Optically pumped semiconductor lasers for atomic and molecular physics

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ABSTRACT

Experiments in atomic, molecular and optical (AMO) physics rely on lasers at many different wavelengths and with varying requirements on spectral linewidth, power and intensity stability. Optically pumped semiconductor lasers (OPSLs), when combined with nonlinear frequency conversion, can potentially replace many of the laser systems currently in use. We are developing a source for laser cooling and spectroscopy of Mg⁺ ions at 280 nm, based on a frequency quadrupled OPSL with the gain chip fabricated at the ORC at Tampere Univ. of Technology, Finland. This OPSL system could serve as a prototype for many other sources used in atomic and molecular physics.

Keywords: semiconductor laser, atomic and molecular physics, tunability, intensity stability

1. INTRODUCTION

The advent of lasers revolutionized atomic molecular and optical (AMO) physics. Modern experiments in precision spectroscopy, laser cooling, optical traps, quantum degenerate gases and quantum information processing (QIP) all rely heavily on lasers. At the same time, laser technology has made enormous progress and is now covering ranges in wavelength and power larger than ever, while efficiency, reliability and control of lasers have steadily improved.

Electrically pumped diode lasers have played a major role in AMO experiments because they are relatively cheap and efficient and can be tuned and controlled to accommodate most of the typical requirements. Unfortunately, not all wavelengths of interest in AMO can be accommodated by electrically pumped diode lasers. Additionally, their relatively small structures often lead to non-ideal mode shapes and limit scalability in power, especially if narrow linewidth, single longitudinal mode operation is desired.

Most of these concerns can be overcome by OPSLs¹ which efficiently convert the light from an inexpensive multi-mode pump diode laser to widely tunable, narrow linewidth light with excellent mode quality and good power scalability. Because of these desirable features, OPSLs are already commercially available at many different colors and in a wide range of powers (several mW to several W). Unfortunately the available lasers do not feature the controls to make them easily tunable, for example, to be on resonance with an atomic transition or to lock them to an external reference, such as a cavity or a molecular transition, to reduce instantaneous linewidth and/or long-term frequency drift. If such features were available, OPSLs combined with nonlinear frequency conversion could provide efficient, compact and relatively inexpensive high-quality light sources for many AMO experiments.

In section 2 we discuss the general requirements for useful sources in AMO physics and lay out a general approach based on OPSL sources and nonlinear frequency conversion. We present a tabular overview of selected transitions in neutral and singly ionized atoms that are widely used in AMO research and could potentially benefit from suitable OPSL sources. In section 3, we briefly review the laser requirements for quantum information processing with trapped ions and introduce specific tasks with Mg⁺ ions within this general scheme that could potentially be addressed by one type of OPSL source. We discuss our effort to develop this particular OPSL, based on gain chips fabricated at the ORC at Tampere Univ. of Technology in Finland. Finally we present our conclusions and outlook in section 4.

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2. AMO LASER SOURCES

In this section we discuss typical tasks for lasers in AMO physics and then introduce a general approach to address these tasks based on OPSLs and nonlinear frequency conversion. We then present a table of specific AMO tasks that outlines the technical specifications necessary to address them with OPSL sources.

2.1 Typical tasks in AMO physics

Many modern AMO physics experiments require laser cooling and fluorescence detection on dipole allowed transitions.²⁻⁵ Laser beams are often used to drive transitions between a ground state in an atom or ion (or in some cases a molecule) and an excited electronic state that decays within a few ns to a few μ s, back to the initial ground state or a set of energetically lower states. These decay times correspond to transition linewidths of 10s of MHz to 100s of kHz. Moreover, atomic transitions typically involve groups of transitions to several sub-levels arising from fine structure (THz to 100s of GHz) and hyperfine structure (10s of GHz to MHz). Some transition frequencies can be tuned with magnetic and electric fields up to 10s of GHz. An ideal laser source should therefore be single longitudinal mode, tunable (preferentially without mode-hops) over the range given by the fine-structure and stable to a fraction of the atomic linewidth. In molecular physics, if one would like to address rotational and vibrational sub-levels, even wider tuning ranges are sometimes required. Transition wavelengths range from the near-infrared to the vacuum UV and less than a mW of power focused into waist sizes of 100 μ m or less (FWHM intensity) is usually sufficient to saturate an allowed dipole transition. In the past, one had to rely on coincidence to find a suitable laser source and often only dye lasers, which are widely tunable but work-intensive and temperamental, were suitable. The situation improved as more types of diode and fiber lasers, combined with advances in nonlinear frequency conversion, have become available. Nevertheless, more versatile sources at many different wavelengths are highly desirable.

