Diffusion of Vacancies in 4H-SiC

(Dated: March 31, 2017)

Abstract

Defect centers in silicon carbide (SiC) have emerged as strong contenders in the quest to realize quantum devices due to the material's lower cost as compared to its counterpart diamond, and due to the micro-fabrication techniques now available and favorable optical emission wavelengths and spin properties. We investigate the diffusion of negatively charged silicon vacancies in 4H-SiC, using density-functional-theory and kinetic Monte Carlo simulations, which should serve as an invaluable guide in controlling defect position within devices.

I. INTRODUCTION

The promise of the nitrogen-vacany (NV) center in diamond as a system for implementing memory storage for a quantum computer has spawned interest in defect centers in related materials such as SiC. SiC is particularly interesting as it is a polymorphic material, exhibiting about 250 known polytypes, which imbues it with a degree of freedom unavailable in diamond. The three most common polytypes, 4H- and 6H-SiC and 3C-SiC, all have spin relaxation times ranging from 8 to 24 ms at 20 K (with 4H-SiC being the highest) and coherence persists up to room temperature[1]. In addition to the long spin coherence, a key feature is the ability to optically address (write in and read out) the spin states. However, much of the luminescence or emission of the defects is diverted into transitions involving scattering processes (and is not purely from the desired spin transitions) at ambient temperatures. Indeed, only about 4% of luminescence is from the desired transitions[2]. A potential solution is to place the defects near cavities on resonance with the desired transitions[3]. Positioning defects, however, is a non-trivial endeavor. The defects would be created at roughly the desired location using the process of focused ion beam implantation, but this process creates a lot of damage. In order to heal the damage the sample is annealed, causing the defects to diffuse and some to be lost through conversion to other species. The purpose of this study is then to assess the probability that the negatively charged silicon vacancy defect in 4H-SiC would be optimally positioned and exist given a certain initial position and a certain number of time steps.

II. COMPUTATIONAL METHODS

There are two main stages to the calculation of the trajectory probability maps. In the first stage, density functional theory calculations will be carried out to obtain the barriers to diffusion for the various pathways and in the second stage kinetic Monte Carlo simulations will be carried out to map out trajectories. We have existing code for the second part in Matlab, which will be converted to C code. The kinetic Monte Carlo simulations will be carried out both with Coulomb interaction and for 1D and 2D random walk test cases (to be compared with theoretical calculations). Without the Coulomb interaction the probability map will be calculated using a breadth first search down the tree of possible transitions (which is O(log(N)), where N is the number of possible pathways). The theoretical calculation of the probability distribution map in space as a function of the number of time steps can be obtained as follows for the 1D and 2D cases.

For the 1D case, we let n_1 be the number of steps to the right and n_2 be the number of steps to the left. The final position (with rightwards being positive) is then given by $d = n_1 - n_2$ and the total

number of steps is $N = n_1 + n_2$. We know that the probability of taking n_1 steps to the right out of a total of N steps is simply given by the number of ways to permute all the steps divided by the product of the number of ways to permute the right steps and the left steps and multiplied by the probability of taking a right step to the power of the number of right steps and the probability of taking a left step to the power of the number of left steps, so $p_N(n_1) = \left(\frac{1}{2}\right)^N \frac{N!}{n_1!n_2!} = \left(\frac{1}{2}\right)^N \frac{N!}{n_1!(N-n_1)!} = \begin{pmatrix} N \\ n_1 \end{pmatrix}$, where we have assumed the probabilities of taking left and right steps are equal. We can uniquely express n_1 and n_2 in terms of d and N by solving the system of two equations, and in doing so we obtain $n_1 = (N+d)/2$, $n_2 = (N-d)/2$. Thus the probability of ending up at a position d after N total steps is simply $P_N(d) = \begin{pmatrix} N \\ (N+d)/2 \end{pmatrix}$.

For the 2D case, we let n_1 be the number of steps to the right, n_2 be the number of steps to the left, n_3 be the number of steps to the upwards and n_4 be the number of steps to the downwards. The probability of taking n_1 steps right, n_2 steps left, n_3 steps up and n_4 steps down is then $p_N(n_1, n_2, n_3, n_4) = \left(\frac{1}{4}\right)^N \frac{N!}{n_1!n_2!n_3!n_4!}$, where $N = n_1 + n_2 + n_3 + n_4$. If we now let $d_x = n_1 - n_2$ be the horizontal position and $d_y = n_3 - n_4$ be the vertical position, then we can solve for n_1, n_2 and n_3 in terms of d_x, d_y, N and n_4 to find,

$$n_1 = (N + d_x - d_y - 2n_4)/2 \tag{1}$$

$$n_2 = (N - d_x - d_y - 2n_4)/2 \tag{2}$$

$$n_3 = d_y + n_4. \tag{3}$$

We note that n_4 can range from 0 to $(N - d_x - d_y)/2$ so,

$$P_N(d_x, d_y) = \sum_{n_4=0}^{(N-d_x-d_y)/2} \left(\frac{1}{4}\right)^N \frac{N!}{((N + d_x - d_y - 2n_4)/2)!((N - d_x - d_y - 2n_4)/2)!(d_y + n_4)!n_4!} \tag{4}$$

$$= \left(\frac{1}{4}\right)^N \frac{\Gamma(N+1)\,{}_2\tilde{F}_1((d_y - d_x - N)/2, (d_x + d_y - N)/2; 1 + d_y; 1)}{\Gamma((N + 2 - d_x - d_y)/2)\Gamma((N + 2 + d_x - d_y)/2)}, \tag{5}$$

where Γ is the gamma function and ${}_2\tilde{F}_1$ is the regularized version of the hypergeometric function ${}_2F_1$.

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