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J. MEIJER[1,✉]
S. PEZZAGNA[1]
T. VOGEL[1]
B. BURCHARD[1]
H.H. BUKOW[1]
I.W. RANGELOW[2]
Y. SAROV[2]
H. WIGGERS[3]
I. PLÜMEL[3]
F. JELEZKO[4]
J. WRACHTRUP[4]
F. SCHMIDT-KALER[5]
W. SCHNITZLER[5]
K. SINGER[5]

Towards the implanting of ions and positioning of nanoparticles with nm spatial resolution

[1] RUBION, Ruhr-Universität Bochum, 44780 Bochum, Germany
[2] Department of Micro- and Nanoelectronical Systems, Institute of Micro- and Nanoelectronics, Technical University of Ilmenau, PF 10 05 65, 98684 Ilmenau, Germany
[3] Institut für Verbrennung und Gasdynamik, Universität Duisburg-Essen, 47057 Duisburg, Germany
[4] 3. Physikalisches Institut, University Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany
[5] Institut für Quanteninformationsverarbeitung, University Ulm, Albert-Einstein-Allee, 89069 Ulm, Germany

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ABSTRACT Decreasing structure sizes in both conventional and quantum solid state devices require novel fabrication methods: we present a technology which allows to implant ions through a small hole in the tip of an atomic force microscope. This technique offers a maskless addressing of small structures using different projectiles at kinetic energies between 0.5 and 5.0 keV. Our method aims to implant single atomic ions, molecular ions or charged nanoparticles with nm resolution. We test the method by implanting N^+ ions into diamond and generating nitrogen-vacancy color centers. The system is operated with a conventional ion gun. However, in future we will employ an ion trap as a deterministic source of cold single ions.

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1 Introduction

Currently, structure sizes in modern nano-electronics reach a few tens of nm and we will certainly see a further miniaturization in the near future. Examples of conventional devices are nanotransistors where structure sizes of less than 30 nm are state of the art. Quantum solid state devices aiming for a future quantum computer also require novel fabrication methods. Examples of these quantum devices such as quantum dots, magnetic impurities, single embedded impurity ions or color centers in solid-state matrices are employed to store and process quantum information. So far, only rudimentary and non-scalable few-qubit processors based on phosphorus dopants have been investigated [1, 2]. Recently, qubit operation at room temperature has been demonstrated using nitrogen-vacancy (NV) centers in diamond crystals [3, 4], which are generated by nitrogen implantation.

✉ Fax: +40-234-32-14215, E-mail: jan.meijer@rub.de

The development of such structures is impeded by the lack of suitable nano-implantation methods. Required is a very high spatial resolution with low beam energy to avoid ion straggling in the substrate. Major challenges are the focusing and the precise alignment of the ion beam into the substrate. A mask technology might be a solution but the application below 5 nm is challenging. Only few groups worldwide are able to meet the technical requirements. A maskless technique such as focused ion beams (FIBs) has the advantage to be very flexible and to allow a quick modification of doping structures. FIBs with a spatial resolution of 3 nm are state of the art and commercially available. The advantage of the FIB as a maskless implanter is based on a liquid metal ion source (LMIS) with high brightness. Different LMISs have been developed and a variety of ions were successfully tested. However, the disadvantage of the LMIS is a large energy spread and the requirement of mass separation. Additionally, the energy spread leads to a chromatic aberration at low beam energies, which in turn spoils a narrow focusing with ion optical elements.

We introduce a new setup based on the combination of an atomic force microscope (AFM) with a collimator and a low-energy ion gun as a method to implant ions with high spatial resolution [5]. The advantage of an AFM collimator is that the implantation becomes independent of the ion-beam focus. The AFM tip easily allows an exact placement of the tip or collimator with an accuracy of better than 1 nm. This combination enables the separation of the ion beam positioning and addressing systems. Additionally, the method is easy to handle and allows using topological markers. Please note that it might be important to implant doping ions precisely with respect to e.g. an electrical control gate [6]. The ion gun can be equipped with a standard source thus allowing the production of a wide range of ion species, especially nitrogen or oxygen. The advantage of the present setup is that it can be combined with different types of sources. The paper gives some experimental details of the method, shows results with maskless

implanted N^+ ions in diamond and discusses the limits of this method. We sketch future improvements and extensions of the method.

