Dynamic control of Purcell enhanced emission of erbium ions in nanoparticles

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The interaction of single quantum emitters with an optical cavity enables the realization of efficient spin-photon interfaces, an essential resource for quantum networks. The dynamical control of the spontaneous emission rate of quantum emitters in cavities has important implications in quantum technologies, e.g., for shaping the emitted photons’ waveform or for driving coherently the optical transition while preventing photon emission. Here we demonstrate the dynamical control of the Purcell enhanced emission of a small ensemble of erbium ions doped into a nanoparticle. By embedding the nanoparticles into a fully tunable high finesse fiber based optical microcavity, we demonstrate a median Purcell factor of 15 for the ensemble of ions. We also show that we can dynamically control the Purcell enhanced emission by tuning the cavity on and out of resonance, by controlling its length with sub-nanometer precision on a time scale more than two orders of magnitude faster than the natural lifetime of the erbium ions. This capability opens prospects for the realization of efficient nanoscale quantum interfaces between solid-state spins and single telecom photons with controllable waveform, for non-destructive detection of photonic qubits, and for the realization of quantum gates between rare-earth ion qubits coupled to an optical cavity.
Quantum network nodes should be able to store quantum information for long durations, to process it using local quantum gates between qubits, and to exchange this information with distant nodes, ideally using photons at telecommunication wavelengths, via an efficient spin–photon interface. Several platforms are currently investigated for the realization of quantum nodes, including atoms, trapped ions, and solid-state systems. Single atoms or solid-state emitters provide a platform for spin–photon interfaces with quantum processing capabilities. The realization of an efficient spin–photon interface is facilitated by the use of an optical cavity in the Purcell regime, which allows channeling the emission from the emitter in the cavity mode while decreasing the spontaneous emission lifetime. In the presence of dephasing, it also increases the indistinguishability of photons from the emitter. However, a strong reduction of the optical lifetime also reduces the available time to realize quantum gates that rely on dipole-blockade mechanisms achieved by driving the emitter to the excited state.

One attractive solution to this problem is to decouple the emitters from the cavity when performing the quantum gates, and coupling it back to emit a single photon at a desired time. The ability to achieve a dynamic modulation of the Purcell factor would therefore be a key ingredient to achieve this type of quantum gates in a high-efficiency spin–photon interface, as well as an essential tool to shape the emitted single-photon waveform and to realize cavity-enhanced non-destructive detection of photonic qubits. In addition, the dynamic resonance modulation also enables addressing inhomogeneously broadened ensembles of single emitters as multi-qubit registers.

The control of collective light emission from ensembles of atoms in free space has been demonstrated. Experiments toward the dynamic control of emission of the cavity-enhanced spontaneous emission rate have been so far mostly performed with semi-conductor quantum dots featuring short optical and spin coherence time, with Raman schemes with single atoms and ions, and by modifying the local optical environment of rare-earth ion-doped (REI) crystals. Related work on fast modulation of erbium ions emission has been performed recently, using electrical modulation of erbium–graphene interaction. Among solid-state materials, REI crystals constitute a promising platform for quantum information processing and networking. REI feature exceptional spin coherence time and store information and narrow optical transitions as spin–photon interface, including at telecom wavelength for erbium ions. They have been used extensively as ensemble-based quantum memories. REI possess permanent dipole moments with different values in the ground and excited states, which enables strong dipolar interactions between nearby ions, opening the door to the realization of quantum gates between two or more matter qubits, using a dipole-blockade mechanism achieved by exciting one ion, e.g., between erbium and another ion species in the context of quantum repeaters.

Recently, rare-earth ions have proven to preserve their coherence properties in nanoparticles. This facilitates their incorporation into microcavities to reach strong Purcell enhancement, as necessary for the emission of coherent indistinguishable single photons. Also, it provides the necessary confinement required to isolate close-by single ion qubits (=10-nm distance), as required for dipolar quantum gates. Following the first demonstration in free space, REIs coupled to phononmonic cavities have also recently led to the detection and manipulation of single rare-earth ions.

