Magnetic Quantum-Phase Control between Two Entangled Macroscopic Nuclear Ensembles

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Heralded generation and manipulation of quantum entanglement between two macroscopic and spatially separated crystals at room temperature is theoretically studied. We show that by combining an x-ray parametric down-conversion source and x-ray interferometry with nuclear resonant scattering techniques, two macroscopic crystals hosting Mössbauer nuclei located each on an interferometer arm can be entangled for few tens of nanoseconds. The coherence time of the entanglement state can be prolonged up to values comparable to the lifetime of a single nuclear excited state, on the order of hundred nanoseconds. A non-mechanical magnetic control of the quantum phase between the two spatially separated entangled nuclear crystals is put forward.

The ability to create entanglement between quantum memories in a heralded manner is vital for quantum communication developments, in particular for quantum repeaters and quantum networks. The search for scalable quantum repeaters has come up with solid-state resources, which require entanglement between quantum memories hosted in spatially separated macroscopical crystals. So far, such macroscopical entanglement has been typically limited in either time duration, working temperature or sample size/number of atoms involved, as shown in Table I that lists some key achievements. As generic feature, these experiments make use of optical photons as entanglement carriers as well as for generation of quantum entanglement in the keV metrology and information technology, and x-ray two-photon absorption extend the frontiers of quantum optics towards higher frequencies. The field of x-ray interferometry, sum-frequency generation of x-ray and optical wave, XFEL and achievements in non-linear x-ray optics, in particular for quantum repeaters and quantum networks is vital for quantum communication applications. We show that a setup comprising nuclear forward scattering (NFS) and x-ray interferometry can be superior to previously employed schemes providing longer coherence times (~100 ns), room temperature handling and larger samples (~10^18 atoms). Furthermore, an alternative function of the same setup involving lattice mechanical excitations in the crystal (phonons) opens the possibility to explore the boundary between quantum realm and the classical world and test decoherence models.

In NFS experiments, a monochromatic x-ray pulse resonant to a nuclear transition energy, i.e., 14.4 keV for 57Fe, coherently propagates through the Mössbauer solid-state sample and is detected in the forward direction. The NFS technique predates the XFEL originally making use of synchrotron radiation (SR) pulses which are monochromatized to meV bandwidths. When impinging on a nuclear crystal, each SR pulse creates at most a single delocalized excitation known as a nuclear exciton state \(|E\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i\vec{k} \cdot \vec{r}_i} |g\rangle |e_i\rangle\). Here, the \(i\)th nucleus at position \(\vec{r}_i\) is excited by the incident x-ray with the wave number \(\vec{k}\), whereas all other \((N-1)\) nuclei stay in their ground state. Intuitively, a single nuclear sample crystal can be divided into two remote parties labeled by \(L\) and \(R\), leading to the formation of the following entangled state \(|ME\rangle\) between two distant parties

\[
|ME\rangle = \frac{1}{\sqrt{2}} \left( |E\rangle_{L} |G\rangle_{R} + e^{i\phi} |G\rangle_{L} |E\rangle_{R} \right),
\]

where \(|E\rangle_{L(R)}\) and \(|G\rangle_{L(R)}\) stand for the \(L(R)\) ensemble being in the excited state \(|E\rangle\) and in the ground state, respectively, and \(\phi\) is the relative phase between the two components. This corresponds to the single-photon version of quantum entanglement, i.e., the entanglement of two field modes by a single photon, that has been successfully used to create heralded quantum entanglement between two crystals with optical photons.

