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Recharging Dynamics of Single Nitrogen-Vacancy Centers in Ultrapure Diamond

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Abstract. Photoluminescence excitation spectra of individual nitrogen-vacancy (NV) centers in diamond are investigated. Recharging dynamics between the neutral (NV^0) and negatively charged (NV^-) states are elucidated and the optimal excitation wavelength for NV^- is found. A sharp resonance at an excitation wavelength of 521 nm facilitates an efficient conversion to NV^0.

Keywords: Nitrogen-vacancy center, photoluminescence excitation, spectroscopy
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INTRODUCTION

The negatively charged nitrogen-vacancy (NV^-) center in diamond is promising for various aspects in the field of quantum optics and engineering due to its extraordinary structural stability and spin properties [1]. Optical excitation and readout of single NV centers are involved in almost all of these experiments. The current challenge in this context is the generation of indistinguishable single photons with NV centers. Under non-resonant excitation there are two major limiting factors present: spectral jumps of emission lines and an unknown dark state of NV^-. So far, there are no systematic investigations of the photo-luminescence (PL) emission of single NV centers according to the excitation wavelength.

EXPERIMENT

We measure photoluminescence excitation (PLE) spectra of single NV centers in ultrapure diamond with a nitrogen atom concentration of less than 5 ppb in the temperature range between T = 10 K and T = 300 K [2]. The experimental conditions are relevant for quantum applications. As a pump source, we use an advanced Er:fiber laser system, which is tunable over the entire visible wavelength range (440 nm < λ_{exc} < 700 nm) and has a pulse duration of 1 ps [3].

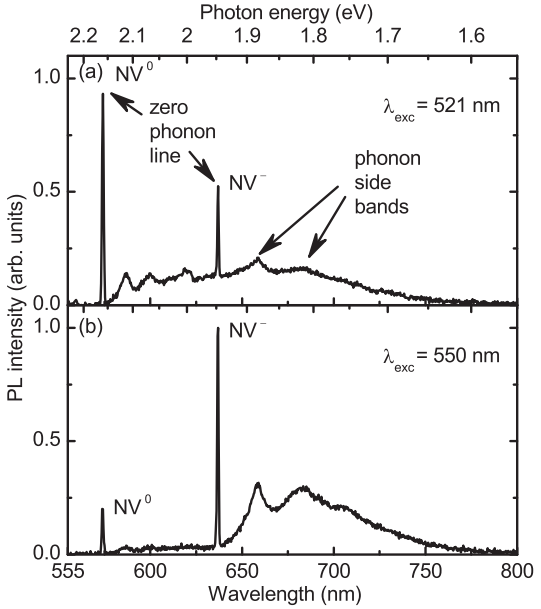

FIGURE 1. Photoluminescence (PL) emission from a single nitrogen-vacancy center under excitation at a wavelength of 521 nm (a) and 550 nm (b) at T = 10 K.

RESULTS AND DISCUSSION

The PL emission of single NV centers is always a

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superposition of features of the NV^- and NV^0 defect center. The PL contribution depends strongly on the excitation wavelength (Fig. 1), but it is unaffected of the excitation regime, i.e. pulsed or cw laser excitation at the same wavelength. PLE spectra of an individual NV color center are assembled in Fig. 2 by plotting the PL intensity of the NV^- (orange circles) and NV^0 (green triangles) charge state as a function of the excitation wavelength (λ_{exc}). The unexpected experimental observations for NV^- are: First, there is no PL signal for $\lambda_{exc} > 575$ nm, which is interpreted as a fast switching to a dark state. Second, by repeating the measurement of NV^- under presence of an additional laser at $\lambda_{exc} = 488$ nm (black squares), efficient PL for $\lambda_{exc} > 575$ nm is observed. Third, the PLE spectrum for $\lambda_{exc} < 575$ nm contains signatures which are expected for the absorption of NV^0.

