Quantum-dot systems prepared by 2D organization of nanoclusters preformed in the gas phase on functionalized substrates

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Abstract. The low-energy cluster beam deposition (LECBD) technique is used to deposit gold nanoclusters preformed in the gas phase on functionalized graphite substrates (highly oriented pyrolitic graphite (HOPG)), to prepare 2D-organized arrays of cluster assembled dots. Functionalized HOPG substrates are obtained using the focused ion beam (FIB) nanoengraving technique to pattern 2D-organized arrays of defects (nanoholes, nanobumps) which act as traps for the diffusing clusters. Depending on the deposition conditions (nature, size and fluence of the deposited clusters) and the functionalized substrates (nature and size of the FIB-induced defects, geometry of the 2D array of defects and temperature during deposition) high-quality quantum-dot arrays can be obtained with well controlled and reproducible morphologies. Kinetic Monte Carlo simulations of the cluster deposition experiments on functionalized substrates allow us to obtain quite good fits of the experimental images performed by tapping mode atomic force microscopy (TMAFM), leading to systematic investigations of the best conditions to realize high-quality quantum dots systems. This combined top-down–bottom-up approach (LECBD-FIB) seems a promising method for preparing high-integration-density devices (\(~\text{Tbit cm}^{-2}\)) well suited for future applications to data storage, nanoelectronics, nano-optics, nanomagnetic systems.
1. Introduction

The design and fabrication of quantum dots systems and the study of their properties are playing an increasingly important role, mainly because of the large number of potential applications in various fields such as high-density information storage (Tbits in $^2$), nanoelectronics, nano-optics, magnetics and magnetoelectronics devices [1]–[3]. Several approaches to produce such systems have been developed, including top-down, bottom-up or combined top-down–bottom-up technologies. However, in most cases, the individual nanosize dots must exhibit specific structure/morphology and properties which are very often difficult to obtain using conventional preparation techniques. Moreover, the specific geometries of the quantum-dot arrays require the use of well suited techniques with spatial resolutions typically in the 1–10 nm range. In this context, we propose an original combined bottom-up–top-down method based on the deposition of functionalized nanoclusters (i.e. with controlled size, morphology and composition) preformed in the gas phase on functionalized substrates to prepare 2D-organized arrays of clusters. The synthesis of clusters in the gas phase using a combined laser vaporization–inert gas condensation source offers some unique advantages mainly related to the non-equilibrium conditions which govern the cluster formation [4]. More specifically, when using the pulsed high-pressure inert-gas condensation method, cooling rates as large as $10^8$–$10^{10}$ K s$^{-1}$ allow the formation of functionalized pure or mixed clusters which cannot be obtained using conventional techniques. Subsequently, the low-energy cluster beam deposition (LECBD) technique [4]–[6], by which clusters with very low energy (gained in the supersonic expansion at the exit nozzle of the source) are deposited on functionalized substrates, is used. In such a low-energy deposition regime, clusters are not fragmented upon impact on the substrate, leading to the formation of supported nanostructures which retain the unique properties of free incident clusters [4]–[7]. Moreover, clusters with diameters as large as a few nanometres (a few thousands of atoms) can easily diffuse on the surface of the substrate depending on the cluster/substrate interaction [8]–[11]. Thus, by controlling the nucleation and growth process of cluster islands on functionalized substrates with 2D arrays of traps (defects), organized arrays of quantum dots could be obtained. One has to notice that the main stages arising in the process described above are completely independent. The synthesis of functionalized clusters takes place in the cluster source while the formation and organization of supported cluster-assembled dots take place in a second step on the substrate. This allows wide choices in the experimental conditions used for preparing clusters and for determining the nature of the clusters (covalent semiconducting, metallic/bimetallic, oxides, ...), as well as for deciding the nature and the preparation/functionalization of the surface of the substrate. In this last case, various nanolithography or nanoengraving techniques can be used to realize the 2D arrays of defects likely to trap the deposited clusters.

