Falsification of Leggett’s model using neutron matter waves

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New Journal of Physics 14 (2012) 023039 (12pp)
Received 15 November 2011
Published 17 February 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/2/023039

Abstract. According to Bell’s theorem, no theory based on the joint assumption of realism and locality can reproduce certain predictions of quantum mechanics. Another class of realistic models, proposed by Leggett, that demands realism but abandons reliance on locality, is predicted to be in conflict with quantum mechanics. In this paper, we report on an experimental test of a contextual realistic model analogous to the model of Leggett performed with matter waves, more precisely with neutrons. Correlation measurements of the spin-energy entangled single-particle system show violation of a Leggett-type inequality by more than 7.6 standard deviations. Our experimental data falsify the contextual realistic model and are fully in favor of quantum mechanics.

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1. Introduction

Although predictions of quantum theory have been verified with high accuracy, there persists a long-standing debate on whether the concepts of quantum mechanics can be extended to yield a deterministic description of nature, e.g. by introducing so-called hidden variables [1]. Experiments based on Bell’s theorem [2] discard certain types of alternative theories to quantum mechanics [3–8], whereas the Kochen–Specker theorem [9–14] accentuates the incompatibility between such alternatives and quantum mechanics. In 2003, Leggett proposed a non-local realistic model [15], and a paper concerning an extension of Leggett’s model appeared later [16].

The results of photon experiments are in conflict with Leggett’s model [17–20]. A natural question arises as to whether it might be that nature uses different models depending on the physical systems. Thus, it is important to test a model à la Leggett on neutrons, in addition to the ones already performed on photon pairs. In this work, we present a neutron matter-wave experiment, where correlations between spin and energy observables of single particles are measured. A clear violation of the contextual realistic model is demonstrated. This confirms that also for massive particles Leggett’s assumptions cannot fully account for quantum predictions.

An EPR-Bell argument [1, 2] is the best-known approach to examine whether local realism complies with quantum theory: one finds that quantum mechanical predictions cannot be reproduced by the class of local realistic theories. Although hidden-variable theories are introduced to maintain a realistic and deterministic description of nature, thereby providing alternative theories of quantum mechanics, experiments [3–7] show a contradiction between these theories and quantum mechanics. Another class of hidden-variable theories is non-contextual hidden-variable theories, in which the measured value \( v[A] \) of an observable \( A \) is assumed predetermined and not affected by a joint (or simultaneous) measurement of an observable \( B \) compatible with \( A \) (\( \{A, B\} = 0 \))—in other words, the measured value \( v[A] \) of observable \( A \) does not depend on the act and specific settings (the context) of the measurement of observable \( B \) [21]. Studies of non-contextual theories were started by Kochen and Specker [10]. Conflicts between non-contextual theories and quantum mechanics have been pointed out theoretically [22] and subsequently confirmed by experiment [8, 11–14].
2. Leggett’s model

In 2003, Leggett proposed a class of non-local hidden-variable theories and proved that his model was incompatible with quantum predictions [15, 23]. The first experimental test was carried out by extending conventional correlation measurements of linear polarizations to elliptical polarizations [17]. In a subsequent experiment, the rotational symmetry assumption of the bipartite correlation was removed [18] and a different approach to applying a finite number of measurement settings was realized [19]: both experiments clearly exhibit the incompatibility of non-local theories with quantum mechanics. Further correlation measurements [20] revealed violations of inequalities derived also from partially predetermined values.

Until now, non-local Leggett models have been examined only experimentally with photons [24, 25]. In this paper we report experiments with massive particles, replacing non-local correlations by correlations between commuting and compatible observables. Starting with an observation of a violation of a Bell-like inequality [8], neutron interferometer experiments [26] have exploited entanglement between degrees of freedom for single neutrons and have accomplished studies of quantum contextuality: Kochen–Specker phenomena [13] as well as GHZ-like entanglement [27] have been demonstrated. Neutron polarimetric experiments [28] are suitable for investigating contextual models à la Leggett due to high intensities, efficient manipulations and insensitivity to ambient disturbances.

