Laser collimation of an atomic gallium beam

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We have laser collimated a gallium atomic beam in one dimension using the linear-perpendicular-linear polarization gradient technique. Operating on the cycling $4p\,^2P_{3/2}(F=3)\rightarrow 4d\,^2D_{5/2}(F=4)$ transition at 294.45 nm, the full angular divergence of the atomic beam was reduced to 0.3 mrad, corresponding to a transverse velocity of ±11 cm/s, about one-half the Doppler cooling limit. The dependence of the cooling efficiency on the laser detuning and power was investigated. Optical pumping of the atoms out of the $4p\,^2P_{3/2}(F=3)$ state by the cooling laser was observed. Repumping schemes were investigated and found to successfully repopulate the cooled $F=3$ hyperfine state.

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Laser standing waves have been used to focus neutral atoms into nanometer scale features during their deposition onto a substrate [1]. This direct atom lithographic scheme has been demonstrated in sodium [2], chromium [3,4], aluminum [5], and cesium [6]. In order to achieve the smallest feature size in direct laser focusing, the incident atomic beam must be collimated transversely to a very high degree, typically less than 1 mrad [7,8]. Laser cooling was used to provide a high degree of collimation without a significant loss of atom flux. In this technique, two counterpropagating laser beams intersect the atomic beam at right angles to generate a one-dimensional optical molasses [9,10]. If the polarization of the optical molasses varies across the path of the atoms, an additional force is exerted on the atoms. This polarization gradient force can cool the atoms to below the Doppler limit [11,12].

In this paper, we report on the one-dimensional laser collimation of a Ga atomic beam with the linear-perpendicular-linear (lin $\perp$ lin) polarization gradient configuration. Gallium is particularly interesting because it is a key element in modern III-V semiconductor diode lasers. The ability to directly focus Ga with laser light during molecular beam epitaxial growth of III-V heterostructures offers a unique method for modulating the Ga density in the growth plane [13]. Thus, the technique has the potential for forming large numbers of quantum wires and quantum dots in a controlled manner. Laser collimation of Ga atoms is the first step towards achieving this goal.

Ga has two stable isotopes, ^{69}Ga and ^{71}Ga, with an isotopic ratio of 60.4% to 39.6%. Both isotopes have nuclear spin 3/2. A cycling transition suitable for laser cooling is the $4p\,^2P_{3/2}(F=3)\rightarrow 4d\,^2D_{5/2}(F=4)$ transition at 294.45 nm, with a natural linewidth $\Gamma=25$ MHz and saturation intensity $I_0=129$ mW/cm^2. See Fig. 1. The Doppler cooling limit is $T_D=600\ \mu$K, corresponding to a rms velocity of 27 cm/s in one dimension. In this experiment we examined mainly the laser cooling of ^{69}Ga in the $4p\,^2P_{3/2}(F=3)$ state, even though ^{70}Ga was partially cooled as well. A laser-induced fluorescence spectrum of the $4p\,^2P_{3/2}\rightarrow 4d\,^2D_{5/2}$ transition is shown in Fig. 1(c) [14].

In our experiment, a thermal beam of Ga atoms was generated by a resistively heated effusive oven source. The $4p\,^2P_{3/2}$ state was metastable and located at 0.103 eV above the $4p\,^2P_{1/2}$ ground state. With an oven operating at 1300 °C, ~13% of the atoms in the beam were ^{69}Ga in the $4p\,^2P_{3/2}(F=3)$ state. An aperture precollimated the beam to 6.2 mrad, for a nominal transverse velocity of ±2.3 m/s. The most probable (longitudinal) speed of the atomic beam was 750 m/s. Figure 1(b) shows the arrangement of the atomic beam and the laser beams. The cooling laser beam intersected the atoms perpendicularly and was retroreflected to form a standing wave. The atoms traveled 34 cm down-

