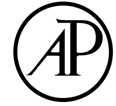
Superlattices and Microstructures, Vol. 20, No. 3, 1996



Nanolithography with an atomic force microscope

M. Wendel, B. Irmer, J. Cortes, R Kaiser,
H. Lorenz, J. P. Kotthaus, A. Lorke
Sektion Physik, Ludwig-Maximilians-Universität München, 80539 München, Germany

E. Williams
IBM Zürich Research Laboratory, 8803 Rüschlikon, Switzerland

(Received 20 May 1996)

A lithographic technique, employing the vibrating tip of an atomic force microscope to mechanically pattern various materials such as photoresist, metals or semiconductors in the nanometre regime has been developed. We use this technique for the fabrication of etch masks as well as for the patterning of evaporation shadow masks. The tip quality has been found to be a crucial factor in the lithographic resolution. We therefore use ultra hard, amorphous carbon tips, which are prepared by electron beam deposition in an electron microscope. With these tips, additionally sharpened in an oxygen plasma, we now succeed in fabricating hole arrays with periods in the 10 nm regime. These hole arrays are transferred to the electron system of a GaAs–AlGaAs heterostructure, and the magneto resistance of such fabricated antidot arrays is discussed.

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In spite of being very young, lithographic techniques employing scanning probe microscopes have already brought substantial progress in the miniaturization of semiconductor devices. A review of various lithographic methods is given for example in Ref. [1]. Lithographic methods using scanning probe techniques can be divided into two categories. On the one hand field or current induced surface modifications are achieved by applying a voltage to a conducting tip of a scanning tunnelling microscope (STM) or an atomic force microscope (AFM) with respect to the substrate. On the other hand there are techniques by which the sample surface is mechanically modified. These latter lithographic methods have the advantage that they are not restricted to conductive materials.

Recently we have introduced a technique to mechanically pattern various materials with the vibrating tip of an atomic force microscope [2]. Our aim is to use this method to laterally confine the electron system of a GaAs–AlGaAs heterostructure or of an InAs–GaSb quantum well on a length scale in the nanometre range. To do this we have developed three different techniques: the patterning of a surface mask consisting of photoresist or metal, the direct lithography of the semiconductor material or the fabrication of an evaporation shadow mask.

To create a photoresist mask on a semiconductor device we first spin a highly thinned photoresist layer

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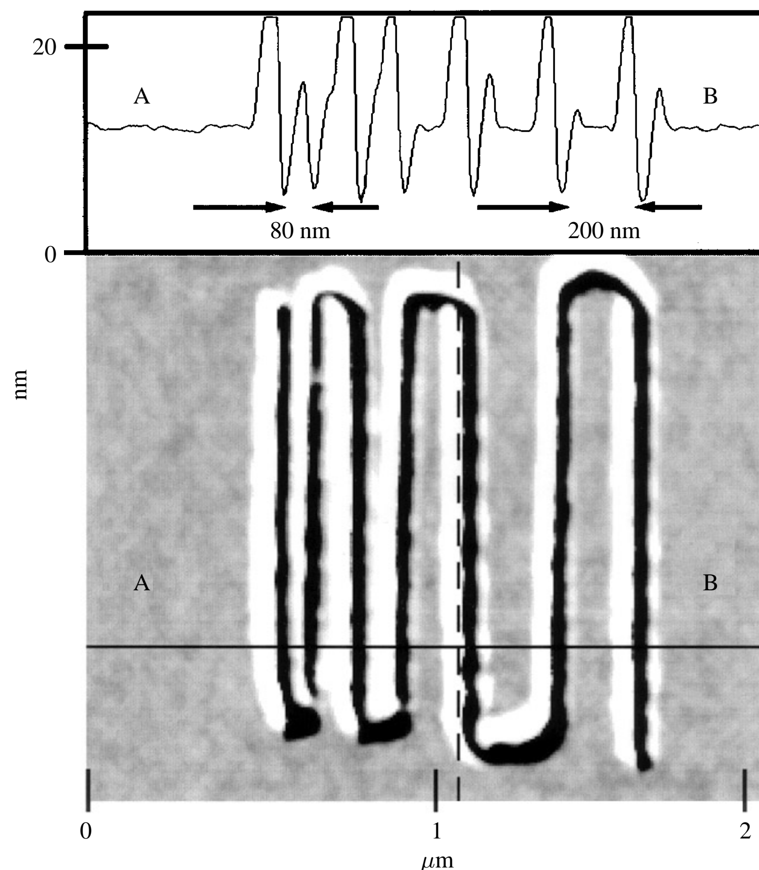

