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The atom pencil: serial writing in the sub-micrometre domain

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ABSTRACT The atom pencil we describe here is a versatile 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 on to 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. 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 sub-micron aperture.

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Atom lithography aims to fabricate lateral structures on micro- and nanometre scales 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 interfering light beams [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 on to a fixed substrate just below it. This *atom pencil* writes arbitrary 2D structures through direct deposition of atoms, with low-energy impact. Developing the technology of the *atom pencil* is motivated by the future need for precision doping of semiconductor [8], spintronic [9] and photonic [10] devices. Although shadow-mask deposition through sub-micron apertures was first reported by Lüthi et al. [11], 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 while 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 a 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 $z_4^{th} = Rv_{th}^2/(a_4 L)$, 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, $\alpha_{div} = 1$ mrad. For estimating the on-axis flux density, we thus calculate the average flux into an area $\pi(\alpha_{div} z)^2$ and, averaging over the thermal longitudinal velocity distribution, we find $F_4 = C \exp(-z/z_4^{th})$, 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 6 cm/s, or 1 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 $|6S_{1/2}, F = 4, m_F = 4\rangle$ quantum state is obtained. The magnetic 4-pole has a

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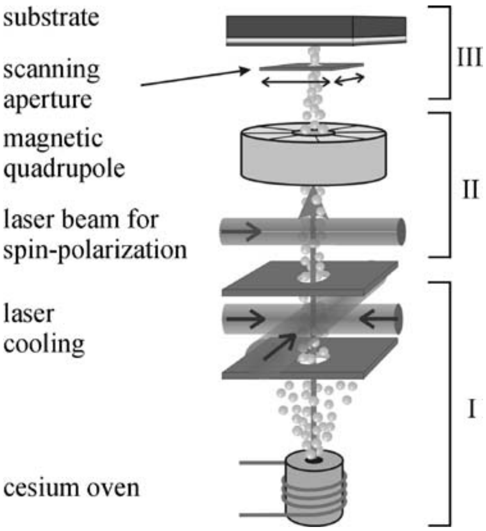

FIGURE 1 The *atom pencil* consists of a transversely collimated atomic beam I, which after a concentration stage II is deposited through a pinhole on to a substrate III

50-mm diameter with an inner bore of 10 mm. It is 20-mm long and provides a field gradient of 2.81 T/cm in the radial direction [12, 13]. The thermal velocity distribution leads to a focal line of about 100-mm length for the 4-pole. At 65-mm separation from the centre 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} cm^{-2} s^{-1}. The divergence at this position is 8 mrad and the atomic beam FWHM is 45 µm. An increased oven temperature could also raise the flux density of the atomic beam, but we have found that the quality of transverse laser cooling is seriously impaired at these higher densities.

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 [13]. 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 $F_6 = Czz_4^{th}/(z_6^{th})^2 \exp(-z/z_6^{th})$. The characteristic focal length of a 6-pole constructed from the same material is now $z_6^{th} \approx 6z_4^{th}$. Comparing F_6 and F_4, the 4-pole concentration exceeds the 6-pole concentration by more than an order of magnitude for $z < z_4^{th}$. These properties are summarized 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 [14, 15]. 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 etch process [16]. For the first experiments with the concentrated atomic beam we used micron-sized stainless steel apertures. A simple structure written by stepping the aperture across the concentration maximum is shown in Fig. 3a. 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. 3b we show a similar structure written with a 400-nm aperture and a 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 the present experiments our key element for writing sub-micron 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 Refs. [17, 18].

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 1 mrad) but with a flux density of 10^{12} cm^{-2} s^{-1}. A SAM-covered substrate was exposed to the cesium atom flux for 15-min intervals up to 1 h 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 µm above the SAM surface and the aperture diameter was 300 nm. Figure 5a and b show AFM images of the etched SAM and line profiles, respectively. They demonstrate that the aperture significantly closes after 30-min exposure.

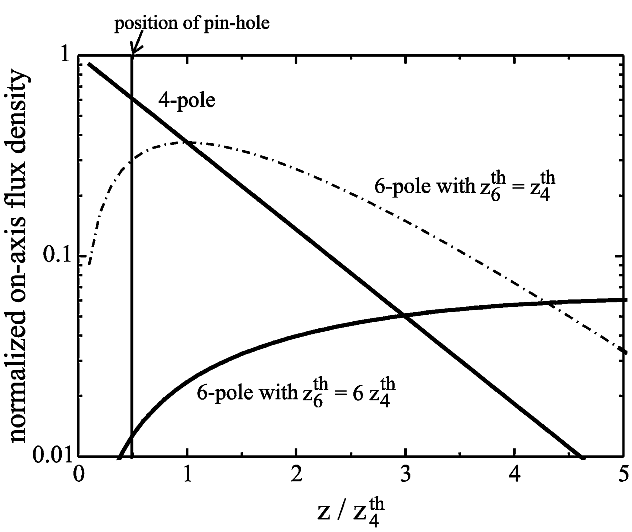

FIGURE 2 Normalized on-axis flux density for a thermal atomic beam focused by a magnetic 4-pole and a magnetic 6-pole with a short focal length (*upper curve*) and a realistic long focal length (*lower curve*). The pinhole is typically positioned at $z = 0.5z_4^{th} = 65$ mm from the centre of the 4-pole

