Bridging the length scales between lithographic patterning and self assembly mechanisms in fabrication of semiconductor nanostructure arrays

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Abstract. We employ focused ion beam patterning of single crystal Si(100) surfaces to template the assembly of Ge(Si) nanostructure arrays. The evolution and final structures of the templated arrays are determined by combinations of transmission electron, low energy electron microscope, focused ion beam and scanning probe microscopies. It is shown how the positions of individual nanostructures may be controlled to the order of 10 nm. However, to achieve controlled spacings between elements that are in the 10 nm range requires careful matching of the characteristic lengths scales of self assembly mechanisms to the length scales of the external lithographic “forcing functions”.

1. Introduction
Several potential nanoelectronic architectures, such as quantum cellular automata [1,2] and coherent spin switches [3] require ultra high precision control of the position, dimension, spacing and electronic/ magnetic doping of individual semiconductor nanoscale elements in extended arrays. This manuscript focuses on the controlled assembly of such nanostructure arrays. Silicon (100) surfaces are patterned by an array of point-by-point focused ion beam pulses, and subsequent epitaxial growth of Ge(Si) nanostructures on the surface are templated to the original ion beam irradiation sites.

It is well established that growth of thin compressively strained epitaxial films leads to roughening of the surface and/or the formation of discrete nanoscale clusters of the epitaxial deposit. For the GeₙSi₁₋ₙ/Si(100) system considered here, the set of transitions that occur during near-equilibrium growth have been established by the work of multiple groups as comprising formation of a thin planar wetting layer of the epitaxial GeₙSi₁₋ₙ, followed by formation of coherent {510} faceted “hut clusters”, transformation to predominantly {311} faceted coherent “dome clusters”, and finally misfit dislocation formation within the clusters. For pure Ge/Si, the wetting layer is about 4 atomic monolayers thick and typical hut cluster dimensions are of order 30 nm. As strain is reduced through increasing Si concentrations in GeₙSi₁₋ₙ epitaxial deposits, the relevant length scales (wetting layer thickness, cluster dimensions etc) increase. It also becomes necessary to increase the substrate temperature to enable adatom diffusion lengths to be sufficiently large to attain these quasi-equilibrium structures (for example, Ge₀.₂Si₀.₈ substrate temperatures of order 750° C enable this
sequence of structural transitions to be maintained [4]), otherwise metastable morphological transitions may be observed, as will be discussed later in this manuscript.

The aim of the templating process is to transfer the symmetry of the focused ion beam (FIB) template pattern to the symmetry of the final nanostructure array as faithfully as possible. Thus for an initial ion distribution \( i(x,y) \) on the surface of the Si(100) substrate, and a final nanostructure distribution \( n(x,y) \), we want as simple as possible a relationship between these two distributions:

\[
n(x,y) = i(x,y) * t(x,y)
\]  

where \( t(x,y) \) represents the “smearing function” between the processes and * is the convolution operator.

Analogous to treatments of the transfer of spatial frequencies through an objective lens in high resolution transmission electron microscopy, it is instructive to consider the arrays as a Fourier summation of periodic components, and to determine what range of spatial frequencies are faithfully transmitted from the “object” (i.e. the templating array, \( i(x,y) \)) to the “image” (the nanostructure array, \( n(x,y) \)) by a transfer function \( T(u,v) \) in Fourier space.

\[
n(x,y) = F\{N(u,v)\} = F\{I(u,v) * T(u,v)\} = i(x,y) * t(x,y)
\]  

Here \( F[\cdot] \) represents the inverse Fourier transform operator; \( u,v \) are the Fourier space coordinates corresponding to real space coordinates \( x,y \); and the upper case function (e.g. \( N(u,v) \)) represents the Fourier transform of the corresponding lower case function (e.g. \( n(x,y) \)). For simplicity, we henceforth consider only real space and Fourier space variables, \( x \) and \( u \), i.e. we will assume isotropic behavior of the templating process with respect to the surface azimuthal direction. Fundamental questions include the maximum spatial frequency \( u_{\text{max}} \) that can be transmitted, i.e. the closest spaced objects (this is critical with respect to electron tunneling and spin transmission between adjacent nanostructures for nanoelectronic applications), and the minimum transmissible frequency \( u_{\text{min}} \) (which speaks to the practical overall dimensions of the array, and the ability to maintain large areas of unoccupied “white space” as needed in parts of the array). Between these values, efficient transfer of all spatial frequencies is required to faithfully replicate complex aperiodic patterns. We point out, however, that one significant difference from contrast transfer function analysis in TEM phase contrast imaging is that for high spatial frequencies we are not defining the ideal transfer function \( T(u) \) to be unity. Rather a major purpose of marrying lithographic templating with self assembly mechanisms is to access length scales below those that can be directly lithographically defined.

