Experimental assessment of non-classicality in a solid-state spin qutrit

Santiago Hernández-Gómez, Stefano Gherardini, Matteo Lostaglio, Amikam Levy, and Nicole Fabbri

1European Laboratory for Non-linear Spectroscopy (LENS), Università di Firenze, I-50019 Sesto Fiorentino, Italy
2Dipartimento di Fisica e Astronomia, Università di Firenze, I-50019, Sesto Fiorentino, Italy
3Istituto Nazionale di Ottica del Consiglio Nazionale delle Ricerche (CNR-INO), I-50019 Sesto Fiorentino, Italy
4Istituto Nazionale di Ottica del Consiglio Nazionale delle Ricerche (CNR-INO), Area Science Park, Basovizza, I-34149 Trieste, Italy
5Institut für Theoretische Physik, Eberhard-Karls-Universität Tübingen, 72076 Tübingen, Germany
6Centre for Theoretical Atomic, Molecular and Optical Physics, School of Mathematics and Physics, Queen’s University Belfast, Belfast BT7 1NN, United Kingdom
7Korteweg-de Vries Institute for Mathematics and QuSoft, University of Amsterdam, The Netherlands
8Department of Chemistry, Bar-Ilan University, Ramat-Gan 52900, Israel

The incompatibility of physical observables is one of the hallmarks of quantum mechanics. This form of non-classicality, encapsulated by negative quasiprobabilities, has been shown to underlie metrological and thermodynamical advantages and it can be related with information scrambling and dynamical phase transitions in many-body systems. In this work, we use a nitrogen-vacancy center in diamond to realize the experimental implementation of a weak two-point measurement scheme to reconstruct the Margenau-Hill quasiprobability distribution, whose negativity implies non-classicality. Finally, we experimentally show work extraction, empowered by non-classicality, beyond the classical case.

It is general folklore that one cannot define (linear) joint probability distributions for non-commuting measurements [1, 2], position and momentum being the most well-known example. It follows as a corollary that there is not a unique way to describe multi-time fluctuations of quantum mechanical quantities, since in general even a single observable not always commute with itself at different times.

Nevertheless, across the quantum sciences, we often deal with fluctuations of quantum-mechanical quantities, especially energy. We treat them by defining correlation functions between events at different times. Quantum mechanically, correlations between two observables at different times are characterized in terms of quantities resembling joint probability distributions for the eigenvalues of the observables. While these quantities are normalized to one and furnish the correct marginal probabilities, describing the statistics of measurement outcomes at single times, they are in general neither real nor positive. Objects with these properties are known as quasiprobabilities, akin to the well-known Wigner function in quantum optics [3]. Particularly relevant in the context of quantum correlations between observables is the Kirkwood-Dirac quasiprobability (KDQ) [4–6].

Introduced as a characterization of the quantum state for discrete quantum systems [4, 5], the KDQ and its real part, the Margenau-Hill quasiprobability (MHQ), have been recently brought back into the spotlight by a series of papers showing their ubiquity in quantum physics. As we survey in [7], these quasiprobabilities underpin analyses of perturbation theory [8–10], quantum currents [11], dynamical phase transitions [12, 13], quantum information scrambling beyond out-of-time ordered correlators [14, 15], non-classical heat-flows between locally-thermal systems [16], fluctuation theorems [17, 18] and more. KDQ and MHQ are also closely related to the concept of weak values, and they have been used in tomographic reconstructions of the quantum density matrix in optical experiments [19, 20]. Furthermore, they also have applications both in the foundations of quantum mechanics [21, 22] –where nonzero negative or imaginary parts are associated to proofs of contextuality [23, 24]– and in quantum metrology leading to advantages in local and postselected setups [25–27].

To our knowledge, experimental access to quasiprobabilities has been limited to weak measurement schemes [20, 28–33] that use an ancillary system. Their main drawback is thus to require the engineering of system-ancilla couplings, which may change depending on the quasiprobability one aims to measure and are usually platform-dependent. It is thus desirable to have the ability to implement an “optimal” measurement strategy that only requires to perform projective measurements, thus without making use of any ancilla.

