Since the pioneering work of Einstein, Podolsky, and Rosen [1] numerous experiments have exploited the concept of Nonlocality which tests local hidden variable theories (LHVTs). The LHVTs are a subset of a larger class of hidden-variable theories namely the noncontextual hidden-variable theories (NCHVTs). Noncontextuality implies that the value of a measurement is independent of the experimental context, i.e. of previous or simultaneous measurements [2,3]. 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 [4].

In the case of neutron experiments, entanglement is not achieved between particles, but between different degrees of freedom. Since the observables in different Hilbert spaces commute with each other, the single neutron system is suitable for studying NCHVTs. Single-particle entanglement, between the spinor and the spatial part of the neutron wave function [5], as well as full tomographic state analyses [6] have already been accomplished. In addition, the contextual nature of quantum theory [7] has been demonstrated using neutron interferometry [8]. Aiming at the preparation of a single-particle multiply entangled state, implementation of another degree of freedom to be entangled with the neutron's spin and path degrees of freedom was a challenge.

The neutron's energy seems to be an almost ideal candidate for this third degree of freedom, due to its experimental accessibility within a magnetic resonance field [9]. 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 [10,11]. The J-C Hamiltonian can be adopted for a system consisting of a neutron coupled to a quantized rf-field [12].

This letter reports on observation of stationary interference patterns, confirming coherent energy manipulation of the neutron wavefunction. This technique provides realization of triple-entanglement between the neutron’s path, spin and energy degrees of freedom.

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

$$
\mathcal{H}_{J-C} = -\frac{\hbar^2}{2m} \nabla^2 - \mu B_0(r) \sigma_z + \hbar (\omega a^\dagger_\omega a_\omega + \frac{\omega}{2} a^\dagger_{\omega/2} a_{\omega/2}) \\
+ \mu \left( \frac{B_1^{(\omega)}(r)}{\sqrt{N_\omega}} (a^\dagger_\omega \tilde{\sigma} + h.c.) + \frac{B_1^{(\omega/2)}(r)}{\sqrt{N_{\omega/2}}} (a^\dagger_{\omega/2} \tilde{\sigma} + h.c.) \right),
$$

with $\tilde{\sigma} = \frac{1}{2} (\sigma_x + i \sigma_y)$. 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 $a^\dagger$ and $a$. Finally, the last term represents the coupling between photons and the neutron, where $N_\omega = \langle a^\dagger_\omega a_\omega \rangle$ 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 $|\psi(r)\rangle$ and the (time-dependent) energy $|E(t)\rangle$ subspaces of neutrons, respectively [13].

The state vectors of the oscillating fields are represented by coherent states $|\alpha\rangle$, which are eigenstates of $a^\dagger$ and $a$. The eigenvalues of coherent states are complex numbers, so one can write $a|\alpha\rangle = |\alpha\rangle = |\alpha| e^{i\phi}|\alpha\rangle$ with $|\alpha| = \sqrt{N}$. 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: $|\Psi_1\rangle = |\alpha\rangle \otimes |\alpha_{\omega/2}\rangle \otimes |\Psi_N\rangle$. 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 superposed at the third crystal plate and the wave function in the forward direction then reads as $|\Psi_N\rangle \propto |\Psi_N^{(I)}\rangle + |\Psi_N^{(II)}\rangle$, where $|\Psi_N^{(I)}\rangle$ and $|\Psi_N^{(II)}\rangle$ only differ by an adjustable phase factor $e^{i\chi}$ ($\chi = N_{ps} b_c \lambda D$, with the thickness of the phase shifter plate $D$, the neutron wavelength $\lambda$, the coherent scattering length $b_c$ and the particle 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 [8]. A
multiply entangled dressed state vector, expressed as
\[
|\Psi(t)\rangle \propto |\alpha_{\omega}\rangle \otimes |\alpha_{\omega/2}\rangle \otimes \frac{1}{\sqrt{2}} \left(|I\rangle \otimes |E_0\rangle \otimes |\rangle\right) + e^{i\chi}|\rangle \otimes e^{i\omega t}|E_0 - \hbar \omega\rangle \otimes e^{i\phi_{\omega/2}|\rangle\rangle},
\] (2)

