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Feature width miniaturization in atom nanolithography with double standing wave layers

Pingping Zhang (张萍萍)*, Yan Ma (马艳), and Tongbao Li (李同保)

Department of Physics, Tongji University, Shanghai 200092, China

*Corresponding author: zpp_7890@163.com

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Periodic nanostructures spaced by half of the wavelength can be obtained by the technology of laser-focused atomic deposition. Experimental result with single standing wave layer is presented, with a periodicity of 213 ± 0.1 nm, a height of 4 nm, and a feature width of 64 ± 6 nm. To further minimize the feature width, focusing and depositing characteristics of double standing wave layers are numerically simulated with optimized particle optics model. It is shown that the spherical aberration is reduced significantly. The predicted feature width is 18.2 nm and the height is approximately 12.6 nm when the powers of the two standing wave layers are 6 and 14 mW, respectively. Well-defined line occurs even when the full-width at half-maximum (FWHM) of transverse angular spread reaches 0.5 mrad.

Figure 2 shows the atomic force microscope (AFM) image of the deposited sample with SL in our group. The wavelength of the laser in vacuum is 425.55 nm with respect to $^{52}\text{Cr}$ atomic transition $S_3 \rightarrow P_4$. The $1/e^2$ radius of the Gaussian beam ($w_0$) is roughly 100 μm. The laser frequency for deposition is blue shifted 500×2 μMHz by an acoustic optic modulator from the resonance. The total input power of the laser is 180 mW, including laser frequency stabilization of 24 mW, laser cooling of 31 mW, and laser focusing of 15 mW. To avoid the diffraction, the silicon substrate is separated from the center of the Gaussian beam by 1.5 $w_0$. After a deposition time of 62 min, the sample is taken out from the vacuum chamber and then examined by AFM. The AFM image of the sample shows the surface three-dimensional (3D) topography of a 5×5-μm area and a height of 4 nm. Periodic chromium lines with mean periodicity of 213 ± 0.1 nm is obtained, and the feature width presented is 64 ± 6 nm. Although the deposited lines of the nanograting are well-defined, the feature width is far from satisfactory.

The arrangement of DL is shown in Fig. 3. The intensity profile of light field is given by

$$I(x, z) = I_1 \exp \left( \frac{-2\pi^2}{w_1^2} \right) \sin^2 (kx + \varphi) + I_2 \exp \left( \frac{-2\pi^2}{w_2^2} \right) \sin^2 (kx),$$

(1)

where $D$ is the separation between the two standing waves, $w_1$ and $w_2$ denote the $1/e^2$ radius of the two laser beams, respectively, and $k = 2\pi/\lambda$, where $\lambda$ is the wavelength. $I_1 = 2P_1/\pi w_1^2$ and $I_2 = 2P_2/\pi w_2^2$ represent the peak intensities of the two standing waves, respectively.
The overlap and the interference can be neglected when $P_1$ and $P_2$ are the powers of the two laser beams. The overlap and the interference can be neglected when $d > 3 \omega_1$. $L$ denotes the distance between the substrate and the Gaussian center of the first SL. $\varphi$ represents the phase of the two layers, which is set to be zero for simplicity.

The optical potential associated with the dipole force in low-intensity and large detuning regime is

$$U = \frac{\hbar \Delta}{2 \ln \left( 1 + \frac{I(x,z)}{I_s} \right) \left( \frac{\Gamma^2}{\Gamma^2 + 4 \Delta^2} \right)},$$

where $\Delta$ is the detuning, $\eta$ is Plank’s constant divided by $2 \pi$, $\Gamma$ is the natural width of the transition, and $I_s$ is atomic saturation intensity. The motion equation of neutral particles in double standing wave light field is given by a set of differential equations

$$\begin{cases}
  x' = \alpha \\
  \alpha' = \frac{1 + \alpha^2}{2(\Delta^2 + \pi^2)} \left( \frac{\partial U}{\partial x} - \frac{\partial U}{\partial z} \right),
\end{cases}$$

where $E_0$ represents the kinetic energy.

Experimentally the atomic beam scattering from the atom oven is precollimated by an aperture intersecting the atomic beam and further collimated by laser cooling techniques. Generally the longitudinal velocity $v_z$ of the chromium beam obeys the Maxwell-Boltzmann statistics and the initial transverse divergence defined by $\alpha = v_x/v_z$ exhibits a Gaussian spread, where $v_x$ is the transverse velocity. The combined density probability relation between $v_z$ and $\alpha$ can be written as

$$P(v_z, \alpha) \propto v_z^d \exp \left( -\frac{v_z^2}{2v_0^2} \left( 1 + \frac{\alpha^2}{\alpha_0^2} \right) \right),$$

where $v_0$ is governed by the oven temperature through

$$0.5m v_0^2 = 0.5k_B T_{oven},$$

where $k_B$ is Boltzmann constant and $T_{oven}$ represents the temperature of the atomic oven. The angle $\alpha_0$ is given by

$$\alpha_0 = \alpha_{FWHM} / \sqrt{\sqrt{2} - 1},$$

where $\alpha_{FWHM}$ is obtained through fluorescence experiments. Furthermore, naturally occurring chromium includes approximately 84% $^{52}$Cr that is free of hyperfine structure. The other isotopes such as $^{53}$Cr and $^{54}$Cr do not couple to the laser field. The portion of isotopes is sensitive to experimental geometry during the cooling process. Here we accept the worst case of 16.2%.

