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**Citation for published version (APA):**

Smakman, E. P., Bree, van, J., & Koenraad, P. M. (2013). Laser and voltage manipulation of bistable Si dopants in the GaAs (110) surface. *Physical Review B*, 87(8), Article 085414.  
<https://doi.org/10.1103/PhysRevB.87.085414>

**DOI:**

[10.1103/PhysRevB.87.085414](https://doi.org/10.1103/PhysRevB.87.085414)

**Document status and date:**

Published: 01/01/2013

**Document Version:**

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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# Laser and voltage manipulation of bistable Si dopants in the GaAs (110) surface

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(Received 19 October 2012; revised manuscript received 21 January 2013; published 8 February 2013)

Bistable behavior of single Si dopants in the (110) surface layer of GaAs was studied with a scanning tunneling microscope (STM). The Si atom acts as either a positively charged substitutional donor or a negatively charged interstitial. Its configuration can switch under the influence of a local biased STM tip. To independently manipulate the charge state, the sample was illuminated by a laser during STM operation. The Si atom can be reversibly switched between its positive and negative charge states by turning the laser on and off, respectively. This process occurs mostly with the photon energy tuned above the band gap of GaAs, indicating that photogenerated electron-hole pairs play an important role in the process. The occupation of the donor and interstitial configurations depends on the carrier dynamics, i.e., the possibility of the electrons to escape or to be captured. If the tip-induced band bending is large enough, it is possible for electrons to tunnel into the conduction band and the donor configuration is observed. Another escape path is created when the sample is illuminated and photogenerated holes can recombine with the bound electrons of the dopant.

DOI: [10.1103/PhysRevB.87.085414](https://doi.org/10.1103/PhysRevB.87.085414)

PACS number(s): 73.20.Hb, 68.37.Ef, 66.35.+a, 78.67.-n

## I. INTRODUCTION

In semiconductor devices, dopant atoms are important for the operation because of the free charge carriers they add to the material. With the scaling down of these structures, properties of individual dopant atoms start to play a role in their functionality. The influence of a single dopant atom on its surrounding environment can be studied with scanning tunneling microscopy (STM). Examples are studies of the deep acceptor Mn,<sup>1,2</sup> the shallow donor Si,<sup>3,4</sup> or the shallow acceptor Si<sup>5,6</sup> in the surface of GaAs. A more complete understanding of the fundamental properties of dopants will assist in the development of so-called solotronic or single dopant electronic devices.<sup>7</sup> In this paper we discuss laser and voltage manipulation of bistable Si dopants in the GaAs (110) surface.

An STM simultaneously collects information about the topography and the electronic properties of a surface, which allows for imaging the charge state of single dopant atoms. Here we focus on Si in GaAs, which is a donor when located on a Ga site and an acceptor when on the position of an As atom. A bright contrast in the STM images corresponds to the Coulomb field of a positively charged Si donor.<sup>8</sup> Ionization of the shallow donor state occurs at positive sample bias voltages, due to the local upward tip-induced band bending (TIBB).<sup>9</sup> Similarly, at these tunneling conditions, Si acceptors in GaAs appear dark because they are negatively charged.

Si atoms in the GaAs (110) surface layer show bistable behavior in STM measurements.<sup>10</sup> The exact depth of Si with respect to the GaAs surface can be determined by topographic height measurements. It was shown that only and all the Si atoms in the topmost surface layer are bistable at low temperature, and below this layer the behavior is not observed. During scanning of the STM tip, the Si can switch between a dark and a bright contrast. Examples of switching Si atoms can be seen in Fig. 1. In the presented STM images, the fast scan direction of the tip is left to right and the slow scan direction is bottom to top. In Fig. 1(a) Si appears with the dark contrast of a negatively charged dopant at relatively low positive sample bias voltage. In Fig. 1(c) a higher voltage is applied, and now a positive charge state is visible. There is an intermediate critical

voltage at which the Si configuration is bistable; see Figs. 1(b) and 1(d). The switching between positive and negative charge occurs within one STM image at random positions during scanning. The exact value of this critical voltage is affected by the tip work function, which controls the amount of TIBB at the surface of the semiconductor.<sup>11</sup> In addition, there is a slight variation between various bistable Si in the surface layer, due to the random distribution of other charged dopants in their neighborhood, leading to a different electrostatic environment.

