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Single-photon generation from a nitrogen impurity center in GaAs

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We have demonstrated single-photon emission from a nitrogen luminescence center in GaAs. An inhomogeneously broadened luminescence band formed by localized centers was observed in the spectral range from 1480 meV to 1510 meV at 5 K in nitrogen delta-doped GaAs. Optical properties of the individual centers were investigated by steady-state and time-resolved micro photoluminescence. We have found that a bright luminescence center emits single photons with a radiative lifetime of 650 ps, which is much shorter than the lifetime of NN pairs in previous reports. © 2012 American Institute of Physics. [doi:10.1063/1.3679181]

Single dopants in semiconductors have currently attracted significant attention because of their potential application to single-spin devices or single-impurity transistors.[1] Single dopants are also promising candidates for a non-classical light source. In recent years, single-photon emission has been demonstrated in impurity centers in semiconductors, such as nitrogen-vacancy centers in diamonds,[2,3] N acceptors in ZnSe,[4] and Te pairs in ZnSe.[5] In principle, single photons with well-defined energy could be produced from homogeneous impurity centers in high quality crystals. To have well-defined energy is an important factor for the coupling with microcavities and photonic crystals, as well as for the generation of indistinguishable photon pairs.[6] Energetically defined single-photon generation has been demonstrated in highly homogeneous NN pairs in GaP.[7] The radiative lifetime of NN pairs in GaP is rather long (56 ns),[8] probably due to the indirect bandgap nature of the host material. For reliable triggered generation of single photons with short time intervals, a shorter radiative lifetime of the emitter is favorable. From this perspective, nitrogen impurity in GaAs may be promising because shorter radiative lifetimes are expected due to the direct bandgap structure of the system. Moreover, sharp photoluminescence (PL) peaks have been observed even in the ensemble PL spectrum in nitrogen delta-doped GaAs.[9] Most luminescence peaks observed in the bandgap in GaAs:N have been assigned to the bound exciton luminescence due to the NN pair centers.[10,11] While spectroscopy on single centers in GaAs:N has been performed,[12–17] single photon generation has not yet been observed in this system. In this letter, we demonstrate single-photon emission from a single nitrogen luminescence center in nitrogen delta-doped GaAs.

The sample was grown at 550 °C on a Cr-O doped semi-insulating GaAs(001) substrate by low pressure metal-organic chemical vapor deposition. Triethygallium and arsine were used as sources for the GaAs epitaxial layers, and dimethylhydrazine was used as a nitrogen dopant. GaAs:N of nominal monolayer thickness was inserted

between 250 nm-thick buffer and cap layers. A nitrogen sheet density of $2.9 \times 10^{12} \, \text{cm}^{-2}$ was determined by secondary ion mass spectroscopy. The densities of other impurities, such as carbon and oxygen, were below the detection limit. The sample was kept in a cryostat. A hemispherical solid immersion lens was attached to the sample surface to improve the spatial resolution and the collection efficiency. The luminescence of the sample was collected with a microscope objective × 40 lens (NA = 0.5) in a confocal optical system and was transferred to an imaging spectrometer with a focal length of 25 cm, equipped with a charge-coupled device (CCD) and a spectrometer with a focal length of 92 cm. A 532 nm continuous-wave (cw) solid-state laser, a semiconductor pulse laser emitting at 405 nm, and a cw Ti:Sapphire laser were used as the excitation light sources for the static, time-resolved, and photoluminescence excitation (PLE) measurements, respectively. Two types of silicon avalanche photodiode (APD) based single-photon detectors were used: one with a faster response (Micro Photon Devices) for PL decay time measurements, and the other with a higher efficiency (PerkinElmer) for photon correlation measurements.

Figure 1(a) shows a macro-PL spectrum of the sample at 5 K, which was obtained with a large pinhole, corresponding to a circular detecting area of diameter 14 μm. In addition to the GaAs exciton peak, labeled E_{ex}, a smooth band, which has been marked by a solid circle, as well as a rough and broad luminescence band were observed below the exciton energy. These two bands are obviously caused by the nitrogen impurity, because they are not observed in samples that do not contain the GaAs:N layer. The broad, rough band arises from localized states, which are due to the nitrogen luminescence centers. This assignment is supported by the presence of many spots in the PL image, shown in the inset, which was obtained using a 10-nm (18 meV) band-pass filter centered at 830 nm (1493 meV). Aiding in this confirmation is the spatial intensity profile of the CCD (Fig. 1(b)) for the PL spectrum (Fig. 1(a)). Since the image was obtained by the imaging spectrometer, the horizontal axis corresponds to the photon energy and the vertical axis corresponds to the position on the sample surface. While the spatial profile at the exciton energy is uniform, as expected, many spots are

