Triple Entanglement in Neutron Interferometric and Polarimetric Experiments

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Abstract. Entanglement is a remarkable peculiarity in quantum mechanics. It occurs in quantum systems that consist of space-like separated parts, or in systems whose observables belong to disjoint Hilbert spaces. The latter is the case in single-neutron systems. Entangled states are renowned for exhibiting non-classical correlations between observables of individual sub-systems. In a perfect Si-crystal interferometer experiment entanglement between three degrees of freedom in a single-neutron system is created. The prepared entanglement of spin, path and energy is induced by interaction with an oscillating magnetic field. The generated Greenberger-Horne-Zeilinger (GHZ) state is analyzed with an inequality derived by Mermin, yielding a value $M = 2.558(4) \leq 2$, which exhibits a clear violation of the classical assumption. In addition observation of a GHZ entanglement, consisting of spin, momentum and total energy, in a neutron polarimetric experiment is presented. Here the advantages of neutron polarimetry, such as high contrast or insensitivity to ambient disturbances, are utilized resulting in final value of $M = 3.936(2) \leq 2$.

1. Introduction

Quantum mechanics is probably the most successful physical theory ever. No other theory has yet given more accurate predictions from elementary-particle physics to the early stages of our universe. However, quantum mechanics only gives probabilistic predictions for individual events. Consequently, according to Einstein, a more complete, deterministic, hidden theory must underlie quantum mechanics [1]. 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]. After this, 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 quantum mechanics [3]. Local hidden variable theories (LHVTs) assume that the outcome of a measurement is predetermined and independent of spacelike separated measurements. Therefore, Bell introduced inequalities which hold for the predictions of any LHVT considered, but are violated by quantum mechanics. Violation of a Bell inequality proves the presence of entanglement and thus, according to Bells theorem, non-local characteristics of the quantum systems. From this, one can conclude that quantum mechanics cannot be reproduced by local hidden variable theories. Only five years later Clauser, Horne, Shimony and Holt (CHSH) reformulated Bells inequalities pertinent for the first practical test of quantum non-
locality [4]. Polarization measurements with correlated photon pairs [5], produced by atomic cascade [6, 7] and parametric down-conversion of lasers [8, 9, 10], demonstrated violation of the CHSH inequality. Up to date several systems [11, 12, 13, 14] have been examined, including neutrons [15].

Not a statistical violation, but a contradiction between quantum mechanics 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 quantum mechanics and local realism are mutually incompatible. Several experimental realizations, using multipartite entanglement have been achieved: among them are photon polarization experiments [18, 19, 20, 21], atoms [22] and trapped ions [23]. The GHZ argument was analyzed in detail by Mermin [24], where a inequality is derived for a state of \( n \) spin-1/2 particles that is violated by quantum mechanic by an amount that increases exponentially with \( n \).

LHVTs are a subset of a more general class of hidden variable theories, namely the noncontextual hidden variable theories. Noncontextuality implies that the value of a dynamical variable is determined and independent of the experimental context, i.e. of previous or simultaneous measurements of a commuting observable [25, 26]. Noncontextuality is a more stringent demand than locality because it requires mutual independence of the results for commuting observables even if there is no spacelike separation [27]. First tests of quantum contextuality, based on the Kochen–Specker theorem [28], have been proposed [29, 30] and performed successfully using trapped ions [31], photons [32] and neutrons [33, 34].

In the case of neutrons entanglement is not achieved between particles but between different degrees of freedom. Since the observables of one Hilbert spaces (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 with multiple degrees of freedom.

2. Interferometric Experiment
Neutron interferometry [35, 36] has become a unique tool for studies on the quantum-mechanical nature of matter waves. A beam of massive particles is coherently split by amplitude division and recombined after passing through different regions of space. During these space-like separation of typically a few centimeters, the neutron’s wavefunction is modified in phases (and amplitude), due to various possible interactions. This interaction may be of nuclear, magnetic, electric or gravitational type, depending on which neutron optical devices are inserted. The topic of neutron interferometry belongs to the field of self-interference, since at a given instant at most one neutron propagates through the interferometer. Using neutron interferometry (IFM) single-particle entanglement, between the spinor and the spatial part of the neutron wave function [15], as well as full tomographic state analysis [37], have already been accomplished.

2.1. Coherent Energy Manipulation
The neutron’s total energy degree of freedom is an almost ideal candidate for a degree of freedom to be manipulated, due to its experimental accessibility within a magnetic resonance field [38]. For this purpose the time evolution of the system is described by a photon-neutron state vector, which is an eigenvector of the corresponding modified Jaynes-Cummings (J-C) Hamiltonian [39]. The J-C Hamiltonian can be adopted for a system consisting of a neutron coupled to a quantized rf-field [40]. Manipulating the total energy provides realization of triple-entanglement between the neutron’s path, spin and energy degrees of freedom.
2.1.1. The corresponding neutron Jaynes-Cummings Hamiltonian

Since two RF-fields, operating at frequencies $\omega$ and $\omega/2$, are involved in the actual experiment, the modified J-C Hamiltonian is denoted as

$$\mathcal{H}_{J-C} = -\frac{\hbar^2}{2m} \nabla^2 - \mu B_0(\vec{r}) \sigma_z + \hbar (\omega \hat{a}^\dagger \hat{a} + \frac{\omega}{2} \hat{a}^\dagger_{\omega/2} \hat{a}_{\omega/2})$$

$$+ \mu \left( \frac{B_1^{(\omega)}(\vec{r})}{\sqrt{N_\omega}} (\hat{a}^\dagger \sigma_+ + \hat{a} \sigma_+) + \frac{B_1^{(\omega/2)}(\vec{r})}{\sqrt{N_{\omega/2}}} (\hat{a}^\dagger_{\omega/2} \sigma_+ + \hat{a}_{\omega/2} \sigma_+) \right),$$