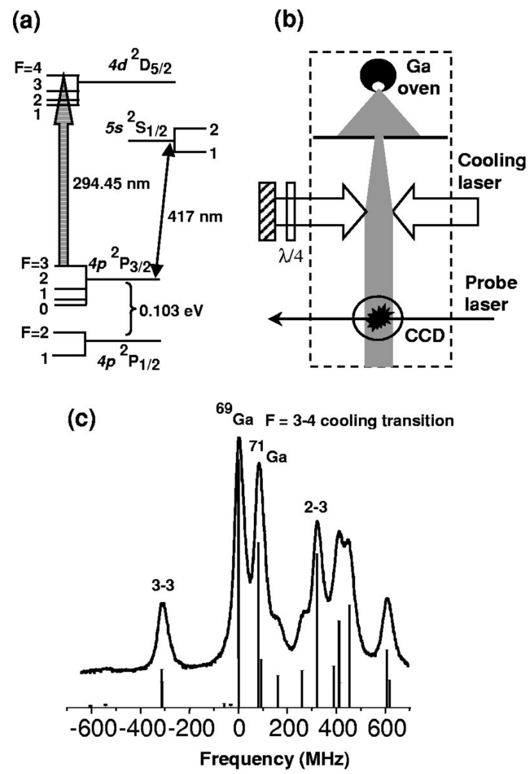

FIG. 1. (a) Energy-level diagram of Ga. (b) Schematic of the laser collimation experiment. (c) Laser-induced fluorescence spectrum of the $4p\,^2P_{3/2}\rightarrow 4d\,^2D_{5/2}$ transition together with the calculated peak locations for Ga (from Ref. [14]). The zero of the frequency scale is set at the $F=3\rightarrow 4$ cooling transition of ^{69}Ga. The other labels indicate relevant ^{69}Ga transitions.

stream where they were illuminated transversely by a weak probe laser to measure the amount of collimation. The probe laser-induced fluorescence from the atoms was collected with a lens and imaged onto a liquid-nitrogen-cooled uv sensitive charge-coupled device (CCD) camera. No external magnetic fields were applied to cancel the Earth's field.

The uv laser light was produced by frequency doubling a cw ring dye laser (Rhodamine 6G) in an external cavity. Utilizing a BBO crystal, ~90 mW of tunable uv light was generated [15]. However, a more typical operating power was around 70 mW. The laser frequency was stabilized by offset locking the fundamental dye laser to a nearby iodine hyperfine transition using saturation spectroscopy [15]. This apparatus used a double-passed acousto-optic modulator (AOM) to shift the laser frequency into resonance with iodine. Changing the drive frequency to the AOM allowed precise tuning of the laser frequency. In this experiment both the cooling beam and the probe beam had the same frequency. The cooling laser beam had a 1 mm$\times$5.5 mm elliptical cross section ($1/e^2$ diameters). It was linearly polarized and passed through a quarter-wave plate on the far side of the cooling region prior to retroreflection. This generated a lin $\perp$ lin polarization gradient in the cooling region. Typical powers in the cooling beam ranged from 40 to 70 mW. The probe laser, also linearly polarized, was circular in cross section with a $1/e^2$ diameter of 1.6 mm and was tens of μW of power.

The cooling laser was aligned perpendicular to the atomic beam by observing the fluorescence due to the incident and the retroreflected beams with a photomultiplier located above the cooling region. The cooling laser frequency was scanned over the transition. If the cooling beam was not perpendicular to the atomic beam, a double peaked fluorescence signal or a shift in the peak center was observed when the retroreflected beam was on. Iterative adjustments of the cooling laser direction were made to overlap the two fluorescence peaks. In this way the cooling laser was aligned perpendicular to the atomic beam to about 1 mrad. The probe laser beam was aligned parallel to the cooling beam by measuring the separation of the two beams on the input side of the atomic beam interaction chamber and at the output side after the beams had propagated 2 m. The beam centers were aligned to better than 1 mm over a 2 m distance, corresponding to less than 0.5 mrad deviation between the two beams.