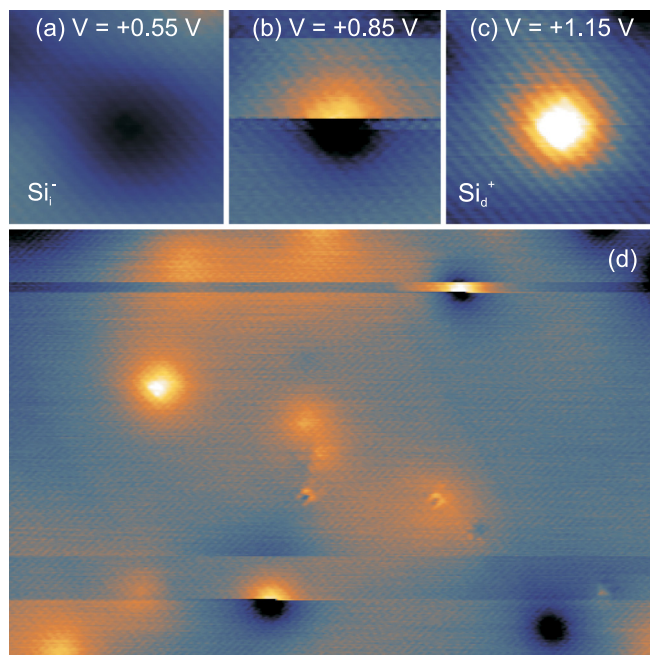


FIG. 1. (Color online) (a)–(c) Example of a bistable Si atom in the GaAs (110) surface, STM topography of  $10 \times 10 \text{ nm}^2$  taken at  $I = 100 \text{ pA}$ . (a) Negatively charged interstitial state  $\text{Si}_i^-$ . (b) Bistable behavior during scanning. (c) Positively charged donor state  $\text{Si}_d^+$ . (d) Two bistable Si atoms surrounded by Si donors (bright) and acceptors (dark). STM topography of  $48 \times 36 \text{ nm}^2$  taken at  $V = +0.75 \text{ V}$  and  $I = 50 \text{ pA}$ .

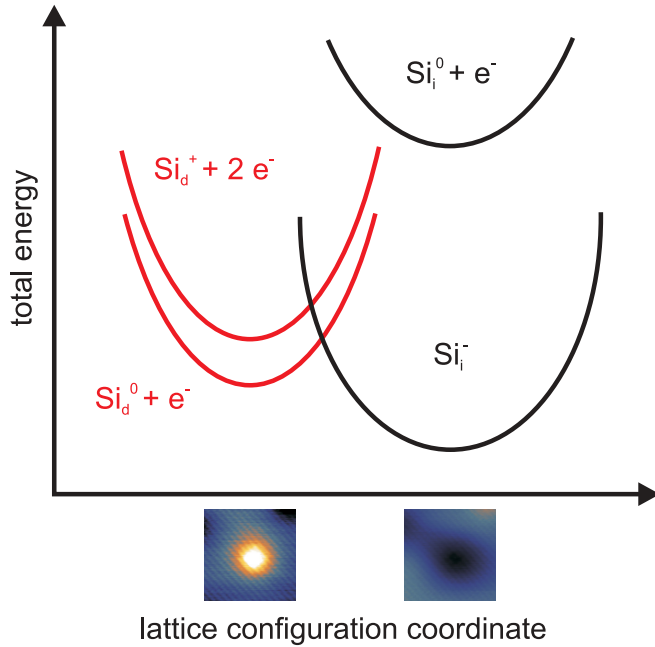


FIG. 2. (Color online) The lattice coordinate energy diagram of the Si atom in the (110) surface layer of GaAs; see the text for explanation.

The STM observations show many similarities to the behavior of the  $DX^-$  center in bulk semiconductors. Here dopants also appear in bistable configurations, which can be related to different lattice positions.<sup>12,13</sup> In the case of the  $DX^-$  center in bulk GaAs under hydrostatic pressure, the negative charge state involves the breaking of one bond between the Si and an As neighbor, combined with a lattice relaxation of the dopant to an interstitial position and a more localized charge distribution compared to the donor configuration. Recent DFT calculations revealed that indeed the Si atom in the GaAs (110) surface can occupy similar bistable configurations,<sup>14</sup> but without any broken bonds. It was shown that a negatively charged Si atom on an interstitial position in the lattice has a total energy of  $\sim 500$  meV lower compared to when it sits on a substitutional donor position in a neutral charge state.

