Robust determination of quantum signatures from weak measurements

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The ability to follow the dynamics of a quantum system in a quantitative manner is of key importance for quantum technology. Despite its central role, justifiable deduction of the quantum dynamics of a single quantum system in terms of a macroscopically observable remains a challenge. Here we show that the relation between the readout signal of a single electron spin and the quantum dynamics of the single nuclear spin is given by a parameter related to the measurement strength. We determine this measurement strength in independent experiments and use this value to compare our analysis of the quantum dynamics with experimental results. We prove the validity of our approach by measuring violations of the Leggett-Garg inequality.

INTRODUCTION

It is well known that properties of quantum and classical stochastic processes are fundamentally different. This is described by the non-commutativity of quantum observables, which are Hermitian operators (matrices), in contrast to random variables (measurable functions) in classical physics. It is, for example impossible to construct a joint probability distribution of two non-commuting observables [1]. However, the Wigner quasi-probability density for quantum position and momentum theoretically can take negative values. In experiments it is not trivial to isolate these negative values [2, 3], because we derive the quantum behaviour from indirect, classical observations of the underlying quantum theory. The challenge at hand is, to find robust methods to derive a quantum behaviour from those measurements. Often this requires some filtering of classic data to reveal the quantum properties of the initial process. Most tests of the quantumness of a system (like e.g., performed in Leggett-Garg Inequality (LGI) test [4] suffer from a "clumsiness loophole" [5], as this filtering process relies on ad-hoc assumptions on the function of the measurement device.

In this paper, we provide a way to isolate the quantum behaviour from a noisy measurement output on a quantum measurement in a highly accurate way. We achieve this by applying weak measurements [6] on single nuclear spins in diamond, an extremely well-controlled and understood quantum system. Our approach is based on a thorough analysis of correlation measurements. We measure those correlation functions for a quantum and separately for a classical signal. By comparing the theoretical expressions for the meter output correlator and the observable correlator, we notice that they differ from each other only by a parameter which marks the measurement strength. This allows us to construct an estimate of the correlation function of the quantum process based on the measurement outputs without introducing an empirical "measurement factor". To validate the effectiveness of our approach, we compare experiments on measuring a quantum signal with a classical signal and compare the LGI for both cases.

RESULTS

We use a single $^{13}$C nuclear spin in diamond as quantum system, probed by an NV center electron spin (see Fig. 1). The electron spin interacts weakly with the single $^{13}$C nuclear spin, through their hyperfine interaction. The electron is read out via projective optical measurements. An important ingredient to weak measurements is that the measurement strength can be varied.

To this end, the interaction between the electron and nuclear spins is controlled by a Knill - dynamical decoupling (KDD-XYn) sequence of periodically spaced $\pi$ pulses, which contains $n$ units of 20 pulses with a pulse interval $\tau$. If $\tau$ is adjusted to the effective nuclear Larmor period $\omega_L$, i.e., $\tau = \pi/\omega_L$, the system evolves under the effective Hamiltonian $H_{\text{eff}} = 2\alpha S_z I_z$, where the Larmor frequency $\omega_L = \gamma B$ is given by the static magnetic field $B \approx 0.25$ T and the nuclear gyromagnetic ratio $\gamma$. The effect of the KDD-XYn sequence is specified by a measurement strength parameter $\alpha = 2N_\text{p}A_\perp \tau$, which depends on the hyperfine interaction off-diagonal component $A_\perp$ between the $^{13}$C and the electron spin as well.

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FIG. 1. Scheme of the experiment. a) The sequential weak measurements $M_i$ are composed of a sensor initialization part, dynamical decoupling (KDD-XY-n) and readout of the sensor state using conventional optical readout of an NV center electron spin. The KDD-XYn filter function is tuned to the Larmor precession of the weakly coupled ($A_{es} \approx 100$ Hz) $^{13}$C nuclear spin, which results in an effective interaction between nuclear and electron spin. The electron spin state is read out after each interaction, which leads to extraction of information about the nuclear spin and its back-action. b) Schematic evolution trajectory of an electron spin state in case the nuclear spin is in the $I_e + I_n$ state. c) Schematic evolution of the nuclear spin during sequential measurements. Initially, the nuclear spin is in the thermally mixed state $|\rho_0 = I_e\rangle$, which is then partially polarized by measurements along the $x$ axis with magnitude $\sin \alpha$. The free precession between the measurements leads to a rotation around the $z$ axis. Subsequent measurements affect the measurement by disturbing both, the $x$ and $y$ component of the Bloch vector, conditioned on the measurement outcome (see Text).