The nuclear exciton in NFS has remarkable properties that support the study of quantum entanglement between macroscopic samples: (1) the size of the nuclear sample crystals subject to x-ray irradiation is macroscopic with a typical dimension of mm×mm×\(\mu\)m and (2) NFS experiments are typically performed at room temperature or even higher temperatures reached in the sample due to the bombardment.
FIG. 1: (Color online) (a) A combination of x-ray interferometry with nuclear resonant scattering and an XPDC setup. X → X + EUV down-conversion occurs within a diamond crystal (yellow cuboid). Subsequently, a converted single x-ray signal photon (red arrow) enters an x-ray interferometer while a converted EUV idler photon (green arrow) reaches detector A producing a click. Beam splitter BS 1 transfers the signal photon into a two-path entanglement state |TPE⟩ = (|1⟩_L|0⟩_R + i|0⟩_L|1⟩_R)/√2. The |TPE⟩ photon is then subject to NFS as it impinges on two 57Fe crystals (green slabs). The nuclear transitions in the latter experience hyperfine splitting under the action of the applied magnetic fields _B_ _L_ and _B_ _R_ (blue short arrows). As the |TPE⟩ single photon is absorbed and shared by the two distant nuclear crystals, the latter are entangled in the state |ME⟩ = (|E⟩_L|G⟩_R + i|G⟩_L|E⟩_R)/√2. The re-emitted signal photon from the nuclear crystals is in turn reflected by the mirror, recombined at beam splitter BS 2 and registered by either detector B or C. (b) 57Fe nuclear level structure. A linear polarized x-ray signal photon drives two Δm = 0 transitions (red arrows). (c)(d) Dynamics of the nuclear currents (rotating orange arrows) on left and right arm of the interferometer induced by the time-dependent magnetic fields _B_ _L_ and _B_ _R_ (blue solid lines), respectively. _B_ _R_ is inverted at switching time _T_ _φ_. (e) Interference pattern _Q_ (see text) at detector B and C for different _T_ _φ_. The light green downward arrow indicates the moment a click at detector A starts the chronometer for _T_ _φ_.
ple crystals, either both detectors A and B, or both detectors A and C simultaneously register the two XPDC photons. Each successful creation of the entanglement state $|\text{ME}\rangle$ is heralded by the click at detector A while no photon is registered at detectors B, C (registering coherent decay of nuclear exciton) or any other detectors monitoring the $4\pi$ emission angle (for photon loss or incoherent, spontaneous decay of the nuclear exciton). The missing count of an x-ray signal photon is attributed to the absorption by the two remote crystals. The absorbed and rescattered signal photon reaches the detectors B or C only later, with a time delay on the order of the nuclear excited state lifetime, i.e., approx. 100 ns. Due to detection efficiency (though in principle very high for x-rays \cite{19}) or incoherent decay processes in $4\pi$ solid angle, the original lack of signal at detectors B and C might just be the consequence of photon loss. However, in this case the click at detector A will not be followed by any further photons registered with detectors B and C (the 14.4 keV photon background is negligible), such that no misleading events may be recorded.

The typical bandwidth of the down-converted photons is around 1 eV \cite{22} corresponding to a pulse duration of 1 fs. This bandwidth is much broader than the linewidth of the interacting nuclear transition such that NFS can be treated as the coherent propagation of an ultrashort pulse through the resonant nuclear media \cite{55, 56}. Due to the hyperfine magnetic field, each $^{57}$Fe 14.4 keV nuclear transition is split into a sextet as illustrated in Fig. 1(b). Linearly polarized x-rays will drive simultaneously the two $\Delta m = 0$ transitions, with Zeeman energy shifts $\pm h \Delta_B$. The coherently scattered photon wavepacket off the nuclear crystals can be written as \cite{49, 55, 56}

$$\psi(t) = \frac{\alpha}{\sqrt{\alpha^2 + 1}} J_1 \left( 2\sqrt{\alpha^2 + 1} \right) \cos(\Delta_B t) e^{-\frac{\alpha^2}{2} t}.$$  \hspace{1cm} (2)

Here, $J_1$ is the Bessel function of the first kind and of the first order \cite{43, 55, 57} caused by multiple scattering \cite{57} or dispersion \cite{55, 56}, $\alpha$ the effective resonant thickness \cite{57} and $\Gamma$ the spontaneous decay rate of the nuclear excited state. Furthermore, the trigonometric oscillation is caused by the quantum beat of the two split nuclear transitions \cite{56, 58}, and the exponential decay term describes the incoherent spontaneous decay of the excited states. The coherently re-emitted x-ray signal photon will then be reflected by an x-ray mirror with near 100% reflectivity \cite{15} and subsequently recombined on beam splitter BS 2. Finally, either detector B or C will register the time-delayed x-ray signal from the two output ports of the interferometer with probabilities that depend on the relative phase between the two paths.

To verify the entanglement between the two nuclear sample crystals, we invoke the method of quantum state tomography \cite{1, 6, 59} to determine the density matrix $\hat{\rho}$ of $|\text{TPE}\rangle$ of the coherently re-emitted single x-ray photon from two targets. In the photon-number basis, $\hat{\rho}$ reads \cite{6, 59}

$$\hat{\rho} = \frac{1}{P} \begin{pmatrix} p_{00} & 0 & 0 & 0 \\ 0 & p_{10} & d_{tpe} & 0 \\ 0 & d_{tpe} & p_{11} & 0 \\ 0 & 0 & 0 & p_{11} \end{pmatrix},$$  \hspace{1cm} (3)