A qualitative lattice coordinate energy diagram of the Si atom in the (110) surface layer of GaAs is shown in Fig. 2. The total energy of the Si is depicted for two different lattice coordinates.<sup>12,14</sup> Also, different charge states are represented. Extra electrons are added in the diagram for a correct energy comparison between the different configurations. On the left, the substitutional donor configuration is indicated in red. There is the neutral charge state plus one electron in the bulk  $Si_d^0$ , and the positive charge state plus two electrons in the bulk  $Si_d^+$ . The energy difference between the neutral and positive donor state can be overcome by the presence of the STM tip through upward TIBB, leading to clear rings of ionization appearing in the STM images.<sup>9</sup> At the other lattice coordinate, the interstitial configuration exists, indicated in black, which is either neutral plus one electron in the bulk  $Si_i^0$  or negatively charged  $Si_i^-$ . The neutral interstitial configuration has not been observed by STM measurements. Important here is that in order to switch between the positive and negative charge states

observed in the measurements, an energy barrier needs to be overcome in either direction.

## II. EXPERIMENTAL

Experiments were performed in an Omicron LT-STM operated at a temperature of 5 K with a base pressure of  $p \leq 2 \times 10^{-11}$  mbar. Electrochemically etched tips made from polycrystalline tungsten wire were used. Further tip preparation was done under ultrahigh vacuum (UHV) conditions, including heating and argon sputtering for improved stability.<sup>15</sup> We studied commercially grown GaAs wafers  $n$ -doped with Si with a concentration of  $\sim 2 \times 10^{18}$   $\text{cm}^{-2}$ . The samples were cleaved in UHV to obtain atomically flat and clean (110) surfaces.

## III. RESULTS AND DISCUSSION

To study the behavior of a Si atom in time, the STM tip is restricted to scan over a fixed single line; see Fig. 3. These repetitive line scans track the charge state and record any switching event on or around the Si atom. This not only allows the determination of switching dynamics in time, but also enables a systematic way to study the variation of tip parameters like bias voltage and tunneling current on a single dopant. Moreover, an optical experiment is possible in which a laser illuminates the sample surface. The laser is turned on or off and the effects on the Si atom are recorded directly as a function of time by the scanning STM tip.

In the STM setup, a  $\lambda = 632$  nm diode laser was focused to an  $\sim 0.1$  mm radius spot on the sample and tip by means of an *in situ* lens in the STM.<sup>16</sup> The effect of illumination with photon energy  $U_p = 1.96$  eV on the configuration of the Si atom is shown in Fig. 4(a). For clarity, only 2 nm of the scanned line on top of the Si atom is depicted as a function of time. The rest of the 60-nm line scan is used to appropriately level the contrast of the Si atom against the GaAs surface. This means the dark and bright contrasts here represent the negative and positive charge states, respectively. The tunneling conditions are chosen such that  $Si_i^-$  is favored when the laser is off. In this case, the bias voltage  $V = +0.50$  V, resulting in a near absence of spontaneous switching to  $Si_d^+$ . When the laser is turned on, the Si atom immediately switches to the bright

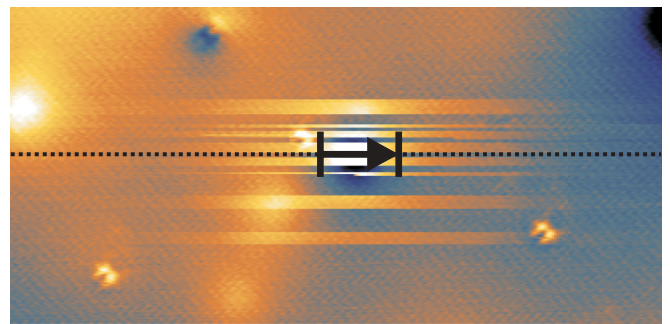


FIG. 3. (Color online) Illustration of the line restriction (dotted line) applied during measurements. Only part of the line scan directly on top of the bistable Si atom (bracketed arrow) is shown in Figs. 4 and 6. STM topography of  $40 \times 20$   $\text{nm}^2$  taken at  $V = +0.75$  V and  $I = 50$  pA.

