Supplementary Information: Experimental signature of initial quantum coherence on entropy production

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In this document we detail the theoretical modelling of the photodynamics of the NV-center (Supplementary Note 1), and we report further experimental data concerning the reconstruction of the probability distribution resulting from the EPM scheme (Supplementary Note 2).

Supplementary Note 1

Theoretical modelling

In our experimental set-up, the photodynamics of the NV center is well described by a seven-level quantum model [1–3], which, however, can be effectively reduced to a two-level quantum system as detailed in the Supplementary Information of Refs. [3]. Here, for completeness of exposition, we report the modelling of this effective two-level open dynamics of the NV center, by resorting to the super-operator formalism [4] and working in the energy eigenbasis.

Let us thus model the NV center as a two-level quantum system subjected to a dissipative dynamics. The dissipative dynamics is induced by the laser pulses and it is described by the linear super-operators $\mathbf{S} \in \mathbb{C}^{4\times4}$ acting directly on the column vector $\mathrm{col}[\rho_t] \in \mathbb{C}^{4\times1}$, with $\mathrm{col}[\rho]$ denoting the vectorization of the density operator $\rho \in \mathbb{C}^{2\times2}$. The super-operator $\mathbf{S}$ is explicitly given by

$$\mathbf{S} = \frac{1}{2}\begin{pmatrix} 2 - p_{\mathrm{abs}}(k_{\mathrm{c}} - p_{\mathrm{d}}\cos\alpha) & p_{\mathrm{abs}}k_{\mathrm{sc}} & p_{\mathrm{abs}}k_{\mathrm{sc}} & p_{\mathrm{abs}}(p_{\mathrm{d}}\cos\alpha + k_{\mathrm{c}}) \\ p_{\mathrm{abs}}(k_{\mathrm{sc}} + p_{\mathrm{d}}\sin\alpha) & 2 - p_{\mathrm{abs}}(1 + k_{\mathrm{s}}) & -p_{\mathrm{abs}}(k_{\mathrm{s}} - 1) & p_{\mathrm{abs}}(p_{\mathrm{d}}\sin\alpha - k_{\mathrm{sc}}) \\ p_{\mathrm{abs}}(k_{\mathrm{sc}} + p_{\mathrm{d}}\sin\alpha) & -p_{\mathrm{abs}}(k_{\mathrm{s}} - 1) & 2 - p_{\mathrm{abs}}(1 + k_{\mathrm{s}}) & p_{\mathrm{abs}}(p_{\mathrm{d}}\sin\alpha - k_{\mathrm{sc}}) \\ p_{\mathrm{abs}}(k_{\mathrm{c}} - p_{\mathrm{d}}\cos\alpha) & -p_{\mathrm{abs}}k_{\mathrm{sc}} & -p_{\mathrm{abs}}k_{\mathrm{sc}} & 2 - p_{\mathrm{abs}}(p_{\mathrm{d}}\cos\alpha + k_{\mathrm{c}}) \end{pmatrix}, \tag{1}$$

where

$$k_{\mathrm{c}} = 1 - (1 - p_{\mathrm{d}})(\cos\alpha)^2, \qquad k_{\mathrm{s}} = 1 - (1 - p_{\mathrm{d}})(\sin\alpha)^2, \qquad k_{\mathrm{sc}} = (1 - p_{\mathrm{d}})\sin\alpha\cos\alpha, \tag{2}$$

p_{abs} is the absorption probability, p_{d} is the probability of population transfer to $|S_z = 0\rangle$, and $\alpha \in [0, \pi/2]$.

The unitary dynamics in between two consecutive pulses is instead described by the linear operator

$$\mathbf{U} = \exp\left(-i\tau\left(H \otimes \mathbb{I}_2 - \mathbb{I}_2 \otimes H^*\right)\right) \tag{3}$$

with $\hbar$ set to 1, $\mathbb{I}_2$ denoting the 2×2 identity matrix, and $H = \omega\,\sigma_z/2$ the Hamiltonian of the effective two-level system.

Considering a total number of pulses N we thus have

$$\mathrm{col}[\rho_t] = \mathbf{L}\,\mathrm{col}[\rho_0] \quad \text{with} \quad \mathbf{L} \equiv (\mathbf{SU})^N. \tag{4}$$

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For a fixed set of values of the parameters $(p_{\mathrm{abs}}, p_{\mathrm{d}}, \alpha, \tau, \omega)$, the super-operator $\mathbf{L}$ governing the open dynamics of the NV center possesses a unique steady-state ρ^* that is characterised by a non vanishing coherence in the energy basis.

