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Tunable lasers for atomic vapor laser isotope separation: the Australian contribution

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High-performance narrow-linewidth tunable lasers are an essential tool for atomic vapor laser isotope separation also known as AVLIS. In this article a description is given of a successful research effort, carried out at Macquarie University in the 1980-1984 period, to develop efficient high-power narrow-linewidth tunable lasers in the visible. Part of this effort was supported by the then Australian Atomic Energy Commission (AAEC). The published literature is cited and some peripheral episodes are revealed for the first time.

Introduction

In a previous article Pryor (*1*) described the contribution of Australian physicists to advanced uranium isotope enrichment. His article centers on two approaches: a phase transition transformation of UF_6 known as the Ward process and molecular laser dissociation using CO_2 lasers. The first approach was a theoretical idea that was not demonstrated in practice but the molecular laser dissociation approach yielded many publications in international journals (*1*). In the same account Pryor writes "The AAEC considered an AVLIS program. Professor Jim Piper, from his university, would have been very happy to develop the copper-vapour pump and the tuned dye lasers. But the cautious hand refrained." Here, the development of efficient narrow-linewidth tunable laser oscillators for AVLIS applications, at Macquarie University, is described from a historical perspective. Also the influence of this oscillator physics, and architecture, program on various laser research efforts around the world are outlined as well as a stealth attempt to have an AVLIS effort adopted by the Australian Federal Government circa 1982.

Early laser resonators

In 1978, following my Honours year at the former *School of Mathematics and Physics* of Macquarie University, I embarked on a doctoral research project to develop an optically-pumped molecular laser under the supervision of J. A. Piper. The pump laser of choice was the copper-vapour-laser (CVL) which was one of the research areas that Piper had specialized in while working with C. E. Webb at The Clarendon Laboratory in Oxford. However, prior to performing these experiments we realized that we needed high-performance tunable lasers to replicate the emission lines of the copper laser at 510.554 nm and 578.213 nm and study the spectroscopy of the molecular gain medium. Thus, I built a nitrogen-laser-pumped tunable dye laser based on the telescopic design of Hänsch (*2*). Due to its relatively long cavity length and its two-dimensional intracavity beam expansion, illuminating the diffraction grating, this laser proved rather sensitive to thermal variations. The need for more compact, stable, and rugged alternatives became painfully clear. As possible alternatives, Jim Piper brought a paper by Hanna *et al.* (*3*), on a single-prism beam expander, and a couple of papers on grazing-incidence designs (*4, 5*). It didn't take me long to implement these designs and to enjoy the benefits of compactness. However, the price to pay was either very low conversion efficiencies or high levels of amplified spontaneous emission (ASE) at the output. High ASE levels originated from coupling the output emission from either the reflection losses at the prism or the reflection losses at the grazing-incidence grating. Since low conversion efficiencies were unacceptable, and ASE is the equivalent of broadband optical noise, something had to be done. The drive towards efficient low-ASE narrow-linewidth tunable laser emission led to the independent development of closed-cavity multiple-prism grating dye lasers (*6*) and shortly afterwards to the introduction of the prism pre-expanded near-grazing-incidence tunable lasers (*7*).

Copper-vapour-laser-pumped narrow-linewidth tunable dye lasers

Soon the main focus of my research became the development of efficient high-power narrow-linewidth tunable dye lasers. In 1982, while partially funded by the AAEC, I began a series of experiments on closed-cavity CVL-pumped multiple-prism grating tunable laser oscillators. At first we employed a low repetition rate TE CVL as the excitation laser (*8*). My knowledge on the TE CVL grew, at the practical level, from my observation of the work of Milan Brandt who was completing his doctoral research on the subject (*9*). These low pulsed-repetition-frequency (prf) experiments were followed by a series of experiments using a high prf (~ 8 kHz) CVL laser as the pump (*10, 11*). Two of the closed-cavity oscillators developed in these experiments are depicted in Figures 1 and 2. Basic emission parameters include tuning in the $565 \le \lambda \le 603$ nm range, laser linewidths of $\Delta\nu \approx 650$ MHz (or $\Delta\lambda \approx 0.0007$ nm at $\lambda \approx 575$ nm) for a 4-5% conversion efficiency. The peak power of this narrow-linewidth emission was ~ 1 kW.

Generalized multiple-prism dispersion theory

The systematic development of multiple-prism grating laser oscillators required the theoretical characterization of the laser linewidth which in turn depends on the overall intracavity dispersion of the multiple-prism grating configuration. However, at the time there was no comprehensive theory and no multiple-prism dispersion equations. In regard to the multiple-prism dispersion Jim Piper suggested that I should "look it up in a book." So I did, and the more that I looked the more I realized that it wasn't out there. At the time I was doing a postdoc with Brian Orr, at the University of New South Wales, and