In this letter we report the experimental implementation of a weak two-point measurement (wTPM) scheme [7, 34] in a solid-state spin qutrit at room temperature. The scheme allows to directly access the MHQ encoding the statistics of energy fluctuations via projective measurements only. By employing this measurement scheme, we show how the driven unitary dynamics allows for the experimental detection of non-classicality in the form of negativity of the MHQ work distribution. Then, by studying the work fluctuations in our closed three-level system, based on the spin qutrit of a nitrogen-vacancy (NV) center in diamond, we observe
peaks of work extraction up to five times those of the standard, and classical, two-point measurement (TPM) scheme. We trace back these enhanced work extraction to the MHQ negativity and thus show how the latter can be exploited as quantum resource for work extraction.

Non-classicality.—The KDQ encodes temporal correlations between quantum observables. Given two observables $A(0) \equiv \sum a_i \Pi_i(0)$ and $B(t) \equiv \sum f_j \Xi_f(t)$ in terms of their eigenvalues $a_i$ and their eigenprojectors $\Pi_i(0)$ and a quantum channel $\mathcal{E}$ describing the system dynamics in the time interval $[0, t]$, the KDQ reads as

$$q_{i_f}(\rho) = \text{Tr} \left[ \rho \Pi_i(0) \mathcal{E}^\dagger(\Xi_f(t)) \right], \quad (1)$$

where $\mathcal{E}^\dagger$ denotes the adjoint of $\mathcal{E}$ and $\rho$ is the quantum state at $t = 0$. In the remits of our experiments, we consider a quantum system that evolves unitarily, so $\mathcal{E}(\cdot) \equiv U(\cdot) \equiv U(\cdot)U^\dagger$ with $U$ a unit operator.

The non-classicality of the KDQ is defined via [6, 35]

$$\mathcal{R}(\mathbf{q}(\rho)) \equiv -1 + \sum_{i,f} |q_{i_f}(\rho)|, \quad (2)$$

where $\mathbf{q}(\rho)$ denotes the KDQ vector containing all the elements of $\{q_{i_f}(\rho)\}$ [7]. For the MHQ distribution, the relevant measure of non-classicality is the negativity. Note that, if any pairs among $\rho$, $\Pi_i$ and $\mathcal{E}^\dagger(\Xi_f)$ are mutually commuting, then Eq. (1) reduces to the probability of observing outcomes $i$ followed by $f$ in a sequential projective measurement of the observables $A(0)$ and $B(t)$. This is also returned by the TPM scheme [36]. In such a case, negative/complex values disappear and a classical stochastic interpretation is possible. It is also important to stress that, in general, non-commutativity is not a sufficient condition for non-classicality, as discussed in [6].

Negativity, heralding non-classicality, is responsible for advantages in quantum metrology and thermodynamics [16, 25–27]. It is thus crucial to identify suitable strategies for its experimental assessment. The latter, however, is not a trivial task. In fact, non-classicality can be diluted under coarse-graining, which makes the initial state commuting with the initial measurement operator [16, 37]. In these classical limits, the KDQ is a positive probability distribution.

Measurement scheme.—In this letter we focus on measuring the MHQ (the real part of KDQ) for work fluctuations during a time-dependent unitary process. We present an experimental implementation of the wTPM measurement scheme [34], which consists of a non-selective 2-outcome projective measurement that checks whether the initial energy is $E_i$ or NOT $E_i$ followed by a projective measurement of the final Hamiltonian. The wTPM joint probabilities then reads

$$p_{i_f}^{wTPM} \equiv \text{Tr} \left[ \mathcal{U}(\rho_{NS,i})\Xi_f(t) \right], \quad (3)$$

where $\rho_{NS,i} = p_i \rho_i + (1 - p_i)\overline{\rho}_i$, $\rho_i = \Pi_i \rho \Pi_i / p_i$, $p_i = \text{Tr} [\rho \Pi_i]$ and $\overline{\rho}_i = (1 - \Pi_i) \rho (\mathbb{I} - \Pi_i) / (1 - p_i)$. Here, NS stands for “non-selective”, and the state $\rho_{NS,i}$ can be obtained by performing non-selective projective measurements with projectors $\Pi_i$ and $\mathbb{I} - \Pi_i$ or, equivalently, by the preparation of the states $\rho_i$ and $\overline{\rho}_i$ with the corresponding probabilities.

