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ATOMIC-VAPOR-LASER ISOTOPE SEPARATION

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Abstract

This paper gives a brief history of the scientific considerations leading to the development of laser isotope separation (LIS) processes. The close relationship of LIS to the broader field of laser-induced chemical processes is evaluated in terms of physical criteria to achieve an efficient production process. Atomic-vapor LIS processes under development at Livermore are reviewed.

Historical Perspective

Since early in this century when isotopes were first discovered, there has been a series of important discoveries that has progressively improved our understanding of atomic (and nuclear) structure and in some cases led to the large-scale production of isotopes.

Not surprisingly, the primary methods for separating isotopes have been based on the bulk transport properties of a medium containing a mixture of isotopes, i.e., differential transport of isotopic components in a multicomponent fluid (gas, liquid, or plasma).

Several of the more important methods of this type are:

- Gaseous diffusion.
- Centrifugation (gas, liquid, or plasma).
- Mass spectrometry.
- Chemical exchange.
- Distillation.

All of these have been used to produce significant quantities of isotopes, but gaseous diffusion (for uranium) and distillation (for deuterium) have operated at the largest scale.

Although spectroscopic differences between isotopes of individual atoms or molecules were known to exist, these differences are quite small. It was not until 1931 when the first spectroscopic isotope separation of deuterium by Urey et al., brought international attention to the importance of spectroscopic differences between isotopes. Photochemistry was being developed somewhat in parallel with the work on isotope separation and during World War II photochemical methods of isotope separation were explored by the Manhattan Project under the direction of Urey. This effort was not totally successful and the bulk transport processes were implemented on a large scale. The following is a list of selected highlights from the first 50 years of isotope separation:

- 1906-1907 Discovery of Radioactive Isotopes (Uranium/Thorium)
  - B.B. Boltwood (1906); H.N. McCoy and W.H. Ross (1907).
- 1913 The Term "Isotopes" Is Coined by F. Soddy
  - "isos" equal, "topos" place.
- 1913-1919 Discovery of Natural Isotopes (Neon)
  - J. J. Thomson (1913) and F.W. Ashton (1919)
- 1931 Discovery of Deuterium Using Spectroscopic Differences
  - H. Urey wins the Nobel Prize.
- 1932-1935 Attempted Photochemical Separation of Hg Isotopes
  - S. Mrozowski (1932, theory) and K. Zuber (1935, experiment).
- 1943-1946 Large-Scale Uranium Enrichment Processes Developed by US Manhattan Project
  - A cast of hundreds; many processes investigated; gaseous diffusion emerges the winner.
- 1953-1956 Successful Photochemical Separation of Hg Isotopes
  - B. Billings, W. Hitchcock, and M. Zelikoff.

After World War II, people continued some research on photochemical processes but made very little progress toward practical production of isotopes. The isotopic differences in electronic, vibrational, and vibronic spectra are energetically very small compared to the energies involved in chemical reactions. Thus the isotopic selectivity is very difficult to maintain when chemical reactions are occurring at the high rate (density) required for large-scale production. This difficulty leads one to consider photophysical processes (e.g., selective photoionization of atoms or photodissociation of molecules) where radiation is used to produce atomic and molecular fragments that can be collected by physical means.

Although using a photophysical vs a photochemical process might improve the prospects for isotope separation based on spectroscopic differences, a major barrier still remained prior to 1960. The primary sources of radiation available at that time were very broadband (or blackbody) radiators in the visible portion of the spectrum where important electronic transitions exist in atoms and molecules. These early light sources were highly incoherent (spatially and temporally) and operated at relatively low intensity.

With the first operation of a laser in 1960 by T. Maiman, a new era began. Throughout the 1960s progress was rapid in the development of lasers and it soon became apparent that high intensity radiation could be generated with an extremely narrow frequency bandwidth and specific lasers could be precisely tuned over a broad frequency range. Precision frequency control combined with high intensity was the key to the use of spectroscopic differences in the separation of isotopes on a production scale.

