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Atomic nanofabrication by laser manipulation of a neutral cesium beam

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Abstract

In this work, we report on the results of a nanolithography experiment with a cold cesium beam. We have realized a brilliant and collimated cesium beam with a low longitudinal velocity (10 m/s) exploiting laser cooling techniques, in particular a pyramidal atomic funnel. The cesium atomic beam has been utilized to pattern gold substrates, using Self Assembled Monolayers (SAM) of thiols as resist, and a wet etching process. The pattern generated by a light mask, a one-dimensional standing e.m. wave, was characterized by diffraction and Atomic Force Microscopy (AFM) measurements, showing the presence of lines spaced half the wavelength of the standing wave (426 nm) with lateral size well below 100 nm.
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1. Introduction

In the past decades we have observed a continuous decrease of the feature size in integrated circuits, well described by the Moore's law, which summarizes the industry's ability to quadruple the number of transistors on a chip every year [1]. Clearly the semiconductors industry is quickly approaching the physical limit of conventional optical lithography, the basic technology of semiconductors industry, located below 100 nm. Even though this will not affect the electronic market for the next decade, it is on the other hand clear that fabrication and manipulation of electronic devices in the range of tens of nanometers requires the application of different concepts and architectures. Among the new fabrication approaches, atom lithography [2] has attracted a growing interest, due to the possibility of producing ordered nanosized structures with a lateral size below the optical diffraction limit.

Atom lithography is in principle similar to optical lithography, the main difference being that in atom lithography neutral atoms play the role that light plays in optical lithography. In other words, while in optical lithography matter is used to manipulate light and generate patterns (for example by using masks, lenses, mirrors), in atom lithography light is exploited to manipulate atoms. The patterns generated by the interaction of atoms with light can be either directly deposited on proper substrates or used to impress a particle sensitive resist, in close analogy with conventional optical lithography.

The main and most powerful aspect of atom lithography is the possibility of utilizing light masks. Roughly speaking, a light mask is a standing configuration of e.m. fields, quasi resonant with an atomic transition, able to guide the atoms, i.e., to focus them onto the substrate. An atom in a quasi resonant non-uniform e.m. field feels a force, known as dipole force, described by the relation

$$F_{\mathrm{dip}} = -\frac{h\gamma^2}{8\delta I_{\mathrm{sat}}}\nabla I(x), \qquad (1)$$

where I_{sat} is the saturation intensity of the atomic transition, γ the natural linewidth, δ the detuning of the e.m. field with respect to the atomic transition and $I(x)$ is the intensity of the e.m. field. In the simple case of a one-dimensional standing wave with wavelength $\lambda = 2\pi/k$, $I(x) = I_0 \sin^2(kx)$, and atoms will be pushed toward the minima or maxima of the standing wave, depending on the sign of the detuning, generating an array of nanolines spaced $\lambda/2$. The advantage of using a neutral particle beam and a light mask is that diffraction does not represent a limiting factor as in optical lithography, since the de Broglie wavelength of a particle beam is, generally, well below 1 nm. Furthermore, limitations related to the periodicity of the light mask can be overcome by using holographic masks [3] or a proper movement of the substrate during the deposition. The main limiting factors are indeed

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related to the divergence of the atomic beam, and to effects of chromatic and spherical aberrations, due to the non-perfect monocromaticity of the particle beam.

However, the advent of laser cooling [4] has offered a series of techniques able to produce beams of cold atoms, with a reduced divergence and a good monocromaticity. In principle, structures with typical dimensions below 10 nm could be produced by atom lithography, although, up to know, structures of 20 nm [5] have been realized with thermal beams, limited by the interactions of the deposited atoms with the substrate.

In this work, we present the results of an atom lithography experiment, where a beam of cold cesium atoms, that is a beam with a reduced longitudinal velocity, is exploited. To the best of our knowledge, this is the first report on atom lithography with cold neutral atoms, previously being reported only for metastable noble gas atoms (Ne) [6]. Cold cesium atoms were deposited, through a light mask consisting of a one-dimensional standing wave, on Self Assembled Monolayers (SAM) of thiols on gold. A post-deposition etching process transfers the pattern generated by cesium atoms to the gold layer. The experimental setup, the atomic beam properties and preliminary analysis of the patterned samples will be discussed in the following sections.