Fig. 1. AFM micrograph of a meander-like structure in photoresist, fabricated by AFM-lithography. The lines consist of thousands of overlapping single holes. Top: Section plot along the indicated line.

(Shipley 1805: thinner = 1: 70–1:15) on a conventionally prefabricated hall bar device, resulting in a film thickness of about 3–15 nm. To reduce the viscosity to an optimal value, the resist is baked at 120–170 °C for from 30 minutes to 5 h. In the lithographic step a software controlled silicon tip, vibrating at its resonance frequency of about 200–300 kHz with an amplitude of some 10 nm, is pushed towards the surface to structure the underlying material. To create holes and lines in photoresist we apply a triangular voltage pulse of about 0.1 second duration to the z-piezo. The estimated maximal force is about 1 μN. Metals such as gold, lead or aluminium need a significantly higher force in the region of several μN. An advantage of the vibration in the so-called tapping mode is that very small lateral forces stress the tips, resulting in a very slow tip degradation. Figure 1 represents a meander-like structure in photoresist, consisting of thousands of overlapping single holes. The line width is about 25 nm (FWHM), the period decreases from 200 nm (right) to 80 nm (left). The section plot indicates that the depth of the groove is the same as the resist thickness.

In testing the lithographic resolution of this technique one recognizes that this method fails in producing structures with periods far below 80–100 nm. The limiting factor is the tip quality. Sharp silicon tips [3] deliver brilliant and reproducible results in the lithography as long as the minimal period is not smaller than this value. Smaller periods are not achievable because of a slight flattening of the brittle tip during the lithographic step. We have found that sharp electron-beam-deposited tips (EBD tips) can overcome this problem [4]. EBD tips

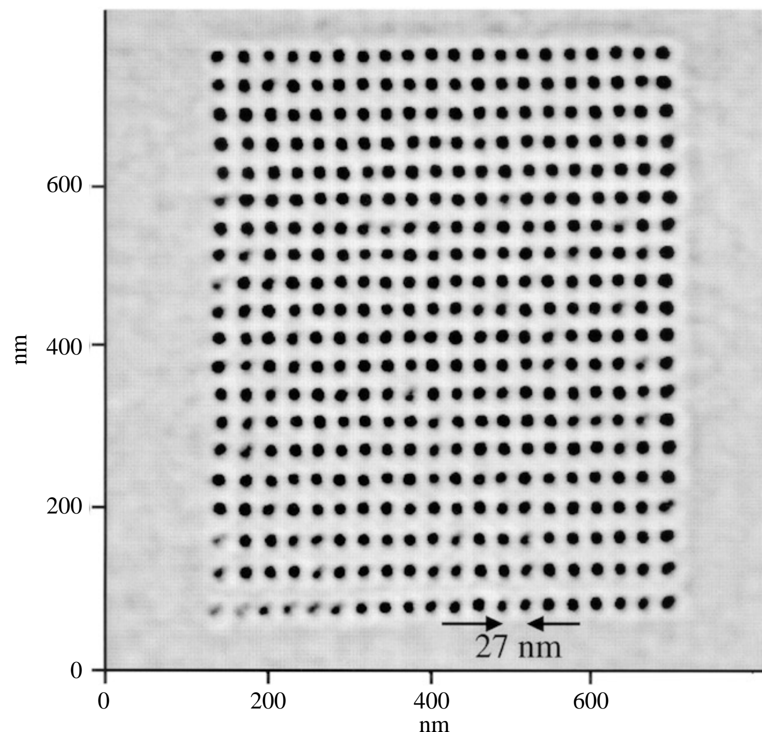

Fig. 2. AFM micrograph of a hole array in photoresist, fabricated with an EBD tip. Period and depth of the holes are 27 nm and 3 nm, respectively.

are extremely hard and durable and do not degrade significantly even after the production more than 100 000 holes in photoresist. Figure 2 depicts an array of holes in a 4 nm thick photoresist layer fabricated by an EBD tip. The period and depth of the holes are 27 nm and 3 nm, respectively. The smallest hole periods we succeeded in fabricating in photoresist until now with an EBD tip are 15 nm.

