On the quantumness of correlations in nuclear magnetic resonance

BY D. O. SOARES-PINTO1, R. AUCCASE2, J. MAZIERO3, A. GAVINI-VIANA4, R. M. SERRA3 AND L. C. CÉLERI3,*

1Instituto de Física de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970 São Carlos, São Paulo, Brazil
2Empresa Brasileira de Pesquisa Agropecuária, Rua Jardim Botânico 1024, 22460-000 Rio de Janeiro, Rio de Janeiro, Brazil
3Centro de Ciências Naturais e Humanas, Universidade Federal do ABC, Rua Santa Adélia 166, 09210-170 Santo André, São Paulo, Brazil
4Centro Brasileiro de Pesquisas Físicas, Rua Dr Xavier Sigaud 150, 22290-180 Rio de Janeiro, Rio de Janeiro, Brazil

Nuclear magnetic resonance (NMR) was successfully employed to test several protocols and ideas in quantum information science. In most of these implementations, the existence of entanglement was ruled out. This fact introduced concerns and questions about the quantum nature of such bench tests. In this paper, we address some issues related to the non-classical aspects of NMR systems. We discuss some experiments where the quantum aspects of this system are supported by quantum correlations of separable states. Such quantumness, beyond the entanglement–separability paradigm, is revealed via a departure between the quantum and the classical versions of information theory. In this scenario, the concept of quantum discord seems to play an important role. We also present an experimental implementation of an analogue of the single-photon Mach–Zehnder interferometer employing two nuclear spins to encode the interferometric paths. This experiment illustrates how non-classical correlations of separable states may be used to simulate quantum dynamics. The results obtained are completely equivalent to the optical scenario, where entanglement (between two field modes) may be present.

1. Introduction

It is widely accepted that nuclear magnetic resonance (NMR) is an experimental technique able to implement an ensemble quantum information processor using nuclear spins. From the creation of NMR pseudo-pure states, several quantum protocols for quantum information processing (QIP) were implemented in such physical systems [1,2]. NMR has also made possible the experimental evaluation of abstract theoretical proposals in quantum information science as well as tests of principle in the foundations of quantum mechanics [3], with a precision rarely obtained through other experimental techniques. Thus, NMR has become a

*Author for correspondence (lucas@chibebe.org).

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powerful experimental approach to problems related to QIP, even though it has been criticized due to the absence of entanglement in most of the systems used up to now [1], besides the scalability issues [2].

The non-existence of entangled states became a problem when the necessity of such a resource was conjectured to make it possible to obtain the advantages of QIP [4]. In this sense, as the states used to implement quantum protocols in NMR, named pseudo-pure states, do not present entanglement except in singular situations, a question about the quantum nature of these implementations was raised [5]. It was also shown, for a large class of protocols, that the existence of entangled states is a necessary (but not a sufficient) condition for quantum computers to present an exponential speed-up over the classical ones [6], although too much entanglement could also be harmful [7–9]. However, there are other important characteristics, such as the effectiveness of the implementation and the manipulation of quantum states, which also play some role in the quantum advantage game. Thus, considering that the principal aspect of NMR is the excellent control of unitary transformations promoted by radiofrequency (RF) pulses, such a spectroscopic technique allows one to obtain effective and singular methods in order to manipulate quantum states to implement QIP protocols. This is part of the reason for the success of NMR QIP experiments.

Notwithstanding, as suggested in earlier studies [5,10–13], the presence of quantum correlations other than entanglement offers additional motivations for successful NMR QIP implementations, although, until now, they have not promoted the exponential speed-up of information processing. It is possible to measure such non-classical correlations in a bipartite system using a quantity called quantum discord [14,15] (for recent reviews about quantum aspects of correlations beyond the entanglement–separability paradigm, see [16,17]). One interesting feature of such correlations is that some mixed-separable quantum states, or non-entangled states, can present non-null discord, indicating the existence of non-classical correlations beyond the entanglement–separability paradigm. These correlations may have a significant role in quantum information science owing to the quantum advantage, when compared with its classical analogues, obtained by protocols based on non-entangled states [10]. These concepts allowed us to study experimentally and theoretically the existence of such correlations in NMR systems, as well as the effects of decoherence (usually modelled as amplitude- and phase-damping channels) over the system’s non-classicality [11–13,18]. Such a general kind of non-classical correlation is a source of quantumness (or a quantum resource) available in room-temperature NMR experiments.

