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The *Atom Pencil*: Serial Writing in the Sub-Micrometer Domain

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The *Atom Pencil* we describe here, is a new tool that writes arbitrary structures by atomic deposition in a serial lithographic process. This device consists of a transversely laser-cooled and collimated cesium atomic beam that passes through a 4-pole atom flux concentrator and impinges onto micron and sub-micron sized apertures. The aperture translates above a fixed substrate and enables the writing of sharp features with sizes down to 280 nm and a fractional depth gradient of $\simeq 0.8/20\,\mathrm{nm}$. We have investigated the writing and clogging properties of an *Atom Pencil* tip fabricated from silicon oxide pyramids perforated at the tip apex with a submicron aperture.

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Atom lithography aims to fabricate lateral structures on a micro- and nanometer scale through controlled motion of atoms [1, 2]. In this method the structure arises from transverse density modulation of an atomic beam induced by the standing wave intensity pattern of an interfering light beam [3]. The density distribution is then transferred to a suitable surface.

The methods of atomic nanofabrication (ANF) [4] were introduced as a convenient method for direct-write parallel structuring. Periodic arrays of lines and dots have been realized in two dimensions (2D) [5]. Even more complex structures have been fabricated [6, 7] although at the price of complicated and fixed mask designs for each individual structure.

We present here a more versatile *serial* writing method in which atoms deposit locally through a translating aperture onto a fixed substrate just below it. This *Atom Pencil* writes arbitrary 2D structures through direct deposition of atoms, with low-energy impact. Although shadow-mask deposition through submicron apertures was first reported by Lüthi et al. [8], their approach suffers from a large divergence of the beam emanating from a thermal effusive source.

In the present experiment we show that a transversely laser cooled atomic beam [3] effectively removes this drawback by reducing atom flux divergence to below 1 mrad maintaining flux density as the atoms travel from source to target. As a second consequence of laser cooling, concentration of the atomic beam by an axial magnetic 4-pole results in further and significant enhancement of the atom flux density at the aperture.

A magnetic 4-pole acts as the analogue of an optical axicon and generates a longitudinal focal line by deflecting atoms with magnetic moment μ through the Stern-Gerlach force in an inhomogeneous magnetic field with gradient $\partial_r|B|$. The corresponding radial acceleration is given by $a_4 = \mu \cdot \partial_r|B|/M$ where M is the atomic mass. For an estimate of the characteristic focal length z_4 we calculate the longitudinal position at which an atom with average longitudinal velocity v_{th} and initial radial position R corresponding to the half-width of the thermal atomic beam crosses the axis. We find $z_4^{\mathrm{th}} = R\,v_{\mathrm{th}}^2/(a_4\,L)$, where L is the length of the 4-pole. The narrow radial width of the focal line expected for a perfectly collimated atomic beam is blurred by the finite divergence of the incoming laser-cooled beam, $\alpha_{\mathrm{div}} \simeq 1\,\mathrm{mrad}$. For estimating the on-axis flux density we thus calculate the average flux into an area $\pi(\alpha_{\mathrm{div}}\,z)^2$; and, averaging over the thermal longitudinal velocity distribution, find $F_4 = C \cdot \exp\left(-z/z_4^{\mathrm{th}}\right)$ where C is a constant. This result indicates that the *Atom Pencil* apertures should be mounted close to the exit of the 4-pole. Numerical trajectory simulations have verified our analytic model.

The setup of the *Atom Pencil* is illustrated in Fig. 1. The cesium atomic beam emanating from an effusive oven at a temperature of 140°C is transversely collimated by optical molasses [3] resulting in an average transverse velocity of $\simeq 6\,\mathrm{cm\,s^{-1}}$, or $\simeq 1\,\mathrm{mrad}$ divergence, and a Gaussian beam density profile with 1 mm full-width at half-maximum (FWHM). By optical pumping a spin polarization of better than 95% in the $|6S_{1/2}, \mathrm{F}=4, \mathrm{m_F}=4\rangle$ quantum state is obtained. The magnetic 4-pole has a 50 mm diameter with an inner bore of 10 mm. It is

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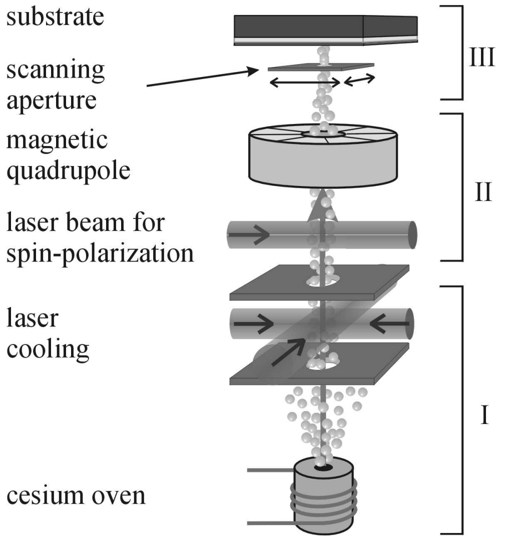

FIG. 1: The *Atom Pencil* consists of a transversely collimated atomic beam (I) which after a concentration stage (II) is deposited through a pin-hole onto a substrate (III).

