All-optical preparation of coherent dark states of a single rare earth ion spin in a crystal

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All-optical addressing and coherent control of single solid-state based quantum bits is a key tool for fast and precise control of ground state spin qubits. So far, all-optical addressing of qubits was demonstrated only in very few systems, such as color centers and quantum dots. Here, we perform high-resolution spectroscopic of native and implanted single rare earth ions in a solid, namely a cerium ion in yttrium aluminum garnet (YAG). We find narrow and spectrally stable optical transitions between the spin sublevels of the ground and excited optical states. Utilizing those transitions we demonstrate the generation of a coherent dark state in electron spin sublevels of a single Ce3+ ion in YAG, by coherent population trapping.

Coherent population trapping (CPT) [1–4] is an all-optical way of coherent manipulation of electron and nuclear spin qubits. CPT and related physical phenomena (slow light, electromagnetically induced transparency, etc.) were initially applied to quantum ensembles. In recent years, however, it has been demonstrated on single fluorescent centers in solids [5–9] resulting in all-optical control of single qubits [10, 11], which is a significant step towards fast and high fidelity control of single qubit spins [12, 13].

Rare-earth ions residing in inorganic crystal have been widely studied and applied in fields ranging from solid-state spectroscopy and laser physics [14] to quantum information processing [15], due to their narrow optical transitions [16, 17] and long spin coherence time [18, 19]. In particular, rare-earth ions in solids are promising systems for quantum information storage and processing [20, 21]. Achievements comprise six hours coherence time of nuclear spins in Eu:Y2SiO5 crystal [22], coherent storage of single photon states in Nd:YVO4 [23] and entangled photon pairs in Nd:Y2SiO5 crystals [24]. In addition to progress made with ensembles of rare-earth ions, the detection of individual ions have been demonstrated in three rare-earth species recently [25–29]. High fidelity spin control of single Ce3+ ion has been demonstrated by applying a resonant microwave field [30]. With this microwave control, the electron spin coherence time was extended from 150 ns to 2 ms by using the dynamical decoupling technique. Extending these finding to all-optical spin control would enable much faster control and make best use of the photon-spin coupling of rare earth ions.

Here, we report on CPT in a "dark" coherent superposition of the electron spin sublevels of a single Ce3+ ion in YAG based on resonant optical excitation. CPT results in lower fluorescence yield of a Ce3+ center when it is excited by two laser fields in two-photon resonance with the ground-state spin transition. Spectroscopic properties of resonant optical transitions of single native ions as well as single implanted ions have been studied. These studies reveal an optical linewidth of $∼2π\times80$ MHz and small spectral diffusion compared to color centers in diamond [31, 32] and quantum dots [33].

Figure 1(a) shows the unit cell of a YAG (Y3Al5O12) crystal. Trivalent cerium ions substitute trivalent yttrium ions and form color centers. Cerium can be found in yttrium-containing crystals as residual impurities. Al-
ternatively, individual Ce can be introduced into the crytal artificially by doping during the crystal growth or by ion implantation. The energy levels of Ce$^{3+}$ in YAG are shown in Fig. 1(b) [26, 34]. Ce$^{3+}$ has only one unpaired electron, and its ground states are located in the 4$f^1$ shell. Electrons in the 4$f$ shell are efficiently screened by closed outer lying 5$s$ and 5$p$ shells. This screening is responsible for the weak interaction between ions and their surrounding environment. The ground state is split into two sublevels $^2F_{5/2}$ and $^2F_{7/2}$ due to spin-orbit coupling. These two sublevels are further split by the crystal field interaction into three and four Kramers doublets, respectively. If an external magnetic field is applied, the degeneracy of these seven Kramers doublets is lifted. The excited state is located in the 5$d$ shell. It splits into five Kramers doublets due to the combined action of the spin-orbit coupling and the crystal field. The energy difference between the two lowest 5$d$ Kramers doublets is approximately 8,000 cm$^{-1}$ [26, 34] and, therefore, the excited states can be optically addressed individually. In addition, the quantum efficiency of the 5$d$ → 4$f$ transitions is close to 100% [35]. The lifetime of the lowest 5$d$ state is 60 ns [26, 36].