In this paper, we present an NMR implementation of an analogue of the well-known single-photon Mach–Zehnder (MZ) interferometer employing two nuclear spins to encode the interferometric paths. The MZ interferometer is an important and very useful tool for applications in quantum optics as well as for fundamental tests of quantum mechanics [19]. Such a device enables us to determine the phase shifts between two paths mediated by two half-silvered mirrors, called beam splitters (BSs). At the single-particle level, wave interference between the probability amplitudes in the two arms of the interferometer plays a crucial role in phase determination. It is worth while to note that it was believed that this interferometer, at the single-quantum level, needed entanglement (between the field modes in both paths of the interferometer) to work properly—in other words,
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for separable input states of the field modes after the first BS—and the output should also be classical. In this case, there is no interference between the two paths of a single quantum. Experimental implementations of interferometry in NMR have already been reported in the literature [3,20,21]. However, such experiments were realized using only one nucleus (one spin-$\frac{1}{2}$ particle) to encode the two interferometric paths. Here, we present an analogue of an MZ interferometer by means of two nuclei that encodes the path information taken by a single quantum. This has some advantages, as discussed later, but our aim here is mainly to discuss the root of the non-classical aspects of NMR systems, as well as to give one more indication of the potential of such a system as a quantum simulator.

The paper is organized as follows. In §2, we describe how one can quantify and measure quantum and classical correlations in highly mixed states in an NMR setup. Section 3 is dedicated to the detection, but not quantification, of quantum correlations. The intention of these two sections is to emphasize the quantum nature of NMR systems, because one needs a quantum system to efficiently simulate another quantum system. The analogue of the single-photon MZ interferometer is implemented in §4. Finally, in §5, we present our final discussions.

2. Measuring quantum and classical correlations in nuclear magnetic resonance

For a typical liquid-state NMR system at room temperature, the gap between the Zeeman energy levels of the nuclei are much smaller than the average thermal energy, $\epsilon = \hbar \omega_L/2nk_B T \sim 10^{-5}$ (with $\omega_L$ being the Larmor frequency), which implies that the density matrix can be written as the expansion [1,22]

$$\rho \simeq \frac{1}{2^n} \mathbb{I} + \epsilon \Delta \rho,$$  \hspace{1cm} (2.1)

where $\mathbb{I}$ is the $2^n \times 2^n$ identity matrix, $n$ is the number of qubits (encoded in spin nuclei) and $\Delta \rho$ is the traceless deviation matrix. This is the well-known high-temperature approximation.

Any manipulation in the earlier-mentioned state, such as state preparation, quantum state tomography, qubit rotations and so on, is performed only over the deviation matrix $\Delta \rho$. This is because such manipulations are, generally, sequences of RF pulses, which are basically unitary transformations, $U$, that act on the density matrix in the following way:

$$U \rho U^\dagger = \frac{1}{2^n} \mathbb{I} + \epsilon U \Delta \rho U^\dagger.$$  \hspace{1cm} (2.2)

By adjusting each pulse length, phase and amplitude, it is possible to obtain very fine control over the density matrix populations and coherences (diagonal and off-diagonal elements of the density matrix, respectively, usually defined in the computational basis). Together with proper temporal or spatial averaging procedures and evolution under spin interactions [23], the RF pulse can be specially designed to prepare any two-qubit computational base states, as well as their superpositions, starting from the thermal equilibrium state [2,24–26]. Although it is possible, in principle, to create any quantum state by means of

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these techniques, it was shown that the obtained state, in general, is a highly mixed one that does not possess entanglement since $\epsilon \sim 10^{-5}$, thus violating the boundary $\epsilon \geq 1/(1 + 2^{n-1})$ for the existence of such correlations [4,6].

