Semiconductors, 2024, Vol. 58, No. 9

Negatively charged nitrogen-vacancy centers in a silicon carbide crystal of $6H$-^{28}SiC

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Received April 19, 2024
Revised August 14, 2024
Accepted October 30, 2024

In this work, high-spin ($S = 1$) color centers in isotopically modified $6H$-^{28}SiC crystal were identified using high-frequency electron paramagnetic resonance techniques. The components of the spin Hamiltonian (g, D, A) of negatively charged nitrogen-vacancy (NV^-) centers are determined and the influence of optical excitation modes on their relaxation characteristics is studied. The obtained results prove the potential possibility of using NV^- defects in $6H$-^{28}SiC for the material implementation of qubits and spin-photonic interfaces.

Keywords: Spin defects, silicon carbide, optical polarization.

DOI: 10.61011/SC.2024.09.59915.6309A

1. Introduction

The quantum mechanical theory has forever changed the classical view of the seemingly familiar and well-studied world, providing the interpretation of physical phenomena and observations in a fundamentally new way. The quantum mechanics once again gives researchers hope for a breakthrough more than a century after its inception, this time in the computational impasse associated with reaching the limit in the productive power of modern computers [1]. It is assumed that the development of quantum technologies in a short time will allow solving a number of interdisciplinary problems and tasks beyond the power of classical analogues [1,2].

The material base of quantum processors can be represented as a set of qubits in a structurally protected matrix, which are the basis for the organization of quantum simulators [2]. The defects in wide-band semiconductor crystals with a high spin state and optical absorption and radiation spectra in the visible and infrared (IR) ranges are particularly prominent candidates among the variety of physical implementations of qubits [3,4]. The effective interaction of the optical excitation quantum and the electronic state of the defect makes the color center an attractive object for creating spin-photon interfaces capable of operating at room temperature together with existing optoelectronic technology. A silicon carbide (SiC) crystal can serve as a host matrix for a large family of color centers, being a semiconductor with a wide range of applications [5,6].

2. Material

This paper studies $6H$-SiC crystals with isotopic enrichment of ^{28}Si nuclei (nuclear spin $I = 0$) grown by high-temperature sublimation from the gas phase to the solid [7]. The concentration of nitrogen in the crystal $6H$-^{28}SiC was $C \approx 10^{17}\,\text{cm}^{-3}$. $6H$-^{28}SiC samples were irradiated with electrons with an energy of 2 MeV and a dose of $4 \cdot 10^{18}\,\text{cm}^{-2}$, after which they were annealed at a temperature of $T = 900^\circ$C in an argon atmosphere for 2 h.

3. Experiment procedure

The samples were studied by high-frequency (94 GHz, W-band) electron paramagnetic resonance (EPR) using a Bruker Elexsys E680 spectrometer. Irreversible decay of the electron spin echo (ESE) was obtained using the Hahn sequence ($\pi/2-\tau-\pi-\tau-$ESE) with pulse duration of $\pi/2 = 40$ ns, $\pi = 80$ ns and pulse interval of $\tau = 1.2\,\mu$s. The transverse relaxation time (T_2) was obtained with a fixed B_0 and an increase in the interval τ in increments of 64 ns. Continuous solid-state lasers with $\lambda = 980$ and 1064 nm with an output power of P up to 500 mW were used.

4. Results and discussion

The EPR signal in the samples below spectrometer sensitivity without irradiation with light over a wide temperature range (297−50 K). When the laser is turned on, the EPR spectra of the studied color centers in the $6H$-^{28}SiC crystal are observed (Figure 1, a and b), consisting of resonant absorption of defects of various nature. The components of interest are highlighted by rectangles (Figure 1, a), which are associated with negatively charged NV^--centers with electron spin $S = 1$. The thin structure with the center of gravity of the spectrum at $g = 2.003$ (the spin nature of magnetism) is caused by the splitting of spin sublevels in