2 Experimental setup

The use of a pierced AFM tip as a collimator for ion-beam implantation has been developed in a cooperation between Berkeley, Bochum and Ilmenau [7]. The method is based on a beam of low-energy ions or particles focused on the cantilever tip of an AFM system (Fig. 1). In the Bochum system an ion source (SPECS, model IQE12/38) can deliver different kinds of ions in an energy range between 0.5 and 5 keV. The source is equipped with a mass separator and an electrostatic lens to focus the ion beam. The beam is guided through a pierced mirror at 45° on which an optical microscope is pointed to achieve an in situ optical inspection. The microscope uses a long working distance objective and is connected to a high resolution and high sensitivity CCD camera outside the vacuum chamber. The resolution of the microscope is about 1 μm and the image covers about 700 μm × 700 μm. Focalization is obtained by moving the objective in a high-precision guide inside the vacuum chamber. The microscope allows an easy and precise alignment between the AFM tip and the beam. To reduce the beam spot size, we use an aperture-lens system close to the AFM cantilever. The ion lens demagnifies the image of an aperture with a diameter between 1 and 20 μm to the surface of the AFM tip. The AFM system (Alpha contact) is based on a piezoresistive cantilever with a hollow tip. On the apex of the tip a hole of some nm diameter is drilled[1] in order to act as a collimator. It is of great importance to drill the hole close to the apex and not into the cantilever in order to minimize the vertical distance between hole and target. A vertical distance of only 5 μm will broaden the beam by a factor of 5 or more depending on the divergence of the beam and scattering effects. Especially for small holes, scattering becomes quite dominant. The AFM enables the alignment of the hole within some nm. The AFM stage itself is positioned using a three-axis PI table (Physik Instrumente (PI) GmbH) with a resolution of 200 nm. Polymethylmethacrylate (PMMA) on silicon is used for the alignment procedure. Due to the incident beam the irradiated PMMA changes its color and the AFM stage can thus be moved to the indicated position. The piezoresistive positioning sensor system [8] allows a very compact AFM system with a resolution of about 1 nm. A single ion detector with a detection efficiency of more than 90% is used to measure the ion current through the AFM tip. The detector is covered by a 1-μm aperture in front of it to ensure that only ions having crossed the tip will be measured. The system is mounted in the piezo-driven table and allows routinely inspecting the hole in the AFM tip. The substrate is placed on a piezotranslation stage with a resolution of 0.1 nm and a moving range of 150 μm in horizontal and 20 μm in vertical directions. To extend the moving range, the setup is mounted on a second step motor driven translation stage with a resolution of 1 μm and a moving range of 11 cm in the horizontal direction. Figure 2 gives a view of the setup inside the vacuum chamber.

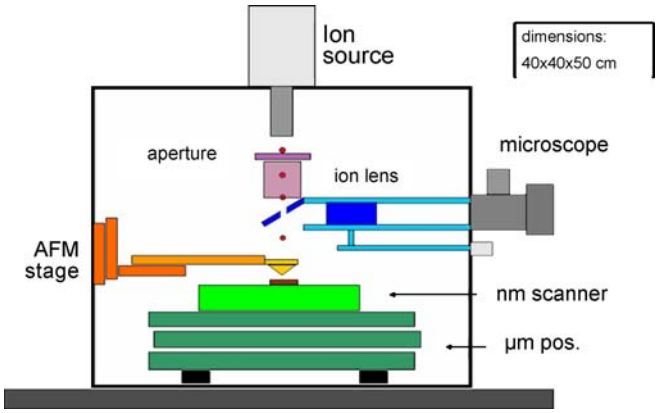


FIGURE 1 From *top* to *bottom*: the ion gun providing different kinds of ions, a micro ion lens with apertures, an optical microscope with a pierced mirror and a table with sub-nm resolution. The pierced AFM tip can be moved precisely in all directions using micromotors. The target is based on a PI piezo table with 0.1-nm resolution and a stepping motor driven table for μm alignment