High average power was of interest to the military and precision frequency control was of interest to research scientists and communications engineers. The discovery of the dye laser in the late 1960s allowed one to achieve frequency control and tunability simultaneously with moderate power levels. There was an ambiance in the international scientific community regarding the potential for using lasers in a practical isotope-separation process. Throughout the 1960s there was a large number of ill-conceived processes proposed and it was not until around 1970 that cogent concepts began to emerge; perhaps the most notable was that of S. Janes and R. Levy, then at the Avco Everrett Research Laboratory, Wilmington, MA. They proposed to use tunable dye lasers to

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selectively photoionize ^{235}U in a stream of atomic uranium vapor. The ionized ^{235}U would be removed from the vapor using electromagnetic forces and collected on plates, while the ^{238}U would flow past the collector plates largely undisturbed by the laser light. Other photophysical processes emerged in the early 1970s, but the only other one to receive serious financial support was based on the concept of selective photodissociation of UF_6. In this process the isotopic selectivity derives from differences in the vibrational spectra. The molecule is vibrationally excited with infrared lasers and then photodissociated into UF_5 and F using ultraviolet lasers. The dissociated product condenses as a powder on a collector and the remaining gas flows past the product collector largely unaffected.

In the early 1970s the laser isotope separation (LIS) program at Livermore began. Though many different laser processes were explored in the early years, we soon focused on the atomic vapor (AVLIS) process.

The AVLIS program is both important to and illustrative of the general application of lasers in chemistry. Figure 1 gives some perspective on how long it takes to get a process into production and shows what we think might happen in the future. Scientific feasibility for uranium LIS was demonstrated in the early 1970s. Development of the process and investigation of the economic potential were continued throughout the 1970s. Intensive economic analyses have been performed in the last few years and we are now entering into a very intensive engineering effort leading to a production plant in the early 1990s.

We believe that in the next decade there will be a much more orderly scientific evaluation of industrial photochemistry. There was a certain amount of euphoria in the 1970s that is beginning to wear off, but we believe there will be more fruitful research in the 1980s and there will be enough major applications of lasers once they have undergone careful examination. We can expect in the 1990s to see industrial photochemistry emerge. AVLIS is expected to be the first use of lasers on an industrial scale and we believe it will help the general acceptance of laser technology by the chemical industry and the general acceptance across the board of laser technology in commercial processes.

General Consideration of Laser Process Economics

In evaluating laser processes, we have found a simple expression to be quite useful:

$$\left(\frac{\$}{\text{unit}}\right)_{\text{Product}} = \left(\frac{\$}{\text{unit}}\right)_{\text{Handling}} + \left[\left(\frac{\text{MJ}}{\text{unit}}\right)\cdot\left(\frac{\$}{\text{MJ}}\right)\right]_{\text{Laser}}$$

It relates unit cost of product to the (life-cycle) cost of laser energy, to the laser energy required per unit, and to the cost of handling the material throughout the process in a form compatible with laser processing. Although we have been primarily concerned with LIS processes, this formulation is equally useful in evaluating a wide range of laser applications such as laser sorting, photochemistry, laser annealing, cutting, or welding.

An analysis of the four terms in this expression can form the basis for judging the viability of a laser process in the market place.

- The unit cost of product must be small compared to that for alternative nonlaser processes. The cost for the laser-based process must be small compared to the forward (operating only) cost to displace existing production, but need only be smaller than the total (operating plus capital) costs for new production. In the case of uranium enrichment, the inefficiency of chemical, aerodynamic, centrifuge, and

