ELSEVIER

Microelectronic Engineering 57–58 (2001) 857–863

MICROELECTRONIC ENGINEERING

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Three-dimensional nanolithography with light forces

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Abstract

We present the first realization of three-dimensional lithography on the nanometer scale using atom optical techniques. It has already been shown that, with atom lithography, two-dimensional lateral structures of 50 nm can be obtained. In our experiment we utilize the resonant enhancement of light forces to address specifically one species of a multi-component beam. Thus a host and a dopant material can be evaporated simultaneously, but only the dopant is focused and thus spatially modulated by the light field. In this paper we show that this patterning of the dopant concentration on the nanometer scale can cause macroscopic effects such as wrinkle pattern formation in a film.

Keywords: Three-dimensional nanolithography; Atom optical techniques

1. Introduction

In recent years the atom–light interaction has been used to cool and trap atoms [1,2] and to realize a great variety of optical elements for atomic beams such as atom-mirrors, beamsplitters and wave guides [3]. In 1992, atom lithography was invented at AT&T Bell Laboratories [4]. This non-conventional method of lateral nanostructuring employs the light forces associated with the near resonant light interaction. Thus far, the generation of one-dimensional [5] and two-dimensional structures have been reported [6–8].

In this paper we present a method which enables us to extend this technique to the third dimension. Three-dimensional nanostructures can be produced by utilizing the material selectivity of light forces. Light forces can be used to manipulate specifically one species of a multi-component atomic beam. Thus the selected atoms can be deposited in a structured way in a host material which is formed by the other homogeneously deposited components. With this technique we are able to grow a film which is structurally doped in three dimensions by using light forces for the two lateral dimensions and flux manipulation in the growth direction.

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0167-9317/01/$ – see front matter
PII: S0167-9317(01)00442-7

2. Principle of structured doping

In atom lithography the optical dipole force is used to manipulate the external motion of the atom to create nanostructures. This force arises from the interaction between the light-induced electric dipole moment and the electric field of the light itself. The resulting force is proportional to the light intensity.

An elementary light field configuration, which can be used to realize general two-dimensional structures, is a standing light wave. It is a realization of a periodic intensity distribution with a periodicity of $\lambda/2$ where λ is the wavelength of the light. The associated light force leads to a deflection of the atoms towards the nodes of the standing light wave if the laser has a higher frequency than the transition frequency (blue detuned). Thus a standing light wave acts as an array of cylindrical lenses with a period of $\lambda/2$. The atoms experience a force large enough for focusing only if the laser is tuned close to an atomic transition frequency so that the induced dipole moment is resonantly enhanced. Therefore, the light force is strongly element selective and can be used to address one specific atomic or molecular species of a multi-component beam.

The basic principle of structured doping is sketched in Fig. 1. A standing light wave tuned close to an optical transition of the dopant (in our case chromium) acts as an array of lenses and leads to a laterally modulated flux of the chromium atoms. Another material, which in our experiments is magnesium fluoride, does not interact with the light field and thus is deposited homogeneously and forms the host material. Therefore, the chemical composition of the resulting film is laterally

