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# A laser-cooled atom beam for nanolithography applications

A. Camposeo, F. Cervelli, A. Piombini, F. Tantussi, F. Fuso\*, M. Allegrini, E. Arimondo

INFM, Dipartimento di Fisica "Enrico Fermi", Università di Pisa, Via F. Buonarroti, 2, I-56127 Pisa, Italy

#### Abstract

We are developing an apparatus for atom lithography with the main objective of to push the space resolution of the technique towards its ultimate limit, expected in the 10 nm range. We exploit an original implementation of laser-cooling techniques to produce a brilliant and collimated cesium beam with low longitudinal velocity. Beam characterization, carried out with a variety of spectroscopic techniques, demonstrates the compatibility of the system with the strict requirements of nanolithography experiments. © 2002 Elsevier Science B.V. All rights reserved.

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### 1. Introduction

Presently, large efforts are being devoted to develop novel fabrication techniques able to push lateral definition of the produced structures well below the 100 nm range. New approaches must be found in order to meet the strict requirements posed by such a challenging goal. Within this context, appealing possibilities are offered by atom lithography (for a review, see for example Ref. [1]).

Roughly speaking, atom lithography is similar to conventional optical lithography, but for the replacement of light with a neutral particle beam. The most striking consequence is that diffraction problems, which actually set the main limitation in the space resolution achievable by optical lithography, are confined to the sub-nm level, thanks to the small value of the de Broglie wavelength in particle beams. Furthermore, the recent development of laser-cooling in the area of atomic physics has demonstrated that atoms can be conveniently and efficiently manipulated through application of suitable electromagnetic fields. Exploitation of laser-cooling technologies makes possible to replace the standard mask of optical lithography with an immaterial mask, consisting of a particular configuration of standing electromagnetic fields. In practice, atoms of a beam can be guided into precisely determined space positions, corresponding to maxima, or minima (depending on the detuning of the focusing radiation with respect to an atomic

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transition resonance) of the standing e.m. field. The sosegregated beam can then be directly deposited to produce spatially ordered nanostructures, or used to impress a particle-sensitive resist. Advantages in terms of lateral definition and control of the space position are ensured by the immaterial mask, and arrays of regularly spaced planes or hexagonal dots have been already produced, with a lateral size below 20 nm [2]. On the other hand, fabrication of more arbitrarily shaped nanostructures is also possible, for instance by exploiting substrate motion, or sophisticated holographic masks [3].

Furthermore, thanks to particle neutrality, problems related to electric forces (see, e.g., charged-beam lithography) are practically negligible, and relatively large diameter beams can be produced in order to preserve the parallel character typical of conventional lithography. Obviously, in order to be used in an atom lithography experiment, the atom beam must meet specific requirements especially in terms of brilliance and collimation. In fact, a large atom flux is needed to have relatively short exposure times, and, similar to what was experienced in standard light optics, beam divergence must be minimized in order for the atoms to be efficiently focused onto the substrate. Moreover, beam monochromaticity (i.e., reduced spread in the longitudinal velocity) is desirable to prevent chromatic aberrations.

In this work, we report on the preparation of an atom beam suitable for nanolithography purposes. The main difference with respect to all systems already in use is the wide exploitation of laser-cooling and trapping techniques [4], aimed at producing a well-collimated and intense beam with a small longitudinal velocity and a highly monochro-

<sup>\*</sup> Corresponding author. Tel.: +39-05-0221-4305; fax: +39-05-0221-4333.

E-mail address: fuso@df.unipi.it (F. Fuso).

matic character. Cesium has been chosen as the atom beam species, mostly motivated by the availability of instruments and methods for an efficient laser-manipulation. Beam properties have been analyzed by various non-obtrusive techniques based on imaging and spectroscopy methods. Our findings can be transferred to beams of other atomic species for which cooling schemes and suitable laser sources are available, including for instance elements of group III (e.g., Ga, In) of direct technological interest.

#### 2. Experimental setup

The experimental set-up, sketched in Fig. 1, can be roughly divided into three main parts, devoted to preparation of the laser-cooled beam, already presented in Ref. [5], to its collimation and to the diagnostics. Further developments of the set-up, in progress at present, foresee an atom focusing stage and a facility for in-situ probe microscopy of as-deposited samples. The whole process takes place in an ultra-high vacuum environment, consisting of a stainless steel chamber evacuated by ion and turbo-dry pumps at a residual pressure below  $10^{-9}$  mbar. In addition, a complete laser set-up has been developed for the laser-manipulation of Cs atoms. It consists of different diode laser sources, operating around the D2 resonance line  $(6S_{1/2} \rightarrow 6P_{3/2} \text{ Cs})$ levels, wavelength around 852 nm). The optical set-up includes a variety of optical and opto-electronics components, which will not be described here, used to stabilize the laser operation and to condition the laser beams prior to interaction with the atoms.

