NMR analog of Bell's inequalities violation test

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Abstract. In this paper, we present an analog of Bell's inequalities violation test for *N* qubits to be performed in a nuclear magnetic resonance (NMR) quantum computer. This can be used to simulate or predict the results for different Bell's inequality tests, with distinct configurations and a larger number of qubits. To demonstrate our scheme, we implemented a simulation of the violation of the Clauser, Horne, Shimony and Holt (CHSH) inequality using a two-qubit NMR system and compared the results to those of a photon experiment. The experimental results are well described by the quantum mechanics theory and a local realistic hidden variables model (LRHVM) that was specifically developed for NMR. That is why we refer to this experiment as a *simulation* of Bell's inequality violation. Our result shows explicitly how the two theories can be compatible with each other due to the detection loophole. In the last part of this work, we discuss the possibility of testing some fundamental features of quantum mechanics using NMR with highly polarized spins, where a strong discrepancy between quantum mechanics and hidden variables models can be expected.

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Contents

1. Introduction

Since the birth of quantum mechanics, interesting questions have been raised, some of them remaining not completely understood. One of the most amazing ones concerns the EPR paradox, raised by Einstein *et al* [1]. In that work, the authors stated that the quantum mechanics theory is not complete, since it does not contain what they called 'elements of reality'. The EPR correlations, which exist in the so-called entangled states, have no dependence on distance, which initially led to the wrong conclusion that they would violate the theory of relativity. One attempt to overcome the strange features of entangled states is to postulate the existence of some supplementary variables outside the scope of quantum mechanics, called 'hidden variables' [2]. A hidden variables model is supposed to reproduce all the quantum mechanical predictions.

However, in 1964 John Bell [3] discovered a conflict between quantum mechanics and the hidden variables theory. Mathematically, this conflict takes the form of a set of inequalities (called Bell's inequalities), which can be violated by entangled states, but are never violated by non-correlated quantum states or classical 'objects'. Recently, there has been increasing interest in the subject of Bell's inequalities, not only to test local realism in quantum mechanics in a variety of contexts, but also because of their connection to quantum communication [4]–[6] and quantum cryptography [7, 8]. Furthermore, Bell's inequalities can be a useful tool to detect entanglement, which is found to be a powerful computational resource in quantum computation [9].

Violation of Bell's inequalities has been verified in various experiments [10]–[18]. The recent developments in the field of nuclear magnetic resonance quantum information processing (NMR-QIP) have shown that NMR is a valuable testing tool for new ideas in quantum information science (for recent reviews, see [19]–[22]). NMR experiments with as many as 12 qubits have been reported [23, 24]. More than 50 years of development has put NMR in a unique position to perform complex experiments, sometimes quoted as 'spin choreography' [25]. Particularly fruitful has been the use of NMR-QIP to simulate quantum systems [22].

In this work, we use an NMR system to simulate a quantum optics experiment. We built a scheme to simulate the violation of Bell's inequalities for N qubits [26, 27], and tested it in the violation of the Clauser, Horne, Shimony and Holt (CHSH) inequality [28] using a two-qubit NMR system. The experimental results were compared to the quantum mechanical theoretical predictions and also to a local realistic hidden variables model (LRHVM), built to explain the correlations observed in NMR experiments [29]. We found that both theories are consistent with our experiment and that is why we refer to the experiment as a *simulation*. The consistency between both theories can be understood by the fact that NMR can detect only a small fraction

of spins due to small polarization at room temperature, a situation that resembles the so-called detection loophole [30].

It is important to stress that the NMR qubits are nuclear spins of atoms bounded together in a single molecule, separated by a few angstroms. Therefore, an NMR experiment is inherently local and cannot be used to prove non-local effects. Furthermore, most NMR-QIP experiments are performed at room temperature in a macroscopic liquid sample containing a large number of molecules, each of them working as an independent 'quantum information processing unit'. In the NMR context, the ensemble of spins constitutes a highly mixed state and their density matrix is not entangled, as demonstrated by Braunstein *et al* [31]. Therefore, our work neither provides an experimental procedure to prove or disprove non-local effects nor reveals entanglement in NMR experiments at room temperature. However, it does provide a way to simulate tests for different Bell's inequalities. The comparison between our experiment and a true quantum optics experiment shows the fidelity of the simulation. Besides, our scheme can be applied to a highly polarized spin ensemble [32]. In this case, true entangled states can be achieved and a contradiction between hidden variables models and quantum theory could be detected.