For a polarimetric test, the criteria of the first experimental study by Gröblacher et al [17] are used. We test the model assuming the system to possess two definite physical properties that correspond to two commuting observables (of a two-dimensional system) in quantum mechanics. The model to be tested here is based on the following assumptions.

(i) All the values of measurements are predetermined.
(ii) States are a statistical mixture of subensembles.
(iii) The expectation values taken for each subensemble obey cosine dependence.

Assumptions (i) and (ii) are common to experimental tests of ordinary non-contextual theories and assumption (iii) is a peculiarity of this model. Here, the result of the final measurement of \( B \) (\( A \)) depends on the setting of the previous measurement of \( A \) (\( B \)): a realistic contextual model is tested in our experiment.

Following the development used in works dealing with non-local hidden-variable models [17, 19] and assuming full rotational symmetry, a similar inequality can be employed to test Leggett’s model in our experiment. Denoting the measurement settings for observables \( A \) and \( B \) by \( \vec{a}_1, \vec{a}_2 \) and \( \vec{b}_1, \vec{b}_2 \), respectively (on the Bloch sphere, see figure 1(a)), the Leggett-like inequality is given by

\[
S_{\text{Legg}} = |E_1(\vec{a}_1; \phi) + E_1(\vec{a}_1; 0)| + |E_2(\vec{a}_2; \phi) + E_2(\vec{a}_2; 0)| \leq 4 - \frac{4}{\pi} \left| \sin \frac{\phi}{2} \right|, \tag{1}
\]

where \( E_j(\vec{a}_j; \phi) \), with \( j = 1, 2 \) (as in [19]), represent expectation values of joint (correlation) measurements at settings \( \vec{a}_j \) and \( \vec{b}_j \) with relative angle \( \phi \). We assume settings \( \vec{a}_1, \vec{a}_2 \) and \( \vec{b}_1 \) to lie in a single (equatorial) plane and \( \vec{b}_2 \) to lie in a plane perpendicular to it: expectation values \( E_1 \) and \( E_2 \) are given by correlations in planes perpendicular to each other. The expectation value \( E_j(\vec{a}_j; 0) \) is derived from joint measurements between \( \vec{a}_j \) and \( \vec{b}_j \) (see figure 1(a)). For a pure singlet state, quantum mechanics predicts the expectation values \( E_j(\vec{a}_j; \phi) = -\vec{a}_j \cdot \vec{b}_j = -\cos \phi \) and for the \( S \)-function \( S_{\text{QM}}(\phi) = 2|1 + \cos \phi| \). Maximum violation is expected at \( \phi_{\text{max}} \sim 0.1\pi \).
resulting in a bound of the Leggett-like inequality $S_{\text{Legg}} = 3.797$ and a quantum value of $S_{\text{QM}} = 3.899$.

3. Experiment

The measurement is based on joint measurements of two commuting observables, $A_{\text{spin}}$ for the neutron’s spin and $B_{\text{energy}}$ for the neutron’s total energy (the sum of kinetic and potential energies), representing two degrees of freedom. Coherent manipulations of the energy degree of freedom and a realization of a triply entangled GHZ-like state (additionally using the path degree of freedom) have already been reported [27]. In the present experiment, a maximally entangled Bell-like state

$$|\Psi_N^{\text{Bell}}\rangle \propto \frac{1}{\sqrt{2}} (|\uparrow\rangle |E_0\rangle - |\downarrow\rangle |E_0 - \hbar \omega\rangle),$$