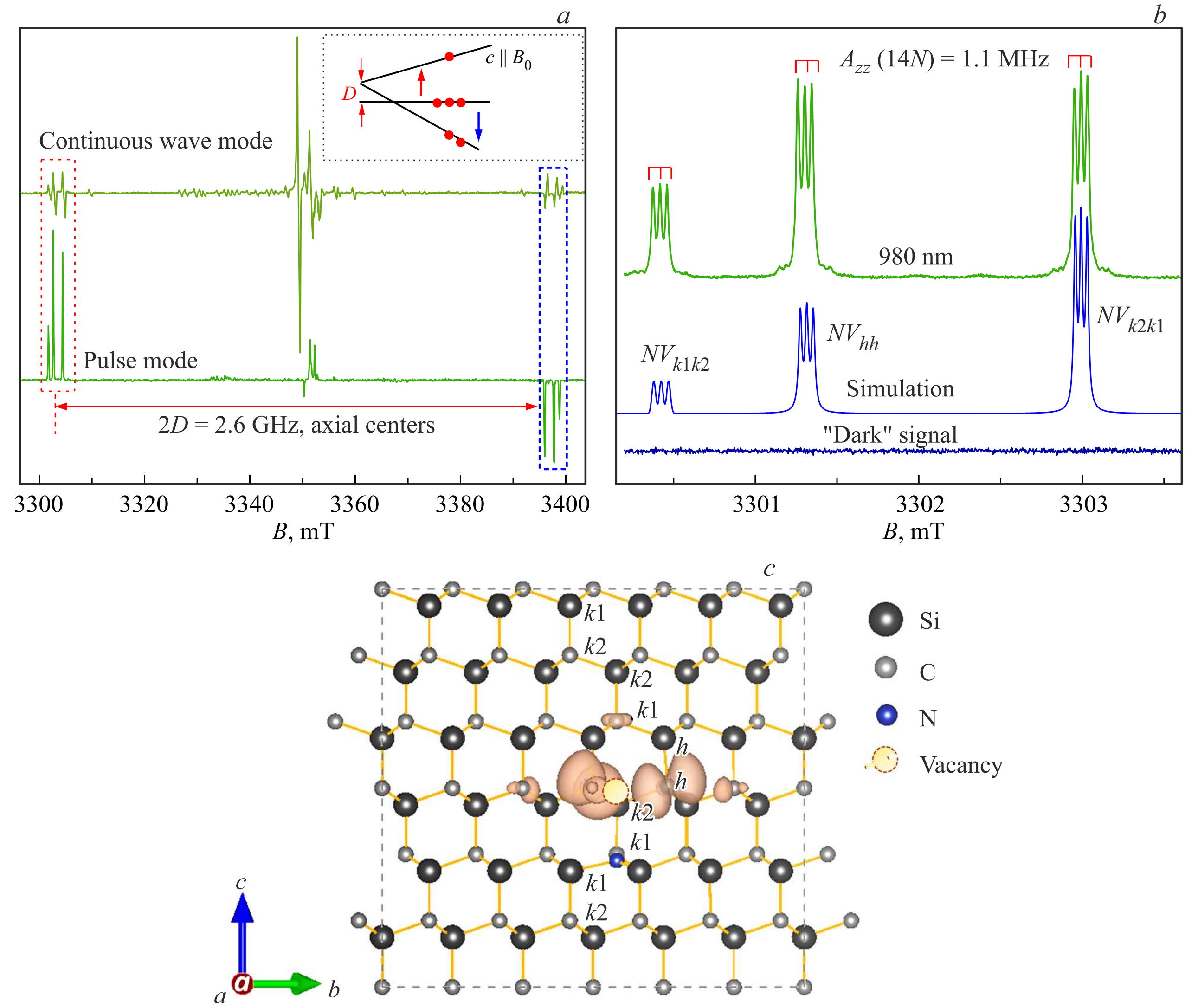

Figure 1. *a* — EPR spectra (980 nm) in stationary (top) and pulsed (bottom) modes at $c \parallel B_0$. The insert shows the spin levels of the defect with $S = 1$ under the action of optical excitation; *b* — low-field component of the fine structure of the NV^--center in the pulsed mode (green line —experiment (980 nm), blue line — simulation with spin Hamiltonian parameters given in the text of the article), highlighted by a rectangle in Figure 1, *a*. „Dark“ signal is represented by purple; *c* — the defect model of the NV^--center in the crystal lattice 6*H*-SiC. (A color version of the figure is provided in the online version of the paper).

a zero magnetic field of magnitude $D \approx 1.3$ GHz due to the spin-spin interaction [8]. Optical excitation leads to a predominant population of the state with $M_S = 0$ (see the box in Figure 1, *a*), thereby forming a population inversion and a microwave radiation signal (high-field component). Given the absence of an EPR signal in crystals without light exposure, it can be assumed that the degree of polarization of the electronic system is close to 100%, necessary for the „initialization“ stage of quantum computers. Figure 1, *b* shows a detailed low-field component of a fine structure consisting of three contributions due to the presence of structurally unequal positions of the NV^--center (k_1k_2, *hh*, k_2k_1) in 6*H*-^{28}SiC (Figure 1, *c*).

A slight difference of the microstructure and distribution of the spin density of defects affects the value of *D*, which makes it possible to selectively excite each color center. Thus, there are three independent qubits in the electronic subsystem in 6*H*-^{28}SiC that are excited at three different frequencies. The distance between the centers NV_{k1k2} and NV_{k2k1} is 70 MHz, which allows the use of standard radio frequency generators for quantum manipulation of the nuclear subsystem or microwave generators for manipulation of electronic qubits. The presence of a magnetic nucleus ^{14}N with spin $I = 1$ near the silicon vacancy leads to the formation of an additional hyperfine splitting $(2I + 1)$ of each component of the fine structure into three equidistant lines of the order of $A_{zz} = 1.1$ MHz. Coherent electron-nuclear coupling provides an additional degree of freedom when conducting experiments with multipulse sequences [9,10] and the potential feasibility