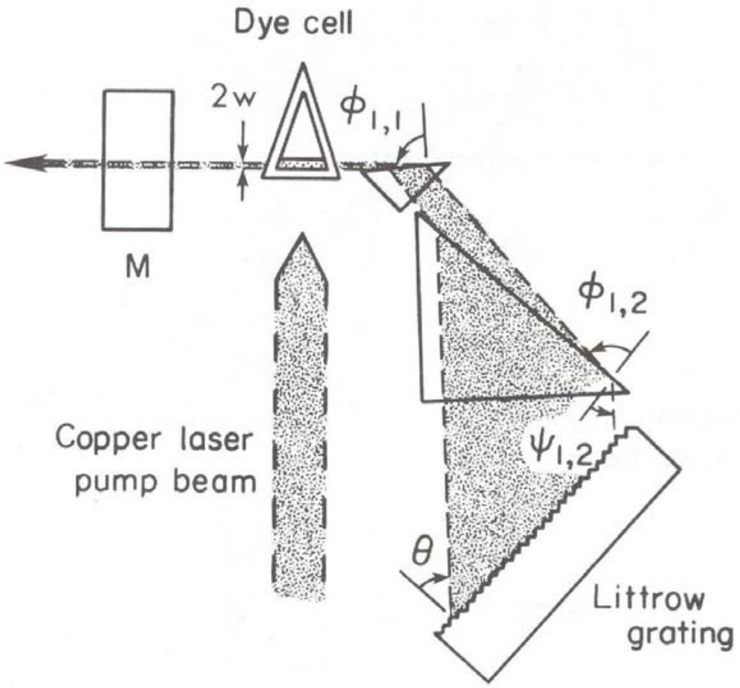

Fig.1. CVL-pumped narrow-linewidth multiple-prism grating tunable laser oscillator. In this closed-cavity dispersive oscillator configuration the beam waist at the gain medium (w) is reduced to achieve single-transverse-mode emission. The resulting beam dimension ($2w$) is then expanded to completely illuminate the diffraction grating. For this particular oscillator the beam magnification factors is $M \approx 100$ (from Duarte and Piper (*10*)).

for a while I would spend the evenings working on this problem. Eventually, I worked out the generalized single- and double-pass multiple-prism-grating dispersion equations thus allowing us to characterize the double-pass linewidth equation (*12*). A following paper extended the treatment to a multiple return-pass analysis (*11*) and a subsequent paper included generalized higher derivatives applicable to pulse compression in femtosecond lasers (*13*) which recently has been extended to a mathematical framework, derived using a Newtonian iterative approach, that provides higher derivatives, in analytical form, at will (*14*). As a footnote I should add that Newton had pictorially, and qualitatively, discussed the first order dispersion of multiple-prism arrays in his book *Opticks* (*15*). Our equations can be nicely used to quantify the dispersion in Newton's prismatic configurations. Besides the use of these equations in laser cavity design (*16*), and pulse compression calculations (*17*), researchers have also applied them in the design of femtosecond laser microscopy systems (*18*, *19*).

Advocacy for an AVLIS effort in Australia

Due to my involvement in the Macquarie science reform movement (*20*) I had developed several contacts among noted Australian politicians and also within the Australian Federal Government at the time. In particular, I was acquainted with the federal minister for education (1975–1979) Senator John L. Carrick whom subsequently became the minister for national development and energy (1979–1983). By then I had studied several reports on AVLIS for uranium and I was very keen to continue the work on the physics and architecture of narrow-linewidth tunable laser oscillators. Thus, on my own initiative, I directly proposed to Sir John the introduction of an AVLIS facility in Australia. Correspondence was interchanged and in one of his letters Sir John Carrick wrote "The Uranium Enrichment Group of Australia (UEGA) assessed a number of competing technologies… and concluded that centrifuge technology would be best suited for an Australian plant… I am assured that UEGA has taken into account the recent advances in laser isotope separation… UEGA was not, however, prepared to commit to a technology still at the R&D stage" (*21*). From his letter it was clear that Carrick took advice from the UEGA group and that a decision had already been taken *not to invest* in AVLIS or alternative laser isotope separation schemes. Albeit he politely suggested a meeting with some senior public servants it was obvious that the decision was final. As Pryor insightfully said in his writings… "*the cautious hand refrained*."

