Observation of Environment-Induced Double Sudden Transitions in Geometric Quantum Correlations

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Correlations in quantum systems exhibit a rich phenomenology under the effect of various sources of noise. We investigate theoretically and experimentally the dynamics of quantum correlations and their classical counterparts in two nuclear magnetic resonance setups, as measured by geometric quantifiers based on trace norm. We consider two-qubit systems prepared in Bell diagonal states, and perform the experiments in real decohering environments resulting from Markovian local noise which preserves the Bell diagonal form of the states. We then report the first observation of environment-induced double sudden transitions in the geometric quantum correlations, a genuinely nonclassical effect not observable in classical correlations. The evolution of classical correlations in our physical implementation reveals in turn the finite-time relaxation to a pointer basis under nondissipative decoherence, which we characterize geometrically in full analogy with predictions based on entropic measures.

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Introduction.-Quantum technology embraces some of the most exciting modern developments in physics, mathematics, engineering, and computer science. These were spurred by the discovery that distinctive quantum features, such as superposition and entanglement, can be exploited as practical resources for novel or enhanced sensing, communication, and computation [1]. Mass adoption of quantum technologies, however, demands a deeper understanding of the ultimate ingredients which enable improvements over classical scenarios and the ability to sustain those properties against the typically detrimental effects of decoherence. Beyond entanglement, subtler forms of nonclassical correlations such as the quantum discord [2,3] have emerged as key quantum signatures [4-6] with operational roles, e.g., in quantum metrology [7–9], entanglement activation [10–12], information encoding, and distribution [13,14]. Studying the behavior of quantum correlations under physical sources of noise is then pivotal to their exploitation for real-world implementation of these and related protocols.

In the presence of decohering environments, quantum discord does not exhibit the phenomenon of sudden death [15–17], and is typically more robust against noise than entanglement [18] (see also Refs. [19,20]). When evaluated on simple Bell diagonal states of two qubits, the traditional entropic measure of quantum discord [2] displays peculiar properties in its decay rate, such as freezing [21] and sudden changes, i.e., abrupt transitions at specific evolution times [22] (see Ref. [23] for a critical study on more general states). Moreover, the entropic classical correlation [3] complementary to the quantum discord has

recently been employed to characterize the emergence of the pointer basis of an apparatus subject to decoherence [24]. This characterization shows that the time τ_E for the emergence of the pointer basis is conceptually distinct from the decoherence half-life τ_D (decoherence time). Experimental investigations of both the freezing and the sudden change phenomena have been realized through different physical setups [25–27], while the emergence of a pointer basis has only recently been observed in an experiment with polarization entangled photon pairs [24].

Alternatively to an entropic approach, quantum correlations in a bipartite quantum state ρ can also be measured and typically more simply evaluated-via a geometric scenario, in terms of the distance between ρ and the closest state with zero discord, where states with zero discord are described by density operators which are left undisturbed by a nonselective measurement over one subsystem. In this context, the trace-norm (Schatten 1-norm) geometric quantum discord (GQD-1) plays a special role [28-33], as it defines a rigorous and physically motivated measure which does not increase under local trace-preserving quantum channels for the unmeasured part (as a consequence of the contractivity of the trace norm [28,34,35]). The GQD-1 thus does not suffer from an inherent problem which affects instead the Hilbert-Schmidt (Schatten 2-norm) geometric quantum discord originally introduced in [36], as well as any Schatten *p*-norm measure of discord for $p \neq 1$. The GQD-1 furthermore inherits the robustness of the entropic quantum discord against noise, because it is also capable of exhibiting freezing [37] and sudden change behaviors [38]. Moreover, it displays a new effect for Bell diagonal states, which is the possibility of double sudden changes [38]. The freezing and single sudden change behaviors for GQD-1 have recently been experimentally verified [39] through the observation of the negativity of quantumness [11], a quantum correlation measure equivalent to GQD-1 for qubit states [31,32].

In this Letter we report the first experimental realization of the double sudden change behavior via two distinct NMR setups, where different Markovian noisy channels are induced by real physical interactions inducing relaxations on partially polarized nuclear spin ensembles. Moreover, we show that these double sudden changes are a genuinely quantum effect, which are forbidden in the geometric classical correlations associated with GOD-1 [40,41]. Geometric quantum and classical correlations take very simple and experimentally friendly expressions for Bell diagonal states, directly given by spin-spin correlation functions in different directions. Our study further allows the establishment of a complete equivalence between the entropic and the geometric characterizations of the emergence of the pointer basis in an apparatus subject to decoherence. The relaxation to the pointer basis at a finite emergence time τ_E is then demonstrated as a by-product in our NMR implementation.