In this work, we demonstrate the dynamical control of the Purcell enhanced emission of a small ensemble of erbium ions in a single nanoparticle. This is achieved by inserting the nanoparticles into a fully tunable high finesse fiber microcavity at cryogenic temperature, whose frequency can be tuned at rates more than 100 times faster than the spontaneous emission lifetime of 10.8(3) ms of the ions by physically moving the fiber mirror with sub-nanometer precision using a piezoelectric device. By measuring the decay of fluorescence counts in the cavity, we infer an average effective Purcell enhancement of 14(1). The decay is compatible with a multiplexponential model describing Purcell enhancement with an ensemble of ions that predicts that 10% of the ions experience an effective Purcell enhancement higher than 74. The Purcell enhancement can be controlled on a time scale of hundred microseconds. In addition, we show that we can shape in real time the Purcell enhanced emission of the ions to control the emitted photons’ waveforms, without perturbing the emitter. Our approach opens the door to a solid-state quantum node with the potential of exhibiting quantum computing and communication capabilities all in a single device.

Results

Description of the experiment. When coupling an emitter to a microcavity the emitter’s lifetime is reduced to \( \tau_c = \tau_r/(C + 1) \), where \( \tau_r \) is the natural lifetime and \( C = L \xi C_0 = L \xi C_0 + Q/\lambda^3 \) is the effective Purcell factor, with \( \lambda \) the emission wavelength, \( Q \) the quality factor of the resonator and \( V_m \) its mode volume, \( \xi \) the branching ratio of the respective transition and \( \Delta(\delta) = (\Delta/2\pi)^2 + (\delta/2\pi)^2 \) is the detuning factor in case the cavity with linewidth \( \Delta \) is detuned by \( \delta \) from the emitter’s resonance. In addition, the collection efficiency of the cavity mode is given by \( \eta = gC/(C + 1) \) where \( \eta \) is the cavity outcoupling efficiency. Thus, fast and near-unity efficiency readout can be achieved for sufficiently large \( C \) and \( \eta \).

Fiber cavities can achieve high Purcell factors up to 10^4, provide open access to the cavity mode for optimal overlap between the ions and the cavity field, and are small and lightweight enough to offer fast frequency tuning. Nanoparticles are a promising platform to study REI as their small size isolates a mesoscopic ion number, such that the large ratio between the inhomogeneous and homogeneous linewidth can allow one to frequency select single ions. Also, their scattering cross section can be small enough such that integration into high-finesse cavities is possible. In particular, we study Er^3+:Y_2O_3 nanoparticles with 200 ppm erbium concentration and an average radius of 75 nm. Y_2O_3 is a host that showed to maintain good crystalline quality in the nanoscale allowing long coherence time for dopants like europium and praseodymium. For Er^3+:Y_2O_3, the optical transition 4f_{15/2}→4f_{11/2} is at 1535 nm with a relatively large branching ratio \( \xi = 0.21 \).

Our setup is based on a fast and fully tunable, stable, cryogenic-compatible Fabry–Perot microcavity (see Fig. 1a). The microcavity is composed of a fiber with a concave structure on the tip, on which a reflective coating is deposited. The other side of the cavity is a planar mirror with the same reflective coating as the fiber, over which the doped nanoparticles are placed after depositing a thin layer of SiO_2 to ensure maximum coupling between the ions and the cavity electric field (see section 1 of Supplementary Information (SI)). The setup allows to move the fiber around the mirror and localize the nanoparticles, and to set the separation between the fiber and the mirror to form a cavity on resonance with the ions. Nanoparticles are located by scattering loss microscopy. We use a nanoparticle with a radius of 91(1) nm (see Fig. 1b) adding a total intra-cavity loss of 2×43(2) parts-per-million (ppm) per round trip and containing close to 11,000 Er^3+ ions in the measured C_2 crystallographic site. The empty cavity has a finesse of 2×10^5, which is reduced to 1.6×10^4 in presence of the nanoparticle. For cavities as short as 3.5 μm we can expect a maximal Purcell factor \( C_{\text{max}} = 320 \), which
Measurement of the inhomogeneous line of the $^4I_{15/2} - ^4I_{13/2}$ transition. We then estimate the probability for an ion to emit in the cavity mode $P = C/C^{\text{max}} = (14(1))/14(1) = 93.2\%$, and the cavity-enhanced branching ratio $\zeta = (C_0 + 1)/(C_0 + 1) = 95.5\%$.