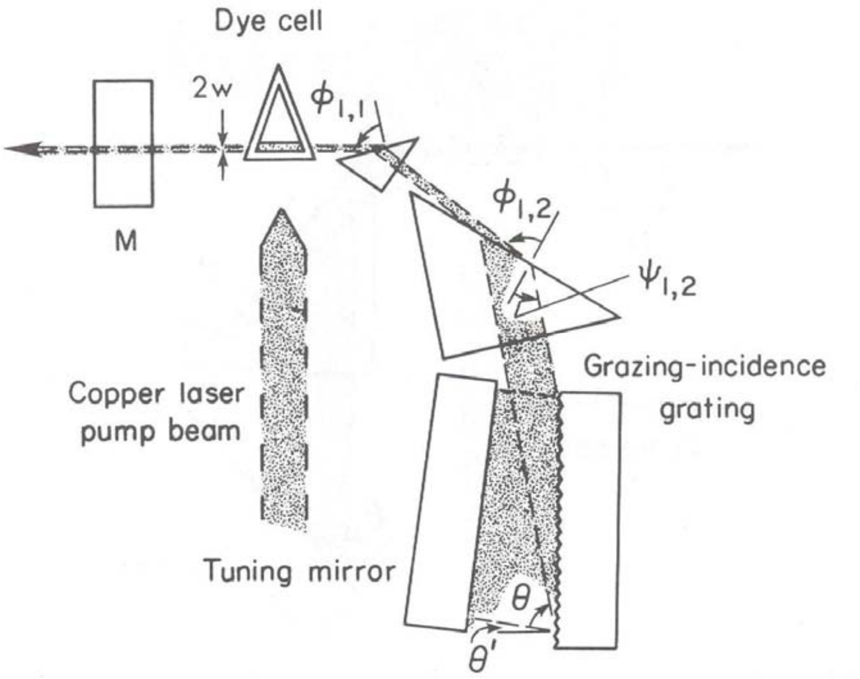

Fig. 2. CVL-pumped narrow-linewidth multiple-prism pre-expanded near grazing-incidence grating tunable laser oscillator. In this closed-cavity dispersive oscillator configuration the diffraction grating is deployed at a higher angle of incidence so that the required intracavity beam expansion is reduced to $M \approx 25$. Lasing at a $\Delta\nu \approx 650$ MHz linewidth corresponds to single-longitudinal-mode emission (from Duarte and Piper (*10*)).

AVLIS efforts in the open literature

The popular literature often associates AVLIS with the American effort at the Lawrence Livermore Laboratory which is best summarized by Bass *et al.* (*22*). The truth is, however, that AVLIS is a far more widespread research and development approach to isotope enrichment. As our papers were published I began to receive numerous postcards with reprint requests from the USA, all over Europe, Israel, and Asia. Among these requests were postcards from the now well-known Bhabha Atomic Research Centre (BARC), in India. Years after sending their reprint requests the Bhabha laser group published several papers describing the use of multiple-prism pre-expanded near-grazing-incidence tunable lasers (see Figure 2) for the efficient generation of low-noise narrow-linewidth tunable laser radiation in the $564 \le \lambda \le 602$ nm portion of the spectrum (*23*). Japanese laser researchers working in the nuclear field also adopted multiple-prism grating oscillator designs in their CVL-pumped tunable laser systems (*24*).

In 1998, when the underground nuclear tests were conducted in India, questions were asked to attempt to elucidate if the researchers at BARC had the capability to achieve laser isotope separation. The answer was in the open literature.

American laser sequel

Around 1982-1983 academic positions in lasers, or optics, were scarce in Australia. Although I was offered a very nice position with the Australian Department of Defense, I decided to accept an assistant professorship at the University of Alabama with funding to do research in infrared lasers. There I demonstrated the use of multiple-prism grating cavities, using ZnSe prisms, to achieve tunable narrow-linewidth emission in high-power TEA CO_2 lasers (*25*). Then, one late morning and out of the blue, I got a phone call from R. W. Conrad, a top physicist with the US Army Missile Command (MICOM), asking me what I knew about narrow-linewidth high-energy dye lasers. That was a long phone conversation that led to an even longer collaboration (1985-2002) on the development of narrow-linewidth high-energy tunable lasers. The pride and joy of this effort was a highly-stable single-longitudinal-mode long-pulse oscillator engineered in an all-invar structure (*26*). This elegantly ruggedized oscillator was successfully tested on a moving vehicle over a rough terrain but, due to the end of Cold War funding, was not integrated to its matching kW-class amplifier stage.

The work I did on multiple-prism grating CO_2 lasers, plus the work of other researchers, proved that these oscillator configurations were universally applicable to tunable lasers. In 1992 Paul Zorabedian, working at Hewlett Packard, successfully applied our multiple-prism grating configurations to semiconductor lasers (*27*).

The experiments at MICOM also led to research into solid-state dye lasers. Paradoxically, using highly purified dye-doped polymers developed in the former Soviet Union I was able to demonstrate, for the first time, tunable narrow-linewidth emission from solid-state dye laser oscillators (*28*). Subsequently, this work led to the development of very compact optimized multiple-prism grating oscillators, tunable in the $550 \le \lambda \le 603\,\mathrm{nm}$ range, yielding high-power single-longitudinal-mode laser emission at a linewidth $\Delta\nu \approx 350$ MHz, for pulses $\Delta t \approx 3\,\mathrm{ns}$ (FWHM) (*29*). The emission of these laser oscillators is at the limit allowed by Heisenberg's uncertainty principle.

AVLIS status

Despite opposition from participating scientists, the Lawrence Livermore uranium AVLIS effort was brought to an end shortly after it was transferred to a commercial entity in 1999. Although the approach was a scientific and technological success apparently this success was not enough to ensure timely commercial viability. At present it is undetermined how many laboratories around the world are engaged in this type of research. The interest is not just on uranium but also on various other atomic species including lithium (see, for example, *30*). A Russian perspective on AVLIS is given by Bokhan *et al.* (*31*).

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