It should be noted that in our experiment, the CCD images did not represent the true geometrical beam size for an uncooled atomic beam. The fluorescence signal of the atoms was dependent on the transverse Doppler shift and the detuning of the probe laser. At zero detuning, atoms near the central portion of the beam contributed more significantly to the fluorescence signal than the atoms in the wings, and the CCD image was narrower than the actual beam width. As the red detuning of the probe laser increased, the CCD images became asymmetric and broadened. However, when the atoms were cooled and the Doppler frequency shift is negligible, all the atoms responded equally to the probe and the CCD images represented the true geometrical sizes.

To determine the divergence and the size of the uncooled Ga beam independent of the CCD imaging, the atoms were deposited onto glass slides at various distances along the beam axis and the deposited spots were measured with an optical microscope. In the transverse dimension, the full angular divergence was measured to be 6.2 ± 0.1 mrad. The atom beam full width at half maximum (FWHM) was found to be 1.79 ± 0.03 mm at the center of the cooling region. In the limit of zero divergence, this would be the size of the atomic beam in the probe region and this is denoted as the geometric limit. The measured beam divergence and sizes were in good agreement with a ballistic trajectory calculation utilizing the configuration of the oven and aperture geometry based on the theory of effusive molecular flow [16].

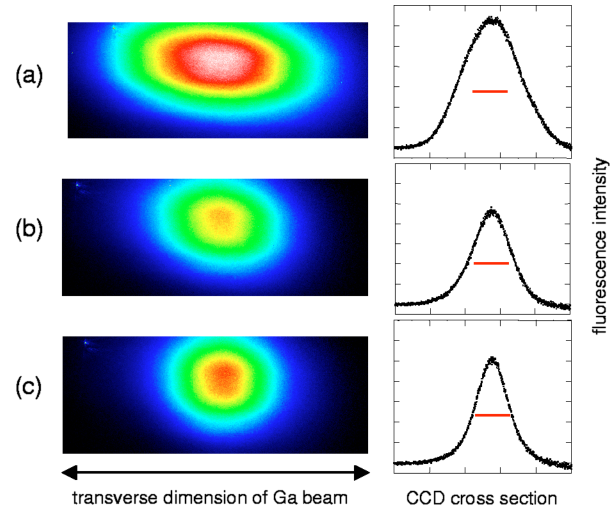

FIG. 2. (Color online) CCD images of laser-induced fluorescence from an atomic gallium beam: (a) no laser cooling, FWHM =3.72 mm, (b) cooling with Doppler molasses only, FWHM =2.30 mm, and (c) cooling with lin $\perp$ lin polarization gradient, FWHM=1.94 mm. Cooling laser 52 mW power, $\Delta=-12$ MHz.

The effect of laser polarization gradient cooling is presented in Fig. 2. CCD images of the atom beam illuminated by the probe laser are shown. In the fluorescence images, the atoms traveled from top to bottom and the probe laser illuminated the atoms from left to right. The cooling laser was 52 mW (peak $I/I_0=9$) and detuned -12 MHz from resonance. Next to the CCD images are intensity profiles taken through the middle of the fluorescence images. Recall that 1.79 mm was the atomic beam's transverse dimension at the center of the cooling region and was denoted earlier as the geometric limit. This limit is indicated by the horizontal mark on each intensity profile. Figure 2(a) shows the uncollimated, normally diverging atomic beam. A slight asymmetry in the fluorescence line shape due to the probe detuning is evident. Figure 2(b) shows the atomic beam with the cooling laser on, but the quarter-waveplate removed. This cooling was from the Doppler molasses only. Figure 2(c) shows the atom beam cooled with the lin $\perp$ lin polarization gradient configuration. Very little divergence of the atom beam occurred over the 342 mm flight path from the cooling region to the probe region. The measured FWHM was 1.94 mm, corresponding to a transverse velocity of 16 cm/s. This is below the Doppler cooling limit. The CCD image showed that the beam was more collimated and brighter on axis than with the Doppler molasses alone. This was a direct observation of the polarization gradient technique extending the cooling into the sub-Doppler regime.

