High-efficiency manipulations of triply entangled states in neutron polarimetry

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Abstract. Entanglement occupies a peculiar position in quantum mechanics (QM). It occurs in quantum systems that consist of space-like separated parts or—more generally—in systems whose observables belong to disjoint Hilbert spaces. The latter is the case with single-neutron systems. Here, we report on a neutron polarimetric experiment, where a triply entangled Greenberger–Horne–Zeilinger state is exploited. The entanglement of spin, momentum and total energy degree of freedom is generated utilizing a suitable combination of radio-frequency and static magnetic fields. An average deviation of expectation values from theory—ideal circumstances—of 0.016(1) confirms the predictions of QM with high accuracy, demonstrating the high-efficiency manipulation of the entangled single-neutron system.
1. Introduction

Quantum mechanics (QM) is one of the most successful physical theories ever. Predictions have been confirmed accurately for various physical systems ranging from elementary particle physics to superconductors. However, QM only gives probabilistic predictions for individual events. To preserve a deterministic realism, Einstein and his co-workers Podolsky and Rosen (EPR) claimed, based on the assumption of local realism, that QM is not a complete theory [1]. According to Einstein, a more complete, deterministic theory must underlie QM. In 1951, Bohm reformulated the EPR argument for spin observables of two spatially separated entangled particles to illuminate the essential features of the EPR paradox [2]. Following this reformulation, Bell proved in his celebrated theorem that all hidden variable theories, which are based on the joint assumption of locality and realism, conflict with the predictions of QM [3]. Local hidden variable theories (LHVTs) assume that the outcome of a measurement is predetermined and independent of spacelike separated measurements. Bell introduced inequalities which hold for the predictions of any LHVT, but are violated by QM. Violation of a Bell inequality confirms entanglement and thus, according to Bell’s theorem, the nonlocal character of the quantum systems. From violations of Bell’s inequality, one can conclude that QM cannot be reproduced by local hidden variable theories. Only five years later Clauser, Horne, Shimony and Holt (CHSH) reformulated Bell’s inequalities pertinent to the first practical test of quantum nonlocality [4]. Polarization measurements with correlated photon pairs [5], produced by atomic cascade [6, 7] and parametric down-conversion of photons [8–10], demonstrated violation of the CHSH inequality. To date, various systems [11–14] have been examined, including neutrons [15].

Not a statistical violation but a contradiction between QM and local hidden variable theories was found by Greenberger, Horne and Zeilinger (GHZ) in 1989 for tripartite entanglement [16, 17]. The GHZ argument is independent of the Bell approach, thereby demonstrating in a nonstatistic manner that QM and local realism are mutually incompatible.

Several experimental realizations using multipartite entanglement have been achieved: among them are photon polarization experiments [18–21], experiments with atoms [22], trapped
ions [23] and neutrons [24]. LHVTs are a subset of a more general class of hidden variable theories, namely noncontextual hidden variable theories. Noncontextuality implies that the value of a dynamical variable is pre-determined and independent of the experimental context, i.e. the act and specific settings of previous or simultaneous measurements of commuting observables [25, 26].

In the case of neutrons, entanglement is achieved between different degrees of freedom and not between individual particles. Since the observables of a particular Hilbert space (describing a certain degree of freedom) commute with observables of a different Hilbert space, the single-neutron system is suitable for studying noncontextual hidden variable theories involving multiple degrees of freedom. In [24], preparation of GHZ states for entangled path, spin and energy degrees of freedom in a perfect crystal neutron interferometer was demonstrated utilizing an inequality derived by Mermin [27]. While such an interferometer is well suited for highly intuitive proof-of-principle demonstrations of quantum phenomena [28, 29], the device is extremely delicate. A more robust approach that can be extended to more degrees of freedom would certainly be of advantage. Neutron polarimetry has been used to demonstrate fundamental quantum-mechanical properties, such as the noncommutation of the Pauli spin operator [30] and a number of geometric phase measurements [31–33]. In recent experiments, a test of Leggett’s model, which is based on nonlocality and realism [34], as well as demonstration of a universally valid uncertainty relation [35] have been performed successfully.

In this paper, a neutron polarimetric experiment concerning triple entanglement of spin, total energy and momentum—featuring just the properties lacking in perfect crystal interferometry—is presented. The advantages of neutron polarimetry, such as insensitivity to ambient mechanical and thermal disturbances yielding high phase stability, are exploited and play a key role in our experiment. High-efficiency manipulations, i.e. state splitting and recombination in the individual subspaces, result in a total efficiency of about 99%.

