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Frequency multi-mode lens for atoms and molecules: Application to nanofabrication

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We present a frequency multi-mode lens with corrected spherical aberration suitable for focusing of both atoms and molecules. The lens is formed by superposing of several harmonics of standing light waves in such a way that to create harmonic potential for all particles crossing the light field. The application to nanolithography is discussed emphasizing the possibility to improve the contrast of deposited structures.

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1. Introduction. The manipulation of atoms with optical fields is a key tool of atom optics. Specific configurations of optical fields can be built to closely resemble optical elements manipulating light rays in conventional optics such as lenses, mirrors, beamsplitters, diffraction gratings, etc. At present due to considerable progress in cooling of molecules [1 – 4] the techniques of optical manipulation developed for atoms can be extended to molecular beams. The focused molecular beams as well as atomic beams may be used in nanolithography for deposition of nanometre-scaled patterns on suitable substrates. In nanolithography with atoms the focusing effect is achieved by exploiting the interaction of atoms with standing light wave resulting in a space modulation of atomic beam in the transverse direction [5]. Thus, the distribution of light intensity is mapped onto the distribution of atoms in a substrate. Similar approach can be used to realize nanofabrication with molecules. Moreover, molecular nanofabrication can be potentially even more advantageous since molecules usually have larger mass and shorter deBroglie wavelength resulting in smaller diffraction sport size.

Focusing with light fields suffers from aberrations smearing focal spot. As in the case of conventional optics, where only paraxial rays can be point-focused, in atom lithography sharp focusing is achieved only for atoms moving near the minima of the standing wave (SW) potential, where the potential is approximately harmonic. These atoms form useful structure, while the atoms crossing the potential far from the minima contribute to the background. This distortion is similar to the spherical aberration in conventional optics. The main consequence of the spherical aberration is the low contrast of the deposited structures, which means that the height of the background is larger than the height of the structures. For example, for Al the contrast is found to be 0.15 [6], for Fe it is equal to 0.3 [7], and for Cr it is 0.8 [8].

The straightforward way to reduce the influence of the spherical aberration is the use of a mechanical mask in addition to optical focusing [9, 10]. This mask blocks non-paraxial particles resulting in a rather sharp focusing. However, such aperturing is unpractical for nanolithography since it inevitably reduces particle flux and thereby decreases the efficiency of deposition. Therefore, one should look for alternative approaches providing for correction of the spherical aberration without loosing particles.

In Ref. [11] it is proposed to use a combination of two SWs separated by some distance to correct the spherical aberration. The first SW prefocuses particles towards the minima of the second potential. Optimizing the intensities of SWs the background has been reduced in approximately two times as compared to a single SW.

This idea of using complex field configurations to improve focusing can be further developed to achieve even better results. In this paper, we propose a scheme of particle focusing based on proper combination of available optical fields so that all particles in the beam feel potential very close to harmonic one.

It should be noted that nanolithography with molecules has not yet been demonstrated. However, due to the similarity of focusing mechanisms the issues typical for atom lithography will certainly appear in molecular lithography too. Since the methods to solve these problems should be suitable both for the atoms and the molecules later on they will be referred as particles.

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2. Frequency multi-mode lens. Before start to discuss focusing with complex light fields let us briefly recall principles used in focusing of particles with a standing light wave. Let us consider two-level particles having mass m and moving in x direction (Fig.1). The

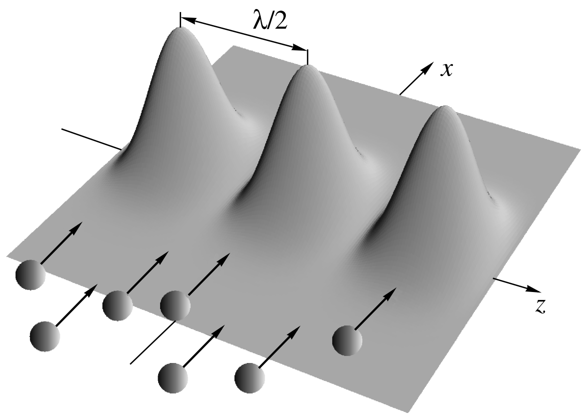

Fig.1. Standing wave potential

SW is formed in z direction due to the interference of two counter-propagating laser beams with frequencies ω. Assuming gaussian profiles of the laser beams the intensity of the lattice field is given by