To even further reduce the lithographic structure size, the EBD tips can be additionally sharpened in an oxygen plasma [4]. Figure 3 shows a transmission electron micrograph of unsharpened (A), and a slightly sharpened EBD tip (B). A high resolution micrograph of the end of the sharpened tip (C) indicates a tip radius of about 5 nm. The dominant central spot in the diffraction picture implies an almost amorphous substance (Fig. 3D). A small microcrystalline part is reflected by several small diffraction peaks (arrows). Energy dispersive X-ray analysis shows that EBD tips consist mainly of carbon and possibly a very small amount of oxygen. Hydrogen can not be detected by this method. Figure 4 shows an array of holes, fabricated by a sharpened EBD tip. To illustrate better the shape of the holes, the picture is inverted so that the holes appear as mounds. The period of the holes is 16 nm. The smallest period we have fabricated up to now is 9 nm.

A method to pattern an electron system below the surface without preparing a mask on the surface is the direct ‘writing’ on a semiconductor surface. This technique has the advantage of working without the additional step of transferring the mask to the electron system. Figure 5 depicts an AFM micrograph of an InAs–GaSb quantum well with the electron system 30 nm below the surface. The 3 nm thick cap layer of this sample consists of InAs. This comparatively soft material can be removed directly during the AFM lithographic process. By removing this layer the electron density of the quantum well is increased about 40% [5]. The first electronic investigations of such fabricated devices are in progress.

The third possibility to employ AFM lithography in the patterning of a two-dimensional electron gas is the