2.1 *AFM-tip-based nanocollimator*

AFM devices based on Wheatstone-bridge piezoresistive cantilevers allow for many applications. The integration of the piezoresistive readout provides the best solution to realize a very compact high-resolution AFM, attractive as compared to the conventional optical beam deflection technique especially in those cases where laser detection may not fit the overall system. Piezoresistive cantilevers are irreplaceable in such areas as high vacuum, AFM arrays of cantilevers and high-frequency small cantilevers and in applications such as microbalances, infrared radiation detection or cryogenic conditions. The extreme sensitivity and short response times of micromechanical cantilevers used for nanoscale AFMs have been extended beyond those of a surface-imaging tool. In previous publications [9, 10], we have demonstrated that the minimum detectable deflection for piezoresistive cantilevers can be as low as 0.01 nm for a 1-kHz bandwidth, confirming that this detection scheme is adequate for AFM operation. Piezoresistive cantilevers with integrated and glued hollow tips were microfabricated using standard CMOS processing and double-sided micromachining techniques. The glued hollow tips are based on SiN and are produced by standard moulding techniques. The thickness of the hollow tips is about 150 nm. The hollow AFM tips with a drilled hole are fabricated by FIB milling with Ga ions at 30 keV. Figure 3 shows a drilled hollow tip glued onto a cantilever. A Faraday cup behind the AFM tip is used to turn off the beam just at the moment when the transmission current indicates that a hole was drilled. The entrance of a typical hole is about 140 nm, the exit in the range of 30 nm or below [5]. The AFM operation with resistive cantilevers allows imaging the surface in contact mode with a resolution in z better than 1 nm. A similar result is achieved for a tip pierced at the apex.

We studied the lifetime of the AFM tips with drilled holes and realized that the holes tend to become closed with increas-

[1] Drilling has been performed by the FIB installation of Institut für Angewandte Festkoerperphysik, Ruhr-Universität Bochum (Prof. A. Wieck). We want to thank Dr. Melnikow for his assistance.

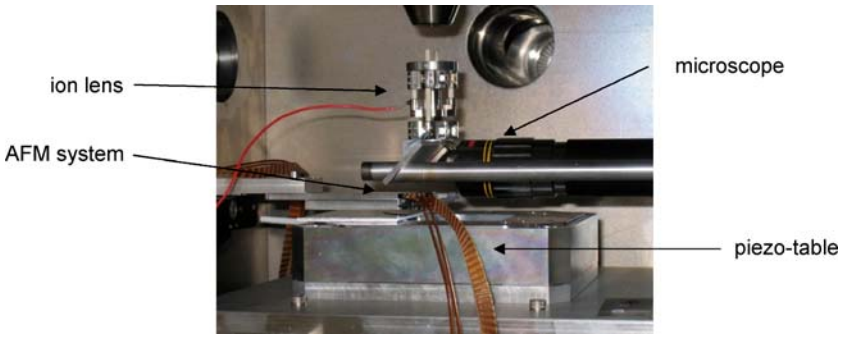


FIGURE 2 A view inside the nano-addressing chamber

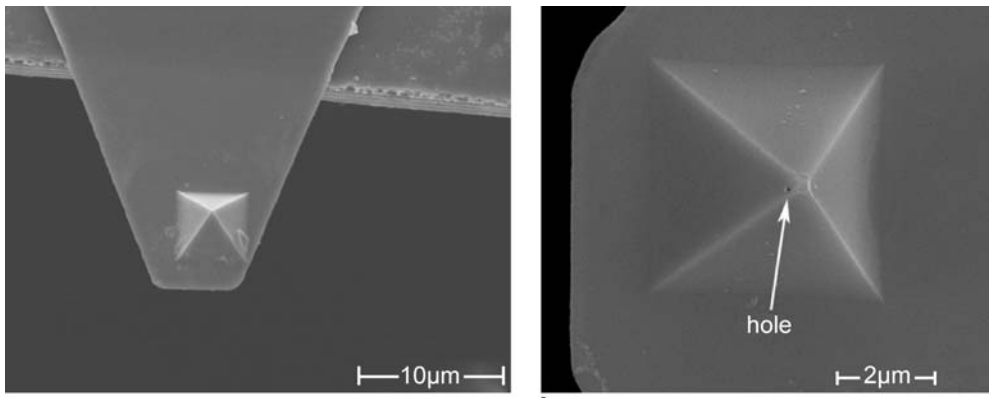


FIGURE 3 (**a**) Glued hollow AFM tip with piezoresistive cantilevers. (**b**) Hole drilled into AFM tip using a focused ion beam (FIB)

