Atomic gallium laser spectroscopy with violet/blue diode lasers

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(Dated: March 31, 2022)

We describe the operation of two GaN-based diode lasers for the laser spectroscopy of gallium at 403 nm and 417 nm. Their use in an external cavity configuration enabled the investigation of absorption spectroscopy in a gallium hollow cathode. We have analyzed the Doppler broadened profiles accounting for hyperfine and isotope structure and extracting both the temperature and densities of the neutral atomic sample produced in the glow discharge. We have also built a setup to produce a thermal atomic beam of gallium. Using the GaN-based diode lasers we have studied the laser induced fluorescence and hyperfine resolved spectra of gallium.

PACS numbers: 42.55.Px; 42.60.-v; 32.30.-r; 03.75.Be

I. INTRODUCTION

Atomic physics has seen a tremendous development in recent years [1]. New research fields include the creation and study of cold atoms [2], Bose-Einstein condensation [3], ion traps and quantum information processing [4]. A crucial role in this progress has been played by radiation sources such as diode lasers based on GaAs technology. In a Littrow configuration an external cavity diode laser (ECDL) represents a stable (both in intensity and frequency), inexpensive and handy source of radiation perfectly suited for atomic physics experiments [5,6].

Recently GaN-based laser diodes [7] emitting in the blue or near ultraviolet region have been commercially available (Nichia Corporation, Japan). Although their main impact is related with telecommunications and optical data storage applications, these new radiation sources are also of great interest in atomic physics. The laser spectroscopy of many atoms and molecules absorbing in this wavelength region can be accessed without the use of complicated frequency doubling systems. Another important perspective is their applications for laser cooling and atom nano-fabrication (ANF) experiments [8]. For this reason we will report some details on the vacuum system and atomic source that we have developed.

II. THE GALLIUM ATOM

The gallium element (Z=31) has two main isotopes $^{69}$Ga (60.1%) and $^{71}$Ga (39.9%), both with nuclear spin I=3/2. It has a melting point of 29.78 °C and a boiling point of 2403 °C. Its electronic configuration is [Ar]3d$^{10}$4s$^2$4p$^1$. This leads to a ground state $^4$P$_{1/2}$. The gross structure is shown in Fig. 1(a) where we have outlined the transitions in the violet and blue region at $\lambda_1 = 403.299$ nm and $\lambda_2 = 417.204$ nm [10].

The lifetime of the first excited state is $\tau = 7.0(0.4)$ ns [11] and the rates for the two decay channels are $\Gamma_1 = 4.9 \times 10^7$ s$^{-1}$ and $\Gamma_2 = 9.2 \times 10^7$ s$^{-1}$ [12,13]. The P-states hyperfine (HF) splittings of both isotopes are precisely known from atomic beam magnetic resonance measurements [13,14] and are shown in Fig. 1(b) and (c). The isotope shifts are $\Delta_{1S,1} = 32.8(3.5)$ MHz and $\Delta_{1S,2} = 39.6(3.5)$ MHz. These values have been measured together with the S-state splittings by Neijzen and Dönszelmann [15] using a Dye laser excitation of the two resonance lines. In Fig. 1(d) and (e) we have reproduced the structure of the gallium resonance lines. The ratio between the HF lines is determined by the isotope composition and by the relative hyperfine linestrength $S_{F \rightarrow F'}$:

$$S_{F \rightarrow F'} = (2F + 1)(2F' + 1) \left\{ \begin{array}{ccc}
F' & F & I \\
J' & F & J \\
I & J & 1
\end{array} \right\}^2 .$$

Note that the present definition excludes all the atomic constant associated to the definition of the absolute
linestrength \[2\]. Another important parameter is the saturation intensity. Extending its definition \[2\] for an open transition by considering the spontaneous emission rate for each decay channel \(\Gamma_i\) \((i = 1, 2)\), we have:

\[
I_{\text{sat},i} = \frac{\pi \hbar c}{3 \lambda^3 \Gamma_i}.
\]

Thus for the two transitions the saturation intensities are \(I_{\text{sat},1} = 15.7 \text{ mW/cm}^2\) and \(I_{\text{sat},2} = 26.4 \text{ mW/cm}^2\).