2. Bell's inequalities and NMR

In this work, we refer to a generalization of the Bell's inequalities for N qubits developed in [26, 27, 33]. It involves the measurement of a set of correlation functions, for which each one of N observers can choose one of the M observables and measurements can yield only two possible values: $s = \pm 1$. Hence, M^N correlation functions, labeled $E(n_1, \ldots, n_N)$, can be constructed, where the index n_i runs from $n_i = 1, \ldots, M$ and denotes the settings of the ith observer. For the NMR case, these observables are projections of the 1/2 nuclear spins along a particular direction labeled n_i. Taking into account the measurement of these observables, it is possible to build different Bell's inequalities, each of them exhibiting contradictions with LRHVM's predictions for some entangled states.

A general expression for the Bell's inequalities can be written as [33]:

$$-L \leqslant \sum_{n_1,\ldots,n_N=1}^{M} C(n_1, \ldots, n_N) E(n_1, \ldots, n_N) \leqslant +L, \tag{1}$$

where $C(n_1, \ldots, n_N)$ are real coefficients, L is some limit imposed by local realism and the correlation functions are given by:

$$E(n_1, \ldots, n_N) = \sum_{s_1,\ldots,s_N=\pm 1} \left(\prod_{j}^{N} s_j \right) P(s_1, \ldots, s_N), \tag{2}$$

where $P(s_1, \ldots, s_N)$ is the probability of the first observer finding the outcome s_1, the second s_2 and so on. In a standard experiment, a set of N correlated particles is prepared in a pure entangled state, and their spin projection onto M different directions is measured by different observers. After a large number of runs, the observers compare their results in order to obtain the probabilities shown in (2) and to verify whether the inequality (1) was violated or not.

At room temperature, NMR experiments are described by density matrices of the kind $\rho_{\text{eq}} \approx (\hat{I} - \beta H)/2^N$, where β is the Boltzmann factor, H is the internal Hamiltonian of the spin system and $\hat{I}$ is the $2^N \otimes 2^N$ identity matrix. In NMR experiments, only the deviation of the
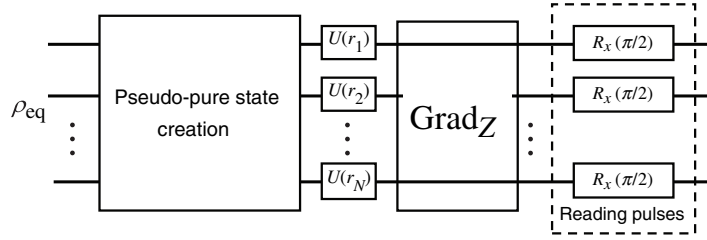
Figure 1. The quantum circuit proposed to simulate the correlation function of equation (2). After the creation of the desired PPS, individual rotations on each spin are used to select the spin projection to be measured and a magnetic field gradient, denoted by grad$_z$, is applied to emulate a projective measurement. Finally, reading pulses are used in order to assess the population values.

density matrix from unity is observed. To simulate the violation of (1) in an NMR quantum computer, the initial state is prepared from the thermal equilibrium into a highly mixed state called pseudo-pure state (PPS) [34]:

$$\rho_{\text{pps}} = \frac{(1-\epsilon)}{2^N} \hat{I} + \epsilon |\psi\rangle\langle\psi|, \tag{3}$$

where $\epsilon \sim 10^{-6}$ is the polarization at room temperature. It is important to remember that the last part of equation (3) represents a pure state and under a unitary transformation it behaves as such. In order to measure the spin projection $\mathbf{r} \cdot \sigma$ (where σ is a vector whose components are the Pauli matrices σ_x, σ_y and σ_z) onto an arbitrary direction $\mathbf{r} = (\cos(\phi)\sin(\theta), \sin(\phi)\sin(\theta), \cos(\theta))$, unitary transformations can be used to rotate the eigenvectors of the operator $\mathbf{r} \cdot \sigma$ onto the computational basis. Since $U^\dagger(\mathbf{r})\sigma_z U(\mathbf{r}) = \mathbf{r} \cdot \sigma$ for $U(\mathbf{r}) = R_y(-\theta)R_z(-\phi)$, by applying the appropriate $U(\mathbf{r})$ on each qubit, we have:

$$\begin{aligned} E(n_1, \ldots, n_N) &= \text{Tr}(\rho_{\text{pps}}\mathbf{r}_1 \cdot \sigma \otimes \cdots \otimes \mathbf{r}_N \cdot \sigma) \\ &= \text{Tr}(\rho'\sigma_z \otimes \cdots \otimes \sigma_z), \end{aligned} \tag{4}$$

where $\rho' = U(\mathbf{r}_1) \otimes \cdots \otimes U(\mathbf{r}_N)\rho_{\text{pps}}U^\dagger(\mathbf{r}_N) \otimes \cdots \otimes U^\dagger(\mathbf{r}_1)$. The above equation tells us that the measurement of $E(n_1, \ldots, n_N)$ can be achieved by rotating each qubit by an appropriate individual rotation and then measuring them all in the computational basis. The projective measurement in the computational basis can be emulated by applying a magnetic field gradient [35], which causes the non-diagonal elements of the density matrix of a heteronuclear spin system to vanish. The density matrix then becomes:

$$\rho' = \frac{(1-\epsilon)}{2^N} \hat{I} + \epsilon \begin{bmatrix} P_{0\cdots0} & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & P_{1\cdots1} \end{bmatrix}. \tag{5}$$

The populations in the second term of (5) represent the probabilities of finding the rotated system in one of the 2^N energy levels. Furthermore, they are also the probabilities $P(s_1, \ldots, s_N)$ shown in (2), which can be recovered from the NMR signal after applying reading pulses to each spin. The signal detected is the average magnetization of the sample over time, which is proportional to the population difference [22, 25]. The acquired signal is then Fourier transformed and normalized by a reference input state. Such a normalization allows the comparison between the experiment and the theoretical results. The scheme to measure
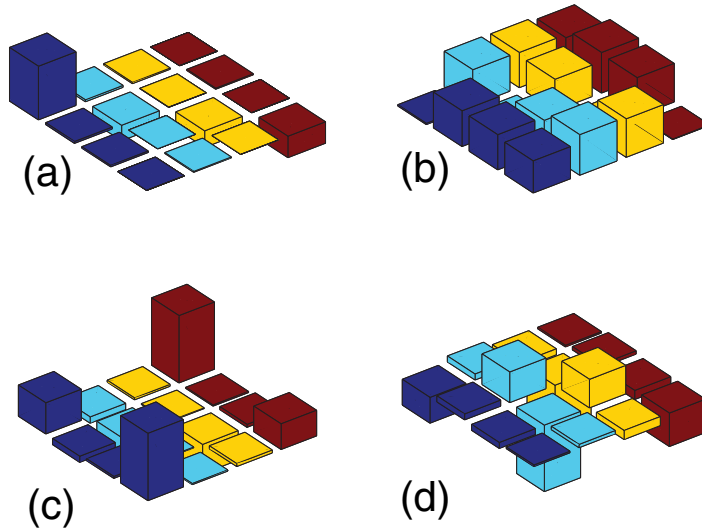
Figure 2. The real part of deviation density matrices determined experimentally for the investigated PPSs (a) $|00\rangle$, (b) $(|00\rangle + |01\rangle + |10\rangle + |11\rangle)/2$, (c) $(|00\rangle + |11\rangle)/\sqrt{2}$ and (d) $(|01\rangle - |10\rangle)/\sqrt{2}$.

correlation functions is shown in figure 1. The circuit must be run for each correlation function appearing in equation (1).

In order to demonstrate our scheme, we used a two-qubit NMR system, namely the nuclear spins of ^{1}H and ^{13}C in chloroform (CHCl$_3$), to simulate the violation of the CHSH inequality [28], which is a special case of (1). It involves the measurement of the quantity

$$\text{CHSH} = E(n_1, n_2) + E(n_3, n_2) + E(n_3, n_4) - E(n_1, n_4), \tag{6}$$

where CHSH is bounded by $-2 \leqslant \text{CHSH} \leqslant +2$ for any LRHVM, whereas the limits imposed by quantum mechanics are given by Tsirelson's bounds $\pm 2\sqrt{2}$ [36]. A particularly interesting situation occurs when the parameters n_1, n_2, n_3 and n_4 label a measurement in the directions $(0, 0, 1)$, $(\sin(2\theta), 0, \cos(2\theta))$, $(\sin(4\theta), 0, \cos(4\theta))$ and $(\sin(6\theta), 0, \cos(6\theta))$, respectively. In this case, quantum mechanics predicts that $\text{CHSH} = 3\cos(2\theta) - \cos(6\theta)$ for the pure entangled state $|\psi\rangle = (|00\rangle + |11\rangle)/\sqrt{2}$ (also called cat state), which results in a maximal violation of the CHSH inequality for $\theta = 22.5°$ and $\theta = 67.5°$.