2. Theory

In our polarimeter experiment, entanglement is created between three different degrees of freedom (spin, total energy and momentum) and is defined via a tensor product tripartite Hilbert space: $\mathcal{H} = \mathcal{H}_{\text{spin}} \otimes \mathcal{H}_{\text{momentum}} \otimes \mathcal{H}_{\text{energy}}$. One Hilbert space is spanned by spin-up and spin-down eigenstates, denoted as $|\uparrow\rangle$ and $|\downarrow\rangle$, referring to a quantization axis along a static magnetic field (pointing in the +z-direction); another is spanned by the two-level quantum system consisting of $|E_0\rangle$, the initial total energy eigenstate, and $|E_-\rangle := |E_0 - \hbar \omega\rangle$, which results from a transition due to emission of a photon of energy $\hbar \omega$ when interacting with a time-dependent magnetic field. The third Hilbert space consists of the momentum eigenstates $|k_+\rangle$ and $|k_-\rangle$ induced by Zeeman splitting in a static magnetic field $B_{\text{ACC}}$. The resulting entangled state of the neutron, given by

$$|\psi_N\rangle = \frac{1}{\sqrt{2}}(|\uparrow\rangle|k_-\rangle|E_0\rangle + |\downarrow\rangle|k_+\rangle|E_-\rangle),$$

is of GHZ form.

A graphical illustration of the setup to prepare and manipulate the GHZ state given in equation (1) is depicted in figure 1.

The state of neutrons with energy $\omega_0$ and momentum $k_0$ propagating in the +y-direction, with polarization in +z, in a static magnetic field with a field gradient parallel to its propagation

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Figure 1. (a) Experimental apparatus for the observation of correlations between spinor, momentum and energy degree of freedom. (b) Energy level diagram for momentum and total energy, including corresponding energy eigenstates $|E_0\rangle$, $|E_-\rangle := |E_0 - \hbar \omega\rangle$ and spin states $|\uparrow\rangle$. A $\pi/2$ radio-frequency (RF) spin-turner creates the spin-energy entanglement followed by the accelerator coil, preparing a GHZ-like state that exhibits entanglement between spin, momentum and energy degrees of freedom.

direction (with $\vec{B}(y) = 0$, $\forall y < 0$ and $\vec{B}(y) = \vec{B}_0(y)$, $\forall y > 0$), can be described by a single plane-wave. The neutron wave function, within the static magnetic guide field $\vec{B}_0 = \omega_0 \hat{z}$, is given by

$$|\psi_{gf}\rangle = e^{ik_{gf}y}e^{-i\omega_0 t}\langle \uparrow|,$$

and $k_{gf} = \sqrt{k_0^2 - \omega_z^2}$ (we work in units $\hbar = 2m = 1$ as in [36]), which is identified in terms of a tripartite Hilbert space with

$$|\psi_{gf}\rangle = |\uparrow\rangle|k_{gf}\rangle|E_0\rangle,$$

where $|\uparrow\rangle$, $|k_{gf}\rangle$ and $|E_0\rangle$ denote the eigenstates in spin, momentum and total energy degree of freedom, respectively.
2.1. State evolution

For a precise description of the state vector when exposed to the various static and time-dependent fields, the setup is split up into regions numbered from I on the left to VII on the right side (see figure 1). After entering the guide field \( \vec{B}_0 = \frac{\omega}{\mu} \cdot \hat{\zeta} \) and leaving region I, the state of the neutron as defined in equation (3) is given by

\[
|\psi_1\rangle = e^{ik_{\text{rf}}a_1} |\uparrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle,
\]

where \( a_1 \) denotes the length of region I. In region II (of length \( a_2 \)), a radio-frequency spin-rotator (RF1), together with the guide field, generates a magnetic field of \( \vec{B}_{\text{rf1}} = \frac{1}{|\mu|} (0, \omega_r \cos(\omega t + \varphi), \omega_z)^T \). It consists of the guide field contribution \( \omega_z / |\mu| \) in the \( z \)-direction and a perpendicular oscillating field of strength \( \omega_z / |\mu| \), frequency \( \omega / 2\pi = 40 \text{ kHz} \) and fixed phase \( \varphi \), generated by the RF coil. A \( \pi / 2 \) spin-rotation is induced (with \( \omega_r \tau_1 = \pi / 2 \), where \( \tau_1 \) denotes the time of flight through the coil), resulting in a spin-energy entanglement which is represented as

\[
|\psi_2\rangle = \frac{1}{\sqrt{2}} e^{ik_{\text{rf}}a_1} (e^{ik_{\text{rf}}a_2} |\uparrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle - ie^{i\pi / \omega} e^{ik_{\text{rf}}a_2} |\downarrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle),
\]