as the number of pulses $N_p = 20n$. The initial state of the nuclear spin in our experiment is not polarized, i.e., the totally mixed state

$$\rho_I = \frac{1}{2}|0\rangle\langle 0| + \frac{1}{2}|1\rangle\langle 1| = \frac{1}{2}I^\alpha + \frac{1}{2}I^\beta = \frac{1}{2} = |I_e\rangle,$$  \tag{1}$$

where $I^\alpha = I_e + I_z$ and $I^\beta = I_e - I_z$ with $I_e = \frac{1}{2}$ being the half of the identity matrix, $S_k = I_k = \frac{1}{2}\sigma_k$, and $\sigma_k$ are Pauli matrices.

The measurement protocol is as follows. The electron spin is optically pumped (see Fig. 1a) into the state $|0\rangle = |m_S = 0\rangle$ or equivalently to $S_x + S_z$, and then rotated by a $(\pi/2)_y$ pulse around the $y$-axis to state $S_x + S_y$. Then, the interaction controlled by the KDD-XYn sequence is applied, which leads to a $(\pi/2)_x$ pulse along the $x$-axis and a final optical readout of the $S_z$ component of the NV sensor. This procedure is repeated sequentially. In between two successive measurements $M_i$ (see Fig. 1a) the target spin undergoes free precession around the $z$-axis with the Larmor frequency $\omega_L$. To obtain reliable statistical information, we conduct a sequence of experimental runs. The interaction between the $^{13}$C nuclear spin and the electron spin maps a signal proportional to the $I_z \sin \alpha$ onto the $S_y$ spin component of the NV electron spin. The final $(\pi/2)_x$ rotation imprints this signal onto the optically readable $S_z$ spin component of the NV center electron spin. We note, that although the initial state of the nuclear spin is given by equation 1, after the first measurement it is partially polarized to the state $I_e + \sin \alpha I_z$. [10]

We study the interaction process between the electron and the nuclear spin in the x-y plane, using a modification of the Baker-Campbell-Hausdorff (BCH) formula [10, 11]. Applying a sequence of weak measurements for a sufficiently small alpha [12], results in the following approximate representation for the amplitude of the $I_x$ and $I_y$ components of the density matrix $\rho_I$:

$$x_N(x_0) \approx x_0 \cos(\omega N t_f) \left(\cos \frac{\alpha}{2}\right)^{2(N-1)}$$  \tag{2}

$$y_N(x_0) \approx \sin(\omega N t_f) \left(\cos \frac{\alpha}{2}\right)^{2(N-1)}$$

Here, $x_0$ is the initial state, $N$ is the number of measurements $M_i$ over which the correlation is taken and $t_f$ is the time interval between the measurements. Note, that we obtained a damped oscillation expression, representing a free precession of the nuclear spin undergoing back-action due to weak measurements. The key in our approach is the calculation of the correlation function from consecutive measurements of $x_N$ and $y_N$ [10]. Taking $I_x$ as key quantity to be measured, we calculate the joint probabilities and the correlation function as:

$$C_{I_x}(N) = \cos(\omega N t_f) \left(\cos \frac{\alpha}{2}\right)^{2(N-1)}.$$  \tag{3}$$

Further, considering the component $S_z$ of the NV electron spin as observable in our experiment, we calculate the autocorrelation of the output process as:

$$C_{S_z}(N) = \sin^2 \alpha \cos(\omega N t_f) \left(\cos \frac{\alpha}{2}\right)^{2(N-1)}.$$  \tag{4}$$

Comparing Eqs. 2 and 4, we find that to restore the correlation function of the quantum bit $C_{I_x}$, it is only necessary to calibrate the correlation function of measurements on the electron spin by $\sin^2 \alpha$.

