

Spin Selective Segregation of Oxygen in Dislocation Cores in ^{29}Si Enriched Silicon Crystals

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Abstract

Plastic deformation of Fz— ^{29}Si crystals enriched with ^{29}Si magnetic isotope causes redistribution of ^{29}Si atoms and $^{29}\text{Si}^{16}\text{O}$ complexes in the surface areas of the crystal. Redistribution of the magnetic isotope correlates well with the density of dislocations introduced by plastic deformation. Oxidization of silicon is forbidden in the absence of the magnetic field due to strict prohibition to change full angular momentum conservation in Si—O electronic pair. The presence of the ^{29}Si nuclear spin allows conversion of the initial triplet state to the singlet state in the Si—O pair. This transition accelerates formation of the stable SiO oxide. The single event of the oxidization was interpreted in terms of quantum computation accordingly with the CNOT quantum gate equivalent to the transformation of the triplet state to the singlet one. The read out of the stored quantum information corresponds to “1” in stable SiO complex or logic “0” in the decomposed Si—O pair.

Keywords

Dislocations; isotopes; qubit; quantum logic.

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Introduction

Approaching the sizes of logic devices to atomic size meets quantum effects transferring modern technologies to the area strongly different from classical electronics. Quantum computing based on pure mono isotope silicon crystals [1 – 5] is of interest for researchers investigating spin-dependent phenomena in silicon and methods of pumping of spin polarization [4 – 8]. Although the main advantage of silicon quantum computers is a long coherency time of electron [9, 10] and nuclear [11] spins, this device still requires overcoming multiple difficulties to realize this intriguing idea. Most efforts are spent to propose a method to create topologically isolated ensembles of electronic keys to read nuclear spin states [12], to develop fast optical initialization of spins [13–16] and to organize read out of the quantum information with no disturbance of the information stored in nuclear qubits. In generally, most of the experimental researches ignore the contribution of the crystal surface and structural defects in real crystals. Despite this in

[17 – 19] reading out electronic key was proposed on the basis of chemical reaction between silicon and oxygen dissolved in a crystal lattice. This oxidization reaction and the amount of oxide $^{29}\text{Si}^{16}\text{O}$ on crystal surface can be controlled by external magnetic field as well as hyperfine magnetic field of the ^{29}Si magnetic nuclei [17]. Nuclear magnetic and electron spin resonances are the main methods of selective access to qubits. These methods were applied to study spin dynamics in isotope enriched Si crystals containing dislocations [20 – 22]. Paramagnetic defects generated by deformation have low enough concentration ($< 10^{18} \text{ cm}^{-3}$) satisfying to the conditions of spin coherency, that still remain appropriate for quantum computations even in plastically deformed crystals [18]. An accurate and systematic study of energy spectrum of dislocations and extended defects in semiconductors was realized by Yu. Ossipyan and his team by electron spin resonance, microwave conductivity, photoconductivity and other methods [23]. Fundamental knowledge on electronic properties of dislocations is necessary to resolve the problems of full

silicon quantum computations. A spin state of dislocations controls their mobility in silicon [23], where dangling bonds exist and contribute to dislocation of electrical and optical properties [24–26]. The introduction of linear dislocations populated by dangling covalent bonds allows designing linear chains containing electron spins. The population of the electron spin can be controlled by optical irradiation in dislocation core. Low diffusion coefficient limits the oxidization rate in silicon. Thus, elementary oxidization events are very rare. At the same moment, a very low concentration of the non-stable Si—O pairs is in the sample. Detection of the pairs by the ESR or NMR spectrometer is not possible due to their low concentration. The accumulation of successful oxidizations during a few hours or even days causes the emergence of macroscopic amount of the oxide available for observation by different methods. The secondary ion mass spectroscopy (SIMS) allows identifying final products of the spin controlled reaction as well as distinguishing the presence of different isotopes in the final stage of the magnetically driven processes. In this article, we used the SIMS to probe the silicon isotopes presence in the subsurface silicon crystal layers after the deformation of the crystals. Thus, the spin-controlled oxidization was indirectly detected by the SIMS following the ideas developed by E. Frankevich and A. Buchachenko in spin chemistry. They studied chemical reactions by indirect detection of the spin-dependent processes through electrical current or a reaction yield product. The aims of this research are to test the possibility of using deformation defects (dislocations and $^{29}\text{Si}^{16}\text{O}$ complexes) to create photo-controlled areas of silicon crystals providing read

out quantum information stored in nuclear spin ensembles (qubits), and to provide the optical monitoring of the reaction between the oxygen atom and silicon in the presence of hyperfine interaction with ^{29}Si nuclei.