(1)

where $m$ and $\mu$ are the mass (1.6749 $10^{-27}$ kg) and the magnetic moment ($-1.913\mu_N$, with $\mu_N = 5.051 \times 10^{-27}$ J/T) of the neutron, respectively and $\sigma_{\pm} = \frac{1}{2}(\sigma_x \pm i \sigma_y)$. The first term accounts for the kinetic energy of the neutron. The second term leads to the usual Zeeman splitting of $2|\mu|B_0$. The third term adds the photon energy of the oscillating fields of frequencies $\omega$ and $\omega/2$, by use of the creation and annihilation operators $\hat{a}^\dagger$ and $\hat{a}$. Finally, the last term represents the coupling between photons and the neutron, where $N_{\omega_j} = \langle \hat{a}^\dagger_{\omega_j} \hat{a}_{\omega_j} \rangle$ represents the mean number of photons with frequencies $\omega_j$ in the RF-field. Note that the first two and the last terms concern the spatial $|\psi(\vec{r})\rangle$ and the (time-dependent) energy $|E(t)\rangle$ subspaces of neutrons, respectively.

The state vectors of the oscillating fields are represented by coherent states $|\alpha\rangle$, which are eigenstates of $\hat{a}^\dagger$ and $\hat{a}$. The eigenvalues of coherent states are complex numbers, so one can write

$$\hat{a}|\alpha\rangle = \alpha |\alpha\rangle = |\alpha\rangle e^{i\phi}|\alpha\rangle \text{ with } |\alpha| = \sqrt{N}.$$  

(2)

Using Eq. (1) one can define a total state vector including not only the neutron system $|\Psi_N\rangle$, but also the two quantized oscillating magnetic fields:

$$|\Psi_I\rangle = |\alpha_\omega\rangle \otimes |\alpha_{\omega/2}\rangle \otimes |\Psi_N\rangle.$$  

(3)

In a perfect Si-crystal neutron interferometer the wavefunction behind the first plate (acting as a beam splitter) is a linear superposition of the sub-beams belonging to the right ($|I\rangle$) and the left path ($|II\rangle$), which are laterally separated by several centimeters. The sub-beams are recombined at the third crystal plate and the wave function in forward direction then reads as $|\Psi_N\rangle \propto |\Psi_I^I\rangle + |\Psi_{II}^I\rangle$, where $|\Psi_I^I\rangle$ and $|\Psi_{II}^I\rangle$ only differ by an adjustable phase factor $e^{i\chi} (\chi = N_{PS}\delta_c \lambda D)$, with the thickness of the phase shifter plate $D$, the neutron wavelength $\lambda$, the coherent scattering length $b_c$ and the atom number density $N_{PS}$ in the phase shifter plate). By rotating the plate, $\chi$ can be varied systematically. This yields the well known intensity oscillations of the two beams emerging behind the interferometer, usually denoted as O- and H-beam [36]. A sketch of the setup is depicted in Fig. 1.

In our experiment, only the beam in path II is exposed to the rf-field of frequency $\omega$, resulting in a spin flip process. The spin flip configuration of the first rf-field ensures entanglement of spin and spatial degree of freedom of the neutron state [15]. Interacting with a time-dependent magnetic field, the total energy of the neutron is no longer conserved after the spin-flip [41, 42, 43, 44, 45]. Photons of energy $\hbar \omega$ are exchanged with the rf-field. This particular behavior of the neutron is described by the 'dressed-particle' formalism [40, 46]. Consequently the two sub-beams $|I\rangle$ and $|II\rangle$ now differ in total energy (see Fig 1(b)). Therefore the neutron state can be considered to consist of the three subsystems, namely the total energy, path and spin degree of freedom. A coherent superposition of $|I\rangle$ and $|II\rangle$ results in the multiply entangled state vector, expressed as

$$|\Psi(t)\rangle \propto |\alpha_\omega\rangle \otimes |\alpha_{\omega/2}\rangle \otimes \frac{1}{\sqrt{2}} \left( |I\rangle \otimes |\uparrow\rangle + e^{i\omega t} e^{i\chi} |II\rangle \otimes e^{i\phi_\omega} |\downarrow\rangle \right).$$  

(4)
According to the arguments above Eq.(4) can be rewritten as

$$|\Psi(t)\rangle \propto |\alpha_\omega\rangle \otimes |\alpha_{\omega/2}\rangle \otimes \frac{1}{\sqrt{2}} (|I\rangle \otimes |E_0\rangle \otimes |\uparrow\rangle + e^{i\chi} |II\rangle \otimes |E_0 - \hbar \omega\rangle \otimes e^{i\phi_\omega} |\downarrow\rangle )$$  (5)

The spin in path $|II\rangle$ is flipped by a RF-flipper, which requires two magnetic fields: A static field $B_0 \cdot \hat{z}$ and a perpendicular oscillating field $B_{(1)}^{(\omega)} = B_{rf}^{(\omega)} \cos(\omega t + \phi_\omega) \cdot \hat{y}$ satisfying the amplitude

