

Opening up three quantum boxes causes classically undetectable wavefunction collapse

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One of the most striking features of quantum mechanics is the profound effect exerted by measurements alone. Sophisticated quantum control is now available in several experimental systems, exposing discrepancies between quantum and classical mechanics whenever measurement induces disturbance of the interrogated system. In practice, such discrepancies may frequently be explained as the back-action required by quantum mechanics adding quantum noise to a classical signal. Here, we implement the “three-box” quantum game [Aharonov Y, et al. (1991) *J Phys A Math Gen* 24(10):2315–2328] by using state-of-the-art control and measurement of the nitrogen vacancy center in diamond. In this protocol, the back-action of quantum measurements adds no detectable disturbance to the classical description of the game. Quantum and classical mechanics then make contradictory predictions for the same experimental procedure; however, classical observers are unable to invoke measurement-induced disturbance to explain the discrepancy. We quantify the residual disturbance of our measurements and obtain data that rule out any classical model by ≥ 7.8 standard deviations, allowing us to exclude the property of macroscopic state definiteness from our system. Our experiment is then equivalent to the test of quantum noncontextuality [Kochen S, Specker E (1967) *J Math Mech* 17(1):59–87] that successfully addresses the measurement detectability loophole.

Leggett–Garg | quantum contextuality | quantum non-demolition measurement

Classical physics describes the nature of systems that are “large” enough to be considered as occupying one definite state in an available state space at any given time. Macrorealism (MR) applies whenever it is possible to perform nondisturbing measurements that identify this state without significantly modifying the system’s subsequent behavior (1). MR allows the assignment of a definite history (or probabilities over histories) to classical systems of interest, but the MR condition can break down for systems “small” enough to be quantum mechanical during times “short” enough to be quantum coherent: times and distances that now exceed seconds (2) and millimeters (3) in the solid state. How can we tell whether a particular case is better described by quantum mechanics (QM) or MR? If there is a crossover between these, what does it represent?

One explanation for the breakdown of MR is that measurement back-action (either deliberate measurements by an experimenter or effective measurements from the environment) unavoidably change the state in the quantum limit, excluding MR due to a breakdown of nondisturbing measurability. This position is supported by “weak value” experiments (4, 5) that explore the transition from quantum to classical behavior as a measurement coupling is varied. Quantum behavior is found under weak coupling, whereas MR-compatible behavior is recovered when strong projective measurements effectively “impose” a classical value onto the measured quantum system (4).

We examine a case in which the back-actions of sequential “strong” projective measurements impose new quantum states that provide no detectable indication of disturbance to a “macro-realist” observer. We show that these states are still incompatible with MR, however, because no possible MR-compatible history can be assigned to the process as a whole. Our experiment can be described as a game played by two opponents (Alice and Bob) who take alternate turns to measure a shared system. The system they share may or may not obey the axioms of MR. For the purposes of the game, Bob assumes he may rely on the MR assumptions being true and only Alice is permitted to manipulate the system between measurements. If Bob is correct to assume MR holds, the game they play is constructed in his favor; however, “paradoxically,” the exact same sequence of operations will define a game that favors Alice when a quantum-coherent description of the system is valid (6).

Experimentally, we use the ^{14}N nuclear spin of the nitrogen vacancy ($^{14}\text{NV}^-$) center ($S = 1$, $I = 1$) in diamond as Alice and Bob’s shared system, enabling us to maintain near-perfect undetectability by Alice of Bob’s observations. The experiment involves pre- and postselection (5, 7) on a three-level quantum system that is known to be equivalent to a Kochen–Specker test of quantum noncontextuality (8). Such tests are only possible in $d \geq 3$ Hilbert spaces (9); here, we use recent advances in the engineering (10) and control (11) of the NV^- system that enable the multiple projective nondemolition measurements that are crucial to observing Alice’s quantum advantage in the laboratory. We describe the game (12) and Bob’s verification of it from the MR perspective, and we then discuss the experiment and results from the QM position. We quantify the incompatibility of our results with MR through use of a Leggett–Garg inequality (1) and discuss the implications of our result.