This paper reports on the preparation of 2D-organized arrays of gold nanoclusters on functionalized HOPG substrates (highly oriented pyrolitic graphite). The focused ion beam (FIB) technique [12, 13] is used to realize rather large ($\approx 100 \times 100$ $\mu$m$^2$) 2D-organized arrays of different nanodefects (nanoholes, nanobumps, ...) with controlled geometries on HOPG substrates prior to gold cluster depositions in the LECBD regime in various conditions (cluster flux, temperature of the substrate). In situ STM–AFM observations of the samples reveal the trapping of gold clusters at the defects, which leads to the formation of stable 2D-organized cluster-assembled dots. Simulations using the DDA model (deposition–diffusion–aggregation) [14], previously developed to study the nucleation and growth process characteristic of the LECBD regime, have been carried out to interpret the experimental results in this particular

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case of gold clusters deposited on functionalized HOPG substrates. From both experiments and simulations, the best conditions for the preparation of high-quality quantum-dot arrays are discussed and extended to other systems of interest.

2. Preparation of FIB-functionalized HOPG substrates

The high-resolution (≤ 10 nm) FIB instrument specially designed and fabricated at the LPN-CNRS, Marcoussis [12] was used for these experiments. This equipment operates with 30 keV gallium ions produced from a liquid metal ion source (LMIS) allowing a close to 8 nm diameter ion probe to carry 6 pA. A high-performance pattern generator with pixel exposure times ranging from milliseconds down to 100 ns is used to scan the beam over the sample surface in order to pattern the HOPG substrates with well defined geometries and ion fluences per point. Before FIB patterning, freshly cleaved HOPG substrates are annealed under ultra-high vacuum at 770 K for 5 h. This treatment results in atomically flat and clean graphite surfaces extending over several μm between steps.

Tapping mode atomic force microscopy (TMAFM) images of FIB-patterned HOPG substrates at room temperature for two different ion fluences ($I_f = 3750$ and 37 500 Ga$^+$ ions/point) and defect periodicities ($L_{def} = 100$ and 300 nm) are displayed in figure 1. Depending on the FIB fluence per point, the morphologies of the nanodefects created on the surface of the crystalline graphite substrate are different. Nanobump-type structures are observed in the case of low point-doses (i.e. 3750 Ga$^+$ ions/point, figure 1(b)), while nanocrater-type structures are observed for higher ion point-doses (i.e. 37 500 ions/point, figure 1(a)). The sputtering yield of the graphite sample is calculated using the SRIM code [15] and a value close to 2 carbon atoms/Ga$^+$ ion is found, in good agreement with the experimental results. For high ion point-doses, nanoholes with a mean diameter of about 30 nm and a depth of about 1 nm are etched in the sample, and redeposition of sputtered C atoms around the holes leads

Figure 1. TMAFM images showing the morphologies of FIB-created defects on HOPG substrates at room temperature with 30 keV Ga$^+$ ions and different ion fluences $I_f$ and periodicities $L_{def}$. (a) $L_{def} = 300$ nm, $I_f = 37 500$ Ga$^+$ ions/point, image scale: 3 μm × 3 μm and inset 0.5 μm × 0.5 μm; (b) $L_{def} = 100$ nm, $I_f = 3750$ Ga$^+$ ions/point, image scale 1 μm × 1 μm.
to the characteristic nanocrater-type morphology observed with a lateral extension of about 50 nm (figure 1(a)). Conversely, for lower ion fluences per point, only ion-induced defects and subsequent local volume variations (spatial extension 30 and 1 nm height, figure 1(b)), corresponding to the transition from highly crystalline graphite to amorphous carbon, are evident. More detailed experimental studies and simulations of the FIB-defect creation and morphology are reported elsewhere [13]. The above-described results illustrate the interesting potential of the high-resolution FIB patterning technique for the functionalization of our substrates, since by changing the ion fluence one can strongly modify the nature of the nanodefects and thus explore a large range of interactions between defects and deposited clusters. Moreover, the periodicity and the geometry of the defect lattice on the HOPG surface can be easily and precisely adjusted. The results are reproducible and the time needed to pattern a large area (0.1 × 0.1 mm$^2$) is relatively short (one to a few tens of seconds in the case of low doses), emphasizing the high damaging efficiency of energetic heavy ions compared to electrons or photons.