The major experimental limitations on the maximum spatial frequencies in the final nanostructure array, \( u'_{\text{max}} \), are: (i) the minimum spacings accessible by the initial patterning function, \( i(x,y) \), i.e. here by the spatial resolution of our focused ion beam pulses, corresponding to a maximum input spatial frequency, \( u'_{\text{max}} \) (ii) the characteristic capture zone dimensions, \( R \), for adatoms during epitaxial growth on the template sites, and (iii) the degree to which more complex nanostructures may be assigned to each lattice point defined by the template array. These last two factors speak to the balance of length scales between the initial lithographic pattern and the characteristic length scales over which nanostructures self assemble during the epitaxial growth process. The primary limitation on \( u'_{\text{min}} \) will be the tendency for spontaneous nanostructure formation away from template sites. Suppression of this process requires either a large adatom diffusion length (although as described later, this can place lower limits on \( u'_{\text{max}} \)), or a mechanism that inhibits nucleation away from templating sites.

2. Experimental

Structures are grown in a set of different UHV growth systems and examined by a range of microscopies; in several cases the growth and characterization systems are combined.

Ge quantum dot (QD) structures described in this manuscript are grown in one of two instruments: (i) A unique Hitachi UHV transmission electron microscope in the laboratory of Frances Ross at IBM.
Thomas J. Watson Research Center [5]. This instrument enables digermane and disilane deposition in the objective pole-piece area, allowing direct TEM observation of the CVD growth process in real time. An FEI 611 FIB column is connected under UHV in an adjacent preparation chamber, and is typically operated at 25 keV, an ion current of 10 pA, and ion pulse times per template feature in the range of hundreds of µs (Figure 1(a)). The ion beam diameter is of order 50 nm. Atomically clean, electron transparent Si(100) samples are prepared using procedures previously described [6]. Growth of Ge QDs from digermane on unpatterned Si surfaces was performed at temperatures in the range 500-600°C and typical digermane partial pressures ~ 10^{-7} Torr. (ii) A SPECS P90 UHV Low Energy Electron microscope (LEEM) [7], equipped with multiple in-situ deposition and surface modification capabilities, with a base pressure of ~ 10^{-10} Torr. Samples are cleaned by in-situ oxide desorption at temperatures ~ 800-1100°C (the lower temperatures are employed to enable pre-existing FIB template patterns to be retained) and Ge deposition is from a Knudsen cell at a substrate surface temperature estimated to be ~ 500-550°C.

For growth and templating of Quantum Dot Molecule (QDM) structures, Ge_{x}Si_{1-x} films were grown using molecular-beam epitaxy (MBE) on (100) Si. 300 µm thick substrates were chemically cleaned and oxidized using a modified Shiraki procedure [8] before being loaded into the growth chamber. The resulting surface oxide was desorbed at a temperature of 750-820°C and a 1000Å Si buffer layer was then grown at 750°C. After buffer growth, the substrate temperature was lowered to the Ge_{x}Si_{1-x} growth temperature (usually 550°C). Following temperature equilibration, Si buffer (typically of order 7 nm thickness) was grown followed by Ge_{x}Si_{1-x} alloy growth at the same temperature. Most of the FIB-templating of Ge(Si) QD and QDM sites on Si(100) surfaces described in this manuscript employs a standard Ga beam using an FEI 200 system, and an FEI 611 column integrated into a UHV-TEM system, as described above. We are also working with an Orsay Canion column [9] equipped with a mass selecting ExB filter, that allows separation of a far wider range of primary ion beams from liquid metal alloy sources (to date, we have generated Au, Si, B, As, Mn and Ge beams from AuSi, PdAsB and Mn(Au)Ge sources).

