European Conference on Optical Communication (ECOC) 2022 © Optica Publishing Group 2022

Entangled States in Nd^{3+} Doped Crystals with Fluorite Structure as Qubits

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Abstract *One- and two-exciton collective states with quantum entanglement in ion pairs in Nd^{3+}: CaF_2 and Nd^{3+}: SrF_2 crystals are studied. It is shown that the strong resonance electrical exchange interaction with spin-flip determines the structure of the luminescence excitation spectra of the one-exciton state.*

Introduction

In a number of previous studies on the use of rare earth ions (REI) doped crystals for quantum computing (QC), the authors considered the use of hyperfine levels as qubits [1-15]. Single qubit gate operations with such qubits require two optical pulses with a very small spectral width of the order of kHz. Therefore, such qubits can operate with millisecond sampling time.

However, as we recently discovered [16], REI can also be used for qubits with GHz frequencies. In this case, much shorter light pulses can be used than in the case of qubits with hyperfine levels. This may allow a much faster QC with nanosecond sampling time corresponding to coherence time of ~10 ns at T ~ 10K. In [16], we proposed to use for QC the ions having 4f levels with small and large values of the Judd-Ofelt parameter $U^{(2)}$. It was shown that the former can be used as qubits, while the latter are suitable for auxiliary states. This allows to perform CNOT and other conditional gate operations. We also draw attention to the fact that the pair centers of REI and other ions can also be used for fast QC [17].

In current report, we pay attention to the fact that the experimentally observed and described here the two-exciton states in the Nd^{3+} pair centers in Nd^{3+}: CaF_2 (M-center) and Nd^{3+}: SrF_2 (M_1- and M_2-centers) are also suitable for fast qubits, and the one-exciton states of these centers can be used as auxiliary states for implementing CNOT. The scheme of performing gate operations in this case is given in Fig. 1.

Due to the large value of the square of the reduced matrix element $U^{(2)} \sim 0.9$, the quadrupole and dipole electronic transitions between the ground $^4I_{9/2}$ and excited $^4G_{5/2}$ states in the Nd^{3+} ion have very high probability, one or two orders of magnitude higher than in the case of other 4f-4f transitions of all REI, and precisely among all transitions from the ground states. This makes the transition suitable for studying quantum states, their cooperative emission and entanglement in pair, triple, quartet and other complex centers of the Nd^{3+} ion [18].

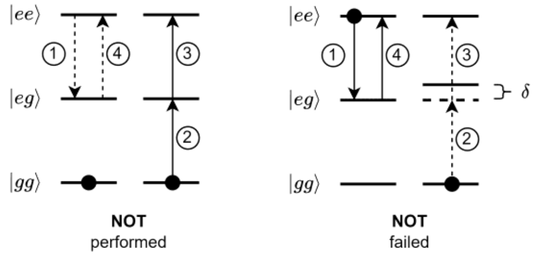

Fig. 1. Scheme of operation of CNOT quantum gate for pair of M-centers. The arrows indicate the excitation of qubits by π-pulses. The number next to the arrow indicates the sequence of a π-pulse in time. The circles show which level of the control qubit is initially occupied. A parameter δ is the offset of the auxiliary level in the target qubit due to a change in the state of the control qubit. The *e* and *g* means excited and ground state, respectively.

In particular, the entangled state $^4I_{9/2}(1) \times {}^4G_{5/2}(1)$ (one-exciton state) formed in the Nd^{3+}: CaF_2 crystal at liquid helium temperatures under laser excitation at the $^4I_{9/2}(1) \rightarrow {}^4G_{5/2}(1)$ transition of the M-center (Fig. 2) has a collective character, which manifests itself in the presence of four spectral peaks in the excitation spectra with the energy difference of the order of one cm^{-1} (see Fig. 3 and [19, 20]). According to [19], the collective origin of the one-exciton state in the M-center is the result of a high rate of resonant exchange of electronic excitation between two Nd^{3+} ions. However, the reason for the appearance of four lines in this spectrum remained unclear in [19]. To elucidate the question of the interaction mechanisms responsible for the observed structure of the one-exciton state of the M-center in the Nd^{3+}: CaF_2 crystal, we measured the excitation spectra of this state using a tunable nanosecond pulsed laser, and for the two-exciton $^4G_{5/2}(1) \times {}^4G_{5/2}(1)$ state we used time-synchronized two-color pulsed laser excitation. Since all the measurements were done at $T = 6.5K$ only the lowest Stark energy level (1) of each manifold was populated.