Nevertheless, such separable states can present other kinds of non-classical correlations that are conjectured to play a relevant role in QIP protocols [10,27,28]. The total correlation contained in a state $\rho_{AB}$ is quantified by the quantum mutual information [29–31]

$$I(A:B) = S(A) + S(B) - S(AB),$$

with $S(X) \equiv S(\rho_X) = -\text{Tr} \rho_X \ln \rho_X$ being the von Neumann entropy and $\rho_A = \text{Tr}_B \rho_{AB}$ being the reduced density operator for subsystem $A$, with an equivalent definition for partition $B$. This is a direct generalization of the classical mutual information, introduced by Shannon to quantify correlations in classical information theory, $I(A:B) = H(A) + H(B) - H(AB)$, with $H(A) = -\sum_k p^A_k \ln p^A_k$ being the Shannon entropy of the probability distribution $\{p^A_k\}$ for the random variable $A$ [32]. In the classical domain, we can rewrite this expression in the equivalent form $J(A:B) = H(A) - H(A|B)$, with $H(A|B)$ being the knowledge we can get from a random variable $A$ when we have measured $B$, i.e. the conditional entropy. This is the point where the quantum and the classical domains break up. While a classical measure does not disturb the system, a quantum one generally does. Therefore, the quantum extension of $J(A:B)$ is not straightforward, but a possible one was considered in Ollivier & Zurek [14] as

$$J(A:B) = S(\rho_A) - S_{\{\Pi^B_j\}}(\rho_{A|B}),$$

with $S_{\{\Pi^B_j\}}(\rho_{A|B}) = \sum_j q_j S(\rho^A_{j})$ being a quantum extension of the classical conditional entropy $\mathcal{H}(A|B)$. Here $\{\Pi^B_j\}$ is a complete set of projective measurements on partition $B$ and $\rho^A_{j} = \text{Tr}_B(\rho_{AB} 1_A \otimes \Pi^B_j)/p_j$ ($p_j = \text{Tr}[1_A \otimes \Pi^B_j \rho_{AB}]$) is the measured reduced density matrix of partition $A$. The difference

$$D(A:B) = I(A:B) - \max_{\{\Pi^B_j\}} J(A:B)$$

was called the quantum discord and is a measure of the quantumness of correlations [14,16,17], because the expressions for $I(A:B)$ and $J(A:B)$ are classically equivalent. We note that the second term on the right-hand side of equation (2.5) can be regarded as a measure of the classical correlations contained in the state $\rho_{AB}$ [15].

Let us go back to the NMR system, where all correlations in the quantum state $\rho_{AB}$ come from the deviation matrix $\Delta \rho_{AB}$. In this context, it is desirable to express the correlation quantifiers as functions of $\Delta \rho_{AB}$. In order to do this, we expand the von Neumann entropy in powers of the parameter $\epsilon$ as

$$S(\rho) = 2 \left(1 - \frac{\epsilon^2}{\ln 2} \text{Tr} \Delta \rho^2\right) + \cdots,$$

where we have used $\text{Tr} \Delta \rho = 0$. As the reduced density operators read $\rho_{A(B)} = \text{Tr}_{B(A)} \rho = 1_{A(B)}/2 + \epsilon \Delta \rho_{A(B)}$, with $\Delta \rho_{A(B)} = \text{Tr}_{B(A)} \Delta \rho_{A(B)}$ being the reduced

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deviation matrix, the marginal entropies become

$$S(\rho_{A(B)}) = 1 - \frac{\epsilon^2}{\ln 2} \text{Tr} \Delta \rho_{A(B)}^2 + \cdots. \quad (2.7)$$

Substituting equations (2.6) and (2.7) into equation (2.3) and keeping terms up to second order, we obtain

$$I(\rho) \simeq \frac{\epsilon^2}{\ln 2} (2 \text{Tr}(\Delta \rho)^2 - \text{Tr}(\Delta \rho_A)^2 - \text{Tr}(\Delta \rho_B)^2), \quad (2.8)$$

which is the desired expression for total correlations.