Experimental

Single crystals of silicon doped with boron ($\sim 10^{13} \text{ cm}^{-3}$) sample No. 1 and sample No. 3 were grown by the floating zone technique and enriched with ^{29}Si isotope. The sample No. 5 of natural isotope concentrations was grown by the Czochralski technique (see Table 1). The samples were $1 \times 4 \times 16 \text{ mm}^3$ sizes, long side was along (110) crystallography orientation. The deformation of the samples was performed by the three point bend until dislocation density $\sim 10^8 \text{ cm}^{-2}$ in the sample center was reached. The deformation temperature was $950 \text{ }^\circ\text{C}$.

The isotope analysis of silicon crystals was performed by the time-of flight secondary ion mass-spectroscopy by spectrometer TOF.SIMS 5 (ION-TOF Germany). The cleaned initial surfaces as well as the surface formed after the ionic etching until the 30 nm depth were also analyzed. The concentrations of isotopes were determined with the high accuracy of $\pm 0.01 \%$. The concentration profiles of three stable Si isotopes ^{28}Si , ^{29}Si , ^{30}Si , as well as $^{29}\text{Si}^{16}\text{O}$, $^{29}\text{SiHO}$, $^{28}\text{Si}^{29}\text{Si}$, $^{29}\text{SiO}_2$, $^{29}\text{Si}_2$, $^{29}\text{Si}_3$ complexes were recorded. The ionic etching was performed by bombardment by Ce^- ions convenient for oxygen complexes revealing. The estimations of the isotopes concentrations are given in Table 1.

Table 1

Parameters of samples

Sample	Active area	Isotope concentrations, %			Deformation
		^{28}Si	^{29}Si	^{30}Si	
Sample No. 1	Surface	0.36	99.58	0.06	Fz— ^{29}Si
	Depth 30 nm	0.045	99.9	0.051	Deformed until $\sim 10^8 \text{ cm}^{-2}$ dislocation density at the central part of sample
Sample No. 3	Surface	2.00	97.89	0.11	Fz— ^{29}Si
	Depth 30 nm	0.19	99.77	0.045	Deformed with low dislocation density ($5 \times 10^5 \text{ cm}^{-2}$)
Sample No. 5	Surface	93.49	3.41	3.10	Cz— ^{28}Si
	Depth 30 nm	93.21	3.79	3.00	Deformed until $\sim 3 \times 10^7 \text{ cm}^{-2}$ dislocation density

Photoluminescence (PL) of the samples placed to helium cryostat was excited by GaAs laser ($\lambda = 920$ nm) and analyzed by the monochromator and cooling Ge photo resistor. At a temperature of 6 K PL the spectra of isotope enriched silicon were compared in crystals with different dislocation density and crystals of natural isotope abundance.

Results and discussion

The main idea of our paper is application of natural chains of paramagnetic centers distributed along dislocations core for creation of the information reading spin keys placed in massive of the nuclear qubits. Dangling bonds along the dislocation core can be labeled by oxygen captured by dislocation paramagnetic centers in the presence of a magnetic field or a hyperfine field. Thus, the silicon-oxygen reaction and the amount of $^{29}\text{Si}^{16}\text{O}$ complexes in the dislocation core control the population of the dangling bonds. The SIMS can distinguish $^{29}\text{Si}^{16}\text{O}$ complexes because Si emission under ionic bombardment depends on the Si environment [11]. Distribution of $^{29}\text{Si}^{16}\text{O}$, $^{29}\text{SiHO}$, $^{29}\text{SiO}_2$, ^{29}Si emission versus the depth of the bombarded layer in the silicon samples are shown in Fig. 1. Deformation of the crystals causes redistribution of the ^{29}Si magnetic isotope as well as oxygen complexes containing this isotope $^{29}\text{Si}^{16}\text{O}$, $^{29}\text{SiHO}$, $^{29}\text{SiO}_2$. This is in good agreement with our previous results manifesting control of the spin-dependent reaction between Si and O by the hyperfine magnetic field of ^{29}Si nuclei [17, 19]. In our previous work, the redistribution of ^{28}Si , ^{29}Si and ^{30}Si isotopes in the surface layers of single Si crystals under plastic deformation was reported [27]. Here, we are going to compare new data on oxide redistribution with the Si atoms depth monitoring. It was shown that oxidation