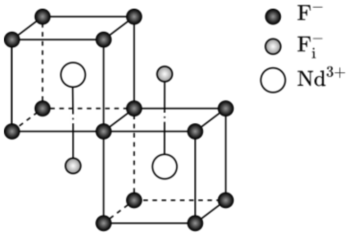

Fig. 2. The structure of the M center in the Nd^{3+}: CaF_2 crystal.

Experimental

The oxygen-free Nd^{3+}(0.6%): CaF_2 (type 1) and Nd^{3+}(0.5%): SrF_2 crystals were grown by the Bridgman-Stockbarger technique in fluorine atmosphere.

The biharmonic pumping was performed in the spectral range of 564–590 nm into the $^4G_{5/2}$ level of Nd^{3+} ion by tunable pulsed Rhodamine 6G dye laser DL-Compact (Estla Ltd., Estonia) with laser line width $\Delta\lambda$ = 0.0065 nm FWHM, pumped by the 2nd harmonics of Nd:YAG (model LQ215, f = 10 Hz, t_p = 14 ns, Solar, Belarus), and Continuum Sunlite OPO system PL 9010, TRP with EX OPO frequency extension module (signal 405–705, idler 715–1750 nm, laser line width $\Delta\lambda$ = 0.003 nm FWHM**)** pumped by 2nd harmonics of Continuum Nd:YAG laser with seeder (f = 10 Hz, pulse duration 5 ns). The wavelength of excitation was controlled by the wavelength meter WS 5 (HighFinesse, Germany) with an accuracy of 0.001 nm. The luminescence was detected by the Shamrock 750 spectrometer system (Andor, Oxford Instruments, UK) with 300, 1200 or 1800 grooves per mm gratings with linear inverse dispersion down to 1.6 nm/mm and the gated Andor iCCD camera iStar DH320T-18H-13 (26 µm pixel size) with Peltier cooling system. The FF01-593/LP-25 edge-filter (Shamrock) was used to limit a laser radiation at the front slit of the monochromator in case of luminescence detection at $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}$ transition. In the case of detection of upconversion fluorescence at the $^4D_{3/2}$ (1) $\rightarrow$ $^4I_{11/2}$ transition of Nd^{3+}, no filter was used. The whole system and the timing of the laser pulses were synchronized with Digital delay/pulse generator DG645/1 (Stanford Research Systems, USA) with the accuracy of 1.5 ns determined by a jitter of the internal pulse generators of Nd:YAG lasers. The samples were placed in the Janis CSS-300S/204N cryostat (USA) and kept at a temperature of 6.5 K. The laser beams combined in one path passed sequentially through two iris diaphragms and focused by a lens (f = 180 mm) to a sample.

Results and Discussion

Using time-resolved site-selective laser spectroscopy, we have confirmed the splitting of the one-exciton $^4G_{5/2}(1)$ x $^4I_{9/2}(1)$ state of the pair M-center of Nd^{3+} ions into four sublevels in Nd^{3+}: CaF_2 crystals at low temperatures (Figs. 3,4, orange curve). Like the authors of [21], we did not find the splitting of two-exciton $^4G_{5/2}(1)$ x $^4G_{5/2}(1)$ state for the doubly excited pair of Nd^{3+} ions of the M-center at our two-photon laser resolution of 0.4 cm^{-1}. The position of main excitation peak simulated by a Gaussian (Fig. 4, blue dashed curve) was consistent with the equation $\omega_{dye} + \omega_{OPO} = 2\omega_0$ regardless of the excitation wavelength. In Fig. 4, the dark green curve shows an example of two-color excitation spectrum when the dye laser was fixed at 579.404 nm.