Samples are studied in-situ and ex-situ from growth by a set of different microscopies spanning lengths scales of interest in two and three dimensions, as summarized in Table I.

| Technique / Instrument | Lateral resolution a | Vertical resolution b | 2D projection, Surface, or 3D e | In situ or Ex-situ d |
|------------------------|----------------------|-----------------------|-------------------------------|--------------------|
| TEM / Hitachi UHV      | ~ 0.3 nm             | Atomic at surfaces    | 2D Proj and Surface           | In-situ            |
| TEM / JEOL 2000 FX     | ~ 0.3 nm             | N/A                   | 2D Proj                       | Ex-Situ           |
| LEEM / SPECS P90       | ~ 5 nm               | Atomic                | Surface                       | In-situ            |
| FIB / FEI 200          | ~ 10 nm              | ~ 10 nm               | Surface, 3D e                 | Ex-situ            |
| AFM / Multiple         | ~ 1 nm               | Atomic                | Surface                       | Ex-Situ            |

Table 1. Imaging methods employed in this work. a Lateral resolution is in the plane of the sample surface. b Vertical resolution is perpendicular to the sample surface. c “2D Projection” is through a thin foil, “Surface” is for images sensitive to near surface monolayers only, “3D” is for a full tomographic reconstruction; d “In situ” means growth and imaging occur in parallel, “Ex situ” means imaging occurs in a different chamber after growth is completed. e For further details see reference [10].

3. Templating of Ge Quantum Dots
The ability to control the nucleation of individual Ge quantum dot clusters by pre-patterning the substrate by FIB features is demonstrated in Figure 1(b,c). Using Ga⁺ ion pulses of order 25 keV energy, 10 pA current and 100 µs duration, a strong correspondence is observed between the initial template sites and the final positions of Ge clusters in the final array.

We have previously examined the fundamental mechanisms that govern this templating process [6,11-13] and concluded that the key process is the development of a transient morphology on the surface that evolves during post implant annealing. The initial implant dose in these particular...
experiments is insufficient to create substantial surface topography (the ion dose within the templating features is of order $10^{14}$ cm$^{-2}$, and the sputter yield is of order 2-3 for these conditions). In fact, under these ultra-low dose implant conditions, the surface actually swells due to damage and amorphization within the implanted volume. However, a fraction of a monolayer of the Si surface is sputtered away.

On annealing, these “surface vacancies” aggregate to form initially a shallow annulus within each implant feature, which then evolves with further annealing into a surface pit, and then the surface eventually replanarizes [6,13]. If the surface nanostructure evolution is terminated at the pit stage (by stopping the anneal or initiating growth), then a strong correlation is observed between the position of the original template sites and the position of the final QDs.

![Image](image_url)

**Figure 1.** (a) As implanted features for 25 keV 10 pA 100 µs Ga$^+$ pulses imaged in weak beam in the UHV - TEM. (b) and (c) AFM images of examples of Ge QD arrays templated to the FIB sites. In (b) “vacancy” sites are indicated with white arrows and a “Frenkel defect” (interstitial QD adjacent to an unoccupied template site) by a black arrow. The pattern in (c) corresponds to part of the arrangement of a simple quantum cellular automata (QCA) adder circuit.

We quantify this correlation as follows. Let $N_Q$ be the total number of quantum dots within a given area of substrate surface (we avoid the term “unit area” here, as the arrays we seek to fabricate may be of greatly varying local density, consistent with circuit architectures), and let $N_t$ be the total number of template sites in the same area. Let $F_n$ be the fraction of template sites that are occupied by exactly $n$ quantum dots. Thus $F_n$ corresponds to the fraction of vacant sites, $F_1$ corresponds to the fraction of template sites that are occupied by exactly one quantum dot (generally the preferred state), and $F_{n>1}$ corresponds to the fraction of multiply occupied sites ($n=2,3,4$, etc.). Finally let $N_i$ be the number of interstitial (i.e. not associated a template site) quantum dots within the given area. Then:

$$N_Q = N_t \sum n F_n + N_i$$

(3)