The NMR experiment was implemented in a Bruker Avance 500 MHz spectrometer in the Bruker BioSpin facility in Germany. The sample contained 99% ^{13}C-labeled chloroform dissolved in deuterated dichloromethane (CD$_2$Cl$_2$), and the concentration was close to 200 mg of CHCl$_3$ per 1 mL of CD$_2$Cl$_2$. PPSs were prepared by the spatial average technique, for which pulse sequences can be found in [35]. The density matrices of the initial states were reconstructed by using the quantum-state tomography technique [37, 38] and the correlation functions were obtained using the measurement scheme summarized in figure 1. The real part of the experimental deviation density matrices ρ_{exp} of the investigated states is shown in figure 2. The deviation $\delta = (\|\rho_{\text{exp}} - \rho_{\text{id}}\|_2)/(\|\rho_{\text{id}}\|_2)$ from the ideal pseudo-pure density matrices ρ_{id} is below 10% in all cases, and the imaginary parts were found to be negligible compared to the real parts. The errors are mainly due to radiofrequency field inhomogeneity and small pulse imperfections. Decoherence is not an important source of error since the time required for the entire experiment ($\sim$15 ms) is much smaller than the estimated decoherence time, $T_1 \sim 5$ s
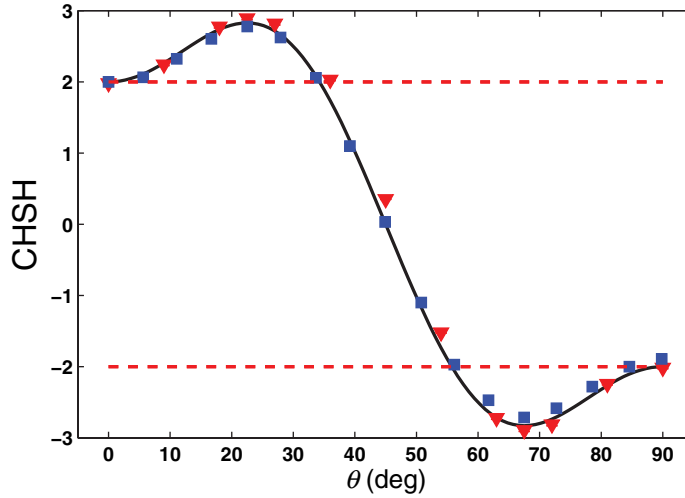
Figure 3. Experimental results for the cat state. ▼, NMR experiment; ■, photon experiment taken from [10]. The solid line is the quantum mechanical prediction.

$(T_1 \sim 15\,\text{s})$ and $T_2 \sim 200\,\text{ms}$ $(T_2 \sim 300\,\text{ms})$ for hydrogen (carbon). From figure 3 it is clear that these sources of error do not cause the experimental points to deviate significantly from the predicted curve.

The experimental results for the cat state can be seen in figure 3, where the CHSH quantity is shown as a function of the angle θ. The experimental results are also compared to the quantum mechanical predictions for a pure cat state and to a photon experiment extracted from [10]. As can be seen, our experiment is in good agreement with both the quantum mechanical theory and the photon experiment.

3. Comparison with a hidden variable model

The density matrix (3) can be decomposed into an ensemble in which a fraction ϵ of the system is in a pure state $|\psi\rangle$ while $(1 - \epsilon)$ is in a completely mixed state. However, this decomposition is not unique. Braunstein *et al* [31] demonstrated that any matrix of the form (3) can be decomposed into a separable ensemble whenever $\epsilon \leqslant 1/(1 + 2^{2N-1})$. This remarkable result shows that although the PPS (3) can be used to implement any quantum computation, it is classically correlated and has a local realistic description.