To test our approach, we first perform sequential measurements on the NV center electron spin influenced by a
FIG. 2. Calibration of fluorescence readout and externally applied classical field. 

a) Experimental protocol for modulation assisted method for determining the fluorescence response of the NV spin readout \( n_a \) and \( n_b \). Sequential measurements with optical readout of the electron spin yields phase information obtained by the interaction with the external signal. The readout \( \pi/2 \) pulse phase is sinusoidally modulated with an amplitude of \( \pi/2 \) and a period of 8 measurement cycles. 

b) The empirically calculated correlation of the centred photon counts numbers trace. The beating in the correlation originates from the presence of two frequencies (see text). The size of the beating is determined by the relative amplitude of the external signal to phase modulation. The solid curve is the best fit of the analytical model of the correlation function which includes phase modulation and the unknown external signal. 

c) Measurement protocol for the estimation of the classical signal correlation function. 

d) Reconstructed correlation function of the classical sinusoidal signal with a stochastic phase

classical external oscillating (linearly polarized) magnetic field \( B_{ac}(t) = B_{ac} \sin(\omega_{ac} t + \phi) \), where \( B_{ac} \) is the amplitude of the magnetic field \( \omega_{ac} = 2\pi f_{ac} \) is the frequency and \( \phi \) is an initial phase. This classical oscillating field mimics the interaction between the nuclear spin and the electron spin which is oscillating due to the Larmor precession of the nucleus. We conduct a sequence of experimental runs \( M_i \) (see Fig. 2a). The time interval between the \( M_i \) is not precisely controlled, so that we can assume that each run corresponds to a different realization of a random phase \( \phi \) between the oscillating field and the measurement. Analysing the sequence of experimental runs as a set of samples of a random process, 

\[
x(t) = \sin(\omega t + \phi),
\]

we calculate the empirical correlation of the output signal with a uniformly distributed \( \phi \) as 

\[
C(\tau) = \frac{1}{2} \cos(\omega \tau).
\]

Further analysis of the experiment with a classical random magnetic field basically follows the approach described above [10]. In this experiment, the effective Hamiltonian is given by 

\[
H_{\text{eff}} = 2\alpha S_z \sin(\omega t + \phi).
\]

We can interpret this Hamiltonian as describing the interaction of the NV sensor with a random sinusoidal signal eq. 5 with uniformly distributed \( \phi \) where \( \alpha = B_z N_p \tau / \pi \) determines the strength of the interaction. Using our approach, we find, that the correlation function of the
output process $z(t)$ has the following form:

$$\langle z(t)z(t+\tau) \rangle = \frac{\alpha^2}{2} \cos(\omega \tau). \tag{8}$$

Comparing expressions 6 and 8, we find that they differ only in the factor $\alpha^2$, which we must extract from a separate experiment. To infer $\alpha$ we additionally apply a phase modulation $\phi_k = \pi/2 \sin(2\pi k/8)$ to the final ($\pi/2$) pulse which modulates the output signal (see Fig. 2 a,b). The correlation of output results shows (Fig. 2.b) the modulated behaviour as a result of two signals – externally applied classical field and modulation of the final ($\pi/2$) pulse. We fit the curve with least square method using an analytical expression:
\[
\min_{n_a,n_b,\alpha} \sum_{k=1}^{200} \left( \left( \langle n_{i+k} \rangle - n_{av}^2 \right) - \frac{(n_a - n_b)^2}{4} \langle S_z(\alpha, \Phi_s) S_z^{i+k}(\alpha, \Phi_s) \rangle \right)^2
\]

(9)

where \( n_{av} = (n_a + n_b)/2 \), and

\[
S_z^i(\alpha, \Phi_s) = \sin \left( \frac{\pi}{2} \sin \left( \frac{2\pi k}{8} \right) + \alpha \cos \left( \frac{k\Phi_s \pi}{4} \right) \right)
\]

(10)

In result we extract both the fluorescence responses of the NV center in \( m_s = 0 \) \((n_a)\), \( m_s = -1 \) \((n_b)\) and the local strength of the RF field (for details see SM). Further we reconstruct the normalized correlation function of the classical signal depicted in Fig. 2d. using [9]

\[
\langle zz \rangle_{emp} = 4 \frac{(nn)n_{av}^2}{(n_a - n_b)^2}
\]

(11)

and Eq. 6,8. We note that the reconstructed correlation function of the classical signal equals the analytically calculated function.