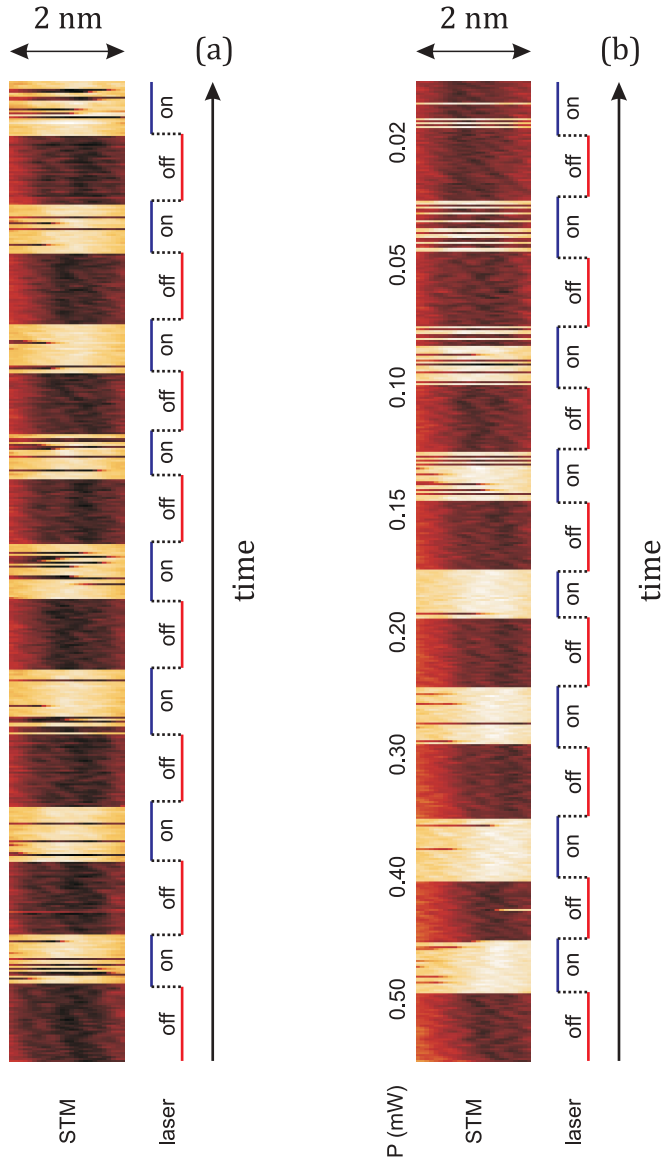


FIG. 4. (Color online) (a) Part of a 12-min-long STM line scan recording the charge state of a Si atom in the GaAs surface. The bias voltage  $V = +0.50$  V, which favors the dark  $\text{Si}_i^-$  if the laser is off.  $I = 50$  pA,  $P = 0.25$  mW, and  $U_p = 1.96$  eV in this experiment. Illumination results in the bright  $\text{Si}_d^+$  being the predominant state. (b) A similar experiment in which the laser power is varied, showing a gradual decrease of the bright state occupation with lower laser power.

state. During illumination, switching back to the dark state occurs sometimes at random positions, but this effect is rare. The effect of varying laser power  $P$  is shown in Fig. 4(b). Lower power gradually leads to less occupation of  $\text{Si}_d^+$ , with the lowest power settings matching almost full occupation of  $\text{Si}_i^-$  when the laser is off.

To understand the physical mechanism behind the observed behavior, we can view the tip-sample system in terms of energy. To incorporate the bound electrons qualitatively in the model, two different energy levels should be distinguished: The substitutional donor state at the surface where the binding energy is  $\sim 40$  meV<sup>17</sup> and the interstitial state located

$\sim 500$  meV below the conduction band minimum.<sup>14,18</sup> Note that both energies differ from the value of 5.6 meV for a Si shallow donor in bulk GaAs. From this, the influence of the tip on the occupation of both states can be understood; see Figs. 5(a) and 5(b). If the TIBB is large enough, the bound electrons from  $\text{Si}_i^-$  can tunnel into the conduction band of the sample, making it possible to switch to  $\text{Si}_d^+$ . However, if the band bending is small, the electrons cannot escape and  $\text{Si}_i^-$  is the favorable configuration.