In the “three-box” quantum game (12), Alice and Bob each inspect a freshly prepared three-state system (classically, three separate boxes hiding one ball) using an apparatus that answers the question “Is the system now in state j ?” (“Is the ball in box j ?”) for $j = 1, 2, 3$ by responding either “true” (1) or “false” (0). The question is answered by performing one of three mutually orthogonal measurements M_j . The game allows Bob a single use of either M_1 or M_2 . Alice is allowed to use only M_3 , and, additionally,

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she is allowed to manipulate the system. Alice is allowed one turn (a manipulation either before or after an M_3 measurement) before Bob to prepare the system and one turn following him. Alice attempts to guess Bob's measurement result, and the pair bet on Alice correctly answering the question, "Did Bob find his M_j to be true?" Alice offers Bob $\geq 50\%$ odds to predict when his M_j is true, although she may "pass" on any given round at no cost when she is undecided.

Bob realizes that if the M_j measurements are performed on a system following MR axioms, Alice must bet incorrectly $\geq 50\%$ of the time, even if Alice could "cheat" by knowing which j -value will be presented (classically, knowing which box contains the ball); with three boxes and his free choice between M_1 and M_2 , Alice is prevented from using her prior knowledge to win with a $>50\%$ success rate. Bob expects to win if the M_j measurements reproduce the behavior of a ball hidden in one of the three boxes. The conditions for this are (a) the M_j measurements are repeatable and mutually exclusive, such that $M_j \wedge M_k = \delta_{jk}$ (classically, the ball does not move when measured); (b) for any trial, $M_1 \vee M_2 \vee M_3 = 1$ (there is only one ball, and it is definitely in one of the boxes); (c) Bob has an equal probability of finding each j -value when measuring a fresh state, with $P_{M_j}(B) = 1/3 \ j \in 1, 2, 3$ (the ball is placed at random); and (d) Alice has no additional means to determine which, if any, M_j measurement Bob has chosen to perform. The conditions a–d serve to prevent Alice from learning Bob's M_j result in any macroreal system. Before accepting Alice's invitation to play, Bob verifies that properties a–d hold experimentally by carrying out M_j measurements. During verification, the game rules are relaxed and Bob is permitted to make pairs of sequential measurements, checking $M_j \wedge M_k = \delta_{jk}$. He is also allowed to measure every M_j , including M_3 , which will be reserved for Alice once betting commences, or he may opt to perform no measurement at all and monitor Alice's response to determine if she can detect a disturbance caused by his measurement (SI Text).

When Bob is satisfied that a–d hold, the game appears fair from his macrorealist standpoint. Bob accepts Alice's wager, and play commences with Alice preparing a state, which Bob measures using either M_1 or M_2 , while keeping his j -choice and M_j result secret. Alice manipulates the system, uses her M_3 measurement, and bets whenever her M_3 result is true. Believing that Alice could only guess his secret result, Bob accepts Alice's wager. Doing so, he finds that Alice's probability of obtaining a true M_3 result is $P_{M_3}(A) \approx 1/9$, independent of his j -choice between M_1 , M_2 , or no measurement. Under MR, Bob could account for this only through Alice using a nondeterministic manipulation that would reduce the information available to her from the M_3 result. To Bob's surprise, when Alice plays, her true M_3 results coincide with

the rounds on which Bob's M_j -result was also true. She passes whenever Bob's M_j result was false. In a perfect experiment, she would win every round she chose to play; in our practical realization, she achieves significantly more than the 50% success rate that would be predicted by MR. To understand Alice's advantage, we must examine the game from a QM perspective.

Alice uses the initial M_3 measurement to obtain the pure quantum state $|3\rangle$, passing on all rounds in which her initial M_3 measurement is false. She applies the unitary \hat{U}_1 , which operates as $\hat{U}_1 = |I\rangle\langle 3| + (\text{orthogonal terms})$, to produce the initial state:

$$|I\rangle = \frac{|1\rangle + |2\rangle + |3\rangle}{\sqrt{3}} \quad [1]$$

Her first turn presents the state $|I\rangle$ to Bob, who next measures M_j on $|I\rangle$, performing a projection. If Bob's M_j result is true, he has applied the quantum projector $\hat{P}_j = |j\rangle\langle j|$, and by finding an M_j result that is false, he has applied $\hat{P}_j^\perp = 1 - |j\rangle\langle j|$. Alice uses her final turn to measure the component of the state left by Bob's measurement along the state $|F\rangle = (|1\rangle + |2\rangle - |3\rangle)/\sqrt{3}$. Bob's projectors on Alice's initial and final states $|I\rangle$ and $|F\rangle$ obey:

$$\left| \langle F | \hat{P}_1 | I \rangle \right|^2 = \left| \langle F | \hat{P}_2 | I \rangle \right|^2 = 1/9 \quad [2]$$

$$\left| \langle F | \hat{P}_1^\perp | I \rangle \right|^2 = \left| \langle F | \hat{P}_2^\perp | I \rangle \right|^2 = 0 \quad [3]$$

for both $j = 1$ and $j = 2$. Alice cannot directly measure $|F\rangle$ but is able to transform state $|F\rangle$ into state $|3\rangle$ with a unitary $\hat{U}_F = |3\rangle\langle F| + (\text{orthogonal terms})$, and she uses her measurement of M_3 as an effective M_F measurement. Alice therefore obtains M_3 -true when Bob's M_j result is true with probability $P_{M_3}(A \cap B) = |\langle 3 | \hat{U}_F \hat{P}_j \hat{U}_1 | 3 \rangle|^2 = |\langle F | \hat{P}_j | I \rangle|^2 = 1/9$ and when Bob's M_j result is false with probability $P_{M_3}(A \cap \neg B) = |\langle 3 | \hat{U}_F \hat{P}_j^\perp \hat{U}_1 | 3 \rangle|^2 = |\langle F | \hat{P}_j^\perp | I \rangle|^2 = 0$. Alice finds that her M_3 result being true is conditional on Bob leaving a component of $|\psi_j\rangle$ along $|F\rangle$; to do so, his M_j result cannot have been false. Alice's probability conditioned on Bob is then $P_{M_j}(B|A) = 1$. Alice bets whenever her M_3 result is true, playing one-ninth of the rounds and winning each round she plays.

Materials and Methods

Our implementation of this game uses the NV⁻ center, which hosts an excellent three-level quantum system for the three-box game: the ¹⁴N nucleus, which has $(2I + 1) = 3$ quantum states (Fig. 1A). Although we cannot (yet)

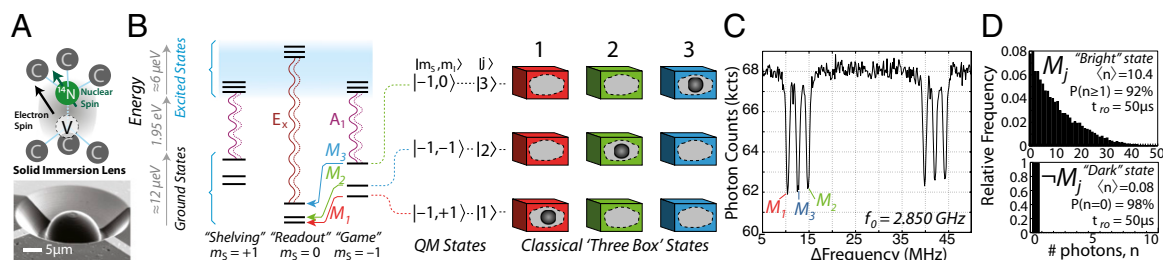


Fig. 1. Three-box game is implemented using the ¹⁴N nuclear spin of the NV⁻ center in diamond, measured using the electron spin. (A) Schematic of the NV⁻ defect in diamond and representative diamond lens used in the measurements. (B) Magnetic moment of the electron spin is quantized into one of three values: $m_s = -1, 0$, or $+1$. These states split into a further three ($m_l = -1, 0$, or $+1$) according to the magnetic moment of the ¹⁴N nuclear spin. The $m_s = \pm 1$ states fluoresce via the A_1 transition, whereas $m_s = 0$ fluoresces via the E_x transition. We use the $m_s = -1$ manifold to hold the three states in the game, conditionally moving the state between $m_s = -1$ and $m_s = 0$ dependent on the nuclear spin sublevel m_l . These three m_l states are taken to correspond to the configurations of a hidden ball. (C) We identify the allowed microwave transitions ($\Delta m_s = 1, \Delta m_l = 0$) that provide the M_j readouts. (D) Photon counting statistics, in each case from 10,000 trials, observed during a typical projective readout indicate the presence (Upper) or absence (Lower) of optical fluorescence, corresponding to outcomes M_j and $-M_j$, respectively.