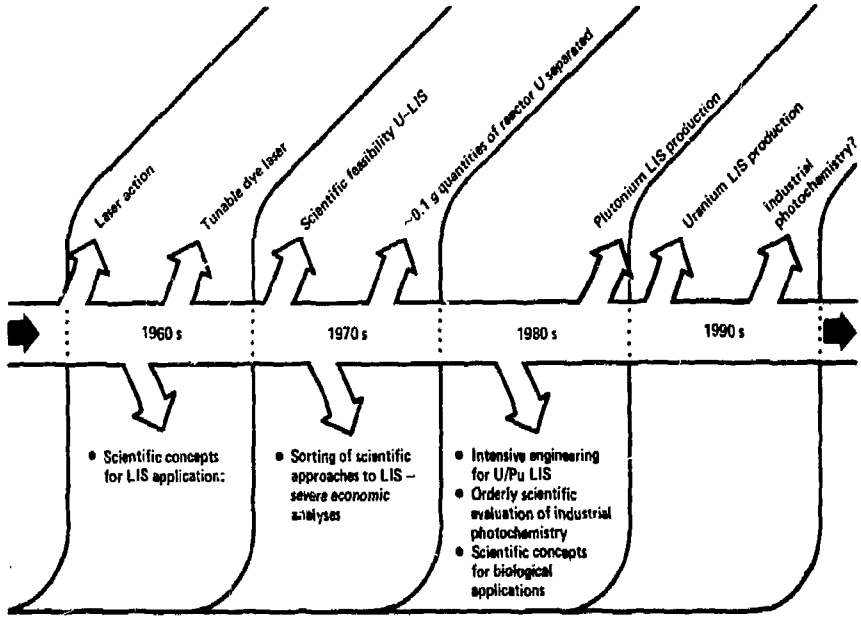

Fig. 1. History and anticipated progress in the science, technology, and application of industrial laser systems.

diffusion processes leads to many repetitive processing steps (staging). This leads to quite large capital costs. Also, in most cases the electric power and other operating costs are high so that, in general, LIS of uranium is competitive for both displacement of existing capacity and construction of new capacity.

- The cost of material handling appropriate to a laser process is strongly influenced by the staging required, i.e., the yield or efficiency of the lasers in a single illumination. In many applications, particularly those involving separation, sorting, and photochemistry, the desired selectivity of the laser necessitates a working medium that may be costly to produce. For example, to have sufficient selectivity in isotope separation the working medium in general must be a fairly dilute gas. The cost associated with producing the proper material conditions for the desired laser interaction, rather than the laser cost itself, can easily exceed the costs associated with the laser and hence dictate the economic viability. This is particularly true where the material must be processed in several stages to attain the desired product quality.
- The laser energy required per unit is a strong function of the selectivity of the irradiation step. Laser energy is quite expensive compared to thermal energy. Thus, if a laser process is to be economical, the energy must be efficiently used in producing the product. This will be the case for driving a chemical reaction only if the number of product particles generated by each photon is considerably in excess of unity. In the case of isotope separation, efficient energy use can be achieved via interaction only with a minor isotope. Any loss in selectivity, resulting in the absorption of laser energy in a manner that does not produce product, tends to drive up the laser-power requirement and hence makes the process less economically competitive. In separating uranium isotopes, for example, it is important to have sufficient selectivity to ensure that little laser energy is absorbed in the major ^{238}U isotope.
- The life-cycle cost of laser energy must be low. The cost of laser systems is often quoted and thought of in terms of capital cost per installed watt. For most high-power laser systems, however, operating costs associated with reliability and maintenance dominate the cost associated with return on capital and depreciation. Thus, laser-based processes require the development of highly reliable low-operating-cost and low-capital-cost systems. The magnitude of these costs depends strongly on the detailed laser-system requirements, such as frequency stability and bandwidth, timing and control, intensity or amplitude waveform, and beam quality.

Table 1. Nominal laser parameters for a practical AVLIS process.

Laser characteristic	AVLIS requirement	Reason for requirement
Pulse	20 to 200 ns	Laser pulse must be short in comparison with atomic-state lifetime
Prf	~10 kHz	Repetition rate must be adequate to illuminate all the flowing vapor
Pulse energy	0.1 to 1 J	Intensity must be sufficient to saturate atomic transitions
Frequency	Tunable	Frequency must be tunable to allow overlap with atomic transitions
Bandwidth	~1 to 3 GHz	Bandwidth should match inhomogeneously broadened atomic line
Frequency stability	~±30 MHz	Good spectral overlap of the laser and atomic line should be maintained
Phase front	~3 λ/D	Beams must propagate through long columns of vapor for efficient photon utilization
Uniformity	~30%	Vapor must be fairly uniformly illuminated

The ultimate issue concerning the success of these laser systems is the total life-cycle cost for producing the highly coherent energy required for the application. The cost of the laser system can be better understood by first considering the basic cost of electricity. At 36 mils/kWh, the cost of electrical power is 1¢/MJ. If a laser system is approximately 1% efficient (as is true for AVLIS copper-vapor lasers), the electrical costs alone are $1/MJ. For an AVLIS laser electro-optical system, we have to include also the losses due to frequency-conversion efficiency, beam combination, and beam transport. These considerations push the net laser-system cost of electrical power into the $1 to $10/MJ range. When we also include the costs of the initial capital investment and regular refurbishment, a reasonable estimate for the total life-cycle cost is in the $10 to $100/MJ range.