We now turn towards the analysis of the quantum signal case where we probe the dynamics of the single \(^{13}\)C nuclear spin. In order to obtain reliable results, we choose a statistically uniform data set (see SM). Furthermore, we calibrate the fluorescence output \( n_a \) and \( n_b \) of the NV center for electron spin in the bright \( m_s = 0 \) and dark \( m_s = -1 \) state. With a detuned filter function of the KDD-XYn from the \(^{13}\)C Larmor frequency, we apply a phase modulation to the final \((\pi/2)\) pulse (see Fig. 3a), similar to how it was done for the classical signal. In a series averaged output, we obtain an oscillating signal, which is fitted with

\[
n(k) = \frac{1}{2} (n_a + n_b) + \frac{1}{2} (n_a - n_b) \sin \left( \frac{\pi}{2} \sin \left( \frac{2\pi k}{8} \right) \right),
\]

(12)

dependent only on \( n_a \) and \( n_b \) (see Fig. 3b). We estimated the empirical photon output correlation function

\[
C_n(N) = \langle n_i n_{i+N} \rangle = \frac{1}{E-N} \sum_i n_i n_{i+N}
\]

and the empirical electron spin correlator

\[
C_S_z(N) = \langle S_z(t) S_z(t + t_f) \rangle = \frac{4(C_n(N) - n_{av}^2)}{(n_a - n_b)^2}
\]

(13)

using the obtained \( n_a \) and \( n_b \) from Fig 3b. We evaluate the parameters \( \alpha \) using the standard least squares method \((\hat{\alpha} = \omega t_f)\) by comparing, the empirically estimated \( C_{S_z}(N) \) to the analytical one (eq. 2). In this way we find an estimate of \( \alpha \) with which eq. 2 approximates the reconstructed correlation function of the output signal in an optimal way (see Fig. 3d). After carefully measuring the calibration constant \( \alpha \) we normalized the empirical correlation function \( C_{S_z}(N) \) by \( \sin^2 \alpha \) and get an estimation of \( C_{L_z}(N) \) (see Fig. 3e). Finally, we apply our reconstructed correlation functions to the analysis of the LGI [4, 5, 13, 14]. To do this, we use as inputs the reconstructed correlation functions of classical (Fig. 2e) and quantum signal (Fig. 3e) and apply the LGI [5, 13]:

\[
LG(\tau) = 2C(\tau) - C(2\tau)
\]

(14)

where for the \( C(\tau) \) the reconstructed correlation function of the classical field \( C_B(N) \) and the reconstructed function of \( C_{L_z}(N) \) are used. For the classical data \( LG(\tau) < 1 \) needs to be valid for all \( \tau \), while the quantum signal is known to violate the LGI. The results are presented in Fig. 4a and show that in the classical case, as expected, the violation of the LGI does not occur while for the case of single nuclear spin the inequality is violated until the influence of damping of oscillations appears. Though the violation is very subtle, it exceeds \( \approx 2 \) standard deviations, and is comparable to previous work on other systems [13].

In summary we developed an accurate method to reconstruct the correlation function of the quantum observable of a single quantum system. Similar attempts have been done before [13], by continuous weak measurements on superconducting transmon qubit, which resulted in a violation of the LGI. However, in all prior work, the observable has been related to the underlying quantum process in a qualitative manner. Here we used the detailed knowledge of the interaction between the measurement system and the quantum system to get a quantitative relation between our measurement result and the underlying quantum behaviour. In addition, i.e., a single electron spin interacting with a nuclear spin has been used before to verify the LGI [15], however in this work a strong projective readout was used and continuous monitoring of the evolution of the quantum system was not possible. The present work thus extends the state of the art two-fold by achieving weak continuous monitoring of a quantum system and relating it to macroscopically observable signal in unambiguous way.

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AUTHORS’ CONTRIBUTIONS

VV and OG developed the initial idea of the experiment. OG performed theoretical work, VV, JM performed the experimental work, HS, SO, JI synthesized
and characterized the sample, VV and OG performed data analysis, VV, OG, JW wrote the manuscript, all authors discussed and commented on the manuscripts.

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Supplementary materials for "Robust determination of quantum signatures from weak measurements"

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EXPERIMENTAL SETUP

The experimental setup consists of a superconducting magnet (Scientific Magnetics) with a room temperature bore adjacent with a confocal microscope. The 12Gs AWG (Keysight M8190) provides microwaves (MW) amplified with a Traveling wave tube amplifier (Hughes 8010H) and radiofrequencies (RF) amplified with an Amplifier Research 150A250 amplifier for control for the spin experiment. The optical readout is performed via a confocal microscope with 520 nm laser diode (Thorlabs) and APD (Perkin Elmer SPCM). The objective lens (NA = 1.35) is positioned via the 3d non-magnetic piezo positioner (n-Point) (See Fig. S1). Photoluminescence is spatially filtered with a pinhole (50 um) and spectrally with a long-pass filter 650 nm (Senrock).