This joint probability is related to the MHQ by [7, 34]

$$\text{Re} q_{i_f}^{wTPM} = p_{i_f} - \frac{1}{2} \left( p_{i_f}^{wTPM} - p_f^{END} \right), \quad (4)$$

i.e., the real part of the KDQ is given by three distinct contributions [38] that stem from applying in three separate sets of runs the wTPM protocol, the TPM scheme ($p_{i_f}^{TPM} = p_i \text{Tr} \left[ \mathcal{U}(\rho_i)\Xi_f(t) \right]$), and a single final measurement of $H(t)$ ($p_f^{END} \equiv \text{Tr} \left[ \mathcal{U}(\rho)\Xi_f(t) \right]$) where END stands for “end-time energy measurement” [39].

Experimental setting.—In order to experimentally witness non-classicality in the MHQ work distribution, let us consider a quantum three-level system evolving under a time varying Hamiltonian.

To achieve this, we use as quantum system the electronic spin of an NV center in bulk diamond at room temperature. NV centers are defects in a diamond lattice with an orbital ground state that is a spin triplet $S = 1$. The degeneracy in $m_S$ is lifted due to the zero field splitting and to the presence of an external bias field aligned with the spin quantization axis. The NV spin qutrit can be optically initialized into $m_S = 0 \,(|0\rangle)$ by illuminating the defect with a green laser [40]. Moreover, the spin state can be readout by detecting the photoluminescence (PL) after the laser illumination, as the PL intensity depends on the spin projection [41, 42]. In addition, on-resonance microwave fields are used to coherently drive the spin, with coherence times up to milliseconds (at room temperature) [43, 44]. By virtue of these properties, NV centers are broadly used for quantum technologies, such as quantum sensing [45–47], quantum information [48, 49] and, recently, for quantum thermodynamics [50–52].

A time varying Hamiltonian is implemented by coherently driving the NV spin with a microwave field with phase changing in time. More specifically, the spin qutrit is driven by a bi-chromatic microwave field on-resonance with both the transitions $|0\rangle \leftrightarrow |+1\rangle$ and $|0\rangle \leftrightarrow |-1\rangle$. In the microwave rotating frame, the Hamiltonian of the system (after the rotating wave approximation) is

$$H(t) = \Omega_1 \left( S_{z1} \cos \phi_1 t + S_{y1} \sin \phi_1 t \right) + \Omega_2 \left( S_{z2} \cos \phi_2 t - S_{y2} \sin \phi_2 t \right), \quad (5)$$

where $h = 1$, $S_{\alpha}$ are the spin operators defined in terms of the Gell-Mann matrices, as detailed in [53], and $\Omega_1$ and $\phi_1$ ($\Omega_2$ and $\phi_2$) are the Rabi frequency and the phase of the driving field for the transition $|0\rangle \leftrightarrow |+1\rangle$ ($|0\rangle \leftrightarrow |-1\rangle$). See also [53] for more details on the energy level structure and the driving fields. To simplify the measurements in the time-varying energy eigenbasis, we remove
The time dependency on one of the Hamiltonian eigenstates by setting $\Omega_1 = \Omega_2 = \Omega$ and $\phi_1 = \phi_2 = \phi$. In the case of $\phi_1 = \phi_2 = \phi$, the interaction between the NV center and the two microwave fields corresponds to a Stimulated Raman Adiabatic Passage (STIRAP) in the two-photon resonance condition [54]. The eigenstates of the Hamiltonian (5) are: $|E_{\pm}(t)\rangle = \frac{1}{\sqrt{2}}(|1\rangle \pm e^{2\pi i\phi} |0\rangle)$, and $|E_0(t)\rangle = \frac{1}{\sqrt{2}} (|1\rangle - |0\rangle)$, with eigenvalues $E_{\pm} = \pm \Omega$ and $E_0 = 0$. Thus, the projectors of the Hamiltonian are $\Xi_k(t) = |E_k(t)\rangle\langle E_k(t)|$ and $\Pi_k(0) = \Xi_k(0)$ for $k = +, 0, -$. Note that $[H(0), H(t)] \neq 0$ for $t\phi/\pi \not\in \mathbb{Z}$. Hence, the system energy changes during the unitary evolution under $H(t)$, meaning that work is exchanged between the NV spin and the microwave field.