The effect of illumination can be considered for a system of metallic tip and semiconductor sample.<sup>19</sup> If the laser is tuned above the band gap of GaAs, electrons and holes are created that result in a photocurrent and a photovoltage. The photocurrent is in the same direction as the tunneling current between tip and sample, so the STM feedback in constant current mode retracts the tip slightly for compensation. The photovoltage counteracts the applied bias voltage. Both effects result in a decrease of the TIBB. To estimate if these effects are relevant in our experiments, we studied the size of donor Si rings of ionization as a function of applied laser power.<sup>9</sup> There was no visible change in ring size with the laser power up to four times higher than the values used in the rest of the experiments, meaning the change in TIBB due to illumination is negligible. Furthermore, a decrease of TIBB cannot explain the observed occupation of the bright state being due to illumination, because this situation would favor the dark state.

An explanation for the observed illumination effect on switching can be found in electron-hole recombination, which is illustrated in Figs. 5(c) and 5(d). Part of the holes that are created by photon absorption accumulate in the region below the tip, because of the local upward TIBB there. Electrons from  $\text{Si}_i^-$  can now recombine with the available holes, creating a path for the electrons to effectively escape the Si atom.

The immediate switching to  $\text{Si}_d^+$  both at high bias voltage and at high laser power suggest that under these conditions the escape of electrons from the dopant atom is a relatively fast process. The relevant timescale is then faster than or in the order of the sample frequency of the STM, which is 1 kHz, because we observe only few dark pixels under these conditions. In the case of an applied bias voltage, the TIBB is the parameter that determines the chance of quantum tunneling for electrons into the conduction band. Under illumination and low TIBB, the photoexcited hole concentration determines the recombination rate.

Switching to  $\text{Si}_i^-$  is strongly dependent on the tunneling current and reaches a frequency of about 1 Hz for  $I = 250$  pA.<sup>10</sup> The switching depends on the availability of electrons, which is favored by a high current just below the tip. At the same time, local TIBB repels electrons, which is also highest just below the tip. This means there is a finite distance away from the center of the Si atom, where the switching to the dark state is most efficient. Another consequence of this is that for certain tips the switching to  $\text{Si}_i^-$  is never possible while the tip is near the dopant. This is a reason why the length of the line traces in the measurements presented here are typically 60 nm, preventing any proximity effect and allowing the Si atom to relax back into its negative charge state with the tip far enough away. We also suspect that ultrasharp tips with an apex of only a few nanometers are required to observe the switching behavior. More blunt