To quantify the classical correlations, we must obtain the measured density operator, which is given by (in a symmetric form) [16,17]

$$\eta = \sum_{i,j} (\Pi_i^A \otimes \Pi_j^B) \rho (\Pi_i^A \otimes \Pi_j^B)$$

$$\equiv \frac{1}{4} + \epsilon \Delta \eta, \quad (2.9)$$

where we have defined the measured deviation matrix as

$$\Delta \eta \equiv \sum_{i,j} (\Pi_i^A \otimes \Pi_j^B) \Delta \rho (\Pi_i^A \otimes \Pi_j^B).$$

Note that we have applied the projection operators on both partitions $A$ and $B$. This is a symmetric version of the classical correlations defined in equation (2.5), and from here on we will adopt such a definition to quantify correlations (see [16,17] for details and discussions about non-classical correlation quantifiers).

Following the reasoning that led us to equation (2.8), we obtain the following expression for the mutual information of the measured state:

$$J(\eta) \simeq \frac{\epsilon^2}{\ln 2} \{2 \text{Tr}(\Delta \eta)^2 - \text{Tr}(\Delta \eta_A)^2 - \text{Tr}(\Delta \eta_B)^2\}, \quad (2.10)$$

and thus for the classical correlation

$$C(\rho) \simeq \max_{\{\Pi_i^A \otimes \Pi_j^B\}} J(\eta), \quad (2.11)$$

where $\Delta \eta_{A(B)} = \text{Tr}_{B(A)} \Delta \eta$. The quantum correlation in the composed two-qubit system can be directly computed from equations (2.8) and (2.11) as [11]

$$Q(\rho) = I(\rho) - C(\rho). \quad (2.12)$$

Equations (2.11) and (2.12) are general and can be used to quantify quantum and classical correlations in every system whose density matrix can be cast in the form given by equation (2.1). Specifically, for NMR systems, we have employed such expansions to study the relaxation dynamics of correlations in two different systems: (i) a sodium dodecyl sulfate (SDS) liquid-crystal sample containing a $^{23}\text{Na}$ quadrupolar nucleus with spin $I = \frac{3}{2}$ [11] and (ii) a liquid sample of $^{13}\text{C}$-enriched chloroform, where we have two spin-$\frac{1}{2}$ systems ($^1\text{H}$ and $^{13}\text{C}$ nuclei) [12]. We can use system (i) to encode two logical qubits, while system (ii) encodes two

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Figure 1. Relaxation dynamics of correlations. The blue circles are the mutual information while the red squares and green triangles represent classical and quantum correlations, respectively. In (a), we consider two logical qubits encoded in the SDS molecule under the action of the same phase-damping environment, while (b) shows the dynamics of the two physical qubits of the chloroform molecule. The insets show a schematic diagram of the molecular structure of the two systems. In both cases, we have prepared the same initial state. For details see [11,12]. (Online version in colour.)

physical qubits. It is worth while to note that, during the decoherence dynamics, the amplitude-damping channel (which describes energy loss) acts individually on each qubit in both systems, while the phase-damping channel (responsible for coherence loss) is a global channel in case (i) and it acts independently over each qubit in case (ii) [18,33]. This difference allowed us to detect, in system (ii), a peculiar behaviour of the decoherence dynamics of quantum discord, i.e. a sudden change in its decay rate [12] (theoretically predicted in [34]).

These results reveal two interesting aspects of NMR systems. First, there are quantum correlations in these systems, which can be quantified, for instance, by the quantum discord. Second, because the relaxation mechanisms present distinct features in both systems, they provide a very interesting platform to investigate the differences introduced in the dynamics by a global and a local environment. An example of this difference is illustrated in figure 1, where the relaxation dynamics of the correlations between two qubits is shown. While, in the case of a global reservoir (figure 1a), we have the expected exponential decay of all kinds of correlations [11], the sudden-change behaviour clearly takes place in the case of independent reservoirs [12], as can be seen in figure 1b (see the caption of the figure for details). Moreover, it is quite remarkable that sudden change still takes place even in the presence of the amplitude-damping channel, indicating that such a phenomenon could be a strong characteristic of the correlations.