In the simplest cases of laser cooling and detection, the dipole transition is a so-called "cycling transition" where the atom always decays to the same ground state from which it was excited. However, in many atoms that are widely studied, the decay from the excited state branches to other levels that may need to be repumped to the initial ground state. This requires one or a few repump lasers mostly with similar technical specifications as the laser cooling/fluorescence detection lasers. Importantly, if the decay branches into a different electronic level, the repump lasers must typically be at very different wavelengths compared to the laser cooling/fluorescence laser. Molecules, with their more complicated level structure typically require even more repump sources, a complication that currently makes laser cooling or fluorescence detection impractical in most species. Many frequently used neutral atoms feature transitions in the infrared to visible range, while the dipole transitions from the ground states of atomic ions occur mainly in the ultraviolet region. Repumping often requires additional lasers with a wide variety of wavelengths.

Ion traps are often loaded from an atomic beam or vapor that was traditionally ionized by electron impact. More recently, laser ionization that resonantly excites the electron through one or more levels into the continuum has been employed.⁶ Laser ionization can be very efficient compared to electron impact ionization and reduces the presence of electrons that can charge surfaces surrounding the trapped ions, but this requires one or more laser sources for each ion species to be produced. Resonant transitions in thermal beams can be Doppler-broadened to a few GHz and require several mW of power into typical beam-waists to saturate the transition and achieve efficient photoionization. In some cases ionization is effectively a two-photon process, with the second photon promoting the electron to the continuum coming from the same laser source. In those cases ionization efficiency rises quadratically with the available power until it saturates. The rise depends on the exact transition rates and may increase even if the first transition is already saturated.

Raman transitions between long lived states are a popular choice for laser manipulations of atoms or ions when long coherence times are desired. This is especially true for experiments on quantum information processing and quantum simulation.⁷ Raman transitions can be used to connect two levels that are (meta-) stable against decay, for example two hyperfine states in the ground-state manifold. The frequency difference of such states is typically in the radio or microwave frequency range, and one can connect them by adjusting the frequency difference of two different laser beams to be resonant with their splitting. The transition rate is increased if the light sources are tuned to be near allowed transitions, but one needs to detune sufficiently to effectively suppress spontaneous decay from nearby excited levels. Large Raman-transition rates and little decoherence can

be achieved by using powerful sources (10s of mW to W) at a detuning of 10s of GHz to a few THz from the nearest excited levels.

Modern experiments, in particular those on degenerate quantum gases such as Bose-Einstein condensates, often employ light induced dipole forces and dipole traps that can either depend on the state an atom or ion is in or can be detuned enough that the dipole force is nearly independent of the atomic state.⁸ State-dependent dipole forces are typically produced with similar detuning and power levels as Raman-transitions, while state independent dipole traps can be much farther detuned. Powerful CO_2 lasers, producing 10 W or more at 10 μ m wavelength, were a popular choice for early far detuned dipole traps; lately, high-power lasers at 1064 nm or 532 nm or sources with fiber amplifiers are often used because of their relatively high power and low intensity noise. Long wavelength lasers typically create "high-field-seeking" traps where the atoms are attracted to the intensity maxima, while "low-field-seeking" dipole traps can be created if the trapping laser is detuned to the blue of the closest transition of the trapped species. In the latter case, the atoms are attracted to intensity minima, therefore the confining light field mode needs to have a minimum surrounded by higher intensity regions, as for the transverse directions of a doughnut mode. Higher order Laguerre-Gaussian modes have been produced as the direct output of OPSLs by using a 2D photonic crystal reflector as a cavity mirror.⁹

The tasks described above require reliable lasers that can fall anywhere inside a large range of wavelengths and operate at very different power levels, preferably with low intensity noise. Up to now, many different types of lasers have been used to perform these tasks, but OPSL- based systems should be a useful substitute for most of them and frequently offer economic, technical and performance benefits.

2.2 General approach with OPSLs

The gain chips of optically pumped semiconductor lasers can be designed for laser systems over a broad range of wavelengths.¹ For example, indium gallium arsenide (InGaAs)-based OPSLs have produced output from 700 to 1200 nm. The range can be further extended by frequency doubling to most of the visible spectrum (350 nm to 600 nm), and tripling and quadrupling can enable continuous coverage from 175 nm in the ultraviolet¹⁰ to 600 nm in the visible. This wide range enables the laser source to be designed to fit the application, while it has previously sometimes been the other way around. An extension of the fundamental range down to 600 nm seems possible and with frequency conversion this would lead to continuous coverage from the far ultraviolet to the near-infrared. OPSLs often allow for several 10s of nanometers tuning range, often wider than other laser types. In many cases, addition of a birefringent filter and an intra-cavity etalon is sufficient to enforce single longitudinal mode operation in OPSLs and their linewidth and amplitude noise can be made very low. Further adding piezoelectric tuning of a cavity mirror enables continuous tuning of the output wavelength. Intra-cavity elements for frequency selection need enough degrees of freedom (for example angle or temperature tuning) to tune the laser on to resonance with atomic transitions and to provide feedback when locking to reference cavities. In addition, OPSLs have good power scalability and peak output levels of more than 15 W have been demonstrated in single longitudinal mode operation.¹¹