The Kraus representation for $\mathbf{L}$ can be uniquely determined by diagonalizing its Choi matrix

$$\mathbf{T} \equiv \sum_{k,j=0}^{1} (E_{kj} \otimes \mathbb{I}_2)\, \mathbf{L}\, (\mathbb{I}_2 \otimes E_{kj}) = \sum_{\ell=0}^{3} \xi_\ell \mathbf{u}_\ell \mathbf{u}_\ell^\dagger \tag{5}$$

where $E_{kj} \equiv |k\rangle\langle j|$, with $|0\rangle \equiv (1,0)^T$ and $|1\rangle \equiv (0,1)^T$, and $(\xi_\ell, \mathbf{u}_\ell)$ denotes the j-th pair (eigenvalue, eigenvector) resulting from the eigenvector decomposition of $\mathbf{T}$. The Kraus operators $\{K_\ell\}$ associated with the open map $\mathbf{L}$ are thus implicitly provided by the following relation:

$$\mathrm{col}\,[K_\ell] = \sqrt{\xi_\ell}\mathbf{u}_\ell \quad \text{such that} \quad \rho(t) = \sum_{\ell=0}^{3} K_\ell\, \rho(0) K_\ell^\dagger\,. \tag{6}$$

One can easily determine that the open dynamics of the NV center is correctly described by three Kraus operators K_ℓ. These Kraus operators can be then considered for the derivation of the backward dynamics as discussed in the main text.

Supplementary Note 2

Experimental data and simulation

As described in the main text, in our experiments we measure the EPM probability $p_f^{\mathrm{fin}}(\rho) = \mathrm{tr}(\Pi_f^{\mathrm{fin}}\Phi(\rho))$ i.e., the probability of obtaining E_f^{fin} when measuring the energy of the system at the final time $t_{\mathrm{fin}} = N\tau$ (N is the number of laser pulses and τ is the time between them), assuming that the system is initialized into the state ρ. In particular, we performed four independent experiments for each of the four initial states: $|0\rangle$, $|1\rangle$, $|+\rangle_y$, and $|-\rangle_y$. The results of such measurements are shown in Fig. 1. Notice that we only measure the probability $p_{f=1}^{\mathrm{fin}}$ associated to the final excited state $|1\rangle$. This is because, for a two level system, the remaining probability is obtained as $p_{f=0}^{\mathrm{fin}} = 1 - p_{f=1}^{\mathrm{fin}}$. As mentioned in the main text, the classical mixtures we are interested in are a convex combination of these four pure states. For example, the EPM probability associated with the initial thermal state $\rho_{\mathrm{th}}^{\mathrm{in}} = \mathrm{e}^{-\beta H_{t_{\mathrm{in}}}}/Z_{\mathrm{in}}$ is obtained as $p_f^{\mathrm{fin}}(\rho_{\mathrm{th}}^{\mathrm{in}}) = p(E_0^{\mathrm{in}})p_f^{\mathrm{fin}}(|0\rangle\langle 0|) + p(E_1^{\mathrm{in}})p_f^{\mathrm{fin}}(|1\rangle\langle 1|)$, where $p(E_i^{\mathrm{in}}) = \mathrm{e}^{-\beta E_i}/Z_{\mathrm{in}}$. Similarly, the probability $p_f^{\mathrm{fin}}(\rho_0)$ for a given initial state (see main text) $\rho_0 = p|1\rangle\langle 1| + (1-p)|+\rangle_y\langle +|$, with $p \in [0,1]$, is obtained as $p_f^{\mathrm{fin}}(\rho_0) = p\, p_f^{\mathrm{fin}}(|1\rangle\langle 1|) + (1-p)p_f^{\mathrm{fin}}(|+\rangle_y\langle +|)$.

In Supplementary Figure 1 we compare the experimental data with the numerical simulation of the dynamics, using the model described in the previous section. The values of the parameters p_{abs}, and p_{d} are selected by minimizing the sum of the squares of the residuals between data and simulation.

In Fig. 2 of the main text we show the results of the irreversible entropy production $\Delta\Sigma$ obtained for an initial state $|+\rangle_y$. In Supplementary Figure 2 we present similar results but for the initial state $|-\rangle_y = (|0\rangle - \mathrm{i}\,|1\rangle)/\sqrt{2}$.