The laser-cooled atom beam is produced out of a pyramidal funnel [6]. This is a particular configuration of mirrors and prisms, assembled in a hollow pyramidal shape (base size 38 mm) with an apical hole  $(1 \times 2 \text{ mm}^2)$ . By sending a single large-diameter laser beam onto the hollow pyramid along its axis (hereafter denoted as *z*-axis), the laser beam configuration of a standard magneto-optical trap (MOT) can be recovered, the quadrupolar static magnetic field required for trapping being provided by a pair of anti-Helmoltz coils wounded around the pyramid. As in any MOT, a repumping laser beam is needed to recycle atoms lost to atomic levels not interested by the cooling transition. More specifically, in our set-up the cooling laser is reddetuned with respect to the  $|F=4> \rightarrow |F'=5>$  hyperfine transition, while the repumper is tuned on the |F=3> $\rightarrow |F'=4>$  transition.

The presence of the apical hole prevents retroreflection of the laser light along the z-axis. Thus, atoms feel an unbalanced force, which pushes them through the hole, so forming a continuous beam. After leaving the funnel, the atoms enter the collimation region, where they undergo interaction with two pairs of countepropagating laser beams crossing orthogonally the atom beam. The collimating mechanism involves the realization of a two-dimensional "optical molasses" [4]. The exchange of momentum between laser radiation and Cs atoms leads to friction forces along the transverse (x and y) directions, which affect the transverse dynamics of the beam. The process depends on many laser parameters, including the detuning with respect to the Cs atomic resonance and the intensity. Furthermore, optical molasses is generally produced with linearly polarized light (in the "lin perp lin" configuration, meaning that in the retroreflection polarization direction is changed by  $90^{\circ}$ ), but the presence of stray magnetic fields in the interaction region (the tails of the MOT quadrupolar field)



Fig. 1. Sketch of the experimental set-up for the preparation of the collimated laser-cooled cesium beam.

suggested investigation of circular polarized radiation ( $\sigma^+/\sigma^-$ , meaning that the direction of the circular polarized light is changed in the retroreflection). In order to control and/or modify the magnetic field in the interaction region, we added a third coil, identical to those used for the pyramidal MOT. By tuning the current flowing in this coil, we could modify the intensity of the static magnetic field and its spatial gradient in the interaction region, which both affect the transverse laser-cooling processes [4].

Atom beam diagnostics is carried out by various methods substantially based on laser excitation. To this aim, a probe laser beam, produced by a laser diode completely adjustable in terms of intensity, polarization and frequency, is sent across the beam at  $\sim 250$  mm distance from the pyramid hole. Results of the interaction, either emission or absorption, are observed and imaged through suitable optical accesses.

#### 3. Results and discussion

The laser-cooled Cs beam leaving the funnel has been fully analyzed, and measurements have been already presented elsewhere [5]. Briefly, an atom beam with a transverse dimension roughly corresponding to the hole crosssection is continuously produced, with an atom flux in excess of  $10^9$  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 limited by the measurement accuracy. The beam divergence is approximately 25 mrad FWHM.

Such a divergence value is not compatible with atom lithography. In the simplest geometrical arrangement, which we intend to exploit, the atom beam is made to cross a onedimensional standing wave produced by superposition of two laser beams. For laser radiation suitably detuned with respect to the D2 Cs transition, atoms are polarized, and interaction with the standing wave leads to the occurrence of dipolar forces, which modify the dynamics of the atom beam along that direction. Since a standing wave is used, the intensity of the guiding force is regularly distributed in space. In other words, a regularly spaced (the spacing being equal to one-half the wavelength, i.e., around 426 nm) sequence of potential wells is produced along a transverse direction. Atoms are guided towards the region of minimum potential, but, for a beam divergence too large, they may escape the potential well. As a consequence, no space segregation of the beam is realized, and no nanostructure can be obtained.

The maximum allowed beam divergence depends on the parameters of the focusing stage, which are not yet fixed in our experiment. A rough estimation suggests that the divergence should be below 1 mrad. Thus, an efficient beam collimation is a key point for the process. It must be noted that exploitation of a laser-cooled beam, as in our apparatus, leads to increase the collimation efficiency. In fact, the mechanisms responsible for momentum exchange in the collimation stage turn out to involve stimulated absorption/emission cycles, being more efficient if many cycles occur in the interaction region. Due to the reduced longitudinal velocity of the beam, a long interaction time can be achieved by keeping the size of the laser beams used for the optical molasses at a reasonable level. We use elliptical beams 12 mm long and 5 mm high, which allow around  $4 \times 10^4$  cycles to take place.