$$I(x,z) = I_0 \exp(-x^2/w_x^2)\cos^2\left(\frac{\omega}{c}z\right), \quad (1)$$

where I_0 is the maximum intensity, w_x is the beam width in x direction, c is the velocity of light. Here we assume that $w_x \ll w_y$. This field creates periodic potential for particles

$$U(x,z) = \frac{\hbar\gamma^2}{8\delta I_s} I(x,z) = U_0 g(x) I_0 \cos^2\left(\frac{\omega}{c}z\right). \quad (2)$$

Here $g(x) = \exp(-x^2/w_x^2)$, $U_0 = \hbar\gamma^2/8\delta I_s$, where γ is the radiative decay rate, δ is the detuning between the laser frequency and particle transition, and I_s is the saturation intensity.

The potential (2) acts on a particle beam as an array of lenses with apertures $\lambda/2$, where λ is the wavelength of the lasers forming the SW. Near its minima at $z_m = m\lambda/2$, where m numbers potential wells, this potential can be approximated by harmonic functions. We restrict our consideration to a single lens extends from $z = -\lambda/4$ to $z = \lambda/4$. Let axis x goes through the center of the lens. Then the force acting on the particles in the vicinity of the potential minimum is proportional to the distance from the axis x: $F_z = 2U_0 I_0 g(x) k^2 z$. We assume that initially particles move parallel to the x axis with the same longitudinal velocities v_x (monochromatic beam). After the interaction with SW these particles will have a velocity in a transverse direction $v_z = 2U_0 I_0 g(x) k^2 w_x z/mv_x$. If the width of the laser beams is small enough (regime of thin lens), so that longitudinal velocities v_x of the particles do not change during the interaction with the SW, the time required for particles with different initial z coordinate to reach the axis x does not depend on this coordinate. As a result the particles moving in the vicinity of the potential minimum parallel to the axis x cross this axis at the same point $x_f = mv_x^2/2U_0 I_0 g(x) k^2 w_x$.

The particles moving in the region where the potential is unharmonic are subject to the weaker and nonlinear transverse force. As a result they are focused at the points situated farther than x_f. This effect is in direct analogy with spherical aberration known in conventional optics. To construct particle lens free from spherical aberration the linear transverse force should be achieved for all particles traveling within the standing wave period. Such a potential can be produced from the experimentally available potentials as follows.

Any periodic continuous function $f(z+2l) = f(z)$ can be decomposed in a Fourier series. If $f(z)$ is an even function it is represented as a cosine series

$$f(z) = \frac{a_0}{2} + \sum_{n=1}^{\infty} a_n \cos\left(\frac{n\pi}{l}z\right), \quad (3)$$

with expansion coefficients

$$a_n = \frac{1}{l}\int_{-l}^{l} f(z)\cos\left(\frac{n\pi}{l}z\right)dz, \quad n = 0,1,2\ldots \quad (4)$$

Aiming to construct parabolic potential we are interested in decomposition of parabolic function for $z \in [-l,l]$, which reads

$$z^2 = \frac{l^2}{3} + \frac{4l^2}{\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\cos\left(\frac{n\pi}{l}z\right), \quad (5)$$

where $2l$ should be interpreted as an aperture of a single lens. Thus, the required ideal potential can be written

$$\tilde{U}(x,z) = U_0 I_0 g(x)\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\cos\left(\frac{n\pi}{l}z\right). \quad (6)$$

The constant part of the potential has been ignored since it does not contribute to the force acting on particles in the transverse direction.

The simplest way to build the potential (6) is to use a combination of potentials (2) produced by SWs at different frequencies ω_n. The potential (6) formed by several SW fields with different frequencies will be referred as frequency multi-mode lens.