2. Experimental setup

The system we realized is schematically represented in Fig. 1. The cesium beam, differently from standard atom lithography experiment where a high temperature oven is exploited as the atomic source, is produced by a pyramidal funnel [7], a simplified magneto-optical trap (MOT) that is able to generate an intense atomic beam, cooled also in the longitudinal velocity component. The atomic beam is further laser cooled in the transverse directions, in order to reduce its divergence and increase the surface flux density (in other words, to decrease deposition time) [8]. Finally, a one-dimensional standing wave is used to focus the cesium atoms in parallel nanolines.

The atomic pyramidal funnel consists of four mirrors arranged in the shape of an inverted pyramid with a small hole at its apex. This system requires only one input laser beam to produce the same configuration of a standard six-beam MOT, in terms of beam direction and polarization. Furthermore, the hole at the pyramid apex creates a region where the force due to the incident trapping laser beam is unbalanced and, consequently, the atoms captured and slowed inside the pyramid volume are pushed away through the hole as they reach this region, generating a continuous beam of cold atoms.

We used diode laser as radiation sources, operating at 852 nm corresponding to the D2 cesium transition ($5S_{1/2} \rightarrow 5P_{3/2}$ Cs levels). The trapping laser, necessary for the funnel operation, is produced by two diode lasers in master–slave configuration, while a DBR diode laser produces the radiation for the repumping process. A third laser diode provides the light needed for the collimation of the atomic beam. A second couple of diode lasers, in master slave configuration, is used to generate the light mask. Trapping, collimating and light mask lasers are frequency stabilized, through saturated absorption spectroscopy, on the $|F=4\rangle \rightarrow |F'=5\rangle$ hyperfine transition of the D2 line, while the repumping laser is locked on the $|F=3\rangle \rightarrow |F'=4\rangle$ transition.

The vacuum system is made of two main chambers separated by a gate valve, typically kept at a base pressure around 10^{-8} mbar. In the first chamber, the mirrors of the pyramid are mounted on a stainless steel holder and the

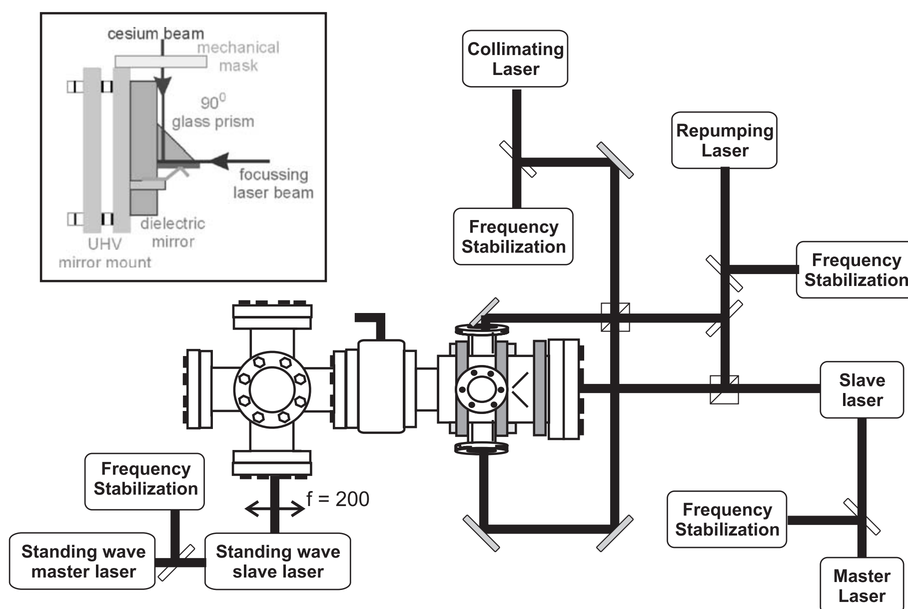

Fig. 1. Sketch of the experimental setup; the inset shows schematically the arrangement for the substrate and mirror holder.

atomic beam is produced and collimated, the collimating laser beam being sent into the chamber through four optical windows. The second chamber houses the substrate holder and the optics for the standing wave used as optical mask.