Figure 1. Bloch sphere descriptions of the observables of the spin and energy measurements: (a) for the Leggett case and (b) for the Bell-Clauser–Horne–Shimony–Holt (CHSH) case. The azimuthal angles of $\vec{a}_1$ and $\vec{a}_2$ are fixed at 0 and $\pi/2$, respectively, for all measurements. For case (a), the measurement direction $\vec{b}_2$ lies outside of the equatorial plane and the directions $\vec{b}_1$ and $\vec{b}_2$ are inclined to $\vec{a}_1$ and $\vec{a}_2$ by $\phi$, one along the equator and the other along the meridian. For case (b), all observables lie in the equatorial plane, and directions of the measurements $\vec{b}_1$ and $\vec{b}_2$ are adjusted to have azimuthal angles $\beta_1$ and $\beta_2$ fixed at $\pi/4$ and $3\pi/4$, respectively.
Figure 2. Experimental setup of the neutron polarimetric test of Leggett’s model. An incident monochromatic neutron beam passes through a polarizer: the outgoing beam is polarized in the $+z$-direction. A pair of rectangular coils produces a homogeneous magnetic guide field $B_0$ ($\sim 13$ G). Two spin rotators induce oscillating magnetic fields to the $y$-direction in the RF regime. The first RF spin rotator (RF1) is used for state preparation: the amplitude and the phase of the sinusoidally oscillating field of RF1 are fixed. The measured observables are set by the second spin rotator (RF2): the amplitude and the phase of the oscillating field of RF2, in addition to its position, are tuned appropriately. A spin analyzer rejects neutrons with $-z$-spin component and the intensity of the remaining neutrons in the $+z$-direction are counted by a high-efficiency detector.

with spin basis states $|\uparrow\rangle$ and $|\downarrow\rangle$, as well as energy basis states $|E_0\rangle$ and $|E_0 - \hbar \omega\rangle$, is generated and subjected to successive energy and spin measurements. The experimental setup is displayed in figure 2.

The experiment was carried out at the research reactor facility TRIGA Mark II of the Vienna University of Technology. The incident beam from the reactor is monochromatized to $\lambda = 1.96$ Å by a precisely oriented pyrolytic graphite monochromator and propagates in the $+y$-direction. Passing through a bent Co–Ti super mirror array, the beam is highly polarized. The same technique is employed to analyze the polarization. A high-efficiency (nearly 100% [29]) $BF_3$ detector records the expectation values given in equation (1).

Two identical radio-frequency (RF) spin rotators are employed, each producing a sinusoidally oscillating magnetic field ($\sim 1$ G for a $\pi/2$ rotation) with $\omega = 40$ kHz. They are about 20 cm long and made of an enameled copper wire wound on PVC pipes (diameter $\sim 4$ cm). Both RF spin rotators are put in a homogeneous and static magnetic guide field ($\sim 13$ G) supplied by two rectangular coils. Under these conditions, the rotating-wave approximation is well justified. After tuning the guide field strength, scans of the magnetic field amplitude exhibit sinusoidal intensity modulations with more than 99% contrast. While the position of the first spin rotator (RF1) is fixed, the second RF spin rotator (RF2) is mounted on a translation table: this enables precise adjustment of neutron flight time between the two spin rotators.

By tuning the rotation angle of RF1 to $\pi/2$, a maximally entangled Bell-like state $|\Psi^\text{Bell}\rangle_N$ is generated. The amplitude and phase of the oscillating magnetic field generated by RF2 are directly associated with the parameters of the measurement: the former enables tuning of the polar angle $\alpha$ and the latter the azimuthal angle $\beta$ of the spin measurement. In addition,
adjustment of the position for RF2 results in precise tuning of the relative phase $\gamma$ between the two energy eigenstates. The spin analyzer removes the down-component of the spin: only neutrons with a final $|\uparrow\rangle$-component reach the detector. Denoting unit vectors representing measurement directions as $\vec{x}[^{\theta, \phi}]$ with polar angle $\theta$ and azimuthal angle $\phi$, the analyzer and RF2 enable to set $\vec{a}_j[\pi/2, \gamma_j]$ for energy (equator of the Bloch sphere) and $\vec{b}_k[\alpha_k, \beta_k]$ for spin (arbitrary directions).