To form this lens one can take N ($N \to \infty$) plane waves with different frequencies ω_n traveling in positive direction of axis z and N identical waves traveling in opposite direction. The intensity of the field produced by these waves can be written as

$$I = \sum_{n=1}^{N} I_{0n}^2 g(x) \left[1 + \cos\left(\frac{2\omega_n}{c}z + \xi_n\right)\right] +$$
$$+\frac{c}{4\pi}\langle \sum_{n,m=1}^{N} E_{0n}E_{0m}g(x)\cos[(\omega_n-\omega_m)t+(\xi_n-\xi_m)]\times$$
$$\times \cos\left(\frac{\omega_n}{c}z\right)\cos\left(\frac{\omega_m}{c}z\right)\rangle_\tau, \qquad (7)$$

where E_{0n} and $I_{0n}=cE_{0n}^2/8\pi$ are the amplitude and the intensity of the nth wave, respectively. ξ_n is the phase determining relative position of different contributions. $\langle ...\rangle_\tau$ denotes the time averaging over the atom-field interaction time τ. The first term in this expression describes the interference of the counter-propagating waves at the same frequencies and the second term corresponds to the interference of the waves at different frequencies. The latter term oscillates at optical frequency and therefore after averaging over τ vanishes. Thus, the intensity of the resulting field is the sum of the intensities of the SWs with frequencies ω_n and intensities I_{0n}. The potential produced by this field resembles that given by Eq. (6) provided that

$$\omega_n = \frac{\pi c}{2l}n, \quad \xi_n = (n+1)\pi, \quad I_{0n} = \frac{I_0}{n^2}. \qquad (8)$$

Eq. (8) sets the relation between frequencies, phases and intensities of different frequency modes – standing waves harmonics. Thus, the particle lens free of spherical aberration can be constructed combining large number of SW harmonics.

As mentioned above to provide for the harmonic potential so many SW harmonics as possible are required. However, in practice only restricted number of harmonics can be used. There are basically two reasons for that. First, it might be difficult to generate many harmonics with sufficient intensity. Second, it could be difficult to find particles interacting equally well with all harmonics. In order the frequency multi-mode lens works it is necessary that the particles have several almost equidistant levels. It is worthnoting, that molecules meet this condition especially well since they have equidistant vibrational levels. Therefore, the frequency multi-mode lens could be especially effective for focusing of molecules.

Figure 2 demonstrates that the limited number of SWs is sufficient to provide for harmonic potential. This figure shows the transverse force acting on particles in the potentials created by different number of SW harmonics. Dotted and dashed curves correspond to *3* and *5* harmonics. For comparison, the solid curve shows the force in a single SW. It is seen that the light field which is the superposition of even five harmonics provides for the force very close to linear one. Therefore, one can expect good correction of spherical aberration focusing particle beam with only few SW components.

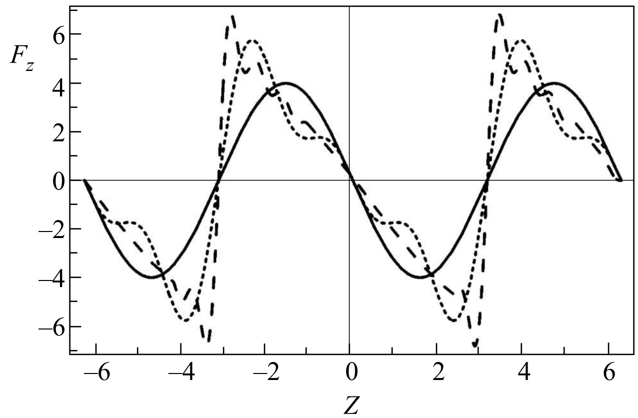

Fig.2. Transverse force acting on particles in the potential created by *1* (solid curve), *3* (dotted curve) and *5* (dashed curve) SWs

Let us denote the parameter describing the interaction of the particle with the n-th SW as U_{0n} and the number of the SWs providing for the potential (6) is to be close to harmonic one as N_h. Then the focusing potential is finally written as

$$\tilde{U}(x,z) = g(x)\sum_{n=1}^{N_h} U_{0n}I_{0n}\cos\left(\frac{\omega_n}{c}z + \xi_n\right). \qquad (9)$$