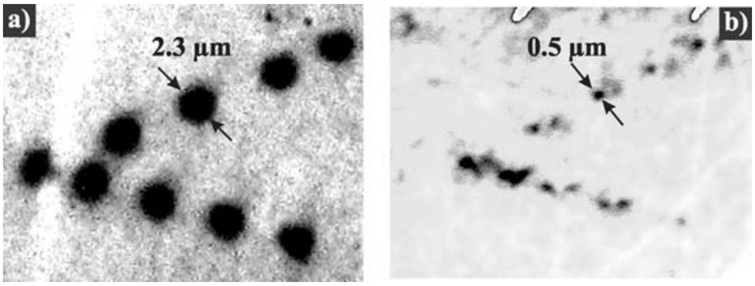

FIGURE 3 Examples of structures consisting of a V-shaped stepped series of dots written with apertures of two different diameters: **a** 1 µm, **b** 400 nm. The exposure time per dot was 180–200 s

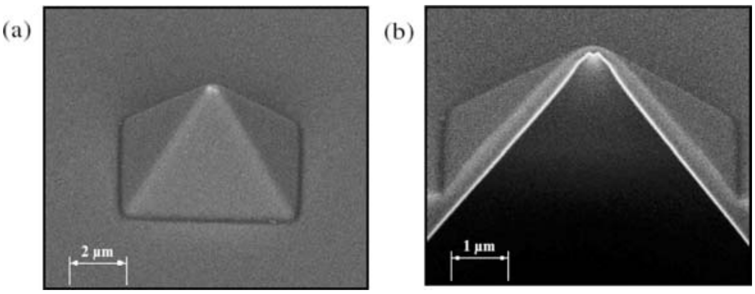

FIGURE 4 Secondary electron microscopy image of the pyramidal tips used in the *atom pencil*: **a** pyramidal structure with a 300-nm hole in the apex of the tip, **b** cross section of the pyramidal tip aperture milled with a focused ion beam

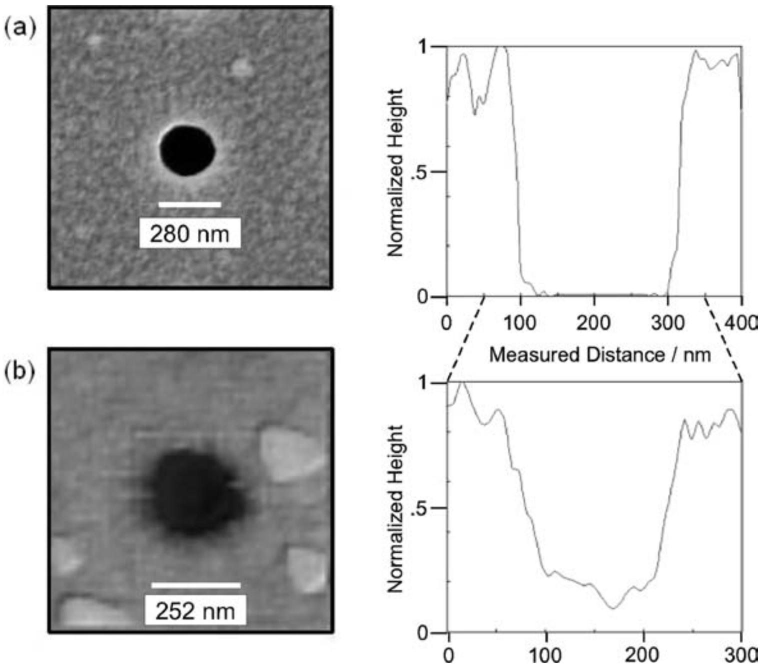

FIGURE 5 **a** AFM image (*left*) and measured profile (*right*) of the etched SAM substrate exposed for 15 min to the cesium atom flux through the pyramidal tip mask. **b** Same conditions after a 30-min exposure. The pyramidal tip is pointed toward the atomic beam source. The mask aperture is 16 µm above the SAM surface; the diameter is 300 nm

After taking successive Cs flux exposures, AFM images and line profiles for up to 1 h, we find that the average clogging rate is 6 nm/min. The line profile of Fig. 5a exhibits very steep flanks. The absolute depth of the hole is about 50 nm. The sharp hole edge contrasts favourably with electron- or ion-beam milling since the profile of these beams is always Gaussian, and the milling rate is therefore always less on the outer periphery than on the beam centre. Furthermore, the thermal Cs beam with its relatively 'soft landing' does not create collateral damage or engender proximity effects between adjacent holes [19]. We have also carried out clogging measurements for the pyramidal aperture oriented in the opposite direction, i.e. pointing toward the SAM substrate. We find the rate of clogging for this case to be about twice the rate for the first case. We surmise that the higher rate of clogging for the second case 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, 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 probability that an atom hitting the periphery of the aperture will adhere to it is about 0.5%. We have also carried out measurements of clogging rate with the same type of masks resistively heated to about 340 K. *These masks show no evidence of clogging over continuous operation for 16 h and intermittent operation for several weeks.*

In conclusion, we can state that the *atom pencil* is a versatile atom writing tool that allows the generation of submicrometre structures. This device can be used to deliver precision quantities of material at the micro- and nanoscales 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 such as Au and Ag as well as increased writing speed should be straightforward to realize.

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