Appl. Phys. B 70, 657–660 (2000) / Digital Object Identifier (DOI) 10.1007/s003400000262

**Applied Physics B**
Lasers
and Optics

# Optical manipulation of group III atoms

**S.J. Rehse**[1], **R.W. McGowan**[2], **S.A. Lee**[1]

[1] Department of Physics, Colorado State University, Ft. Collins, CO 80523, USA
(Fax: +1-970/491-7947, E-mail: rehse@lamar.colostate.edu)
[2] Imation Enterprises Corp., 1 Imation Place, Oakdale, MN 55128-3414, USA

Received: 29 September 1999/Published online: 5 April 2000 – 

**Abstract.** We present details for the laser manipulation of group III atoms, specifically aluminum, gallium, and indium. The practical considerations of accomplishing this manipulation are discussed and alternative schemes are presented for each species. The possibility of using such an optical technique for composition modulation during semiconductor growth for the fabrication of quantum wire and quantum dot structures is also discussed.

**PACS:** 42.50.Vk; 03.75.Be; 81.15.Hi

In the last several years, the use of neutral atomic beams to fabricate nanoscale features has become an active area of research [1]. Neutral atoms are attractive because of their small de Broglie wavelengths ($< 1$ nm) and because the atoms have internal energy levels that can be accessed with lasers. It is possible to optically manipulate the atoms with laser light that is frequency-tuned close to an atomic transition [2]. In particular, a standing wave of near-resonant laser light can be used to form a periodic array of cylindrical lenses for atoms. If the standing-wave laser beam is positioned directly over a substrate, the atoms traveling though the laser beam experience a periodic light force and are focused into narrow lines during deposition onto the substrate [3]. This method is shown schematically in Fig. 1. Several atomic species have been optically focused, including sodium [3, 4], chromium [5–7], and aluminum [8]. Theoretical analysis of the physics underlying the optical cooling and focusing of the atoms predicts that it is possible to focus the atoms to less than 10 nm in width [9, 10]. To date, lines as narrow as 15 nm with a contrast of 10 : 1 have been achieved in Na [4]. Using multiple standing waves, a two-dimensional array of dots was demonstrated in Cr [6].

Direct deposition via optical focusing of neutral atoms is a novel approach to form periodic nanofeatures. The group III atoms (Al, Ga, and In) are technologically interesting, in that they are the primary constituents of III-V semiconductor devices. Optical focusing of group III atoms offers a unique opportunity for the controlled growth of nanowire and nanodot structures during molecular beam epitaxy (MBE). This paper discusses the requirements for the optical manipulation and focusing of these group III atoms and considerations for MBE growth.

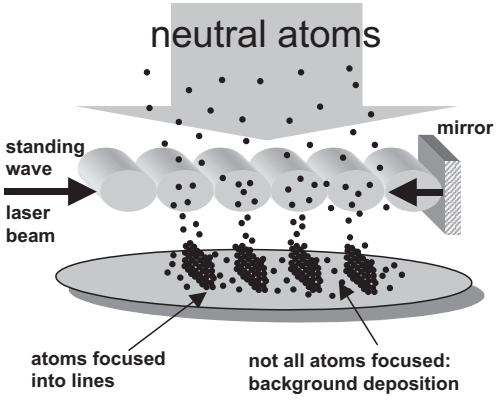


**Fig. 1.** Standing wave of laser light acts as an array of cylindrical lenses to focus atoms during deposition onto a substrate. Not all of the atoms can be focused, resulting in the formation of features on top of a background

## 1 Optical atom manipulation

There are two types of light forces that can be utilized for atom manipulation [2]: the spontaneous force and the dipole or gradient force. The spontaneous force is the result of momentum transfer from a light beam to an atom through the repeated absorption and subsequent spontaneous emission of photons. This is a dissipative force used extensively in atom cooling and trapping. The dipole force arises from the interaction between the induced atomic dipole moment and the local laser electric field. This is a conservative force that is the result of photon momentum transfer through the coherent process of absorption and stimulated emission. The dipole force is ideal for atom optics applications such as atom focusing.