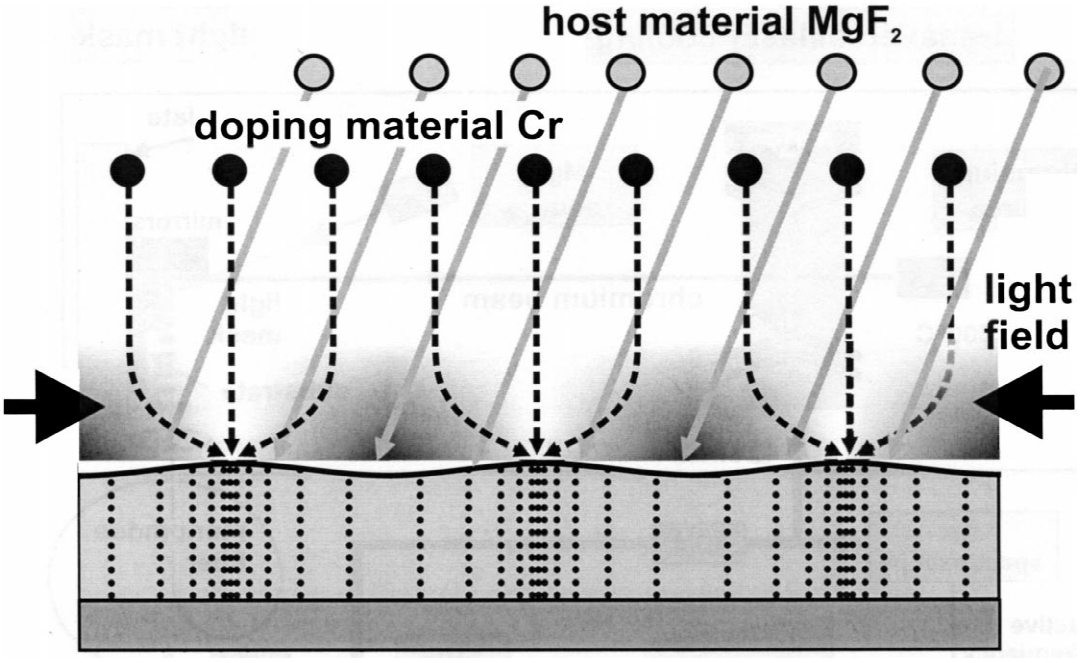

Fig. 1. Principle of structured doping. A multi-component beam, which in our experiment consists of chromium and magnesium fluoride, traverses a standing light wave. Only the chromium atoms are focused within the standing light wave because the light frequency is tuned close to an atomic transition of chromium leading to a resonantly enhanced light force.

modulated corresponding to the pattern of the light field. Combining this lateral patterning with the widely used technique of structured doping in the growth direction allows a straightforward realization of three-dimensional nanolithography.

3. Experimental setup

Our experimental setup is depicted in Fig. 2. The chromium beam is realized by sublimating chromium in a high temperature ($T = 1600°C$) effusion cell in a vacuum chamber (10^{-6} mbar). The distance from the end of the cell to the substrate (silicon) is 120 cm.

It is known from geometrical optics that the focal spot size depends on the divergence of the beam impinging on the lens. In order to obtain small focal spots for the chromium beam one has to collimate the divergent effusive chromium beam produced by the source. In our experiment this is achieved by using polarization gradient laser cooling [9] to reduce the transverse atomic velocity. This leads to an atomic beam with a residual divergence of less than 0.4 mrad.

The light mask is realized by retroreflecting a focused laser beam which is brought into the vacuum chamber by an optical fiber to obtain a good beam shape. The diameter of the standing light wave is about 200 μm. The frequency is tuned close (200 MHz) to the chromium transition $^7S_3 \rightarrow {}^7P_4$ at