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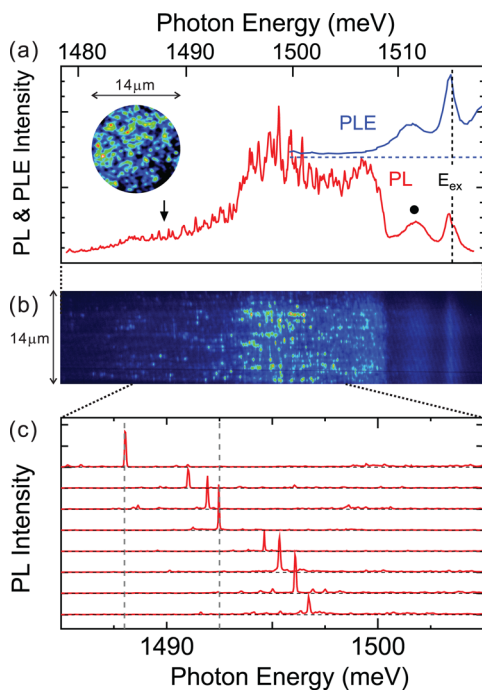
FIG. 1. (Color online) (a) PL spectrum of nitrogen delta-doped GaAs at 5 K. The luminescence band, ranging from 1480 meV to 1510 meV, is due to the nitrogen luminescence centers. The PLE spectrum detected at 1488 meV, marked by an arrow, has a peak at 1512 meV, in addition to exciton absorption E_{ex}. Inset: PL image taken with a 10-nm (18 meV) band-pass filter for the wavelength of 830 nm (1493 meV). (b) Corresponding spatial intensity profile on the CCD. Localized and delocalized states can be clearly distinguished by the presence of the spots. (c) Examples of μ-PL spectra. Vertical dashed lines represent the NN pair energies of NN_F in Ref. 18 and the "1.4925 eV line" in Ref. 9.
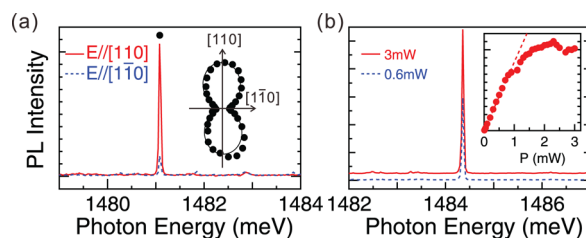

FIG. 2. (Color online) (a) Polarization resolved PL spectrum of a single center. The PL spectrum is strongly polarized along the [110] crystal axis and has only a single peak. Inset: Polar plot of the peak intensity. The angle dependence has been fitted by $a \cos^2 \theta + b$ for a:b = 1:0.14, where θ is the angle between the polarization direction and [110] crystal axis. (b) PL spectrum of a single center below and above the saturation excitation power. The spectrum still has a single peak in the saturation regime. Inset: PL intensity vs. excitation power.

observed for the rough band. It is also evident from the data that the PL spectrum of the nitrogen luminescence center in our sample was inhomogeneously broadened, in contrast to a previous report.[9] The small band, marked by a solid circle in Fig. 1(a), appears to be a continuous and delocalized state, since we see no spots for this band in the spatial intensity profile. The result of PLE measurement, which was detected at about 1488 meV, is consistent with this speculation because the PLE signal, along with the exciton absorption, peaks at 1512 meV. Spatially resolved PL (μ-PL) spectra were obtained by inserting a small pinhole in the confocal plane that selects a 0.6 μm diameter area. Examples of μ-PL spectra are presented in Fig. 1(c). Each spectrum is practically due to a single luminescence center. Although the first and fourth spectra, from the top, match the energies of the NN pairs in the literature,[9,18,19] which are represented by vertical dashed lines, we believe that this is just a coincidence, and that these energies are not associated with the same NN pairs that are described in the literature because the polarization property of the line is substantially different, as we will discuss later. Figure 2(a) shows the polarization resolved PL spectrum of a single luminescence center. The width of the peak is below the resolution limit of our system (45 μeV). The PL spectrum of the single center was found to be strongly linearly polarized along the [110] crystal axis. Strong linear polarization along the [110] or [1$\bar{1}$0] crystal axis is a common property of many of the luminescence centers investigated in this work.