superpose a physical ball under three separate boxes, real-space separation is not essential to the three-box argument. Alice and Bob can bet on any physical property of a system for which MR assigns mutually exclusive outcomes; for instance, a classical gyroscope revolving about one of three possible axes is not simultaneously revolving about the second and third axes. By using rf pulses (13), we can readily prepare the ^{14}N angular momentum into a superposition of alignment along three distinct spatial axes, providing three “box states” that are presumed to be mutually exclusive in the macrorealist picture. We work in the electron spin $m_S = -1$ manifold and assign eigenvalues of nitrogen nuclear spin m_I to the box-states j according to (a) $|m_I = -1\rangle \sim |j = 1\rangle$, (b) $|m_I = +1\rangle \sim |j = 2\rangle$, and (c) $|m_I = 0\rangle \sim |j = 3\rangle$ (Fig. 1B).

Preparation and readout of the ^{14}N nuclear spin is provided via the NV⁻ electronic spin ($S = 1$). We use selective microwave pulses to change m_S conditioned on m_I , reading out the electron spin in a single shot and with high fidelity (11), by exploiting the electron spin-selective optical transitions of the NV⁻ center. The spin readout achieves 96% fidelity and takes $\approx 20\ \mu\text{s}$, which is much shorter than the nuclear spin inhomogeneous coherence lifetime of $T_2^* \gg 1\ \text{ms}$ at $T = 8.7\ \text{K}$, enabling three sequential readout operations during a single coherent evolution of the system, as required for our three-box implementation. We achieve all steps of the quantum experiment well within the coherence time of our system, and therefore make no use of refocusing rf pulses.

The full experimental sequence is shown in Fig. 2, with further details provided in *SI Text*. The initial state $|3\rangle$ is prepared by projective nuclear spin readout using a short-duration ($\approx 200\ \text{ns}$) optical excitation. The subsequent experiment is then conditioned on detection of at least one photon during the preparation phase, which heralds $|3\rangle$ with $\geq 95\%$ fidelity (Fig. 1D) at the expense of $\leq 1\%$ preparation success rate. Once $|3\rangle$ is heralded, all subsequent data are accepted unconditionally. After initialization, Alice transforms the state $|3\rangle$ into $|I\rangle$ via two rf pulses (*SI Text*) and hands the system to Bob, who measures M_1 or M_2 . A further four rf pulses transform $|F\rangle$ to $|3\rangle$, and Alice performs her final M_3 measurement while statistics about Alice and Bob’s relative successes are recorded.

We quantify the discrepancy between MR and QM by constructing a Leggett-Garg function for our system, defined as

$$\langle K \rangle = \langle Q_1 Q_2 \rangle + \langle Q_2 Q_3 \rangle + \langle Q_1 Q_3 \rangle \quad [4]$$

where Q_j are observables of our system with values ± 1 , recorded at three different times, derived from Alice and Bob’s measurements (1). We assign $Q_j = +1$ whenever an M_3 result is true (or could be inferred true in the MR picture) and assign $Q_j = -1$ otherwise. The initially heralded state $|3\rangle$ fixes the value of $Q_1 = +1$ always, and values for Q_2 and Q_3 are taken directly from Bob and Alice’s measurement results. The Leggett-Garg function is known to satisfy $-1 \leq \langle K \rangle \leq +3$ for all MR systems (1), and for the present system, we can show that $\langle K \rangle$ is related to Bob and Alice’s statistics (*SI Text*) as follows:

$$\langle K \rangle = \frac{4}{9} (1 - P_{M_1}(B|A) - P_{M_2}(B|A)) - 1 \quad [5]$$

where $P_{M_j}(B|A)$ is the probability that Bob finds the M_j result true, given that Alice has also found her final M_3 result true. MR asserts that M_1 and M_2 are mutually exclusive events, whereas QM does not, such that:

$$\text{MR} : P_{M_1}(B|A) + P_{M_2}(B|A) \leq 1 \quad [6]$$

$$\text{QM} : P_{M_1}(B|A) + P_{M_2}(B|A) \leq 2 \quad [7]$$

Under QM assumptions, Eq. 5 satisfies $\langle K \rangle \geq -13/9 = -1.44$, possibly lying outside the range compatible with MR.