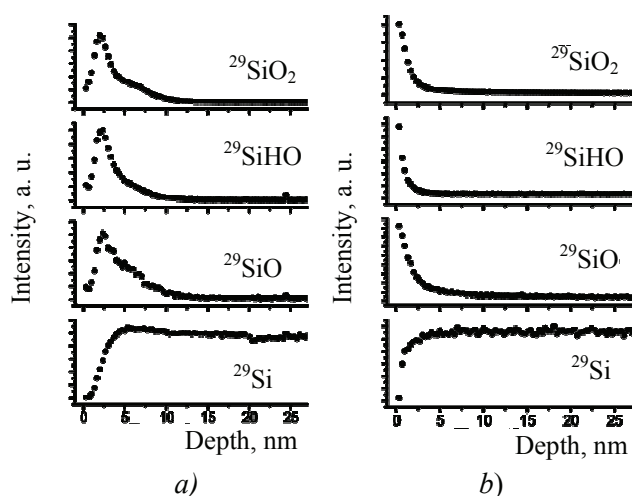


Fig. 1. Profiles of atomic distribution in deformed sample 1 (a) and sample 3 (b) with low dislocation density

of silicon atoms mainly occurred for those of them that contained ^{29}Si magnetic nucleus. In this work we had confirmed the data concerning the deformation induced migration of ^{29}Si atoms involved in the oxygen complexes. In addition, we determined what oxygen complexes are subjected to the influence of the hyperfine magnetic field of ^{29}Si nuclei (Fig. 1). This fact follows from the distributions of the $^{28}\text{Si}^{29}\text{Si}$ complexes different in the deformed Fz— ^{29}Si and Cz— ^{29}Si crystals containing $3 \times 10^7 \text{ cm}^{-2}$ dislocations (Fig. 2). In the deformed Cz— ^{28}Si samples with low magnetic isotope concentration the profile of $^{28}\text{Si}^{29}\text{Si}$ secondary ions was the depth independent in contrast with Fz— ^{29}Si crystals. A similar distribution of ^{28}Si in crystal with different concentrations of dislocations independent on ^{29}Si enrichment (Fig. 3) was also a good confirmation of the ^{29}Si importance in oxidization. The profile of ^{29}Si distribution strongly depends on deformation and presence of ^{29}Si , in contrast with ^{28}Si . Atoms ^{29}Si participate in oxidation twice as fast as the atoms containing nonmagnetic