The light mask, a one-dimensional standing wave, is produced by retro-reflecting a laser beam with a mirror mounted on the substrate holder (see inset of Fig. 1), to allow precise alignment of the standing wave. In order to increase the intensity of the standing wave (I_0) and reduce the interaction time and, consequently, the number of spontaneous emission events that may lead to broaden the deposited structures, the standing wave laser is focused with a cylindrical lens ($f=200$). Thus, the standing wave laser beam has a waist of 4 mm in the direction perpendicular to the axis of the atomic beam, and a waist of 110 μm in the parallel one. With the typical parameters of the light mask (a power of 60 mW and a detuning of 1 GHz) the number of spontaneous emissions is ~ 6. The substrate is clamped on the mirror, in order to prevent any relative motion, and two 90° prisms ensure the proper orthogonal mutual alignment between the standing wave, the atomic beam and the substrate (see inset of Fig. 1).

3. Atomic beam characterization

The laser-cooled Cs beam leaving the funnel has been fully analyzed, and measurements have been already presented elsewhere [7]. Briefly, an atom beam with a transverse dimension roughly corresponding to the hole cross-section (around 2 mm) is continuously produced, with an atom flux of 4×10^9 atoms/s. Longitudinal velocity of the beam is in the range 8–12 m/s depending on the operating parameters, with a 1.5 m/s spread corresponding to the measurement accuracy. The beam divergence is approximately 25 mrad FWHM.

Such a divergence value is not compatible with atom lithography. The maximum allowed beam divergence depends on the parameters of the focusing stage; a rough estimation suggests that the divergence should be below 5 mrad. Thus, an efficient beam collimation is a key point for the process. Furthermore, a reduced divergence will also allow us to enhance the flux density, i.e., the number of atoms per unit of surface and time, that corresponds to shorter deposition time. Our collimation stage [8] is based on an optical molasses, a configuration of two retro-reflected laser beams, mutually orthogonal, and also perpendicular to the atomic beam axis. The collimation follows interaction of the atoms with these quasi-resonant laser fields and the consequence of several absorption and spontaneous emission cycles is a net decrease of the atom transverse velocity, i.e., beam collimation. Two polarization configuration has been exploited for the collimation laser beams, the "lin⊥lin" and the "$\sigma^+\sigma^-$", the latter being more efficient in the presence of stray magnetic fields, as in our case. Also in the collimation

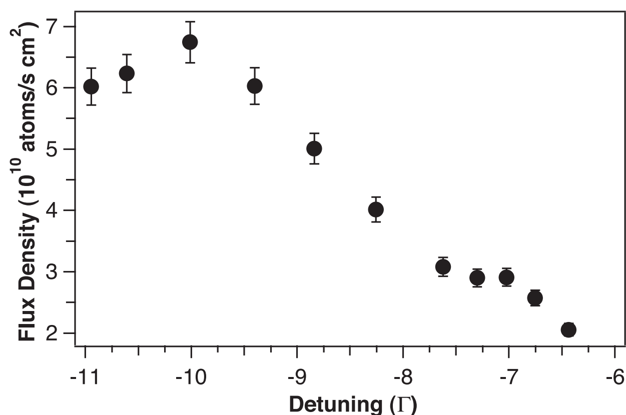

Fig. 2. Flux density vs. collimating laser detuning, reported in units of Γ, the natural linewidth of the Cs D2 transition.

stage, an increase of the atom/laser interaction time (around 1.2 ms) is provided by the use of elliptical beams, 12 mm long and 5 mm high.

