Formation of adatom density gratings on a substrate by channeling of an atomic beam through a pulsed-laser standing wave under a completely off-resonant condition

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We show that a transversely uncooled atomic beam can be focused by a microsecond laser standing wave to form adatom density gratings on a substrate with good contrast ratio under a completely off-resonant condition ($\Delta\omega/\omega \sim 1$). © 1997 Optical Society of America

Two of the most exciting areas of research in atomic physics and optical physics are the cooling and manipulation of atoms with lasers. In the past decade these two aspects of laser interaction with neutral atoms have led many groups to produce periodic atomic-density distributions or density gratings by channeling a laser-cooled atomic beam (in the transverse direction) through a laser standing wave.[1,2] Recently, this technique was demonstrated[3–6] as a possible new scheme for submicrometer or even nanometer-scale lithography. Another useful application of these atomic-density gratings is in the study of surface-diffusion kinetics of adatoms.[7] One can follow the decay of the diffraction of a probe laser beam from these density gratings as a result of adatom diffusion.[8–11] The decay rate can be shown to be linearly proportional to the diffusion rate. So far the atomic- or molecular-density gratings used in surface-diffusion studies have been produced solely by laser-induced thermal desorption with the interference pattern of a laser pulse pair.[9,11,12] The thermal-desorption method is problematic when adatoms can no longer be thermally desorbed without optically damaging the substrate surface by the heating laser pulses. Therefore the laser standing-wave method complements the laser-induced thermal-desorption method in producing adatom-density gratings for the surface-diffusion studies.

In making the laser standing-wave method a versatile technique, it is desirable that its effectiveness not depend critically on the spectroscopic specifics of the atom in question so that one can use one (inexpensive) laser system to produce a density grating of any material. The current laser standing-wave technique does not meet this requirement, as it exploits sharp atomic transitions of atoms and thus relies on the tunability and monochromaticity of cw lasers. In the studies reported by Timp and co-workers and by Solomon *et al.*, one tunable laser was used to transversely cool the atomic beam, and a second tunable laser was used to form the standing wave for channeling of the atom beam.[1–5] The effectiveness of the transverse cooling and the subsequent channeling require a near-resonance condition such that the frequencies of the cw lasers with extremely narrow linewidths must be tunable within a range 10^6–10^9 Hz near a sharp atomic-transition frequency. This condition requires not only very detailed information on the atomic spectra but also single-frequency laser systems that can be finely tuned near the atomic transition ($\Delta\omega/\omega \sim 10^{-6}$). So far only alkali atoms, chromium, and aluminum have been successfully focused with laser standing waves.[1–5] As spectroscopic information on a metal atom must be known in detail and appropriate single-frequency laser systems are required for sufficient focusing and cooling, the current focusing technique will be problematic if we are to produce density gratings of atoms at will based the needs of materials research.

The spectral dependence of the laser standing-wave channeling can be lifted if one can work in a completely off-resonant condition with $\Delta\omega/\omega \sim 1$. In this case much less atomic spectral information and at the same time much less sophisticated laser systems are needed. The severe price to pay, however, is (1) the loss of a near-resonance enhancement of laser standing-wave focusing power by a factor of $\omega/\Delta\omega \sim 10^6$ and (2) the loss of the transverse laser cooling of atomic beams before they are channeled through a laser standing wave and as a result a potentially complete loss of contrast. In this Letter we show that in a completely off-resonant condition with $\Delta\omega/\omega \sim 1$ the focusing power lost by a cw laser standing wave can be restored by use of a pulsed-laser standing wave with a much higher electric field. We propose an experimentally practical arrangement that implements this concept and is applicable to all atoms or atomic clusters. We show that even with a transversely uncooled atomic beam one should be able to produce adatom density gratings on a substrate with good contrast ratios.

We first note that for a thermally produced atomic beam the longitudinal velocity of the atoms is roughly $v_l = 7.4 \times 10^4$ cm/s. Since the width of an optical standing-wave region is 0.1 cm or less, the interaction time is only $\tau_i = L_t/v_l = 1.3$ μs. Consequently one should be able to use a microsecond pulsed-laser standing wave with a much higher electric field to regain the lost focusing power in a completely off-resonant condition. This method is readily achievable with existing commercial laser systems. We now

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estimate the single-pulse energy of a microsecond laser that achieves the same focusing power as cw lasers in the near-resonant condition.[1–5] Let a beam of atoms propagate along the z direction. A laser standing wave is set up along the y direction with a width $L_t = 0.1$ cm. At an off-resonant condition only the dipole force on an atom in the beam that is exerted by the standing-wave electric field needs to be considered. The time-averaged dipolar potential energy of the atom is given by $U = -\mathbf{p}\cdot\mathbf{E} = -\alpha'|\mathbf{E}|^2$, and

$$U(y) = -\frac{8I_0}{c}\left(\frac{\alpha'}{\epsilon_0}\right)\sin^2\left(\frac{2\pi y}{\lambda}\right). \tag{1}$$