In general our goal is to suppress the terms $F_n$ ($n\neq 1$) and $N_i$ over the widest possible range of spatial frequencies, $F(u,v)$. The ability to achieve this goal will depend on both the energetic and kinetic landscapes encountered by surface adatoms during epitaxial growth. The energetic landscape will be determined largely by the strength of attachment of adatoms to the template sites, relative to the strength of attachment to other possible nucleation sites. The kinetic landscape will be dominated by adatom diffusion kinetics on the surface, which will be functions of substrate temperature and partial pressure of the depositing species.
Suppression of the $N_i$ term requires suppression of other heterogeneous nucleation sites, and the presence of an energetic landscape that suppresses spontaneous nucleation on the surface. In the present system, the latter condition is achieved by controlling the wetting layer thickness. For unmodified Si(100) surfaces the wetting layer thickness is about 4 monolayers, depending on growth conditions. Local modification with the Ga$^+$ focused ion beam reduces the wetting layer thickness to about 2 monolayers [13]. Thus, if the net amount of Ge deposited is between 2 and 4 monolayers, nucleation occurs on template sites, but not spontaneously in between. This in principle eliminates the $N_i$ term. Suppression of higher order ($n>1$) $F_n$ terms requires tuning of the template dimensions to the QD size. In development of the template morphology by post-implantation annealing, we find that growth on features that have evolved to the annular stage can result in multiple nuclei per template site ($n>1$), while growth at the nano-pit stage corresponds to the $n=1$ condition [6,13]. The final factor in equation (3) is the $n=0$ or vacancy state. This is the dominant “defect” in our QD arrays (see for example Figure 1b), with populations that grow with increasing spatial frequency of the template sites, as will be subsequently discussed.

In an extensive series of experiments [14], we have examined the filling of templates sites as function of spatial frequencies in the pattern. For these experiments, we maintain a constant number of template sites per unit area, but vary the maximum spatial frequencies by adjusting the inter-dot spacings within templates of four quantum dots in a square lattice as illustrated in Figure 2. It is observed that as the inter-dot spacing $d$ becomes smaller, the fidelity of pattern transmission decreases dramatically.

Figure 2. (a) Schematic of template array designed to investigate template filling as a function of template site separation. The spacing between QDs within a four-fold motif, $d$, varies but the total number of template sites per unit area of substrate remains constant. (b) Illustration of the simplified “capture length ($R$)” model. $r$ is the radius of an individual template feature.

Figure 3(a) shows how as the spacing between features in the four fold cluster reduces, the fraction of “vacancy sites” substantially increases. This may be understood on the basis of the concept of a capture zone, that bridges the length scales between the lithographic pattern and the local propensity for self assembly of the Ge quantum dots. The capture zone, of radius $R$ (capture length), is illustrated in Figure 2(b). The basic assumption of this model is that the nucleation of an island inhibits further nucleation within the capture length $R$ (the diffusion length of Ge adatoms is expected to be large compared to the site separation under the growth conditions employed in our experiments [15]). Initially, we allow islands to nucleate anywhere with equal probability within circles of radius $r$ centered on each nominal FIB implant site position, Figure 2(b). Once this first island has nucleated within its circle of radius $r$, it is then assumed that no other islands can nucleate within a radius $R$ centered at the first island. Thus, the exclusion area can partially overlap with the neighboring nucleation sites, and the probability of another island nucleating on an adjacent site is assumed to be proportional to the remaining available area. As described in Reference 14 and illustrated in Figure 3(b), this model can reproduce the fundamental feature of our experimental observations.