Table 1 provides examples of selected tasks in AMO physics, but it should be stressed that this table is not comprehensive. Many more transitions used in AMO experiments, for example narrow optical clock transitions requiring exquisite laser stabilization, are not listed in this table. The table is sorted by atomic mass, and states the type of task as well as the laser wavelength, transition linewidth and possible OPSL sources. The necessary power levels close to the transition frequency vary by task; for most resonant dipole transitions used in laser cooling and fluorescence detection, less than 1 mW is sufficient. High efficiency photoionization requires on the order of 10 mW, Raman interactions and dipole forces require several 10s of mW up to several W. The approximate short-term stability (stability over times less than 1 s) required from the OPSL source should be well below the transition linewidth and long-term drifts need to be controlled to stay resonant with the transition of choice. For resonantly enhanced nonlinear frequency conversion, the frequency stability of the input light has to be sufficient to not convert frequency fluctuations into amplitude fluctuations due to the bandwidth of the enhancement resonator, which often has relatively high finesse to improve conversion efficiencies.

Species	Task	Wavelength/nm	Trans. linewidth	Source/nm
He*	LC, FD, DP	1083	1.6 MHz	1083
Li	LC, FD, RA, DP	671	5.9 MHz	671
Be	LC, FD, PI	235	85 MHz	Q 940
Be ⁺	LC, FD, RA, DP	313	19.4 MHz	T 939, D 626
Ne*	LC, FD	640	8.2 MHz	640
Na	LC, FD, RA, DP	589	8.2 MHz	D 1178
Mg	LC, FD, RA, DP, PI	285	73 MHz	T 855, Q 1140
Mg ⁺	LC, FD, RA, DP	280	41.3 MHz	T 840, Q 1120
К	LC, FD, RA, DP	766	6.2 MHz	766
Ca	LC, FD, RA, DP, PI	422	35 MHz	D 844
Ca ⁺	LC, FD, RA, DP	393,397	23 MHz	Т 1179, Т 1191
Ca ⁺	RP	850, 854, 866	23 MHz	850, 854, 866
Cr	LC, FD	426	5 MHz	D 852
Cr	RP	649, 658	7, 20 Hz	649, 658
Kr*	LC, FD	812	4.6 MHz	812
Rb	LC, FD, RA, DP	780, 795	6 MHz	780, 795
Sr	LC, FD, RA, DP, PI	461	32 MHz	D 922
Sr ⁺	LC, FD, RA, DP	407, 421	21.5 MHz	D 814, 842
Sr ⁺	RP	1033, 1092	21.5 MHz	1033, 1092
Ag	LC, FD, RA, DP	328	20.7 MHz	Т 984
Cd	LC, FD, PI	229	90 MHz	Q 916
Cd ⁺	LC, FD, RA, DP	214, 227	50.5 MHz	Q 856, 908
Xe	LC, FD, RA, DP	882	6 MHz	882
Cs	LC, FD, RA, DP	852	5 MHz	852
Ba	LC, FD, RA, DP, PI	554	17.5 MHz	D 1108
Ba ⁺	LC, FD, RA, DP	455, 493	20.1 MHz	D 910, 986
Ba ⁺	RP	650	20.1 MHz	650
Yb	LC, FD, RA, DP, PI	399, 556	29, 0.18 MHz	Т 1197
Yb ⁺	LC, FD, RA, DP	328, 369	19.7 MHz	T 984, 1107
Yb ⁺	RP	935	19.7 MHz	935
Fr	LC, FD, RA, DP	718	8 MHz	718

Table 1. Selected tasks in AMO physics: Noble gas species annotated with * are in metastable levels. Tasks are abbreviated as follows: LC means laser cooling, FD fluorescence detection, RP repumping, RA Raman-transitions, DP dipole forces/traps, and PI photoionization. In the "Source" column, D means doubled, T tripled and Q quadrupled.