We use the mks unit system. Here α' is the atomic polarizability at the off resonant condition. I_0 and λ are the intensity and the wavelength of the incident laser for the standing wave, respectively. ϵ_0 is the vacuum permittivity. From the potential energy we find the force $f(y) = -\partial U(y)/\partial y$ on the atom:

$$f(y) = -\frac{16\pi I_0}{c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right)\sin\left(\frac{4\pi}{\lambda}y\right). \tag{2}$$

After the atom passes through the standing-wave region, its transverse velocity in the y direction changes by $\Delta v_y = f(y)\tau_i/m = -(16\pi I_0\tau_i/mc\lambda)(\alpha'/\epsilon_0)\sin(4\pi y/\lambda)$, where m is the mass of the atom. As a result the density of the atomic beam develops a periodic structure with a periodicity of one-half the standing-wave wavelength. Let a substrate be at a distance of $S_1 = 0.2$ cm behind the standing-wave region. An adatom density grating with the highest contrast ratio is formed when

$$\frac{16\pi I_0\tau_i}{mv_l c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right) \approx \frac{\lambda}{4\pi S_1}. \tag{3}$$

To estimate the atomic polarizability of atoms at the off-resonant condition, we use the bulk-phase optical constants $\tilde{n} = n + i\kappa$ of the material at the wavelength of the standing wave. It is easily shown that $\alpha'/\epsilon_0 = (n^2 - \kappa^2 - 1)/N_b$, where N_b is the bulk-phase density of the atoms. At $\lambda = 1.0$ μm we find that $\alpha'/\epsilon_0 = 2\times10^{-28}$ m^3 for Si, $\alpha'/\epsilon_0 = 6\times10^{-28}$ m^3 for Ni, and $\alpha'/\epsilon_0 = 2\times10^{-28}$ m^3 for Cu. From expression (3), and using $\lambda/4\pi S_1 = 4\times10^{-5}$ and $\tau_i = 1.3$ μs, we find that the required laser intensities would be $I_0 = 3\times10^7$ W/m^2 for Si, $I_0 = 2\times10^7$ W/m^2 for Ni, and $I_0 = 7\times10^7$ W/m^2 for Cu. Using a 100-μs pulsed-laser system (from Light Age, Inc., Somerset, N.J.) and focusing the beam to a waist of $d_{\mathrm{sw}} = 0.1$ cm, we would need a single-pulse energy of only 2–10 mJ, which is very modest considering that commercially available laser systems have single-pulse energies of as much as 10 J at 100 μs. This result means that even if the atomic polarizabilities are underestimated by as much as 2 orders of magnitude we should still be able to form high-quality density gratings of any material with the available single-pulse energy.

We next propose a practical experimental arrangement that implements this concept and use this setup to illustrate how the contrast of the atomic-density grating is mostly preserved even with a transversely uncooled atomic beam. The proposed setup is shown in Fig. 1 (not to scale). A pulsed atomic beam is produced by nanosecond laser ablation of a material target of interest placed at a distance $S_0 = 20$ cm from the optical standing-wave region. The ablation laser beam is focused on the target to a diameter of $d_0 = 25$ μm. An aperture of diameter $d_A = 0.1$ cm is placed in the atomic-beam path right before the standing-wave region. The pulsed atomic beam is normally incident upon a substrate that is placed at $S_1 = 0.2$ cm from the center of the standing-wave region. For the optical standing wave a 100-μs pulsed-laser beam at $\lambda = 1.0$ μm intercepts the atomic beam along the y direction. The laser beam is focused by a cylindrical lens to have a waist $d_{\mathrm{sw}} = 0.1$ cm in front of the substrate. A flat reflection mirror is placed at the opposite side of the substrate from the incident laser. The reflected and the incident beams form the standing wave that channels the atomic beam. The arrival of the pulsed atomic beam and the pulsed optical standing wave is synchronized by control of the firing of the ablation and the standing-wave lasers. We take the effect of thermal velocity distribution in the transverse direction into consideration by allowing the emission angle of atoms from the target to vary uniformly by $d_A/2S_0 = 2.5$ mrad. Let α be the emission angle projected in the y–z plane of an atom from a point on the target at y coordinate y_i. The atom will arrive on the substrate at a location along the y direction:

$$y_f = y_i + \alpha_i(S_0 + S_1) - \frac{16\pi I_0\tau_i}{mv_l c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right)S_1 \times \sin\left[\frac{4\pi}{\lambda}(y_i + \alpha_i S_0)\right]. \tag{4}$$

From the center of the ablated area on the target with $y_i = 0$, we obtain the location of the deposited atoms as a function of emission angle α_i:

$$y_{f0} = \alpha_i(S_0 + S_1) - \frac{16\pi I_0\tau_i}{mv_l c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right)S_1\sin\left(\frac{4\pi}{\lambda}\alpha_i S_0\right). \tag{5}$$

If we choose $(16\pi I_0\tau_i/mv_l c\lambda)(\alpha'/\epsilon_0) = (1.1\lambda/4\pi S_1)$ such that the beam is optimally focused at the substrate surface, Eq. (5) gives a periodic density distribution along the y direction on the substrate that