3. Cluster preparation and depositions on functionalized HOPG substrates to form 2D-organized arrays

Gold clusters with a mean size centred on 2.9 nm (750 atoms: Au$_{750}$) are prepared using the specially designed equipment schematically represented in figure 2. A cluster generator based on a combined laser vaporization/gas condensation source has been developed to produce intense supersonic jets of nanoclusters with sizes ranging from a few tens to a few thousands of atoms (diameter ≈1 to a few nanometres), typically. Briefly, a Nd–YAG laser (λ = 532 nm, pulse duration ≈ a few nanoseconds, frequency ≤ 30 Hz) is used to vaporize the high-purity gold rod mounted in the source chamber (figure 2). Combined with the laser pulse, a low-pressure (a few tens of mbars) continuous flow of helium is injected in the source to rapidly cool the plasma generated at the target surface and to nucleate clusters, which are subsequently completely cooled and stabilized in the supersonic expansion taking place at the exit nozzle of the source. The main feature of this type of cluster source compared to other ones currently used (thermal, sputtering) is the very high cooling rate of up to about 10$^{10}$ K s$^{-1}$ which governs the formation of original nanoscale systems in non-equilibrium conditions. From the key parameters of the source (laser pulses, continuous gas-flow pressure, geometries of the nucleation chamber in the source and the nozzle), it is possible to control the cluster size distributions measured in the high-resolution time-of-flight (TOF) mass spectrometer mounted immediately after the skimmer (figure 2). In this case, neutral clusters are photoionized using a tunable excimer-dye laser, then deviated and accelerated at the entrance of the TOF line.

After the first stage of free cluster studies in flight, only neutral clusters having the very low energy gained in the supersonic expansion at the exit of the source (∼ 0.25 eV/atom in the case of gold clusters) are deposited on substrates in the UHV deposition chamber mounted on line with the cluster generator–TOF arrangement (figure 2). In this case, the complete distributions of neutral clusters (nearly Gaussian in shape and rather narrow), centred on Au$_{750}$, are directly deposited on the substrates to grow films. The incident average flux ($F$) and the equivalent thickness deposited ($t$) are controlled with a quartz balance. The temperature of the sample holder in the UHV deposition chamber can be adjusted in order to control the cluster diffusion on the HOPG surface. Heating of the FIB-patterned HOPG substrates (770 K for 2 h in situ in the UHV chamber) prior to cluster depositions can also be performed to clean the surface layer of the substrates depending on their transfer conditions (in air or in vacuum) from the FIB
Figure 2. Schematic view of the cluster generator based on a combined laser vaporization/inert gas condensation source, associated with a TOF mass spectrometer for the studies of free clusters and a UHV deposition chamber for cluster-assembled film preparations. The source can operate with one or two independent laser/target arrangements for the production of pure clusters as well as mixed clusters. Some characterization techniques of the films are available, e.g. *in situ* in UHV:RHEED diffractometry, STM–AFM near-field microscopy, XPS–ISS spectrometry. A UHV transfer system is used for sample transfers to *ex situ* equipment. An electron-beam evaporator in the UHV chamber is used for codeposition experiments to produce films of clusters embedded in various media.
Figure 3. UHV STM images (1 µm × 1 µm) of the morphologies of 0.2 nm thick gold-cluster films deposited at room temperature: (a) on an HOPG substrate without defect; (b) on an HOPG substrate with a random distribution of nanoholes created before deposition by 1.5 keV Ar⁺-ion bombardment and subsequent annealing in air. The high magnification image (25 nm × 25 nm) inset in (b) shows a group of three gold clusters (in white in the figure) trapped at an ion-induced nanohole (in black in the figure).