**Figure 1.** (b) Schematic view of the experimental setup for stationary observation of interference between two rf-fields. Showing the arrangement of two radio-frequency flip coils (the first within one path of the skew-symmetric Mach-Zehnder-type neutron interferometer and the other driven by half the frequency behind the interferometer), accelerator coil and $\pi/2$ spin-turner (the purpose of the DC spin-flipper is explained in Sec. 2.2.1). (c) Energy level diagram of the two interfering sub-beams $|I\rangle$, $|II\rangle$ during their passage through the different static field regions ($B_{0,0}/2$), including corresponding energy eigenstates $|E_0\rangle$, $|E_-\rangle := |E_0 - \hbar \omega\rangle$ and spin states $|\uparrow\rangle, |\downarrow\rangle$ and taking into account the spin flips at rf-frequencies $\omega$ and $\omega/2$. (a) Bloch-sphere descriptions to depict evolutions of each quantum state (i.e., spin, path and energy degrees of freedom) in the GHZ measurement. The directions of the projective measurements $P_j$ are depicted by thick red arrows in Bloch spheres.
and frequency resonance condition

\[ B_1^{(\omega)} = \frac{\pi \hbar}{\tau |\mu|} \text{ and } \omega = \frac{2|\mu|B_0}{\hbar} \left(1 + \frac{B_1^2}{16B_0^2}\right), \tag{6} \]

where \( \mu \) is the magnetic moment of the neutron and \( \tau \) denotes the time the neutron is exposed to the RF-field. The second term in \( \omega \) is due to the Bloch-Siegert shift [47]. The oscillating field is produced by water-cooled RF-coil, operating at a frequency of \( \omega/2\pi = 58 \text{ kHz} \). The static field is provided by a uniform magnetic guide field \( B_0^{(\omega)} \sim 2 \text{ mT} \), produced by a pair of water-cooled Helmholtz coils. The state vector of the neutron acquires a phase \( \phi_0 \) during the interaction with the oscillating field \((B(t) = B_1 \cos(\omega t + \phi_0))\), induced by the action of the operators \( \hat{a}_\omega \) and \( \hat{a}_\omega^\dagger \) in the last term of Eq. (1). The neutron part of the total state vector in Eq.(5) represents a path-energy-spin entanglement within a single-neutron system.

The two sub-beams are recombined at the third plate, which is described by the projection operator \( \hat{O}^{(P)} = \frac{1}{2}(|I\rangle + |II\rangle)\langle I| + \langle II|\), resulting in a time-dependent state vector due to the different energies of the two partial wavefunctions. Due to the orthogonality of the energy and spin eigenstates the measured polarization is zero and no intensity modulations are observed in the H-beam:

\[ F_H(t) = \left( \cos(\chi + \pi - \omega t - \phi_\omega), \sin(\chi + \pi - \omega t - \phi_\omega), 0 \right). \tag{7} \]

This behavior [38] is related to the spinor precession known from zero-field spin-echo experiments [42, 43].

The beam recombination of the O-beam is followed by an interaction with the second RF-field, with half frequency \( \omega/2 \). Here the operator

\[ \hat{O}^{(E)} = \frac{1}{\sqrt{2}}|E_0 - \hbar \omega/2\rangle\langle E_0 - \hbar \omega/2| + \langle E_0 - \hbar \omega|, \tag{8} \]

represents mathematically the energy transfer and the total state vector is given by

\[ |\Psi_f\rangle \propto |\alpha_\omega\rangle \otimes |\alpha_{\omega/2}\rangle \otimes (|I\rangle + |II\rangle) \otimes |E_0 - \hbar \omega/2\rangle \]
\[ \otimes \frac{1}{\sqrt{2}} \left( e^{i\phi_\omega/2} |\downarrow\rangle + e^{-i\omega T} e^{i\chi} e^{i(\phi_\omega - \phi_{\omega/2})} |\uparrow\rangle \right). \tag{9} \]

where \( \phi_\omega \) and \( \phi_{\omega/2} \) are the phases induced by the two RF-fields. The second RF-flipper is operating at \( \omega/2\pi = 29 \text{ kHz} \), which is half the frequency of the first RF-flipper. The oscillating field is denoted as \( B_1^{(\omega/2)} \cos((\omega/2)t + \phi_{\omega/2}) \cdot \hat{y} \), and the strength of the guide field was tuned to \( B_0^{(\omega/2)} \sim 1 \text{ mT} \) in order to satisfy the frequency resonance condition. By choosing a frequency of \( \omega/2 \) for the second RF-flipper, the time-dependence of the state vector is eliminated since both components acquire a phase \( e^{\pm i\omega/2(t+T)} \), depending on the spin orientation. Only a constant phase offset of \( e^{-i\omega T} \) remains, which is referred to as zero-field phase, with \( T \) being the neutron’s propagation time between the centre of the first and second RF-flipper coil, remains in the stationary state vector [48]. Hence the second RF-flipper compensates the energy difference between the two spin components, by absorption and emission of photons of energy \( E = \hbar \omega/2 \). Thus, the energy difference between the orthogonal spin states is compensated by choosing a frequency of \( \omega/2 \) for the second RF-flipper, resulting in a stationary state vector, which is graphically illustrated in (see Fig.1(c)). Hence also the time dependence of the polarization vector is eliminated:

\[ \vec{P}_O^{\text{fin}} = (\cos \Delta_{\text{tot}}, \sin \Delta_{\text{tot}}, 0), \tag{10} \]
with
\[
\Delta_{\text{tot}} = (\chi - 2\phi_{\omega/2} + \phi_{\omega} - \omega T),
\] (11)
consisting of the phases induced the path (phase shifter \(\chi\)), spin (phases of the two RF-fields \(\phi_{\omega}, \phi_{\omega/2}\)), and energy manipulation (zero-field phase \(\omega T\)).