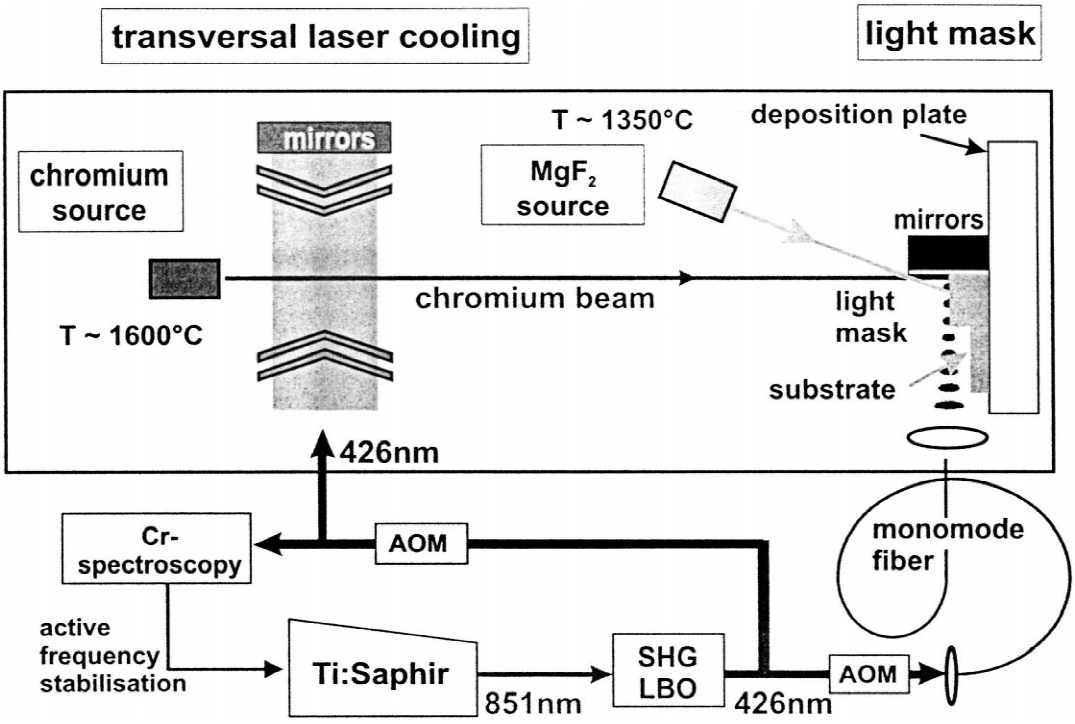

Fig. 2. Experimental setup. The chromium and MgF_2 beams are realized with high temperature effusion cells inside a vacuum system. The required light frequency corresponding to a vacuum wavelength of $\lambda = 425.6$ nm for resonant interaction with chromium is realized with second harmonic generation (SHG) of a Ti:Sapphire laser. The detuning of the standing light wave of 200 MHz from resonance is achieved with an acousto-optical modulator (AOM).

$\lambda = 425.6$ nm to address specifically ^{52}Cr atoms. This light field configuration acts as an array of cylindrical lenses and leads to a laterally varying chromium flux with a periodicity of $\lambda/2 = 213$ nm.

The host material, magnesium fluoride, is evaporated with another high temperature ($T = 1350°C$) effusion cell. The magnesium fluoride molecules are not affected by the light mask and thus form an homogeneous host material which is structurally doped with chromium in the transverse direction. The chromium flux (about 1 nm/min) and the magnesium fluoride flux (about 3 nm/min) is monitored with a rate monitor during deposition.

The realized thin films are analyzed with an optical microscope and with an atomic force microscope (AFM) outside the vacuum chamber.

4. Experimental results and discussion

The thin laterally structured Cr:MgF_2 films are experimentally produced by evaporating magnesium fluoride and chromium at the same time. After 15 min of evaporation the samples are taken out of the vacuum chamber and analyzed with an optical microscope. In the following we will describe our observation of wrinkle formation after the sample was taken out of the vacuum. We will show that this macroscopic effect allows us to deduce the success in realizing a structurally doped material using light forces.

A typical optical microscope picture is shown in Fig. 3a and reveals the formation of wrinkles which show up as bright lines. Clearly, one can see that the wrinkle pattern changes over the field of view from rough honeycomb structures to lines. A closer look with an atomic force microscope (see Fig. 3b) reveals that the straight wrinkles are formed where the chromium concentration is spatially modulated. This can be deduced from the surface topography which is modulated with a period of $\lambda/2 = 213$ nm. The AFM picture further reveals that the wrinkles on the micrometer scale are guided by the spatially modulated chromium concentration on the nanometer scale.

Analyzing the area with the honeycomb structures shows no topography on the nanometer scale, implying a spatially homogeneous chromium concentration. We performed further experiments with pure chromium and pure magnesium fluoride films but no wrinkle formation was observed. Thus the wrinkles occur only in a film consisting of Cr and MgF_2. This experimental result combined with the alignment of the wrinkles along the structured dopant concentration is a clear indication of the first successful creation of a laterally structured doped film.

Typical scales of the wrinkles in the region of the light mask are a width of about 2 μm and a height of about 500 nm. The spacing between two adjacent lines is on average 10 μm. Outside the light mask the wrinkles have a width of about 4 μm. The size of a cell in the honeycomb structure is about 10 μm × 10 μm.