Results

Bob picks a secret j -value and maps the corresponding nuclear spin projection to the electron spin by applying a microwave π -pulse to drive a transition from one of the $m_S = -1$ states ($|j\rangle$ is $|1\rangle$ or $|2\rangle$) into the $m_S = 0$ manifold. He then uses optical measurement of the E_x fluorescence to determine m_S . Absence of fluorescence (“ E_x -dark” NV⁻) implies $-M_j$ and collapses the electron state into $m_S = -1$ while performing \hat{P}_j on the nuclear spin (Fig. 3A, *ii*). We find that nuclear spin coherences within $m_S = -1$ are unaffected by the $-M_j$ readout process.

Detection of $n \geq 1$ photons during Bob’s 20- μs readout projects the electron into $m_S = 0$ and corresponds to an M_j result that is true. In such events, there is an $\approx 70\%$ chance the electronic spin will be left in an incoherent mixture of $m_S = \pm 1$ following readout, due to optical pumping (11). Conditional on Bob’s M_j result being true, we take care to undo the mixing effect as follows. We first pump the electron spin to $m_S = 0$ by selective optical excitation of $m_S = \pm 1$ (via a laser resonant with the A_1 transition), followed by driving a selective microwave pulse from $m_S = 0$ to $m_S = -1$ (Fig. 1C). This procedure is effective because the optical fluorescence preserves the nuclear spin populations m_I that encode the game eigenstates in $\geq 70\%$ of cases (Fig. 3B). Bob performs repeated pairs of measurements, verifying from a macrorealist’s perspective that performing M_j is equivalent to opening one of the three boxes containing a hidden ball. Bob finds the probability for each M_j is $\approx 1/3$ (Fig. 3A, *i*). Bob performs consecutive M_j observations and verifies that finding M_j ($-M_j$) true on one run implies that the subsequent measurement of M_j ($-M_j$) will also be true (Fig. 3B and C), gathering statistics over $n = 1,200$ trials for each combination.

Once Bob has measured in secret, Alice predicts his result by mapping $|F\rangle$ to $|3\rangle$ and performing M_3 . Alice accomplishes this via: $|F\rangle \rightarrow |I\rangle \rightarrow |3\rangle$. The Berry’s phase associated with 2π rotations (14) provides the map $|F\rangle \rightarrow |I\rangle$ via two rf pulses that change the signs of the $\{|1\rangle, |3\rangle\}$ and then $\{|2\rangle, |3\rangle\}$ states. State $|3\rangle$ then acquires two sign changes yielding $|F\rangle$ up to a global phase. The map \hat{U}_I from $|I\rangle$ to $|3\rangle$ is then achieved by inverting the order and phase of Alice’s initial \hat{U}_I pulses (*SI Text*).

Alice and Bob compare their measurement results during $n = 2 * 1,200$ rounds of play, distributed evenly across Bob’s two choices of M_j measurement, as well as during a further 1,200 rounds in which Bob performs no measurement whatsoever.