3.Particle focusing. In order to study focusing with the frequency multi-mode lens constructed with several SW harmonics it is convinient to solve numerically the differential equations for the deviation of particles from $z=0$ axis, $z(x)$, and the slope of the tracjectory, $\theta = dz(x)/dx$,

$$\theta = \frac{dz(x)}{dx}, \qquad (10)$$

$$\frac{d\theta(x)}{dx} = \frac{1+\theta^2}{2(E-\tilde{U})}\left[\theta\frac{d\tilde{U}}{dx} - (1+\theta^2)\frac{d\tilde{U}}{dz}\right]. \qquad (11)$$

Here, E is the kinetic energy of the particles before the interaction with the laser field, the potential $\tilde{U}(x,z)$ is given by Eqs (9) and (8). To simplify the problem it is assumed that the particles interact equally well with all SW harmonics, so that $U_{0n}=U_0$. In this case, the potential $\tilde{U}$ and the energy E can be measured in units of $U_0 I_0$. Coordinates are measured in units of $\lambda/4$.

The result of the modeling is shown in Fig.3. To be more illustrative the trajectories of particles within two elementary lenses are presented. For modeling $E \approx 10^5$

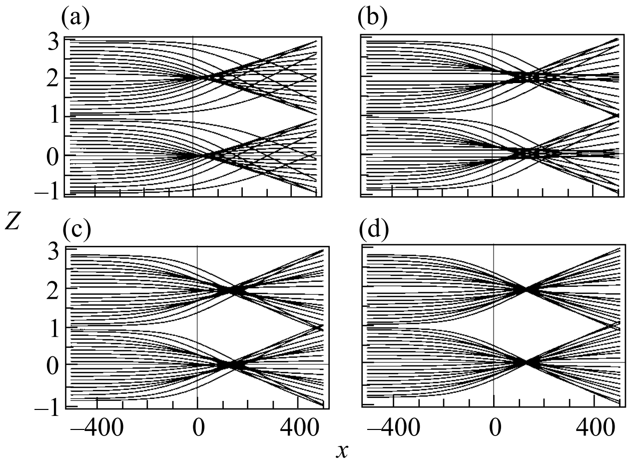

Fig.3. Focusing of particles with the potential created by *1* (a), *2* (b), *3* (c) and *5* (d) SW harmonics

has been used. Fig.3a demonstrates focusing with the lens created by a single SW. It is seen that only trajectories of particles moving in the vicinity of the axis of the lens are point-focused. All other particles form the background. In this case, one has two alternatives: to deposit useful structures of relatively small size against a strong background (in the figure the substrate should be placed somewhere at $x = 50$) or to reduce the background due to the significant increase of the structures width. In latter case the substrate should be positioned somewhere at $x = 200$. The increase of the number of SW harmonics results in a gradual decrease of the background (Fig.3b (2 harmonics), c (*3* harmonics), and d (*5* harmonics)). It is seen that the lens formed by *3* SW harmonics already provides for a good enough focusing ensuring high contrast of the deposited structures.

For quantitative description of the focusing one can introduce the so called localization factor. We define it as a mean-square deviation of the particles positions from the axis x

$$L(x) = \frac{1}{N_t} \sum_{i=1}^{N_t} z_i^2(x, z_i'). \tag{12}$$

Here, N_t is the number of trajectories inside a single lens with the aperture $2l$, $z_i(x, z_i')$ describes the trajectory of a particle with initial coordinates $x \to -\infty$ and $z = z_i'$, which can be found from Eqs. (10). Introducing continuous linear density of trajectories $\rho(z')$ one can replace summation in Eq. (12) with integration

$$L(x) = \frac{1}{N_t} \int_{-l}^{l} z^2(x, z') \rho(z') dz' = \frac{1}{2l} \int_{-l}^{l} z^2(x, z') dz'. \tag{13}$$

The right-hand side of this equation is obtained assuming uniform distribution of the trajectories, so that $\rho(z') = N_t/(2l)$. Figure 4 represents the localization factor as a function of x coordinate in the vicinity of its global minimum. The solid curve shows localization in the case of the focusing with a single SW, dashed and dotted curves corresponds to the focusing with the lenses formed by 2 and 3 SW harmonics, respectively. Using frequency multi-mode lens formed by several SW harmonics allows one to improve the localization in approximately two times. Particle lens formed by larger number of harmonics does not provide for the better localization: in Fig.4 the curve corresponding to 3 harmonics coincides with that corresponding to 5 harmonics.