Interestingly, as described in Reference [14], the transition from the low to high vacancy regions for a given capture length $R$ occurs at template site spacings that are largely independent of the template feature size, $r$. Thus the challenge with increasing the fidelity of the templating process at high spatial frequencies is not predominantly a factor of improving $r$. This parameter (determined primarily by the quality of the focused ion beam optics in our experiments and the feature sizes that...
evolve at each template point during post implantation annealing [13]) controls the accuracy in positioning the nuclei (as it is assumed the initial nuclei can form anywhere within the template site) but not how closely they can be spaced. The minimum attainable spacing is primarily controlled by $R$. In comparing the results of our model to experimental observations, the best agreements between model and experiment occurs for values of $R$ that are of order 100 nm. While the magnitude of $R$ is expected to be governed by the same fundamental parameters, it does not appear to be equivalent to the adatom surface diffusion length, which from values in the literature (e.g. [15]) and from the fact that we can accurately populate much sparser template arrays (lower spatial frequencies), is considerably larger. Rather it appears to represent a critical length scale over which the local gradients in chemical potential are sufficient to drive Ge adatoms towards the clusters. However, given the finite time and temperature scales of these experiments, kinetic parameters clearly play a substantial role. For example, increasing the digermane partial pressure during growth reduces the “lifetime” of an adatom on a terrace before it encounters high densities of other atoms on the surface. This increases the probability that nucleation occurs in parallel on multiple closely spaced sites, and reduces the capture zone radius. As described in Reference [14], this indeed increases the fidelity of island attachment to template sites at higher spatial frequencies.

Another key parameter will be the diffusivity of adatoms between template sites. Of course, this is controlled partially by temperature, but it will also be controlled by surface structure. For example the Si(100) 2x1 surface reconstruction is known to have different adatom diffusivities parallel and perpendicular to the dimer rows [16]. The net adatom diffusivity will also depend critically upon the surface step density / terrace size between template sites, and recent LEEM results in our group suggest that this may be a potential route to controlling the size of the capture zone. Shown in Figure 4 is a LEEM image of the surface structure of a Si(100) surface that has been patterned with a Si

![Figure 3](image_url)
focused ion beam and then annealed to temperatures \( \sim 850^\circ \) C in the LEEM. This temperature is determined from observation of the LEED pattern to be just sufficient to generate a reconstructed surface, without creating so much surface mobility that the FIB features planarize. Note also that the Si ion doses per FIB feature are much higher than we typically use for Ga FIB templating of Ge quantum dots. The images are formed by placing an aperture around one of the orthogonal 2x1 reconstruction reflections, such that terraces with the dimer rows aligned to diffract into the selected diffraction spot appear bright, and adjacent terraces with the dimer rows rotated 90 degrees appear dark. The boundaries between the bright and dark terraces thus represent monatomic steps. It is seen that large terraces extend between adjacent template sites, with very low step densities between the template sites. This is believed to be due to the pits acting as efficient sinks for surface steps, as previously demonstrated [17], and should provide the ability to “tune” adatom diffusivity by controlling step densities between template sites, through the length and temperature of annealing steps and through the initial pit dimensions (which will control its ability to absorb steps).

We can thus summarize our progress to this point by saying that the interplay between surface energetic, kinetics and lithographically induced topology currently limits the maximum attainable QD spatial frequencies in these patterns comprising a single QD element on each lattice basis point to \( \nu_{\text{max}} \sim 10^7 \) m\(^{-1}\). The minimum attainable spatial frequency, \( \nu_{\text{min}} \) is controlled by the suppression of off-site “interstitial” nucleation between template sites that arises from the reduction of the Stranski Krastanov wetting layer thickness as a function of the implanted Ga density [13], as discussed earlier in this manuscript. For regularly spaced arrays of template sites, the wetting layer thickness changes from 1.75 ± 0.25 monolayers for a template site center-center spacing of 180 nm to 2.25 ± 0.25 monolayers for a template center-center spacing of 360 nm to 3.75 ± 0.25 monolayers for an unmodified Si (100) surface. The corresponding average Ga doses per unit area of substrate are \( 2 \times 10^{13} \) cm\(^{-2}\), \( 2 \times 10^{12} \) cm\(^{-2}\) and 0. While it is somewhat surprising that such modest doses (only a fraction of which will, of course, reside at the surface) can modify the wetting layer thickness, this phenomenon is of great advantage in that by growing between about 2 and 4 monolayers (depending upon the exact template feature density), nucleation occurs on the template sites, but not on unpatterned regions. In principle, this extends the minimum transferrable spatial frequency down to zero, although the difference between wetting layer thicknesses for patterned and unpatterned features will become vanishingly small as the template feature density / dose is sufficiently reduced. However, in practice we find that nucleation is suppressed over length scales of at least microns to tens of microns, which in terms of the interaction distances for the mechanisms relevant to the nanoelectronic device architectures we are considering (spin exchange, tunneling etc) is essentially infinite.