III. LASER SPECTROSCOPY IN A Gallium GALVATRON

Our study of atomic gallium lines was performed using two diode lasers at 403 nm and 417 nm (Nichia NDHV310APB) with 30 mW nominal power, housed in a DL100 system purchased from TOPTICA Photonics (Munich, Germany). The highly stable mounting and the diode control units for the diode temperature and current enable to have a mode-hops free range larger than 10 GHz (i.e. over the all frequency range of the piezo scan). The experimental setup used in our first investigation is shown in Fig. 2. The source of neutral gallium atoms is an hollow cathode discharge lamp (galvatron) made by Hamamatsu (series L2783-31Ar-Ga). The see-through cathode has a cylindrical shape with a hole of 3 mm diameter and 15 mm length. The maximum current applicable to the discharge before breaking is limited to 6 mA. Gallium atoms are sputtered from the cathode surface by energetic argon ions produced in the neutral buffer gas (argon). The equilibrium between ion production and recombination process leads to a gas of neutral atomic gallium forming in the cathode region. The argon gas pressure is fixed by the company to a value of 3 torr to enable a low current operation of the galvatron.

The laser absorption was monitored independently for the two transitions sending a probe beam of about 0.3 \(I_{\text{sat},1}\) and 0.2 \(I_{\text{sat},2}\) through the plasma generated in the galvatron. The signal was detected using photodiodes (Thorlabs DET110). A frequency calibration of the scan was obtained using a confocal Fabry-Perot interferometer with a free spectral range of 300 MHz (Coherent Spectrum Analyzer 33-6552).

We performed a systematic study of the absorption signals of gallium transitions as a function of the galvatron current for both transitions. We measured the absolute absorption of the probe by looking at its transmission signal straight from a photodiode. The maximum absorption at the line center we could detect was always below 3%. Hence the sample is optically thin. To enhance the signal and look for saturation effects we aligned a pump beam of about 20 \(I_{\text{sat},1}\) and 11 \(I_{\text{sat},2}\) (from the same laser source) with the probe in a counter-propagating beam configuration. The pump was chopped at high frequency (1 Khz) and the probe signal was detected by means of a lock-in amplifier. Figures 3 (a) and (b) show Doppler broadened absorption spectra obtained in this configuration at 403 nm and 417 nm respectively.

In a standard alkali vapor cell with no collisional broadening, using a counter-propagating beam configuration yields to the detection of the Doppler-free homogeneous lineshapes. This is a standard technique based on the saturation of the atomic transition by the pump used for example to actively stabilize infrared ECDLs in alkalis experiment (see for example \[2\]). The situation is more complex in a galvatron because of velocity changing collisions (VCC) occurring in the glow that can decrease or prevent the saturation of a specific velocity class (in particular the zero velocity class responsible for the appearance of the Doppler-free peaks) \[21\]. Velocity changing collisions distribute atoms among the different velocity classes and thus produce a broad pedestal in the saturated absorption spectrum. When the mass ratio between the perturber and active atom (argon and gallium in our case) is not too large, a single collision completely thermalizes the atomic velocity distribution. This is called strong collision regime \[21\]. The pedestal shape in this case is a Gaussian with a width changing with the buffer gas pressure. In the case of strong collisions and high intensity pump the width equals the Doppler width \(\Delta\nu_D\) and the VCC signal becomes independent of pump intensity. For high buffer gas pressure (as in our case) the strong VCC actually destroys the Doppler-free signal.

In our spectra no sub-Doppler feature was visible and the absorption lines were well described by a superposition of gaussian profiles. Indeed the buffer gas pressure of 6 torr in our galvatron causes the disappearance of the sub-Doppler features. This cannot be avoided because a low current operation is needed due to the low melting point temperature of gallium.