**Measuring work quasiprobabilities.**— Assuming that the initial state $\rho$ is a pure state, one can reconstruct the MHQ work distribution by measuring a set of conditional probabilities of the form

$$p(f|\psi) \equiv \text{Tr} \left[ \mathcal{U}(|\psi\rangle\langle\psi|)\Xi_f(t) \right].$$

Eq. (6) denotes the probabilities to obtain $E_f(t)$ from measuring $H(t)$ at time $t$, conditioned on having initialized the NV spin qutrit into a given pure state $|\psi\rangle$. The specific pure state $|\psi\rangle$ in which the system is initialized depends on the scheme that we want to implement (EPM, TPM, or wTPM), as detailed below. With our experimental setup, we can measure $p(f|\psi)$ following the protocol described in Fig. 1. In this protocol, the qutrit is prepared into a pure state $|\psi\rangle$. Then, it evolves under the time-varying Hamiltonian $H(t)$ in the time interval $[0, t]$. At the end of the protocol, in order to readout $p(f|\psi)$, we apply to the quantum system a microwave gate $\mathcal{R}_f$, such that $\mathcal{R}_f(\Xi_f) = |00\rangle\langle00|$, and then we measure the PL intensity. As mentioned before, the PL value depends on the spin projection, specifically, it indicates the probability that the qutrit is in $|0\rangle$: $\text{Tr} \left[ \mathcal{R}_f(\mathcal{U}(|\psi\rangle\langle\psi|))|00\rangle\langle00| \right] = \text{Tr}\left[\mathcal{U}(|\psi\rangle\langle\psi|)\mathcal{R}_f^{-1}(|00\rangle\langle00|)\right] = p(f|\psi)$. Note that, since the eigenstates of the Hamiltonian change in time, the gate $\mathcal{R}_f$ depends on the final time $t$. For a given initial state $\rho$ we perform independent experiments for several values of $t$ and for each of the three Hamiltonian projectors $\Xi_f(t)$.

Due to the low photon collection efficacy and shot noise of the detector, each of these experiments is repeated around $10^6$ times to obtain the average PL value and subsequently $p(f|\psi)$.

Given the initial and final non-commuting observables, it is always possible to find an initial state $\rho$ such that the corresponding MHQ distribution entails non-classicality [7]. We follow this logic to find an initial pure state to witness non-classicality when the MHQ work distribution is measured. In particular, for the Hamiltonian (5) with $\Omega \simeq (2\pi)2.2$ MHz and $\phi \simeq 1.09 \Omega$, we set the initial pure state $\rho = |\xi\rangle\langle\xi|$ that approximately maximizes the negativity [55], where $|\xi\rangle = \sum_i \sqrt{p_i} e^{2\pi ja_i} |E_i(0)\rangle$, with $j^2 = -1$, $p_i =
the classical limit (dotted line) and entering into the non-classicality region (blue area). This region is bounded from above by the negativity is obtained as

\[
\sum_i |\text{Re} q_{if}|. \quad \text{In (a), (b) the work MHQs are always positive, while in (c) there is a case with negative values, serving as a witness of non-classicality. This negative values of the MHQ imply that } \sum_f |\text{Re} q_{if}| \text{ is not constant in time (in contrast with the marginal values } \sum_i |\text{Re} q_{if}| \approx 0.23, \text{ and will lead to non-classicality values different from zero.} \]