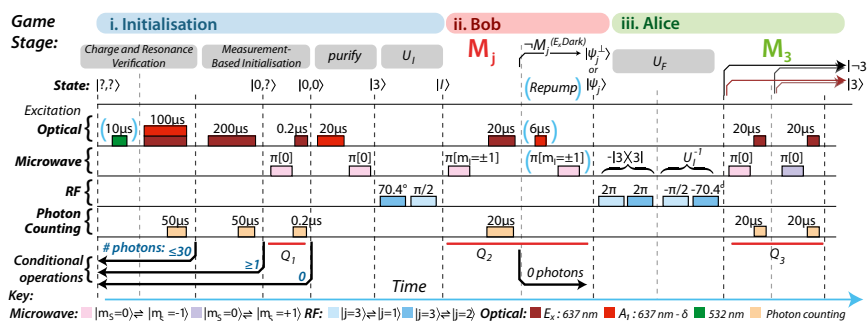


Fig. 2. Microwave, rf (RF), and optical pulse sequence implementation in the three-box experiment. (i) Initialization consists of preparing the NV⁻ state via charge-state verification and measurement-based initialization into state $|3\rangle$, followed by purification of $m_S = 0$ and application of \hat{U}_I . (ii) Bob’s measurement M_j consists of moving the population from $m_S = -1$ to $m_S = 0$ conditioned on m_I , indicated as $\pi[m_I]$ in the figure, followed by monitoring of E_x fluorescence. If fluorescence is observed, a “repopulating” sequence via the spectrally resolved A_1 fluorescence ($\lambda_{A_1} - \lambda_{E_x} = \delta = 1.89\ \text{pm}$) resets $m_S = -1$ while leaving m_I unchanged and ready for Alice’s measurement. (iii) Alice’s measurement consists of the unitary \hat{U}_F , followed by readout of M_3 in the $m_S = -1$ and $m_S = +1$ sublevels. Further details on the experimental sequence are provided in *SI Text*.

Alice finds her final M_3 result is true in $\approx 15\%$ of cases, independent of Bob's choice of measurement context between M_1 and M_2 or neither measurement (Fig. 4A). Among those $\approx 15\%$ of cases in which Alice's M_3 result is true and she chooses to bet, Bob finds she wins $\geq 67\%$ of such rounds for either of Bob's choices between measuring M_1 and M_2 (Fig. 4B), confounding the macrorealist expectation. The principle source of error in our experiment arises from imperfect control of the nuclear spin (SI Text).

We quantify the Leggett–Garg inequality violation in our experiment by determining $\langle K \rangle$ from estimates of $P_{M_j}(B|A)$, finding $\langle K \rangle = -1.265 \pm 0.23$, corresponding to a $\approx 11.3 \sigma$ -violation of the Leggett–Garg inequality under fair sampling assumptions and to a $\approx 7.8 \sigma$ -violation in a “maximally adverse” macrorealist position in which all undetermined measurements are assumed to represent Alice “cheating” and are reassigned to minimize the discrepancy between QM and MR predictions (SI Text).

Discussion

Our results unite two concepts in foundational physics: Leggett–Garg inequalities (1) and pre- and postselected effects (7) in a quantum system to which the Kochen–Specker no-go theorem applies (9). Previous experimental studies of the Leggett–Garg inequality have used ensembles (15, 16), have made assumptions regarding process stationarity (17, 18), or have required weak measurements (4) to draw conclusions, whereas the existing studies of the three-box problem cannot incorporate measurement nondetectability (19, 20), presenting a loophole that allows classical noncontextual models to reproduce the quantum statistics (8). We have studied the three-box experiment on a matter

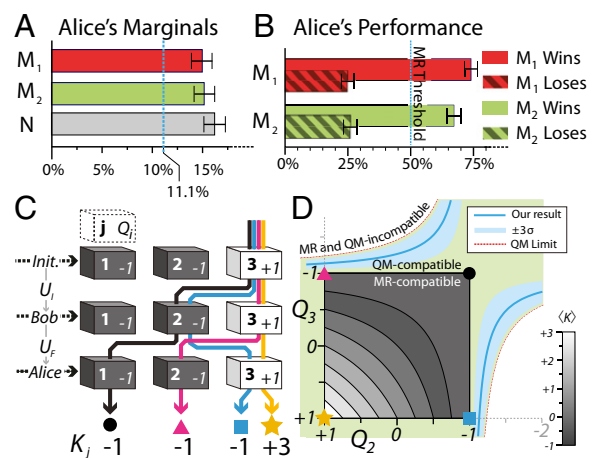


Fig. 4. Violation of a Leggett–Garg inequality in the three-box game. (A) Alice's measurement M_3 is independent of Bob's choice to perform measurement M_1 , M_2 , or neither (N). (B) Observations of Bob and Alice are correlated to indicate the probability that Bob has (or has not) seen state M_j , given that Alice has seen M_3 , determining who “wins” the game. Alice's probability of winning exceeds 50% for both of Bob's choices M_1 and M_2 . (C) Four MR-compatible histories that extremize $\langle K \rangle$ are illustrated by four trajectories passing through different boxes during the game. A trajectory entirely within the white $j = 3$ boxes has $Q_{(1, 2, 3)} = +1$ and yields $\langle K \rangle = +3$. Histories that visit other boxes yield $\langle K \rangle = -1$. (D) $\langle K \rangle$ values of the four paths are shown in the corners of the (Q_2, Q_3) graph. Values for $\langle Q_2 \rangle$ and $\langle Q_3 \rangle$ from MR-compatible experiments must lie inside the shaded square satisfying $-1 \leq \langle K \rangle \leq 3$. Our measurements lie on the cyan curve outside this region but within the region allowed by QM.