In Fig. 3 (c) and (d) we have plotted the absorption signals while changing the galvatron current showing the growth of the atomic density in the glow. Absorption profiles were obtained both with and without pump beam (i.e. without lock-in amplification). We have verified that the two situations, hence the FWHM and lineshapes, are completely equivalent. In Fig. 3 we showed the ones obtained using the lock-in amplification to enhance the signals and have a better resolution to determine FWHM, amplitude and centers of the lines even at lower currents.

a. Analysis: An accurate analysis has been made by taking into account the HF components and the different isotope abundances. We fit the observed curves as a sum of several gaussians (one for each HF transition), imposing the same width for all curves \(\Delta\nu_D\). This is the Doppler width resulting from the temperature of the gas. Moreover we fix the distance between the centers and the ratio between the amplitudes following the predicted HF structure of Fig. 1 (d) and (e). Figure 4 (a) shows the dependence on the galvatron current for the \(\Delta\nu_D\) derived from this analysis. The uncertainty on the data points from the fit is of the order of 0.2%. The width
of the lines (hence the temperature) appears to be constant within the range of current explored. The width $\Delta \nu_D$ obtained from the multiple HF fit is consistently lower than the widths obtained from single gaussian profiles for both 403 nm and 417 nm transitions as expected. From the Doppler width we can deduce the temperature of the neutral gas by the relation (22):

$$T = \frac{M}{2R \log 2} \left( \frac{c \Delta \nu_D}{2\nu_0} \right)^2$$  \hspace{1cm} (3)

where $M$ is the molar weight, $R$ the gas constant, $c$ the speed of light and $\nu_0$ the frequency of the transition. Considering all the $\Delta \nu_D$ values calculated for each transition, the temperature estimated was $T = 390$ K with an uncertainty of 4%. This takes into account the fluctuations on the width obtained from different spectra.

The absorption of light passing through an atomic sample of length $L$ and absorption coefficient $\alpha(\omega, z)$, is expressed by the Lambert-Beer’s law (22):

$$I_T(\omega) = I_0 \exp \left[ -\int_L \alpha(\omega, z) \, dz \right].$$ \hspace{1cm} (4)

In our case this can be simplified by assuming a uniform density of neutral atoms in the plasma and optically thin samples. Hence introducing the absorption cross-section $\sigma_{ik}(\omega)$ for the fine structure transition $i \rightarrow k$ and the lower state density of absorbing atoms $n_i$ we can express the absorbance as:

$$\frac{\Delta I(\omega)}{I_0} \approx n_i L \sigma_{ik}(\omega).$$ \hspace{1cm} (5)

In our case to have a correct estimate of the atomic density of length $L$ and absorption coefficient $\alpha(\omega, z)$, is expressed by the Lambert-Beer’s law (22):

$$\frac{\Delta I(\omega)}{I_0} \approx n_i L \sigma_{ik}(\omega).$$ \hspace{1cm} (5)

where $R$ is the gas constant, $M$ the molar weight and $g$ the degeneracy of the fine structure levels. For our measurements the temperature in the glow was constant and we can use the value deduced from the Doppler width to obtain the absorption cross-section for the two transitions $\sigma_1 = 0.457 \cdot 10^{-12}$ $m^2$ and $\sigma_2 = 0.475 \cdot 10^{-12}$ $m^2$ with a 2% uncertainty coming from the uncertainty on the temperature. Figure 4 (b) shows the atomic density in the glow evaluated from each spectrum using Eq. 5 (with $L = 15$ mm being the cathode length). The population in the $P_{3/2}$ state (extracted from the 417 nm spectra) is about two times larger than the population in the $P_{1/2}$ state (extracted from the 403 nm spectra). In fact our measurements are made in a galvatron where the neutral atomic sample is created by sputtering and recombination of gallium ions from the cathode. The population in the two P ground levels depends upon the recombination process that creates highly excited atoms decaying in the ground states through spontaneous emission. Thus this is mainly determined by the branching ratio for the S to P transitions and much less by the Boltzmann factor or by optical pumping.