Finally, the spin is rotated back by an angle \(\delta = \pi/2\) (in the \(\hat{x}, \hat{z}\) plane) to the \(\hat{z}\)-direction by use of a \(\pi/2\) static field spin-turner. Due to a spin dependent reflection within a Co-Ti multilayer supermirror, the spin is analyzed along the \(\hat{z}\)-direction, denoted as a projection operator \(\hat{O}^{(S)} = |\uparrow\rangle \langle \downarrow|\).

The interference oscillations can be written in form of
\[
I_0 \propto 1 + C \cos(\chi + \Phi - \omega T),
\] (12)
introducing the contrast \(C\) and the relative (spin) phase \(\Phi = \phi_{\omega} - 2\phi_{\omega/2}\). Typical interference patterns, when scanning \(\chi\), for different settings of \(\phi_{\omega}\), are depicted in Fig. 2 (a). In the O-beam an average fringe contrast of \(C = 52.4(2)\) % is achieved, whereas no interference fringes are observed in the H-detector, where no further manipulations are applied. However, even for \(\phi_{\omega} = \phi_{\omega/2} = 0\) the relative phase \(\Phi\) exhibits an offset, as seen in Fig. 2 (a). This is due to the zero-field phase contribution \(\omega T\), but also due to dynamical phase contribution, resulting from

Figure 2. (a) Typical interference patterns of the H- and the O-beam. In the H-beam no interference fringes are observed due to orthogonal spin states in the interfering sub-beams, whereas the O-beam exhibits time-independent sinusoidal intensity oscillations, when rotating the phase shifter plate \(\chi\). A phase shift occurs by varying \(\phi_{\omega}\). Here \(\phi_{\omega/2}\) is kept constant at \(\phi_{\omega/2} = 0\). (b) Relative phase \(\Delta \Phi^\pm\) vs. \(\phi_{\omega}\), and (c) \(\Delta \Phi^\pm\) vs. \(\phi_{\omega/2}\). The sign of the phase depends on the chosen initial polarization. An additional tunable accelerator coil compensates the phase contributions of the constant zero field phase \(\omega T\) and the dynamical phase accumulated by Larmor precession.
Larmor precession within the guide field regions $B^{(\omega)}_0 \cdot \hat{z}$ and $B^{(\omega/2)}_0 \cdot \hat{z}$, denoted as $\delta_{\text{GF} \ I}$ and $\delta_{\text{GF} \ II}$, which have not been mention yet. These contributions are compensated by an additional Larmor precession within a tunable accelerator coil with a static field, pointing in the $\hat{z}$-direction. Thus by choosing

$$\delta_{\text{ACC}} = -(\delta_{\text{GF} \ I} + \delta_{\text{GF} \ II} - \omega T)$$

no more offset is observed for $\phi_\omega = \phi_{\omega/2} = 0$, as shown in Fig. 2 (b) and (c).

2.1.2. Phase measurement results

The experiment was carried out at the neutron interferometer instrument S18 at the high-flux reactor of the Institute Laue-Langevin in Grenoble, France. Here a silicon perfect-crystal monochromator is installed permanently in the neutron guide to monochromatize the incident neutron beam to a mean wave length of $\lambda_0 = 1.91$ Å with the monochromaticity $\Delta \lambda / \lambda_0 \sim 0.015$. The cross section of the beam is roughly 5x5 mm$^2$. Birefringent magnetic field prisms are used to polarize the incident beam in $\hat{z}$-direction, before the beam enters the interferometric setup. The angular separation can be used such that only the spin-up (or spin-down) component fulfills the Bragg-condition at the first interferometer plate (beam splitter).

It is possible to invert the initial polarization simply by rotating the interferometer by a few seconds of arc, thereby selecting the spin-down component to enter the interferometer, which is expected to lead to an inversion of the relative phase. Thus a relative phase difference $\Delta \Phi^\pm = \pm \phi_\omega \mp 2\phi_{\omega/2}$, where $\pm$ denotes the respective initial spin orientation is observed. Figure 2 (a) shows a plot of the relative phase $\Delta \Phi^\pm$ versus $\phi_\omega$, with $\phi_{\omega/2} = 0$, and a phase shift $\Delta \Phi^\pm$ caused by a variation of $\phi_\omega$. As expected, the slope is positive for initial spin up orientation (1.007(8)), and negative for the spin down case (-0.997(5)). In Fig. 2 (b) $\phi_{\omega/2}$ is varied, while $\phi_\omega$ is kept constant, yielding slopes of -1.995(8) and 1.985(7) (since $\Delta \Phi$ is shifted by $-2\phi_{\omega/2}$), depending again on the initial beam polarization.

The arrangement of two RF-flippers of frequencies $\omega$ and $\omega/2$ can be interpreted as an interferometer-scheme for the neutron's total energy. Due to energy splitting the first RF-flipper generates a superposition of two coherent energy states, similar to the action of the first beam-splitter of a Mach-Zehnder interferometer, where a single beam is split spatially into two coherent sub-beams. The second flipper compensates the energy difference and therefore acts as a beam analyzer equivalent to the last beam-splitter of the interferometer. The results of the experiment discussed in this Section can be found in more detail in [48].