Optimized particle optics model[15] where the initial condition is stochastically selected is used to simulate the deposition process in the double Gaussian SLs. The initial condition $(x_i, v_i, \alpha_i, T_i)$ for each trajectory is chosen on a grid with Monte Carlo method. Obviously the initial position $x_i$ is uniformly distributed. The longitudinal velocity $v_i$ and the initial transverse angle $\alpha_i$ just obey the combined probability density relation defined by Eq. (4). $T_i$ determines whether an atom scattering out from the oven is $^{52}$Cr or not. If a $^{52}$Cr atom comes out, we would resort to differential Eq. (5) to trace the corresponding trajectory; otherwise the trajectory should be a straight line because there is no coupling between the laser field and the atom. At the image plane, a histogram of the probabilities is accumulated as a function of position, resulting in a predicted flux distribution at the surface.

The spontaneous emission rate for $^{52}$Cr is $\Gamma = 5 \times 2\pi$ MHz and the saturation intensity $I_s = 85$ mW/cm$^2$. The detuning is $\Delta = 250 \times 2\pi$ MHz blue shifted from the resonance. Let $T_{oven} = 1823$ K, $\alpha_{FWHM} = 0.16$ mrad, $\omega_1 = \omega_2 = 60 \mu$m, $d = 3\omega_1$ and $L = 4.2\omega_1$ respectively. Obviously best focusing process occurs when the two focal planes coincide with the substrate. From the viewpoint of particle optics theorem, the laser power required for bringing the focal plane at the object plane is
given by [11]

\[ P_{\text{focus}} = \alpha \frac{\pi E_0 I_s \Delta}{\eta l^2 k^2}, \]  

(7)

where \( \alpha \) is the excitation parameter. According to Eq. (7), the laser powers that bring the two focal planes at the substrate are \( P_1 = 6 \text{ mW} \) and \( P_2 = 14 \text{ mW} \), respectively.

Figure 4 shows the calculated trajectories of atoms in DL and its corresponding feature widths. As can be seen from Fig. 4(a), the spot size of \( f_2 \) is much smaller than that of \( f_1 \), which we believe arises from the reduction of the spherical aberration. In this configuration, the first layer prefocuses the atoms towards the minima of the sinusoidal potential of the second layer. The light intensity of the second layer \( I_2(x, z) \) is proportional to \( \sin^2(kx) \) which can be expanded as

\[ I_2(x, z) \propto \sin^2(kx) \]

(8)

When the prefocused atoms cross the second standing wave, they see closely the parabolic part of the potential which should result in a reduction of the overall spherical aberration. In addition, the feature widths obtained in Fig. 4(b) for both SL and DL cases are 30.5 and 18.2 nm, respectively, when the total amount of the deposited atoms is \( 5 \times 10^4 \) in one period. The heights are approximately 8.5 and 12.6 nm for both cases. In comparison with the result with SL shown in Fig. 4(b), a significant reduction of feature width for DL case occurs. Also the height is enhanced considerably. Thus with the prefoocusing scheme, a much better deposition result appears, which makes DL a rather competitive candidate for the following experiment.

Since DL mechanism offers a far stronger focusing property than SL, we may expect focusing with relatively large transverse angular distributions. Figure 5 shows the transverse angular dependence of the feature width for both SL and DL cases. The circles represent the trend of SL. The predicted flux is taken from the histogram at \( f_1 \) which corresponds to SL. As the angular increases from zero, the feature width shows a rapid increase. When \( \alpha_{\text{FWHM}} \) is 0.38 mrad, the feature width reaches as large as 100 nm, which is improper for the experiment because the transverse kinetic energy exceeds the depth of the potential well. The squares correspond to the trend of DL. The slope is much smaller that that of SL. As can be seen from Fig. 6(a), well-defined line can still be seen even when \( \alpha_{\text{FWHM}}=0.5 \text{ mrad} \) for DL case, with a feature width of 68.5 nm. While in Fig. 6(b), the feature width for SL reaches as large as 146.3 nm. Thus demand for laser cooling scheme in DL configuration may be reduced considerably.
In Conclusion, latest experimental result with SL is presented in this letter. The feature width obtained is $64 \pm 6 \text{nm}$ and the height of the deposited lines is 4 nm. In order to further reduce the feature width and improve the quality of the focusing process, focusing and depositing characteristics of DL are numerically studied via optimized particle optics model. In comparison with SL, a significant reduction in feature width is predicted even when the transverse angular spread is relatively large. Thus DL is a competitive candidate light mask for the atom lithography experiment.

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