3. Witnessing the quantumness of correlations in nuclear magnetic resonance

In §2, we discussed the quantification of quantum and classical correlations in composite systems. This is very expensive information because a full quantum
Quantumness of correlations in NMR state tomography, beyond the numerical optimization process to compute equation (2.11), must be performed. However, there are many situations in which we do not need to know how much correlation a certain state possesses; instead, it is enough to know only its nature. In other words, we just have to distinguish between classical and quantum correlations. In the same spirit as what happens to entanglement, some non-classical correlation criteria and observable witnesses were proposed in [35–40] and experimentally verified in [13,41,42]. A classicality (or non-classicality) witness is regarded as an observable (or a set of observables) that can be directly measured in an experimental setup. Depending on its expected value, we know whether the state has quantum or only classical correlations. The measurement of such a witness in an NMR system revealed directly the non-classical nature of its highly mixed state [13,41].

In what follows, we describe a non-classicality witness and its experimental measurement performed in an NMR setup at room temperature [13]. In the study of Maziero & Serra [40], it was shown that the mean value of the operator

$$W_{\rho} = \sum_{i=1}^{3} \sum_{j=i+1}^{4} |\langle O_i \rangle_\rho \langle O_j \rangle_\rho| = 0$$

(3.1)

is a sufficient condition for classicality of correlations for a wide class of two-qubit systems, expressed by

$$\rho = \frac{I^{AB}}{4} + \frac{1}{4} \sum_{i=1}^{3} (A_i \sigma_i^A \otimes I^B + B_i I^A \otimes \sigma_i^B + C_i \sigma_i^A \otimes \sigma_i^B).$$

(3.2)

Here $O_i = \sigma_i^A \otimes \sigma_i^B$ for $i = 1, 2, 3$; $O_4 = \sum_{i=1}^{3} (z_i \sigma_i^A \otimes I^B + w_i I^A \otimes \sigma_i^B)$; $\sigma_i^{A(B)}$ is the $i$th Pauli operator acting on subsystem $A(B)$; and $A_i, B_i, C_i, z_i, w_i \in \mathbb{R}$, with $z_i$ and $w_i$ randomly chosen and constrained such that $\sum_i z_i^2 = \sum_i w_i^2 = 1$.

In the case of the Bell-diagonal class of states

$$\rho_{bd} = \frac{I^{AB}}{4} + \frac{1}{4} \sum_{i=1}^{3} C_i \sigma_i^A \otimes \sigma_i^B,$$

(3.3)

$W_{\rho_{bd}} = 0$ is also a necessary condition for the absence of quantumness in the correlations of the composite system (for this case $\langle O_4 \rangle_{\rho_{bd}} = 0$ [40]).

One interesting fact about such a witness is that it is possible to rewrite the observables in equation (3.1) in terms of one component of the magnetization in one subsystem as $\langle O_i \rangle_\rho = \langle \sigma_1^A \otimes I^B \rangle_\xi_i$, with $\xi_i = U_{A \rightarrow B} [R_{n_i}(\theta_i) \rho R_{n_i}^\dagger(\theta_i)] U_{A \rightarrow B}^\dagger$, where $R_{n_i}(\theta_i) = R_{n_i}^A(\theta_i) \otimes R_{n_i}^B(\theta_i)$, $R_{n_i}^{A(B)}(\theta_i)$ being a local rotation by an angle $\theta_i$ around direction $n_i$ on subsystem $A(B)$, where $\theta_1 = 0$, $\theta_2 = \theta_3 = \pi/2$, $n_2 = y$, $n_3 = z$, and $U_{A \rightarrow B}$ is the CNOT gate, with subsystem $A$ being the control qubit. This fact leads to a straightforward implementation of this witness in the NMR scenario, because the one-qubit magnetizations are the natural observables for these systems. In fact, the witness in equation (3.1) was experimentally implemented in a room-temperature NMR two-qubit system [13], directly revealing the non-classical aspects of highly mixed states.
4. Mach–Zehnder interferometer

An interesting way to test non-classicality in NMR systems can be provided by an analogue of the well-known single-photon MZ interferometer. In order to perform such an interferometer and test the role of correlations, we employ two nuclei to encode the two-interferometric-path information. As already mentioned, this approach differs significantly from the previous interferometric measures implemented in the NMR scenario [3,20,21], in which the two-path information is encoded in just one nuclear spin. Moreover, because distinct nuclei have, generally, distinct relaxation times, it becomes possible to study the environment-induced phase shift between the two paths. This environment-induced phase shift may find applications, for example, in thermometry [43] and in the quantum illumination protocol [44].