where $|\rangle, |\rangle$ denote the neutron’s up and down spin states referred to the chosen quantization axis. The state vector of the neutron acquires a phase $\pm \phi_{\omega}$ during the interaction with the oscillating field, given by $B(t) = B_1 \cos(\omega t + \phi_{\omega})$, induced by the action of the operators $a_{\omega}$ and $a_{\omega}^\dagger$ in the last term of Eq. (1). The neutron part of the total state vector is represented by a path-energy-spin entanglement within a single neutron system. At the last plate of the interferometer (region 4) the two sub-beams are recombined, which is described by the projection operator $\hat{O}^{(E)} = \frac{1}{2}(|I\rangle + |II\rangle)(|I\rangle + |II\rangle)$. Due to the orthogonality of the energy and spin eigenstates the polarization is zero and no intensity modulations are observed in the H-beam, which is plotted in Fig. 2. A time-resolved measurement (see [9]) can reveal the dynamic behavior of the polarization expressed as
\[
\hat{P}_O(t) = \left(\cos(\chi - \omega t - \phi_{\omega}), \sin(\chi - \omega t - \phi_{\omega}), 0\right). \quad (3)
\]

This phenomenon has been measured separately [9], and is related to the spinor precession known from zero-field spin-echo experiments [15, 16].

The beam recombination is followed by an interaction with the second rf-field, with half frequency $\omega/2$, in region 5. Mathematically the energy transfer is represented by the operator $\hat{O}^{(E)} = \frac{1}{\sqrt{2}}(|E_0 - \hbar \omega/2\rangle \langle E_0 - \hbar \omega/2| + |E_0\rangle \langle E_0|)$, respectively. The total state vector is given by
\[
|\Psi_1\rangle \propto |\alpha_{\omega}\rangle \otimes |\alpha_{\omega/2}\rangle \otimes (|I\rangle + |II\rangle) \otimes |E_0 - \hbar \omega/2\rangle
\]
\[
\otimes \frac{1}{\sqrt{2}} \left(e^{i\phi_{\omega/2}|\rangle\rangle} + e^{i\omega T} e^{i\chi} e^{i(\phi_{\omega/2} - \phi_{\omega}|\rangle\rangle}\right), \quad (4)
\]

where $\phi_{\omega}$ and $\phi_{\omega/2}$ are the phases induced by the two rf fields and $\omega T$ is the zero-field phase, with $T$ being the neutron’s propagation time between the two rf-flippers [22]. 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. Hence the time dependence of the polarization vector is eliminated:
\[
\hat{P}_1 = (\cos \Delta_{\text{tot}}, \sin \Delta_{\text{tot}}, 0), \quad (5)
\]

where $\Delta_{\text{tot}} = (\chi - 2\phi_{\omega/2} + \phi_{\omega} + \omega T)$, consists of the phases induced by the path (phase shifter $\chi$), spin (phases of the two rf fields $\phi_{\omega}, \phi_{\omega/2}$), and energy manipulation (zero-field phase $\omega T$). The principle of energy compensation is visualized in Fig. (b). As seen from $\Delta_{\text{tot}}$ in Eq. (5)}
of the three degrees of freedom can be manipulated independently and the associated observables are separately measurable.

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.

After applying a projection operator $\hat{P}^{(S)} = |\uparrow\rangle\langle\uparrow|$ to the spin (region 6), the stationary interference oscillations are given by $I_0 \propto 1 + \nu \cos(\chi + \Phi + \omega T)$, introducing the fringe visibility $\nu$ and the relative phase $\Phi$. The relative phase can be calculated as $\Phi = \phi_\omega - 2\phi_{\omega/2}$. In the following experiment we demonstrate the coherence property of the modified J-C manipulation defined in Eq. (11) as well as the phase dependence expressed above.