Light-force focusing of neutral atoms was first demonstrated in 1992 by Timp et al. at Bell labs, who used a standing wave of laser light to focus a beam of Na atoms into parallel lines during deposition onto a Si substrate [3]. In this way a pattern was written directly onto a substrate without the use of any masks.

Numerical simulations of the conditions encountered in an actual atomic beam [9, 10] show that the deposited linewidth is primarily determined by the transverse collimation of the atomic beam. Therefore the divergence of the atom beam must be as close to zero as possible in order to achieve narrow lines. It is necessary to optically cool the transverse motion of the atomic beam to near zero before focusing. The cooling requirement makes the optical focusing of atoms more complicated. This is because in cooling, it is necessary to have a closed transition, i.e., one in which the spontaneous emission always returns an atom to the same state from which the absorption begins. If a non-closed transition is used the atoms will be lost to a different level after a few cooling cycles. Many atoms do not have a closed transition starting from the ground state, and this has been a limitation in extending the technique of laser focusing to all atoms.

## 2 Optical manipulation of the group III atoms

A group III atom has the electronic configuration $ns^2np$ and its ground state consists of two fine-structure levels, $np\,^2P_{1/2}$ and $np\,^2P_{3/2}$. There is no closed transition suitable for cooling from the true ground state, $np\,^2P_{1/2}$ level. However, we have identified a transition that starts from the second ground state $np\,^2P_{3/2}$ to the $nd\,^2D_{5/2}$ level which approximates a closed transition suitable for cooling. In a thermal atomic beam generated from an effusive oven source, the two ground states are statistically populated and some of the atomic population will be in the correct cooling state. A list of the relevant parameters for group III atoms is given in Table 1.

To make the focusing of group III atoms technologically important, it is necessary to examine the required results that would allow the potential formation of nanowires and nanodots during MBE growth. As a specific example, consider the formation of quantum wires in AlGaAs. A 30% change in the Ga density will produce a 0.25 eV difference in the band-gap energy. This should be sufficient for quantum confinement [11]. This corresponds to a contrast of 1 : 3 in the focused features. Here, contrast is defined as the ratio of the feature height (not including background) to the background. This is a very modest requirement and the presence of a background is not necessarily detrimental. Assuming Ga lines with a full width at half maximum of 20 nm, a 1 : 3 contrast ratio means that we need to focus $\approx 4\%$ of the total Ga population into the peaks. This point is very important for focusing group III atoms, since, in general, it is possible to manipulate only a fraction of the atoms in the atomic beam.

### *2.1 Light-force focusing of aluminum*

In 1995, we focused Al atoms using a cw UV laser [8]. This was the first time a group III atom had ever been optically manipulated. A cw dye laser was frequency doubled in an external cavity to generate tunable UV laser light in the 309-nm region. A 1D grating of Al lines was deposited onto a Si substrate and then analyzed with an atomic force microscope (Fig. 2). The grating lines were spaced by 155 nm, had a FWHM of $\approx 70$ nm and were approximately 3 nm tall. The successful manipulation of Al provides the basis for the present discussion of focusing gallium and indium.

In fabricating the Al grating, the standing-wave laser was red detuned 240 MHz from resonance. However the sodium [12] and chromium [9] groups have demonstrated much narrower grating features by using high-intensity, blue-detuned ($\approx 2$ GHz) standing waves. It is difficult to produce a high-intensity UV laser at the Al wavelength that can meet this requirement. This limitation suggests the use of alternative wavelengths for optical focusing, if possible. Such alternative transitions do exist in the group III atoms and will be discussed later, in the context of focusing gallium and indium.