(d) Experimental measurements (green circles) of the negativity [Eq. (2)] as a function of time, where the solid green line represents the simulated data. For almost all the interaction time the negativity is larger than zero, hence overcoming the classical limit (dotted line) and entering into the non-classicality region (blue area). This region is bounded from above by

\[
\sqrt{d} - 1 \quad (\text{dashed line}, \text{where } d = 3 \text{ is the dimension of the system’s Hilbert space})
\]

For the measurement of \( P_i^{\text{END}} \), we initialize the system in \( \rho \), let it evolve unitarily as described above and then perform an energy projective measurement at time \( t \). The results are shown in Fig. 2c. In contrast, to reconstruct \( \rho_{\text{TPM}}^{f} \) and \( \rho_{\text{wTPM}}^{f} \), the conditional probabilities \( p(f|i) \) and \( p(f|\bar{i}) \) are measured, by initializing in the quantum states \( \rho_i \) and \( \bar{\rho}_i \) respectively. Notice that, since \( \rho \) is a pure state, then \( \bar{\rho}_i \) is also a pure state for any \( i \).

We plot the results obtained from these measurements in Fig. 2d,e. The TPM probabilities are thus computed by combining the measured conditional probabilities for the initial states \( \rho_i \) with the initial probabilities \( p_i \equiv \text{Tr}[\rho_i \Pi_i] \), such that, as usual, \( \rho_{\text{TPM}}^{f} = p_i p(f|i) \). Similarly, we combine the measured \( p(f|i) \) and \( p(f|\bar{i}) \), so as to obtain the wTPM probabilities \( \rho_{\text{wTPM}}^{f} = p_i p(f|i) + (1 - p_i) p(f|\bar{i}) \).

We can now obtain the MHQ work distributions by using all the previous measurements as dictated by Eq. (4). The results are shown in Fig. 3a-c. The only work MHQ showing negative values is \( \text{Re} q_{if} \) with \( f' = - \) and \( f = + \), i.e., the quasiprobability for the qutrit to move from the lowest energy state \( |E_-(0)\rangle \) to the highest energy state \( |E_+(t)\rangle \). From Eq. (2) we can then determine the non-classicality quantified by the negativity of the measured work distribution. As one can observe in Fig. 3a,b (see black dashed line), all the MHQs with \( i \neq - \) sum up to a constant positive value that corresponds to \( s = \sum_f |\text{Re} q_{if} | + \text{Re} q_{0f} = 0.770 \pm 0.021 \). Accordingly, the negativity is obtained as

\[
\mathcal{N}(q(\rho)) = -1 + s + \sum_f |q_{-f}|,
\]

and its experimental values are shown in Fig. 3d.

**Work extraction.**— In Fig. 4, we compare the experimental data for the average work values

\[
\langle W \rangle_{\text{TPM}} = \sum_{i,f} \text{Re} q_{if} (E_f - E_i) \quad \text{and} \quad \langle W \rangle = \sum_{i,f} \text{Re} q_{if} (E_f - E_i). \]

Note that the latter coincides with the standard definition of average work that is returned by applying the unitary generated by \( H(t) \).

In our experiments the peaks in average extracted work coincide with the peaks in negativity. The reason is that under our experimental conditions (\( \phi_1 = \phi_2 = \phi \)), all the negativity is associated to the quasiprobability of the largest exciting transition, \( \Delta E_{-+} = 2\Omega \). Classically this transition contributes to the work done on the system but, due to negativity, it instead contributes to work extracted, thus leading to the observed peak. Numerical simulations [53] show that our experimental conditions are both close to maximizing the negativity of \( \text{Re} q_{-+} \) as well as maximizing the extracted work. The upshot is that thermodynamic transformations aiming at maximizing the extracted work should target maximizing the negativity of the most exciting transitions, which is distinct from maximizing \( \mathcal{N}(q(\rho)) \) itself.