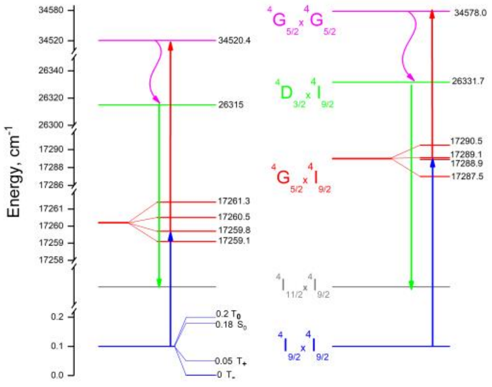

Fig. 3. Energy level diagram illustrating the excitation and detection transitions of upconversion luminescence of the pair M center in the Nd^{3+}: CaF_2 (left) and M_1 center in the Nd^{3+}: SrF_2 (right) crystals. The one-exciton levels consist of four sublevels shown on an enlarged scale.

In the Nd^{3+}: SrF_2 crystal at T = 6.5 K, two types of the pair centers, M_1 and M_2 were found, spectrally shifted relatively to each other by about 2 $см^{-1}$ (Fig. 5). One can conclude that there is a significant inhomogeneous splitting of the one-exciton state of the pair centers of the Nd^{3+} ions in this crystal. As in the case of the pair M-center in Nd^{3+}: CaF_2 crystals, the observed splitting of the one-exciton state for the pair M_1 and M_2 centers in the Nd^{3+}: SrF_2 crystal is several cm^{-1}. This splitting is determined by strong resonance electric exchange interaction between the ions. Due to the absence of such an electric resonance exchange interaction between two excited Nd^{3+} ions, the width of single spectral line of the two-exciton state of the pair M_1 center, as well as for the pair M-center in the Nd^{3+}: CaF_2 crystal, is mainly determined by inhomogeneous broadening and exchange spin-spin interaction. The excitation spectrum of the one-exciton state in Nd^{3+}: CaF_2 consist of four main peaks with different intensities. Two central peaks with the energy difference of 0.7 cm^{-1} have the highest and the lowest intensity among all four peaks. Unlike Nd^{3+}: CaF_2, the excitation spectra of the one-exciton state in Nd^{3+}: SrF_2 consist of three broader peaks (Fig. 5), where the two central peaks are merged due to their smaller splitting comparing to the splitting in Nd^{3+}: CaF_2, while the overall width of the spectra and the difference in the intensities of these peaks is larger. This difference can be explained by stronger spin-orbit interaction in Nd^{3+}: SrF_2 compared to Nd^{3+}: CaF_2 due to stronger relativistic effects of electron motion in the heavier nucleus of Sr^{2+}.

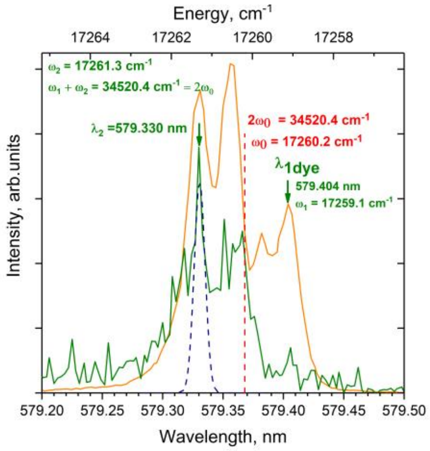

Fig. 4. Fluorescence excitation spectra of the one- and the two-exciton states of the 0.6% Nd^{3+}: CaF_2 crystal at T = 6.5K measured by scanning tunable OPO at the $^4I_{9/2}(1) \rightarrow {}^4G_{5/2}(1)$ transition: NIR fluorescence recorded at the $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}(1)$ transition at 863.56 nm – orange curve; upconversion UV fluorescence recorded at the $^4D_{3/2}(1) \rightarrow {}^4I_{11/2}(1)$ transition at 380.98 nm: under simultaneous excitation by tuned OPO and dye laser fixed at 579.404 nm (shown by arrow) – dark green curve; Gaussian peak positioned at 579.331 nm according to $\omega_{\mathrm{dye}} + \omega_{\mathrm{OPO}} = 2\omega_0$ – blue dashed curve. The position of ω_0 is shown by vertical dashed line.