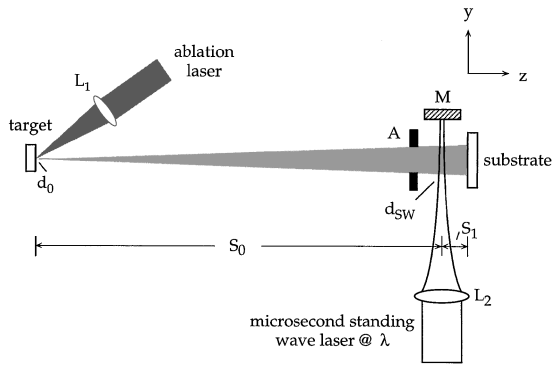

Fig. 1. Proposed experimental arrangement for production of adatom density gratings on a substrate by focusing a transversely uncooled atomic beam with a microsecond pulsed optical standing wave. L_1, spherical lens; L_2, cylindrical lens; M, reflector; A, aperture.

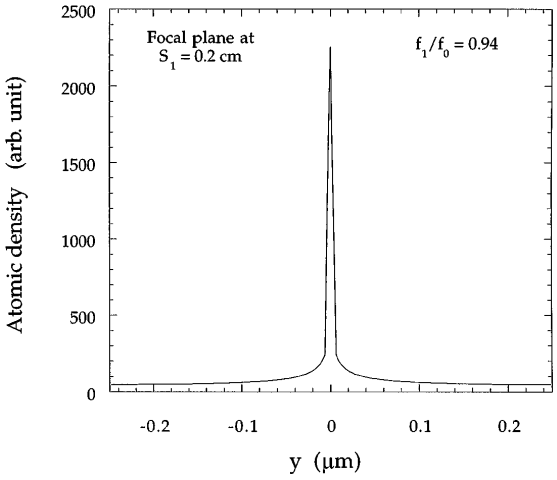

Fig. 2. Calculated atomic density grating on a substrate within one period (0.5 μm). The substrate is at a distance $S_1 = 0.2$ cm behind the standing wave. The ratio of the first spatial Fourier coefficient f_1 to the mean density f_0 is 0.94. We used a constant longitudinal velocity v_l in the calculation.

peaks at

$$y_{f0}^{(m)} = \frac{m\lambda}{2}\left(1 + \frac{S_1}{S_0}\right), \tag{6}$$

with a periodicity $(\lambda/2)(1 + S_1/S_0)$. The distribution within one period is shown in Fig. 2. In this calculation we used a constant longitudinal velocity v_l similar to the procedure that McClelland *et al.* used to calculate Fig. 6A of Ref. 4. Our result of an extremely narrow linewidth reproduces that of McClelland *et al.* In reality the linewidth is broadened when the Maxwell–Boltzmann distribution of longitudinal velocities is taken into consideration, as was shown by McClelland *et al.*[4] From a point away from the center of the target with a finite y_i, Eq. (4) also gives a periodic density distribution on the substrate, except that the peaks at

$$y_f^{(m)} = y_{f0}^{(m)} - \frac{y_i S_1}{S_0} \tag{7}$$

are shifted from $y_{f0}^{(m)}$ by a distance $y_i S_1/S_0$. Since $S_1/S_0 = 1 \times 10^{-2}$ and y_i is less than $d_0/2 = 12.5$ μm, we have $y_i S_1/S_0 \leq 0.125$ μm, which is still much smaller than the periodicity $(\lambda/2)(1 + S_1/S_0) \approx 0.5$ μm of the atomic-density grating. For surface-diffusion studies, one is mostly interested in the first spatial Fourier component, $f_1(y) = f_1 \cos(4\pi y/\lambda)$, of the atomic-density grating. Assume that the coefficient f_1 from a point source at the center of the target is f_{10}. When the point source is spread out to a circular spot with a diameter $d_0 = 25$ μm while the total emitted atomic flux is maintained, it is easily shown that $f_1 = f_{10}[\sin(2\pi d_0 S_1/\lambda S_0)/(2\pi d_0 S_1/\lambda S_0)] \approx 0.64 f_{10}$. This result is still very good for surface-diffusion studies.[11]

With the target distance $S_0 = 20$ cm from the standing wave and the Maxwell–Boltzman longitudinal velocity distribution centered at $v_l = 7.4 \times 10^4$ cm/s, the temporal width of the pulsed atomic beam is roughly 100 μs. With a 100-μs laser standing wave, the channeling effect is valid for the entire atomic beam.

In conclusion, we have shown that, under a completely off-resonant condition with $\Delta\omega/\omega \sim 1$, one can focus a transversely uncooled atomic beam with a microsecond pulsed-laser standing wave to form an adatom density grating on a substrate with a high contrast ratio. We have suggested an experimental arrangement using commercially existing laser systems that implements this idea. We have shown that such a method of producing adatom density gratings is applicable to all atoms or atomic clusters with the same experimental arrangement, which is particularly significant if we apply this technique to the study of surface-diffusion kinetics of adatoms based on materials-research requirements rather than on the availability of detailed atomic spectral information and high-resolution laser systems. This method complements the laser-induced desorption technique for making adatom density gratings and thus extends the optical diffraction method for surface-diffusion investigation to essentially all adsorbate–substrate systems.[8]

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