3.1. Violation of the Bell-Clauser–Horne–Shimony–Holt inequality

A measurement of the Bell-CHSH-like inequality is carried out first. The Bell-CHSH inequality (see, for instance, [3]) is written in the form

$$S_{CHSH} \equiv |E(\vec{a}_1, \vec{b}_1) + E(\vec{a}_1, \vec{b}_2) - E(\vec{a}_2, \vec{b}_1) + E(\vec{a}_2, \vec{b}_2)| \leq 2,$$

(3)

where $E(\vec{a}_j, \vec{b}_k)$ denotes the expectation value of joint measurements with settings $\vec{a}_j$ and $\vec{b}_k$ (see figure 1(b)). The rotation angles of both spin rotators are set to $\pi/2$, and thus all settings of $\vec{a}_j$ and $\vec{b}_k$ lie in the equatorial plane of the Bloch sphere (see figure 1(b)). The maximum violation is expected for directions $\vec{a}_1[\pi/2, 0]$, $\vec{a}_2[\pi/2, \pi/2]$, $\vec{b}_1[\pi/2, \pi/4]$ and $\vec{b}_2[\pi/2, 3\pi/4]$. For our measurements, the azimuthal angles $\gamma_j$ of the energy measurement directions $\vec{a}_i[\pi/2, \gamma_j]$ are fixed at values $\gamma_1 = 0$, $\gamma_2 = \pi/2$ and $\gamma_j' = \gamma_j + \pi$, whereas the azimuthal angles $\beta_k$ of the spin measurement directions $\vec{b}_k[\pi/2, \beta_k]$ are scanned. In practice, this means that the phase of RF2 is scanned at fixed positions of RF2. Typical intensity oscillations are displayed in figures 3(a) and (b). With a maximum intensity of about 300 neutrons per second, sinusoidal oscillations with extremely high contrast, all above 98% and some reaching almost 99%, are obtained. Following the procedure described in appendix B, the four expectation values of the joint measurements are extracted from the intensity modulations at angles $\beta_1 = \pi/4$, $\beta_2 = 3\pi/4$ and $\beta_k = \beta_n + \pi$. The experimental value of the Bell-CHSH-like inequality is $S_{CHSH} = 2.781(15)$, which is clearly above the boundary 2 by about 53 standard deviations. This experiment confirms a high signal-to-noise ratio and the reliability of our setup.

3.2. Violation of the Leggett inequality

For a measurement of the Leggett-like inequality, equation (1), we require correlation measurements between settings outside the equatorial plane. We vary the parameter $\phi$, which represents the angle of deviation of $\vec{b}_2[\pi/2 - \phi, \pi/2]$ from $\vec{b}_1[\pi/2, \pi/2]$ along the meridian by varying the amplitude of RF2, and of $\vec{b}_1[\pi/2, -\phi]$ from $\vec{b}_1[\pi/2, 0]$ along the equator by varying the phase of RF2 (see figure 1(a)). The polar angle setting is $\pi/2$ for the measurement directions in the equatorial plane and $\pi/2 - \phi$ for the direction $\vec{b}_2$. As mentioned above, the maximum discrepancy between Leggett’s model and quantum mechanics is expected at the angle $\phi_{max} \sim 0.1\pi$ with directions $\vec{a}_1[\pi/2, 0]$, $\vec{a}_2[\pi/2, \pi/2]$, $\vec{b}_1[\pi/2, -\phi_{max}]$ and $\vec{b}_2[\pi/2 - \phi_{max}, \pi/2]$.

Typical intensity oscillations are depicted in figure 3. They are obtained by scanning the azimuthal angle $\beta_k$ for the polar setting $\alpha_1 = \pi/2$ (see figures 3(a) and (b)), and $\alpha_2 = \pi/2 - \phi$ (with $\phi$ is fixed) (see figure 3(c)). Contrasts of the oscillations in figure 3(c) are reduced as the model predicts. For the Leggett-like inequality, equation (1), we have to determine four expectation values. Three of them, $E_1(\vec{a}_1; \phi) \equiv E(\vec{a}_1, \vec{b}_1)$, $E_1(\vec{a}_1; 0) \equiv E(\vec{a}_1, \vec{b}_1)$ and $E_2(\vec{a}_2; 0) \equiv E(\vec{a}_2, \vec{b}_2)$, are extracted from the intensity modulations at the vertical lines with