Geometric correlations.—Let us begin by considering a two-qubit system *AB* described by a density operator ρ . We will focus here on the particular case of two-qubit Bell diagonal states, whose density operator is of the form $\rho = \frac{1}{4} [\mathbb{I} \otimes \mathbb{I} + \sum_{i=1}^{3} c_i \sigma_i \otimes \sigma_i]$, where \mathbb{I} is the identity matrix, $\{\sigma_i\}$ are the Pauli matrices, and $\{c_i = \langle \sigma_i \otimes \sigma_i \rangle\}$ are spinspin correlation functions. Geometric quantifiers of quantum (namely, GQD-1) and classical correlations between *A* and *B* can be defined through the trace distances [40]

$$Q_G(\rho) = \inf_{\chi} \operatorname{tr} |\rho - \chi| = \operatorname{tr} |\rho - \chi_{\rho}|, \qquad (1)$$

$$C_G(\rho) = \operatorname{tr}[\chi_\rho - \pi_\rho], \qquad (2)$$

where χ_{ρ} is a classical-quantum state [42] emerging from a projective measurement on subsystem A that minimizes Eq. (1), whereas $\pi_{\rho} = \rho_A \otimes \rho_B$ represents the product of the local marginals of ρ . In this case, $\rho_A = \rho_B = \mathbb{I}/2$ and $\chi_{\rho} = (\mathbb{I} \otimes \mathbb{I} + c_j \sigma_j \otimes \sigma_j)/4$, where *j* is such that $|c_j|$ corresponds to the maximum of the set { $|c_i|$ }. Defining c_- , c_0 , and c_+ as the minimum, intermediate, and maximum of { $|c_i|$ }, respectively, Eqs. (1) and (2) reduce to [40]

$$Q_G = c_0, \qquad C_G = c_+.$$
 (3)

It is worth mentioning that the classical part in Eq. (3) is monotonically related to the expression obtained in Ref. [41] through an alternative definition for the geometric trace-norm classical correlation, which takes into account an extra optimization to find out the closest product state.

Decoherence.—We will consider the system-environment interaction through the operator-sum representation formalism [1]. In this context, we will take the evolution of the quantum state ρ as described by a trace-preserving quantum channel given by $\varepsilon(\rho) = \sum_{i,j} (E_i^A \otimes E_j^B) \rho(E_i^A \otimes E_j^A)$ E_j^B)[†], where $\{E_k^s\}$ is the set of Kraus operators associated with a decohering process of the qubit *s*, with the tracepreserving condition reading $\sum_k E_k^{s\dagger} E_k^s = \mathbb{I}$. We provide in Table I the Kraus operators for the two quantum channels considered in this work: phase damping (PD) and generalized amplitude damping (GAD).

The PD decoherence process preserves the Bell diagonal form of the density operator ρ . For the case of GAD, the Bell diagonal form is kept for $\gamma_s = 1/2$. In this situation, as $p_s = 1 - \exp[-t/T_s]$ is a function time t, with T_s the relaxation time of the qubit s, we can write

$$\varepsilon(\rho) = \rho(t) = \frac{1}{4} \bigg[\mathbb{I} \otimes \mathbb{I} + \sum_{i=1}^{3} c_i(t) \sigma_i \otimes \sigma_i \bigg], \quad (4)$$

where the time-dependent correlation function $c_i(t)$ is given in Table II in terms of the initial value $c_i(0) = c_i$ and of the decoherence time $\tau_D = T_A T_B / (T_A + T_B)$.

Since $\rho(t)$ preserves the Bell diagonal form, we can directly obtain the dynamics of the geometric quantum and classical correlations from Table II by using the respective expressions $Q_G(t) = c_0(t)$, $C_G(t) = c_+(t)$. Thus, we can get general patterns of the geometric correlations as functions of the parameters $\{c_j \equiv c_j(0)\}_{j=-,0,+}$ and τ_D . Depending on the values assumed by these parameters, Q(t) and C(t) may present nonanalyticities caused by crossings among $|c_1(t)|$, $|c_2(t)|$, and $|c_3(t)|$, which give rise to the occurrence of double sudden change and emergence of the pointer basis phenomena.

