Fabrication of nano-structural arrays by channeling pulsed atomic beams through pulsed-laser standing-waves under off-resonant condition

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We show that it is feasible to produce one- and two-dimensional nano-structure arrays by passing microsecond pulsed atomic beams through microsecond laser standing-wave patterns under completely *off-resonant* condition. This method enables fabrication of vertically heterogeneous nanostructures such as multilayers with one pulsed laser system. © *1999 American Institute of Physics.* [S0003-6951(99)03204-0]

In recent years, new forms of materials that are made from conventional ones but reduced in one dimension (1D) (i.e., thin films) or two dimensions (2D) (i.e., thin wires) or even three dimension (3D) (i.e., dots or particles) have drawn considerable attention in both the material research community and industry. These new material forms not only promise ever increasingly large scale integration of conventional devices, they also promise new devices since different physics are expected to be at work when the length scales along the reduced dimensions are of the order of nanometers (10^{-9} m). In particular, nano-particles that are confined in all three dimensions have unique (mesoscopic) electronic properties and in turn optical and in some cases magnetic properties that differ from those of their bulk (macroscopic) or atomic (microscopic) counterparts. The uniqueness presumably comes from the quantum confinement of electrons that is in between those of the bulk and the atoms and most importantly can be tailored or ''tuned.''

Many lithographic techniques have been developed to fabricate nano-structures. Most involve irradiation of photo-resist-covered templates such as Si wafers with photons, electrons, or ions and then chemical etching to form patterns with nanometer-scale structures. Subsequent evaporation of functional materials onto the patterned templates leads to desired nano-structures. Chou and coworkers recently developed a nano-imprinting technique, a nonirradiative method, to form patterns on polymer templates or photo-resists before further material processing that leads to desired nano-structures.[1] Timp and coworkers and others demonstrated another scheme for a nanometer scale lithography by channeling continuous wave (cw) atom beams through laser standing-wave patterns.[2–5] Without the need of physical masks, this method has been used to produce 1D and 2D nano-structural arrays over macroscopic scale either directly or with photo-resists as a means to further reduce the sizes of nano-structures.[2–11] In order to reduce the smearing effect due to the transverse velocity spread of atomic beams from spatially extended thermal oven sources, the beams have to be transversely cooled before channeling.[2–4] For laser cooling to be effective, the frequency of the cooling laser ω has to be tuned to a suitable transition frequency ω_0 of the atoms within 100 MHz or $|(\omega_0-\omega)/\omega_0|\sim 10^{-6}$. The frequency of the standing-wave laser is also within a few hundred MHz from ω_0. The predominant channeling effect from the laser standing-wave pattern is the induced electric dipole force on the atomic beam. The magnitude of the force is inversely proportional to the frequency detuning or $|\omega_0/(\omega_0-\omega)| \sim 10^6$. Implementation of such a technique at the present time depends upon the detail atomic spectral information and the availability of suitable cw lasers whose frequencies are within a few GHz or a few hundred MHz from a usable atomic transition frequency. So far only alkali atoms, chromium, and aluminum have been successfully focused with laser-standing waves.[2–4] This is clearly problematic if we are to use this technique as a viable means to fabricate nano-particle arrays or nano-wires based upon needs of materials research.

It is desirable that the effectiveness of an atom channeling method does not depend critically on spectroscopic specifics of the atom in question so that one can use *one* laser system to fabricate nano-structural arrays of various materials. The spectral dependence can be lifted if one uses laser-standing waves at a completely *off-resonant* condition with $|(\omega_0-\omega)/\omega_0|\sim 1$. The prices to pay or the problems are (1) at the same laser intensity, the focusing power is reduced by a factor of 10^6; (2) one can no longer pre-cool atomic beams in the transverse direction(s) before channeling. The latter can cause a complete loss of contrast and deem such a proposition useless. The first issue has been addressed in an earlier publication.[12] In this communication, we will address the second issue and show how one can fabricate large arrays of nano-wires or nano-particles by using microsecond atomic beams through pulsed laser-standing waves.