**Table 1.** Relevant parameters for the optical manipulation of group III atoms

| Stable isotopes | $^{27}$Al | $^{69}$Ga | $^{71}$Ga | $^{115}$In | $^{113}$In |
|---|---|---|---|---|---|
| % abundances | 100% | 60.4% | 39.6% | 96% | 4% |
| Nuclear spin I | 5/2 | 3/2 | | 9/2 | |
| Ground-state fine-structure splitting | 0.014 eV | 0.103 eV | | 0.275 eV | |
| Thermal population @ 1500 °C | in $3p\,^2P_{3/2}$ $(F=4)$ 25% | in $4p\,^2P_{3/2}$ $(F=3)$ 22% | | in $5p\,^2P_{3/2}$ $(F=6)$ 8% | |
| Cooling and focusing transition | $3p\,^2P_{3/2}$ $(F=4)$ $\rightarrow 3d\,^2D_{5/2}$ $(F=5)$ | $4p\,^2P_{3/2}$ $(F=3)$ $\rightarrow 4d\,^2D_{5/2}$ $(F=4)$ | | $5p\,^2P_{3/2}$ $(F=6)$ $\rightarrow 5d\,^2D_{5/2}$ $(F=7)$ | |
| Transition wavelength | 309.4 nm | 294.4 nm | | 325.7 nm | |
| Saturation intensity $(m=F\rightarrow m=F+1)$ | 57 mW/cm$^2$ | 127 mW/cm$^2$ | | 78 mW/cm$^2$ | |
| Optical pumping transition | $3p\,^2P_{1/2}$ $(F=3,2)$ $\rightarrow 4s\,^2S_{1/2}$ $(F=3)$ | $4p\,^2P_{1/2}$ $(F=2,1)$ $\rightarrow 5s\,^2S_{1/2}$ $(F=2)$ | | $5p\,^2P_{1/2}$ $(F=5,4)$ $\rightarrow 6s\,^2S_{1/2}$ $(F=5)$ | |
| Transition wavelength | 394.5 nm | 403.4 nm | | 410.3 nm | |
| Alternative focusing transition | $3p\,^2P_{3/2}$ $(F=4)$ $\rightarrow 4s\,^2S_{1/2}$ $(F=3)$ | $4p\,^2P_{3/2}$ $(F=3)$ $\rightarrow 5s\,^2S_{1/2}$ $(F=2)$ | | $5p\,^2P_{3/2}$ $(F=6)$ $\rightarrow 6s\,^2S_{1/2}$ $(F=5)$ | |
| Transition wavelength | 396.2 nm | 417.3 nm | | 451.3 nm | |

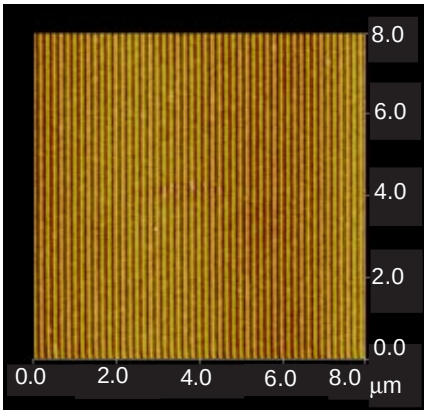


**Fig. 2.** AFM image of aluminum deposited on Si using a 309-nm cw laser. The single-beam laser power was 14 mW, waist diameter 55 μm, and detuned −240 MHz from resonance. The total grating area was 1 mm × 0.1 mm, and was deposited in 30 min. The *lines* are separated by 155 nm with an average peak height of 3 nm, and a fullwidth half maximum of 80 nm. The background is 20 nm

### *2.2 Light-force focusing of gallium*

The lower lying energy states of Ga are shown in Fig. 3. A transition that starts from the second ground state $4p\,^2P_{3/2}$ $(F=3)$, to the $4d\,^2D_{5/2}$ $(F=4)$ level, at 294.4 nm is suitable for both cooling and focusing. This light can be generated by frequency doubling a dye laser with an ammonium dihydrogen arsenate (ADA) crystal. The thermal population of this second ground state is ≈ 20%. The remaining atoms will form a background deposition that is unmodulated by the laser.