Laser Isotope Separation of Uranium

The specific application of lasers to the separation of uranium isotopes is illustrative of the requirements placed on a laser-system design by both the process physics and the operating constraints of the production plant. The performance parameters required for efficient use of the laser radiation by the uranium vapor are shown in Table 1. Tunable dye lasers satisfy well the requirements for tunability, bandwidth control, and frequency stability. Similarly the requirements on pulse duration and pulse-repetition frequency (prf) suggest using a laser as the excitation source for the dye lasers. Copper-vapor lasers (CVLs) were selected as the pump source because of their high prf and relatively high operating efficiency. Apart from these laser specifications, there are a number of system requirements that must also be satisfied to achieve the overall economics required for a viable commercial process. High availability of the laser subsystems is required if good use of the installed capital equipment (both lasers and separators) is to be obtained. The goal of the system designer is to obtain high availability with a minimum of installed equipment, while at the same time using a system architecture that places reasonable requirements on component reliability.

The design approach for our basic CVL-pumped dye laser system that accommodates these availability and reliability requirements is shown in Fig. 2. The laser system consists of two major modules, the waveform generator and the power amplifier. The waveform-generator module provides the temporal, spatial, and spectral waveform required to drive the photoionization process. The waveform-generator output power is amplified by the power-amplifier module, which in turn provides sufficient power to illuminate the uranium vapor in the separator modules.

The waveform generator consists of several dye laser oscillators whose frequencies and spectral bandwidths are precisely controlled. Each is tuned to the exact frequency of the particular uranium transition required in the photoionization process. Electro-optic hardware serves to acquire, monitor, and control the frequency of each of these lasers. Since failure of a single waveform generator is sufficient to terminate plant production, a redundant waveform generator is provided to ensure that this subsystem has a high degree of availability.

Once the basic frequency has been generated by the dye laser oscillator, it is amplified in a power-amplifier chain. This master-oscillator/power-amplifier (MOPA) configuration was chosen for the dye lasers because it permits tailoring of the laser-beam quality, spectral content, etc., at low power levels where conversion efficiency is unimportant.

The power to excite these dye lasers is provided by CVLs. These lasers pump both the oscillators and preamplifiers in the waveform generator as well as the high-power dye amplifiers located in the power-amplifier modules. This laser-system configuration allows the final amplifier stages to operate with high extraction efficiency so that the overall conversion efficiency from CVL light to tunable radiation can attain a value that approaches the theoretical Stokes efficiency.

A MOPA chain configuration was also selected for the CVLs because it allows high extraction efficiency and accommodates the short excitation lifetime of the upper laser level. The CVL MOPA chains are supported on large concrete monolithic structures that also supply integrated services such as buffer gas, cooling water, electrical power, and diagnostics and controls. Each chain consists of a small-bore oscillator and four large-bore series amplifiers (small bore ≈ 3-cm diam, large bore ≈ 8-cm diam). Lasers with these diameters have already been demonstrated in our laboratories. Larger-diameter lasers are currently under consideration. In the MOPA chains, each amplifier contributes ~100 W to the chain output power. Each power-amplifier module includes nominally 30 MOPA chains. High availability from each set of CVL MOPA chains is provided by one or more redundant chains that are operated hot and ready to supply power. If a failure in any chain occurs, a series of mirrors can be moved to replace the failed beamline with light from a redundant chain. Repairs to the failed hardware (or to hardware receiving normal maintenance) can be made while the redundant chain is in operation.

Status of the CVL Components and Systems

Each of the laser modules discussed in the previous section is assembled from a relatively small number of building blocks that include:

- Dye master oscillator.
- Dye amplifiers.
- CVLs.
- Optical-transport systems.
- Support structures.
- Diagnostics and controls.