DIAMOND SAMPLE

The \( \langle 111 \rangle \)-oriented diamond slice \((2 \text{ mm} \times 2 \text{ mm} \times 88 \mu \text{m})\) is obtained by laser-cutting and polishing from a high crystalline quality, 99.995 \% \(^{12}\text{C}\)-enriched, type-IIa high-pressure and high-temperature (HPHT) crystal. In the original crystal, single NV centers were created by 2 MeV electron irradiation \((1.3 \times 10^{11} \text{ cm}^{-2})\) at room temperature and subsequent annealing \((1000^\circ \text{ C for 2 h in vacuum})\). The sample is positioned on a coplanar waveguide to deliver the MW and RF signal to the NV site. (see Fig S2). The B field is aligned with the NV orientation.

HYPERFINE FIELD CALIBRATION

To find a proper NV center for our experiment we examined around 20 NV centers and checked their nuclear spin environment. A set of calibration measurements for the NV center we used to calibrate the weakly coupled nuclear spin \(^{13}\text{C}\).
In addition to the absence of the ENDOR signal, we check the nuclear bath spectroscopy via high order DD resonance [2]. By going to the highest possible resonance (in our case - 21th) with KDD-XY30 sequence, we determine no visible splitting of the $^{13}C$ bath peak (Fig. S5,S6). That means that the $A_{zz}$ coupling is below 0.37 kHz. From the position of the peaks as a function of resonance number we determine the Larmor frequency with high precision (Fig. S7). Next, we check that the 1-st peak has a coherent interaction by changing the number of pulses N in the KDD-N sequence and observe coherent oscillations for NV2 (Fig. S8). We derive $\tau = 3.92033(2) \mu s$ from the best fit of the curve. From the fitting of Fig. S7, we got also the linear dependence of the resonance position as a function of its order: $a \cdot k + b$ with $a = 0.186682(1)$ and $b = 10^{-6}$. Having the value for the width of the 21st resonance we obtain an upper bound on the $A_{zz}$ coupling as:

$$
\tau_k \approx \frac{(2k + 1)\pi}{\omega_l + A_{zz}/2} \\
\delta \tau_k = \frac{(2k + 1)2\pi}{2\omega_l} - \frac{(2k + 1)2\pi}{2\omega_l} \approx \frac{A_{zz}}{\omega_l} \\
\frac{\delta \tau_k}{\tau_k} \approx \frac{A_{zz}}{\omega_l} \\
A_{zz} < \frac{\delta \tau_k}{\tau_k} \omega_l = 0.37(2\pi) \text{kHz}
$$

(1)

To further study the interaction with the bath, we obtain a 2D image of KDD-XY number and period of the interpulse sequence. The obtained figure resembles a 2D rabi oscillations with detuning. It could be seen from Fig. S9, that during the DD, the NV is moderately coupled to a single nuclear spin via the $A_{xx}$ interaction, which gives coherent oscillation in a broad region of tau intervals, while the bath interaction results in coherence fast decay after the first full oscillation. As soon as the detuning of tau is larger than the average $A_{xx}$ coupling to the bath, only coherent interaction is visible. The 2D data presented on Fig. S9 was fitted to extract the center of the fringes and the effective coupling to the nuclear spin. Results are presented in the table I.

Phase locking via $\tau$ sweep

To estimate the $\tau$ of the DD with higher precision we utilize a Hamiltonian interpolation technique [3] Since the $A_{zz}$ term is very subtle its accurate identification requires a calibration to observe it via the sequential weak measurements, and observe the decay of the correlation function, similar to [4]. At the resonance condition, a phase matching occurs, which results in reduction of the decay rate of the correlation function and a phase transition, a phase locking of the nuclear spin precession. We design a sequence, parametrized with
FIG. S4. ENDOR spectroscopy of 2 NV centers. NV5 is example with detectable nuclear spin coupling for total interrogation time of 100 µs. NV2 is the NV center used in the current research without detectable $A_{zx}$ coupled nuclear spin within the 100 µs total interrogation time.