In summary, lithography can be used to template Si(100) surfaces over spatial frequencies:

\[
0 \leq \nu' \leq 10^5 \text{ m}^{-1}
\]  

The resulting transfer function limits the range of attainable spatial frequencies to:

\[
0 \leq \nu'' \leq 10^7 \text{ m}^{-1}
\]  

Figure 4. Dark field LEEM image (aperture around \( \frac{1}{2},0 \) reconstruction spot) from 2x1 reconstructed Si(100) surface which has been periodically patterned with pits (arrowed) by a Si focused ion beam. Field of view is 5 \( \mu \)m.
This attenuation of higher spatial frequencies is due to the characteristic length scales of self assembly, specifically the effective capture cross-section for adatoms of a nucleated cluster.

4. Templating of Complex Nanostructures: The Quantum Dot Molecule

For the nanoelectronic device architectures we are considering, the relevant interaction lengths (i.e. for spin exchange/interaction or electron/hole tunneling) are substantially less than the minimum QD spacings we can reliably transfer through the templating processes described in section 3. As a solution, we seek to assemble more complex nanostructures with the necessary length scales built into each component through self-assembly mechanisms. To develop the analogy of a crystallographic lattice, we would then be realizing the equivalent of a complex basis on each lattice point.

Such a basis is the Quantum Dot Molecule (QDM), a complex nanostructure that evolves spontaneously during growth under conditions of limited adatom mobility in the Ge$_x$Si$_{1-x}$/Si(100) system [18-21]. It comprises a four fold set of quantum dots bound elastically to a central pit, Figure 5(a). The evolution of the QDM follows the following trajectory (illustrated with specific numbers for growth by Molecular Beam Epitaxy growth of Ge$_{0.3}$Si$_{0.7}$/Si(100) at 550°C and a growth rate of about 1 Ångstrom s$^{-1}$). (i) First, the Ge$_{0.3}$Si$_{0.7}$ growth is planar up to a wetting layer thickness ~ 5 nm. (ii) At this point, strain relieving pits start to form, which are shallower than the film thickness and are thus assumed to not be directly associated with contamination at the original growth surface. (Although morphological relief of strain in compressively stressed films is more generally achieved through islanding, pits are also effective strain relief mechanism, e.g. [22]). (iii) With further film growth, the pit widens and deepens onto $\{510\}$ facets and quantum dots start to grow at each pit edge. (iv) The quantum dots elongate along the edge of the pit and eventually form a continuous wall surrounding the pit. (v) At this point lateral growth of the QDM stops, and $\{510\}$ facets define both the pit and QDs. On further growth, the structure maintains its shape and dimensions on the surface until the film thickness is large enough for formation of interfacial misfit dislocations. The QDM is then destabilized. For these specific film stoichiometries and growth conditions, the QDM size measured between outer edges of opposite QDs is 220 nm. This dimension scales with strain to produce increasingly smaller QDMs at higher Ge concentrations (e.g. 120 nm at Ge$_{0.5}$Si$_{0.5}$). X-Ray

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{AFM images of QDM structures in 20 nm Ge$_{0.3}$Si$_{0.7}$/Si(100) grown at 540-550°C, 0.09 nm/s. (a) Spontaneous formation on unpatterned surface, (b1,b2,b3) Growth of regular arrays on FIB patterned substrates. Note untemplated QDMs in b1 (circled), (c) Template pattern illustrating suppression of untemplated QDMS in proximity to template features. Image edges are along in-plane <001> directions in (a) and <011> directions in (b), (c).
\end{figure}
synchrotron experiments have demonstrated that the QDMs are Ge rich with respect to the surrounding matrix, and will thus confine carriers (holes) due to lower bandgap. [23]

While on an unpatterned surface, these nanostructures form quasi-randomly at densities ~ 10^8 cm^-2, we can template their formation by FIB patterning to “seed” the initial pits. Because we have no FIB column integrated with the MBE system in which these structures are grown (and because the growth rates and compositions are not accessible to the CVD processes in the UHV-TEM where the quantum dot arrays described in section 2 were grown), we FIB template ex-situ to the growth chamber, i.e. before substrate cleaning and growth. This process requires parallel optimization of many parameters [21]: the size of the ex-situ FIB seeding pits, the subsequent wafer cleaning conditions, the initial Si buffer layer growth conditions, and the final Ge_xSi_1-x growth conditions. We have established conditions where initial Ga^+ FIB template features of just a few nm deep can survive the substrate cleaning process, propagate through subsequent Si buffer layer and Ge_xSi_1-x film growth, and successfully template QDM structures.