Double sudden change.—For a two-qubit Bell diagonal state evolving under Markovian local noise preserving their Bell diagonal form as described by the PD or GAD channels, the quantum geometric correlation (GQD-1) is able to exhibit at most two sudden changes as a function of time. Indeed, it can be observed in Table II that, for both PD and GAD channels, $|c_1(t)|$ and $|c_2(t)|$ display the same decay rate, which means that they do not cross as functions of t. Therefore, only the crossings $|c_3(t)| = |c_1(t)|$ and $|c_2(t)|$ are allowed, implying at most two nonanalyticities in the intermediate value $c_0(t) = Q_G(t)$. The analytical expressions for the critical points $t = t_1^*$ and $t = t_2^*$ ($t_2^* > t_1^*$) associated with this double sudden change behavior are indicated in Table III. These results generalize the expressions obtained in Ref. [38] for the particular case $T_A = T_B$.

On the other hand, the crossings $|c_3(t)| = |c_1(t)|$ and $|c_3(t)| = |c_2(t)|$ allow for at most a single nonanalyticity in

TABLE I. Kraus operators for the PD and GAD quantum channels, where the parameters p_s and γ_s represent decoherence probabilities.

Channel	Kraus operators
PD	$E_0^s = \sqrt{1 - p_s/2}\mathbb{I}, \ E_1^s = \sqrt{p_s/2}\sigma_3.$
GAD	$E_0^s = \sqrt{\gamma_s} \begin{bmatrix} 1 & 0 \\ 0 & \sqrt{1 - p_s} \end{bmatrix}, E_2^s = \sqrt{1 - \gamma_s} \begin{bmatrix} \sqrt{1 - p_s} & 0 \\ 0 & 1 \end{bmatrix},$ $E_1^s = \sqrt{\gamma_s} \begin{bmatrix} 0 & \sqrt{p_s} \\ 0 & 0 \end{bmatrix}, E_3^s = \sqrt{1 - \gamma_s} \begin{bmatrix} 0 & 0 \\ \sqrt{p_s} & 0 \end{bmatrix}.$

TABLE II. Correlation function $c_i(t)$ for the PD and GAD quantum channels in terms of c_i and τ_D . For GAD, we fixed $\gamma_s = 1/2$.

Channel	$c_1(t)$	$c_2(t)$	$c_3(t)$
PD	$c_1 \exp[-t/\tau_D]$	$c_2 \exp[-t/\tau_D]$	<i>c</i> ₃
GAD	$c_1 \exp[-t/2\tau_D]$	$c_2 \exp[-t/2\tau_D]$	$c_3 \exp[-t/\tau_D]$

the geometric classical correlations $C_G(t) = c_+(t)$. This sudden change occurs at times t_2^* and t_1^* for the PD and GAD channels under the necessary and sufficient conditions $c_+ > |c_3| \neq 0$ and $|c_3| > c_0 \neq 0$, respectively. This implies that the double sudden change is, for Bell diagonal states, a genuinely quantum effect, which is unattainable for C_G . Since the original entropic classical correlation C_E [3] is monotonically related with C_G within the whole class of Bell diagonal states [40], its dynamics $C_E(t)$ also presents at most one single sudden change. Moreover, as the original entropic quantum discord [2] is given by $Q_E =$ $T_E - C_E$, with the entropic total correlation T_E being a smooth function, we conclude that $Q_E(t)$ is unable to display double sudden changes.

Emergence of the pointer basis.—A quantum apparatus \mathcal{A} measuring a system \mathcal{S} suffers decoherence through contact with the environment, collapsing into a possible set of classical states known as the pointer basis [43]. The emergence of these pointer states is attributed to the instant of time τ_E at which the entropic classical correlation C_E between \mathcal{A} and \mathcal{S} becomes constant [24]. In other words, τ_E is the necessary time for the information about \mathcal{S} to be accessible to a classical observer. In particular, if the system \mathcal{AS} under decoherence is described by Eq. (4), the geometric classical correlation C_G can be used to characterize τ_E , since C_G and C_E are monotonic between themselves for Bell diagonal states. In fact, from Table II it is easily seen that $C_G(t) = c_0(t)$ becomes constant in finite time only for the PD channel at the sudden change point

$$\tau_E = \tau_D \ln[c_+/|c_3|] \qquad (c_+ > |c_3| \neq 0), \qquad (5)$$

which is in complete agreement with τ_E found in Ref. [24] by using C_E . Depending on the ratio $c_+/|c_3|$, the pointer basis can emerge at time τ_E smaller or larger than the decoherence time τ_D . On the other hand, the GAD channel does not allow for the emergence of a pointer basis at finite time.