where \( k_{\text{rf}} = \sqrt{k_{\text{gf}}^2 + 2\omega_z - \omega} = \sqrt{k_0^2 + \omega_z - \omega} \equiv k_0 + \frac{\omega_z - \omega}{v}, \) where \( v = 2k_0 \) denotes the velocity of the neutron. Now the state enters the acceleration field \( \vec{B}_{\text{acc}} = \frac{\omega}{|\mu|} \cdot \hat{\zeta} \), where the triply entangled GHZ state is generated, denoted as

\[
|\psi_{\text{III}}\rangle = |\psi_{\text{GHZ}}\rangle = \frac{1}{\sqrt{2}} e^{ik_{\text{rf}}a_1} (e^{ik_{\text{rf}}a_2} e^{i\pi / \omega} |\uparrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle - ie^{i\pi / \omega} e^{ik_{\text{rf}}a_2} e^{i\pi / \omega} |\downarrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle),
\]

where \( k_- = \sqrt{(k_{\text{gf}})^2 - \omega} \) and \( k_+ = \sqrt{(k_{\text{rf}})^2 + \omega} \). After the accelerator coil the second RF field (RF2), operating with the magnetic field \( \vec{B}_{\text{rf2}} = \frac{1}{|\mu|} (0, \omega_r \cos(\omega t + \alpha), \omega_z)^T \), induces again a \( \pi / 2 \) spin-rotation \( (\omega_r \tau_2 = \pi / 2) \), where \( \tau_2 \) denotes the time of flight through the second coil. After passing through the analyzer, which projects onto the \( |\uparrow\rangle \)-component of the state, and after leaving the guide field, the final state yields

\[
|\psi_{\text{fin}}\rangle = \frac{1}{2} e^{-i\omega (a_1 + a_2)} e^{-i\omega L''} e^{-i\pi / \omega} e^{i\pi / \omega} |\uparrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle - \frac{1}{2} e^{i\omega (a_1 + a_2)} e^{i\omega L''} e^{i\pi / \omega} e^{-i\pi / \omega} |\uparrow\rangle |\psi_{\text{gf}}\rangle |E_0\rangle,
\]

where we have used the following approximations:

\[
k_{\text{gf}} - k_0 \equiv -\omega_z / v, \hspace{1cm} k_+ - k_0 \equiv -\omega_z / v - \omega / v,
\]

\[
k_{\text{rf}} - k_0 \equiv -\omega_z / v, \hspace{1cm} k_- - k_0 \equiv -\omega_z / v - \omega / v + \omega_a / v.
\]

Finally, the measured intensity is calculated as

\[
I = \langle \psi_{\text{fin}} | \psi_{\text{fin}} \rangle = \frac{1}{2} [1 - \cos(\gamma + \delta + \beta + \alpha - \xi)],
\]

where \( \gamma = -\omega L'' / v \), with \( L'' = a_2 + L + L' \) being the phase accumulated in energy sub-space, \( \delta = -2\omega_z L'' / v = -\omega_z L'' / v \) is the Larmor phase, \( \beta = -2\omega_a L'' / v \) is the momentum phase, \( \alpha \) is the spin phase and \( \xi \) is there for compensation, which will be explained in detail in section 3.2. A detailed description of the neutron’s state vector in all regions can be found in the appendix.
2.2. Measurement procedure

The observable for a spin measurement in the equatorial plane of the Bloch sphere (see figure 2) is given by the projector

\[ \hat{P}^{(S)}(\alpha) = \frac{1}{2}(|\uparrow\rangle + e^{-i\alpha}|\downarrow\rangle)(\langle\uparrow| + e^{i\alpha}\langle\downarrow|), \]  

where \( \alpha \) denotes the azimuthal angle of the spin measurement direction. In the same manner the projection operator for the momentum subspace is given by

\[ \hat{P}^{(k)}(\beta) = \frac{1}{2}(|k_+\rangle + e^{-i\beta}|k_-\rangle)(\langle k_+| + e^{i\beta}\langle k_-|) \]  

and for the energy measurement we use the observable

\[ \hat{P}^{(E)}(\gamma) = \frac{1}{2}(|E_0\rangle + e^{-i\gamma}|E_{-}\rangle)(\langle E_0| + e^{i\gamma}\langle E_{-}|) \]  

where \( \beta \) and \( \gamma \) denote the azimuthal angles in the \( xy \)-planes of the Bloch spheres associated with energy and momentum subspace, respectively. The experimentally measured intensity \( I \) in equation (9) corresponds to the expectation value of the joint measurement observable \( P^{(S)}(\alpha) \otimes P^{(k)}(\beta) \otimes P^{(E)}(\gamma) \) with respect to the GHZ state \( |\psi_N\rangle \), given by equation (1):

\[ I(\alpha, \beta, \gamma) = \langle \psi_N | P^{(S)}(\alpha) \otimes P^{(k)}(\beta) \otimes P^{(E)}(\gamma) | \psi_N \rangle = \frac{1}{8}(1 + \cos(\alpha + \beta + \gamma)). \]  