b. Optical pumping: Another important aspect in the gallium lambda system is the optical pumping within the two P ground states. We have verified the role played by it in the galvatron absorption spectra by looking at the absorption of the weak probe (0.3 $I_{sat,1}$) tuned on the 403 nm lines in presence of a strong (20 $I_{sat,1}$) pump beam at 403 nm (from the same laser) or a repump beam tuned at 417 nm (specifically on the $P_{3/2}$ $\rightarrow$ $F' = 2$ transition and 11 $I_{sat,2}$). When the pump was superposed to the probe we could observe a 33% decrease of the absorption signal. On the other hand when the repump was superposed we could observe an increase of the probe absorption up to 55% of the original signal. Finally when both pump and repump were aligned there was an increase of the probe absorption of about 35%. All these values and line shapes did not change by having the beams co-propagating or counter-propagating confirming the crucial role of velocity-changing collisions in smearing out the saturation effects.

In principle an alternative option for spectroscopy investigation is to monitor the discharge current i.e. to detect an optogalvanic signal (23). Sub-Doppler resolution in the gallium lines was shown in (23) by the use of optical-optical double resonance spectroscopy with optogalvanic detection. In this work two frequencies at 417.2 nm and 641.4 nm acted on the $6p^2P_{1/2}$ $\rightarrow$ $5s^2S_{1/2}$ level cascade. Saturation of the blue line was achieved in a hollow cathode cooled with liquid nitrogen to reduce the discharge noise and pressure. We have verified that with our setup (no liquid nitrogen cooling) we were able to detect an optogalvanic signal. However this reproduced the Doppler broadened profiles for both transitions with no sub-Doppler features visible.
IV. LASER INDUCED FLUORESCENCE IN A THERMAL GALLIUM BEAM

c. Atomic beam: The vacuum system has been carefully planned according to the following criteria: keep the system as simple and standard as possible within the current UHV technologies; have the possibility to extend and implement the setup for ANF into a molecular beam epitaxy (MBE) system; allow the optical access needed for the laser cooling and manipulation of gallium atoms. The vacuum chamber can be divided in three regions: i) production of the gallium atomic beam; ii) collimation (through mechanical and eventually optical means) and probing; iii) optical focusing and deposition on a substrate.

The atomic source is a gallium effusion cell (dual filament type DFC-63-60-300-WK by CreaTec-Fischer, Erligheim, DE). The PBN (Pyrolytic-Bore-Nitrate) crucible has been specially designed in order to have a horizontal atomic beam and to avoid damaging the oven by gallium reacting with the heating elements. The crucible is made of four parts: an outer standard cylindrical crucible, an inner boat shaped part where gallium is evaporated, a spacer and a 1 mm insert placed at the lip of the outer crucible (that is always kept at a temperature 120 °C higher than the inner part to avoid gallium condensation).

To keep a good ultra-high vacuum we use two ion pumps (40 l/s in the oven region and 55 l/s in the cooling and deposition region) that are switched on after a roughing and bake-out of the system is obtained. This gives a pressure of about \(5 \times 10^{-10}\) mbar in the deposition region (if required a better UHV could be achieved by the use of a non evaporable getter pump). The effusion cell is generally operated at a temperature of 1100 °C (the gallium vapor pressure at this temperature is \(6 \times 10^{-2}\) mbar). The atomic beam coming out of the cell is collimated by a 1 mm skimmer placed 5 cm after the effusive source. According to Knudsen law this source provides a flux of \(1 \times 10^{14}\) atoms/s with a most probable velocity of \(v_p = 690\) m/s along the longitudinal direction.