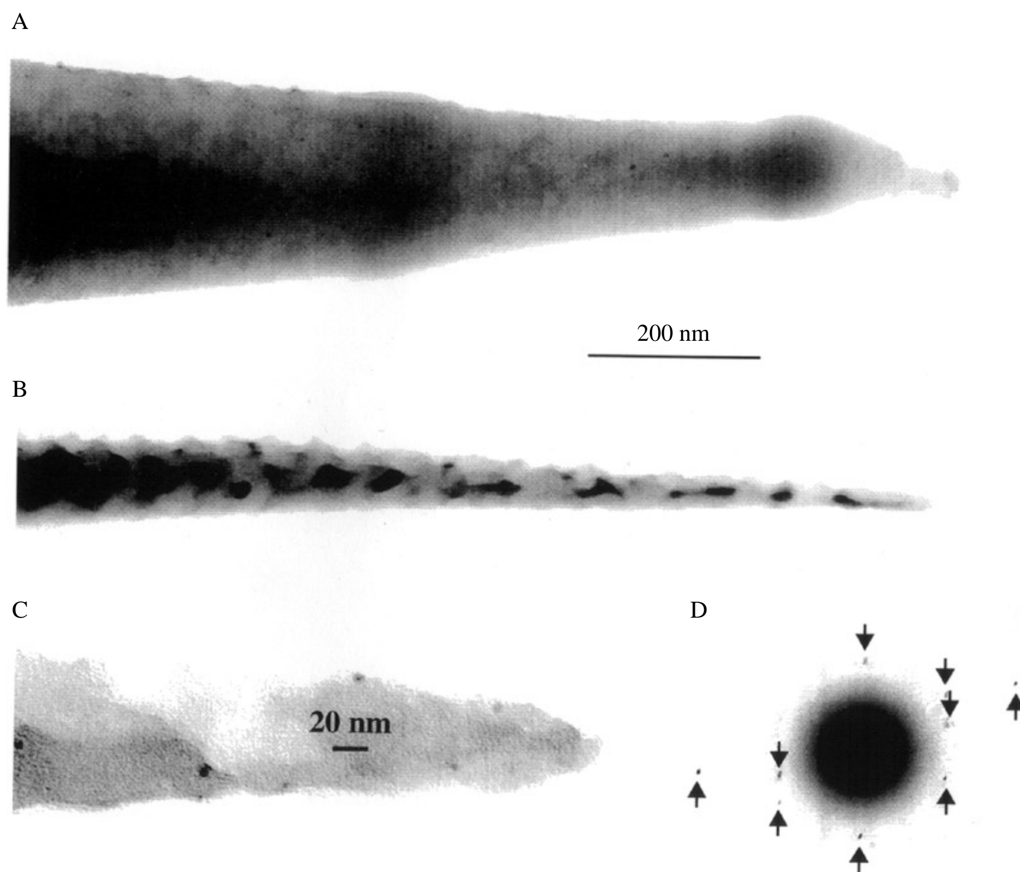

Fig. 3. Transmission electron micrograph of an unsharpened electron beam deposited tip (A) and a slightly sharpened tip (B). C, a high resolution picture of the end of the sharpened tip. The diffraction pattern indicates an amorphous structure with very small crystalline parts (D).

fabrication of an evaporation mask. A related AFM basing lift-off technique has been presented recently [6]. Figure 6 depicts schematically the preparation steps. The sample surface is coated by a several hundred nm thick photoresist layer, followed by a thin (of the order of 10 nm) gold or chromium film and a thin (about 10 nm) photoresist layer. In the first step the thin photoresist layer is structured by AFM-lithography, while the harder metal film is not affected (A). Afterwards the metal film is etched through the holes in the photoresist. We use a thinned KI solution to remove gold and a thinned solution of $KFe(CN)_6$ as a chromium etchant. The thick photoresist film underneath is removed in the unprotected areas by an oxygen plasma (B). We then evaporate an arbitrary material through the holes in the metal film (C(i)). Evaporating a second time through this shadow mask after tilting the sample duplicates the pattern and creates a dot array with a complex base (C(ii)). By carefully selecting the tilting angle it is even possible to reduce the period of the mask. In the final step the evaporation mask is removed in hot acetone (D). Figure 7 depicts an AFM micrograph of an array of NiCr dots, deposited through one of the first such fabricated evaporation masks, consisting of a 10 nm thick gold film. Period and diameter of the dots are 450 nm and 30 nm, respectively, and the thickness is about 15 nm. The reduction to smaller periods and the fabrication of a complex base is in progress.

To pattern the two-dimensional electron system of a GaAs–AlGaAs heterostructure or of a InAs–GaSb

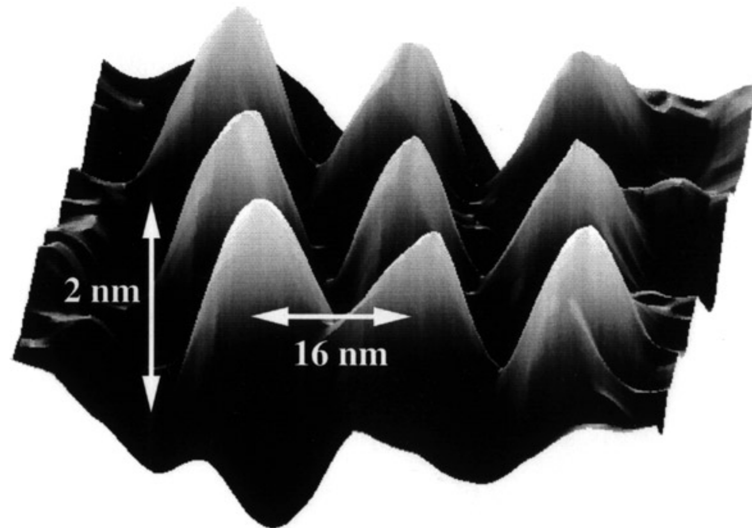

Fig. 4. Hole array in photoresist with 16 nm period and 2 nm depth. To better illustrate the shape of the holes, the AFM picture is inverted, the holes therefore appear as mounds.