Setting \( \alpha, \beta \) and \( \gamma \) at values 0 and \( \pi \) or \( \pi/2 \) and \( 3\pi/2 \), the Pauli operators \( \sigma_x \) and \( \sigma_y \) can be decomposed into projection operators as

\[ \sigma_x^{(i)} = \hat{P}^{(i)}(0) - \hat{P}^{(i)}(\pi), \]

\[ \sigma_y^{(i)} = \hat{P}^{(i)}\left(\frac{\pi}{2}\right) - \hat{P}^{(i)}\left(\frac{3\pi}{2}\right), \]  

where \( \hat{P}^{(i)}(j) \), with \( i = (S), (k), (E) \) and \( j = \alpha, \beta, \gamma \), denotes the projection operators onto an up-down superposition on the equatorial plane on the spin, momentum and energy Bloch spheres.

Figure 2. Bloch-sphere description including measurement settings (azimuthal angles) \( \alpha, \beta \) and \( \gamma \) for spin, momentum and total energy subspace, respectively. The settings determine the projection operators used for joint measurements in the experiment.
spheres (see figure 2). Each expectation value $E(\sigma_x^{(S)} \sigma_y^{(k)} \sigma_z^{(E)})$ is experimentally determined by a combination of normalized count rates using appropriate settings of $\alpha$, $\beta$ and $\gamma$, for instance,

$$E(\sigma_x^{(S)} \sigma_y^{(k)} \sigma_z^{(E)}) = E\left(\alpha : (0; \pi), \beta : \left(\frac{\pi}{2}; \frac{3\pi}{2}\right), \gamma : \left(\frac{\pi}{2}; \frac{3\pi}{2}\right)\right)$$

$$= \langle \Psi_{\text{GHZ}} | (\hat{P}(0)^{(S)} - \hat{P}(\pi)^{(S)}) \otimes \left(\hat{P}\left(\frac{\pi}{2}\right)^{(k)} - \hat{P}\left(\frac{3\pi}{2}\right)^{(k)}\right) \otimes \left(\hat{P}\left(\frac{\pi}{2}\right)^{(E)} - \hat{P}\left(\frac{3\pi}{2}\right)^{(E)}\right) | \Psi_{\text{GHZ}} \rangle = \frac{A}{B}$$

(15)

with

$$A = \left( N\left(0, \frac{\pi}{2}, \frac{\pi}{2}\right) - N\left(\pi, \frac{\pi}{2}, \frac{\pi}{2}\right) - N\left(0, \frac{\pi}{2}, \frac{3\pi}{2}\right) + N\left(\pi, \frac{\pi}{2}, \frac{3\pi}{2}\right) \right.
- N\left(0, \frac{3\pi}{2}, \frac{\pi}{2}\right) - N\left(\pi, \frac{3\pi}{2}, \frac{\pi}{2}\right) + N\left(0, \frac{3\pi}{2}, \frac{3\pi}{2}\right) + N\left(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}\right) \right)$$

(16)

and

$$B = \left( N\left(0, \frac{\pi}{2}, \frac{\pi}{2}\right) + N\left(\pi, \frac{\pi}{2}, \frac{\pi}{2}\right) + N\left(0, \frac{\pi}{2}, \frac{3\pi}{2}\right) + N\left(\pi, \frac{\pi}{2}, \frac{3\pi}{2}\right) + N\left(0, \frac{3\pi}{2}, \frac{\pi}{2}\right) + N\left(\pi, \frac{3\pi}{2}, \frac{\pi}{2}\right) + N\left(0, \frac{3\pi}{2}, \frac{3\pi}{2}\right) + N\left(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}\right) \right),$$

(17)

where for example $N(0, \pi/2, \pi/2)$ is the count rate for spin phase $\alpha = 0$, momentum phase $\beta = \pi/2$ and total energy phase $\gamma = \pi/2$.