The single exponential fit does not describe well the data for the Purcell enhanced case suggesting that emitters experiencing different enhancement contribute to the detected signal (see dashed red line in Fig. 2a–c). To investigate this further, we derive a model describing our experiment (see section 6 of SI). The model considers a maximum Purcell factor $C^{\text{max}}$ which is then reduced due to the randomly oriented dipole moment of the emitters, the finite extension of the particle, and fluctuations of the cavity resonance. From the model we can perform a dynamic and a static analysis. The dynamic approach is based on

**Fig. 1 Description of the experiment.** a Schematic drawing of the tunable fiber-based microcavity. The fiber mirror can be moved in 3D, allowing us to set the length of the cavity and locate a nanoparticle inside the cavity. b Map of scattering loss introduced by a single nanoparticle when scanning the cavity mode across it. c Measurement of the inhomogeneous line of the $^4I_{15/2} - ^4I_{13/2}$ transition. The solid line is a Gaussian fit, yielding a full-width-half-maximum linewidth of 15(2) GHz (error bars represent one standard deviation). d The optical setup is described in more detail in the SI. We use a second laser at 790 nm to actively stabilize the cavity (see section 3 of SI). We calculate the effective Purcell factor $C = C_0 / C^{\text{max}} = 31(1)/93.2\% = 95.5\%$.
the probability of an ion to emit into the cavity mode which is proportional to $e^{-\int_{t_0}^{t} C(t') \, dt'}$ where $C(t')$ is the experienced Purcell factor as function of time. The static approach starts by estimating the distribution of Purcell factors in the system $p(s = C/C_{\text{max}})$, from which we reconstruct a decaying curve proportional to $\int_0^s p(s) e^{-s C_{\text{max}}/\tau_0} \, ds$. While the first method is a direct simulation of the experiment, which is sensitive to the frequency of the cavity vibration, from the second method we can obtain a distribution of Purcell factors and estimate the maximum enhancement present in the system (see section 6 of SI).

The magenta and blue lines in Fig. 2a–c are the models following the dynamic and static analysis, respectively, when considering expected parameters from our system but reducing $C_{\text{max}}$ to 150 in order to obtain a satisfying description of the experimental data. Figure 2d is the probability $P_{c>C}$ that a given ion experiences a Purcell factor $c$ greater than $C$ calculated from the estimated $p(s)$. From this model, we infer that 50% of the ions experience a Purcell factor larger than $C = 15$ consistent with the measured $\tau_c = 0.74(1)$ ms, and that at least 10% of the ions experience a Purcell factor larger than 74. The models describe the data with noticeably more accuracy than the single exponential (see RMSE calculations in Fig. 2b). In particular, they succeed to reproduce the decay at early times, confirming the presence of Purcell enhancement larger than 15.

Black points in Fig. 2a corresponds to photons generated by a few hundred ions due to power broadening, and by decreasing the input power to 300 pW, a number as low as 10 ions could be detected with a signal-to-noise ratio around 5 (see section 7 of SI). High-fidelity single ion detection could be achieved by using more efficient superconducting nanowire single-photon detectors.