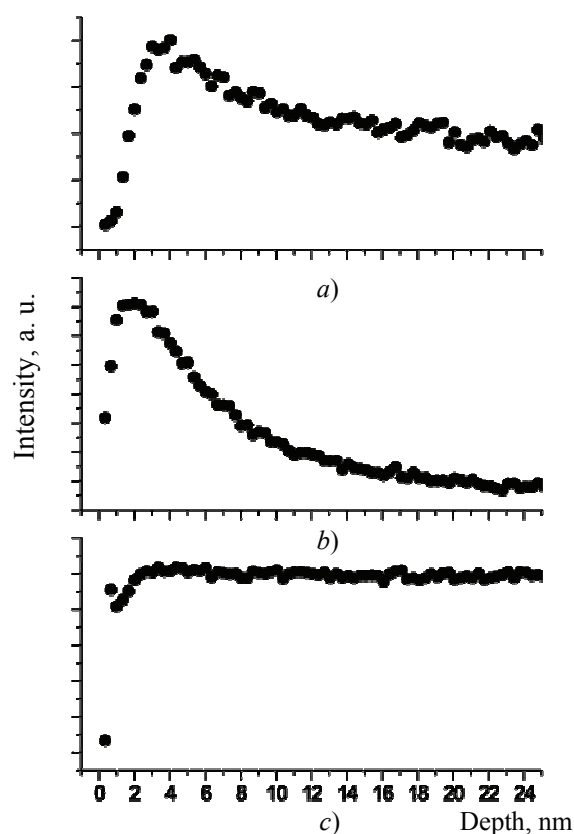


Fig. 2. Profiles of distribution of secondary ions of $^{28}\text{Si}^{29}\text{Si}$ in three types of crystals:

a – in isotope enriched deformed sample 1;

b – in isotope enriched sample 3

(with low dislocation density ($5 \times 10^5 \text{ cm}^{-2}$));

c – deformed sample 5 with natural abundance of isotopes

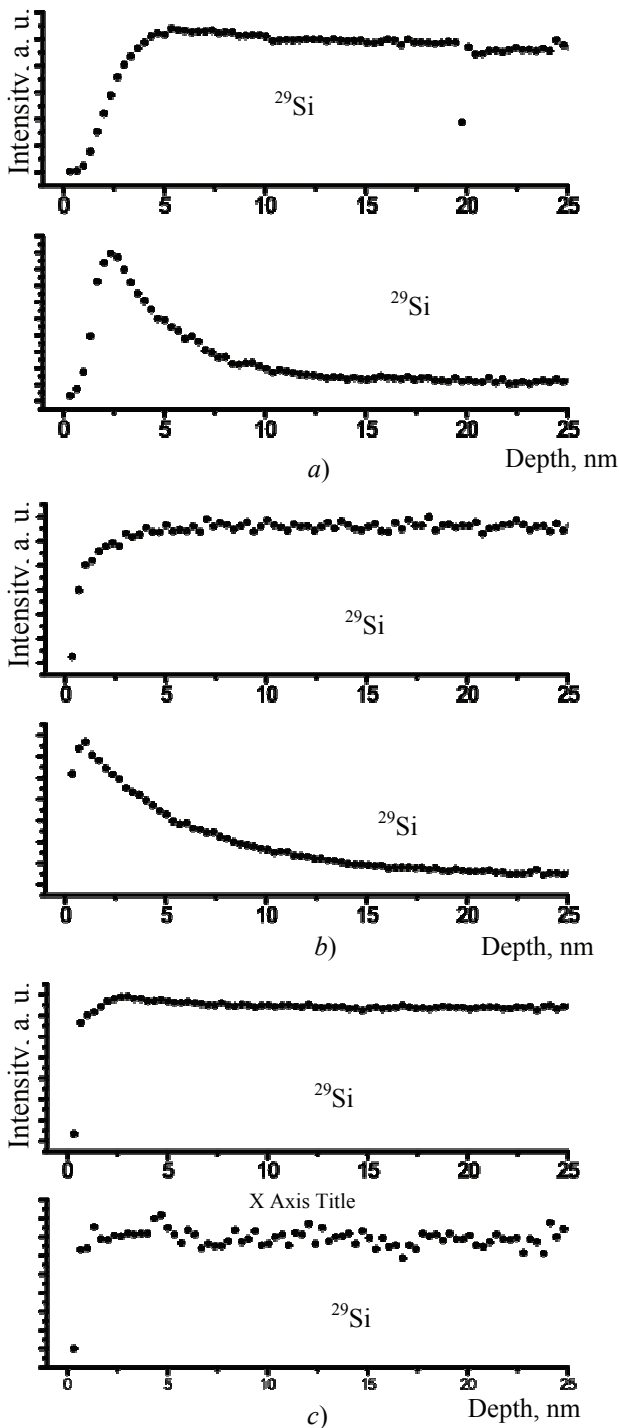


Fig. 3. Distributions of magnetic ^{29}Si and non magnetic ^{28}Si isotopes in three types of crystals:
a – in isotope enriched deformed sample 1;
b – with low dislocation density sample 3;
c – deformed sample 5 with natural abundance of isotopes

nuclei ^{28}Si , ^{30}Si due to the hyperfine interaction [17, 19]. One can conclude that the change of the elastic constants and the dilatation of the crystal lattice due to the change of the isotope nuclei size can be a reason of the described above effects, because ^{28}Si , ^{30}Si

behaviors are similar, although both of them are different from ^{29}Si .