3. OPSL FOR QUANTUM INFORMATION PROCESSING EXPERIMENTS WITH MAGNESIUM IONS

In this section we will briefly describe general requirements for laser sources used in QIP with trapped ions. We will discuss a specific example, the laser-driven transitions that are relevant when using magnesium ions in QIP implementations. Finally, we present some preliminary results achieved with an OPSL source that we are currently developing in our laboratory, in collaboration with ORC.

3.1 Laser requirements for quantum information processing with trapped ions

QIP¹² with trapped ions was started with the seminal 1995 paper by Cirac and Zoller.¹³ They showed how tools and concepts developed in ion trap research can be used to implement a universal set of quantum gates, meaning that any algorithm viable on a quantum computer can be decomposed into a sequence of gates from this set. This proposal instantly made trapped ions a candidate system for the scalable implementation of quantum information processing. Several overview articles on the fundamentals and subsequent development of the field are available, see for example Ref.s 14–16. For the purposes of this article it is only necessary to know that trapped ions are often produced from neutral atomic beam sources by exciting the neutral atom via one or several resonant dipole transitions followed by a final excitation step to the continuum to ionize the atom. It is envisioned that thousands to millions of qubits will be needed in a useful large scale QIP system, therefore large numbers of ions need to be loaded efficiently. While today thermal atomic beams are typically used for loading, large-scale systems will possibly require pre-cooling and collection of neutral atoms before they are ionized,¹⁷ further increasing the number of lasers required.

The ions need to be cooled and optically pumped into a well defined initial quantum state. This again requires lasers to resonantly drive dipole transitions. The quantum logic operations performed on the ions can be divided into single-qubit manipulations and two-qubit gates. The highest quality single qubit manipulations have been done with microwaves,^{18, 19} but the highest quality two-qubit gates relied on far detuned Raman-laser interactions.²⁰ Achieving higher quality two-bit gates at even larger detuning and thus larger power, along with the need for massively parallel operations will put a premium on robust and scalable low-amplitude-noise, high-power laser sources.

3.2 Transitions in neutral Magnesium and Magnesium ions

Our QIP work requires laser sources for exciting several transitions in neutral and singly ionized magnesium. A transition from the $2p^{6}3s^{2}$ ¹S ground state to the excited $3s^{3}p^{1}P^{0}$ state around 285 nm in neutral Mg is the first step to photo-ionize atoms from a thermal beam to load ion traps,⁶ see Figure 1 a). The second photon into the continuum is either from the same laser or from the laser that also cools the Mg⁺ ions (below). Photoionization requires 1-10 mW into a waist of 20-60 μ m (FWHM intensity). The photoionization wavelength is too long to be amplified with available fiber amplifiers, so doubled dye lasers at 570 nm or a quadrupled Raman fiber amplifier at 1140 nm, seeded with an external grating stabilized diode laser, have been used. Once magnesium is ionized and trapped, cooling and fluorescence detection require driving transitions from the ${}^{2}S_{1/2}$ ground state to the first excited ${}^{2}P_{1/2}$ and ${}^{2}P_{3/2}$ states in Mg⁺ ions near 280 nm, see Figure 1 b). The resonant transitions can be saturated with power significantly below 1 mW into waist sizes of order 10-100 μ m (FWHM intensity). However, additional cooling beams that are detuned by several hundred MHz and at power levels between 100 μ W and 1 mW are often used to improve cooling of highly motionally excited ions, for example right after ionization or after a collision with background gas in the trap. Raman transitions are driven by a pair of laser beams that are detuned by 100 GHz-1 THz from the resonant transitions and require much higher power, up to 100s of mW into similar waist sizes as above, see Figure 1 c). These transitions have traditionally been addressed by frequency doubling dye lasers operating around 560 nm and more recently with either diode or fiber-laser seeded fiber amplifiers around 1120 nm that are subsequently frequency quadrupled. The fiber amplifiers operate on the long-wavelength edge of their amplification range and this has limited output powers to below 2 W and lead to problems with fiber darkening and amplified spontaneous emission.