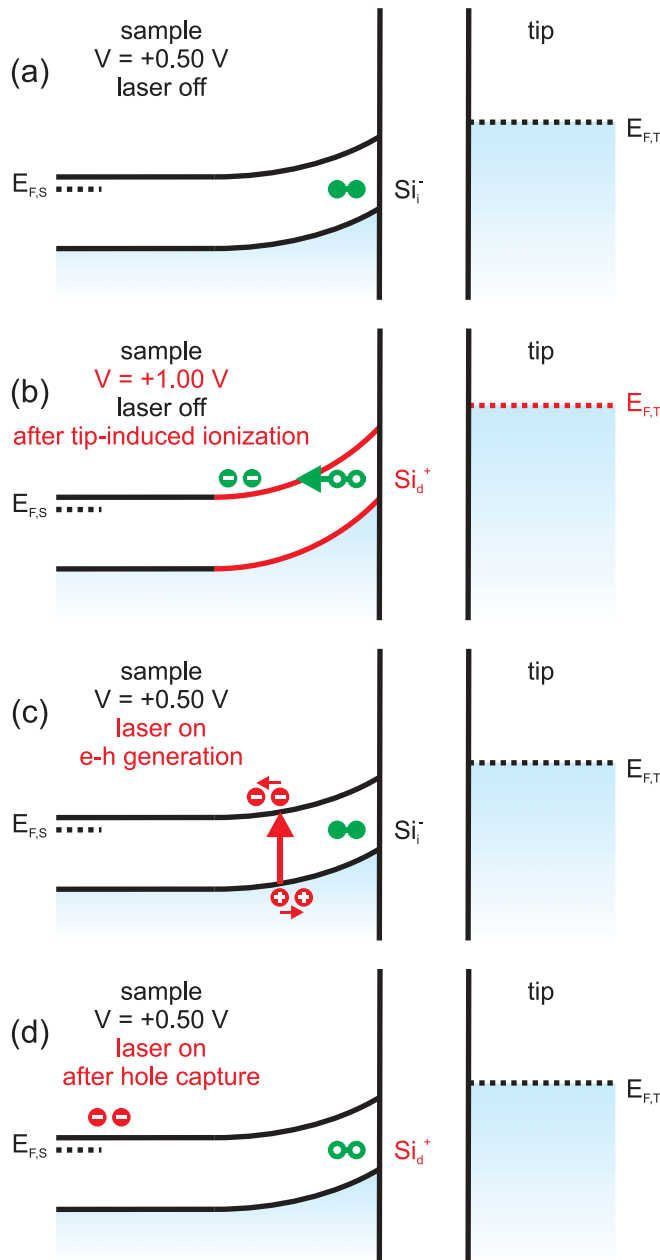


FIG. 5. (Color online) (a) The tip-sample band image without illumination. The interstitial level is indicated by the solid green line, with two electrons occupying this level. At these tunneling conditions,  $\text{Si}_i^-$  is favored. (b) When the TIBB is increased the electrons can escape by tunneling into the conduction band. (c) The effects of illumination: Electrons and holes are created. The holes accumulate in the region of TIBB. (d) When the holes recombine with the bound electrons on the Si, the atom switches to  $\text{Si}_d^+$ .

tips already influence the Si configuration at a large distance, making it impossible to record any change in the charge state.

To confirm that the switching indeed depends on holes originating from absorbed photons, the sample was again illuminated with a laser, but now with varying photon energy. This was accomplished by using a tunable Ti:sapphire laser, enabling a wavelength range of  $\lambda = 780\text{--}920\text{ nm}$ . If the bright state occupation depends on the availability of holes in the