Analysis of the beam dimension was accomplished by emission fluorescence imaging. The atom beam was illuminated by weak quasi-resonant radiation (detuning  $\approx \Gamma$ ,  $\Gamma$ being the spontaneous emission rate) in a 0.6-mm-wide cigar-shape beam sent along the *x*-direction. The emission was collected by a scientific CCD observing along the *y*direction. A Gaussian fit of the fluorescence profile was then carried out in order to derive the beam spatial profile. Care was devoted to prevent any saturation effect of the cesium atomic transition and probe beam retroreflection has been used to minimize dynamical effects on the atom motion (e.g., beam deflection).

The effects of collimation are clearly visible in Fig. 2, which reports the fluorescence emission profiles for the same probe laser position when the collimation lasers are switched off (uncollimated) and on (collimated). Collimation lasers had a detuning of  $-9.7\Gamma$ , the power for each beam was 3 mW and the polarization was linear ("lin perp lin"). The collimated beam appears spatially compressed with respect to the uncollimated one. Furthermore, its position is slightly modified as a consequence of laser manipulation in the collimation stage.

The beam divergence was then evaluated by analyzing the emission profiles as a function of the position. To this aim, the position of the cigar-shaped probe beam was moved along the z-direction, and the beam dimension was evaluated as a function of the position. Beam divergence was derived by a linear fit of the beam dimension. An example of our results is shown in Fig. 3. The beam divergence is reported as a function of the static magnetic



Fig. 2. Atom beam profile with collimation lasers off (uncollimated) and on (collimated). The emission signal of the uncollimated beam has been multiplied by a factor 5 for the sake of clarity.



Fig. 3. Atom beam divergence as a function of the static magnetic field in the interaction region. Values reported in the horizontal axis were derived theoretically from the intensity of the current flowing in the coils. The divergence of the uncollimated beam is reported for reference (dashed line).

field (directed along the *z*-axis) in the center of the interaction region calculated according to the current flowing in the coils. Collimation laser beams had in this case  $\sigma^+/\sigma^$ polarization, their power and detuning being 3 mW and  $- 8.2\Gamma$ , respectively. The data demonstrate that it is possible to achieve an almost complete freedom in determining the transverse dynamics of the beam, by simply modifying the static field in the interaction region: we could even change the beam behavior from divergent to convergent (positive or negative values of the divergence in Fig. 3, respectively).

According to our data, a residual divergence equal to zero within the error bar can be achieved, with a beam size around 4 mm FWHM (see also Fig. 2). A detailed interpretation of our findings on the basis of laser-cooling processes, which is beyond the scope of the present paper, demonstrates the crucial role of all collimation parameters.

The density of the atom beam after collimation was measured by absorption spectroscopy. After crossing the atom beam, the probe radiation, kept at an intensity well below the saturation of the optical transition, was collected by a photodiode. When needed to improve the measurement sensitivity, frequency modulation technique was adopted. The beam turned out to have a density around  $5 \times 10^6$  atoms/cm<sup>3</sup>, which corresponds to an atom flux slightly below the value measured right after the funnel [5], but in

any case of the order of  $10^9$  atoms/s. A rough estimation, based on a reasonable efficiency of the focusing process, indicates that, with as similar flow, 1 mm<sup>2</sup> portion of substrate may be patterned in several tens of minutes to obtain an array of parallel planes 20 nm wide and a few nm high.

# 4. Conclusions

We have built a laser-cooled cesium atom beam with features compatible with the use in an atom lithography experiments in terms of divergence and flux. Its reduced longitudinal velocity and low translational temperature are expected to lead to a highly efficient atom guiding onto the substrate. Once the atom focusing stage is completed, our experimental apparatus will allow us to produce by direct deposition nanostructures (array of parallel planes) whose lateral definition will be limited mainly by particle diffusion on the substrate. In turn, the properties of our beam will enable a detailed investigation of the growth processes in a relatively unexplored regime, where the kinetic temperature of the impinging particles is much smaller than that of the substrate.

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#### References

- Special Issue on "Nanomanipulation of atoms", in: D. Meschede, J. Mlynek (Guest eds.), Appl. Phys., B 70 (5) (2000) 649–739.
- V. Natarajan, R.E. Behringer, G. Timp, Phys. Rev., A 53 (1996) 4381;
  W.R. Anderson, C.C. Bradley, J.J. McClelland, R.J. Celotta, Phys. Rev., A 59 (1999) 2476.
- [3] M. Mützel, et al., Phys. Rev. Lett. 88 (2002) 083601.
- [4] H.J. Metcalf, P. van der Straten, Laser Cooling and Trapping, Springer-Verlag, New York, 1999.
- [5] A. Camposeo, et al., Opt. Commun. 200 (2001) 231.
- [6] J.J. Arlt, et al., Opt. Commun. 157 (1998) 303.