20 mm long and provides a field gradient of 2.81 T/cm in the radial direction [9].

The thermal velocity distribution leads to a focal line of about 100 mm length for the quadrupole. At 65 mm separation from the center of the 4-pole we find a typical flux density enhancement of a factor of 37, corresponding to a typical flux density of $10^{14}\,\mathrm{cm}^{-2}\mathrm{s}^{-1}$. The divergence at this position is 8 mrad, the atomic beam FWHM is 45 μm.

It is interesting to note that the 4-pole is superior to a 6-pole, the analogue of an optical lens and a natural candidate for focusing applications [10]. However, the thermal velocity distribution causes strong degradation of the focusing properties. A calculation analogous to the 4-pole case for the on-axis flux density yields $F_6 = C \cdot z z_4^{\mathrm{th}}/(z_6^{\mathrm{th}})^2 \exp(-z/z_6^{\mathrm{th}})$. The characteristic focal length of a 6-pole constructed from the same material is now $z_6^{\mathrm{th}} \simeq 6 z_4^{\mathrm{th}}$. Comparing F_6 and F_4 the 4-pole concentration exceeds the 6-pole concentration by more than an order of magnitude for $z < z_4^{\mathrm{th}}$. These properties are summarized and plotted in Fig. 2.

In this study we used a cesium atomic beam and a lithographic process in which the cesium modifies the chemical properties of a nonanethiol self-assembled monolayer (SAM) resist [11, 12]. After exposure, chemical wet etching transfers the structure into the gold layer supporting the resist. Systematic studies have determined the writing efficiency of Cs flux on a nonanethiol SAM and determined the optimum conditions for the subsequent chemical wet etch process [13]. For the first experiments with the concentrated atomic beam we used micron sized stainless steel apertures. A simple structure written by stepping the apertures across the concentration maximum is shown in Fig. 3(a). The writing time per dot was 180-200 s. The structure size of 2.3 μm is in accordance with the aperture-substrate separation of 200 μm and the residual atomic beam divergence. In Fig. 3(b) we show a similar structure written with a 400 nm aperture and 200 s exposure time per dot. From the spot size of the written structure we infer an aperture substrate separation of about 12 μm. In present experiments our key element for writing submicron structures is a miniaturized aperture integrated into a hollow pyramidal tip. Figure 4 shows a typical structure. Pyramidal tip fabrication is based on two etch processes, a chemical etch to form the pyramid and a plasma etch to form the aperture. Details are described in [14, 15].

An important consideration for practical application of the *Atom Pencil* is the writing rate vs. the clogging rate of the tip. We investigated the clogging rate of a pyramidal tip mask similar to the one in Fig. 4 with a second transversely cooled and collimated (but not concentrated) cesium atomic beam (divergence $\simeq 1\,\mathrm{mrad}$) but with flux density of $\simeq 10^{12}\ \mathrm{cm}^{-2}\ \mathrm{s}^{-1}$, two orders of magnitude less than the flux-concentrated beam. A SAM-covered substrate was exposed to the cesium flux for 15 minute intervals up to one hour and the spot size after the etching process was measured by atomic force microscopy (AFM). The clogging rate was measured with the pyramid principal axis aligned along the atomic beam Cs flux and with the tip pointing toward the atomic beam source. The tip was $16\,\mu$m above the SAM surface and the aperture diameter was 300 nm. Figure 5 (a),(b) shows AFM images of the etched SAM and line profiles, respectively. The images and line profiles clearly show that the aperture significantly closes after 30 minutes exposure; and, after taking successive Cs flux exposures, AFM images and line profiles for up to one hour, we find the average clogging rate is $\simeq 6\,\mathrm{nm\,min}^{-1}$. We have also carried out clogging measurements for the pyramidal aperture

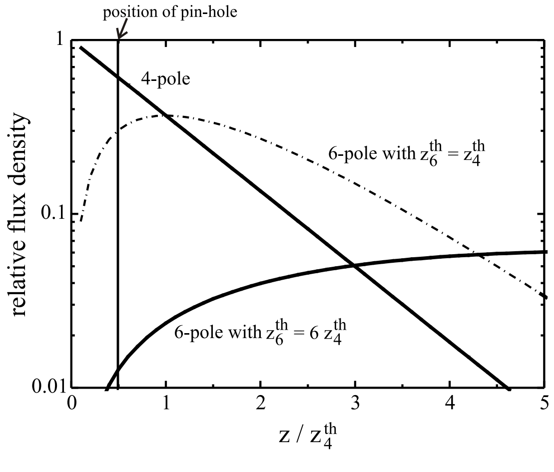

FIG. 2: Relative longitudinal on-axis flux density for a thermal atomic beam focused by a magnetic 4-pole, a magnetic 6-pole with short focal length (upper curve), and more realistic long focal length (lower curve). The pinhole is typically positioned at $z \simeq 0.5\, z_4^{\mathrm{th}} = 65\,\mathrm{mm}$ from the center of the 4-pole.