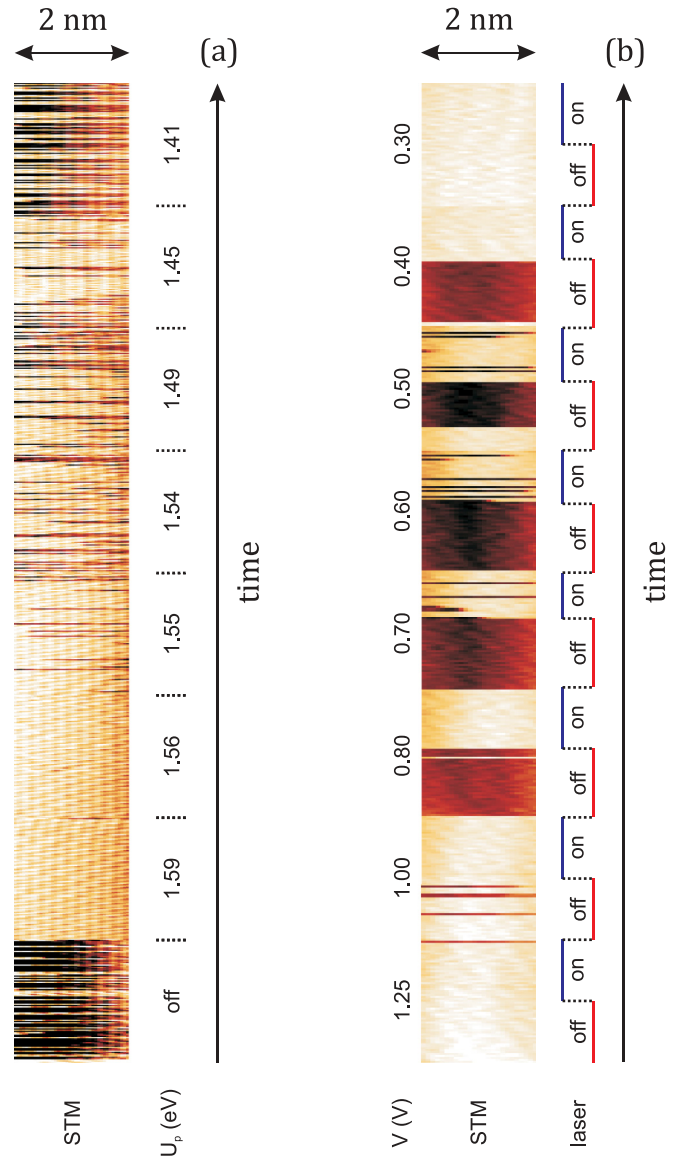


FIG. 6. (Color online) (a) Part of a 12-min-long STM line scan recording the charge state of a Si atom in the GaAs surface showing the effect of varying photon energy, with  $V = +0.65\text{ V}$ ,  $I = 100\text{ pA}$ , and  $P = 0.20\text{ mW}$ . (b) A similar scan in which the bias voltage is varied.  $I = 50\text{ pA}$ ,  $P = 0.25\text{ mW}$ , and the photon energy is fixed at  $U_p = 1.96\text{ eV}$ , well above the band gap of GaAs.

surface region, tuning the photon energy below the band gap of GaAs should stop this effect. In this experiment, the results of which are shown in Fig. 6(a), the laser power was kept constant at  $P = 0.25\text{ mW}$  and the photon energy was varied from  $U_p = 1.41$  to  $1.59\text{ eV}$ . The band gap of GaAs at  $T = 5\text{ K}$  is  $U_g = 1.52\text{ eV}$ , so this range investigates the effects of illumination below and above the band gap.

At photon energies above the band gap, an almost full occupation of the bright state is visible, corresponding to the measurements of that in Fig. 4(a). However, at subsequent lower photon energies below the band gap, the bright state occupation becomes less until it reaches the laser off level at  $U_p = 1.41\text{ eV}$ . This supports the idea that electron-hole

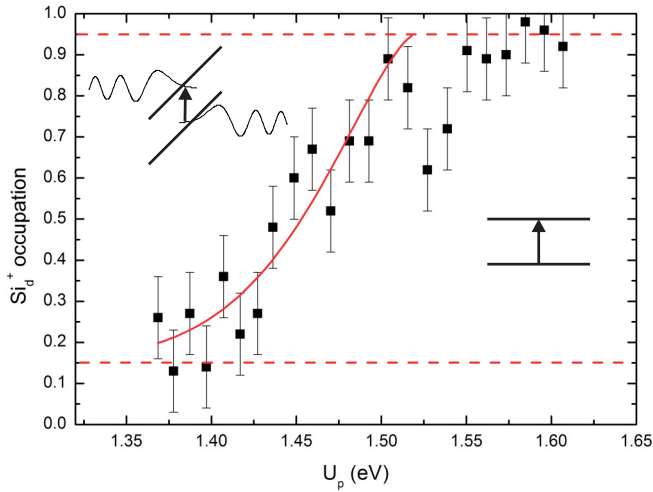


FIG. 7. (Color online) The occupation of  $\text{Si}_d^+$  as function of photon energy. The dotted red lines indicate levels of 15% and 95% for the dark to bright spontaneous switching and the reverse process, respectively. The solid red line is a fit of below band gap photon absorption in a semiconductor under influence of an electric field using Eq. (1).  $V = +0.50$  V and  $I = 250$  pA for all data points. The two schematics illustrate the photon absorption in a semiconductor with and without an added electric field, on the left and right, respectively.

pairs created during illumination play an important role in the dynamics of the bistable Si atom.

In Fig. 7 the occupation of  $\text{Si}_d^+$  is shown as a function of photon energy. Each data point in the graph represents the statistics of 12 minutes under the same illumination and tunneling conditions. It reveals a gradual decrease of the occupation with the photon energy tuned further below the band gap of GaAs. It does not fall directly to zero below the band gap, and we propose that this effect can be explained by local TIBB. With above band gap excitation, holes are created everywhere in the illumination area of the sample. Diffusion then brings the holes near the region of the TIBB, where they can recombine with the bound electrons of the Si atom. When the laser is tuned below the band gap, only in the region with enough TIBB can photons still be absorbed. This is due to the electric fields from the tip, creating a strong local Franz-Keldysh effect. An electron can be photoexcited from the valence band that is shifted up, to the conduction band with a smaller shift slightly farther away from the tip, where the electron and hole wave functions still overlap. The created hole is then available for recombination with electrons on the Si atom.