cluster-assembled nanostructures in the form of ramified islands is a proof of the capability of the deposited clusters to diffuse. Such diffusion effects have previously been reported for various cases (i.e. Sb clusters on HOPG) [6, 16], and interpreted from molecular dynamics simulations [8, 17]. Moreover, using kinetic Monte Carlo (KMC) simulations (DDA model: deposition–diffusion–aggregation) [14], the morphologies of the LECBD films are well fitted, allowing us to deduce the cluster diffusion coefficient. In the case of Au750/HOPG at room temperature represented in figure 3 a diffusion coefficient as large as $10^{-8}$ cm² s⁻¹ is estimated.

The second key condition in the preparation of 2D-organized arrays of cluster-assembled dots on functionalized substrates is the capability of specific surface defects to trap the diffusing clusters. This is well evidenced in figure 3(b) in the case of ion-induced defects on an HOPG substrate. In fact, the deposition conditions in the case of figure 3(b) are similar to those of figure 3(a) while the morphologies of the cluster-assembled nanostructures are significantly different in both cases. This is due to the presence of defects on an HOPG surface (nanoholes, see inset in figure 3(b)) created by 1.5 keV Ar⁺ ions and subsequent annealing in air which efficiently act as traps for the diffusing gold clusters. Some gold clusters trapped at the border of an ion-induced nanohole are clearly observed in the high-magnification image presented in the inset of figure 3(b).

Finally, since both above-mentioned conditions (cluster diffusion and ion-induced traps) are verified in the case of Au750/HOPG, we can expect to succeed in the preparation of 2D-organized arrays of clusters by using FIB-patterned substrates rather than randomly ion-bombarded substrates as in the case of figure 3(b). TMAFM images of 0.05 nm thick gold-cluster films deposited on various FIB-patterned HOPG substrates at 400 K to favour their diffusion are presented in figure 4. In all cases (FIB-patterned HOPG with $I_f = 3750$ or 37 500 Ga⁺ ion/point and $L_{def} = 300$ or 500 nm), the trapping of clusters on or around the FIB-induced defects...
Figure 4. TMAFM images (2 $\mu$m $\times$ 2 $\mu$m) showing the morphologies of 0.05 nm thick gold-cluster films deposited on FIB-patterned HOPG substrates at 400 K with different ion fluences $I_f$ and periodicities $L_{def}$. (a) $L_{def} = 300$ nm, $I_f = 37500$ Ga$^+$ ions/point; inset (500 nm $\times$ 500 nm), (b) $L_{def} = 300$ nm, $I_f = 3750$ Ga$^+$ ions/point, inset (750 nm $\times$ 750 nm); (c) $L_{def} = 500$ nm, $I_f = 37500$ Ga$^+$ ions/point.

leads to the formation of 2D-organized arrays of cluster-assembled dots. However, the quality of the quantum-dot arrays is affected by the FIB-induced defect morphologies (nanocraters or nanobumps, see section 2) directly related to $I_f$, and by the lattice distance $L_{def}$. In the case of large FIB-induced defects (nanocraters for large $I_f$, figure 1(a)), clusters are distributed around the nanocraters (see high-magnification image in inset of figure 4(a)) leading to a rather large lateral expansion of the corresponding cluster-assembled dots. More compact cluster islands are observed on an HOPG substrate patterned with a low FIB fluence (nanobump for low $I_f$, figure 1(b)). On the other hand, nucleation of small cluster islands between FIB defects is observed in the case of a large $L_{def}$ substrate (figure 4(c), $L_{def} = 500$ nm) while this effect is limited in the case of a lower $L_{def}$ substrate (figure 4(b), $L_{def} = 300$ nm).