In this section, we have compared our results to an explicit LRHVM [29]. This model is constructed to predict the quantum mechanical expectation values of any bulk-ensemble NMR experiment that accesses only separable states. The most general type of transformation of quantum states (unitary or not) can be described via operator sum representation $\rho \rightarrow \sum_k E_k \rho E_k^\dagger$ $(\sum_k E_k^\dagger E_k = \hat{I})$ [9]. With this formalism it is possible to simulate every step of our experiment, taking the elements of operation E_k as the model's parameter. Starting from the NMR equilibrium density matrix ρ_{eq}, we simulated every spectrum and analyzed them in the same way as we did for experimental data. Relaxation effects were taken into account using the elements of operation described in [39].
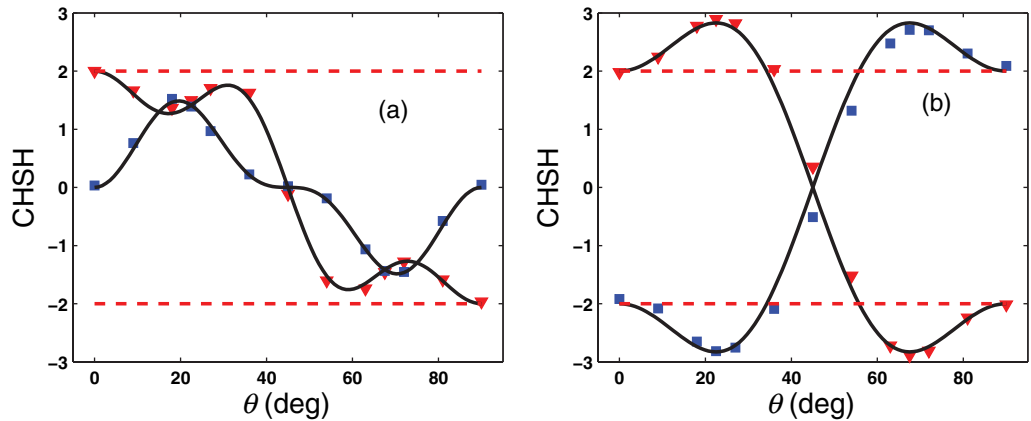
Figure 4. Experimental results of the CHSH quantity as a function of the angle θ. (a) ▼, $|00\rangle$; ■, $(|00\rangle + |01\rangle + |10\rangle + |11\rangle)/2$. (b) ▼, $(|(00\rangle + |11\rangle)/\sqrt{2}$; ■, $(|01\rangle - |10\rangle)/\sqrt{2}$. The continuous lines are the predictions of the LRHVM described in [29]. The NMR data shown here are the same as those in figure 3.

In figure 4(a), the experimental results for the states $|00\rangle$ and $(|00\rangle + |01\rangle + |10\rangle + |11\rangle)/2$ compared to the predictions of the model described in [29] are shown. Since these states are separable, there is no violation of the CHSH inequality, as predicted by the quantum mechanics theory and the LRHVM.

The experimental results for the states $(|00\rangle + |11\rangle)/\sqrt{2}$ and $(|01\rangle - |10\rangle)/\sqrt{2}$ are shown in figure 4(b). Here we found a violation of the CHSH inequality in good agreement with quantum mechanical theory. Additionally, our results are also in good agreement with the LRHVM. The fact that our experimental data are compatible with both theories may appear puzzling. However, it can be understood, noticing that NMR is only sensible for the deviation part of (3), which behaves like a 'pure entangled state'.

This situation resembles the detection loophole, usually discussed in the context of optics. Generally in experiments testing Bell's inequalities[4], imperfections of the experimental apparatus lead to only a small sub-ensemble of the total number of produced entangled particles actually being detected. The question to be asked is whether the measured events are a faithful representation of the whole system. In principle, the detected sub-ensemble could contain a distribution of hidden variables different from the total ensemble. Thus, it is possible for the detected sub-ensemble to violate some Bell's inequality, even if the total ensemble does not, a situation for which one can state that the sub-ensemble 'simulates' the violation of Bell's inequalities. This problem, first noticed in [30], is called the detection loophole. Generally, to overcome the problem, the fair sampling hypothesis, which states that the detected sub-ensemble indeed represents the whole system, is invoked.

In the case of NMR experiments, we cannot invoke such a hypothesis, since the non-detected spins are known to be in highly mixed state and not in the desired entangled state. Furthermore, NMR-QIP has a known LRHVM which is in good agreement with the experimental observations, as shown in figure 4(b). Thus our experiment is indeed a simulation.