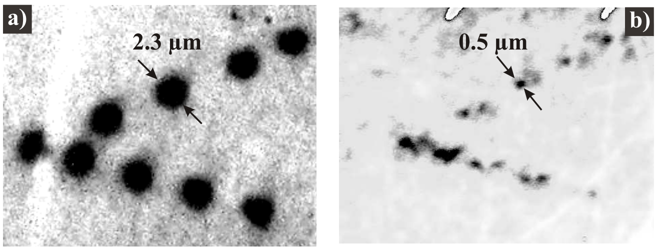

FIG. 3: Examples of structures consisting of a V-shaped stepped series of dots written with apertures of two different diameters: (a) 1 μm, (b) 400 nm. In (b) the distance aperture–substrate was well below 200 μm and the exposure time per dot was 180-200 s.

oriented in the opposite direction so that the tip points toward the SAM substrate. We find the rate of clogging for this case about twice the rate for the first case of the tip pointed upstream toward the atomic beam source. We surmise that the higher rate of clogging for the second case with the tip pointed toward the substrate arises from the Cs flux scattered by the interior pyramid walls and constrained to accumulate around the aperture within the hollow tip. In the first case with the tip pointed toward the atomic beam source the Cs flux striking the pyramid walls near the aperture can rebound away from the pinhole without constraint and therefore will accumulate more slowly around the aperture entrance. From a simple model of atom linear accretion around the periphery of the aperture we estimate that the "sticking coefficient," the probability that an atom hitting the periphery of the aperture will adhere to it, is about 0.5%. We are presently investigating the possibility of further reducing the clogging rate by direct resistive heating of the pyramid structure. A detailed study of the clogging rates for these pyramidal masks will be published elsewhere.

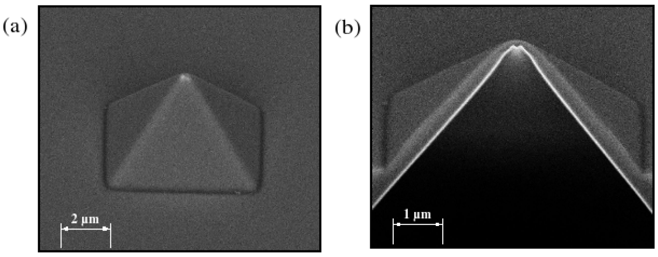

FIG. 4: (a) A secondary electron microscopy (SEM) image of the pyramidal tips used in the *Atom Pencil*. Panel (a) shows one of the pyramidal structures with a 300 nm hole in the apex of the tip. The base of the pyramid, as shown after the wet-etch process, is 3.5 μm on a side. The overall base of the pyramidal structure is 22 μm on a side. Panel (b) shows a focused ion beam (FIB) milled cross-section of the pyramidal tip aperture.

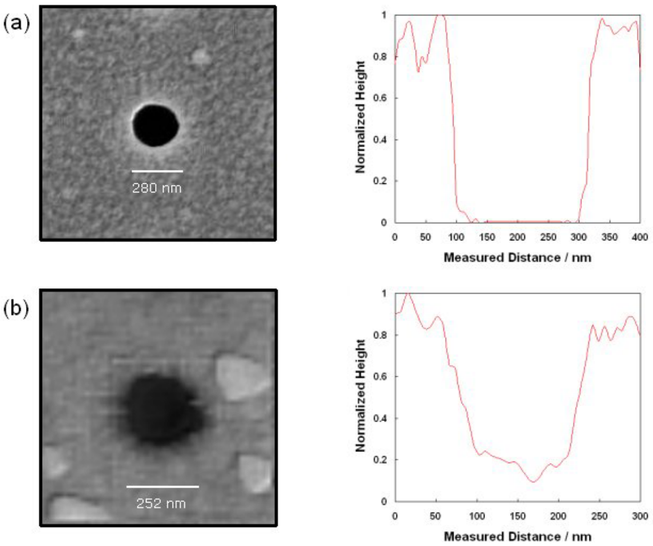

FIG. 5: (a) AFM image (left panel) and measured profile (right panel) of the etched SAM substrate exposed for 15 minutes to the Cs atom flux through the pyramidal tip mask. Note that the measured line profile exhibits a very sharp fractional vertical (depth) gradient, $\simeq 0.8/20\,\mathrm{nm}$. (b) Same conditions after a 30 minute exposure. The pyramidal tip is pointed toward the atomic beam source. The mask aperture is $16\,\mu$m above the SAM surface; the diameter is 300 nm. From the line profiles taken at 15 minute intervals over one hour the average clogging rate $\simeq 6\,\mathrm{nm\,min}^{-1}$.

In conclusion we can state that the *Atom Pencil* is a versatile atom writing tool that allows the generation of sub-micrometer structures. This device can be used to deliver precision quantities of material at the micro- and nanoscale to an active surface at much lower energy than ion implantation techniques and might find application in precision doping of technologically useful materials. The technique can be easily adapted to a planar 2D array of atom pencils thus significantly increasing the yield of written figures. Extension of the atom pencil to other atomic species accessible to laser cooling and increased writing speeds should be straightforward to realize.

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