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Figure 3. Typical intensity modulations as a function of azimuthal angle of the spin observables ($\beta$). Extremely high contrast oscillations are obtained. (a) Oscillations obtained from correlation measurements in the equatorial plane. The azimuthal angle $\gamma_1$ of the energy observable is tuned to 0 and $\pi$. The points on the vertical lines $\beta_1 = -\phi$ and $\beta'_1 = \pi - \phi$ are used to determine the correlations $E_1(\vec{a}_1; \phi)$ of $\vec{a}_1$ with $\vec{b}_1$, and the lines $\beta'_1 = 0$ and $\beta'_1 = \pi$ are used for correlations $E_1(\vec{a}_1; 0)$ of $\vec{a}_1$ with $\vec{b}_1$. (b) Oscillations obtained from correlation measurements in the equatorial plane. The azimuthal angle $\gamma_2$ of the energy observable is tuned to $\pi/2$ and $3\pi/2$. Points on the vertical lines $\beta_2 = \pi/2$ and $\beta'_2 = 3\pi/2$ determine the correlations $E_2(\vec{a}_2; 0)$ of $\vec{a}_2$ with $\vec{b}_2$. (c) Oscillations obtained from correlation measurements outside the equatorial plane. The azimuthal angle $\gamma_1$ of the energy observable is tuned to $\pi/2$ and $3\pi/2$, and the polar angle to $\pi/2 - \phi$. The points on the vertical lines $\beta_2 = \pi/2$ and $\beta'_2 = 3\pi/2$ give the out of plane correlation $E_2(\vec{a}_2; \phi)$ of $\vec{a}_2$ with $\vec{b}_2$.

$\beta_1 = -\phi$, $\beta'_1 = -\phi + \pi$, $\beta''_1 = 0$ and $\beta'''_1 = \pi$ in figure 3(a) and $\beta_2 = \pi/2$ and $\beta'_2 = 3\pi/2$ in figure 3(b). For the derivation of the last expectation value, $E_2(\vec{a}_2; \phi)$, the curves from figure 3(c) are used at the vertical lines $\beta_2 = \pi/2$ and $\beta'_2 = 3\pi/2$ by taking into account the periodicity of polar-angle dependence. Finally, the $S_{\text{Legg}}$-value of the Leggett-like inequality is determined as $S_{\text{Legg}} = 3.8387(61)$ at $\phi = 0.104\pi$, which is clearly larger than the boundary $3.7921$: the violation is more than 7.6 standard deviations.

In order to see the tendency of the violations, the parameter $\phi$ is tuned to eight different values between 0 and $0.226\pi$. Again, the azimuthal angle $\beta_k$ of the sets $\vec{b}_k$ is scanned. The $S_{\text{Legg}}$-value of the Leggett-like inequality is determined as described above. Figure 4 shows a plot of the experimentally determined $S_{\text{Legg}}$ together with the limit of the contextual model as well as the quantum mechanical prediction, calculated for a contrast of 99%. The experimental values follow the quantum mechanical prediction, and this clearly confirms the violation of Leggett’s model for matter waves.

Our result derived from the neutron polarimetric experiment is in excellent agreement with quantum theoretical predictions. All errors include statistical and systematic errors: the systematic errors in these experiments are much smaller than those for interferometric experiments [8, 13], where systematic errors mainly result from phase instability of the
Figure 4. $S$-values as a function of the deviation angle $\phi$ to test Leggett’s alternative to quantum theories. The red (solid) curve is the prediction of quantum mechanics and the dashed line gives the boundary provided by Leggett’s model. The $S_{\text{Legg}}$-value is determined as $S_{\text{Legg}} = 3.8387(61)$ at $\phi = 0.104 \pi$, which is clearly larger than the boundary 3.7921 by more than 7.6 standard deviations.

interferograms. It should be mentioned that, as in other experimental tests of Leggett’s and Bell’s inequalities, all expectation values (as well as intensity modulations in our experiments) are measured successively by using ‘equivalently prepared’ samples.