It is known that a thin film deposited on a substrate by molecular-beam epitaxy can exhibit wrinkles or cracks due to compressive or tensile stress. The wrinkle formation can be understood as the interplay between compressive stress and the adhesion of the film on the substrate. As soon as the compressive stress becomes too large to be supported by the adhesive forces binding film and substrate together, part of the film lifts off. Buckling occurs in the delaminated portion of the film. It is known from the literature that magnesium fluoride and chromium exhibit only tensile intrinsic stress [10]. The question arises of why we observe a compressive stress for Cr:MgF_2 films.

In thin solid films one can observe three different sorts of stress: internal stress, which develops

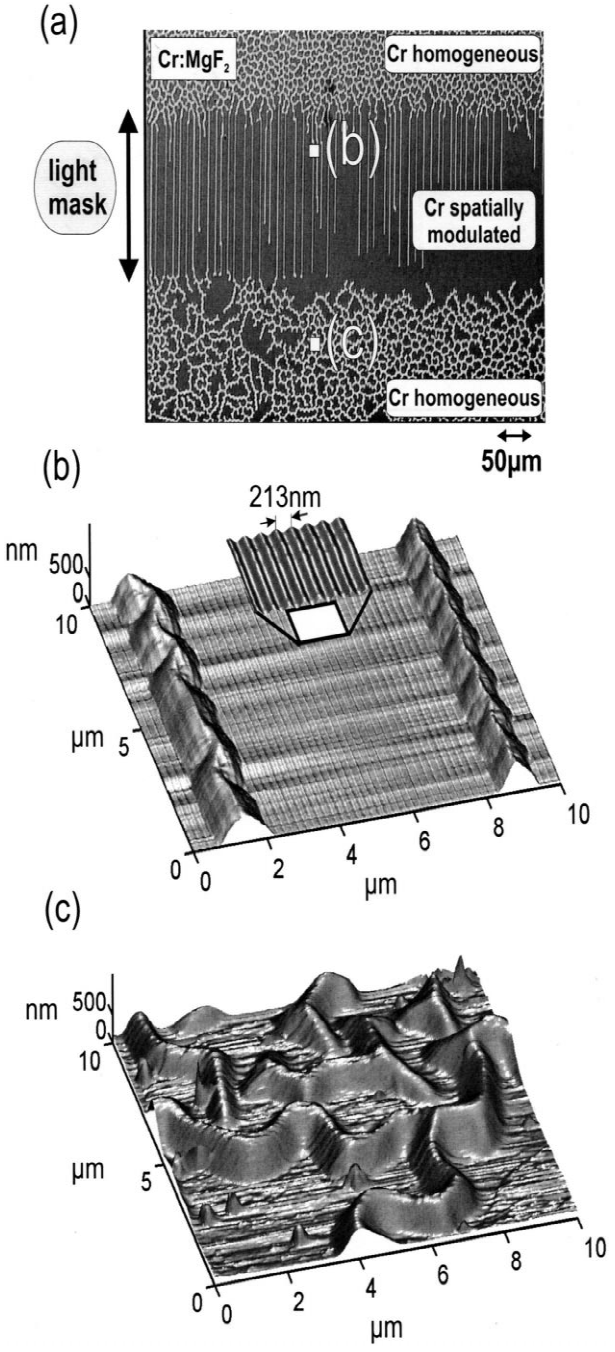

Fig. 3. Wrinkle pattern in a Cr:MgF_2 film. (a) An optical microscope image of the film. Bright lines represent wrinkles. The pattern changes from a honeycomb structure for positions outside the structured doping region to a well-regulated pattern in the region of the light mask. (b, c) Atomic force microscope images of the film at the positions indicated in (a). (b) The topography of the film where chromium was periodically focused and thus reveals a periodic structure with a period of $\lambda/2 = 213$ nm. Clearly, the wrinkles are aligned parallel to these nanostructures. (c) No periodic nanostructuring and thus the wrinkles are in arbitrary directions. The small modulation of the topography is an artifact of the AFM.