Oxygen complexes are non-equilibrium structural defects migrating to the dislocation core in the energy profile of the distorted crystal lattice. This migration minimizes free energy of the crystal. The analysis of photoluminescence of dislocation levels $D_1 - D_4$ allows monitoring of the spin dependent processes involving magnetic nuclei and reading paramagnetic keys. For this purpose, low temperature measurements of photoluminescence were carried out in the crystals (Fig. 4). The intensities of the $D_1 - D_4$ dislocation luminescence bands were strongly different due to different dislocation densities in the studied samples. An increase in the dislocation density leads to the enhancement of the luminescence intensity as well as to the appearance of the long wave shoulder at D_1 line caused by the oxygen segregation in the dislocation cores during their movement through the crystal [28]. In [20, 21], a correlation between temperature dependencies of the D_1 line intensity and the magnetic moment and the magnetic susceptibility of the plastically deformed crystals was revealed. This fact allows us to verify the interrelation between the centers responsible for D_1 luminescence and paramagnetic structural defects in the isotope enriched $^{29}\text{Si}:\text{B}$ crystals. It seems to be reasonable to suppose that the early observed magnetic and microwave magnetic field effect on the dislocation mobility [23] has a direct relation to the elementary processes reported in this article. The effect of the external and/or hyperfine magnetic fields on the oxidization of silicon in the areas close to the dislocation cores is able to change the structure and the potential relief of the dislocation obstacles. Thus, the change of the dislocation mobility

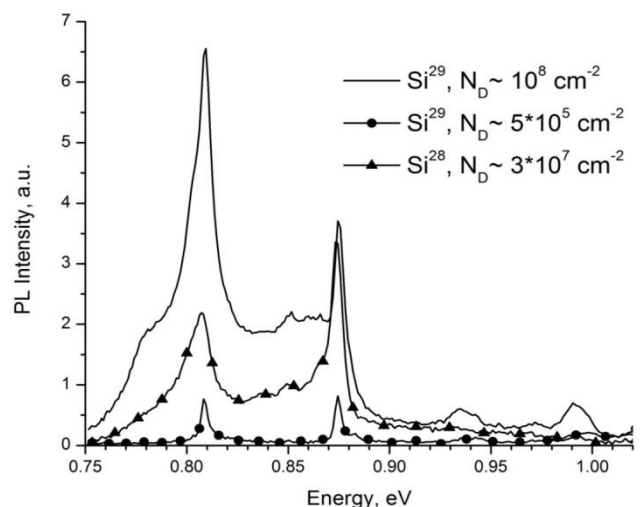


Fig. 4. Spectra of dislocation photoluminescence at 6 K in the crystals with different dislocation densities (see legend)

under the magnetic field can be judged as an indicator of spin-dependent processes stimulated in the dislocation core. Direct experimental evidence of this fact was proposed in [30].

The calculation of probability of different states in three spin system ($S_1 = \frac{1}{2}$ spin of O, $S_2 = \frac{1}{2}$ spin of Si, $I = \frac{1}{2}$ spin of Si nucleus) was performed by spin composer instrument, proposed by IBM [31]. The superconductive quantum computer operating at 15.736 mK performed the calculation of probabilities of the 3-qubit states (results see in Fig. 5, 6). Eight eigenstates of the three spin system are $|000\rangle$, $|001\rangle$, $|010\rangle$, $|011\rangle$, $|100\rangle$, $|101\rangle$, $|110\rangle$, $|111\rangle$ and probabilities of their mixing during classical random score are shown in Fig. 5 (Bloch sphere) and Fig. 6 (population of the states). One can estimate a very small fraction of $|100\rangle$ state due to the saving of the system in the initial $|000\rangle$ state. A small admixture of the $|100\rangle$ state indicates a negligible role of the decoherency as it takes place in the silicon crystal lattice.