Now, let us briefly review the single-photon MZ interferometer in the optical scenario, schematically shown in figure 2. In this figure, BS is a 50:50 beam splitter (one half-silvered mirror), M is a mirror, and $D_A$ and $D_B$ are one-photon sensitive detectors. The phase difference $\phi$ between paths $A$ and $B$ can be due to an environment-induced phase or just a controlled one. In what follows, the indices $A$ and $B$ must be understood as two spatial field modes (referring to distinct paths taken by the photon).

Let us suppose that the input (pure) state (before the first BS in figure 2) is given by

$$|\Psi_i\rangle = |0\rangle_A \otimes |1\rangle_B.$$  \hspace{1cm} (4.1)

The state after the first BS is

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|0\rangle_A \otimes |1\rangle_B + i|1\rangle_A \otimes |0\rangle_B).$$  \hspace{1cm} (4.2)

Note that this can be seen as an entangled state of the two field modes (that encode path information). The coherent superposition of both paths after the first
BS is essential for the interferometer to produce an interference pattern [19]. In our experiment, the information about both paths will be encoded in two distinct nuclei and, when talking about correlations between these nuclei, we mean the coherent superposition paths $A$ and $B$.

The final state, after the second BS (figure 2), will be given by

$$|\Psi_f \rangle = \cos \left( \frac{\phi}{2} \right) |0\rangle_A \otimes |1\rangle_B - \sin \left( \frac{\phi}{2} \right) |1\rangle_A \otimes |0\rangle_B. \quad (4.3)$$

From here on we will omit the tensor product symbol to simplify notation. Equation (4.3) shows the interference pattern between the two paths and reflects the wave-like behaviour of the one-photon state. The probability to detect the photon in the detector $D_B$ (the probability of detecting the initial state at the end of the interferometer) is given by $\cos^2(\phi/2)$. If we introduce a non-destructive detector in any of the paths before the second BS, this interference pattern disappears and we observe the particle character of the quantum system. This is one of the most well-known representations of Bohr’s complementarity principle. It is interesting to note that we could modify the present scheme to study this principle quantitatively by means of an inequality relating the which-way information (particle) and the fringe visibility of the interferometer (wave), as suggested in [45].

We implement such an interferometer by employing a two-qubit system comprising nuclear spins of $^1\text{H}$ and $^{13}\text{C}$ atoms in a $^{13}\text{C}$-enriched chloroform molecule (CHCl$_3$). The sample was prepared by mixing 50 mg of 99 per cent $^{13}\text{C}$-labelled CHCl$_3$ in 0.7 ml of 99.8 per cent CDCl$_3$ in a 5 mm NMR tube. Both samples were provided by Cambridge Isotope Laboratories Inc. The experiments were performed at 25°C using a Varian 500 MHz Premium Shielded ($^1\text{H}$ frequency), at the Brazilian Centre for Physics Research. A Varian 5 mm double resonance probe-head equipped with a magnetic field gradient coil was used.

In our NMR version of the MZ interferometer, we have exactly the same situation as in the optical standard version. The two paths are encoded in two different nuclei, with just one quantum of excitation. The nuclear spin in the ground state represents the vacuum field state, while the nuclear spin in the excited state represents the field excitation (the one-photon state). We choose the $^{13}\text{C}$ nucleus to encode the path in which we apply the controlled phase (path $A$ in figure 2), while the $^1\text{H}$ nucleus encodes the reference path (path $B$ in figure 2). This choice was motivated by the fact that the carbon nucleus has the smaller transverse relaxation time (see [12,13] for details about the relaxation times for our system). It is not important for the present experiment, but can be explored by experiments studying the effects of decoherence in just one path, like the thermometry mentioned earlier [43]. Here, the phase difference will be provided by a controlled $z$-rotation applied to the carbon nucleus.

