Ultrafast nonlinear optical effects induced by nitrogen-vacancy centers in type-IIa diamond

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Introduction

The optical and magneto properties of the negatively charged nitrogen-vacancy (NV) center in diamond has been extensively investigated because of its application to quantum sensing, such as magnetic and electric fields and to quantum information technologies. In most cases, near-infrared luminescence upon the excitation by green laser can be controlled by the irradiation by microwave, which tunes the optical transition path between the excited and ground states. To extend the optical property of diamond over the current generation of linear optical regime, one requires ultrashort laser pulses, which enables one to induce nonlinear optical effect, such as the nonlinear refraction and nonlinear absorption. Almeida et al. recently reported on nonlinear optical spectra in high-purity diamond using femtosecond laser pulses [1]. Because of the absence of defect-related bands below the band-gap (Eg = 5.5 eV), the two nonlinear optical effects were enhanced for $E = E_g/2$. Although there are several investigations on nonlinear optical effects in diamond and nano-diamonds [2], the effects of the NV center on the nonlinear optical phenomena have not yet been examined. Here we explore the NV center induced nonlinear optical effects in pure diamond, specifically optical Kerr effect (OKE) and two-photon absorption (TPA) in transparent region using 800 nm light, at which NV related bands will be sensitive.

Method

We carried out Z-scan measurement to examine nonlinear optical effects, such as OKE and TPA [3]. The Z-scan measurements were performed using a closed aperture mode. To investigate the time-domain response from the OKE and TPA due to the NV centers near the sample surface, reflection-type pump-probe measurements for the diamond samples were also carried out at room temperature. The light source used was a femtosecond amplifier system, which produces 40 fs pulses at a central wavelength of 800 nm at 100 kHz repetition rate. The samples used were type-IIa diamond crystal fabricated by CVD method. To introduce NV centers 30 keV nitrogen ions (N^+) were implanted into the diamond samples at the N^+ dose levels of $2.0\text{x}10^{11}$ and $1.0\text{x}10^{12}$ ions/cm^2 (Fig. 1). The implanted depth deduced from the Monte Carlo calculation (TRIM) was about 30–40 nm. After that, the samples were annealed at 900–1000 ºC in an argon atmosphere for 1 hour to produce NV centers.

Results and discussion

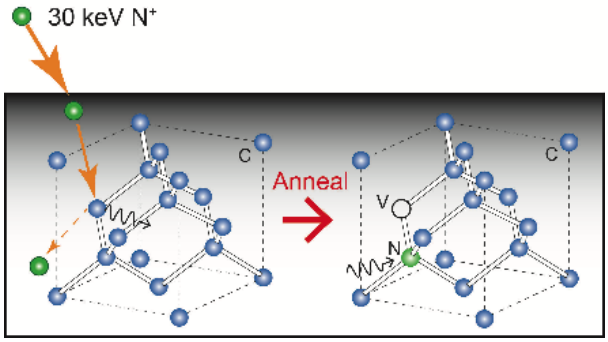

FIG. 1 Schematic for the production of NV centers.

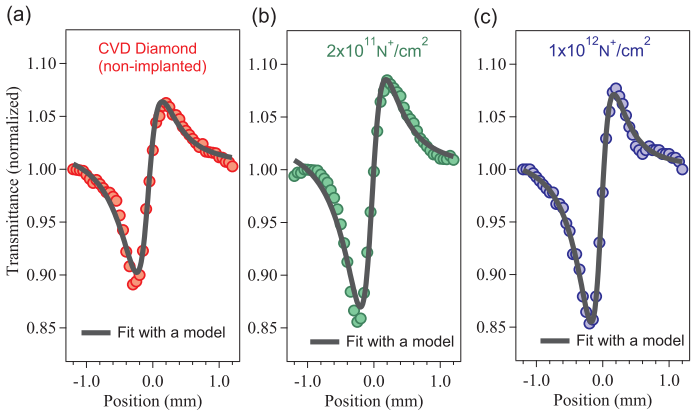

FIG. 2 The Z-scan results at the laser fluence of 20 mJ/cm^2.

Figure 2 shows the results of the Z-scan measurements obtained for different N^+ dose levels. Anti-symmetric line shape, whose peak and valley have not the same amplitude, was observed for all the samples. The anti-symmetric lineshape, with a valley greater than the peak, indicates an existence of two-photon absorption [1]. The intensity of the transmittance change becomes larger when the NV centers were introduced at $2.0\text{x}10^{11}$ and $1.0\text{x}10^{12}$ N^+ /cm^2. In particular, the valley at z = -0.2 mm exhibits ≈1.3 times larger transmission change compared to the pure diamond (non-implanted), indicating that absorption by NV centers plays a central role in the enhancement of the signals in Figs. 2(b) and 2(c). In addition, pump-probe reflectivity data demonstrate that the OKE signal is strongly enhanced for $1.0\text{x}10^{12}$ N^+ /cm^2. We suggest that the strong enhancement of the OKE is possibly originated from cascading OKE, where the high-density NV centers effectively break the inversion symmetry near the surface region of diamond [4].

References

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