**Conclusions.**— We have experimentally realized the wTPM measurement scheme [7, 34] and reconstructed the Margenau-Hill quasiprobability work distribution for a coherently driven spin qutrit. The strategy used to obtain the MHQ requires a combination of three independent measurement schemes that we implement in an NV center in diamond driven by a microwave field acting as a work reservoir. We experimentally witness non-classicality in the process and we relate the specific negativity distribution realized in the experiment to peaks in extracted work. This shows that the negativity of the work quasiprobability distribution can be effectively used as a quantum resource in the task of work extraction.
Our endeavour indicates that quantum effects and non-classicality in the work distribution and beyond (see [7]) can be experimentally investigated through quasiprobabilities using a scheme with similar resource requirements as the TPM. Accordingly, our work represents a step forward in the direction of experimentally investigating genuine non-classical phenomena in a variety of platforms, in particular those in which TPM measurements have been already performed.

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* hernandez@lens.unifi.it
† lostaglio@protonmail.com

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Supplemental Material: Experimental assessment of non-classicality in a solid-state spin qutrit

DETAILS ON THE EXPERIMENTAL SETUP

As described in the main text, the three level system realized for our experiments is based on the spin triplet \( S = 1 \) of the orbital ground state of an NV center, with Hamiltonian

\[
\mathcal{H}_{NV} = \Delta S_z^2 + \gamma_e B S_z ,
\]

(S1)

where \( \Delta = 2.87 \text{GHz} \) is the zero-field-splitting, \( \gamma_e \) denotes the electron gyromagnetic ratio, and \( B \) is a bias magnetic field aligned with the NV quantization axis \( z \) (determined by the orientation of the NV defect in the diamond lattice).

The spin triplet is driven by two continuous on-resonance microwave (MW) fields addressing the transitions \( |0\rangle \leftrightarrow |+1\rangle \) and \( |0\rangle \leftrightarrow |-1\rangle \). Hence, overall the spin dynamics can be described by the Hamiltonian

\[
\mathcal{H}(t) = \mathcal{H}_{NV} + (\Omega_1 \cos(\omega_1 t + \varphi_1(t))|+1\rangle\langle 0| + \Omega_2 \cos(\omega_2 t + \varphi_2(t))|0\rangle\langle -1|) + \text{h.c.},
\]

(S2)

where \( \Omega_1 \) and \( \Omega_2 \) are the Rabi frequencies for the transitions \( |0\rangle \leftrightarrow |+1\rangle \) and \( |0\rangle \leftrightarrow |-1\rangle \), respectively. In addition, \( \omega_1 = \Delta \pm \gamma_e B \) denote the frequencies, and \( \varphi_1(t) \) and \( \varphi_2(t) \) are the time varying phases of the MW fields. The energy level structure of the qutrit and its interaction with the MW fields is depicted in Fig. S1(a).

In the microwave rotating frame (defined by the unitary transformation \( V = \exp(jt(\omega_1|+1\rangle\langle 1| + \omega_2|-1\rangle\langle -1|)) \)), with \( j^2 = -1 \) and after applying the rotating wave approximation, the Hamiltonian \( \mathcal{H}(t) \) reads as

\[
H(t) = \Omega_1 \left(S_{z1} \cos \varphi_1(t) + S_{y1} \sin \varphi_1(t) \right) + \Omega_2 \left(S_{x2} \cos \varphi_2(t) + S_{y2} \sin \varphi_2(t) \right).
\]

(S3)

The Hamiltonian (S3) is defined in terms of the spin operators \( S_{z1} = \frac{1}{\sqrt{2}} \lambda_1, S_{y1} = \frac{1}{\sqrt{2}} \lambda_2, S_{x2} = \frac{1}{\sqrt{2}} \lambda_6, S_{y2} = \frac{1}{\sqrt{2}} \lambda_7 \), where \( \lambda_i \) are the Gell-Mann matrices:

\[
\lambda_1 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_2 = \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \lambda_6 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \quad \lambda_7 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}.
\]

(S4)

In our experiments, we select the time varying phases so that they change linearly in time, i.e., \( \varphi_1(t) = \phi_1 t \) and \( \varphi_2(t) = -\phi_2 t \). Therefore, Eq. (5) in the main text is recovered from Eq. (S3). The energy level structure in the MW rotating frame is sketched in Fig. S1(b).