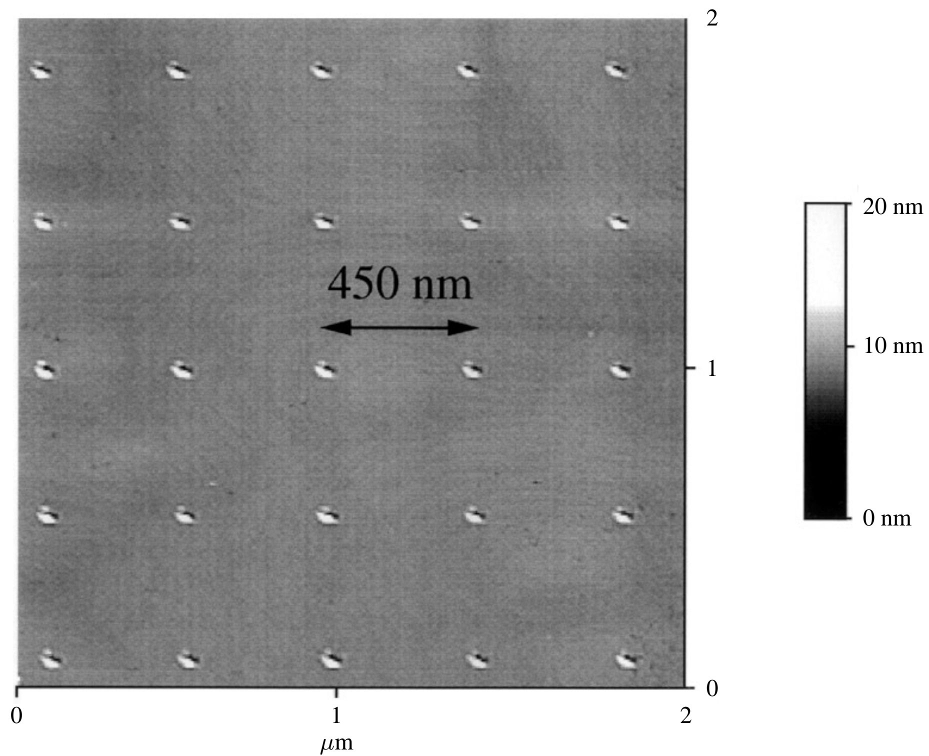

Fig. 5. Holes, fabricated directly in an InAs cap layer of an InAs/GaSb quantum well. The hole period and the depth are 450 nm and 3 nm, respectively. The holes totally penetrate the InAs cap layer.