The experiment was carried out at the neutron interferometer instrument S18 at the high-flux reactor of the Institute Laue-Langevin in Grenoble, France. A monochromatic beam, with mean wavelength $\lambda_0 = 1.91\ \text{Å} (\Delta \lambda/\lambda_0 \sim 0.02)$ and 5x5 mm$^2$ beam cross-section, is polarized by a bi-refringent magnetic field prism in the $z$-direction, see Fig. (11a) region 1. In a non-dispersive arrangement of the monochromator and the interferometer crystal 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) in region 2. Behind the beam splitter the neutron’s wave function is found in a coherent superposition of $|\Psi^{(I)}_N\rangle$ and $|\Psi^{(II)}_N\rangle$, and only $|\Psi^{(I)}_N\rangle$ passes the first rf-flipper mounted in one path of the interferometer. Acting like a typical NMR arrangement, rf-flippers require two magnetic fields: A static field $B_0 \cdot \hat{z}$ with $B_0 = \hbar \omega T/(2|\mu|)$ and a perpendicular oscillating field $B_1^{(\omega)} \cos(\omega t + \phi_\omega) \cdot \hat{y}$ with amplitude $B_1^{(\omega)} = \pi \hbar/(2\tau |\mu|)$, where $\mu$ is the magnetic moment of the neutron and $\tau$ is the time the neutron requires to traverse the rf-field region. The oscillating field is produced by a water-cooled rf-coil with a length of 2 cm, operating at a frequency of $\omega/2\pi = 58\ \text{kHz}$. The static field is provided by the uniform magnetic guide field $B_0 \sim 2\ \text{mT}$, which is produced by a pair of water-cooled Helmholtz coils. However, outside the rf-coil the Larmor precession around the static magnetic guide field induces an additional phase.

The two sub-beams are recombined at the third plate (region 4) resulting in a time-dependent state vector due to the different energies of the two partial wavefunctions. Since the two superposed spin states are orthogonal, no intensity modulation is observed, as seen at the H-detector. In contrast, the O-beam (forward direction) passes the second rf-flipper, operating at 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 about 1 mT in order to satisfy the frequency resonance condition.

This flipper compensates the energy difference between the two spin components, by absorption and emission of photons of energy $E = \hbar \omega/2$. The phases of the two guide fields and the zero-field phase $\omega T$ were compensated by an additional Larmor precession within a tunable accelerator coil with a static field, pointing in the $z$-direction. Finally, the spin is rotated back to the $\hat{z}$-direction by use of a $\pi/2$ static field spin-turner, and analyzed along the $z$-direction due to the spin dependent reflection within a Co-Ti multi-layer supermirror. Typical interference patterns are depicted in Fig. (2). In the O-beam a fringe contrast of 52.4(2)% is achieved, whereas no oscillation was observed in the H-detector, where no further manipula-
tions were applied.

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. In order to observe a relative phase shift, in practice it is necessary to perform a reference measurement. This is achieved by turning off the rf-flipper inside the interferometer, thus yielding the relative phase difference \( \Delta \Phi^\pm = \pm \phi_\omega + 2\phi_\omega/2 \), where \( \pm \) denotes the respective initial spin orientation. Figure 3(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. 3(b) \( \phi_\omega/2 \) is varied, while \( \phi_\omega \) is kept constant, yielding slopes of -1.995(8) and 1.985(7), depending again on the initial beam polarization.

At this point the geometric nature of \( \Delta \Phi^\pm \) should be emphasized. Within the rf-flipper that is placed inside the interferometer, the neutron spin traces a semi-great circle from \(|\uparrow\rangle\) to \(|\downarrow\rangle\) on the Bloch sphere and returns to its initial state \(|\downarrow\rangle\) when passing the second rf-flipper. This procedure is repeated along different semi-great circles when varying \( \phi_\omega \) or \( \phi_\omega/2 \) respectively. The two semi-great circles enclose an angle \( \phi_\omega - \phi_\omega/2 \) and hence a solid angle \( \Omega = 2(\phi_\omega - \phi_\omega/2) \). The solid angle \( \Omega \) yields a pure geometric phase \( \Phi^\pm_G = \Omega/2 \) as in [21][25].

Our work can be seen within a framework related to tripartite entanglement. There are two non-equivalent classes of tripartite entanglement represented by the Greenberger-Horne-Zeilinger (GHZ) state [26][27] and the W state [28] when the three quantum subsystems have non-local correlations. Classification of a GHZ-like state in a single neutron system will be the subject of forthcoming work. In addition, we claim that preparation of other types of triple entanglement can be realized using neutron interferometry and spin precession. For instance creation of a W state can be achieved with rf-flippers within a double loop interferometer. It is worth noting, that the operation of the rf-flipper within the interferometer could be interpreted as a "CNOTNOT-gate", with path as control qubit and energy and spin as target qubits.

In summary, we have established a technique of coherent energy manipulation, by utilizing the neutron interferometer in combination with two rf-fields to observe time-independent interference patterns. Energy splitting provides an additional degree of freedom, available for multiple entanglement of path, spin and energy of the neutron. Our data verify theoretical predictions and illustrate the significance of single particle entanglement.

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