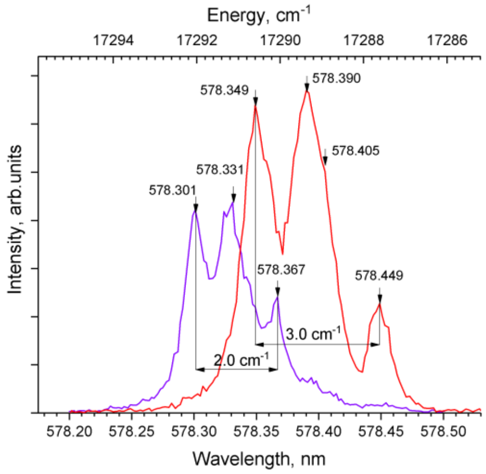

Fig. 5. Fluorescence excitation spectra of the one-exciton states of the 0.5% Nd^{3+}: SrF_2 crystal at T = 6.5K when scanning with the tunable dye laser at the $^4I_{9/2}(1) \rightarrow {}^4G_{5/2}(1)$ transition and detected: at 863.30 nm with a gate delay of 20 ns and a gate width 300 µs (M_1 center) – red curve; and at 863.36 nm with a gate delay of 650 µs and a gate width of 350 µs (M_2 center) – violet curve.

Coherent exchange of electronic excitation

The exchange of electronic excitation between Nd^{3+} ions with conservation of spins can mix the states with the same spin. In this case, the transfer of electronic excitation occurs between pairs of states in the following four pairs [22]: 1) between singlet states, 2) between T_0 -components of triplet states with opposite spin directions in the two Nd^{3+} ions, 3) between $T_{+,-}$ -components of triplet states with the same spin direction in both Nd^{3+} ions. For all of these pairs of states, the exchange of electronic excitation results in the formation of cooperative dark and bright states. In REI, the neglect of spin-flip transitions is not justified because of the rather strong spin-orbit interaction V_{SO} [23, 24]. This interaction causes mixing of bright (dark) singlet states $\left|\Psi_S\right\rangle_{b,d}$ with dark (bright) triplet states $\left|\Psi_{T_-}\right\rangle_{d,b}$. Both, bright and dark triplet states $\left|\Psi_{T_0}\right\rangle_{b,d}$ and $\left|\Psi_{T_+}\right\rangle_{b,d}$ remain unmixed in the first order of spin-orbit interaction V_{SO}. Thus, we get four one-exciton levels with spin-flip and four levels without spin-flip. Of the 32 possible transitions between 8 sublevels of the one-exciton state and 4 sublevels of the ground state, only 12 are allowed. If we neglect the difference in the frequency of states with different spins, then some lines coincide and only four lines remain.

Based on this point, the excitation spectra of the one-exciton states of the M centers in Nd^{3+}: CaF_2 and Nd^{3+}: SrF_2 crystals were numerically simulated, and a good agreement with experimental observations was obtained.

Conclusion

We have proved that the reason for the observed splitting of the one-exciton states of a pair of Nd^{3+} ions in Nd^{3+}: CaF_2 and Nd^{3+}: SrF_2 crystals is a strong resonance exchange of electronic excitation with spin-flip.

We can assume the possibility of fast (GHz) quantum computing (QC) using Nd^{3+} optical pair centers in the Nd^{3+}: SrF_2 and Nd^{3+}: CaF_2 crystals.

Acknowledgements

We wish to thank Estonian Science Foundation for support with grant #PRG347.

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