Quantum memories (QM) are essential components in quantum information processing. They enable storage and on-demand retrieval of quantum states and allow using fast but short-lived processing qubits, or photonic states that are excellent carriers of quantum information but are difficult to store. QM for light find applications in linear optics quantum computing, as well as in quantum communications and networks, where they could enable distribution of entangled states over long distances using quantum repeater architectures. There is also a growing interest in spin based quantum memories that store micro-wave photons which in turn can be interfaced to superconducting qubits. In the solid state, optical and microwave QM based on inhomogeneously broadened ensemble are actively investigated in rare earth (RE) ion doped crystals and diamonds containing NV centers. These two systems are well adapted to highly multimode storage, where multiple photons with large bandwidths are stored for long times. Moreover, high efficiency can be obtained by coupling these centers to a cavity, overcoming their weak interactions with photons, either for spin or optical transitions. A natural protocol to implement QM in inhomogeneous ensembles is the spin or photon echo, which recovers the initial excitation of the system by applying a π pulse to the storage transition. This inverts the atomic or spin phase evolution and results in a collective emission, the so-called echo. However, this scheme does not allow low-noise operation, a key parameter for quantum memories, which must store photonic qubits like single photons. This is because the collective emission occurs in an inverted medium which produces a too large spontaneous emission at the memory output. To avoid this situation, several protocols have been proposed and experimentally investigated. However, they require spectral tailoring, which requires a long lived storage level and can reduce bandwidth, or particular spatial phase matching conditions, that are difficult to combine with a cavity. Another possibility is to use fast frequency tunable cavities, that may be technologically challenging for micro-wave high Q cavities or in the optical domain.

We consider an ensemble of centers in a crystal with an inhomogeneously broadened optical or spin transition showing a linear Stark effect. The ensemble has an inversion symmetry, that can be intrinsic to the host or created by separating the sample in two parts for which the electric field is reversed. Because of the inversion symmetry, a given electric field will produce a posi-
tive frequency shift for half of the centers, and a negative one for the other half. The SEMM principle is shown in Fig. 1. We assume that the whole sequence takes place within a time much shorter than the centers’ occupation and coherence lifetimes (T1 and T2, respectively) to preserve a high storage fidelity. Initially, all centers are in the same state. At time t1, a single input photon is absorbed by the ensemble, and the wavepackets start to dephase relative to each other because of the inhomogeneous broadening. At time t2, an electric field E is applied to induce a phase shift 2πΔ = 2πEk = π/2 to half of the centers and therefore −π/2 to the other half, because of the ensemble’s inversion symmetry. k is the linear Stark coefficient of one of the subgroups related by the inversion symmetry. The wavepackets divide in two groups with opposite phase shifts, as shown on the Bloch sphere 2 of Fig. 1. At time t3, a π pulse is applied to the transition, and for t > t3, the ensemble polarization or magnetization P(t), summed over all centers, is then proportional to:

\[ P(t) \propto \int_{-\infty}^{+\infty} e^{i\omega \delta t} \cos(2\pi \Delta T_s) d\omega, \]  

where \( \omega \) is the frequency of the transition (centered at \( \omega = 0 \)), \( \delta t = 2t_3 - t_1 - t \) and \( T_s \) is the Stark pulse length. As in a 2-pulse echo experiment, the inhomogeneous broadening is rephased at \( t_4 = 2t_3 - t_1 \), but \( P(t_4) \) vanishes for \( 2\pi \Delta T_s = \pi/2 \) or \( E = 1/(4kT_s) \). There is therefore no collective emission (echo) at \( t_4 \). To recover the input photon from the memory, a second electric field pulse is applied at \( t_5 \), as well as a second π pulse at \( t_6 \). The polarization at \( t > t_6 \) is proportional to:

\[ P(t) \propto \left[ \int_{-\infty}^{+\infty} e^{i(2\pi \Delta T_s - \omega \delta t' - 2\pi \Delta T_s)} d\omega \right] + \int_{-\infty}^{+\infty} e^{i(-2\pi \Delta T_s - \omega \delta t' + 2\pi \Delta T_s)} d\omega, \]  

where \( \delta t' = 2t_6 - t_4 - t \). At \( t_7 = 2t_6 - t_4 \), the inhomogeneous broadening is again rephased, whereas the Stark phase shifts cancel, which gives \( P(t_7) = P(t_1) \). This collective emission or echo is the output of the memory and is identical to the initial input (Fig. 1). Thanks to the two π pulses, this emission occurs in a non-inverted medium, which avoids spontaneous emission at the time and in the mode of the memory output. This is required for the memory to operate in the quantum regime 10. Another fundamental source of noise is due to spontaneous emission at \( t_4 \), which would lead to a collective emission at \( t_7 \), because of the π pulse at \( t_6 \), with no relation with the memory input 12. This unwanted echo is however cancelled by the ±π/2 phase shift produced by the Stark pulse at \( t_5 \), in the same way as the echo at \( t_4 \) is suppressed by the Stark pulse at \( t_2 \) (see Eq. 1).

Until now, we assumed that the magnitude of the frequency shift induced by the electric field is the same for all centers. However, variations in each center environment will cause a distribution of the Stark coefficients. Moreover, the electric field will also be to some degree spatially inhomogeneous over the sample. This could limit the SEMM to a (small) sub-ensemble of centers. We examine this question below by considering a distribution of Stark coefficients. Electric field inhomogeneities can be treated in the same way. Assuming no correlation between the transition broadening Γ and the Stark distribution, the polarization after a square electric field pulse of duration \( T_s \) and amplitude \( E \) is:

\[ P = \int_{-\infty}^{+\infty} \cos(2\pi kET_s) g(k) dk \]  

where \( g \) is the normalized distribution of the Stark coefficients (\( \int g(k) dk = 1 \)) for one of the subgroups related by the inversion symmetry. We have therefore \( P = \Re(\mathcal{g}) \), where \( \mathcal{g} \) is the Fourier transform of \( g \). P = 0 will occur for Stark pulse amplitude and duration satisfying:

\[ \Re(\mathcal{g}(ET_s)) = 0. \]  

In the case, of a symmetric distribution centered on \( k_0 \), \( g(k) = g_1(k - k_0) \), where \( g_1(x) = g_1(-x) \). P is given by:

\[ \Re(\mathcal{g}(ET_s)) = \cos(2\pi ET_s k_0) g_1(ET_s) \]  

![FIG. 1. The SEMM sequence. (a) microwave or optical fields. The memory input is at \( t_1 \), π pulses at \( t_3 \) and \( t_6 \), and the output at \( t_7 \). (b) electric field. The Stark pulses at \( t_2 \) and \( t_5 \) produce a phase shift that cancels the collective emission at \( t_4 \). (c) Bloch sphere representations of the wavepackets evolution at the points labeled in (a) and (b). For clarity, the input pulse has an π/2 area.](image-url)
and cancels for \( k_0ET_\pi = 1/4 \), i.e. a central phase shift of \( \pi/2 \), independently of the width of the Stark distribution \( g(k) \). As shown in the supplemental material, condition 4 can be satisfied for any Stark coefficient distribution, unless a large fraction of centers have a zero Stark shift. After cancellation of the intermediate echo at \( t_4 \), the second Stark pulse at \( t_5 \) is identical to the first one at \( t_2 \), but induces an opposite phase shift in each wave packet. This results in a complete recovery of the initial input for any distribution \( g(k) \). This would not be the case if an additional \( \pi/2 \) phase shift was applied, leading to an overall \( \pi \) shift, even in the case of a symmetric Stark distribution (see supplemental material). In SEMM, the memory bandwidth is therefore only limited by the \( \pi \) pulses fidelity over the ensemble of centers. This is in sharp contrast with protocols based on transition broadening by electric fields [17,20], in which the bandwidth is directly dependent on the magnitude of the Stark shifts that can be induced. SEMM has no such limitations, and in the microwave or rf ranges, where \( \pi \) pulses of high fidelity and bandwidth can be readily obtained, the entire ensemble inhomogeneous linewidth can be used, as shown in the following.