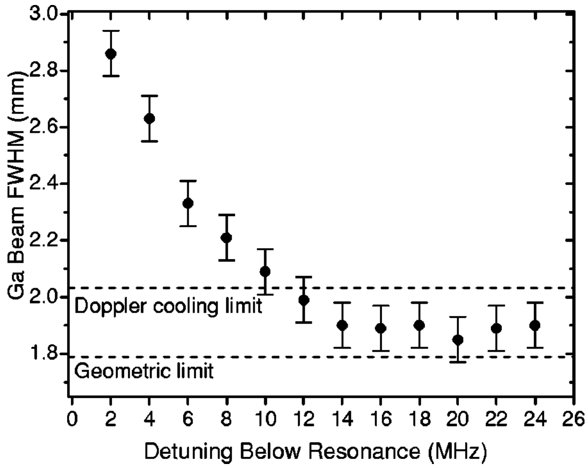

FIG. 3. Collimation of the Ga beam as a function of the cooling laser detuning below resonance.

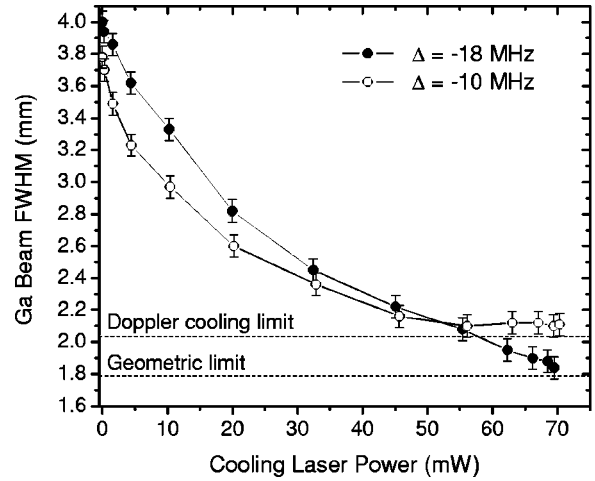

FIG. 4. Collimation of the Ga atomic beam for two different detunings as a function of the total power in the cooling laser.

The lin $\perp$ lin cooling efficiency as a function of cooling laser detuning is shown in Fig. 3. Here the cooling laser power was 68 mW. The detuning of the laser was accomplished by manually changing the drive frequency of the double-passed AOM in the saturated absorption apparatus. Since the full width of the Ga transition in the uncooled atomic beam was typically 35 MHz, the setting of the zero detuning had an uncertainty of $\sim$3 MHz. The cooling laser and the probe laser were parallel to better than 0.5 mrad. Thus the uncertainty of the zero detuning point due to misalignment of the lasers was at most 1.3 MHz. In discussing the detuning of the cooling laser, a negative or red detuning denotes tuning the laser frequency below resonance. A positive detuning (laser frequency above resonance) is denoted as blue detuning. In our experiment, a red detuning always resulted in a reduction of the atom beam size, corresponding to cooling, whereas a blue detuning always caused an increase in the size of the Ga beam, corresponding to heating of the atoms.

The collimation of the Ga beam improved with laser detuning from 0 to −14 MHz. Sub-Doppler cooling was achieved for detunings beyond −12 MHz ($\sim\Gamma/2$). From −14 to −24 MHz ($\sim\Gamma$), the collimation was relatively independent of the detuning and the cooled atom beam had an average FWHM size of 1.89 ± 0.02 mm. Subtracting off the geometric limit, this gave a size increase of 0.10 ± 0.04 mm over a distance of 34 cm. The divergence of the cooled atomic beam was found to be 0.3 ± 0.1 mrad (full angle). The corresponding nominal transverse velocity was 11 ± 4 cm/s, about half of the Doppler cooling limit.

Larger red detunings were also used, but the quality of the CCD images started to deteriorate since the probe illuminating laser was also further from resonance. In general it was observed that the cooling did not improve. The relative independence of the cooling with detuning could be attributed to the influence of the neighboring $F=3\rightarrow3$ transition, which was only 12.5Γ to the red side of the $3\rightarrow4$ cooling transition. [See Fig. 1(c).] As the red detuning increased, the excitation of the $3\rightarrow3$ transition also increased. Investigation of the optical pumping will be discussed later.