To increase the number of useful atoms in the beam, an optical pumping scheme may be utilized. A laser operating at 403.4 nm (a diode laser frequency doubled using a LBO or BBO crystal [13]) can be used to pump the atoms in the $4p\,^2P_{1/2}$ state to the $5s\,^2S_{1/2}$ $(F=2)$ state. From this 5*s* state, the atoms will then decay into either one of the two ground states. If the interaction time between the laser pump beam and the atom beam is sufficiently long, all the atoms that decay back to the $4p\,^2P_{1/2}$ state will eventually be pumped into the $4p\,^2P_{3/2}$ state. In this way, an atom beam can be prepared that has almost 58% of all the atoms in the requisite $4p\,^2P_{3/2}$ $(F=3)$ state.

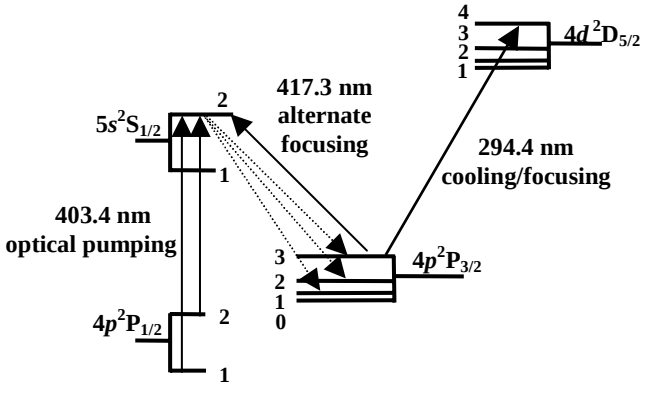


**Fig. 3.** Level diagram of lower-lying atomic structure in gallium, including the hyperfine structure. Energy splittings are not to scale

Ga has two stable isotopes, $^{69}$Ga and $^{71}$Ga, with natural abundances of 60% and 40%, respectively. The cooling transition in $^{71}$Ga is blueshifted 81 MHz from the transition in $^{69}$Ga [14]. Using an appropriately red-detuned laser, the $^{69}$Ga atoms could be cooled for subsequent focusing while the $^{71}$Ga atoms would contribute to the background deposition.

There is no reason why one has to use the same transition for cooling and for focusing. As mentioned before, laser focusing of atoms utilizes the dipole force. Since the laser is detuned sufficiently far from resonance to prevent spontaneous emission, any transition from the already cooled atoms would work for focusing. A particularly good transition is the $4p\,^2P_{3/2}$ $(F=3) \rightarrow 4s\,^2S_{1/2}$ $(F=2)$ at 417.3 nm. LBO or BBO crystals doubling a tunable diode laser can be used to generate this wavelength.

### *2.3 Light-force focusing of indium*

Indium has the most complicated hyperfine structure of the atoms listed in Table 1 because of its 9/2 nuclear spin. The $5p\,^2P_{3/2}$ $(F=6) \rightarrow 5d\,^2D_{5/2}$ $(F=7)$ transition at 325.7 nm can be used for both cooling and focusing. The hyperfine structure of the excited state has been measured by Zimmermann [15]. Laser light at this wavelength can be produced by utilizing a $LiIO_3$ crystal to frequency double a dye laser. There are several differences between indium and gallium. Indium has two stable isotopes $^{113}$In (4%) and $^{115}$In (96%). This isotope ratio is much more advantageous than gallium. However, the ground-state splitting of In is approximately 0.275 eV, roughly 20 times larger than Al and three times greater than Ga. This fact makes an optical pumping scheme extremely important, as an even smaller percentage of the atoms from a 1500 °C oven will be in the $5p\,^2P_{3/2}$ level. A pumping scheme similar to Ga could be utilized, with a BBO frequency-doubled diode laser used to produce the 410.3-nm light required for the pumping.