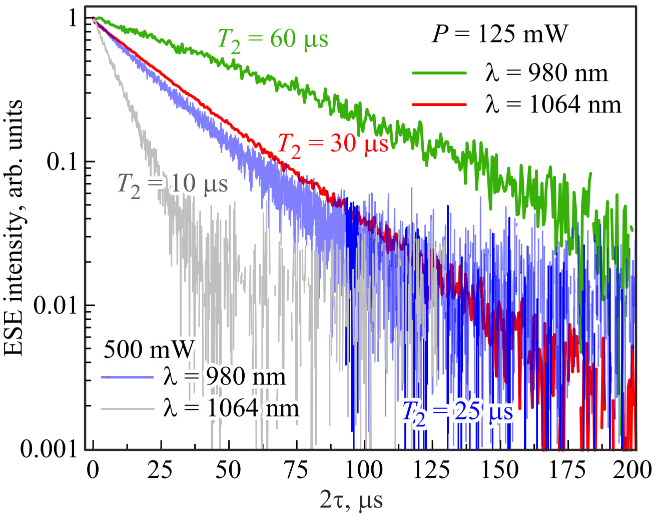

Figure 2. Transverse magnetization decay curves of NV^--centers on a semi-logarithmic scale at $T = 150$ K under the action of optical excitation with a power of 500 mW (blue and gray colors) and 125 mW (red and green colors) for wavelengths 980 (blue and green lines) and 1064 nm (gray and red lines). (A color version of the figure is provided in the online version of the paper).

of quantum algorithms/operations based on spin defect data [11].

An important property of quantum computers is the reversibility of the action of quantum gates. The simplest phenomenon for observing reversibility in spin systems is the electron spin echo [11]. The method consists in recording an electron spin echo when microwave pulses of a given duration are applied with the possibility of inverting the decay processes of spin magnetization due to the inhomogeneous distribution of local magnetic fields. Time boundaries are created in the Hahn sequence with an increase of the inter-pulse interval, within which the loss of spin coherence due to irreversible spin-spin interaction becomes significant. Thus, the lifetime of qubits implemented on spin defects is determined by irreversible spin-spin relaxation processes, in contrast to time-reversible processes (interaction with an inhomogeneous distribution of local magnetic fields) T_2^*. The approximation of these dependencies by the exponential law makes it possible to obtain the phase coherence time of a spin packet, which is one of the important characteristics of any qubit (Figure 2). The polarization of $M_S = 0$ state is associated with quantum transitions under the action of laser radiation from three main levels $M_S = -1,\ 0,\ +1$ to one of the three $M_S = 0$ through several intermediate levels. If we consider a single NV^--center, then such transitions will lead either to a complete restoration of longitudinal magnetization, in the case of excitation through the conduction band, or to a shift in the precession phase due to the nonzero lifetime of states at excited levels, in the case of excitation through the levels of isolated defects. Moving on to the ensemble of spins, on average such processes will cause additional decoherence of the spin package and, consequently, a decrease of the spin-spin relaxation time. Since the number of such transitions is related to the number of optical quanta (optical pumping power) and the energy of one quantum, we conducted a study of the decay of transverse magnetization from laser power and wavelength. In addition, this will have an impact on the probability of such a transition, which depends on the intensity of the absorption line at the laser wavelength associated with NV^--centers. The laser radiation was supplied through the fiber in continuous mode, and pulse width modulation with a period of 10 μs was used to reduce the power. Thus, when the signal was accumulated 8192 times, an average decay of the transverse ensemble magnetization was obtained. Figure 2 shows the relaxation curves of a spin defect under the action of continuous radiation with wavelengths of 980/1064 nm and a power of 125/500 mW. It was found that the effect of the dependence of the radiation frequency is minimal.

Additional measurements were carried out when the optical radiation power was attenuated to 125 mW using pulse width laser modulation. The phase coherence time was doubled with a 4-fold change in the average laser power. This fact indicates that the 125 mW laser power is close to optimal (in Figure 2, green), and irreversible decay of phase coherence is caused by spin-spin relaxation and spin diffusion, which in turn manifests itself as a deviation from the straight line [12]. Under optimal conditions ($T = 150$ K, 980 nm, 125 mW), the transverse relaxation time for an ensemble of particles with $C_N \approx 10^{17}\,\mathrm{cm}^{-3}$ is $T_2 = 60\,\mu$s, which is already more than for previously studied defects at a lower crystal temperature (boron vacancy in *hBN* — 15 μs [4] and divacancies in 4*H*-SiC — 40 μs [5] for $T = 7$ K).

5. Conclusion

Thus, the results obtained in this paper by the photoinduced EPR method show that the NV^--center in the 6*H*-^{28}SiC is a promising platform for the implementation of quantum information technologies based on spin-photon interactions.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by A.Akhtyamov