The cooling as a function of interaction length was investigated by translating a razor blade through the cooling laser beam. The cooling efficiency as a function of the total power in the truncated laser beam was shown for two different detunings in Fig. 4. For the smaller detuning (−10 MHz) only $\sim$50 mW was needed before the collimation of the beam ceased at a beam FWHM of 2.05 mm. For a larger detuning (−18 MHz) it required at least 70 mW to cool the atoms to the minimum (1.79 mm). It was further observed that the on-axis flux of the atomic beam did not improve with cooling. In the case of Fig. 4, as the degree of collimation improved with increasing power, the total fluorescence steadily decreased, indicating a loss of atoms from the $4p\,^2P_{3/2}(F=3)$ cooling state. For the smaller detuning, 68%

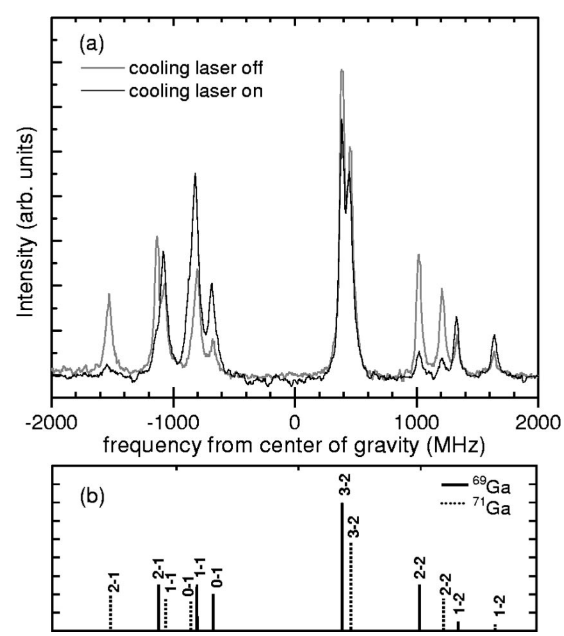

FIG. 5. (a) The laser-induced fluorescence spectrum obtained by 417 nm excitation with the cooling laser on (dark line) and off (light line). (b) The calculated line spectrum for both isotopes of Ga.

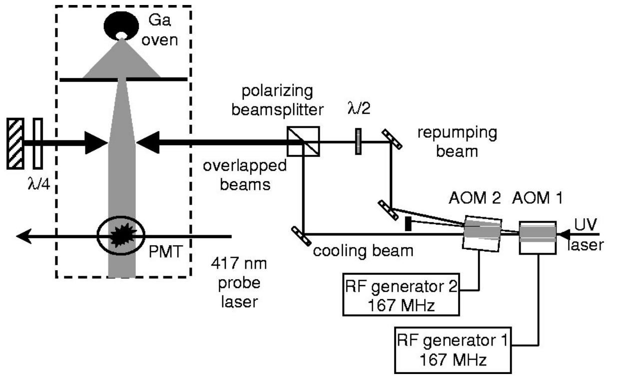

FIG. 6. Laser arrangement for cooling and repumping of a Ga beam.

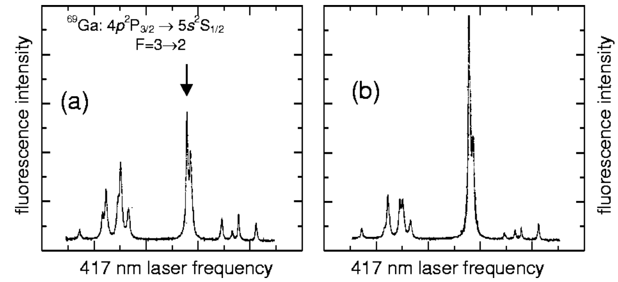

FIG. 7. 417 nm laser-induced fluorescence spectra taken with (a) only a cooling laser on and (b) both a cooling and repump laser on. The repump laser detuned +334 MHz from the cooling laser and power =4.5 mW.

of atoms survived and for the larger detuning, 57% of the atoms survived. This pointed to optical pumping by the cooling laser. Off-resonant excitation of the $F=3\rightarrow 3$ transition and subsequent decay to the $F=2$ lower state resulted in the loss of the cooled atoms. It should be noted that the $F=2\rightarrow 1$ transition was only $\sim 2\Gamma$ from the cooling transition and was also pumped. Thus the net effect of the optical pumping was to transfer atoms from the $F=3$ cooled state to the $F=1,0$ states.