Separation between individual QDs within the cluster can be controlled by arresting growth at the appropriate point of QD elongation along the pit edges, and we can thereby control the separation to be ≤ 10 nm, i.e. u'_max > 10^8 m^-1. Equally, as the QDMs evolve to a narrow distribution of shapes and sizes [19], we can accurately control the spacing between QDMs, as illustrated in Figure 5(b).

The major limitation associated with QDM templating is that while we can successfully template QDMs with very high fidelity (i.e. the vast majority of templated sites result in growth of an associated QDM and there is thus a low “vacancy” concentration), there is no mechanism for suppressing off-site nucleation of QDMs (“interstitials”) as they will continue to form randomly in unpatterned regions. Thus as illustrated in Figure 5(c) we can typically suppress QDM formation only within adatom diffusion lengths / capture lengths of templated sites. This limits the minimum accessible spatial frequency, u'_min, in the FIB-templated QDM patterns. In practice this untemplated nucleation typically (but not always) occurs at length scales of order a µm or greater from template sites, and would then not interfere with operation of the intended nanoelectronic architecture. However, this lack of a mechanism inhibiting off-site nucleation is a limitation to using QDMs as a basis on the template lattices we construct.

In summary of this section, for lithographic templating over spatial frequencies:

0 ≤ u' ≤ 10^7 m^-1  \hspace{1cm} (6)

We can assemble structures with a range of spatial frequencies

10^6 ≤ u' ≤ 10^8 m^-1 \hspace{1cm} (7)

In this case control of self assembly enables extension of the maximum spatial frequencies that can be accessed in the final array, but the lack of a mechanism for suppressing nucleation off template sites limits the minimum spatial frequencies that can be transferred from the template pattern.

5. Summary and Continuing Work

We have demonstrated how focused ion beam templating of Si(100) surfaces enables controlled nucleation of Ge(Si) nanostructures in patterns of any desired complexity and with high fidelity transfer of a broad range of spatial frequencies. The maximum spatial frequency – which is the more critical limit for nanoelectronic device architectures - that can be transferred from template pattern to nanostructure array depends on subtle interplays between the length scales forced by the template pattern and the mechanisms driving cluster self assembly on the surface during growth. Depending on this interplay the maximum spatial frequency in the nanostructure array can be either enhanced or attenuated with respect to that contained within the template array. The minimum spatial frequency is largely dictated by the presence or absence of a mechanism that inhibits nucleation off the template sites. For the nanostructures discussed here, spatial frequencies from essentially zero to 10^8 m^-1 can be designed into the final nanostructure pattern.

Our current work is focusing on the electronic and magnetic functionalization of these nanostructures. The work reported in this paper uses a Ga^+ focused ion beam to template the surfaces.
The use of Ga\textsuperscript{+} is virtually ubiquitous in conventional liquid metal ion source (LMIS) FIBs because of its low melting point and low vapor pressure at the melting point. However Ga\textsuperscript{+} is a shallow acceptor in Si and Ge, and will thus dope the substrate and QDs wherever it is present. For any practical fabrication of prototype nanoelectronic devices, we need to be able to template with electronically non-invasive species (such as Si or Ge) and to be able to intentionally dope with nanoscale precision to introduce extra carriers or spins into the system (e.g. with Mn, B, or As). Using a mass-selecting FIB column from Orsay Physics \cite{9}, we have succeeded in generating beams of Si, Ge, Mn, As and B from AuSi, Mn(Au)Ge and PdAsB sources respectively. To date beam resolution is of order 30-50 nm (limited primarily by vibrations in our home-built system) and we can deliver ion pulses from a few ions to numbers large enough for any templating or doping purposes. We are also exploring in detail structural, optical and electronic recovery of FIB implanted and annealed structures to understand recovery and activation in these structures.

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