A dye MOPA chain is required in the waveform generator for each frequency in the photoionization process. Figure 3 shows a chain with two preamplifiers. In our laboratories we have repeatedly demonstrated master oscillators with performance parameters that are equal to or better than our goals for plant operation. We have accumulated over 1000 h of operation on individual oscillators, although we have not yet made an effort to extend continuous operation times beyond ~15 h.

The preamplifiers provide high-fidelity amplification of the input signal, maintain good spatial beam quality of the input beam, and produce sufficient output power to provide efficient optical extraction in the succeeding power amplifiers. Figure 4 shows a photograph (a) and diagram (b) of an operating dye amplifier. Power amplifiers are very similar in both size and shape to the preamplifier shown. The dye/solvent fluid flows rapidly through the pump illumination region such that fresh dye is illuminated with each pump pulse. The input/output piping for a power amplifier is much larger than for a preamplifier to accommodate the larger flow. Dye preamplifiers and power amplifiers are relatively small devices, varying from approximately 30 to 45 cm in height for the hardware shown in Fig. 4. The active illumination zone is also quite small since, at the repetition rates of interest, the power per unit volume that can be extracted from such a device is approximately 10,000 W/cm^3. To date, the highest output power we have obtained from this type of dye laser is 55 W, operating at 6 kHz with 45% conversion efficiency. We have not yet operated a dye laser at the full plant average-power goal, because we have not yet had adequate installed CVL power for excitation. However, we have operated well above our single-pulse energy goals (with good beam quality),

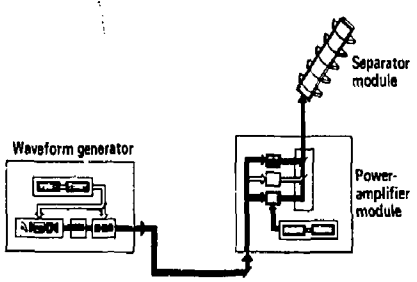

Fig. 2. Conceptual relationship between major AVLIS modules.

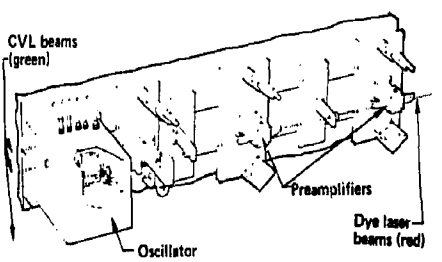

Fig. 3. Elements within a typical dye laser MOPA chain.

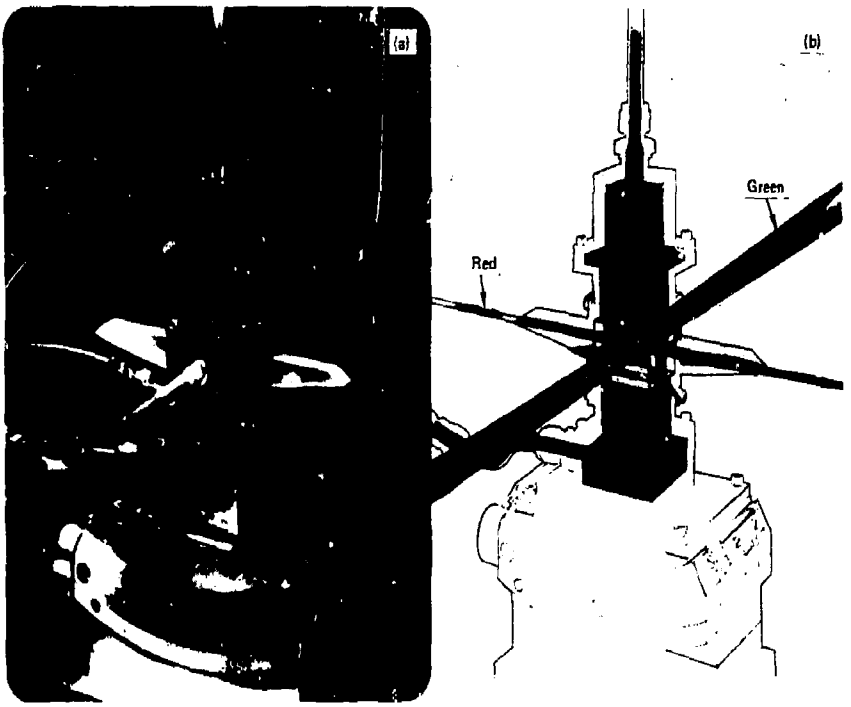

Fig. 4. Dye laser amplifier, shown with mutually orthogonal directions for excitation, flow, and amplification.

while simultaneously confirming with an interferometric diagnostic that similar performance could be achieved at a high repetition rate.