Since a thermal atom traverses through a sub-millimeter wide laser standing-wave region in less than a microsecond, the focusing power is easily restored by ''squeezing'' the optical energy in an otherwise cw laser beam into short bunches with a time duration close to the atom transit time. The squeezing is accomplished by using a microsecond pulsed laser. A proposed experimental setup is shown in Fig. 1.[12] A microsecond pulsed atomic beam is produced by laser ablation of a material target with a nanosecond or preferably microsecond optical pulse. The target is placed at a distance S_0 from the optical standing wave region. The ablation laser beam is focused on the target to a small diameter of d_0. An

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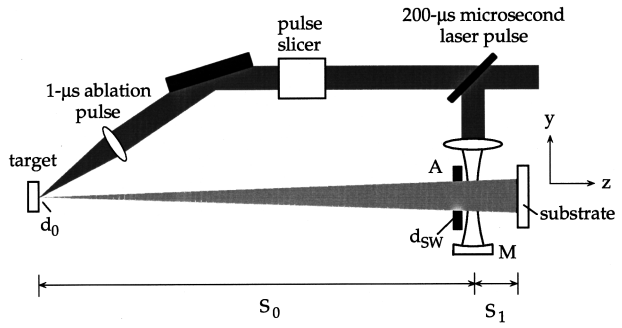

FIG. 1. Proposed experimental arrangement to fabricate nano-wire or nano-particle arrays on a substrate by focusing a transversely *uncooled* pulsed atomic beam with 200 μs laser standing waves. The pulsed atomic beam is produced by ablation with 1 μs laser pulses. *M*: spherical reflector. *A*: aperture with a diameter d_A.

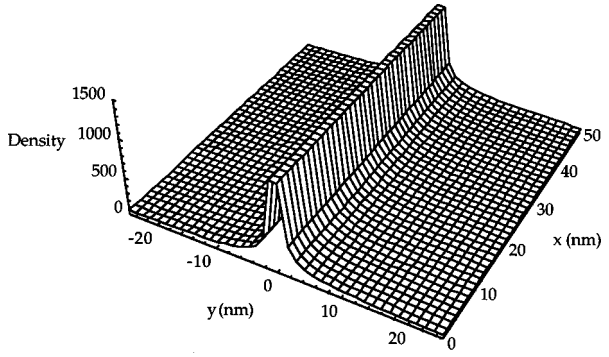

FIG. 2. Calculated atomic density profile (or nano-wire) near the center of the substrate (as shown in Fig. 1) over an area of 50×50 nm. We have used a constant longitudinal velocity $v_{/\!/}$ in the calculation.

aperture of diameter d_A is placed in the atomic beam path (along *z*-direction) right before the standing wave region so that only normally incident portion of the atom plume is channeled through. The atomic beam is afterward incident upon a substrate placed at a short distance S_1 from the standing wave region. For channeling, we propose to use a 200-μs laser pulse and focus it with a cylindrical lens along *y*-direction to a small waist d_{sw} in front of the aperture and reflect the beam from the opposite side with a spherical reflector. The reflected and the incident beams form a standing-wave pattern that channels the atomic beam. The arrivals of the pulsed atomic beam and the pulsed optical standing wave are easily synchronized. We can also use a portion of the same microsecond laser to simultaneously form a standing wave along the orthogonal direction (*x*-direction) or two additional standing waves that are 120° apart in the $x-y$ plane to make nano-particle arrays with square or triangular lattice.

Without any transverse cooling however, it is not obvious that the atom density distribution fabricated by such a laser standing-wave channeling method would still have features of nanostructure.[2–4] It is important to note that the smearing effect due to the Maxwellian velocity distribution in the transverse direction is because (1) the width of the standing-wave region is finite so that an atom may traverse horizontally a distance much larger than tens of nanometers inside the standing-wave region and thus the atom lens effect is severely distorted; (2) the beam source is extended in the transverse direction(s). By controlling these two factors, we show it feasible to obtain nano-wire or nano-particle arrays without the benefit of transverse cooling.