It is instructive to recall the well-known property of compatible measurements of (commuting, $[A, B] = 0$) observables: the second ($B$) measurement does not destroy the previous information obtained in the first ($A$) measurement. From this, one can conclude that ‘$A$ and $B$ measurements do not interfere—the term compatible is indeed deemed appropriately’ [30]. Quantum indefiniteness, i.e. that individual properties cannot be defined, fully [17–19] or partially [20], under non-local conditions is confirmed in experiments with correlated photon pairs. In our experiment such an indefiniteness becomes explicitly visible under contextual conditions, which refer to compatible measurements: correlations between measurements are observed even though they are compatible and non-interfering.

It is an open question how these correlations come from indefinite properties through non-interfering measurements.

4. Conclusion

In summary, we present a polarimetric experiment with neutron matter waves to study Leggett’s model. The correlations for conventional Bell-CHSH settings, where all vectors representing observables are lying in a single plane, have been measured: the mean contrast reached 98.5%, which is the highest correlation obtained between commuting observables of massive particles. Furthermore, we measured correlations between observables not lying in a single plane in order to study a Leggett-like inequality. The parameter $\phi$ for the deviation angle was varied close to the point of maximum violation: the values follow the quantum mechanical predictions and clearly violate the Leggett-like inequality. This, in turn, confirms quantum indefiniteness under the contextual condition for massive particles.

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Acknowledgments

We acknowledge support from the Austrian Science Fund (FWF) through contract numbers P21193-N20, P20265-N20 and T389-N16 (Hertha–Firnberg position of KD-R).

Appendix A. Theory of measurement observables

The observable for a spin measurement is given by the projector

$$A_{\text{spin}} = \left( \cos \frac{\alpha}{2} | \uparrow \rangle + e^{i\beta} \sin \frac{\alpha}{2} | \downarrow \rangle \right) \left( \cos \frac{\alpha}{2} | \uparrow \rangle + e^{-i\beta} \sin \frac{\alpha}{2} | \downarrow \rangle \right),$$  \hspace{1cm} (A.1)

where $\alpha$ and $\beta$ denote the polar and the azimuthal angle of the spin measurement direction, respectively. For the energy measurement we use the observable

$$B_{\text{energy}} = \frac{1}{2} (| E_0 \rangle + e^{i\gamma} | E_1 \rangle) (| E_0 \rangle + e^{-i\gamma} | E_1 \rangle),$$  \hspace{1cm} (A.2)

where $| E_1 \rangle \equiv | E_0 - \hbar \omega \rangle$ and $\gamma$ gives the azimuthal angle in the equatorial plane. The experimentally measured intensity $I$ corresponds to the expectation value of the joint measurement observable $A_{\text{spin}} \otimes B_{\text{energy}}$ with respect to the state $| \psi_{\text{Bell}} \rangle$, given by equation (2). We find that

$$I(\alpha, \beta, \gamma) = \langle \psi_{\text{Bell}} | A_{\text{spin}} \otimes B_{\text{energy}} | \psi_{\text{Bell}} \rangle = \frac{1}{4} (1 - \sin \alpha \cos (\beta + \gamma)).$$  \hspace{1cm} (A.3)