In the experiment, a [1 1 0] orientated ultrapure YAG crystal (Scientific Materials) is used. An external magnetic field ($B \approx 450$ G) is applied perpendicular to the laser beam direction, so that four optical transitions between the two pairs of spin states of the 4$f$ and 5$d$ levels are allowed, as shown in Fig. 1(c) [26]. A Λ scheme, which is a requirement for CPT, can be formed by optically mixing both ground states to either of the excited spin states. Single Ce$^{3+}$ ions are detected under a home-built high resolution confocal microscope at cryogenic temperature ($T \approx 3.5$ K). A solid immersion lens (SIL) is milled by focused ion beam on the surface of the YAG crystal to improve the spatial resolution and the fluorescence collection efficiency of the confocal microscope [37] (see Fig. 1(c)). The laser scanning microscopy image shown in Fig. 1(d) is obtained through non-resonant pulsed excitation (440 nm) with a frequency-doubled femtosecond Ti:Sapphire laser. Each bright spot corresponds to an individual native Ce$^{3+}$ ion. The emitted photons associated with broad phonon sideband are detected by an avalanche photodiode (APD) in a spectral range between 500-625 nm (see our previous work [30]). A tunable single-mode narrow linewidth (~500 kHz) continuous wave (CW) laser (wavelength of 489.15 nm, Toptica Photonics DL pro) is used to resonantly excite single Ce ions.

A portion of the emission spectrum of a single Ce$^{3+}$ ion in the vicinity of its zero-phonon line (ZPL), measured by a high resolution spectrometer, is shown in Fig. 2(a). A sharp zero-phonon line (ZPL) is located at 489.15 nm and accompanied by a red-shifted phonon sideband partly shown in the figure [38].

A CW laser is swept across the ZPL position to obtain the excitation spectrum of the single Ce ion shown in Fig. 2(b). Four individual optical excitation-transition lines are well resolved. These lines correspond to four different optical transitions between the lowest Kramers doublets of the ground state and of the excited state with the assignment indicated in Fig. 2(b). The full width at half maximum (FWHM) of the optical transitions is $\sim 2\pi \times 80$ MHz, which is broader than the lifetime limited linewidth $2\pi \times 4$ MHz. This broadening is caused mainly by the strong $^{27}$Al nuclear spin bath, i.e. it represents an intrinsic property of the host material [30].

By monitoring the fluorescence during each successive frequency sweep through the resonant transitions, we observe stable optical resonance lines without obvious spec-
sequences. The pulsed laser is used to bring the Ce
pulsed and CW laser excitation. (b) Scheme of the laser pulse
FIG. 3. (a) Fluorescence intensity of a single Ce ion under
Ce praseodymium ions in solids [28, 29, 39].
trol of single Ce ion spins possible comparable to single
tions and spectral stability makes precision optical con-
ble spectrum. The combination of narrow optical transi-
3+ of the implanted Ce ions is
Supplemental Material). The optical transition linewidth
contains a broad peak with a dip going down nearly to the
Ce:YAG is ionized by CW laser excitation when the
wavelength is 448 nm. The repetition rate is 2.5 MHz and
the average laser power is 10 µW/cm². A blue diode laser
(451 nm) with 150 µW power is used as the CW laser.
tral diffusion, as shown in Fig 2(c). It indicates that
native single Ce ions have a surprisingly good spectral
ability under resonant excitation.
In addition to the native single Ce centers, the spec-
tral stability of Ce ions created by ion implantation has
been investigated (see Fig. 2(d)). With high-dose implan-
tation, we found about 100 ions in one confocal spot (see
Supplemental Material). The optical transition linewidth
of the implanted Ce ions is \(\sim 2\pi \times 150 \text{ MHz} \), increased
mainly due to extra strain introduced by ion implanta-
tion. Since this linewidth is much narrower than the
inhomogeneous width of \(\sim 2\pi \times 550 \text{ GHz} \), it is possible to
address single Ce\(^{3+}\) ions by tuning the excitation laser
wavelength into resonance with the optical transitions.
Compared to other solid-state systems, e.g. defects in
diamond and quantum dots [6 8 9 31 32], implanted
single Ce\(^{3+}\) presents narrow optical transitions and a sta-
ble spectrum. The combination of narrow optical transi-
tions and spectral stability makes precision optical con-
trol of single Ce ion spins possible comparable to single
praseodymium ions in solids [28 29 30].

In order to measure the excitation spectrum of a single
Ce\(^{3+}\), a CW laser is applied to resonant excitation as
well as a low repetition rate, femtosecond laser with high
peak intensity. If a single Ce\(^{3+}\) is excited with CW laser
only, its fluorescence intensity shows a smooth decay and
quickly goes to background level in a few seconds, as
shown in Fig. 3(a). The decay curve in Fig. 3(a) is the
observation of a fluorescence time traces of the single
Ce ion under CW laser excitation only. Surprisingly, in
contrast to all other single emitters a gradual decay of
the fluorescence is observed and not as usual a step-wise
bleaching. In order to explain such gradual bleaching
of a single Ce\(^{3+}\) ion, we propose a model involving two
competing processes: 1) photo-ionization from Ce\(^{3+}\) into
Ce\(^{4+}\) and 2) restoration of Ce\(^{3+}\) by taking an electron
from a nearby deep donor. As long as there are enough
deep donors in the vicinity of the ion, the cerium remains
in its trivalent state and fluoresces. However, a gradual
reduction of the number of donors reduces the probability
of restoring the trivalent state of cerium. This leads to a
gradual decrease of the fluorescence intensity. The charge
dynamics of single ions observed here is consistent with
previous observations in ensembles [30 40 41]. It also
explains why attempts of detecting single Ce ions under
CW laser excitation were unsuccessful [26].