The photon absorption  $\alpha$  in a semiconductor under influence of an electric field is described by an Airy function. The below band gap part is of the form<sup>20</sup>

$$\alpha \sim e^{-(8\pi\sqrt{2m_{eh}^*})/(3heE_s)(U_g - U_p)^{3/2}}, \quad (1)$$

where  $m_{eh}^*$  is the exciton effective mass,  $e$  the electron charge, and  $E_s$  the electric field at the surface induced by the STM tip.  $E_s = \Phi_{\text{TIBB}}/(ew)$ , where  $\Phi_{\text{TIBB}}$  is the TIBB potential energy at the surface and  $w$  is the depletion length in the semiconductor. The depletion length can be estimated using

a typical work function for W of  $\Phi_{\text{WF}} = 4.50$  eV,<sup>11</sup> which results in  $w = 19$  nm.<sup>21</sup>

Fitting Eq. (1) to the occupation of  $\text{Si}_d^+$  in Fig. 7 yields  $\Phi_{\text{TIBB}} = 0.7 \pm 0.4$  eV. The fit follows the data quite well, and the obtained value for  $\Phi_{\text{TIBB}}$  is in the expected order of magnitude.<sup>11,21</sup> The extraction of the value for the TIBB through this optical method might prove to be more exact than when using only electrical information obtained with the STM.

In another experiment, the sample bias voltage was varied with the photon energy fixed above the band gap of GaAs; see Fig. 6(b). For every voltage, the laser was turned on and off to study the influence on the Si configuration. For relatively high voltage, e.g.,  $V = +1.00$  V,  $\text{Si}_d^+$  is predominant, regardless of the presence of illumination. This is because the sample is biased above the critical voltage at which the electrons from the interstitial level can tunnel into the conduction band through upward TIBB; see Fig. 5(b). For intermediate positive  $V$ , the dark state is favorable when the laser is off, as is shown also in Fig. 4. Illumination creates holes that allow the electrons from  $\text{Si}_i^-$  to recombine and escape the atom. For low bias voltages, e.g.,  $V = +0.30$  V, Si can be put in the bright state by turning on the laser, but it does not switch back to the dark state when the laser is turned off. This can be understood by considering the energy barrier that has to be overcome in order to change the configuration of the dopant; see Fig. 2. When the injection energy, that is  $eV$ , becomes too small, the atom cannot cross the energy barrier from  $\text{Si}_d^+$  back to  $\text{Si}_i^-$ . This is because the energy provided by the electrons is not enough to relax the atom into a different lattice position, probably by either inelastic excitation of phonons or quantum tunneling.<sup>10</sup> Therefore, scanning at low bias voltage allows us to determine the charge state of the Si atom without the STM tip affecting it.

#### IV. CONCLUSION AND OUTLOOK

By combining optical measurements with STM, we were able to study the dynamics of the bistability of the Si atom in the (110) surface layer of GaAs. The Si charge state is affected by the laser and the applied bias voltage. The electrons from  $\text{Si}_i^-$  can escape via tunneling into the conduction band at high TIBB or, under illumination, via electron-hole recombination, both resulting in  $\text{Si}_d^+$  being favorable. At low bias voltage, the Si atom does not switch its configuration due to the energy barrier that is in between the two charge states.

To further investigate the bistable Si dynamics in a future experiment, we intend to perform time-resolved measurements of the switching frequency. For instance, the dependence of the frequency on the tunneling current can verify that two electrons are involved in the switching process. Furthermore, an analysis of the residence times of the  $\text{Si}_i^-$  and  $\text{Si}_d^+$  states can show that the switching process is fully random and allows the extraction of the relevant relaxation times.<sup>22</sup>

#### ACKNOWLEDGMENTS

The funding for this work was provided by FOM (Grant No. 09NSE06). We thank A. Y. Silov for valuable discussions.

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