2.2. GHZ-States in Neutron Interferometry

The neutron part of the multi entangled statevector given by

$$|\Psi_N\rangle \propto \frac{1}{\sqrt{2}} \left( |\text{I} \rangle \otimes |E_0\rangle \otimes |\uparrow\rangle + |\text{II} \rangle \otimes |E_0 - h\omega\rangle \otimes |\downarrow\rangle \right),$$

denotes a Greenberger-Horne-Zeilinger (GHZ)-like state [16, 17]. A contradiction between quantum mechanics and local hidden variable theories for the GHZ state is found only for perfect situations (which cannot be realized experimentally), an inequality is used to demonstrate the peculiarities of the GHZ state. Mermin analyzed the GHZ argument in detail and derived an inequality pertinent for an experimental test of local hidden variable theories [24]. Since our experiment, GHZ-like entanglement exists between degrees of freedom in a single-neutron system we test noncontextual hidden variable theories using a sum of four expectation values defined as

$$M = E(\sigma_x^{(P)}\sigma_x^{(E)}\sigma_x^{(S)}) - E(\sigma_x^{(P)}\sigma_y^{(E)}\sigma_y^{(S)}) - E(\sigma_y^{(P)}\sigma_x^{(E)}\sigma_y^{(S)}) - E(\sigma_y^{(P)}\sigma_y^{(E)}\sigma_x^{(S)}),$$

(15)
where $\sigma_{x,y}^{(i)}$ are the Pauli operators in path, total energy and spin subspace. The value of $M$ is bounded by 2 for any noncontextual hidden variable theory, whereas quantum mechanics predicts an upper limit of 4 for the GHZ state. The Pauli operators used in Eq.(15) can be decomposed as

$$
\begin{align*}
\sigma_{x}^{(i)} &= \hat{P}^{(P)}(0) - \hat{P}^{(P)}(\pi), \\
\sigma_{y}^{(i)} &= \hat{P}^{(P)}(\pi/2) - \hat{P}^{(P)}(3\pi/2),
\end{align*}
$$

with $\hat{P}^{(P)}(\alpha)$, $\hat{P}^{(E)}(\beta)$ and $\hat{P}^{(S)}(\gamma)$ being the projection operators onto an up-down superposition on the equatorial plane in path, total energy and spin Bloch sphere. The azimuthal angle is given by an angle parameter $\chi$, $\gamma$ and $\alpha$, respectively. The operators are defined as

$$
\begin{align*}
\hat{P}^{(P)}(\chi) &= \frac{1}{\sqrt{2}} (|I\rangle + e^{-i\chi}|I\rangle) (|I\rangle + e^{i\chi}\langle I|) \\
\hat{P}^{(E)}(\gamma) &= \frac{1}{\sqrt{2}} (|E_0\rangle + e^{-i\gamma}|E_0 - \hbar\omega\rangle) (|E_0\rangle + e^{i\gamma}\langle E_0 - \hbar\omega|) \\
\hat{P}^{(S)}(\alpha) &= \frac{1}{\sqrt{2}} (|\hat{\chi}\rangle + e^{-i\alpha}|\hat{\psi}\rangle) (|\hat{\chi}\rangle + e^{i\alpha}\langle \hat{\psi}|),
\end{align*}
$$

where $\chi$, $\gamma$ and $\alpha$ are the azimuthal angles on the Bloch spheres depicted, having only the values 0 and $\pi$ or $\pi/2$ and $3\pi/2$, since only $\sigma_x$ and $\sigma_y$ occur in Eq.(15) (see Fig. 1). Each expectation value $E(\sigma_{x,y}^{(P)}\sigma_{x,y}^{(E)}\sigma_{x,y}^{(S)})$ is experimentally determined by a combination of normalized count rates, using appropriate setting of $\chi$, $\gamma$ and $\alpha$. For instance

$$
E(\sigma_{x}^{(P)}\sigma_{y}^{(E)}\sigma_{y}^{(S)}) = E(\alpha : (0; \pi), \beta : (\frac{\pi}{2}; 3\pi), \gamma : (\frac{\pi}{2}; \frac{3\pi}{2})) \\
= \langle \Psi_{GHZ}|(\hat{P}(0)^{(P)} - \hat{P}(\pi)^{(P)}) \otimes (\hat{P}(\pi/2)^{(E)} - \hat{P}(3\pi/2)^{(E)}) \otimes (\hat{P}(\pi)^{(S)} - \hat{P}(3\pi)^{(S)})|\Psi_{GHZ}\rangle = \frac{A}{B}
$$

with

$$
\begin{align*}
A &= \left( N(0, \frac{\pi}{2}, \frac{\pi}{2}) - N(\pi, \frac{\pi}{2}, \frac{\pi}{2}) - N(0, \frac{3\pi}{2}, \frac{3\pi}{2}) + N(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}) \right) \\
&\quad - \left( N(0, \frac{3\pi}{2}, \frac{\pi}{2}) - N(\pi, \frac{\pi}{2}, \frac{\pi}{2}) + N(0, \frac{3\pi}{2}, \frac{3\pi}{2}) + N(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}) \right),
\end{align*}
$$

and

$$
\begin{align*}
B &= \left( N(0, \frac{\pi}{2}, \frac{\pi}{2}) + N(\pi, \frac{\pi}{2}, \frac{\pi}{2}) + N(0, \frac{3\pi}{2}, \frac{3\pi}{2}) + N(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}) \right) \\
&\quad + \left( N(0, \frac{3\pi}{2}, \frac{\pi}{2}) - N(\pi, \frac{3\pi}{2}, \frac{\pi}{2}) + N(0, \frac{3\pi}{2}, \frac{3\pi}{2}) + N(\pi, \frac{3\pi}{2}, \frac{3\pi}{2}) \right),
\end{align*}
$$

where for example $N(0, \pi/2, \pi/2)$ is the count rate for $\chi = 0$, $\gamma = \pi/2$ and $\alpha = \pi/2$.

