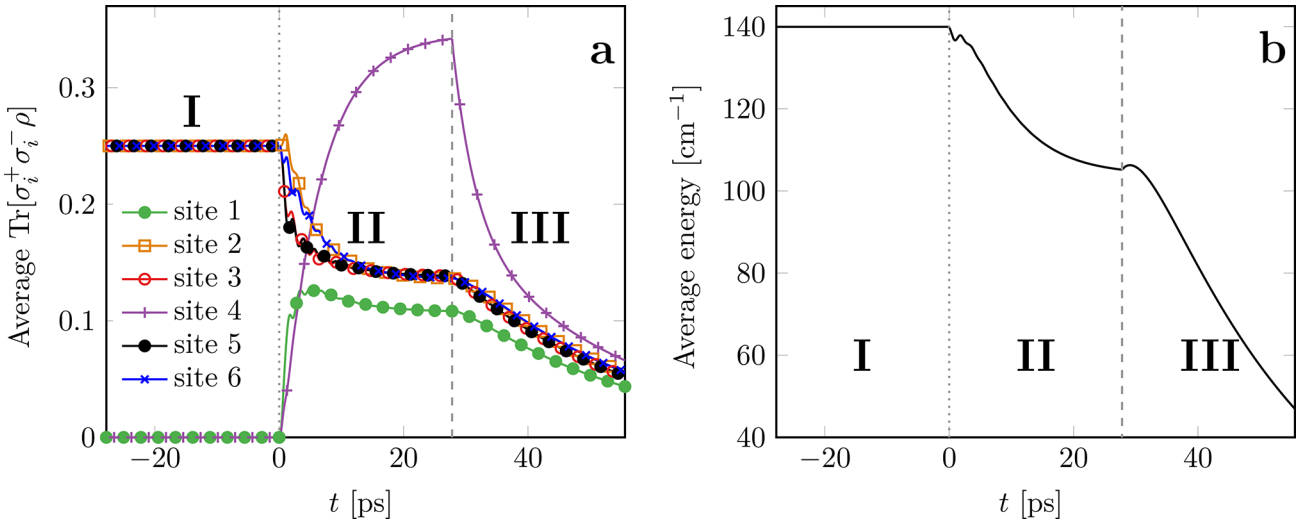

Figure 1. Many-body Redfield results for time-dependent populations and average energy. The initial conditions for region I is the dark state ρ_{dark} and for region III is the steady state of region II. The parameters for the system are $\epsilon = 200$ cm^{-1}, $\epsilon_1 = 250$ cm^{-1}, $\epsilon_4 = 0$ cm^{-1}, $t = -60$ cm^{-1}, $\lambda_b = 35$ cm^{-1}, $\lambda_p = 10$ cm^{-1}, $\Gamma = 0.1$, $\omega_c = \omega_p = 106$ cm^{-1} (Lorentz–Drude spectral density cutoff), $T = T_p = 300$ K (chosen same as Figure 1 in ref 2).

with $\sigma_i^{\pm}$ being spin-1/2 Pauli matrices. As in the single-excitation picture, the system presents a symmetry defined by the unitary operator $\Pi = \exp[i\pi(\sigma_1^+\sigma_1^- + \sigma_4^+\sigma_4^- + (\sigma_2^+\sigma_6^- + \sigma_3^+\sigma_5^- + \text{Hc}))]$.

We have calculated the populations for each site, defined as $\text{Tr}[\sigma_i^+\sigma_i^-\rho]$, as well as the average energy of the system. The populations found in Figure 2 of ref 2 are very similar to the ones we obtain in Figure 1a. It is important to note here that instead of this definition of populations, we could have chosen the projection of the many-body reduced density matrix ρ in the single-excitation subspace, i.e., $\langle i|\rho|i\rangle$. However, these again show the same behavior (not shown). The imbalance between sites 1 and 4 is mainly due to the counter-rotating terms that are not ignored in the Redfield dynamics.[8] Besides the populations we also calculate the average energy of the battery $\text{Tr}[H\rho]$ in Figure 1b. Note that the average energy of the battery is not given by the sum of site populations weighted by the on-site energy, i.e., $\sum_i \epsilon_i \text{Tr}[\sigma_i^+\sigma_i^-\rho]$. This is because the energy is also stored between the sites due to the hopping t and site-coherences.

The system evolution is divided into three different regimes that correspond to the regions in Figure 1. Region I is without a probe or sink ($\chi = 0$ and $\Gamma = 0$), Region II is with only a probe ($\chi = 1$ and $\Gamma = 0$), and Region III is with both probe and a sink ($\chi = 1$ and $\Gamma = 0.1$). Once the probe is attached (region II), the system's energy decreases, and it is not merely rearranged to the various sites. In terms of battery operation, energy is extracted from the system (working substance) during the discharging phase $\chi = 1$ instead of being rearranged to be ready for extraction. The energy reduction is without a load (sink) being attached, meaning this would result in a "leaky" battery. Once the sink is attached to site 4, the exit site's population decreases as claimed in ref 2. Since the bath and the probe are kept at the same temperature the resultant $t \to \infty$ state is passive, which would make it impossible to extract work even if unitary transformations (instead of attaching a sink) are performed in the discharging phase.[1]

From the above analysis, we conclude that when the symmetry is broken (discharging phase), there is not just an energy redistribution in the system, but the probe exchanges energy with the system. For the number conserving probe we find that the attachment of the probe makes it a *leaky* battery. The sink takes an infinite time to completely drain the battery since energy is provided by the probe and baths. We have tried other number nonconserving probes and found the probe can even give energy to the system. In no case was the energy of the system conserved except for pure-dephasing probe, which would not change the populations of the system. Our analysis based on the exact many-body fully quantum dynamics and energetics raises serious doubts on whether the *p*-benzene system proposed in ref 2 is a practical loss-free quantum battery.

Finally, we want also to point out that the symmetry operator in eq 9 of ref 2, earlier stated in ref 5, is wrong as it is proportional to the identity operator. The correct one is

$$\hat{\Pi} = |1\rangle\langle 1| + |4\rangle\langle 4| + (|2\rangle\langle 6| + |3\rangle\langle 5| + \text{Hc}) \tag{4}$$

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Notes

The authors declare no competing financial interest.

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