![Graphs of NV5 and NV2](image)

FIG. S5. High-order KDD-XY-30 (totally 600 $\pi$ pulses) (maximum contrast) spectroscopy of the nuclear spin bath. No trace of $A_{zx}$ coupling, though clear indication of negative contrast, caused by coherent interaction via the $A_{zx}$ terms.

![Graph of measurement strength sweep](image)

$\tau$, which, if tau is chosen correctly, is perfectly synchronized for phase matching. This results in a nuclear spin free precession angle between subsequent measurements equal to odd number of $\pi$. Upon sweeping the $\tau$ of the DD we observe, that when the $\tau$ is resonant the correlation function is not decaying. The width of this resonance could be reduced with reducing the measurement strength. We find that $\tau = 2240.18(2)$ of AWG steps, or $\tau = 2240.18(2) \cdot 1/12$ ns $= 0.18668(1)$ µs which corresponds to the tau estimated from the slope as $\tau_{slopes} = 0.186682(1)$ µs within the error bars.

**Phase locking via $t_f$ sweep**

Alternatively, fixing the inter-pulse interval $\tau$ at resonance condition, and sweeping the free precession angle by precisely controlling the time interval added between the measurements, the phase locking feature also allows us to make an estimate on the perpendicular component of the hyperfine term. As it is described in reference [4], the orange line was fitted with the curve which resulted in the value of $A_{zx}$. We provide the value and the error in the table I. For KDD-XY-5:

$$\alpha = N_p A_{zx} \tau \pi = 0.14(1)\pi$$

$$N_p = 100$$

$$\tau = 2240.2/12\text{ ns}$$

**Measurement strength sweep**

As shown in Fig. S12, we check the dependence of the decay of the correlation function on the measurement strength, i.e., number of pulses. As a result, we fit the decay constant with $\alpha^2/4$ dependence, which gives us an approximate determination of the $A_{zx}$ coupling.
2. The phase acquired during the KDD sequence is estimated as

\[ \Phi = \cos(\omega t + \phi) \text{sinc}(N_p \tau \pi \delta). \]  

PARAMETERS OF THE SEQUENCE

Sequence scheme timing:
1. Electron spin green laser readout time: 300 ns
2. 14N polarization: 151\(\mu\)s
   (a) \(C_n R OT_e\) to electron spin 4\(\mu\)s
   (b) \(C_n R OT_n\) to nuclear spin 50\(\mu\)s
   (c) Laser repolarization. 300\(\mu\)s

Repeat a,b,c + 1\(\mu\)s wait time.
3. 10\(\mu\)s of waiting time
4. KDD5 is 35\(\mu\)s

The free precession time \(t_f\) is then estimated as the total time of the sequence.

CLASSICAL FIELD CALIBRATION

To calibrate the classical field amplitude, we perform AC sensing of 2 MHz AC external field at various output power of the external generator from -18 to 2 dbm. We apply the KDD5 sensing sequence. The algorithm is as following:

1. \(\rho_e = S_e + S_x\) state is prepared with \(\pi/2\) pulse which rotates initially polarized \(S_e + S_z\) state towards the equator.
2. The phase acquired during the KDD sequence is estimated as

\[ \Phi = \cos(\omega t + \phi) \text{sinc}(N_p \tau \pi \delta). \]  

a

FIG. S6. Zoom into the 21st resonance of the KDD-XY30 sequence for the NV2. The resonance reveals no \(A_{zz}\) coupled nuclear spins and allows for determination of Larmor frequency with higher precision.

FIG. S7. DD Resonance position in microseconds as a function of its order.

FIG. S8. Demonstration of coherent coupling in NV2. Dependence of the evolution on number of pulses in KDD-N sequence, shows the coherent oscillations for NV2, while for other NVs without signal in the ENDOR sequence, the decay results in loss of coherence, meaning that the \(A_{zz}\) coupling is also weak.

FIG. S9. The 2D image of KDD numbers vs the inter-pulse time. The image shows a coherent evolution of the single weakly coupled nuclear spin. The resonance to the bath is also visible, resulting in a dissipative dynamics.
FIG. S10. Precise determination of resonant $\tau$ of the DD sequence. Left – dependence of the decay of the correlation function on the interpulse time $\tau$. Right – the no-decay correlation function in the region of phase matching condition.