This theoretically derived intensity function can be compared to the intensity function calculated from plane wave theory for the experimental setup shown in figure 2 (we work in units of $\bar{\hbar} = 2m = 1$, with $m$ the mass and $| \mu |$ the magnetic moment of the neutron, see also [31]). The first RF flipper generates the magnetic field $\hat{B}^{(1)} = \frac{1}{| \mu |} (\omega^{(1)} \cos(\omega t + \zeta), \omega^{(1)} \sin(\omega t + \zeta), \omega_z)^T$, consisting of a guide field $\omega_z/| \mu |$ in the $z$-direction and a rotating field in the $xy$-plane of strength $\omega^{(1)}/| \mu |$, frequency $\omega$ and fixed phase $\zeta$ induces a $\pi/2$ spin-flip ($2\omega^{(1)}\tau_1 = \pi/2$, where $\tau_1$ denotes the time of flight through the first coil). The incoming plane wave $| \psi_0 \rangle = e^{i k^{(2)} y} | E_0 \rangle | \uparrow \rangle$ is transformed into the state

$$| \psi_{\text{RF1}} \rangle = \frac{1}{\sqrt{2}} (e^{i k^{(1)} x} | E_0 \rangle | \uparrow \rangle - i e^{i \zeta} e^{i k^{(1)} y} | E_1 \rangle | \downarrow \rangle),$$  \hspace{1cm} (A.4)

with $k_+ = \sqrt{k_0^2 - \omega_z^2}$ and $k_{\text{rf}} = \sqrt{k_+^2 + 2\omega_z^2 - \omega^2}$. The second RF flipper coil produces the magnetic field $\hat{B}^{(2)} = \frac{1}{| \mu |} (\omega^{(2)} \cos(\omega t + \tilde{\beta}), \omega^{(2)} \sin(\omega t + \tilde{\beta}), \omega_z)^T$, where $\tilde{\beta}$ indicates the adjustable phase of the oscillating field. The distance between the two RF flippers is denoted by $L$. Behind RF2 we obtain

$$| \psi_{\text{RF2}} \rangle = \frac{1}{\sqrt{2}} \left( e^{i k^{(2)} L} \left( \cos \frac{\alpha}{2} e^{i k^{(2)} y} | E_0 \rangle | \uparrow \rangle - i \sin \frac{\alpha}{2} e^{i k^{(2)} y} e^{i \tilde{\beta}} | E_1 \rangle | \downarrow \rangle \right) ight.$$

$$\left. - i e^{i \zeta} e^{i k^{(2)} L} \left( - i \sin \frac{\alpha}{2} e^{i k^{(2)} y} e^{-i \tilde{\beta}} | E_0 \rangle | \uparrow \rangle + \cos \frac{\alpha}{2} e^{i k^{(2)} y} | E_1 \rangle | \downarrow \rangle \right) \right),$$  \hspace{1cm} (A.5)

where the rotation angle $\alpha/2 = \omega^{(2)} \tau_2$ is tuned by adjusting the field strength $\omega^{(2)}$ ($\tau_2$ is the time of flight through the second coil) and $y = y' + L$. Passing the analyzer, which projects onto the $| \uparrow \rangle$-component of the state, leads to

$$| \psi_{\text{proj}} \rangle = \frac{1}{\sqrt{2}} \left( \cos \frac{\alpha}{2} e^{i k^{(2)} L} - i e^{i \zeta} \sin \frac{\alpha}{2} e^{i k^{(2)} L} e^{-i \tilde{\beta}} \right) e^{i k^{(2)} y} | E_0 \rangle | \uparrow \rangle.$$  \hspace{1cm} (A.6)

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After leaving the guide field we obtain

$$|\psi_{\text{out}}\rangle = \frac{1}{\sqrt{2}} \left( e^{ik_1L} - e^{ik_2L} \right) |E_0\rangle |\uparrow\rangle$$

$$= \frac{1}{\sqrt{2}} \left( e^{-i\frac{\pi}{2}L} - e^{i\frac{\pi}{2}L} \right) e^{i(k_2\gamma - k_1\gamma)} |E_0\rangle |\uparrow\rangle,$$

where we have used the approximations $k_1 \approx k_0 - \frac{\omega_z}{v}$, $k_2 \approx k_0 + \frac{\omega_z}{v}$ ($v = 2k_0$ denotes the neutron velocity). The measured intensity oscillations are given by