To study this pumping, the CCD camera was replaced by a photomultiplier tube and the uv probe laser was replaced by a 417 nm diode laser tuned to the $4p\,^2P_{3/2}\rightarrow 5s\,^2S_{1/2}$ transition. This was a better transition for studying the effects of optical pumping because there were fewer hyperfine lines and they overlapped less. Figure 5(a) is the laser-induced fluorescence spectrum obtained by 417 nm excitation of the Ga atoms with the cooling laser upstream turned on and off. The power in the 417 nm beam was 0.6 mW and the power in the uv cooling beam was 45 mW. Figure 5(b) is a calculated spectrum for both isotopes to assist in identifying the transitions. The pumping of atoms from the $4p\,^2P_{3/2}(F=3)$ state to the $F=0,1$ states was observed by the decrease in intensity of the $3\rightarrow 2$ transition and the accompanying increase in the other transitions.

Two repumping schemes were investigated. In the first case, a repumping laser tuned to the $4p\,^2P_{3/2}(F=2)\rightarrow 4d\,^2D_{5/2}(F=3)$ of ^{69}Ga was used. A portion of the cooling laser (20 mW) was picked off and frequency shifted +334 MHz by two AOMs in tandem to be in resonance with the $F=2\rightarrow 3$ transition. The repump beam was overlapped with the cooling laser beam in a polarizing beam splitter. Laser-induced fluorescence spectra were taken downstream with the 417 nm laser operating at a power of 0.4 mW. This arrangement is shown schematically in Fig. 6. The effect of repumping is shown in Fig. 7. The 417 nm spectrum in Fig. 7(a) was taken with only the cooling laser on and no repump laser. The spectrum in Fig. 7(b) was obtained when both the cooling and repump lasers were on. It is seen that the repump laser was able to transfer almost all of the $F=2$ atoms to the $F=3$ state. The dependence of the repumping efficiency on power in the repump laser beam was also studied. About 5 mW of repump power was sufficient to empty the $F=2$ state and transfer the atoms to the $F=3$ state. Even at very low powers (<1 mW) the repump laser was capable of putting a significant number of atoms into the $F=3$ state.

The optical pumping of atoms from the $F=3$ state by the cooling laser is sensitive to the total amount of cooling laser power. This effect is seen by the difference in the spectrum in Fig. 5(a), taken with 45 mW, and in Fig. 7(a) which was taken with 20 mW. Due to a lack of cooling laser power while performing the optical pumping studies, no systematic study of repumping efficiency as a function of power in the cooling laser was performed. However, in the event of a powerful cooling beam (>60 mW), depopulation of the $F=2$ state by the cooling laser to the $F=1,0$ states (as mentioned above) might be significant. We therefore investigated an alternate scheme that involved pumping the atoms from the $F=0$ and $F=1$ states. The uv repump beam was shifted by +400 MHz from the cooling transition to allow pumping on both the $F=0\rightarrow 1$ and $F=1\rightarrow 2$ transitions (separated by only 20 MHz) in ^{69}Ga. This scheme was also able to increase the population of the $F=3$ state and was $\sim 0.75\times$ as effective as pumping directly on the $F=2\rightarrow F=3$ transition.

In summary, using the cycling $4p\,^2P_{3/2}(F=3)\rightarrow 4d\,^2D_{5/2}(F=4)$ transition at 294.45 nm, a Ga atomic beam was laser collimated to 0.3 mrad (full angle) using the lin $\perp$ lin polarization gradient configuration. The transverse velocity of the atoms was reduced to 11 cm/s, about half of the Doppler cooling limit. The one-dimensional kinetic energy of the atoms was reduced to ~ 6 neV. This level of collimation should be sufficient for a laser standing wave focusing on Ga atoms and provides the first step towards a unique method for modulating the atomic density in the growth plane.

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