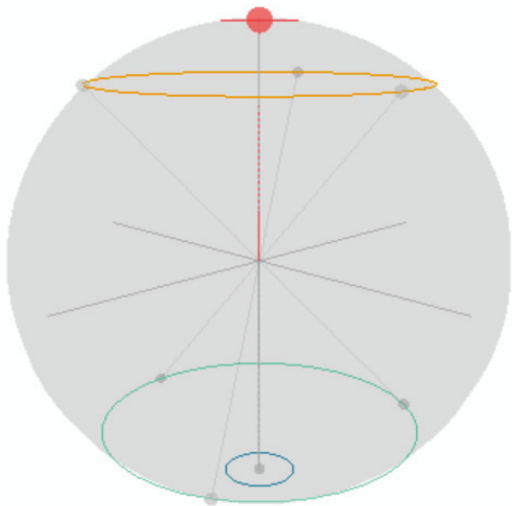


Fig. 5 Results of realization of random score on Bloch sphere with eigenstates marked by points

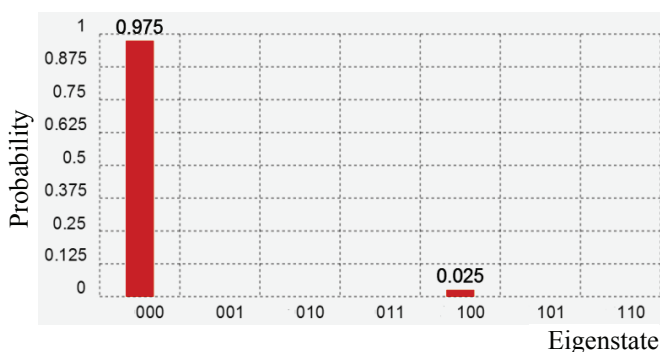


Fig. 6. Results of evaluation of real quantum IBM computer for 3 spin system

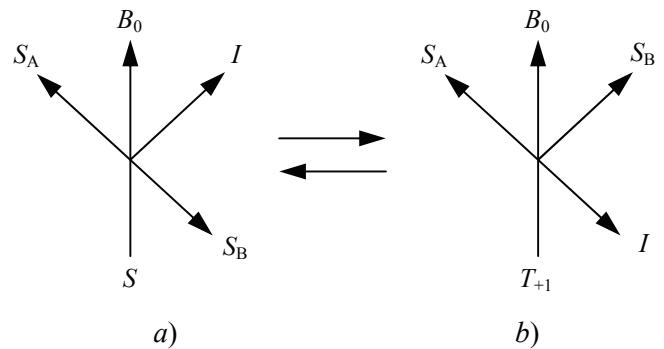


Fig. 7. Scheme of spin-dependent formation of $^{29}\text{Si}^{16}\text{O}$:
a – singlet state; b – triplet state

The conventional gate providing transformation of the two electron spin system from a singlet state (Fig. 7a) to a triplet state (Fig. 7b) is the Controlled-NOT, or CNOT. Nuclear spin controls NOT transition. The examples of this transformation are given in [31].

Thus, the photo controlled electronic states of dislocations and other deformation defects allows one to dissolve the main problem of quantum computations, i.e. the suppression of the spin decoherency resulting from the electron–nuclear interaction in the introduced spin “key” by doping atoms. The possibility of the rapid filling of electronic levels on dislocation under the light emission allows creating an optical shutter keeping a nuclear subsystem isolated from thermal fluctuations. This method provides rapid access to the stored information. Thus, a combination of a long storage of the nuclear spin polarization with the photo controlled key based on the optically controlled deformation defect gives advantages in quantum computing due to the combination of microwave readout and the optical population/depopulation of the keys opening for a short time window.

Conclusions

The formation of $^{29}\text{Si}^{16}\text{O}$ complexes passed through a spin-selective stage. The distribution of these complexes in the near surface layers was determined by the SIMS. In the ^{29}Si enriched crystals the amount of the deformation defects was larger in comparison with the crystals of natural isotope abundance. The dislocation movement caused the redistribution of the isotopes contained in the subsurface layers. The variation of the isotope concentration under the plastic deformation correlated well with the dislocation density.

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