d. Atomic fluorescence: By sending a laser beam tuned on the gallium resonances at 403 nm or 417 nm orthogonal to the atomic beam direction, we detected the laser induced fluorescence (LIF) at right angles to both the atomic beam and the direction of laser light using either a CCD camera or a photodiode with a collection optics. The top parts of Fig. 4 show two images of our gallium atomic beam obtained using the 403 nm ECDL tuned on resonance with the \(P_{3/2}, F = 2 \rightarrow S_{1/2}, F' = 1\) hyperfine transition for the two different isotopes. From the analysis of these gaussian profiles and the geometry of the beams we extracted information on the width of the atomic beam and obtained a divergence of \(15 \pm 2\) mrad. This result is consistent with an estimate of about 20 mrad obtained from the geometry of the collimating holes.

The LIF signal generated by the laser beam has been also collected on a photodiode and analyzed using a phase sensitive detection (PSD) for the two transitions independently. A reference modulation frequency (1 KHz) is sent to the ECDL current while the LIF signal is recorded through lock-in amplification. A scan was sent to the ECDL piezo in order to observe the hyperfine resolved spectra of gallium at 403 nm and 417 nm. Typical signals are shown in Fig. 4. The (a) and (b) plots have been obtained with laser intensity of about 20 \(I_{sat, 1}\) and 11 \(I_{sat, 2}\) respectively. The 100 MHz broadening of the derivative signals is not only due to laser power broadening but also to the applied dither on the laser current needed to enhance the signal to noise ratio. In fact lowering the applied dither and the laser power we have split the closest doublets as shown in Fig. 4(c) and (d). These spectra were obtained with an intensity of 1.5 \(I_{sat, 1}\) and 1 \(I_{sat, 2}\) respectively. The width reduces to 50 MHz, that agrees well with what is expected from the beam divergence. We verified that at large amplitude of the dither the spectra were fitted by a gaussian profile, while lowering the dither the dispersive signals become closer to multiple lorentzian derivative signals. In fact we expect the PSD spectra to be derivatives of Voigt profiles. A study of line shapes in frequency-modulation spectroscopy is beyond the scope of this paper. For a comprehensive investigation in atomic media see for example the work by Xia et al. [26].

V. CONCLUSIONS AND PERSPECTIVES

External cavity diode lasers at 403nm and 417nm have been tested and tuned on resonance with gallium transitions. Laser absorption spectroscopy in a galvatron has proved to be a useful tool for a quick tuning of the ECDLs. We have used these laser sources to study absorption spectroscopy in a gallium hollow cathode exploring the atom density production and Doppler width (hence temperature) as a function of discharge current. Although sub-Doppler features were not visible in a simple saturated absorption configuration, the gallium galvatron has proved to be a very useful tool to perform absorption spectroscopy of atomic gallium with the ECDLs in the blue. We have also built the vacuum system and produced a collimated thermal atomic beam of gallium in a UHV environment. Using the ECDLs we have performed LIF detection with a phase sensitive technique enhancing the hyperfine resolved spectra of gallium.

Our interest in GaN-based diode lasers lies mainly in their use for laser cooling and atom nano-fabrication. There is no closed transition from the true ground state suitable for laser cooling and two alternatives are possible: a two-colour laser cooling using diode lasers at 403 nm and 417 nm on the P states [10] or laser cooling at 294 nm (by frequency doubling a Dye laser), where a closed transition exists between the \(P_{3/2}\) and \(D_{5/2}\) states (this scheme is in use at Colorado State
Our aim is to use the blue and violet diode chips to investigate two-colour laser cooling on the P-S transitions. The peculiarities of the gallium lower energy levels open up many interesting problems regarding the best cooling scheme to be used for the atomic beam collimation. From our first divergence measurements we can estimate the initial width of the transverse velocity distribution to be close to the velocity capture range \( v_c = \lambda \Gamma_i / 2\pi \) (i=1,2) \[2\], 9.2 m/s for gallium. If we consider a Doppler cooling on one of the transition at 403 nm or 417 nm, \( v_c \) for gallium is \( \sqrt{\Gamma_i} = 0.4 \) mrad well suited for ANF. An interesting perspective is the use of two-colour laser cooling in a similar fashion as in the work by Rooijakkers et al. \[28\] on helium atoms. In the case of helium we have a cascade three-level system. Optimum cooling parameters were found, showing that the cooling force is an order of magnitude stronger than for a two-level system. In the gallium case we have a lambda system involving the P-S transitions. We plan to perform a study of the two-colour force for gallium and compare it with the \textit{standard} two level situation.