The pulse sequence used to implement the analogue of the MZ interferometer is shown in figure 3. The first block of this figure is used to prepare the deviation matrix in a state analogous to $|\Psi\rangle = |0\rangle|1\rangle$, which means that the hydrogen nucleus is in the ground state while carbon is in its excited state. Some words about the meaning of this analogy are in order. In §2, we have mentioned that the spin state is represented by the highly mixed density operator shown in equation (2.1). This happens because the ratio of the magnetic energy to the
Figure 3. Pulse sequence for the implementation of the analogue of a single-photon MZ interferometer in a two-nuclei NMR system. The thicker filled bars represent $\pi/2$ pulses, the thinner bars indicate $\pi/4$ pulses and the grey bars indicate $\pi/6$ pulses with the phases as shown (negative pulse phases are indicated by a bar over the symbol). $\Delta = 1/J$ and the time periods in the state preparation represent free evolutions under the $J$ coupling [1]. (Online version in colour.)

thermal energy is $\epsilon \sim 10^{-5}$; so the density operator was decomposed into two contributions: the white noise term and the deviation matrix term. Recalling that the control over the system is given by RF pulses, which are represented by unitary operations (besides gradient pulses), we only act on the deviation matrix term (second term in equation (2.1)). We then encode all the information of the state $|\psi\rangle$ in this matrix. At the end, we will have a quantum dynamics equivalent to that of a pure state.

After the state preparation, there are three consecutive blocks that are used to implement the MZ interferometer. The first and the third ones play the role of the BSs. We have constructed a sequence of 128 pulses with modulated frequencies and amplitudes by means of the GRAPE optimization method [46] to implement the following action on both qubits:

$$\text{BS} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & i & 0 \\ 0 & i & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix},$$

which represents the BS operation. The time duration of the entire sequence in order to implement such an operation is 4.5 ms. Finally, the three pulses between the BSs implement a rotation on the carbon nucleus about the $z$-axis by an angle $\phi$. After the pulse sequence depicted in figure 3, we performed a full quantum state tomography. We repeated the protocol in figure 3 for different phase shifts.

The experimental results are presented in figure 4, where we plot the probability of detecting the initial state $P_i = |\langle \psi_i | \psi_j \rangle|^2$ as a function of the phase shift $\phi$. The theoretical predictions from equations (4.1) and (4.3) are in good agreement with the experimental results, showing that the interference expected for the interferometer can be obtained in the context of the highly mixed states NMR system. It is remarkable that the tiny coherence present in our experiment (of order $\epsilon$) enables the interferometer to work properly. The visibility of the

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interferometer (which quantifies the contrast of the interference in any system that has wave-like character) is near to unity, without entanglement between the two paths (the two nuclear spins in this case). We may argue that the reason for this is the quantum correlation of separable states (quantified by the quantum discord) present in the composite nuclear spin system.

The small deviation between the observed and the expected probability in figure 4 is mainly due to the fact that we are continuously applying an RF pulse on the carbon spin to obtain the controlled phase shift between the two paths. This pulse sequence presents some fluctuations that introduce noise into the system. Moreover, the pulse is not exactly in resonance with the qubit transition frequency, which contributes to the phase mismatch between theory and experiment. We observe that there are two decoherence processes involved here. The first one is that caused by the usual amplitude- and phase-damping channels, which are always present, but that have a negligible effect for the current experiment due to the fact that it occurs in a time much shorter than the characteristic relaxation times of the system. All the observed decoherence is due to the imperfections of the RF pulses employed to generate the phase difference $\phi$ between the two paths.

To give some support to our statement that quantum correlations of separable states are responsible for the successful simulation of the single-photon interferometer, in figure 5 we plot the visibility and the quantum discord between the two qubits (paths) in a slightly modified experiment, where we vary the amount of quantum discord after the first BS operation in the pulse sequence of figure 3. To obtain this result, we performed two experiments. In the first one, we allow the system to evolve freely (under the natural decoherence) during a certain period of time $\tau$ just after applying the first BS operation and then we perform a full quantum state tomography. From the obtained deviation matrix, we compute the quantum discord through equation (2.12). By varying the time $\tau$ of the decoherent evolution, we obtain different amounts of quantum discord, and
Figure 5. Quantum discord (red squares) and visibility (blue circles) of the MZ interferometer for different decoherence periods $t$. Note that, although both quantities are represented on the same vertical axis, they are completely different quantities. While the visibility is a dimensionless quantity, the quantum discord is measured in units of $\epsilon^2 / \ln 2$ bit. The purpose of the graph is only to show that the fade of the interference pattern is accompanied by the vanishing of the quantum discord between the two qubits (paths of the interferometer). (Online version in colour.)