As a proof of concept, we investigated our protocol in a rare earth doped crystal, Eu\(^{3+}\):Y\(_2\)SiO\(_5\)(Eu:YSO), in which Eu\(^{3+}\) ions sit in a \( C_1 \) symmetry site and the crystal symmetry (\( C_{2h} \)) includes an inversion operation. In this material, we recently observed a linear Stark effect on the ground state hyperfine transitions of the Eu\(^{3+}\) isotope, which has a nuclear spin \( I = 5/2 \) [25]. In the present work, rf excitations were stored and retrieved using the ground state \( \pm 1/2 \leftrightarrow \pm 3/2 \) transition at 34.58 MHz [\( k = 0.43 \text{ Hz}/(\text{V/cm}) \)], using a 0.1% \(^{151} \text{Eu} \) doped sample inserted into a coil [Fig. 2 (a)]. Spin echoes were optically detected by Raman heterodyne scattering [20] using a laser resonant with the Eu\(^{3+}\) \( ^7F_0 \leftrightarrow ^5D_0 \) transition at 580 nm. Electric fields parallel to the \( D1 \) crystal optic axis were applied across the 1mm thick sample on which two brass electrodes were placed. All experiments were carried out at 3.5 K. A small static magnetic field of about 48 G was applied in the \( D1 \) direction to increase the spin coherence lifetime to 25 ms. Other experimental details can be found in Refs. [20] and [27].

The sequence we used is shown in Fig. 2 (b). We first investigated suppression of echo 1 after the first rf \( \pi \) pulse by applying a Stark pulse of varying length [Fig. 2 (c)]. The experimental data, normalized by the echo intensity at zero field, could be well fitted by the equation \( I = (\cos(2\pi\Delta T_\pi))^2 \). The minimum echo intensity corresponds to a suppression \( \mu = 1.5 \times 10^{-5} \). This was obtained in a sample with no accurate polishing or parallelism, which is likely to produce inhomogeneous Stark shifts. The observed very low residual echo intensity therefore confirms the above analysis. The lowest achievable echo suppression is limited by parameters fluctuating in time. In our setup, we estimate that the dominating ones were voltage noise, as well as slow fluctuations in temperature and laser intensity and frequency, as signals were averaged over 200 shots. Echo suppression is particularly important in decreasing the collective emission at the memory output time caused by rephased spontaneous emission (see above). This spontaneous emission can be large when a cavity is used. For example, in a microwave resonator, the Purcell effect and the gain due to the inverted medium result in a number of spontaneous photons equals to \( n_{sp} = F(e^{Fd} - 1) \), where \( F \) is the cavity finesse and \( d \) the memory opacity [12]. Our experimental value of \( \mu \) would allow operation at the single photon level for a cavity with \( F \approx 100 \) (see suppl. material). Such a resonator would be suitable for an impedance matched memory [12] or a strongly coupled one, which has to switch between high and medium finesse to avoid super-radiance during the microwave pulses [13].

The complete SEMM sequence was then studied by adding the second Stark and \( \pi \) pulses to retrieve the memory output [echo 2 in Fig. 2 (b)]. To optimize the signal to noise ratio, the input of the memory was a \( \pi/2 \) pulse. The signals recorded at zero electric field are shown in Fig. 2 (d), upper trace. Besides the intermediate and final echoes, we also observed a stimulated echo after the second \( \pi \) pulse. The stimulated echo was separated from the memory output by choosing \( t_2 - t_1 < (t_4 - t_2)/2 \). When the Stark pulses were applied, the intermediate echo was strongly suppressed [Fig. 2 (d)]. The stimulated echo was suppressed too, since it results from a population grating that forms from the pulses at \( t_1 \) and \( t_2 \). The second Stark pulse does not induce any additional phase shift on populations and the stimulated echo is suppressed by the first Stark pulse. The memory output, echo 2, is retrieved with an intensity essentially identical to what is observed when no electric field is applied (see below). The bandwidth of the memory is about 40 kHz limited by the length (24 \( \mu \)s) of the \( \pi \) pulses. This matches well the 32 kHz inhomogeneous width of the \( \pm 1/2 \leftrightarrow -3/2 \) transition at 34.58 MHz [25].