during deposition; thermal stress, which is due to the different thermal coefficients of expansion of substrate and film; and external stress, which can be caused by water absorption or chemical reaction after taking the sample out of the vacuum chamber. The analysis of the samples directly after the deposition revealed no wrinkle formation, which implies that the necessary internal stress does not build up during the deposition. Since the temperature of the samples does not change during the deposition, thermal stress does not play a role in our experiment. The compressive stress must be due to external stress. This is confirmed by the fact that the wrinkles only begin to grow after the sample is taken out of the vacuum chamber. Additionally, we were able to show that the growth of wrinkles could be accelerated by cooling the substrate and thus increasing the water concentration on top of the film. This indicates that the growth of wrinkles is due to water absorption leading to a compressive stress overcoming the intrinsic tensile stress.

Aligned wrinkles only occur if the growth velocity of the wrinkles is very slow (in the range of hours) which is the case when the samples are stored under normal air conditions. For that situation the observed wrinkle pattern is shown in Fig. 4a. The speed of formation of the wrinkles can be accelerated by cooling the substrate and thus increasing the amount of water on the surface. This leads to a faster build up of the internal compressive stress. In that situation, one can observe the growth of the wrinkles in real time (in the range of a few minutes). Under these conditions the observed buckling of the film even forms a honeycomb structure (see Fig. 4b) in the region of the light mask. This behaviour is very similar to the crack pattern formation investigated by Yuse et al. [11]. In that work it was shown that the crack pattern depends critically on the propagation speed of the cracks. For slow speeds, straight to regular cracks were observed, whereas for fast propagation speeds the cracks started to split into two and more branches.

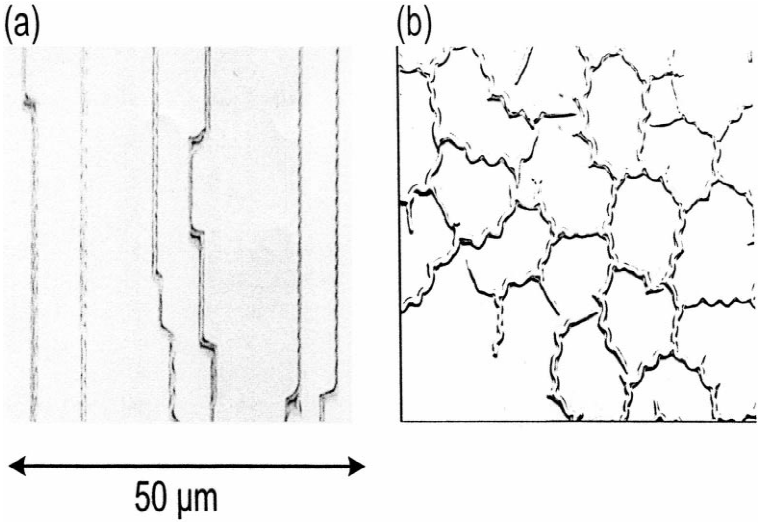

Fig. 4. Optical microscope images of wrinkle patterns for different growth velocities. The pattern shown in (a) was formed over several hours, while the structure in (b) built up within a few minutes. Obviously, the pattern is only guided by the nanostructured dopant concentration if the compressive stress is introduced slowly. Related effects have been observed in crack formation [11].

5. Conclusion

In this paper we report the first successful realization of structured doping in the lateral direction using light forces. The indication for the success in the realization of a laterally structured doped material is a characteristic wrinkle pattern formed on the film. The pattern shows long straight wrinkles where the material is periodically doped and reveals a honeycomb pattern where no structured doping has occurred. We have shown experimentally that the buckling of MgF_2:Cr films is due to compressive stress built up via the absorption of water in the film.

The extension of structured doping to true three-dimensional lithography is straightforward by combining the demonstrated lateral patterning with the standard structuring techniques in growth direction, e.g. flux switching. We are currently working on this extension of atom lithography.

Acknowledgements

This work was funded by the Deutsche Forschungsgemeinschaft (SFB 513) and the Optik-Zentrum Konstanz.

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