3. Experiment

3.1. Setup

The experiment was carried out at the tangential beam port of the 250 kW research reactor facility TRIGA Mark II of the Vienna University of Technology (TU Vienna). A neutron beam, incident from a pyrolytic graphite crystal, of mean wavelength $\lambda = 1.99$ Å and spectral width $\Delta \lambda / \lambda = 0.02$, is polarized to $\sim 99\%$ along the $+z$-direction, by reflection from a bent Co–Ti supermirror array. The polarizer is adjusted to higher incident angles so that the second-order harmonics in the incident beam are suppressed. A $^3$He monitor detector is used for normalization in order to correct statistical fluctuations of the reactor power, and a BF$_3$ detector with high efficiency (nearly 100% [37]) is used for count rate detection. A final maximum intensity of about 500 counts s$^{-1}$ at a cross-section of $10 \times 10$ mm$^2$ was recorded. To avoid unwanted depolarization, a uniform guide field from coils arranged in the Helmholtz configuration, pointing in the $+z$-direction with a strength of about 13 G, is applied over the entire setup. A schematic illustration of the experimental arrangement is depicted in figure 1(a).
3.2. Measurement strategy and results

Each expectation value $E(\sigma_{x,y}^{(S)}\sigma_{x,y}^{(k)}\sigma_{x,y}^{(E)})$ is determined by a combination of normalized count rates, due to an appropriate setting of $\alpha$, $\beta$, and $\gamma$, as defined in equations (15)–(17). Since all projection operators required in the experiment only differ in phase between the two orthogonal states, i.e. lie in the $xy$-plane on the Bloch sphere, they can be realized by simple phase manipulations. By appropriately tuning the phase in the individual subsystems the desired azimuthal angle on the respective Bloch sphere is adjusted.

The spin measurement direction on the equatorial plane of the respective Bloch sphere (see figure 2) is tuned by the spin-phase $\alpha$, being the phase of the oscillating field within the second RF spin-rotator ($\vec{B}^{(rf2)} = \frac{1}{|\mu|}(0, \omega_r \cos(\omega_t + \alpha), \omega_z)\hat{z}$), which determines the azimuthal angle. The corresponding projection operator is defined in equation (10). Since the analyzing supermirror rejects neutrons with $-z$ spin component and all spin states lie in the $xy$-plane, a $\pi/2$ spinor-rotation has to be performed. Typical intensity modulations when scanning $\alpha$ are depicted in figure 3. Since the efficiencies of the manipulations, including state splitting and recombination, are considerably high, an average contrast of 0.9844(9) was achieved.

The measurement direction on the momentum Bloch sphere (also lying in the equatorial plane) is set by the momentum phase $\beta$, i.e. the propagation time within the accelerator coil or the field strength, corresponding to the physical action of the projection operator defined from equation (11). The acquired phase in momentum space is given by $\beta = \int B_{acc} \cdot ds$. In practice, the strength of the magnetic field was varied for experimental convenience. The accelerator coil, just like the uniform guide field, is realized in the Helmholtz configuration, for better field homogeneity.

Figure 3. Typical intensity oscillation when varying the spin phase $\alpha$ for different settings of the momentum phase $\beta$ and energy phase $\gamma$.
This one-detector scheme has already been used in previous experiments [2]. In practice this is not the case, due to the so-called Bloch–Siegert shift [2]. A change in 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 as $\Delta \delta = \omega_L \Delta L/v$. Here $\omega_L$ is the Larmor frequency and $v$ the velocity of the neutrons ($\sim 2000 \text{ m s}^{-1}$). To retrieve a pure tuning of the energy phase $\gamma$ this additional Larmor phase has to be compensated for. As seen from equation (9) this can be done by setting $\zeta$, which is the phase of the oscillating magnetic field in RF1 $(\mathbf{B}^{(rf)}{[\hat{z}]} = \frac{\mu}{\mu} (\omega \cos(\omega t + \zeta)) \cdot \hat{y})$, to $\zeta = \Delta \delta$. As this compensation depends on the relative position ($\Delta L$) of RF2, the associated Larmor precession angle at each position has to be determined in an individual measurement using two DC instead of RF-$\pi/2$ spin rotators and without the accelerator coil. 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 [38]. If the resonance condition were exactly $\omega = \omega_L$, Larmor- and energy-phase, for spin- and energy-subspace, respectively, would make phase contributions for the state, of the same amount but opposite directions, since $\gamma = \omega L''$ and $\delta = -\omega_L L''$. Thus, when varying $L$ there should be no intensity modulation. In practice this is not the case, due to the so-called Bloch–Siegert shift [39]. By tuning the momentum phase $\beta$ and the energy phase $\gamma$ at $0, \pi/2, \pi$ and $3\pi/2$, sixteen spin phase $\alpha$ scans were carried out, which is plotted in figure 3. The individual results for each expectation value are summarized in table 1.