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Figure 5. Comparison between experimental (a) and KMC-simulated (b) morphologies of cluster deposits on functionalized substrates performed under the same conditions (periodicity $L_{\text{def}} = 300$ nm, deposited thickness $t = 0.05$ nm, and substrate temperature during deposition $T = 400$ K). (a) TMAFM image of Au$_{750}$ deposited on an FIB-functionalized HOPG substrate, (b) KMC simulation of Au$_{750}$ on a substrate patterned with an array of specific trapping sites (●) with irreversible sticking (ideal traps).

Finally, for the preparation of high-quality 2D arrays of quantum dots it seems clear that small FIB defects are preferable to limit the lateral expansion of the cluster-assembled dots. This implies the choice of low $I_f$ values to pattern the HOPG substrates with 30 keV Ga$^+$ ions, taking into account their high damaging efficiency [13]. Moreover, the best choice of the $L_{\text{def}}$ value can be deduced from KMC simulations [14] of the low-energy cluster deposition on patterned surfaces. Assuming the diffusion of incident clusters and compact islands with sizes up to 20 clusters [17] and introducing specific trapping sites on the surface of the substrate with irreversible sticking (ideal traps), one obtains good fits of the TMAFM images as shown in the example of figure 5. For these simulations, the diffusion coefficient of compact cluster islands with size $S$ ($D_s$) is deduced from one of the individual clusters ($D_1$) using a power law in the form $D_s = D_1 S^{-\gamma}$, with $\gamma = 2/3$, and this has been experimentally and theoretically verified [8]. Finally, the best arrays are obtained when all cluster islands are nucleated on FIB-induced defects and no islands are formed between defects. This condition can be expressed in the form:

$$(N_{\text{isl/fs}})/(N_{\text{def/fs}}) = 1$$

with $N_{\text{isl/fs}}$ the density of cluster islands formed on an FIB-functionalized substrate and $N_{\text{def/fs}}$ the density of defects created by FIB on the functionalized substrate. The evolution of the above-mentioned ratio in equation (1) versus $N_{\text{isl/non fs}}/N_{\text{def/fs}}$, as deduced from the simulations using the DDA model [14], is represented in figure 6. $N_{\text{isl/non fs}}$ is the density of islands formed on virgin substrate (non-FIB-functionalized) which can be experimentally measured or theoretically deduced using the relation obtained from the DDA model [14]: $N_{\text{isl/non fs}} \propto (F/D_1)^{\chi}$, with $\chi \approx 1/3$. We remark in figure 6 that on the left-hand side of the vertical dotted line equation (1) is satisfied ($N_{\text{isl/fs}}/N_{\text{def/fs}} = 1$), while on the right-hand side of this line the existence of cluster islands nucleating between FIB-induced defects is predicted. This is in agreement with the experimental observations reported in figure 4 since the points corresponding to the
Figure 6. Predictions from KMC simulations of the evolutions of the ratio \( N_{isl/fs}/N_{def/fs} \) (black circles) and the mean size of cluster islands (red circles) as a function of the ratio \( N_{isl/non/fs}/N_{def/fs} \). Dotted and solid curves are guides for the eyes. Note that the growth of the films is stopped when all the defects of the surface are filled by at least one incident cluster. Two TMAFM images corresponding to the experimental cases presented in figures 4(a) and (c) are inset at the corresponding points (large grey circles) on the curve of \( N_{isl/fs}/N_{def/fs} \) showing the good agreement between experimental and simulated results.

Experimental cases of figures 4(a) and (c) fit the predicted curve quite well. More specifically, the point representing the case of figure 4(a) is located in the centre of the region of figure 6 where high-quality arrays without cluster islands between defects are expected, which is confirmed by the TMAFM image. In the case of figure 4(c), the corresponding point in figure 6 is located close to the frontier at \( N_{isl/non/fs}/N_{def/fs} = 1 \), indicating the large probability of nucleating cluster islands between defects in this case, which is confirmed by the TMAFM image. In this last case, the distance between the FIB defects (500 nm) is sufficiently large compared to the mean free path of the diffusing clusters to allow the nucleation of cluster islands between defects. Finally, from figure 6 it is possible to predict the best experimental conditions to obtain high-quality cluster arrays. The adjustable parameters to fit these optimal conditions are: the cluster flux \( \dot{F} \) and the diffusion coefficient of the deposited clusters \( D_1 \), which control the value of \( N_{isl/non/fs} \), and the distance between the FIB defects which fixes the value of \( N_{def/fs} \). Note that the diffusion coefficient of the supported clusters can be easily adjusted from the temperature of the substrate during deposition. In all cases we will have to realize the condition: \( N_{isl/non/fs}/N_{def/fs} \ll 1 \) (figure 6).