3.3 Realization of an OPSL source

A suitable OPSL for QIP with magnesium ions²¹ should meet several critical requirements. The wavelength for photoionization at 285 nm and for manipulation of magnesium ions around 280 nm are conveniently close by coincidence so they should be addressable with one type of OPSL. This requires single mode operation of the laser around 1119 nm and 1140 nm and a tuning range that spans these wavelengths. We only require continuous tuning over approximately 1 GHz around the resonances of neutral and ionized magnesium; mode-hops while tuning in between resonances are not a problem. The task demanding the highest power of approximately 100 mW is the



Figure 1. Relevant transitions in neutral and ionized magnesium: a) Photoionization of neutral magnesium. The second excitation step requires a photon with wavelength < 375 nm, this can either be another photon from the source resonantly driving the first step at 285.2 nm or from the Doppler cooling source around 280 nm. b) Resonant transitions for Doppler cooling, optical pumping and repumping of Mg⁺. The ground state hyperfine splitting of 1.79 GHz can be bridged with electro-optical or acousto-optical means. Transitions to different excited P levels, split by the 2.75 Thz fine-structure, require separate laser sources. c) Two-photon stimulated Raman-transitions between long lived hyperfine ground states that are used for qubits. The detuning Δ from the excited ${}^{2}P_{3/2}$ state can be positive or negative. To drive high quality two-bit gates, Δ needs to be on the order of the fine structure splitting and power levels on the order 100 mW to 1 W are required to drive high quality 2-qubit gates.

excitation of Raman-transitions with a detuning of the order 1 THz from the ion resonances around 280 nm. The power required at 1120 nm depends on the efficiency of two subsequent steps of second harmonic generation (SHG). We estimate that a fundamental power of 1-2 W at 1120 nm should be sufficient. Since our resonant SHG cavities from 560 nm to 280 nm have relatively narrow linewidth, we target a linewidth of significantly less than 1 MHz in 1 second of averaging at 1120 nm. Finally, we would like to achieve intensity stability below 1% at 280 nm. Even with no additional intensity noise in the SHG processes, amplitude fluctuations are roughly doubled in every SHG step. Therefore the relative intensity noise at the fundamental should be well below 0.25%. Based on these requirements, we have designed a standing-wave laser cavity that incorporates a water-cooled top-emitting gain chip with a diamond heat spreader developed and grown at the ORC²¹ and a critical type I phase-matched LBO crystal for intra-cavity doubling to 560 nm. The basic layout of the cavity is shown in Figure 2. The folded linear cavity contains a birefringent filter that can be tuned by changing its temperature as well as by slight changes of its alignment around Brewster's angle. Single longitudinal mode operation is achieved with an etalon that can be temperature and/or angle tuned. The 20-mm-long LBO crystal is AR coated at 1118 nm and 559 nm. We pump the gain chip with up to 11 W from a multi-emitter diode laser bar at 808 nm. Without the LBO crystal in the cavity, we achieve single-mode operation and up to 2 W at the fundamental wavelength. After inserting and tuning the LBO for peak power we achieve 1.2 W at 560 nm. The output is roughly evenly distributed in the two output directions both with high overlap to a TEM00 mode. To reduce drift and improve the frequency stability, we lock the laser to an external reference-cavity using the Hänsch-Couillaud technique;²² a mirror mounted onto a piezo-electric element is used to feed back onto the laser frequency. We are currently working to improve the frequency and amplitude stability of the system. Once the laser is sufficiently stable, it should be straightforward to resonantly double the output a second time to 280 nm and then test the system in practice by photoionizing neutral magnesium and exciting resonant and Raman-transitions in Mg⁺.

4. CONCLUSIONS AND OUTLOOK

OPSLs could become useful sources in AMO physics. For this to happen, stable, single longitudinal mode OPSLs that are tunable by birefringent filters, etalons and piezo-mirrors are required. If nonlinear wavelength conversion, either intra-cavity or external, is achieved, OPSLs can cover a very wide range of wavelengths including most of those frequently used in AMO physics (see Table 1). The high efficiency, relative simplicity,



Figure 2. Layout of OPSL cavity including a birefringent filter (BRF), etalon, and LBO crystal for doubling the infrared fundamental output of the laser to the yellow-green. The visible output is distributed into two beams that are coupled out in two different directions via mirrors that are highly reflective between 1116-1120 nm and highly transmitting between 558-571 nm. The piezo element on the folding mirror allows for continuous tuning of the laser and is used to provide feedback when locking the laser to an external reference cavity.

scalability, and robustness of OPSLs, combined with low intensity noise and straightforward techniques for frequency stabilization, would make OPSLs competitive with or superior to many laser systems used in AMO physics today. Nevertheless, suitable OPSL systems are, to the best of our knowledge, rarely found in AMO research laboratories and not commercially available so far.

As a proof-of-principle, and to fill a pressing need in our community (QIP with trapped ions), we have started to develop an OPSL system for photoionization of neutral magnesium and experiments with magnesium ions in collaboration with the ORC, which is providing suitable gain chips to us. Our first results are encouraging. Although we are not a laser development group, our contribution is to test OPSL technology under conditions common to many AMO experiments. We hope that this encourages commercial development, and greater access to this promising technology.

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