### Table 1. Experimentally determined expectation values of $E(\sigma_x^{(S)} \sigma_y^{(k)} \sigma_x^{(E)})$, together with predictions of QM.

| Observable | $\alpha$ | $\beta$ | $\gamma$ | QM Settings | Determined Values |
|------------|----------|----------|----------|--------------|------------------|
| $\sigma_x^{(S)} \sigma_y^{(k)} \sigma_x^{(E)}$ | (0; $\pi$) | (0; $\pi$) | (0; $\pi$) | 1 | 0.9843(12) |
| $\sigma_y^{(S)} \sigma_y^{(k)} \sigma_x^{(E)}$ | (0; $\pi$) | ($\pi/2$; $3\pi/2$) | ($\pi/2$; $3\pi/2$) | $\beta$ | -0.9839(10) |
| $\sigma_y^{(S)} \sigma_y^{(k)} \sigma_x^{(E)}$ | ($\pi/2$; $3\pi/2$) | (0; $\pi$) | ($\pi/2$; $3\pi/2$) | $\gamma$ | -0.9840(10) |
| $\sigma_y^{(S)} \sigma_y^{(k)} \sigma_x^{(E)}$ | ($\pi/2$; $3\pi/2$) | ($\pi/2$; $3\pi/2$) | (0; $\pi$) | $\beta, \gamma$ | -0.9837(11) |

The measurement direction on the energy Bloch sphere is tuned by the energy phase, which depends on the position of the second RF-$\pi/2$ spin-rotator, mounted on a motorized translation stage. Adjustment of the position of RF2 results in precise tuning of the relative phase $\gamma$ between the two energy eigenstates (see equation (12)). A change in 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 as $\Delta \delta = \omega_L \Delta L/v$. Here $\omega_L$ is the Larmor frequency and $v$ the velocity of the neutrons ($\sim 2000 \text{ m s}^{-1}$). To retrieve a pure tuning of the energy phase $\gamma$ this additional Larmor phase has to be compensated for. As seen from equation (9) this can be done by setting $\zeta$, which is the phase of the oscillating magnetic field in RF1 $(\mathbf{B}^{(rf)}{[\hat{z}]} = \frac{\mu}{\mu} (\omega \cos(\omega t + \zeta)) \cdot \hat{y})$, to $\zeta = \Delta \delta$. As this compensation depends on the relative position ($\Delta L$) of RF2, the associated Larmor precession angle at each position has to be determined in an individual measurement using two DC instead of RF-$\pi/2$ spin rotators and without the accelerator coil. 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 [38]. If the resonance condition were exactly $\omega = \omega_L$, Larmor- and energy-phase, for spin- and energy-subspace, respectively, would make phase contributions for the state, of the same amount but opposite directions, since $\gamma = \omega L''$ and $\delta = -\omega_L L''$. Thus, when varying $L$ there should be no intensity modulation. In practice this is not the case, due to the so-called Bloch–Siegert shift [39]. By tuning the momentum phase $\beta$ and the energy phase $\gamma$ at $0, \pi/2, \pi$ and $3\pi/2$, sixteen spin phase $\alpha$ scans were carried out, which is plotted in figure 3. The individual results for each expectation value are summarized in table 1.

### 4. Discussion and conclusion

#### 4.1. Discussion

A key feature of our experiment is that spin, momentum and energy represent independent degrees of freedom, which can be individually (coherently) manipulated. It is worth noting here that, when a DC spin-flipper is placed subsequent to an RF spin-rotator, only spin is flipped, leaving the energy degree of freedom untouched. This is clear evidence that the spin and energy degrees of freedom are independent.

Since the experiment was carried out with single particles a successive detection scheme with one detector is needed for the experiment. All intensities required for the calculation of the corresponding expectation value were recorded one after the other, as described in section 2.2. This one-detector scheme has already been used in previous experiments [15, 40].
The main limitation in our experiment of the initial degree of polarization. For an even higher contrast a more efficient polarizer/analyzer system has to be developed. With this modification, see, for example, [41] where a degree of polarization $P = 0.997(1)$ has been achieved, the loss of contrast in our experiments is expected to decrease at least by one order of magnitude.

Compared to our previous experiment, where a GHZ state was prepared in an interferometric experiment [24] and analyzed by an inequality derived by Mermin [27], we want to point out the novelties of the present work: in our polarimetric experiment other degrees of freedom are utilized to prepare a GHZ state. The spatial separation obtained in the interferometer (path I and path II) is replaced by the momentum degree of freedom ($|k_\pm\rangle$) — an accelerated and a decelerated component. The stability of the polarimeter scheme allows for highly efficient manipulations of the quantum system, resulting in a precise determination of expectation values. Instead of testing an inequality to rule out certain realistic models, as in [24], we focus on an accurate comparison of the obtained results with the predictions of QM. This work aims to contribute to the advancement in the wide field of quantum communication and quantum information processing [42].