[4] Up to now, there is only one experiment [12] reporting a violation of Bell's inequality without the detection loophole.
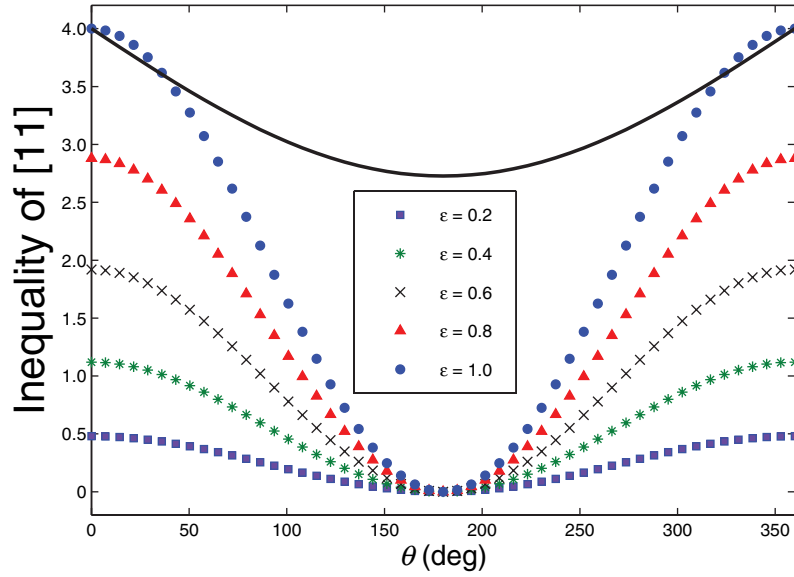
Figure 5. Computer simulation of the violation of the inequality proposed in [11] for various values of the spin polarization ϵ. The solid line represents the limit imposed by hidden variables; the points above the solid line may not have a realistic description.

4. Conclusions

In summary, we have successfully simulated a violation of a Bell's inequality using classical means. The fidelity of our simulation was tested by comparing our results with those of a photon experiment. We also show that we can produce the exact same set of data by using an LRHVM and quantum mechanics. This result can be viewed as an experimental demonstration of how the two theories can be compatible with each other due to the detection loophole. We must emphasize that such an LRHVM is valid only for NMR experiments and not for the photon experiment, although both curves are coincident. Besides, our protocol can be used to simulate or predict results for different Bell's inequality tests, with distinct configurations and a larger number of qubits. Checking Bell's inequalities goes beyond to the hidden variable problem. One example is given in [40], which demonstrates that some Bell's inequalities can act as dimensional witnesses, quantities that can be used to infer the dimensionality of the Hilbert space.

It is important to mention that the same experiment carried out in a highly polarized spin ensemble would not present the same features. It is known [31] that entangled states of the form (3) exist for $\epsilon > 1/(1 + 2^{N/2})$. Recently, a nearly pure NMR quantum entangled state was achieved with polarization $\epsilon = 0.916 \pm 0.019$ [32]. The reported entanglement of formation of such a state was 0.822 ± 0.039.

From the experimental point of view, the difference between the same PPS with low and high polarizations is the intensity of the NMR signal. A remarkable fact is that the former has a known LRHVM and the other does not. Thus, in a highly polarized spin sample, the LRHVM model [29] does not reproduce the quantum mechanical expectation values and a true violation of Bell's inequalities is expected. We must emphasize that even when the NMR state is truly

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entangled, it could not be used to prove non-local effects since NMR is always a local system. However, tests of the realism hypothesis could be performed.

Particularly interesting for NMR are those inequalities that do not require a space-like separation between the entangled particles, such as those recently studied in [11]. In figure 5, we show a computer simulation of the violation of the inequality found in [11]. We simulated the scheme described in this paper using NMR density matrix (3) for various values of spin polarization. The solid line represents the limit imposed by the realism; the region above the limit does not have a realistic description.

Other interesting inequalities are those which do not require entanglement [41][5], such as the temporal Bell's inequalities [42], for which recent proposals based on weak measurements [43, 44] could be adapted to NMR systems.

Besides the ability to simulate quantum systems, we believe that NMR quantum computation could also be used to perform real tests of some fundamentals of quantum mechanics. This subject has been less exploited experimentally outside the scope of optics. However, the ability to generate a highly spin-polarized ensemble allied to the high degree of control could place NMR in a unique position in quantum information science.

Acknowledgments

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[5] Note that even in this case, the polarization must be enhanced, because the LRHVM described in [29] also rules out the violation of such inequalities for $\epsilon \leqslant 1/(1 + 2^{2N-1})$.

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