Our physical interpretation is summarized in Fig. 3(a). Laser optical excitation produces switching between the neural and the negatively charged state in both directions, starting exclusively from the electronically excited states of NV^0 and NV^- [denoted with stars in Fig. 3(a)]. Efficient NV luminescence is detected only in the joint absorption region of both charge states (B) where continuous charge switching is possible. In region (A) and (C) the switching cycles are broken and the center is transferred into its dark state, respectively.

We suggest an intrinsic two-step model for both charge conversion processes [Fig. 3(b)]. The excitation from the ground (GS) to excited state (ES) is followed either by a transition of a hole (h^+) into the valence band for NV^0 (left) or an excitation of an electron (e^-) into the conduction band for NV^- (right).

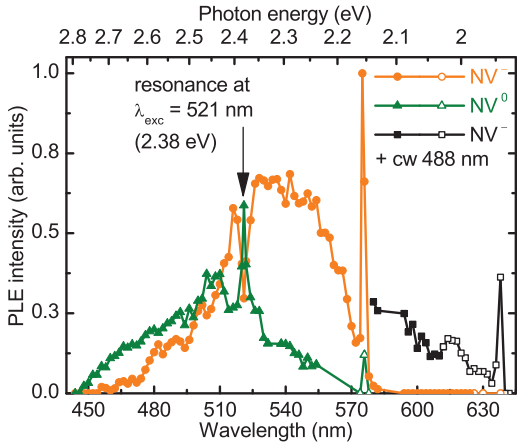

FIGURE 2. Photoluminescence excitation (PLE) spectra of NV^- (orange circles), NV^- with an additional repump laser at $\lambda_{exc} = 488$ nm (black squares), and NV^0 (green triangles) of a single center at T = 10 K. Filled (open) symbols correspond to zero phonon line (rescaled phonon sideband) intensities.

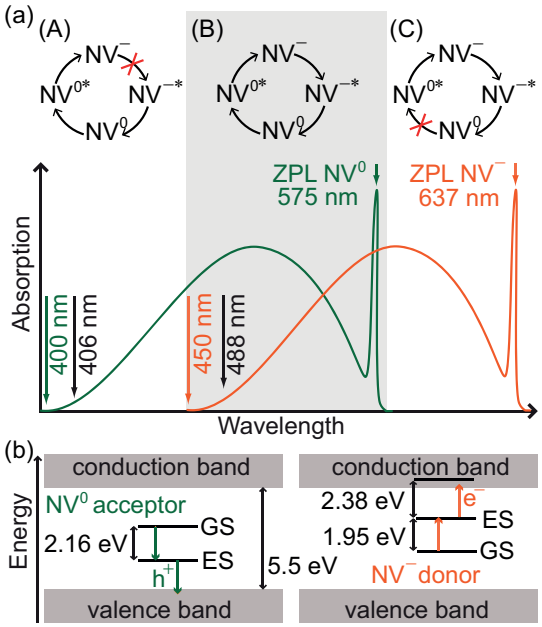

FIGURE 3. (a) Schematic illustration of the absorption spectra of NV^0 (green) and NV^- (orange) along with recharging cycles (top). (b) Simplified electronic energy schemes of the NV center. An additional electronic state of NV^- is located in the conduction band of diamond.

The optimal excitation wavelength for the luminescence emission of NV^- is found at $\lambda_{exc} = 575$ nm. Furthermore, we observe a sharp resonance in the PLE spectra at $\lambda_{exc} = 521$ nm (2.38 eV) which allows an efficient way to convert some NV centers mainly to their neutral form NV^0. We assign this observation to an electronic transition of NV^- originating from its excited state and leading to an energy level located in the conduction band of diamond [Fig. 3(b)]. Not every single NV exhibits the feature at $\lambda_{exc} = 521$ nm and whereas its occurrence correlates with the PL lifetime of NV^-. The shortening of the radiative lifetime of NV^- in combination with the absence of the resonance can be understood as a consequence of random strain of the diamond host lattice.

In conclusion, our analysis provides unexpected and new physical insight into the dynamic operation of NV centers. Our results are relevant for all applications of NV centers involving optical excitation and readout. They pave the way to an efficient generation of indistinguishable photons.

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