The second curve (discontinuous line) in figure 6 gives the mean size of cluster islands trapped at defects as deduced from KMC simulations. We remark that the lowest cluster-island size attainable in the region of high-quality 2D arrays (\( N_{isl/non/fs}/N_{def/fs} \ll 1 \)) is of the order of 5–10 clusters when all FIB defects are filled with clusters. This is due to the growth mode of these cluster islands which is governed by a Poisson law [14]. Consequently, using this technique of cluster deposition on functionalized substrates to prepare 2D arrays of quantum
Figure 7. KMC simulations of the morphological evolutions (size and shape) of the cluster islands located on the defects patterned on the surface of the substrate ($L_{\text{def}} = 300$ nm) with the deposited thickness $t$. (a) $t = t_c$, (b) $t = 100 \, t_c$. The critical thickness $t_c$ corresponds to the lower thickness of the deposit required to fill all the surface defects by at least one incident clusters.

dots, we realize that we have no chance of preparing an array with one single cluster per defect. However, for various applications, a dot consisting of a group of clusters strongly coupled is suitable (i.e. magnetic nanostructures) [18]–[20].

As mentioned above, the nature and the morphology of the FIB-induced defects influence the morphology of the cluster islands. However, for a defined defect morphology, other adjustable parameters can be used to control the cluster-assembled dot sizes and morphologies. In particular, the deposited thickness ($t$) can be used to monitor the mean cluster-island size as shown in figure 7. We can define a critical deposited thickness ($t_c$) corresponding to the lower deposited thickness required to fill all the FIB-induced defects with at least one incident cluster (figure 7(a)). Then, if we assume that the FIB defects are the exclusive nucleation centres, above $t_c$ the growth of the cluster islands trapped on these defects will take place as observed in figure 7(b). Thus by changing $t$ one can control the mean size on the islands created on defects. Note that for large deposited thicknesses, ramified islands are formed which can evolve towards more compact islands (sometimes more suitable for applications) by increasing the substrate temperature during cluster deposition or by annealing the sample after deposition. In this case, the coalescence of adjacent gold clusters [21] at the origin of this evolution is improved.

4. Conclusion

The LECBD technique seems promising for the synthesis of original nanostructures and nanostructured systems. Simple nanosystems based on pure clusters for fundamental studies as well as functionalized systems based on more complex mixed clusters for different applications can be easily prepared and stabilized in well controlled and reproducible conditions. The combination of the bottom-up approach to synthesize original nanostructures from clusters preformed in the gas phase and the top-down FIB nanoengraving technique to functionalize the substrates allows us to prepare 2D-organized arrays of quantum dots with rather well defined geometries and morphologies. The three main stages involved in the preparation of such systems are completely independent leading to a large choice of experimental conditions at each stage.
The cluster synthesis takes place in the gas phase in non-equilibrium conditions well suited to prepare functionalized nanoparticles. The functionalization of the substrate, especially using the FIB technique, offers some interesting potential ways of controlling the geometries of the 2D arrays and the nature and morphologies of the defects to trap the deposited clusters. Finally, the cluster deposition on the functionalized substrates in the LECBD regime is characterized by a memory effect of the free cluster structures and properties, since in the low-energy deposition experiments clusters are not fragmented upon impact on the substrate. All these aspects are well illustrated in this paper which reports the first experiments to prepare 2D arrays of nanostructured dots on FIB-functionalized HOPG substrates from clusters preformed in the gas phase. Results from KMC simulations using the DDA model are in good agreement with the experimental ones, making it possible to predict the 2D-array morphologies and to improve the experimental conditions with a view to obtaining high-quality organized systems.