Fig. 2 Purcell enhanced lifetime measurements. a Black points and green crosses correspond to the natural and Purcell enhanced lifetime measurements. Dash-dotted red line: Single exponential fit. Solid magenta and dashed blue line are the dynamic and static model, respectively. b Gray lines correspond to one and two standard deviation calculated by summing all the counts in the detection window $[t_0, t]$ of the Purcell enhanced lifetime measurement (black points), where $t_0$ is the time at which the photon counting is started, and considering Poissonian counting statistics. Root-mean-square error (RMSE) of the single exponential, the static, and the dynamic models, also calculated as function of the detection time window. The single exponential fit deviates by more than two standard deviations at earlier times, while the model and the reconstructed decaying curve stay below this threshold. c Zoom of the first 150 $\mu$s of the plot shown in (a). Error bars represent one standard deviation calculated from Poisson counting statistics. d Estimated probability $P_{c>C}$ that a given ion decays with a Purcell factor $c > C$.

Dynamical control of Purcell enhanced emission. We now explain our strategy to tune the cavity resonance in a time scale faster than the spontaneous lifetime. First, we set the 1535 nm excitation laser on resonance with the center of the inhomogeneous line of the ions. Then, we use a second laser at 790 nm to stabilize the length of the cavity. The 790-nm wavelength is tuned such that 50% transmission level of the positive slope overlaps with the center of the 1535 nm cavity resonance (see Fig. 3a). By a fast change of the voltage offset $V$ from $V_p$ to $V_n$ or vice-versa, and by switching the sign of the feedback action, we can stabilize the cavity to the positive and negative sides of the 790-nm fringe at will (the feedback is off during the transient phase). The voltage is varied as $\sin^2 \left( \frac{\pi V}{\Delta V_{\text{fwhm}}} \right)$ and the process happens during a time $S_{790} = 300 \mu$s for our realization. Between the two locking positions, the total fiber displacement $d_f$ corresponds to the full-width at half-maximum of the 790 nm cavity fringe, that is, $d_f = \Delta_{790} = 790 \text{ nm} \approx 600 \text{ pm}$ ($F_{790} \approx 700$, reduced due to the particle losses).
This displacement is 12 times larger than $\Delta_{1535} \approx 50$ pm, such that the detuning between the erbium ions and the 1535-nm cavity is $\delta = 12\Delta_{1535}$. Using the Lorentzian lineshape of the cavity, one can estimate a maximum reduction of Purcell factor $C \approx \mathcal{L}(\delta) \approx 1/577$ and for the fluorescence count rate of $1/630^{47}$ (see section 4 of SI).

Next, we show the implementation of this technique and how it can be used to extract the natural lifetime $\tau_n$ of a single nanoparticle while isolating the effect of the cavity from any other lifetime reduction process. We first set the cavity on resonance with the ions ($V = V_p$) and turn on the excitation laser at 1535 nm for $t_d = 300$ μs (Fig. 3b). Immediately after, we detune the cavity ($V = V_s$) and start to collect photons. After a time $t_d$, we set the cavity back on resonance ($V = V_p$). Figure 3c shows the counts as function of time for $t_d = 5, 16, \text{ and } 28$ ms. As shown in Fig. 3c, the total counts decrease while increasing $t_d$. When the cavity is off-resonance with ions, the excited state population decays with the natural lifetime $\tau_n$, and therefore less ions contribute to the signal compared to $t_d$. In order to extract the natural lifetime, we repeat the measurements for $t_d$ in a range from 1 to 28 ms and calculate the decay rate of the excited state $S_{1535}$ at $t_d$ + 5 ms time window (crosses in Fig. 2). We observe a single exponential decay of $S_{1535}$ for $t_d = 5, 16, \text{ and } 28$ ms.