Finally, let us note that we can describe the system dynamics in a different rotating frame for which the Hamiltonian is time independent. In fact, instead of the MW rotating frame, we can express Eq. (S2) by transforming it in the rotating frame determined by the unitary transformation \( V = \exp(jt(\omega_1|+1\rangle\langle 1| + (\omega_1 + \omega_2)|-1\rangle\langle -1|)) \). In this new frame and after the rotating wave approximation, the time independent Hamiltonian is:

\[
\mathcal{H} = \Omega_1 S_{z1} - \phi_1 S_{z1} + \Omega_2 S_{x2} + \phi_2 S_{x2} ,
\]

(S5)

where \( S_{z1} = |+1\rangle\langle +1| \) and \( S_{x2} = |-1\rangle\langle -1| \). The diagram of the energy level structure for this new rotating frame is shown in Fig. S1(c). Observe that this diagram represents a Stimulated Raman Adiabatic Passage (STIRAP) experiment for a three level ladder scheme [S54].

FIG. S1. Energy diagram for the NV center spin triplet. (a) In the laboratory frame according to the Hamiltonian \( \mathcal{H}(t) \) in Eq. (S2). (b) In the MW rotating frame, as described by the Hamiltonian \( H(t) \) in Eq (S3) (or Eq. (5) in the main text). (c) In a rotating frame where the Hamiltonian is effectively time-independent, as in Eq (S5). It is worth noting that in the frame (c), the time-varying phase of each MW field can be reinterpreted as detuning.
ROLE OF NON-CLASSICALITY FOR ENHANCED EXTRACTABLE WORK

In Fig. (4) in the main text we show the comparison between the work values $\langle W \rangle$ associated with the MHQ distribution and the average $\langle W \rangle_{\text{TPM}}$ corresponding to the TPM scheme. In doing this, we also make use of the experimental data obtained from the implementation of the weak two-point measurement (wTPM) scheme.

From the figure, we can observe that the presence of non-classicality, in the form of negativity of the MHQs, entails a larger extractable work that is maximum when also $\mathcal{R}(\rho)$ takes its maximum value. The physical explanation of this effects relies in the fact that non-classicality is able to transform the average work done by the system into extractable work, and vice versa. Let us analyze this statement more in detail. For the sake of simplicity, from here on we call $\text{Re} q_{ij}$ as $z_{ij}$. The average work $\langle W \rangle \equiv \sum_{i,f} z_{ij} (E_f - E_i)$ of a MHQ work distribution can be equivalently written as

$$
\langle W \rangle = \sum_{i,f} \mu_{ij} ||z|| \text{sgn}(z_{ij}) (E_f(t) - E_i(0)),
$$

(S6)

where the set of $\mu_{ij} \equiv |z_{ij}|/||z||$ forms a classical probability distribution respecting the Kolmogorov’s axioms of probability theory, $||z|| \equiv \sum_{i,f} |z_{ij}|$ that is denoted as total negativity ($||z|| = 1$ if no MHQs are negative), and $\text{sgn}(\cdot)$ is the sign function that is equal to $+1$ if $(\cdot)$ is positive and $-1$ otherwise. Hence, by defining $\overline{E}_f(t) \equiv ||z|| \text{sgn}(z_{ij}) E_f(t)$ and $\overline{E}_i(0) \equiv ||z|| \text{sgn}(z_{ij}) E_i(0)$, we can interpret the average MHQ work $\langle W \rangle$ by means of a classical stochastic process defined by the set of probabilities $\{\mu_{ij}\}$, i.e.,

$$
\langle W \rangle = \sum_{i,f} \mu_{ij} \left(\overline{E}_f(t) - \overline{E}_i(0)\right).
$$

(S7)

One can thus compare Eq. (S7) with the average TPM work $\langle W \rangle_{\text{TPM}}$, and then explain why non-classicality (negativity of MHQs in our experimental case-study) can entail a larger amount of extractable work. In fact, when negativity of the single term $q_{ij}$ is present, positive work terms $E_f(t) - E_i(0) \geq 0$ (corresponding to work done by the system) changes sign and they transforms in $\overline{E}_f(t) - \overline{E}_i(0) < 0$, i.e., work that can be extracted from the system. Moreover, always in case of negativity, the effective energies $\overline{E}$ are obtained by multiplying $E$ for $||z|| \geq 1$. Thus, the extractable work originated by non-classicality is larger –in absolute value– than the corresponding positive work terms (done by the system) that enter the TPM work average.