Besides these technical challenges, further studies in the field of quantum contextuality are anticipated, for instance by using multi-energy splitting in a single-particle system. Considering a scheme with multi-step energy manipulations by applying different frequencies $\omega_i$ for the RF spin-rotators (see, e.g., [43]), a large number of ‘artificial’ (instead of natural) energy levels become accessible. Finally, we consider a similar procedure for ultra-cold neutrons in a storage experiment as in [44]. Here the multiple transition frequencies $\omega_i$ are applied successively (in time), promising the generation of multilevel quantum systems (of the order of $10^3$), which could be used for quantum information processing.

4.2. Conclusion

In this paper, the demonstration of high-efficiency manipulations of individual subsystems—degrees of freedom—together with an accurate comparison with quantum theory is presented. The deviation of less than 2% from the theoretical expectation values clearly confirms the predictions of QM of a spin–momentum–energy entangled state in a single-neutron system.

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Appendix. Complete state vector evolution

The entanglement is created within the polarimeter between three different degrees of freedom (spin, momentum and total energy) and can be described with a tripartite Hilbert space $\mathcal{H} = \mathcal{H}_{\text{spin}} \otimes \mathcal{H}_{\text{momentum}} \otimes \mathcal{H}_{\text{energy}}$. The resulting entangled state vector of the neutron represents a GHZ-like state denoted as

$$|\psi_{\text{GHZ}}^N\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle |k_-\rangle |E_0\rangle + |\downarrow\rangle |k_+\rangle |E_-\rangle).$$

(A.1)
A schematic illustration of the setup, divided into seven regions, is depicted in figure A.1. In the following, the occurring state vectors corresponding to each region are derived in detail. The incoming beam is polarized in the up-direction

$$|\psi_{in}| = e^{ik_0 x} e^{-i\omega t} |\uparrow\rangle,$$  \hspace{1cm} (A.2)

which is described in a tripartite Hilbert space as

$$|\psi_{in}| = |\uparrow\rangle |k_0\rangle |E_0\rangle.$$ \hspace{1cm} (A.3)

Entering the guide field $\vec{B}_0 = \frac{\omega}{\mu} \cdot \hat{z}$ gives

$$|\psi_{gf}| = |\uparrow\rangle |k_{gf}\rangle |E_0\rangle,$$ \hspace{1cm} (A.4)

which results in

$$|\psi_1| = e^{ik_0 a_1} |\uparrow\rangle |k_{gf}\rangle |E_0\rangle,$$ \hspace{1cm} (A.5)

at the end of region I, with $k_{gf} = \sqrt{k_0^2 - \omega^2}$, and $a_1$ being the length of region I. In region II the first RF-spin flipper coil RF1 generates a magnetic field of $\vec{B}_{rf1} = \frac{1}{|\mu|} (0, \omega r, \cos(\omega t + \zeta), \omega z)^T$ consisting of a guide field $\omega z / |\mu|$ in the z-direction and an oscillating field in the y-direction of strength $\omega r / |\mu|$, frequency $\omega$ and fixed phase $\zeta$. It induces a $\pi / 2$ spin-flip ($\omega r \tau_1 = \pi / 2$, where $\tau_1$ denotes the time of flight through the first coil) and we get for the state vector

$$|\psi_2| = \frac{1}{\sqrt{2}} e^{ik_{gf}(a)} (e^{ik_0 a_2} |\uparrow\rangle |k_{gf}\rangle |E_0\rangle - i e^{i\zeta} e^{ik_{gf}(a)} |\downarrow\rangle |k_{gf}\rangle |E_0\rangle),$$ \hspace{1cm} (A.6)

where $k_{gf} = \sqrt{k_{gf}^2 + 2\omega r - \omega z} = \sqrt{k_0^2 + \omega z - \omega r} = \sqrt{k_0^2 + \omega z - \omega r}$ is the effective wave number. Now the state enters the acceleration field $\vec{B}_{acc} = \frac{\omega s}{|\mu|} \cdot \hat{z}$ and we get

$$|\psi_3| = |\psi_{GHZ}| = \frac{1}{\sqrt{2}} e^{ik_{gf}(a)} (e^{ik_0 a_2} e^{ik_{gf}(a)} |\uparrow\rangle |k_0\rangle |E_0\rangle - i e^{i\zeta} e^{ik_{gf}(a)} e^{ik_{gf}(a)} |\downarrow\rangle |k_0\rangle |E_0\rangle),$$ \hspace{1cm} (A.7)