Collimation turns out to reduce the divergence of the atomic beam below 2 mrad, a value limited by the measurements uncertainty. The effect of the collimation stage is also a decrease of the transverse dimensions of the atomic beam from 1 cm (FWHM) to 3 mm in the deposition region. As shown in Fig. 2, where the flux density is plotted against the collimating laser detuning, the flux density reaches values up to 6.5×10^{10} atoms/(cm^2 s) for a laser detuning of -10Γ (Γ being the Cs D2 natural linewidth), a value more than one order of magnitude grater than for the uncollimated beam.

4. Deposition results

To test the suitability of the cold atom beam for atom lithography experiment, we have deposited the cesium atoms onto an organic SAM. The SAM we used is a thiol, specifically nonanethiol ($CH_3(CH_2)_8SH$), which spontaneously forms a monolayer on gold substrates. We used commercial substrates (ARRANDEE™), consisting of a gold layer 75 nm thick, grown on a borosilicate substrate, with a Cr adhesion layer (5 nm thick). The gold substrates, once cleaned in piranha solution, were immersed in a solution of ethanol and nonanethiol (1–5 mM), and the organic layer was completely formed after 24 h.

The SAM, due to the CH_3 head group, is normally hydrophobic, but upon contact with Cs undergoes a chemical reaction that locally destroys this hydrophobic character. After exposure to the Cs beam, the SAM resist is treated by standard chemical wet etching to permanently transfer the pattern produced onto the substrate. The etching solution we used is made of distilled water, KOH 1 M, $K_2S_2O_3$ 0.1 M, $K_3Fe(CN)_6$ 0.01 M, $K_4Fe(CN)_6$ 0.001 M.

In the first stage of the experiment we used commercial TEM Ni-grids as mechanical masks to pattern the gold

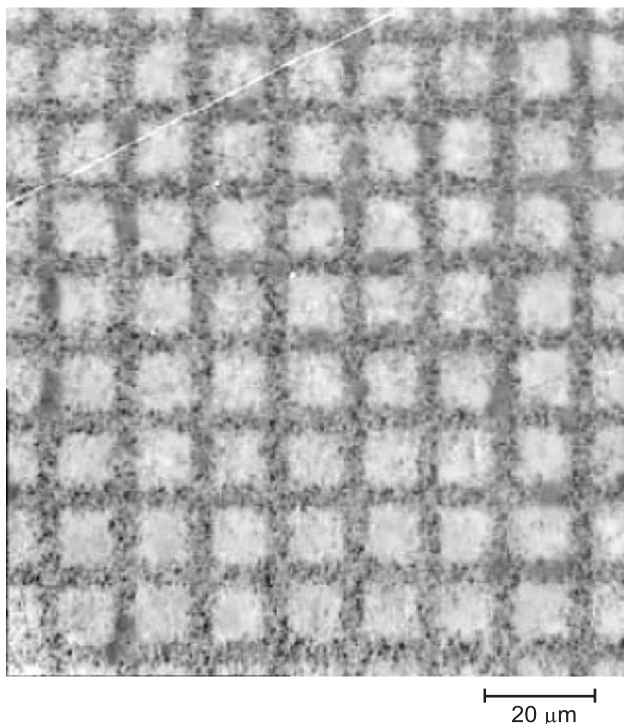

Fig. 3. Optical microscope image (dark field) of a patterned gold substrate. The deposition time was 2 h. The used mask was a Ni TEM grid, 1500 lines/in., pitch 16.5 μm, bar width 5 μm.

substrate. These grids have features in the μm range. They were put in contact with the SAM layer and exposed to the cold cesium beam for up to 3 h. After the etching process the mask pattern was transferred to the gold substrate through the etching process and the samples analyzed with an optical microscope. Fig. 3 shows a typical sample, well reproducing the mask pattern, thus demonstrating the feasibility of SAM impression by our cold cesium beam.