### 2.2.1. Measurement procedure

As seen from $\Delta_{tot}$ in Eq.(11) in principal each of the three degrees of freedom can be manipulated independently and the associated observables are separately measurable. A schematic illustration of the setup, together with a Bloch sphere description of the associated measurement directions as path, spin and total energy are depicted in Fig. 1. The projective measurements of each degree of freedom are carried out exploiting to phase manipulations
between the eigenstates of the individual sub systems, resulting in the desired azimuthal angle on the Bloch sphere.

Path phase: The phase manipulation of the path subspace is accomplished with the auxiliary phase shifter made of a parallel-sided Si plate. Typical oscillations when rotating the phase shifter slab are depicted in Fig. 3.

Spin phase: In this experiment the tunable accelerator coil was used to tune the spin phase $\alpha$. In particular the spin phase is accumulated by Larmor precession within the static magnetic field of the accelerator coil ($B_{ACC} \cdot \hat{z}$) with $\alpha = \omega_L \tau_{ACC}$, where $\omega_L$ is the Larmor frequency depending on $B_{ACC}$ and $\tau_{ACC}$ is the propagation time through the accelerator coil.

Energy phase: As explained above the manipulation of the energy phase is achieved by zero-field precession between the two RF-flippers. The first RF spin-flipper induces the energy difference $\hbar \omega$, which is balanced by the second RF flipper by choosing a frequency of $\omega/2$, resulting in the zero field phase difference $\gamma = \omega T$. Here $T$ can be varied, since the second RF coil is mounted on a translation stage. This displacement of the second RF-flipper is the crucial point in this experiment, since by increasing the distance between the RF-flippers not only the zero field phase $\gamma = \omega T$ is changed, but also the Larmor precession angle within the static guide field $B_0$ which induces an additional undesired spin phase contribution $\alpha' = \omega_L (B_0)$.

However the aim is to address the zero field precession independently from Larmor precession. This is achieved by an auxiliary DC-flipper, which is mounted on the same translation stage subsequently to the second RF-flipper. Thus no additional phase shift, induced by Larmor precession, resulting from the change of $\Delta d$ is observed. Phase contributions with the same sign occur in the regions between first and second RF-flippers ($d + \Delta d$) and between DC-flipper and the DC $\pi/2$ spin-rotator (denoted $d' - \Delta d$), compensating each other. Thus the total Larmor precession angle remains constant ($(d + d')\omega_L (B_0)$), thought changing the position ($\Delta d$) of the second RF-flipper. The displacement only affects the zero field precession angle $\gamma = \omega T \propto \omega(d + \Delta d)$. To demonstrate this individual tuning of Larmor and zero field precession an independent polarimetric experiment was carried out [49].

Figure 3. Typical interference patterns of the O-detector obtained by varying the path phase $\chi$. The phases $\alpha$ and $\gamma$, for the spin and the energy, respectively, are tuned at 0, $\pi/2$, $\pi$ and $3\pi/2$ in order to accomplish project measurements associated with $\hat{P}^j(0)$, $\hat{P}^j(\pi/2)$, $\hat{P}^j(\pi)$ and $\hat{P}^j(3\pi/2)$, with $j =$ spin, path and energy.
Table 1. The four experimentally determined expectation values and the final $M$ value.

| Observable Settings | Determined Values |
|---------------------|------------------|
| $\sigma_y^{(P)} \sigma_y^{(E)} \sigma_y^{(S)}$ | $\chi,$ $\gamma,$ $\alpha$ |
| $(0; \pi)$ | $(0; \pi)$ | $(0; \pi)$ | 0.659(2) |
| $\sigma_y^{(P)} \sigma_y^{(E)} \sigma_y^{(S)}$ | $(0; \pi)$ | $(\pi/2; 3\pi/2)$ | $(\pi/2; 3\pi/2)$ | -0.603(2) |
| $\sigma_y^{(P)} \sigma_y^{(E)} \sigma_y^{(S)}$ | $(\pi/2; 3\pi/2)$ | $(0; \pi)$ | $(\pi/2; 3\pi/2)$ | -0.664(2) |
| $\sigma_y^{(P)} \sigma_y^{(E)} \sigma_y^{(S)}$ | $(\pi/2; 3\pi/2)$ | $(\pi/2; 3\pi/2)$ | $(0; \pi)$ | -0.632(2) |

$M = 2.558(4)$

2.2.2. Experimental results

For the required projective measurements spin phase $\alpha$ and energy phase $\gamma$ each was tuned at $0, \pi/2$ and $3\pi/2$, while path phase scans $\chi$ (i.e., oscillation measurements) were performed. The resulting sixteen oscillations are depicted in Fig. 3, the dashed lines denote the values $\chi = 0, \pi/2$ and $3\pi/2$, which are required for the determination of $M$, as defined in Eq.(15). The average contrast of the oscillations were just below 70 per cent, which is clearly above the threshold visibility of 50 per cent, required for a violation of the Mermin like inequality.