where $k_0 = \sqrt{(k_{gf})^2 - \omega^2}$ and $k_+ = \sqrt{(k_{gf})^2 + \omega^2}$. After the acceleration field we obtain

$$|\psi_4| = \frac{1}{\sqrt{2}} e^{ik_{gf}(a)} (e^{ik_0 a_2} e^{ik_{gf}(a)} L |\uparrow\rangle |k_{gf}\rangle |E_0\rangle - i e^{i\zeta} e^{ik_{gf}(a)} e^{ik_{gf}(a)} L |\downarrow\rangle |k_{gf}\rangle |E_0\rangle).$$ \hspace{1cm} (A.8)

The second RF field RF2 operates with the magnetic field $\vec{B}_{rf2} = \frac{1}{|\mu|} (0, \omega r, \cos(\omega t + \alpha), \omega r, \omega z)^T$ and induces again a $\pi / 2$ spin-flip ($\omega r \tau_2 = \pi / 2$, where $\tau_2$ denotes the time of flight through the second coil) and we get for the state vector

$$|\psi_5| = \frac{1}{\sqrt{2}} e^{ik_{gf}(a)} (e^{ik_0 a_2} e^{ik_{gf}(a)} L |\uparrow\rangle |k_{gf}\rangle |E_0\rangle - i e^{i\zeta} e^{ik_{gf}(a)} e^{ik_{gf}(a)} L |\downarrow\rangle |k_{gf}\rangle |E_0\rangle).$$ \hspace{1cm} (A.9)
through the second coil, which gives

\[ |\psi_V\rangle = \frac{1}{2} e^{ik_{gf}a_1} (e^{ik_{gf}a_2} e^{ik_{L}} e^{i(k_{gf}a_3)} |\uparrow\rangle |k_{gf}\rangle |E_0\rangle - i e^{i\alpha} e^{ik_{gf}a_3}|\downarrow\rangle |k_{gf}\rangle |E_0\rangle + e^{ik_{gf}a_3}|\downarrow\rangle |k_{gf}\rangle |E_0\rangle) \]

(A.9)

Passing through the analyzer, which projects onto the |\uparrow\rangle-component of the state, leads to

\[ |\psi_{V1}\rangle = \frac{1}{2} e^{ik_{gf}a_1} e^{ik_{gf}a_2} e^{ik_{L}} \left( e^{i(k_{gf}a_3)} |\uparrow\rangle - i e^{-i\alpha} e^{ik_{gf}a_3} e^{ik_{L}} |\downarrow\rangle \right) |k_{gf}\rangle |E_0\rangle , \quad (A.10) \]

After leaving the guide field, we obtain

\[ |\psi_{VII}\rangle = |\psi_{fin}\rangle = \frac{1}{2} e^{-i\frac{\pi}{2}(\alpha_3+\alpha_4+\alpha_5)} \times \left( e^{-i\frac{\pi}{2}(\alpha_2+L'+L)} e^{-i\alpha} e^{-i\frac{\pi}{2}(\alpha_2+L'+L)} e^{i\frac{\pi}{2}(\alpha_2+L'+L)} - e^{i\alpha} e^{-i\frac{\pi}{2}(\alpha_2+L'+L)} e^{i\frac{\pi}{2}(\alpha_2+L'+L)} \right) |\uparrow\rangle |k_0\rangle |E_0\rangle , \quad (A.11) \]

where the following approximations have been used:

\[ k_{gf} - k_0 \approx -\frac{\omega_2}{v}, \quad k_+ - k_0 \approx -\frac{\omega_2}{v} - \frac{\omega_a}{v}, \quad \]

\[ k_{gf} - k_0 \approx \frac{\omega_2}{v} - \frac{\omega_2}{v}, \quad k_- - k_0 \approx \frac{\omega_2}{v} - \frac{\omega_2}{v} + \frac{\omega_a}{v}. \quad (A.12) \]

The observed intensity is calculated from |\psi_{fin}\rangle as

\[ I = \langle \psi_{fin} | \psi_{fin} \rangle = \frac{1}{2} \left[ 1 - \cos(\gamma + \delta + \beta + \alpha - \xi) \right], \quad (A.13) \]

with energy phase \( \gamma = \omega_2 \frac{L+L'}{v} \), Larmor phase \( \delta = -2 \omega_2 \frac{a_2+L+L'}{v} \), momentum phase \( \beta = -2 \omega_2 \frac{L}{v} \), spin phase \( \alpha \) and \( \xi \) is there for compensation, as explained in detail in the main text.

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