The length of the memory output pulse was 24 \( \mu \)s with or without the Stark pulses, showing that SEMM preserves the full bandwidth, as expected. The frequency shifts due to the Stark field were however only \( \pm 58 \) Hz, corresponding to \( \approx 15 \text{ V} \) applied across 1 mm.

We also performed quantum state tomography to study the influence of the Stark pulses [28]. Input states \( \pm X, \pm Y \) were created by varying the phase of the \( \pi/2 \) pulse, whereas \( +Z \) corresponded to no input pulse. The \( \sigma_X \) and \( \sigma_Y \) components of the output density matrix were determined by analyzing the real and imaginary parts of the output pulse. The \( \sigma_Z \) component was measured by an additional echo sequence following the output pulse. The upper row of Fig. 3 shows the output density matrices for the \( +X, -Y \) and \( +Z \) input states for the SEMM sequence without the Stark pulses. Although the sequence should operate as the identity operation, devi-
FIG. 2. Measurements on $^{151}$Eu$^{3+}$:Y$_2$SiO$_5$ nuclear spins. (a) scheme of the sample with attached electrodes to create electric fields. The coil is used to produce rf pulses and the laser to detect spin coherences. (b) Experimental SEMM scheme. Delays were $t_2 - t_1 = 4.5$ ms, $t_4 - t_2 = 11.5$ ms. (c) Normalized echo 1 intensity as a function of the length of a Stark pulse of 16.5 V amplitude. Squares: experimental data; solid line: fit, see text. (d) Echoes observed with or without electric field in the SEMM sequence.

Table I. Site symmetry, coherence lifetime, Stark coefficient and field for SEMM (assuming $T_2 = T_s$) for centers in various hosts with global inversion symmetry.

| System         | Site sym. | $T_2$ (ms) | $k$ (Hz cm/V) | $E_0$ (V/cm) |
|----------------|-----------|------------|---------------|--------------|
| Optical trans. | Eu$^{3+}$:Y$_2$SiO$_5$ | $C_1$ | 2.6 | 27000 | 0.005 |
| Electron spin  | Eu$^{3+}$:CaWO$_4$ | $S_4$ | 0.05 | 399 | 12 |
| Nuclear spin   | 151Eu$^{3+}$:Y$_2$SiO$_5$ | $C_1$ | 26 | 0.1 | 9.6 |

In conclusion, we introduced a memory protocol for ensembles of atoms or spins that involves two rephasing pulses to avoid producing an output in an inverted medium. The intermediate collective emission, as well as the rephased spontaneous emission, are cancelled by a Stark induced linear phase shift of centers related by an inversion symmetry. The protocol is thus low-noise and suitable for a quantum memory. Moreover, large storage bandwidths are possible since the cancellation process is insensitive to inhomogeneities in Stark coefficients or the electric field. The protocol has been investigated in RE nuclear spins in a single crystal, where we found a strong echo suppression of $1.5 \times 10^{-5}$. Opposite Stark
phase shifts are produced to recover the memory output, which ensures a high fidelity, experimentally confirmed by quantum state tomography. SEMM could be used to store optical or microwave photons with high efficiency in atoms and/or spin transitions coupled to cavities. This includes NV centers in diamond and rare earth doped crystals, which are currently among the most promising solid-state quantum memories.

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† philippe.goldner@chimie-paristech.fr

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