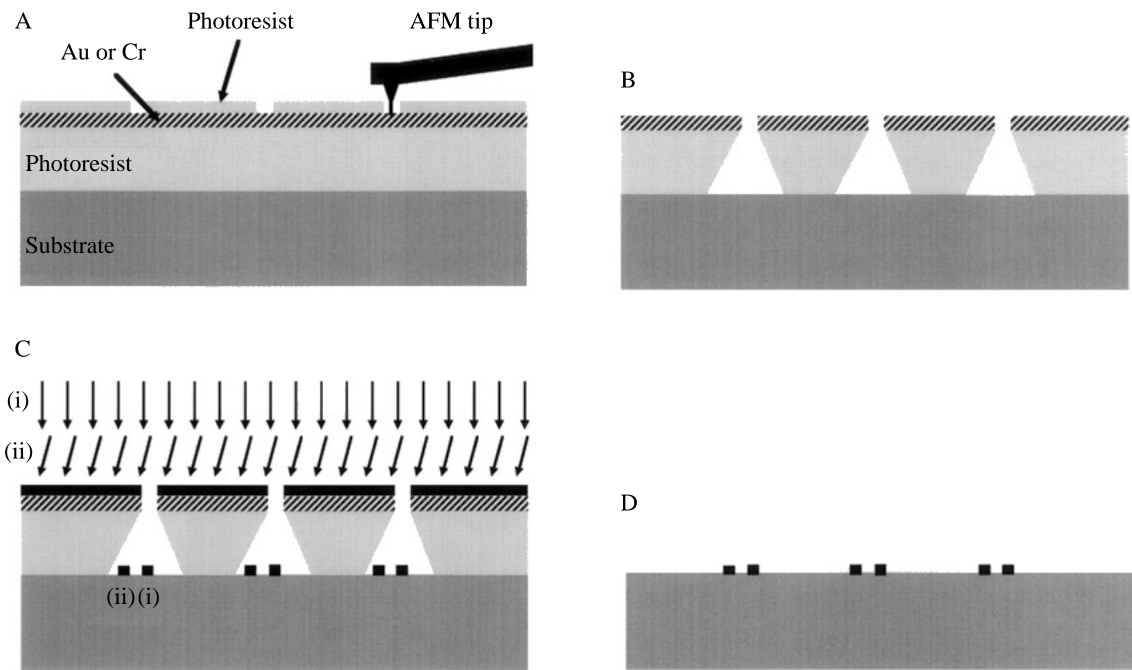

Fig. 6. Schematic plot of the fabrication sequence for an evaporation shadow mask for lift-off purposes. A, The upper photoresist film of a multilayer consisting of thick photoresist, thin gold or chromium and a thin photoresist film is patterned by AFM-lithography. B, The metal layer is etched through the holes and the thick resist film is removed in an oxygen plasma. C, Evaporation through the holes (i). A second evaporation step with a tilted sample leads to a duplication of the pattern (ii). D, Removal of the evaporation mask.

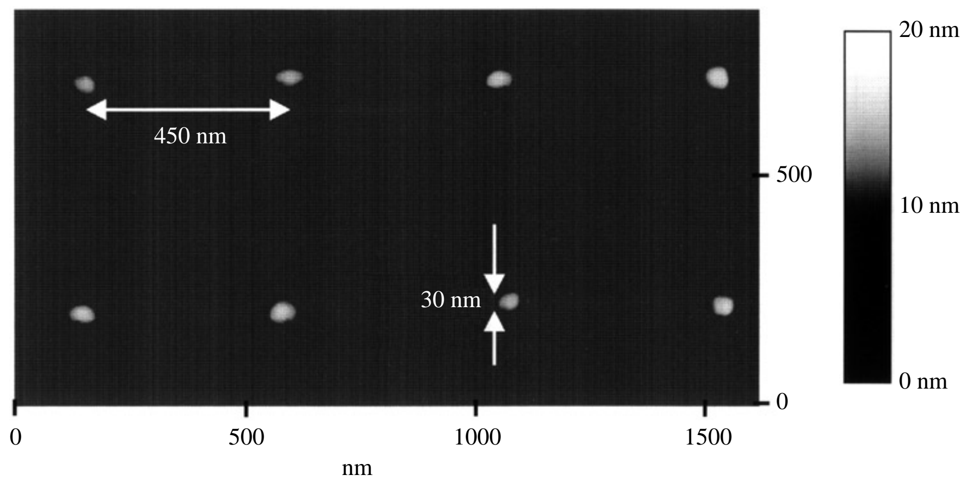

Fig. 7. AFM micrograph of a dot array deposited through an evaporation shadow mask consisting of 10 nm thick gold. Period and diameter of the NiCr dots are 450 nm and 30–35 nm, respectively. The height is about 15 nm.