where $p_{ij}$ is the probability of detecting $i$ photons from the left crystal and $j$ photons from the right one. Furthermore, $d_{tpe}$ is the coherence between the two components of $|\text{TPE}\rangle$ and $P = \text{Tr}(\hat{\rho})$. The concurrence $C = \max\{0, \frac{1}{2} \sqrt{\text{det}((\hat{\rho} - \hat{\rho}_{\text{min}}))}\}$ from a measured $\hat{\rho}$ then quantifies a lower bound for entanglement such that $C = 1$ for maximal entanglement and $C = 0$ for a pure quantum state \cite{6, 59, 60}. With the approximation $p_{00} \approx 1 - (p_{01} + p_{10} + p_{11})$, the diagonal terms can be determined experimentally by conditional measurements that distinguish between photons scattered by the $L$ or $R$ samples, e.g., by removing the second beam splitter BS 2. What concerns the coherence term $d_{tpe}$, it has been shown that this can be approximated as $V(p_{01} + p_{10})/2$ \cite{6, 59}, where $V$ is the visibility of the interference fringe at detectors B and C. The latter results from having the remitted single photon interfere with itself on beam splitter BS 2 for different phase shifts and can be experimentally determined. Typically, an additional Si phase shifter or a vibrating crystal are used to mechanically vary the phase between the two arms in an interferometer \cite{37, 47}. In what follows we demonstrate a magnetic, non-mechanical solution for phase modulation that directly and locally controls the nuclear dynamics in each ensemble and can provide an indication of controlling entanglement between the two remote parties.

The rotating orange arrows in Fig. 1(c-d) depict the time evolution of the nuclear transition current matrix elements as defined Ref. \cite{46}. The nuclear currents in the two crystals are simultaneously driven by the down-converted signal photon. Due to the Zeeman shifts $\pm h \Delta_B$, the two pairs of nuclear currents in the two samples evolve in directions determined by the sign of the corresponding energy shift and accumulate a phase $\phi(t) = \int_0^\tau \Delta_B(t) dt = \Delta_B \tau$ as a constant $\Delta_B$ is introduced. For a pair of currents in a certain crystal, a phase jump of $-2\phi$, associated with a time reversal effect \cite{61}, can be induced by inverting one of the applied magnetic fields at $\tau = T_\phi$ \cite{17, 18}. As only $B_R$ is inverted at $t = T_\phi$, the right mode turns into $\cos(\phi - \Delta_B t)$ that corresponds to $\cos(\phi + \Delta_B t + \Phi)$ with a phase jump $\Phi = -2\phi$, whereas the left wavepacket is still proportional to $\cos(\phi + \Delta_B t)$. The interference fringe can be analyzed as following \cite{62}

$$\left( \begin{array}{c} \tilde{a}_{\text{out}} \\ \tilde{b}_{\text{out}} \end{array} \right) = \left( \begin{array}{cc} 1 & 1 \ i & i \end{array} \right) \left( \begin{array}{cc} -1 & 0 \\ 0 & -1 \end{array} \right) \times \left( \begin{array}{c} \psi_R(t) \\ 0 \ \psi_L(t) \end{array} \right) \left( \begin{array}{cc} 1 & 1 \ i & i \end{array} \right) \left( \begin{array}{c} \tilde{a}_{\text{in}} \\ \tilde{b}_{\text{in}} \end{array} \right).$$  \hspace{1cm} (4)

Here, $\psi_R(t) = \frac{\psi(T_\phi + t)}{\cos[\Delta_B(T_\phi + t)]}$ and $\psi_L(t) = \psi(T_\phi + t)$. Matrices on the right hand side of Eq. 4 in turn correspond to the action of beam splitter BS 2, mirror, NFS in samples L and R and beam splitter BS 1 on the incident XPDC field. As the field $\tilde{b}_{\text{in}}$ is in the vacuum state, the intensities $Q_B = \int_0^\infty (\tilde{a}_{\text{out}}^\dagger \tilde{a}_{\text{out}}) dt \propto \sin^2 \phi$ and $Q_C = \int_0^\infty (\tilde{b}_{\text{out}}^\dagger \tilde{b}_{\text{out}}) dt \propto \cos^2 \phi$ at detector B and C, respectively, are plotted in Fig. 1(e) with $\alpha = 1$, $\Gamma = 1/141$ GHz for $^{57}$Fe and $\Delta_B = 30\Gamma$. Because of the collectively enhanced decay \cite{35}, the absorbed x-ray photon is not likely to be retained in the nuclear ensemble as excitation longer than...
the lifetime of single nuclear excited state. The coherence 
time of the entanglement between two crystals is approx. 60 
ns in Fig. [1]c). However, it has been shown that the speed-
up decay can be coherently turned off and on via a sequence 
of rotating [46] or switching off the hyperfine magnetic field 
[17]. While Ref. [17] remains so far only a theoretical pro-
posal, Ref. [46] reports the successful experimental demonstra-
tion of prolonging the nuclear exciton lifetime to $1/\Gamma$ by 
rotating an externally applied magnetic field of 10 G which 
in turn controls the internal hyperfine magnetic field inside a 
$^{57}$FeBO$_3$ crystal. Such a scheme could also be used to extend 
the coherence time of the presented entanglement setup by us-
ing $^{57}$FeBO$_3$ crystals as nuclear samples and a magnetic field 
rotation setup.