Surprisingly, a femtosecond laser featuring a high peak
intensity restores the population of donors, which helps
the Ce ion pumping back to the Ce\(^{3+}\) charge state (see
Fig. 3(a) and (b)). Therefore, to keep the Ce ion pho-
tostable, we apply CW and pulsed lasers simultaneously
in the experiments. Details of these charge dynamics are
discussed in the Supplementary Material.

From the four different optical transitions of single Ce
ions, a \(\Lambda\) system can be formed, consisting of two ground
states and either one of the excited states. In experi-
ments, we choose the \(\Lambda\) system with transitions D1 and
D3. To observe CPT, the pump laser frequency is fixed
on the transition line D1, and the frequency of the probe
laser is swept around the frequency of the D3 transition.
Simultaneously, the fluorescence intensity of the single
Ce\(^{3+}\) ion is monitored, which is shown in Fig. 3(b). It
contains a broad peak with a dip going down nearly to the
background level. The total width of the peak is consis-
tent with the optical transition linewidth (Fig. 2(b)). The
dip is centered exactly at the D3 transition, indicating
that the ground state population is coherently trapped
in a dark state.

The observed dip width is around \(2\pi \times 35 \text{ MHz} \), caused
by several sources of decoherence, including the intrin-
sic linewidth of the ground state spin transition and
the laser power induced broadening. To understand
this \(2\pi \times 35 \text{ MHz} \) CPT linewidth, we perform optically
detected magnetic resonance (ODMR) on the ground
states, to obtain the intrinsic linewidth of the ground
state spin transition. We use laser excitation resonant
with the D3 transition to initialize the ion into the spin
up state. Microwave (MW) radiation is applied through
the wire located next to the position of the ion under
investigated. Then, the MW frequency is swept through
resonance of the ground-state spin transition. The power
of both, laser and MW is kept low to avoid power broad-
ening (tens of microwatts of laser power and \(\sim 1 \text{ dBm}
microwave power according to the waveguide structure).
For the off-resonant MWs, the electron spin stays in the
spin up state and the fluorescence level is low. Once the

![FIG. 3. (a) Fluorescence intensity of a single Ce ion under pulsed and CW laser excitation. (b) Scheme of the laser pulse](image)
MW field is in resonance with the spin transition, the electron is pumped back to the spin down state, resulting in a higher fluorescence yield (see Fig. 4(a)). From the observed high-contrast ODMR signal, we deduce a fidelity of the initialization of more than 98%. The initialization fidelity is much higher than the 50.2% population at 3.5 K. The inhomogeneous linewidth of the spin transition is $2\pi \times 8.4$ MHz in agreement with our previous measurements \cite{30}. This value is smaller than the linewidth of the CPT dip, indicating the resonance is broadened by laser power.

By fitting the measured fluorescence intensity with the solution for the density matrix equations of the four-level system, we obtain excellent agreement between theory and experimental results (see Supplementary Material). From the fits, we can estimate the driving strength to be $2\pi \times 62$ MHz for the D1 transition. The pump laser induced Rabi splitting with a much weaker dip, as shown in Fig. 4(b), further indicates that the observed dip corresponds to the successful formation of a coherent dark state.

To quantify the power broadening effect, we measured the CPT dip width for various laser powers. The linewidths are linearly dependent on the laser power, as shown in Fig. 4(c), in agreement with expectations. Through a linear fitting, we extract a linewidth $\sim 10.7 \pm 3.2$ MHz without laser broadening. The value is consistent with an intrinsic linewidth 8.4 MHz, obtained from the ODMR measurement.

In conclusion, we showed narrow-linewidth, resonant transitions and good spectral stability for both native and focused ion beam implanted single Ce ions. Based on these optical properties, we demonstrated all optical formation of steady-state coherent dark states of single Ce$^{3+}$ by the CPT technique. All-optical control of a single spin qubit based on a Ce$^{3+}$ can thus be realized by dynamically manipulating coherent laser fields. In addition, on-chip photonic circuits for this system which adds another critical element for their use in quantum technology.

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