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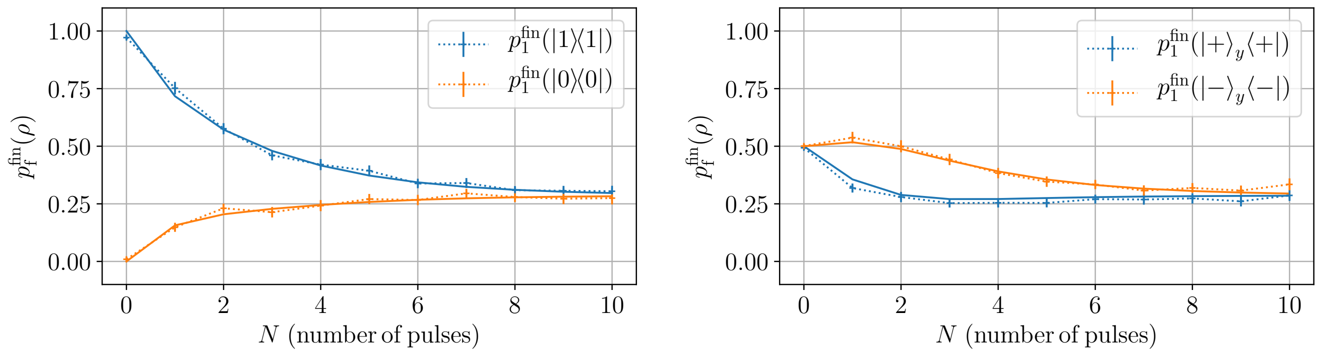
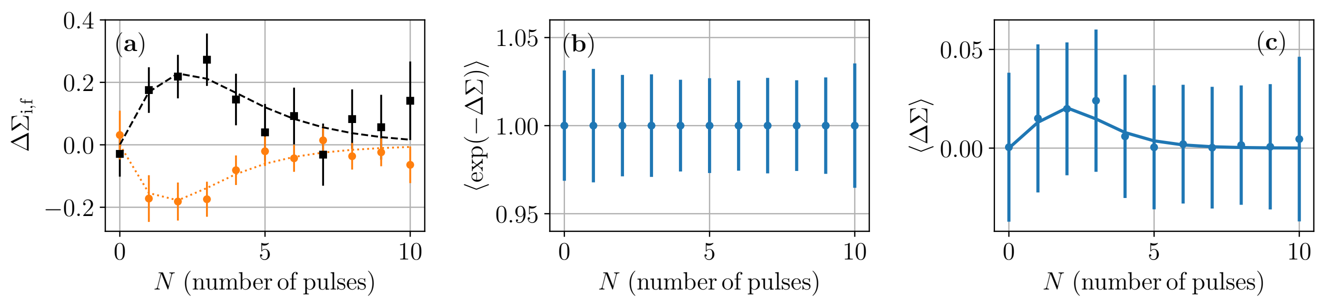


Supplementary Figure 1. **EPM final probabilities.** Measurements of the EPM protocol as a function of the number N of pulses for the initial pure states $\{|0\rangle, |1\rangle, |+\rangle_y, |-\rangle_y\}$. The crosses with error bars are the experimental data (dotted line is a guide to the eye). Solid lines represent the numerical simulation of the dynamics described in the previous section, using the parameters $p_{\mathrm{abs}} = 0.700$ and $p_{\mathrm{d}} = 0.255$. As mentioned in the main text, the other parameters are $\alpha = \pi/4$ (i.e., $\delta = -\Omega$), $\tau\omega \simeq (2\pi)0.9$, and $\tau = 190$ ns. The error bars correspond to the standard deviation of the normalized photo-luminescence (see Methods in the main text).



Supplementary Figure 2. **Coherence-affected entropy production.** Panel **(a)**: Experimental values (markers with error bars) of the irreversible entropy production. Black squares are $\Delta\Sigma_{1,1} = \Delta\Sigma_{0,1}$, and orange bullets are $\Delta\Sigma_{1,0} = \Delta\Sigma_{0,0}$. Dashed black line and dotted orange line are the corresponding numerical simulations. Panel **(b)**: Experimental verification of the fluctuation theorem $\langle \mathrm{e}^{-\Delta\Sigma} \rangle_{\Gamma} = 1$ [Eq. (6) of the main text], for the coherence-affected irreversible entropy production, as a function of the number N of pulses used to drive the dynamics of the NV center. Panel **(c)**: Average experimental coherence-affected entropy production as a function of N (blue circles). In all panels, the experimental data are plotted against the predictions (from the numerical simulations) that are obtained by taking $|-\rangle_y = (|0\rangle - \mathrm{i}\,|1\rangle)/\sqrt{2}$ as the initial quantum state.