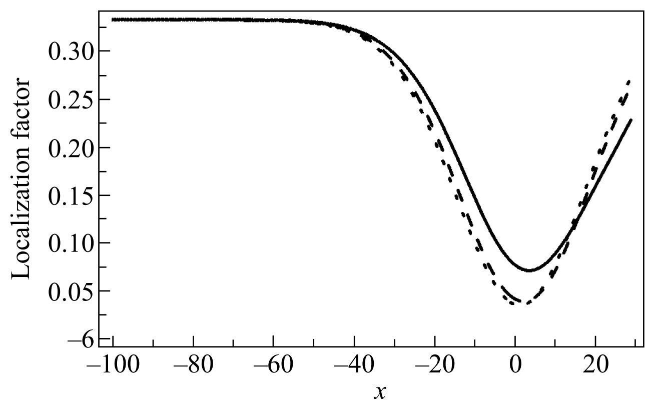

Fig.4. Localization factor for *1* (solid curve), *2* (dashed curve), *3* and *5* (dotted curve) SW harmonics

To estimate the height of the background the particle density at the focal plane can be calculated. Assuming uniform distribution of particles before interaction with the lens field the particle density can be written as

$$p(x_f, \tilde{z}) = \frac{1}{2l} \int_{-l}^{l} \delta[\tilde{z} - z(x_f, z')] dz'. \tag{14}$$

Here $(x_f, \tilde{z})$ is the particle coordinates in the focal plane. Fig.5 shows the particle density at the focal plane for focusing with *1* (solid curve), *3* (dashed curve) and *5* (dotted curve) SW harmonics. The inset represents zoomed right wing of the curves. In the case of a single SW one observes narrow enough central part of the distribution against strong background. The increase of the number

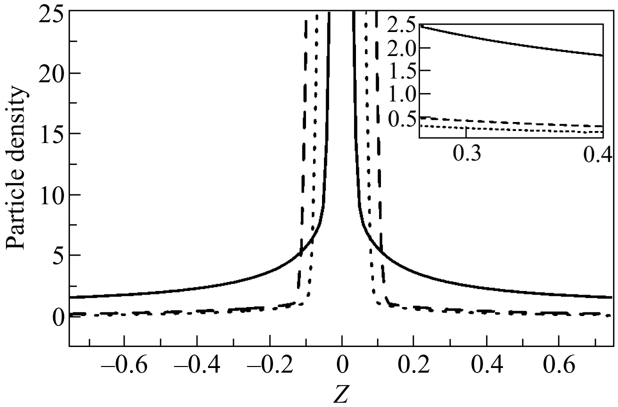

Fig.5. Density of particles focused with *1* (solid curve), *3* (dashed curve) and *5* (dotted curve) SW harmonics

of harmonics from *1* to *5* results in the reduction of the background by a factor of ~ 5. In addition, one can see some broadening of the central part due to the particles formerly producing the background. It is seen from the inset that for *5* harmonics the particle density outside the central part becomes smaller than *1*, which means that the probability to find particles at these points is close to zero.

4. Conclusion. To conclude, the frequency multi-mode lens for focusing of atoms and molecules is proposed. The advantage of this lens is the reduced spherical aberration. The correction of spherical aberration is achieved due to the combination of several standing wave harmonics providing for the potential close to the parabolic one. This lens can be especially effective for focusing of molecules. The application of the frequency multi-mode lens for the deposition of periodic nanostructures is discussed. It is shown that the focusing of particles with the lens formed by *5* SW harmonics allows one to enhance the contrast of the deposited structures in *5* times as compared with the focusing with a single SW.

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