We restrict our consideration to the case where there is only one standing-wave pattern along *y*-direction. Let α_i be the emission angle of an atom in the $y-z$ plane from a point on the source target at *y*-coordinate y_i. The range of the emission angle is determined by d_A/S_0. To eliminate the first cause of the smearing effect, we require that the width of the standing-wave region is small, such that the maximum lateral displacement of the atom inside the standing-wave region $d_A d_{SW}/2S_0$ is less than a few tens nanometers. In this case, the atom arrives on the substrate at

$$y_f = y_i + \alpha_i(S_0+S_1) - \frac{16\pi I_0 \tau_i}{m v_{/\!/} c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right) S_1 \sin\left[\frac{4\pi}{\lambda}(y_i+\alpha_i S_0)\right], \quad (1)$$

m, $v_{/\!/}$, α', $\tau_i = d_{SW}/v_{/\!/}$ are the mass, the longitudinal velocity, the optical polarizability (in MKS unit), and the transit time of the atom, respectively. ϵ_0 is the vacuum permittivity. I_0 and λ are the peak intensity and the wavelength of the standing-wave laser, respectively. From the center of the source target with $y_i = 0$, we have

$$y_{f0} = \alpha_i(S_0+S_1) - \frac{16\pi I_0 \tau_i}{m v_{/\!/} c\lambda}\left(\frac{\alpha'}{\epsilon_0}\right) S_1 \sin\left[\frac{4\pi}{\lambda}\alpha_i S_0\right]. \quad (2)$$

From Eq. (2), we find that atoms emitted from the center of the source target form a periodic array of nano-wires on the substrate at a distance $S_1 \approx (\lambda/4\pi)/[16\pi I_0 \tau_i / m v_{/\!/} c\lambda(\alpha'/\epsilon_0)]$. When under or critically focused, the *n*th maximum of the atom density appears precisely at $y_{f0}^{(n)} = (n\lambda/2)(1+S_1/S_0)$ and the periodicity is $(\lambda/2)(1+S_1/S_0)$. We numerically calculate the nano-wire profile using $S_0 = 500$ nm, $S_1 = 1$ mm, $d_{SW} = 40$ μm, and $\lambda = 1000$ nm. The profile near the center of the aperture on the substrate is shown in Fig. 2 over a 50×50 nm area on the substrate. The full-width-at-half-maximum (FWHM) is less than 4 nm. If we use two standing-wave patterns that are orthogonal to each other, the calculated atom density profile changes from a nanowire as shown in Fig. 2 to a nano-particle with a nominal width of 4 nm. In Fig. 3, we show the nano-wire profiles at different locations on the substrate. The FWHM of the density maximum remains less than 4 nm over an area of 1 mm diameter. Beyond the distance of $r = 0.63$ mm from the center of the aperture, the width of the atom density peak decreases quickly. At this distance, the displacement of the atom in the standing-wave region is $r d_{SW}/S_0 = 60$ nm and is significantly larger than a few nanometers. With the laser wavelength at $\lambda = 1000$ nm, it is not beneficial to reduce the width of the standing-wave region much below 40 μm, as the length of the confocal region over which the beam waist remains roughly the same as the minimum $d_{SW} = 40$ μm will be reduced to less than 1 mm. Since the smearing effect does not depend on the wavelength of the standing-wave laser, it is feasible to increase the density of the resultant nano-wire array without changing the size of the wires by using shorter wavelengths.

We can control the second cause of the smearing effect by controlling the effective size of the source target. From a point y_i away from the center of the target, Eq. (1) gives rise