Complementary experiments on the nucleation and growth process of cluster-assembled dots on FIB-induced defects are in progress and novel systems of practical interest are being prepared based on mixed clusters for applications in high-density integration devices typically in the Tbits cm$^{-2}$ range. For that purpose, the results obtained in the specific case of gold clusters/HOPG can be easily extrapolated to a large number of other systems which satisfy to both key conditions, i.e. easy diffusion of supported clusters in the experimental conditions adopted and the existence of stable defects on the substrate which are efficient in trapping the diffusing clusters.

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References

[1] Orlov A O, Amlani I, Bernstein G H, Lent C S and Snider G L 1997 Science 277 928
[2] Sun S, Murray C B, Weller D, Folsk L and Moser A 1989 Science 287 1989
[3] Andres R P, Bein T, Dorogi M, Feng S, Henderson J L, Kubiak C P, Mahoney W, Osifchin R G and Reifenberger R 1996 Science 272 1323
[4] Perez A, Mélinon P, Dupuis V, Prevel B, Bardotti L, Tuilleon-Combes J, Masenelli B, Treilleux M, Pellarin M, Lermé J, Cottancin E, Broyer M, Jamet M, Negrier M, Tournus F and Gaudry M 2001 Mater. Trans. (special issue on nano-metals 1) 42 1460
[5] Binns C 2001 Surf. Sci. Rep. 44 1
[6] Brechignac C, Cahuzac P, Carlier F, De Fructos M, Masson A, Mory C, Colliex C and Yoon B 1998 Phys. Rev. B 57 2084
[7] Haberland H, Insepov Z and Moseler M 1995 Phys. Rev. B 51 11 061
[8] Deltour P, Barrat J L and Jensen P 1997 Phys. Rev. Lett. 78 4597
[9] Bardotti L, Prevel B, Treilleux M, Mélinon P and Perez A 2000 Appl. Surf. Sci. 164 52
[10] Hou Q, Hou M, Bardotti L, Prével B, Mélinon P and Perez A 2000 Phys. Rev. B 62 2825
[11] Bardotti L, Prével B, Mélinon P, Perez A, Hou Q and Hou M 2002 Phys. Rev. B 62 2835
[12] Gierak J et al 1999 Nuclear instruments and methods Phys. Res. A 427 91

New Journal of Physics 4 (2002) 76.1–76.12 (http://www.njp.org/)
[13] Gierak J, Mailly D, Hawkes P, Jede R, Bruchaus L, Bardotti L, Prevel B, Mélinon P, Perez A, Hyndman R, Ferré J, Mougın A, Jamet J P, Chappert C, Mathet V, Warin P and Chapman J 2002 J. Vac. Sci. Technol. at press

[14] Jensen P 1999 Rev. Mod. Phys. D 71 5

[15] Ziegler J F 1992 Handbook Ion Implantation Technology (Amsterdam: Elsevier)

[16] Bardotti L, Jensen P, Hoareau A, Treilleux M and Cabaud B 1995 Phys. Rev. Lett. 74 4694

[17] Lewis L, Jensen P, Combe N and Barrat J L 2000 Phys. Rev. B 61 16084

[18] Perez A, Dupuis V, Tuaillon-Combes J, jamet M, Négrier M P, Mélinon P, Wernsdorfer W, Barbara B and Thirion C 2001 Proc. Int. Symp. on Cluster Assembled Materials (IPAP Conf. Series 3) pp 34–8

[19] Binns C, Baker S H and Maher M J 2002 Phys. Status Solidi a 189 339

[20] Sun S and Murray C B 1999 J. Appl. Phys. 85 4325

[21] Combe N, Jensen P and Pinpinelli A 2000 Phys. Rev. Lett. 85 110