We now proceed with estimates on the possible production 
rate of heralded macroscopic entanglement. The key require-
ment here is that the XPDC source produces down-converted 
x-ray signal photons with energies within the width of the nu-
clear excited state, where the nuclear resonance absorption 
exceeds by orders of magnitude the atomic background pro-
cesses [49]. With a resonance cross section of $\sigma = 2.5$ Mbarn 
for the 14.4 keV transition of $^{57}$Fe, already a nuclear sample 
of 20 $\mu$m thickness is likely to absorb all incoming resonant 
photons. Assuming 100 % detection efficiency [19], the flux 
$R_E$ of produced signal photons within the nuclear linewidth 
equals the rate of heralded entanglement creation. The flux 
can be estimated as $R_E = \xi_s \Delta E_n / \Delta E_s$, where $\Delta E_n = 29.3$ 
neV is the linewidth of the considered $^{57}$Fe nuclear transition, 
and $\Delta E_s = 1$ eV and $\xi_s$ are the bandwidth and the 
flux, respectively, of the down-converted signal photons [22]. 
According to Ref. [22], $\xi_s \propto |\chi_{111}^{(2)}| I_p$, where $I_p$ is 
the photon density of the pump field, and $|\chi_{111}^{(2)}|$ the 111 Fourier 
coefficient of the second order nonlinear susceptibility for a 
diamond (111) crystal [64, 65]. By introducing $\omega_p = \omega_s + \omega_i$ 
[22] and the law of cosines [60], we obtain for the susceptibility 

$$ |\chi_{111}^{(2)}| \approx \frac{N e^3 F_{111}^r}{4 \varepsilon_0 m^2 \omega_s \omega_i^2 (2 - \omega_s^2 - \omega_i^2)} $$

(5)

where $\omega_p$, $\omega_s$, and $\omega_i$ are the angular frequencies of pump, sig-
nal and idler photons, respectively, $N$ is the number density of 
unit cells, $F_{111}^r$ the linear structure factor of bound elec-
trons [22, 64] and $\hat{Q}_{111}$ the 111 reciprocal lattice vector of the 
XPDC diamond crystal. Further parameters are $m$ the electron 
mass, $\varepsilon$ the electron charge, $\varepsilon$ the speed of light and $\varepsilon_0$ the 
vacuum permittivity. Given $\hbar \omega_s = 14.4$ keV and $\hbar \omega_i = 100$ 
eV, $|\chi_{111}^{(2)}| \approx 10^{-20}$ C/N $\sim 10^{-16}$ statcoulombs/dyne, hav-
ing the same order of magnitude as for the case of $\hbar \omega_s = 10.9$ 
keV reported in Ref. [22]. Since for the latter SR pulses were 
used as pump field, the pump photon density can be enhanced 
by considering an XFEL pulse. Fortunately, diamond crys-

tals are robust and do not experience lattice damage from ex-
posure to intense XFEL radiation [14]. Considering a train 
of XFEL pulses with $10^{12}$ photons/pulse and repetition rate 
f = $2.7 \times 10^4$ [67], on a spot size of 0.0005 mm$^2$, we ob-
tain $I_p = 5.5 \times 10^{18}$ photons/s/mm$^2$. By simple scaling we 
then obtain $\xi_s = 2.9 \times 10^9$ signal photons/s with a band-
width of 1 eV, resulting in a production rate $R_E$ of around 1 
Hz for the heralded creation of entanglement. We note that 
the signal photon rate is low enough to allow sufficient po-
tential recording time (several hundreds ns) between single 
shots. Further attention is required for avoiding losses by air 
absorption of the heralding EUV photon [23] and also for the 
mechanical alignment of the setup, with XPDC source, beam 
splitters and mirrors all having angular acceptances of $\mu$rad 
[13, 22, 36, 37].

Our scheme for heralded generation of quantum entangle-
ment between two macroscopical nuclear sample crystals re-
lies on Mössbauer nuclear transitions. In practice, quantum 
effects of the collective nuclear excitation have been shown 
to be preserved or even induced by vibrating nuclear crystals 
[19, 47, 68]. Nuclear resonant inelastic coherent scattering 
[69, 70], for instance, would allow the creation of entangle-
ment in the mechanical motion of a macroscopic system 
similar to the results reported in Ref. [71], but with increased 
coherence time and several orders of magnitude increase in 
the number of involved atoms. The additional feature required 
from our setup is the detection of phonons in the sample, cor-
responding with meV energy resolution for the signal photon. 
We expect that heralded entanglement using x-rays and 
nuclear transitions can thus open a new research avenue for 
both applied ideas related to quantum technology as well as 
more foundational studies of the boundary between the quantum 
and classical worlds.