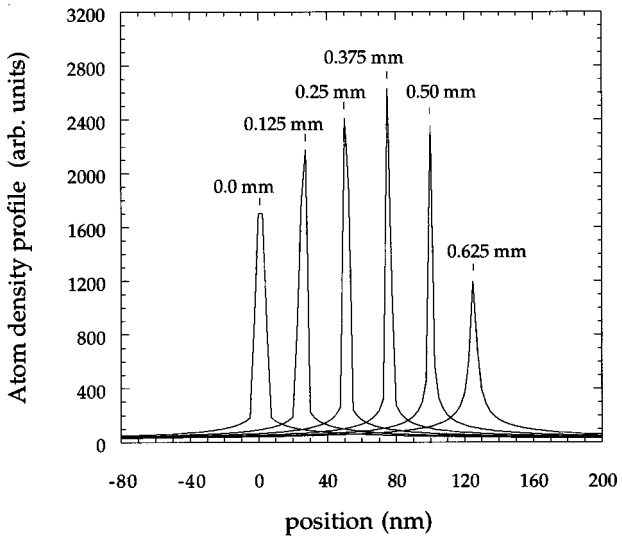

FIG. 3. Calculated atomic density profiles as a function of the distance away from the center of the substrate. The profiles are shifted from one another for clarity. Beyond $r=0.625$ mm, the width of the nanowire broadens drastically.

to a similar nano-wire distribution on the substrate but the maxima are now shifted from $y_{f0}^{(n)}$ to[12] $y_f^{(n)}=y_{f0}^{(n)}-y_i S_1/S_0$. Since y_i varies from $-d_0/2$ to $+d_0/2$, the nano-wire profiles shown in Figs. 2 and 3 are broadened by $d_0 S_1/S_0$. Using $S_0=500$ mm and $S_1=1$ mm, we need to limit the diameter of the laser ablation area on the source target to $d_0=5$ μm in order to limit the broadening to under 10 nm. This is achievable with atomic beams produced by laser ablation methods.

We next consider the standing-wave laser system and the ablation laser system that optimize the effectiveness and yield of such a nano-fabrication method.

Microsecond laser system for forming standing-wave patterns: This deals with the fact that the ablation-produced atomic beam has a thermal longitudinal velocity distribution. With the target distance $S_0=500$ mm from the standing-wave region and a Maxwellian distribution of the longitudinal velocity centered at $v_{/\!/}=7.4\times10^4$ cm/sec, the temporal width of the ablation-produced atomic beam is roughly 250 μs. It is thus desirable to use a pulsed laser system with a duration around $\tau_{SW}=250$ μs and a single-pulse energy up to 100 and 200 mJ.[12] Such a system will be adequate to channel atoms or molecules of most solid materials. It should be possible to operate such a laser system at a repetition rate as high as tens of kHz to boost the deposition yield. Because the pulse duration of the standing-wave laser is of hundreds of microseconds, we can shape the temporal envelope of the pulse electro-optically such that $I_0\tau_i/v_{/\!/}=I_0 d_{SW}/v_{/\!/}^2$ in Eq. (2) and in turn the focusing power is made a constant for the entire beam. As a result the thermal longitudinal velocity distribution will not further smear out the contrast.

Microsecond laser system for laser ablation: To boost the deposition yield, it is best to use ablation pulses with the longest possible duration. The latter is limited to 1 μs, since for most elemental solids and alloys the thermal diffusion lengths during 1 μs are around 5 μm. For a 1 μs laser pulse focused down to a diameter of $d_0=5$ μm, we only need a single-pulse energy of 0.1 mJ to heat up the source target by 3000 K. The 1 μs ablation laser pulses may be obtained by slicing a portion of the standing-wave laser pulses electro-optically.

We now discuss further the usefulness and limitation of such a technique. Since the useful area with nano-structural arrays produced with this technique is only 1 mm^2 at a time, it may be problematic to use it for large-scale high-yield fabrication. Using a 5 kHz microsecond laser and an ablation deposition rate of 0.01 monolayers per pulse, we expect to make 50 monolayers per second per square millimeter. This will present a severe limit to large-scale yield fabrication. At this point, it seems more suited for mechanistic investigation of collective, as well as individual behaviors of nano-wire or nano-particle arrays. For this purpose, this technique complements current *e*-beam lithographic techniques. Another possible limitation is that there is always a layer of uniformly deposited materials in between sharp nanostructures due to aberration of the laser standing wave as atom lenses. However if the desired mesoscopic properties of nano-wire or nano-particle arrays are distinctly different from the macroscopic properties of the bulk-like materials deposited in between, these ''background'' materials should not affect the performance or the characterization of the nanostructural arrays.

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