Measured intensity oscillations were fitted to sinusoidal curves by applying a least squares method. The four expectation values, as defined in Eq.(15), were extracted from the fit curves. Statistical errors were estimated to be about ±0.001, taking all fit errors from single measurement curves into account. One set of measurements consists of thirty-two oscillation measurements, since intensities with and without spin flipper (reference measurement) were recorded. This was done in order to allow corrections of the path phase $\chi$ instability, afterward. Four sets of thirty-two oscillations to reduce statistical errors. A final result of $M = 2.558 \pm 0.004$, exhibiting a clear violation of the classical limit $M = 2$ of the noncontextual border. The individual results of the four expectation values, together with settings of variables and the final value of $M$, are listed in Tab. 1. A more detailed discussion of this measurement is given in [50].

3. Polarimetric Experiment

Neutron polarimetry has several advantages compared to perfect crystal interferometry. It is insensitive to ambient mechanical and thermal disturbances and therefore provides better phase stability. Efficiencies of the manipulations, including state splitting and recombination, are considerably high (typically > 98\%) resulting in a better contrast compared to interferometry. In addition, while single-crystal interferometers accept neutrons only within an angular range of a few arc seconds, which leads to a significant decrease in intensity, polarimeters accept beams with a broad momentum distribution. Neutron polarimetry has been used to demonstrate fundamental quantum-mechanical properties. Just to mention the noncommutation of the Pauli spin operator [51] and a number of geometric phase measurements [52, 53, 54]. In recent experiments a test of an alternative model for non-local correlations [55], as well as demonstration of a universally valid uncertainty relation [56] has been performed successfully.

3.1. GHZ-States in Neutron Polarimetry

In this experiment we report on a correlation measurement in the regime of neutron polarimetry, where three independent degrees of freedom, namely total-energy, momentum and spin, are utilized and manipulated separately. The motion of a free neutron propagating in +$y$-direction, with polarization in arbitrary direction, in a static magnetic field with a field gradient parallel to its propagation direction (with $\vec{B}(y) = 0, \forall y > 0$ and $\vec{B}(y) = B_0 \cdot \vec{e}_z = B_z(y), \forall y > 0$) is
where the polarization vector is given by

\[ \vec{P} = \langle \Psi | \hat{\sigma} | \Psi \rangle, \]

where the azimuthal angle \( \alpha \), being the relative phase between \( | \uparrow \rangle \) and \( | \downarrow \rangle \), equals \( 2y \Delta k \). The contrast of the measured oscillations, due to Larmor precession of the polarization, vanishes for large \( \Delta k \), when the spatial separation of the wave packets exceeds the coherence length \( (\ell_c \sim \lambda^2 / \Delta \lambda) \). This is referred to as longitudinal Stern-Gerlach effect. From this effect one can infer that another two-level system consisting of accelerated and decelerated part shows up. This system can be included in our representation by

\[ | \Psi_{\text{Bell}} \rangle = \frac{1}{\sqrt{2}} (| k_- \rangle \otimes | \uparrow \rangle + | k_+ \rangle \otimes | \downarrow \rangle), \]

where \( | k_\pm \rangle \) is the eigenvector of the momentum operator \( \hat{P}_\pm = \hbar \hat{k}_\pm \) with

\[ \hat{k}_\pm | k_\pm \rangle = k_\pm | k_\pm \rangle. \]

### 3.2. Experimental setup and strategy

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 \( \lambda = 1.99 \text{ Å} \) and spectral width of \( \Delta \lambda / \lambda = 0.02 \), is polarized to \( r \sim 99 \% \) 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 \(^3\text{He}\) monitor detector is used for normalization in order to correct statistical fluctuations of the reactor power. A BF\(_3\) detector with high efficiency. (more than 99 \%) is used for count rate detection. A final maximum intensity of about 500 counts/s at a cross section of 10 x 10 mm\(^2\) was recorded. To avoid unwanted depolarization a uniform guide field in Helmholtz configuration, pointing in +z-direction with a strength of about 13 Gauss is applied over the entire setup. A schematic illustration of the experimental arrangement is depicted in Fig.4(a).

In the present experiment the spin-energy entanglement is achieved by a single RF-\( \pi/2 \) spin-rotator. The RF-flipper is operating at a frequency of \( \omega_{rf} / 2\pi = 40 \text{ kHz} \). The effective field, perpendicular to the initial polarization, is adjusted such that it induces a spin flip with a probability of \( 1/2 \) \( (B_1^{(\omega)} = \pi \hbar / (2 \tau |\mu|) \), with \( \mu \) as magnetic moment and \( \tau \) as propagation
time in RF-field region). Therefore only the flipped spin component is affected by the energy
manipulation yielding an entangled state vector, which can be represented as a Bell state

\[ |\Psi_{\text{Bell}}\rangle = \frac{1}{\sqrt{2}} \left( |E_0\rangle \otimes |\uparrow\rangle + |E_0 - \hbar \omega\rangle \otimes |\downarrow\rangle \right). \]  
(27)

Together with the time dependent interaction within the first RF-coil and the static magnetic
pointing in +z-direction, denoted as \( B_{\text{acc}} \), the total system can be described by a GHZ-like state given by

\[ |\Psi_{\text{GHZ}}\rangle = \frac{1}{\sqrt{2}} \left( |\uparrow\rangle \otimes |k_-\rangle \otimes |E_0\rangle + |\downarrow\rangle \otimes |k_+\rangle \otimes |E_0 - \hbar \omega\rangle \right). \]  
(28)