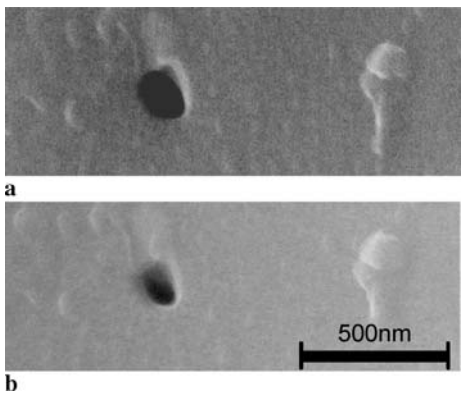


FIGURE 4 (**a**) Hole in the AFM tip, before ion-implantation procedure; (**b**) after ion-implantation procedure of Ar and N with a dose of 4×10^{16} ions/cm^2

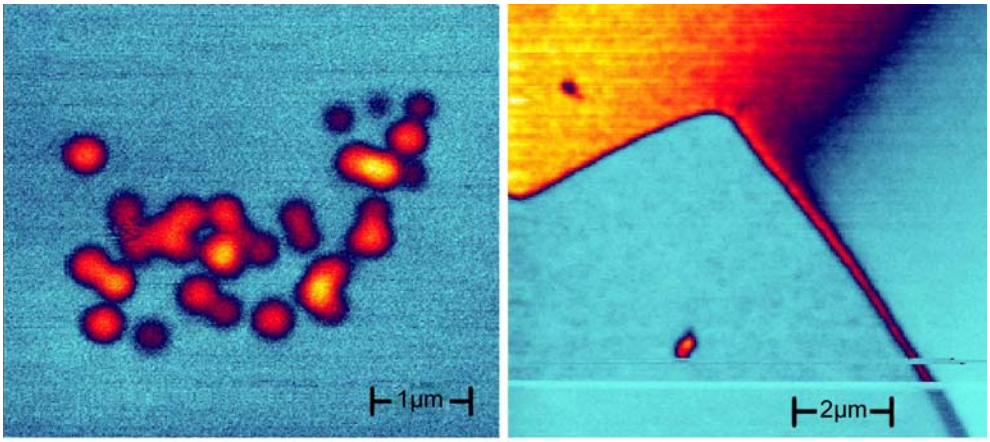
ing ion exposure (see Fig. 4). Under typical working conditions the drilled tip can be used for several hours. Whether this effect depends on the cone of the FIB drilled hole (i.e. from which side the hole is drilled into the tip) is currently being investigated. First results also show a strong dependence of this effect on the impact angle of the ion beam. The hole-size reduction under ion-beam exposure could be exploited for manufacturing holes smaller than the limit of state of the art FIB milling devices.

3 Application: production of NV centers

Our goal is the production of quantum devices by implantation of single nitrogen ions. The long coherence time of nitrogen-vacancy (NV) centers of ~ 0.6 ms for the electron spin even at room temperature allows studying quantum effects [11, 12]. We showed the fabrication of single NV centers using implantation of N^+ [3]. A mutual dipole–dipole coupling of nearby NV centers has also been shown [4]. Color centers find applications as single-photon sources [13, 14] (and for cavity quantum electrodynamic experiments with micro-optical resonators [15]) or as ultra-sensitive magnetometers.

Conditions to build devices of this sort are challenging: implanting within a lateral accuracy of a few nm may be possible when the diameter of the hole inside the AFM tip is reduced to this size. It remains to be shown that the hole does not become clogged, and that proximity effects do not spoil the spatial resolution. The second question is the yield of implanted N^+ ions into NV centers, which should be as high as possible. Experiments with ion implantation at high energy (2 MeV) showed a yield of 20 to 100%, which is reduced for low-energy implantation to below 2.5% [16]. Figure 5a shows a picture of the fluorescence from a single NV center fabricated by $^{15}N^+$ implantation at high energy using a microprobe system [17] and low kinetic energy using our nano-addressing tool through an AFM tip (Fig. 5b). The implantation is performed with $^{15}N^+$ to distinguish the implanted ions from natural ^{14}N-isotope content of diamond samples. The depth of the vacancy production plays an important role in the NV center production process, when the sample is annealed at a temperature of 800 K. We speculate that N^+ ions near the surface may not easily combine with a vacancy as they are trapped at the surface during annealing. Thus, they are no longer available to produce NV centers.

