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Citation: Appl. Phys. Lett. 81, 2160 (2002); doi: 10.1063/1.1507838
View online: http://dx.doi.org/10.1063/1.1507838
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Single spin states in a defect center resolved by optical spectroscopy

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(Received 30 May 2002; accepted for publication 24 July 2002)

Individual paramagnetic defect centers in diamond nanocrystals have been investigated by low-temperature high-resolution optical spectroscopy. Narrow fluorescence excitation spectral lines have been found, indicating transitions between individual spin sublevels. Spectral diffusion is explained by cross relaxation among spin sublevels and by the presence of excited electrons in the conduction band of diamond. The relaxation times are in the millisecond range. The system may be useful for quantum information processing with individual electron spins. © 2002 American Institute of Physics. [DOI: 10.1063/1.1507838]

Paramagnetic centers in wide-gap semiconductors are becoming increasingly interesting because of their possible application in quantum computation and quantum cryptography. Optical addressable defect centers (color centers) are particularly easy to study because the sensitivity in optical experiments is high enough to detect a single center. Because of their simple chemical composition (sometimes only a single impurity atom or vacancy) they may be considered as atomic-like systems. However, the optical properties of defects in, for example, diamond compared with those of atomic systems, are considerably more complex. In addition to radiative decay, electron-phonon interactions and other matrix-induced relaxation processes contribute to the optical properties of electronic transitions. Here we report on the determination of the homogeneous linewidth of a single nitrogen–vacancy (N–V) center in diamond.

The N–V defect consists of a substitutional nitrogen atom and a vacancy in an adjacent site of the diamond lattice with trigonal symmetry $C_{3v}$. Theoretical modeling and positron annihilation experiments show that the charge state of N–V defect is negative. It exhibits a strong dipole-allowed $E \rightarrow A$ optical transition with a zero-phonon line at 1.945 eV (637 nm), allowing to detect individual centers optically. Electron spin resonance and hole burning experiments have shown that the electronic ground state of the N–V center ($^3A$) is paramagnetic. The coupling of the two unpaired electron spins in the diamond crystal field splits the triplet ground state into $Z(m_z=0)$ and $X, Y(m_z=\pm 1)$ sublevels separated by 2.88 GHz. This fine structure is not accessible using conventional nonselective optical spectroscopy because of a strong (1000 GHz) inhomogeneous broadening of the zero-phonon line. Low temperature spectroscopy of single centers has the potential to unravel this fine structure splitting and thus gives hope to determine the spin state of a single electron spin.

Previous single center spectroscopy in large diamond crystals at low temperature has revealed unexpected limitations. First, the presence of a singlet metastable state $^1A$ (see Fig. 1) reduces the fluorescence intensity of the N–V centers below a temperature of 80 K at resonant excitation. Second, the light emitted by N–V centers in bulk diamond is hard to extract because of the high index of refraction of diamond ($n \approx 2.4$). In this letter we show that use of diamond nanocrystals containing N–V centers is an efficient way to overcome both problems. The intersystem crossing of N–V defects causing shelving effect is reduced in diamond nanocrystals, allowing one to detect individual centers with resonant excitation. In addition, subwavelength size of diamond nanocrystals makes surface reflection effect irrelevant.

Single defect measurements were performed using a home-built low temperature confocal microscope (for details of the experimental setup see Ref. 9). As sample material diamond powder was used (De Beers) with a grain size of 250 nm, made of synthetic high-pressure type 1b diamonds. Vacancies have been created by irradiation with 2 MeV electrons. The irradiation dose was $10^{17}$ e/cm$^2$.

Figure 2 shows the fluorescence emission spectrum of a single nanocrystal at $T=2$ K. The two sharp zero-phonon lines correspond to fluorescence emission from single N–V centers present in the nanocrystal. Diamond nanocrystals show an approximately four times wider spread of the spectral line positions of a single center compared to bulk dia-

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FIG. 1. Schematic representation of energy levels of N–V center.
High-resolution optical spectroscopy of individual N–V centers was performed by scanning a single mode ring dye laser (resolution 1 MHz) across the pure electronic transition. Holographic 637 nm Notch and 695 nm redpass filters have been used to reject laser stray light and Raman scattering. Fluorescence was detected by a single photon counting avalanche photodiode. A typical excitation line of single N–V centers was performed by scanning a single mode ring dye laser, respectively. Spectrum 3 shows high-resolution 637 nm laser sweep across zero-phonon transition of single defect.