As in Ga, an alternative focusing possibility exists. The $5p\,^2P_{3/2}$ $(F=6) \rightarrow 6s\,^2S_{1/2}$ $(F=5)$ transition at 451.3 nm could be accessed with a $KNbO_3$ frequency-doubled Ti:sapphire laser [16]. The isotope shift from this transition was measured by Zaal et al. [17] to be 255.4 MHz ($^{113}$In is redshifted). Again, the desire to use a high-intensity focusing laser provides a strong motivation for utilizing this alternate transition.

## 3 Extension of optical focusing to crystal growth

### *3.1 Quantum devices in semiconductor heterostructures*

The group III atoms are the key building blocks of modern III-V semiconductor diode lasers, and the ability to directly focus Ga and In offers uniquely new opportunities for the fabrication of ultralow-threshold quantum wire and quantum dot lasers. The ability to control carrier confinement in III-V semiconductors is paramount in the design of such diode lasers. In quantum well lasers, one-dimensional quantum confinement of charge carriers in the active medium has already led to an order of magnitude reduction in the threshold current, with higher differential gain, narrower linewidths, and better wavelength stability with temperature when compared

to their bulk laser counterparts. In addition, a high flexibility in controlling the output wavelength is obtained by controlling the quantum well width and composition. The improvements in the performance in quantum well lasers are mainly due to the changes in the density of states and the modification of the band structure with increased confinement [18].

### *3.2 Laser focusing during MBE*

The optical focusing of group III atoms provides a fundamentally new approach for controlling the spatial composition of atoms during the growth of III-V heterostructures. Specifically, the direct optical focusing of Ga atoms during MBE growth of AlGaAs quantum well lasers leads to a lateral modulation of the Ga density. Since the band-gap energy of AlGaAs is reduced with increasing Ga composition, the lateral modulation of the Ga density results in the formation of lateral arrays of low-band-gap wires or dots separated by high-band-gap barriers. To accomplish this, the light beams for focusing would be introduced non-intrusively into a MBE chamber directly in front of the substrate. If the focusing laser is chopped on and off during growth, a composition-modulated heterostructure (layers of laterally density-modulated crystal sandwiched between layers of non-modulated crystal) could be created.

Quantum dots or wires so produced would be highly uniform in periodicity and in density modulation. The large number of such features that would be produced is also advantageous. A practical quantum dot device should incorporate a large number of dots to compensate for the very small optical radiation from each dot. The optical focusing technique writes a large number of dots simultaneously. As an example, consider two standing waves of laser beams crossing at 90° to form a two-dimensional optical potential. With a laser beam of 100 μm, approximately half a million dots can be deposited simultaneously in a matter of minutes.

### *3.3 Self-organized quantum dots*

Another interesting system for consideration is the growth of InAs strained layers on a GaAs substrate. The large lattice mismatch of 7% between these two materials leads to the growth of isolated 3D islands. The islands have quantum confinement dimensions ($\approx$ 10–30 nm) and are being considered as candidates for quantum dot lasers [19]. There is evidence that these islands (referred to as "self-organized quantum dots") form at the sites where indium aggregates. The very abrupt change from a 2D to a 3D growth mode occurs within a single monolayer, indicating that a 5%–10% change in the local In atom density will be sufficient to effect nucleation sites for quantum dot formation [20]. The optical focusing technique may create a periodic array of nucleation or "sticking" sites for the growth of quantum dots on the surface of the substrate. One problem with current strain-induced growth techniques of quantum dots is that the size and location of the isolated islands are uncontrolled. As mentioned before, uniformity in periodicity and density modulation are two of the advantages of using the optical focusing technique. This application potentially offers a way to exercise control over both the size and location of the quantum dots.

## 4 Summary

We have discussed the requirements for the optical manipulation of three group III atoms: Al, Ga, and In. Furthermore, we have considered the possibility of the incorporation of such an optical technique into semiconductor crystal growth with the goal of creating quantum wires or dots. The use of light-force manipulation of neutral atoms provides exciting possibilities in the creation of composition-modulated heterostructures.