However, work on novel techniques to enlarge the efficiency by co-implantation and electron and photon irradiation is under way. Also, the use of other color centers or nickel–



a b

FIGURE 5 (**a**) A group of NV centers produced by MeV implantation using a nanoprobe implantation system. (**b**) A NV center produced by the nano-addressing system with an ion gun having an ion kinetic energy of 2 keV

nitrogen complexes [18] seems to be a possible way since they do not need an additional vacancy.

4 Future developments: deterministic ion implantation and nanoparticle addressing

Typically, solid-state devices are doped in order to tailor their electrical properties. Once the devices reach dimensions such that the Poissonian number fluctuations of dopants become too high, their operation will be largely affected. For example, with a typical doping concentration of 10^{15} /cm^3, the gate region of a nano-FET contains only 30 doping atoms such that the relative number fluctuation reaches 20%. For quantum devices, e.g. single embedded ions carrying a nuclear spin to encode quantum bits, it is essential to reach the absolute control of the number of dopants.

One may detect the event of the single-ion impact: the detection of secondary electrons works in an efficient way only for energies above 30 keV. At low energies, highly charged ions can be used as the impact generates an electron shower [19], but these ions may cause severe damage to the surface [20]. In some cases, it is possible to measure electron–hole pairs from a single ion impact in semiconductors, if prepared as a Si-diode structure [21]. However, the method is restricted to dedicated structures and materials. Our method for a single-ion source observes the dopant ion prior to the implantation by laser light fluorescence in a Paul trap. A single ion can be identified and manipulated such that its temperature is reduced to a fraction of a mK, corresponding to 10^{-7} eV. In turn, chromatic aberration is expected to be largely reduced [3]. The method is universal, and should work for almost all types of atomic ions.

The advantage of the presented system is that almost all types of sources can be used. Beside ions, nanoparticles as well as molecules can be collimated.

Especially, a singly charged nanoparticle could be produced inside a plasma reactor combined with a velocity measurement and size-selecting system [22]. This source is now available and its combination with highly resolved deposition is in preparation. The aim of the project 'Cluster jet' is the development of a technique that allows for the addressing of countable nanoparticles with a spatial resolution of 10 nm or less to a pre-structured surface.

The nanoparticle beam will be transferred to the AFM chamber using a tube and ion lenses. This horizontal beam will be connected to the focusing unit using a 90° bending system. The bending system will be installed between the ion source and the ion optical elements (scanning units, lenses, quadruplet systems, etc.). We expect that the pre-alignment procedure of the optical components will simplify the adjustment of the nanoparticle beam. For a particle beam a degeneration of the tip hole due to sputtering effects is not expected. Nevertheless, the holes may also become clogged by the nanoparticles themselves. If this happens, an additional µ-lens will be necessary. However, a ray-tracing calculation shows that a focusing of nanoparticles below 50 nm is possible at the given conditions.

5 Summary

We presented a new technique that allows maskless implantation of single ions at low kinetic energy with nm resolution. The method is based on a pierced AFM tip that acts as a collimator system in the nm range. The spatial resolution is given by the size of the hole. Holes with 30-nm diameter can be fabricated routinely. As a result of our first experiments, the size of the holes became smaller during the irradiation with 2- to 5-keV ions. This effect may be useful to deliberately reduce the diameter. The system allows the implantation of ions with an kinetic energy of 500 eV produced by a conventional ion source. The combination of an AFM table and a positioning table using stepper motors enables an easy handling for different applications and an addressing within 1-nm distance to the target. One application is the implantation of N^+ ions to produce NV centers in diamond. Experiments showed that the efficiency of NV production needs to be optimized. To implant a pre-determined number of ions, the method is currently improved by using an ion trap as cold source. In addition, the system can be equipped with a nanoparticle source for implanting nanoparticles with high spatial resolution.

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