It is a pleasure to thank A.Camposeo, E.Cerboneschi, C.J.Foot, F.Fuso, S.A.Lee, M.Lindholdt, D.Meschede, M.Oberthaler, O.Prudnikov, U.Rasbach, A.Sasso, C.Vasi for useful discussions. We are also in debt to E.Andreoni, D.Arigo, G.Lupò, N.Puccini, G.Spinella for technical support.

This work is funded by CNR (Consiglio Nazionale delle Ricerche) and by the NANOCOLD project of the IST Program of the EC through the Nanotechnology Information Devices Initiative.

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FIG. 1: In (a) simplified Grotrian diagram of the investigated Ga atom transitions. (b) and (c) depict details of the hyperfine splittings (in MHz) for the two stable isotopes. (d) and (e) show the relative line strength $S_{F \rightarrow F'}$ of the hyperfine components, indicated by the height of the columns and the number in brackets after the $F - F'$ quantum numbers.
FIG. 2: Experimental setup for absorption spectroscopy in a gallium galvatron. The light from the laser diode (ECDL) is split using quarter waveplates and beam splitter cubes (BSC), then aligned in a crossed-beam configuration and then detected using photodiodes (PD). The probe signal is enhanced by lock-in amplification. The frequency calibration is obtained using a confocal Fabry-Perot interferometer with a free spectral range of 300 MHz.
FIG. 3: Doppler broadened absorption spectra at 403 nm and 417 nm obtained in crossed-beam configuration for a current of 4.7 mA in the galvatron (see text). The strong velocity changing collisions (VCC) in the cathode region prevent the formation of sub-Doppler features. In (a) and (b) the absorption signal (black lines) is well described by a superposition (gray solid line) of gaussian profiles (gray dashed lines). In (c) and (d) we have studied the gallium atomic production in function of the galvatron current for both transitions. A range of galvatron currents between 1.6 mA and 4.7 mA was explored in steps of ~0.4 mA.
FIG. 4: (a) Study of the optical lines FWHM versus the galvatron current. From the Doppler broadened spectra we can extract the FWHM performing a multiple fit that considers HF and isotope components. (b) Particle density extracted from the Lambert-Beer's law for an optically thin medium. By accounting for HF and isotope composition of the lines the atomic density in each P state and isotope was estimated versus the galvatron current.

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FIG. 5: On the top part, CCD images of the gallium atomic beam. On the bottom part vertical profiles. The atomic beam direction is perpendicular to the page of the images (as shown in the top left image), the laser beam is sent horizontally and orthogonal to the atomic beam. The laser induced fluorescence (LIF) signal at 403 nm has been detected orthogonally to both directions. The laser was tuned on resonance with the \( P_{1/2}, F = 2 \to S_{1/2}, F' = 1 \) hyperfine transition of the two isotopes.
FIG. 6: Hyperfine resolved spectra of gallium at 403 nm (a) and 417 nm (b) by phase sensitive detection. The LIF signal is collected on a photodiode while the laser frequency is scanned. Close-ups of the $F = 2 \rightarrow F' = 2$ and $F = 1 \rightarrow F' = 1$ doublets (c) at 403 nm and $F'' = 3 \rightarrow F' = 2$ (d) at 417 nm with lower power and dither to avoid line broadening. At lower intensity the signals are fitted better with a lorentzian than with a gaussian model.