*Acknowledgements.* This work was supported by the National Science Foundation under Grants No. PHY9312572 and No. PHY9732498. The authors would like to acknowledge Carmen Menoni, Prabhu Thiagarajan, and Gary Robinson for thoughtful discussions.

## References

1. J.H. Thywissen, K.S. Johnson, R. Younkin, N.H. Dekker, K.K. Berggren, A.P. Chu, M. Prentiss, S.A. Lee: J. Vac. Sci. Technol. B **15**, 2093 (1997)
2. For a discussion of light forces and applications, see for example the following special issues: J. Opt. Soc. Am. B **2**, 1707 (1985); J. Opt. Soc. Am. B **6**, 2023 (1989); Appl. Phys. B **54**, 321 (1992). See also: Proc. SPIE, Vol. 2995, ed. by M.G. Prentiss, W.D. Phillips (SPIE, Bellingham, Washington 1997)
3. G. Timp, R.E. Behringer, D.M. Tennant, J.E. Cunningham, M. Prentiss, K.K. Berggren: Phys. Rev. Lett. **69**, 1636 (1992)
4. V. Natarajan, R.E. Behringer, G. Timp: Phys. Rev. A **53**, 4381 (1996)
5. J.J. McClelland, R.E. Scholten, E.C. Palm, R.J. Celotta: Science **262**, 877 (1993)
6. R. Gupta, J.J. McClelland, Z.J. Jabbour, R.J. Celotta: Appl. Phys. Lett. **67**, 1378 (1995)
7. U. Drodofsky, J. Stuhler, B. Brezger, T. Schulze, M. Drewsen, T. Pa, J. Mlynek: Microelectron. Eng. **35**, 285 (1997)
8. R.W. McGowan, D.M. Giltner, S.A. Lee: Opt. Lett. **20**, 2535 (1995)
9. J.J. McClelland: J. Opt. Soc. Am. B **12**, 1763 (1995)
10. K.K. Berggren, M. Prentiss, G.L. Timp, R.E. Behringer: J. Opt. Soc. Am. B **11**, 1166 (1994)
11. S. Adachi: J. Appl. Phys. **58**, R1 (1985) We assume here that the background composition is $Al_{0.4}Ga_{0.6}As$, and the wire composition is $Al_{0.2}Ga_{0.8}As$
12. V. Natarajan, R.E. Behringer, D.M. Tennant, G. Timp: J. Vac. Sci. Technol. B **13**, 2823 (1995)
13. B. Beier, D. Woll, M. Scheidt, K.-J. Boller, R. Wallenstein: Appl. Phys. Lett. **71**, 315 (1997)
14. S.J. Rehse, W.M. Fairbank Jr., S.A. Lee: to be published
15. P. Zimmermann: Z. Phys. **233**, 21 (1970)
16. I. Biaggio, P. Kerkoc, L.-S. Wu, P. Gunter: J. Opt. Soc. Am. B **9**, 507 (1992). For details on how to achieve this frequency with an LBO crystal, see also: D. Woll, B. Beier, K.-J. Boller, R. Wallenstein, M. Hagberg, S. O'Brien: Opt. Lett. **24**, 691 (1999)
17. G.J. Zaal, W. Hogervorst, E.R. Eliel, J. Bouma, J. Blok: J. Phys. B **11**, 2821 (1978)
18. For a review of recent advances in quantum well lasers, see *Quantum Well Lasers*, ed. by P.S. Zory, Jr. (Academic Press, New York 1993)
19. T. Saitoh, A. Hashimoto, M. Tamura: J. Appl. Phys. **71**, 3802 (1992); J.M. Moison, F. Houzay, F. Barthe, L. Leprince, E. Andre, O. Vatel: Appl. Phys. Lett. **64**, 196 (1994); D. Leonard, K. Pond, P.M. Petroff: Phys. Rev. B **50**, 11687 (1994); Y Nabetani, T. Ishikawa, S. Noda, A. Sasaki: J. Appl. Phys. **76**, 347 (1994)
20. R.P. Mirin: private communication