Each of the three degrees of freedom can be manipulated independently and the associated observables are separately measurable, which is essential for determining a sum of four expectation values defined in Eq.(15) where \( \sigma^{(i)}_{x,y} \) are the Pauli operators in spin, total energy and momentum subspace. The projection operators, required for the decomposition of the Pauli operators (see Eq.(16)) are defined as

\[ \hat{P}^{(S)}(\alpha) = \frac{1}{\sqrt{2}} (|\uparrow\rangle + e^{-i\alpha}|\downarrow\rangle)(\langle\uparrow| + e^{i\alpha}\langle\downarrow|) \]
The measurement direction of the momentum phase is tuned by the propagation time within the accelerator coil. The acquired phase in momentum space is given by

$$\pi/2 \text{ spinor-rotation is performed. Typical intensity modulations when scanning the second RF spin-rotator denoted as } B_2. \text{ The measurement direction of the momentum phase } \beta \text{ and energy phase } \gamma \text{ is tuned by the position of the second RF-} \pi/2 \text{ spinor-rotation again in the } \hat{x}\hat{y} \text{-plane. Typical intensity oscillations when scanning } \alpha \text{ are depicted in Fig. 5, where an average contrast of 0.9835(27) was achieved.}

The measurement direction of the momentum phase is tuned by the propagation time within the accelerator coil. The acquired phase in momentum space is given by

$$\beta = \int B_{acc} ds.$$ 

However in practice the strength of the magnetic field was varied, instead of the length, due to experimental convenience. The accelerator coil, just like the uniform guide field, is realized in Helmholtz configuration, for higher field homogeneity and accurate alignment of accelerator and guide field.

The measurement direction of the energy phase is tuned by 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. 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 as \Delta \delta = \omega L \Delta L/v.

**Figure 5.** Typical intensity oscillation when varying the spin phase \(\alpha\) for different setting of the momentum phase \(\beta\) and energy phase \(\gamma\).

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

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

where \(\alpha\), \(\beta\) and \(\gamma\) are the azimuthal angles on the Bloch spheres, having only the values 0 and \(\pi\) or \(\pi/2\) and \(3\pi/2\), sine only \(\sigma_x\) and \(\sigma_y\) (see Fig. 4 (b)). Each expectation value \(E(\sigma_{x,y}^{(S unavoidable 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 ($\sim$ 2000m/s). To retrieve a pure tuning of the energy phase $\gamma$ this additional Larmor phase has to be compensated as in the interferometer experiment. This is done by appropriately tuning the phase of the oscillating magnetic field in RF1, yielding a reversed spin phase shift of $-\Delta \delta$. 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. 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.

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. Thus, when varying $L$ there should be no intensity modulation. In practice this does not hold, due to the so called Bloch-Siegert shift [47].

The second RF-$\pi/2$ spin-turner converts the $\hat{x}$-component of the polarization back to the $\hat{z}$-direction to be analyzed by the second supermirror. This is expressed by applying a projection operator for the spin

$$\hat{P}^{(S)} = | \uparrow \rangle \langle \uparrow |.$$  

Finally the stationary intensity oscillations are given by

$$N(\alpha, \beta, \gamma) = \frac{1}{2} \left( 1 + C \cos(\alpha + \beta + \gamma) \right), \quad (30)$$

with $C$ being the contrast, experimentally determined as 0.9844(9). 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 (which are plotted in Fig. 5) were carried out for a determination of $M$, yielding a final value $M = 3.936(2)$. The individual results for each expectation value are summarized in Tab. 2.

In conclusion, the obtained violation of a Mermin-like inequality for a triply entangled GHZ-like state in a single-neutron system clearly confirm the prediction of quantum mechanics in explicit terms, seen in the final value $M = 3.936(2) \gg 2$. The deviation of less than 2 per cent from the theoretical value $M_{\text{theo}} = 4$ is worth noting here. This work has been partly supported by the Austrian Science Foundation, FWF (P17803-N02).

### 4. Conclusion and Outlook

Multi-entanglement has been studied in perfect Si-crystal interferometer experiments as well as polarimetric measurements. Entanglement of various degrees of freedom has been prepared and analyzed in detail. In Section 2.1 a technique of coherent energy manipulation in neutron interferometry has been presented. By utilizing the interferometer in combination with two RF fields time-independent interference patterns have been observed. After successful preparation and manipulation of a triply entangled GHZ-like state (i.e. entanglement of spin, path and energy degree of freedom), presented in Section 2.2, further investigation in the field of multi-entanglement in neutron interferometry are anticipated. In a next step preparation of a W-state, which requires three RF-flippers inside a neutron interferometer operating at different

### References

[47] Bloch, Siegert shift.
frequencies, is expected. In addition, generation of these typical triply entangled (pure) states will allow further mixture of these states. For instance measurements of gradual translation from the GHZ-state to the W-state are of special interest. At the second stage, neutron interferometer measurements will be carried out in order to determine and distinguish a variety of entangled states, by means of state tomography or entanglement witness.

Concerning neutron polarimetry, in addition to the experiments with spin-energy entangled states, induced by RF-flippers, multi-energy splitting, using Ramsey’s resonance method of separated oscillating fields [57], will be topic of a forthcoming experiment.

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