In order to determine the number of N–V defects present in nanocrystallites we performed fluorescence antibunching experiments on individual nanocrystals (data shown in inset of Fig. 2). The contrast of the fluorescence intensity autocorrelation function indicates that each narrow line visible in fluorescence emission corresponds to the fluorescence of a single N–V defect. The contrast of autocorrelation function (0.5) shows the presence of two N–V centers.

The fluorescence excitation spectrum of a single N–V center at 2 K is shown in Fig. 3. This is probably due to enhanced strain in the lattice of nanocrystals. The zero-phonon transition at 514 nm of the first excited state has a Lorentzian shape. The linewidth of around 150 MHz is mostly determined by the shortest component of the radiative decay (2 and 8 ns). Illumination of the sample with the 488 nm line of an Ar+ laser, as in previous experiments, leads to a dramatic increase of the linewidth. A possible explanation is the Stark effect induced spectral broadening that is caused by the interaction of the N–V center with electrons in the conduction band. Atomic nitrogen is always present in high concentration (200 ppm) in type Ib diamond. Nitrogen is a deep electron donor, 2 eV below the conduction band. It is known, however, that nitrogen electron may be photoexcited into the conduction band. Since the N–V center is charged (N–V-), the narrow excitation lines of single defects are extremely sensitive to the charge fluctuation in their vicinity, giving rise to the large linewidth reported previously, when a blue laser line (488 nm) had been used for deshielding of the 1A state. Since the spectral width of single center is much narrower than the zero-field splitting in the ground state, the excitation line shown in Fig. 3 marks a specific spin configuration of the defect.

In Fig. 4, the fluorescence intensity of a single N–V defect is plotted as a function of time. When the excitation laser is in resonance with the 1A–3E transition, the fluorescence exhibits a telegraph behavior. The duration of bright intervals decreases upon increase of excitation intensity, indicating a photoinduced process. This blinking effect may be related to the internal spin dynamics of the center. Upon resonant excitation, the center performs excitation-emission cycles between resonant spin sublevels of the 1A–3E state (see Fig. 1), resulting in intense fluorescence bursts. During this excitation cycle the spin memory is preserved. The intersystem crossing yield to the 1A singlet state is small, but when this transition takes place spin memory is lost and the defect may escape to a different spin sublevel of the ground state. After the spin flip, the center is no longer in resonance with the excitation laser because of the differences in ground and excited state fine structure splitting. Hence, intersystem crossing may lead to the dark interval in fluorescence emission. The system finally comes back to resonance as a result of the spin-lattice relaxation in the ground state.

In summary, individual N–V defects in diamond nanocrystals have been investigated by low-temperature optical spectroscopy. Individual centers reveal excitation linewidth around 150 MHz, which is sufficiently small to resolve a specific spin state of the center. The selected spin state may

FIG. 2. The fluorescence emission spectrum recorded at 2 K, for a bulk diamond crystal containing N–V centers (1) and diamond nanocrystal (2). Excitation was at 514 nm. Inset shows fluorescence autocorrelation function for the nanocrystal. The contrast of autocorrelation function (0.5) shows the presence of two N–V centers.

FIG. 3. The fluorescence excitation spectrum of a single N–V center at 2 K. Spectra 1 and 2 have been recorded with and without deshieving 488 nm laser, respectively. Spectrum 3 shows high-resolution 637 nm laser sweep across zero-phonon transition of single defect.

FIG. 4. Duration of bright intervals in the fluorescence emission as a function of excitation intensity. Inset is the time trace of single N–V center fluorescence intensity. Excitation was resonant with 637 nm zero-phonon line. Temperature was 2 K.
be observed for up to a second under constant optical excitation. The present study can be an important step towards quantum computation with pure spin states.

This work was supported by DFG and NATO research grants. The authors acknowledge Dr. F. Philipp and Dr. W. Spengel for the irradiation of diamonds.