MAXIMIZING THE AMOUNT OF EXTRACTABLE WORK: NUMERICAL ANALYSIS FOR GENERIC PARAMETERS OF THE NV CENTER HAMILTONIAN

As described in the main text, the interaction between a NV center with an on-resonance microwave field results in the Hamiltonian [Eq. (5)] (expressed in the MW rotating frame) that, after the rotating wave approximation, reads as

$$
H(t) = \Omega_1 (S_{x1} \cos \phi_1 t + S_{y1} \sin \phi_1 t) + \Omega_2 (S_{x2} \cos \phi_2 t - S_{y2} \sin \phi_2 t).
$$

(S8)

Here, our aim is to identify the parameters for which the negativity of $\text{Re} q_{ij}$ is minimized, the average MHQ work $\langle W \rangle$ is minimized (by including its sign), and the measure $\mathcal{R}$ of non-classicality is maximized. In order to achieve this, we run numerical simulations for 10000 different sets of random parameters $\{\Omega_1^R, \phi_1^R, \Omega_2^R, \phi_2^R\}$, such that $\{\Omega_1, \phi_1, \Omega_2, \phi_2\} = \{\Omega_1^R, \phi_1^R, \Omega_2^R, \phi_2^R\}$, and we calculate $\text{min}[\text{Re} q_{ij}]$, $\text{min}[(\langle W \rangle)]$, and $\text{max}[\mathcal{R}]$ for each set of parameters. The $\text{min}[]$ and $\text{max}[]$ are calculated over the time interval $t \in (0, 2\pi/\sqrt{2(\Omega_1^2 + \Omega_2^2) + \phi_1^2})$. The longest time value in this interval corresponds to a period of the dynamics in the case with $\phi_2 = \phi_1$. In addition, the considered random parameters correspond to random floating-point numbers in the intervals $\Omega_1^R \in [1,20]$ MHz, $\phi_1^R \in [-20\Omega_1^R, 20\Omega_1^R]$, $\Omega_2^R \in [1,20]$ MHz, and $\phi_2^R \in [-20\Omega_2^R, 20\Omega_2^R]$. For each set of parameters, the initial state is a random pure state $\rho = |\xi(\xi)|, \text{with } |\xi\rangle = a e^{i\phi_a}|1\rangle + b e^{i\phi_b}|0\rangle + \sqrt{1 - a^2 - b^2}(-1)$, where $a, b, \phi_a, \phi_b$ are random floating-point numbers in the intervals $[0,1], [0, \sqrt{1 - a^2}]$, $[0, 2\pi]$, and $[0, 2\pi]$ respectively.

The results of the numerical simulations are summarized in Fig. S2. From these results it is evident that the condition $\phi_1 = \phi_2$ allows for the minimization of the negativity of $\text{Re} q_{ij}$, as well as the minimization (with sign) of the average MHQ work $\langle W \rangle$. In contrast, in order to maximize the non-classicality measure $\mathcal{R}$, it is more convenient to select the parameters of the system Hamiltonian such that $\phi_1 \neq \phi_2$. 
FIG. S2. Results of the numerical simulations. In both panels (a)-(b), each empty blue circle represents the result for a set of random parameters of the system Hamiltonian and a random initial pure state. For each empty blue circle there are two orange crosses corresponding to the cases $\phi_1 = \phi_2 = \phi_R^1$ and $\phi_1 = \phi_2 = \phi_R^2$. Instead, the red circle with the errorbars represents the experimentally measured values, as detailed in the main text. In both panels $\omega \equiv \sqrt{\Omega_1^2 + \Omega_2^2}$. Finally, in panel (b), the horizontal line denotes the upper bound of the non-classicality measure $\mathcal{N}$. Such a bound is equal to $\sqrt{d} - 1$, where $d = 3$ is the dimension of the Hilbert space of the system [S7].