Finally, we demonstrate that this technique can be operated with a bandwidth high enough to shape the spontaneous emission of the erbium ions. Figure 3d shows the recorded counts as function of the detection time while the cavity is tuned on/off resonance for 1 ms (2.5 ms) times. Figure 3e,f shows a zoom-in of the falling and rising edges. The solid lines are a model using the piezo displacement, $S_{790}$, and an effective 1535-nm cavity linewidth to account for cavity instability. Defining the switching time $S_{1535}$ as the time needed to decrease the count rate by a factor of 10, we extract from the model $S_{1535} = 85(15)$ μs (see section 4 of SI). This value is more than two orders of magnitude shorter than the natural population decay time, and a factor of 9 shorter than the Purcell enhanced decay time $\tau_c$ of the ionic ensemble. For future experiments with single erbium ions (or with other ions with shorter lifetime), much higher Purcell factors and consequently much shorter $\tau_c$ will be required. Therefore, much shorter switching times are desirable. Several improvements in our experiment are possible to decrease $S_{1535}$. First, the current measured value is slower than the expected time of 67 μs, which can be explained by the limited cavity stability (see section 4 of SI). Improvements on the cavity stability will therefore directly impact $S_{1535}$. Another possibility is to increase the finesse of the cavity at 1535 nm (and therefore decrease $\Delta_{1535}$) as also required for increasing the Purcell factor. Finally, the switching time $S_{790}$ could be decreased by designing a system with higher mechanical eigen frequencies or by iterative learning algorithms to minimize added noise. Altogether, we estimate that values of $S_{1535}$ of a few microseconds could be achievable by combining these improvements (see section 4 of SI).

**Discussion**

In conclusion, we have demonstrated the dynamical control of the Purcell enhanced emission of a mesoscopic ensemble of
erbium ions confined in a nanoparticle embedded in an open fiber-based microcavity. By controlling the position of the fiber mirror, we have shown that we can control the cavity resonance and therefore the Purcell factor at a rate more than 100 times faster than the natural decay rate of the ions with the potential of reaching microseconds switching times. By varying the speed at which the fiber is displaced during the switching process, our setup would allow arbitrary shaping of the temporal waveform of the emitted photons. Combined with single-ion addressing, this ability will enable the generation of fully tunable narrowband single photons, and provides a route to quantum processing using single rare-earth ions.

While this paper was under review, we became aware of a related work showing Purcell enhanced emission of erbium dopants on a 19 μm thin film coupled to an optical cavity.50

Data availability
The data that support the findings of this study are available at Zenodo with the identifier https://doi.org/10.5281/zenodo.3736425.

Received: 23 March 2020; Accepted: 28 April 2021; Published online: 11 June 2021

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Acknowledgements
This project received funding from the European Union Horizon 2020 research and innovation program under Grant Agreement No. 712721 (NanoOQTech) and within the Flagship on Quantum Technologies with Grant Agreement No. 820391 (SQUARE). ICFO acknowledges financial support from the Government of Spain (PID2019-10685SRB-100; Severo Ochoa CEX2019-000910-S), Fundació Cellex, Fundación Mir-Puig, and Generalitat de Catalunya (CERCA, AGAUR), from the Gordon and Betty Moore Foundation through Grant No. GBMF7476 to H.d.R., from the European Union’s Horizon 2020 research and innovation program under the Marie Skłodowska-Curie grant agreement No. 665884. B.C. acknowledges the financial support of The Spanish Ministry of Science, Innovation and Universities (MCIU) and State Research Agency (AEI) through grant ICI-2017-33514. D.H. acknowledges funding by the BMFB in project Q.Link.X.
Author contributions
B.C. and C.D. designed and performed experiments, and analyzed data. S.L., D.S., A.F., and P.G. designed and performed the nanoparticles fabrication. T.H. and D.H. provided the fiber cavity and made the original design for the nanopositioner in cryogenic environment. B.C. and C.D. upgraded the design and built the nanopositioners. B.C. modeled the data, with input from C.D., D.H., and H.d.R. B.C., C.D., and H.d.R. wrote the paper, with contributions from all coauthors. H.d.R., D.H., and P.G. conceived the experiment and supervised the project.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-021-23632-9.

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Peer review information Nature Communications thanks the anonymous reviewer(s) for their contribution to the peer review of this work.

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