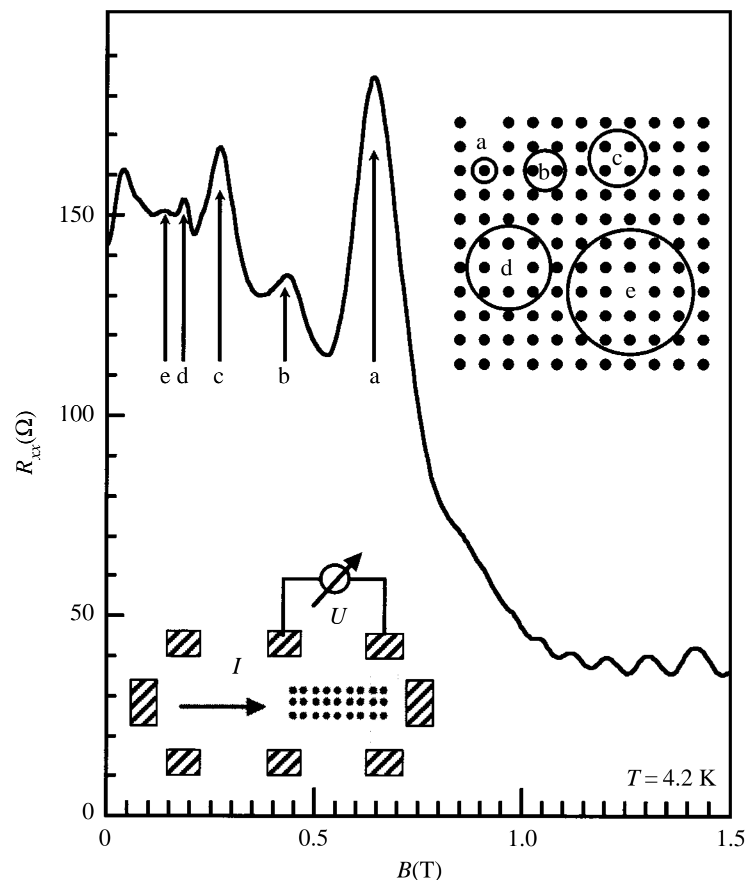

Fig. 8. Magneto-resistance of an AFM fabricated antidot array with 450 nm period. Several well pronounced maxima, the so-called commensurability oscillations, can be detected (arrows).

quantum well, the mask, fabricated by structuring a surface layer or by an evaporation mask, has to be transferred to the electron system. To do this we either use a wet etch step or the ion beam irradiation technique (IBI). In IBI, accelerated neon ions deplete the two-dimensional electron system of a heterostructure below unprotected surface areas. This method has the advantage of creating very small lateral depletion lengths of less than 20 nm. Details of this technique are published elsewhere [7].

Figure 8 shows the magneto-resistance of a hole array, transferred to the electron system by etching 80 seconds in the basic solution $H_2O : NH_4OH : H_2O_2 = 1000 : 3 : 1$. The period of this so-called antidot arrays is 450 nm, the topographic diameter of the holes is about 80 nm, the depth is 30 nm. One clearly recognizes a series of maxima in the low field magneto-resistance of the antidot array, which cannot be seen in unstructured samples and clearly indicates the successful transfer of the mask pattern to the electron system. These maxima were first observed and explained by Lorke [8] and Weiss [9]. They are seen at magnetic fields where the cyclotron orbit of the electrons is commensurate with the hole array. The arrows indicate these commensurability conditions and the orbits which lead to the increased resistance are also plotted. Detailed investigations on such antidot arrays with a systematically varied complex base give new information about the electron transport through the device and will be published elsewhere [10].

Antidot arrays with smaller periods defined by AFM lithography have been transferred to the electron system

by the IBI technique. Up to now we have investigated arrays with periods down to 85 nm [11]. Although the period of these antidot arrays is of the order of the Fermi wavelength, the position of the commensurability maxima in the magnetic field can still be explained by the classical picture. The fabrication of even smaller periods and the investigation of these devices at very low temperatures of $T = 25$ mK are in progress.

In conclusion we describe a technique using the vibrating tip of an atomic force microscope for lithographic purposes. By this method we have fabricated etch and evaporation masks for semiconductor devices or directly structured the semiconductor material. The lithographic resolution can be significantly increased by the use of electron beam deposited tips. With these tips, additionally sharpened in an oxygen plasma, we have fabricated hole arrays in photoresist with periods down to 9 nm. The lithographic defined masks are transferred to the electron system by wet etching or by ion beam irradiation. The magneto-resistance of such fabricated antidot arrays is discussed.

Acknowledgement—Financial support from the Volkswagenwerk-Stiftung is gratefully acknowledged.

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