Experimental results.—We performed two distinct experimental implementations based on NMR setups, each of them realizing different physical quantum channels. Our aim was to explore separately either the PD or GAD channels, since they are inequivalent concerning the emergence of the

TABLE III. Critical points t_1^* and t_2^* in terms of the parameters c_- , c_0 , c_+ , and τ_D . The conditions provided are necessary and sufficient for the occurrence of double sudden transitions.

Channel	t_1^*	t_2^*	Conditions
PD	$\tau_D \ln[c_0/ c_3]$	$\tau_D \ln[c_+/ c_3]$	$ c_3 = c$
GAD	$2\tau_D \ln[c_3 /c_0]$	$2\tau_D \ln[c_3 /c]$	$c_{+} \neq c_{0} \neq c_{-} \neq 0$ $ c_{3} = c_{+}$ $c_{+} \neq c_{0} \neq c_{-} \neq 0$

pointer basis. The state of a NMR two-qubit system in the high temperature approximation is given by $\rho = \frac{1}{4} \mathbb{I}_4 + \epsilon \Delta \rho$, where $\epsilon = \hbar \omega_L / 4k_B T \sim 10^{-5}$ is the ratio between the magnetic and thermal energies at room temperature, ω_L is the Larmor frequency, k_B is the Boltzmann constant, and T the temperature [44,45]. All measurements and transformations affect only the deviation matrix $\Delta \rho$, which contains the available information about the system state. The unitary operations over $\Delta \rho$ are implemented by radio frequency pulses and evolutions under spin interactions, with an excellent control of rotation angle and phase. Additionally, NMR offers very reliable quantum state tomography, which can be used to obtain a full characterization of $\Delta \rho$ [46–48]. It is worth remarking that, since in NMR experiments only the deviation matrix is detected, the calculations of the correlation matrix elements $\{c_i\}$ are done in units of ϵ .

The first set of NMR experiments were performed on a liquid state Carbon-13 enriched chloroform sample (CHCl₃) at room temperature, with the two qubits being encoded in the ¹H and ¹³C spin-1/2 nuclei, and employing a Varian 500 MHz spectrometer. The relevant relaxation times were measured as $T_1^C \approx 12.46$ s, $T_2^{C^*} \approx 0.15$ s for ¹³C, and $T_1^H \approx 7.53$ s, $T_2^{H^*} \approx 0.27$ s for ¹H. Since $T_1 \gg T_2^*$ for both qubits and the system decoherence was evaluated within a maximum evolution time of approximately 0.5 s, the relaxation mechanism is described effectively by a PD channel with $T_A = T_2^{H^*}$ and $T_B = T_2^{C^*}$. A Bell diagonal state, with the coefficients $c_1 = 0.49$, $c_2 = 0.20$, and $c_3 = 0.067$, was obtained by applying the pulse sequence illustrated in Fig. 1(a), where the pseudopure state $|00\rangle\langle 00|$ was prepared as described in Refs. [1,49]. The resulting deviation matrix was of the form $\Delta \rho = c_1 \sigma_x \otimes \sigma_x +$ $c_2\sigma_y \otimes \sigma_y + c_3\sigma_z \otimes \sigma_z$, where $c_1 = -2\cos\theta$, $c_2 =$ $-2\cos\alpha$, $c_3 = -2\cos\theta\cos\alpha$, with tunable α and θ . After the initial state preparation, the system was left to evolve for a time τ_i and quantum state tomography [46] was performed to reconstruct the evolved deviation matrix. The process was repeated for different values of τ_i in order to monitor the state relaxation. Figure 1 shows the time dependence of the quantum and classical geometric correlations as well as the individual correlation functions, calculated from the measured deviation matrices. The continuous lines correspond to theoretical predictions calculated according to the expressions in Table II. Note that Q_G clearly exhibits a double sudden transition at critical points $t_1^* = (0.107 \pm 0.006)$ s and $t_2^* = (0.198 \pm 0.006)$ s, which are compatible with the theoretical values $t_1^* = 0.105$ s and $t_2^* = 0.192$ s, as predicted by Table III. The emergence of the pointer basis in C_G is also evident at $\tau_E = t_2^*$.