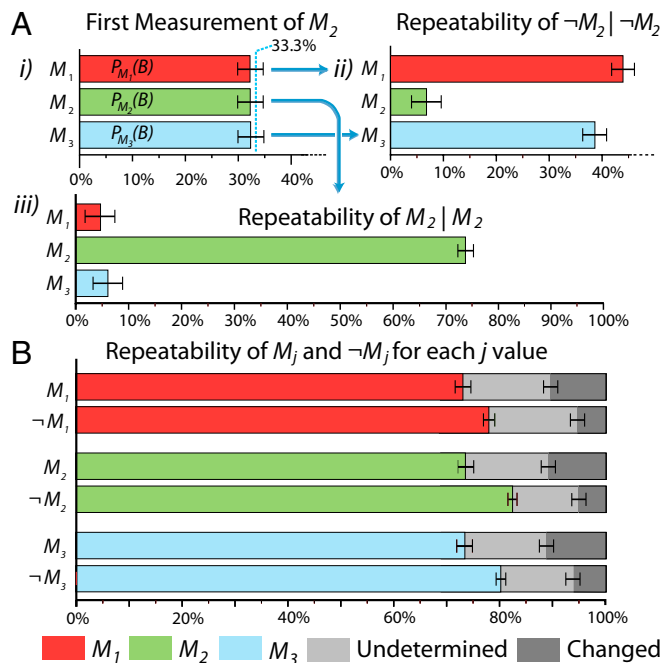


Fig. 3. Bob verifies the nondetectable nature of the M_j measurements. (A) Measurement within the $m_s = -1$ manifold only. (i) Bob's measurement results when observing the state $|l\rangle$ in the $|j\rangle$ basis are independent of the j -value selected to within experimental error. Repeatability is illustrated by plotting the result of a second M_j measurement within $m_s = -1$, conditioned on (ii) the result $-M_2$, or (iii) the result M_2 . (B) Repeatability of each M_j measurement is studied within the $m_s = -1$ manifold; a finite probability exists for the electron spin to branch into the $m_s = +1$ manifold, yielding an undetermined reading, and for the nuclear spin to flip producing a “definitely changes” outcome. Error bars show $\pm 2 \sigma/95\%$ confidence intervals.

system, as originally conceived (12) and developed (6) in terms of sequential, projective nondemolition measurements, and we therefore reexamine the conclusions that can be drawn when using this improved measurement capability.

Two assumptions underpin MR: (i) macroscopic state definiteness and (ii) nondisturbing measurability. In previous studies, it has been possible to assign violations of the Leggett–Garg inequality to a loss of nondisturbing measurability in both optical (4) and spin-based (16) experiments. The disturbance due to measurement can sometimes be surprisingly nonlocal (21), and it has been suggested that detectable disturbance is a necessary condition for violating a Leggett–Garg inequality in all cases (22, 23). We improve this result, clarifying that detectable disturbance is a necessary condition for violating the Leggett–Garg inequality in two-level quantum systems but is not required in the three-level system studied here (SI Text).

We show from the statistics of the measurement outcomes that Alice cannot detect Bob's choice to measure or not (Fig. 4A); thus, our measurements involve no detectable disturbance, whereas the statistics from the three-box game violate a Leggett–Garg inequality. We are therefore able to rule out the macrorealist's assumption i of state definiteness, a result unobtainable from previous studies of two-level quantum systems.

Our experiment makes use of a three-level quantum system in which Bob's choice between M_1 and M_2 represents a choice of measurement “context” in the language of Kochen and Specker (9). If Bob is able to keep his measurement context secret, a macrorealist Alice could only use a “noncontextual” classical theory to describe the experiment. It is known that every pre- and postselection paradox implies a Kochen–Specker proof of quantum contextuality (8). It has been argued that measurement disturbance provides a loophole to admit noncontextuality into classical models [in addition to finite measurement precision (24, 25)]; all classical models presented to date that exploit this loophole give rise to detectable measurement disturbances. In

our experiment, Bob's intervening measurement introduces no disturbances detectable by Alice and cannot be accounted for by existing classical models.

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