the results are shown by the red squares in figure 5. In the second experiment, we again, after the first BS, allow the system to evolve freely under the action of the environment. However, instead of performing the state tomography, we proceed with the sequence shown in figure 3 and compute the fringe visibility of the interferometer,

$$V = \frac{\max \langle \Psi_f | \hat{D}_A | \Psi_f \rangle - \min \langle \Psi_f | \hat{D}_B | \Psi_f \rangle}{\max \langle \Psi_f | \hat{D}_A | \Psi_f \rangle + \min \langle \Psi_f | \hat{D}_B | \Psi_f \rangle},$$  

with $\hat{D}_A$ and $\hat{D}_B$ being the detector operators. As we can see from figure 5, the decay of the discord is clearly accompanied by the decay in the visibility, showing that, without discord, we cannot obtain a visible interference pattern from the simulated MZ interferometer.

5. Final discussions

QIP either for computing or for communication will probably lead to a technological revolution in this century. Having appeared in the early 1980s, quantum computation and quantum information evolved faster as an area of basic research in physics and as a promising technological alternative. In fundamental physics, the concepts of quantum information have stimulated a huge number of new ideas and results, which have deepened our knowledge about the nature of quantum behaviour. Particularly exciting are the relations of quantum correlations and natural resources for computation and communication. The discovery of quantum algorithms, new forms of communication and cryptography led to a remarkable experimental development in order to test these applications.
Up to now, almost all quantum algorithms and communication protocols have been tested on systems with a few qubits. However, for large-scale applications of quantum devices, it is necessary to study the generation, handling and storage of quantum states in such systems. We may say that the experimental technique that has stood out in this context is NMR, because it allows a quite precise manipulation of quantum states. This precise control of spin states is due to the RF technology developed over decades. It has been used in an inventive way for NMR methods and contributed to the impressive and fast success of the technique in QIP [1,2]. NMR also allows, through the manipulation of nuclear spin qubits, the simulation of more complex systems [47–51].

Despite the successful use of NMR in QIP, some years ago the impossibility of the existence of entangled states in such an experimental technique at room temperature was demonstrated. By that time, it was supposed that this kind of quantum correlation was intrinsically linked with the speed-up of the quantum algorithms. Thus, one question could be raised: What is the quantum aspect of NMR states that allows quantum dynamics and its application in QIP? This question is partially answered by the existence of quantum correlations in separable states, as revealed, for example, by the quantum discord. As we have argued in this paper, such correlations seem to occur naturally in room-temperature NMR systems, as far as can be easily quantified or witnessed by the methods discussed here, and are responsible for the quantumness of the system. The analogous single-photon MZ interferometer presented in the last section provides a clear illustration of the non-classicality present in NMR highly mixed states. The coherence present in the deviation matrix allows for a path interference analogous to that obtained in the optical context where an entangled state (between two field modes) is present. In other words, we may argue that the separable state (non-entangled) quantum correlations present in the two-qubit NMR state are the resource that enables the interferometer to work properly with a visibility near to unity.

In summary, the phase coherence between two nuclear spins in the NMR highly mixed states can encompass non-classical correlations (as measured by the quantum discord). Such quantum correlations are available for performing quantum dynamics (path interference), allowing the simulation of several systems, such as, for instance, the single-photon MZ interferometer. The discussions presented here may open the way for new and very exciting tests of the quantum aspects of nature and of protocols in information science that exploit this kind of non-classicality, which is present even in the NMR highly mixed states scenario.

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References

2 Jones, J. A. 2011 Quantum computing with NMR. Prog. NMR Spectrosc. 59, 91–120. (doi:10.1016/j.pnmrs.2010.11.001)
7 Gross, D., Flammia, S. T. & Eisert, J. 2009 Most quantum states are too entangled to be useful as computational resources. Phys. Rev. Lett. 102, 190501. (doi:10.1103/PhysRevLett.102.190501)
Quantumness of correlations in NMR