The implementation of a system in which the relaxation is fully described by a GAD channel was achieved using a spin-3/2 NMR quadrupolar system. A spin-3/2 in the presence of a strong static magnetic field is described by four energy levels, which can be indexed as $|00\rangle$, $|01\rangle$, $|10\rangle$, $|11\rangle$ to represent a two-qubit system [47,50–52]. The energy separation between the levels as well as the system relaxation are dictated by the interaction between the

(a)



FIG. 1 (color online). (a) Schematic representation of the pulse sequence employed to obtain a deviation matrix in the form of a Bell diagonal state. (b) Experimentally reconstructed block diagrams for real and imaginary parts of the deviation matrix related to the Bell diagonal initial state with $c_1 = 0.49$, $c_2 = 0.20$, and $c_3 = 0.067$. The curves in (c) denote the time evolutions of quantum (Q_G , bullet) and classical (C_G , triangle) correlations, respectively. The dots represents the experimental results and the solid lines are the theoretical predictions. In the inset we detail the time evolutions of $|c_1|$ (yellow upward triangles), $|c_2|$ (blue squares), and $|c_3|$ (purple downward triangles) experimentally obtained for the PD decoherence process.

nuclear quadrupole moments with electric field gradients produced by the charge distribution in their surroundings [44]. The relaxation for this system is described by two channels: a GAD channel as in Table I and a global PD channel, acting simultaneously on both logical qubits, with Kraus operators given in Ref. [53]. The global PD channel does not act on the cross-diagonal terms of the deviation matrix, which means that for Bell diagonal states the decoherence is completely dictated by the GAD channel [53]. The experiments on the spin-3/2 system were performed with sodium nuclei in a liquid crystal sample at room temperature using a Varian 400 MHz spectrometer. The relevant relaxation times were estimated as $T_A =$ $T_B \approx 0.012$ s and the initial Bell diagonal state $[c_1 =$ $0.08, c_2 = 0.14$, and $c_3 = 0.16$, Fig. 2(a)] was prepared using a set of numerically optimized radio frequency pulses obtained by the strong modulated pulse technique [54]. Figure 2 shows the time dependence of the quantum



FIG. 2 (color online). (a) Experimentally reconstructed block diagrams for real and imaginary parts of the deviation matrix related to the Bell diagonal initial state with $c_1 = 0.08$, $c_2 = 0.14$, and $c_3 = 0.16$. The curves in (b) denote the time evolutions of quantum (Q_G , bullet) and classical (C_G , triangle) correlations, respectively. The dots represents the experimental results and the solid lines are the theoretical predictions. In the inset we detail the time evolutions of $|c_1|$ (yellow upward triangles), $|c_2|$ (blue squares), and $|c_3|$ (purple downward triangles) experimentally obtained for the GAD decoherence process.

and classical correlations as well as the correlation functions, as calculated from the measured deviation matrices following the same procedure described before. Note also here that Q_G exhibits a double sudden transition, corresponding to the crossings $|c_2(t)| = |c_3(t)|$ and $|c_1(t)| = |c_3(t)|$, at critical points $t_1^* = (0.0020 \pm 0.0005)$ s and $t_2^* = (0.0081 \pm 0.0005)$ s. According to Table III, the theoretical critical points are $t_1^* = 0.0016$ s and $t_2^* = 0.0083$ s. As expected, the presence of a single sudden change in the C_G curve is also observed, but there is no emergence of the pointer basis at finite time in this case.

Conclusions.—We performed a detailed analysis of *bona fide* geometric measures of quantum and classical correlations in paradigmatic two-qubit Bell diagonal states realized experimentally by room temperature NMR setups, in the presence of physical sources of phase damping and generalized amplitude damping noise. We observed recently predicted distinctive features of quantum correlations such as double sudden changes in their dynamics [38] and saturation of classical correlations denoting a relaxation to a pointer basis [24]. Future investigations will be devoted to assess whether the finite-time emergence of the pointer basis, established here for geometric correlations complementing the case of entropic ones [24], can be regarded as a universal feature common to all valid quantifiers of classical correlations, along the lines of Ref. [37].

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week ending 20 DECEMBER 2013

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