$$I_{\text{meas}} = \langle \psi_{\text{out}} | \psi_{\text{out}} \rangle = \frac{1}{2} \left( 1 - 2 \cos \frac{\alpha}{2} \sin \frac{\alpha}{2} \right) \theta \left( e^{-i\frac{\pi}{2}L} e^{i(y_{\text{RF}} + \delta - \zeta)} \right)$$

$$= \frac{1}{2} \left( 1 - \sin \alpha \cos \left( \frac{L}{v} - \frac{L}{v} + \delta - \zeta \right) \right) = \frac{1}{2} (1 - \sin \alpha \cos (\gamma + \beta)), \quad \text{(A.8)}$$

where $\beta = \tilde{\beta} + \delta - \zeta$. For the phase setting $\zeta = -\pi/2$ the state $|\psi_{\text{RF1}}\rangle$, equation (A.4), corresponds to the theoretically proposed state $|\psi_{\text{Bell}}\rangle$, equation (2). The Larmor phase $\delta = -2\omega_z \frac{L}{v} = -\omega_L \frac{L}{v}$ as well as the additional phase $\zeta$ can be compensated for by adjusting the phase $\tilde{\beta}$ of RF2. The energy phase $\gamma = \omega_L \frac{v}{v} = \omega \tau$ solely depends on the oscillation frequency $\omega$ of the RF fields and the flight time $\tau = L/v$ or distance $L$ between the two RF coils.

Appendix B. Experimental details

As mentioned in this paper, adjustment of the position of RF2 results in precise tuning of the relative phase between the two energy eigenstates. However, a change of the position of RF2 by $\Delta L$ induces an undesired additional relative phase between the two spin eigenstates, due to Larmor precession within the guide field, denoted by $\Delta \delta = \omega_L \Delta L / v$. Here $\omega_L$ is the Larmor frequency and $v$ the neutron velocity ($\sim 2000 \text{ m s}^{-1}$). To retrieve a pure tuning of the energy phase $\gamma$ this additional Larmor phase has to be compensated for. This is achieved by appropriately tuning the phase of the oscillating magnetic field in RF1, resulting in a reversed spin phase shift of $-\Delta \delta$. As this compensation depends on the position ($\Delta L$) of RF2, the associated Larmor precession angle at each position has to be determined in a separate measurement with two DC spin rotators replacing the RF $\pi/2$ spin rotators. By shifting the position of DC2 ($\Delta L$), pure Larmor precession is observed, from which the Larmor precession angle, in terms of $\Delta L$, is determined.

Monochromatization of the beam is based on Bragg reflection from a mosaic crystal. In order to suppress unwanted influences from the harmonics, such as $\lambda/2, \lambda/3, \ldots$, a slight offset for the incident angle of the polarizer was employed, causing a decrease of intensity of the beam. Finally, a maximum intensity of about 300 neutrons per second was achieved.

Each of the four expectation values of joint measurements $E(\tilde{a}_j, \tilde{b}_k)$ is determined from the count rates acquired at the settings $\tilde{a}_j$ and $\tilde{b}_k$. In particular, an expectation value is calculated from count rates $N(\tilde{a}_j, \tilde{b}_k)$ by

$$E(\tilde{a}_j, \tilde{b}_k) = \frac{N(\tilde{a}_j, \tilde{b}_k) + N(\tilde{a}_j, \tilde{b}_k) - N(\tilde{a}_j, \tilde{b}_k) - N(\tilde{a}_j, \tilde{b}_k)}{N(\tilde{a}_j, \tilde{b}_k) + N(\tilde{a}_j, \tilde{b}_k) + N(\tilde{a}_j, \tilde{b}_k) + N(\tilde{a}_j, \tilde{b}_k)}, \quad \text{(B.1)}$$
where $\vec{a}_j' = [\pi/2, \gamma_j']$ and $\vec{b}_k' = [\alpha, \beta_k'] = [\alpha, \beta_k + \pi]$. Using four expectation values as defined in equation (B.1), $S_{\text{Legg}}$ and $S_{\text{CHSH}}$ are calculated according to equations (1) and (3), respectively.

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