The second stage of the experiment was the realization of the light mask. As explained above, the intensity gradient dipole force (Eq. (1)), acting as an array of nano lenses, channels the atoms into the nodes of the standing wave and focuses the atoms onto the substrate surface. Nanolines can be produced spaced precisely half the wavelength of the standing wave (426 nm). In order to have macroscopic information on the periodicity of the produced patterns, we characterized our samples by optical diffraction measurements, the pattern generated on the gold substrate acting as a diffraction grating for visible light. According to the well known diffraction law:

$$\cos(\theta_i) + \cos(\theta_d) = -\frac{m\lambda}{d} \tag{2}$$

$m = 0, \ \pm 1, \ \pm 2, \ \ldots$ being the diffraction order, λ the wavelength of the radiation, θ_i and θ_d the incident and diffracted angles, respectively, the grating period d can be

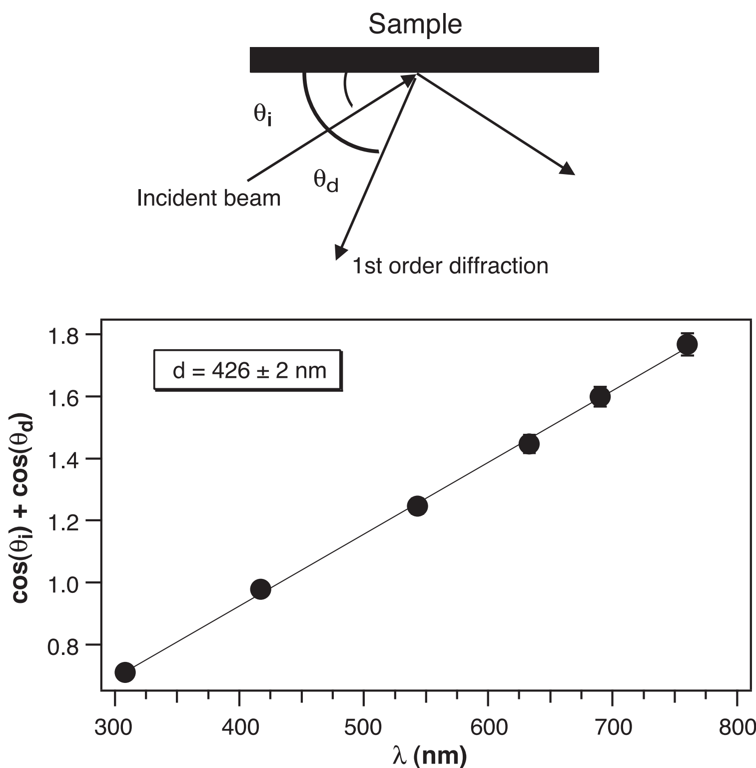

Fig. 4. Optical diffraction measurements of a sample patterned with a light mask. The drawing schematically represents the measurement principle.

easily derived from measurements at different wavelengths. An example of results for a sample exposed to the cold beam for 2 h is reported in Fig. 4. Optical diffraction was clearly visible at any wavelength, and the expected periodicity of 426 nm was found.

We have also carried out preliminary Atomic Force Microscopy (AFM) analysis of our samples. Results, not reported here, are in good agreement with the optical diffraction analysis: an array of parallel trenches spaced 426 nm is observed on the gold surface, with typical lateral size around 50 nm FWHM. Such a value appears to be strongly limited by the grain size of the gold substrate ($\sim$ 100 nm), by the local quality of the SAM, and by the isotropic nature of the etching process.

5. Conclusions

To summarize, we have built a cold cesium beam for nanolithography experiment. The beam, exploiting a pyramidal atomic funnel and further collimation by optical molasses, exhibits a flux of 4×10^9 atoms/s, a divergence less than 2 mrad and a longitudinal velocity around 10 m/s. We have utilized the beam to pattern a gold substrate, exploiting an organic SAM as resist and physical and light masks to generate the patterns. The analysis of our samples shows the presence of periodic structures with features around 50 nm, so demonstrating the possibility of using cold atoms for nanolitography purposes.

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