FIG. S11. Free precession angle sweeping, and phase locking effect (left). The decay of the autocorrelation vanishes as the phase acquired by the target spin between successive measurements approaches $\pi$. 
FIG. S12. Decay of the correlation function for KDD1,2,3,5

|       | CSTE (ENDOR) | 1st order KDD - N rabi ($A_{xx}$) | Weak Measurement | Phase locking in weak measurements | Correlation function fitting |
|-------|--------------|-----------------------------------|------------------|-----------------------------------|-----------------------------|
|       |               | Tau sweep 21st resonance ($A_{zz}$) | decay            |                                   |                             |
| $A_{xx}$, kHz | NA          | 9.4(2)                           | 7.8(6)           | 7.9 (5)                           | 9.5(7)                      |
| $A_{zz}$, kHz | $<10$        | $<0.37$                           | NA               | NA                                | NA                          |
| $\tau$, $\mu$s | 0.186682(1) |                                   |                  |                                   | 0.18668(1)                  |

TABLE I. Various method to estimate the hyperfine components of the single weakly coupled nuclear spin of NV2.
3. The \( \pi/2 \) pulse rotates the state around \( y \)-axis (again) and converts the X component of the state to the readable Z component. Hence

\[
S_z = \cos \Phi = \cos (\alpha \cos(\omega t + \phi) \operatorname{sinc}(N_p \tau \pi \delta)).
\] (4)

4. We perform a series of measurements without controlling the phase and time, hence we average over the argument of \( \cos(\omega t + \phi) \) which we denote as \( \phi_s \) to derive the averaged result [5] \( S_z = \int_{\phi_s} \cos (\alpha \cos(\phi_s) \operatorname{sinc}(N_p \tau \pi \delta)) d\phi_s = J_0(\alpha \operatorname{sinc}(\delta N_p \tau \pi)) \).

5. The readout of the state \( S_z \) is not ideal. It has losses of contrast due to the finite \( T_1 \) of the nuclear spin under the readout and instability of the charge state reduces the full possible contrast by constant \( K \). Hence, we obtain an expression

\[
S(\tau) = S_1 - S_2 = KJ_0(\alpha \operatorname{sinc}(N_p \tau \pi \delta)) \tag{5}
\]

with which we fit to the absorption spectra of the external signal (see Fig. S14a).

Where \( \alpha \) is the normalized field strength \( B_{rf} \), \( \phi \) is the phase of the signal at the beginning of the sequence, \( \tau \) is dynamical decoupling pulse spacing, i.e., the half period \( 0.5f^{-1} \) of the artificial signal. The \( S_z \) component readout is performed via the memory enhanced single-shot readout scheme [6]. After fitting the absorption profiles, we get a calibration of signal strength for various input powers on the signal generator for our setting (see Fig. S14b), fitted with expression \( \alpha = \alpha_0 \exp \left(\frac{P_{dbm}}{k}\right) \). We compare the absorption method with a correlation measurement with the phase modulation scheme presented in the main text of the manuscript and find an agreement between them. For the -17dbm signal power which was used for the experiment we got \( \alpha_{abs} = 0.94(5) \) and \( \alpha_{corr} = 0.88(4) \) rad respectively.

### DATA SET

We collect a series of 1000 measurements, each of those is a coherent series of 200000 sequential weak measurements. The size of 200 000 of each coherent series is motivated by hardware limitations. We then select only streams from 200 to 900 series. We combine all these measurements in one unified dataset for calculation of the correlation function of the photon counts. The walking average with window size equal to one series is depicted on Fig. S15a. We also show the blue shaded region is measurement data taken for analysis. The initial and fi-
FIG. S14. External field amplitude calibration. a) Absorption profiles of KDD5 sequence as a function of inter-pulse spacing $\tau$ for various signal amplitudes. b) The extracted parameter $\alpha$ of the classical signal as a function of signal power on the RF generator.

The final data was discarded due to initial heat up of the setup. The final data set was discarded due to long-term instabilities in the optical collection path and drift in average photon counting.

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FIG. S15. Data set for LGI violation experiment.  

**a)** The walking average photon number output with averaging window size of $2 \cdot 10^5$ measurements.  

**b)** The statistical distribution shows that standard deviation of the photon counts is within the Poisson distribution, showing that the relative standard deviation within $2 \cdot 10^5$ measurements follows $1/\sqrt{n_{av}}$. 