The observations of strong linear polarization and areal density of the centers lead us to believe that the luminescence center is [110] (or [1$\bar{1}$0])-oriented NN pairs. However, the luminescence center has unusual spectral features that are not common for a NN pair. It is reported that the luminescence from a NN pair bound exciton exhibits fine structure splitting due to exchange and local crystal field interactions.[20,21] Since the local crystal field is closely related to the atomic configuration of the luminescence center, the fine structure splitting and their polarization direction are of particular importance to determine the atomic configuration of the center. For example, for a [110]-oriented NN pair, four linearly polarized components, which are [110] and [1$\bar{1}$0]-polarized bright exciton components and the corresponding dark components, are expected if we look the NN pair from [001] direction. Such a fine structure splitting has been observed in polarization-resolved PL spectra from a single NN pair in GaAs,[12,14,15] as well as in GaP.[8] The PL spectrum in Fig. 2(a) has a single peak, in contrast to the above examples. Under low temperature and low excitation power, the number of observed emission peaks could be reduced by thermalizing the population to the lowest state. To investigate this possibility, we inspected the excitation power dependence of the polarized PL spectrum and found that no additional component emerged as shown in Fig. 2(b). In addition, we found that the spectral shape was insensitive to changes in temperature. It is not likely that the luminescence arises from charged excitons bound to a NN pair[16] because PL from a charged exciton does not show significant linear polarization.

PL decay curves of single centers were measured using an APD and a pulsed semiconductor laser with a pulse width of 52 ps. Convolution analysis was applied in order to reproduce the experimental data. Though the PL intensity of single centers in steady-state was distributed, the majority of the luminescence decay times of the centers was shorter than 2.5 ns at 5 K. The shortest decay time observed in our sample was 650 ps, as shown in Fig. 3(a). Photon correlation measurements were performed for a luminescence center with a short decay time using a Hanbury Brown-Twiss setup with pulsed optical excitation at 50 MHz. Figure 3(c) shows photon correlation of a luminescence center at 1492.49 meV, which has been spectrally selected by a narrow band-pass filter. The strong reduction of the central peak unambiguously
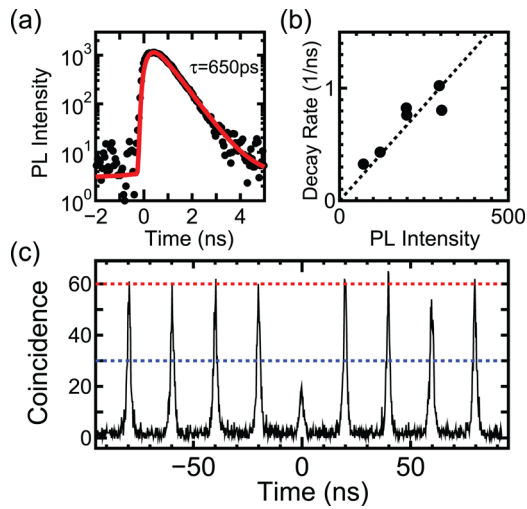
FIG. 3. (Color online) (a) PL decay of a single center that has the shortest decay time, 650 ps. Also shown, in red, is the convoluted fitting curve. (b) PL decay rate vs. PL intensity in the saturation regime under cw excitation. (c) Distribution of coincidence counts for a single impurity center in nitrogen delta-doped GaAs.

sity, the localized luminescence center could be assigned to a NN-pair-like center. However, the atomic arrangement of the observed luminescence centers is not explored in this work. Bright centers in static PL spectra tend to show faster radiative decay rates, where the radiative decay rate of the brightest center is more than one order of magnitude faster than that of a NN pair in GaAs.

confirms the single photon generation from a single nitrogen luminescence center in GaAs. The residual signal at the central peak is due to the background luminescence that was not entirely rejected by the band-pass filter.[22]

The observed short lifetime is not the result of the fast non-radiative energy relaxation mechanism because PL intensity was constant below 10 K. Thermally activated non-radiative mechanisms are negligible at 5 K. Moreover, bright centers in static PL spectra tend to exhibit shorter decay times, as shown in Fig. 3(b). The decay rate is approximately proportional to the maximum PL intensity in steady-state, which indicates that the observed PL decay time is equivalent to the radiative lifetime. Note that the radiative lifetime of the centers studied in this work is much shorter than those of NN pairs in GaAs (about 10 ns)[10] and GaP.[8]

In summary, single-photon emission was demonstrated using a single nitrogen impurity center in GaAs. In accordance with the observed linear polarization and the areal den-

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