Development of adequate CVLs has been an important goal of the AVLIS program since late in 1974.* A progression of the lasers that have been developed is shown in Fig. 5. In 1975-76 CVLs in our laboratories could produce only 1 to 2-W average power. This was increased to 10 to 20 W in 1977 and in recent years we have demonstrated large-bore oscillator performance at well above 100 W (amplifier power is typically 30% higher than oscillator power). One of these 100-W lasers is shown operating in Fig. 6.

In pursuit of a laser system that would be capable of enrichment experiments and would provide a demonstration of laser electro-optic integration, we began in 1975 to design and assemble a prototypical AVLIS laser system. As in our AVLIS plant design, the system consists of two major entities, a waveform generator (SPP-II) and a power amplifier (Venus); this SPP-II/Venus system is shown in Fig. 7. This composite system demonstrates all of the basic

*Development and construction of CVLs at LLNL has been pursued by LLNL and at General Electric Co. under contract to LLNL.

Fig. 5. Evolution (since 1974) in the size and power capability of CVLs.

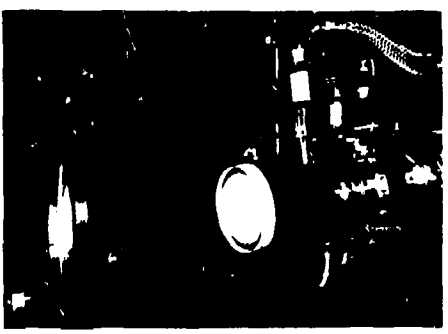

Fig. 6. Large-bore CVL oscillator operating at approximately 90-W output power.

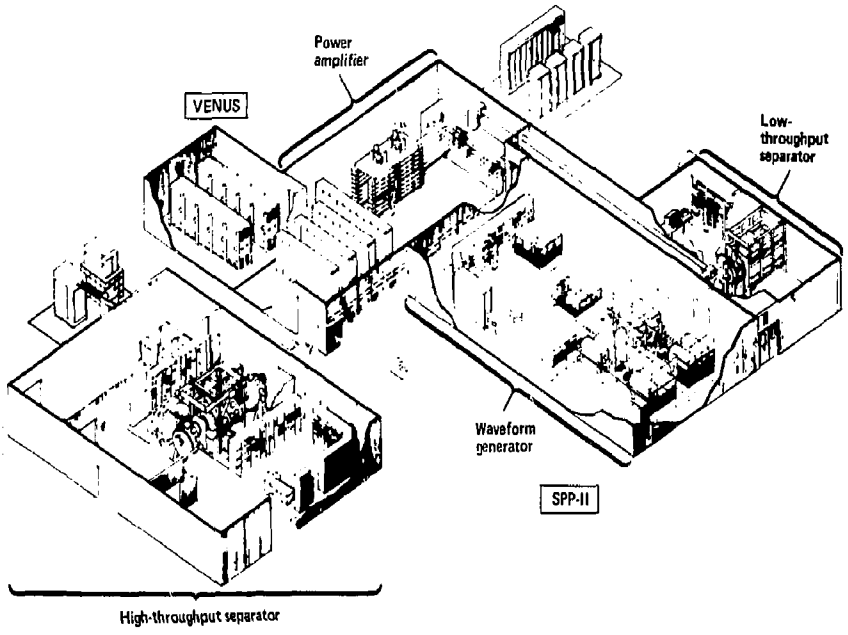

Fig. 7. Major on-line facilities of the LLNL AVLIS program.

functions of a plant laser electro-optical system. It includes 40 CVLs, 5 dye master oscillators (for fast and convenient frequency agility), 7 dye preamplifiers, and a full complement of transport optics, support structures, and diagnostics and controls. Dye master oscillators are held on frequency with automatic controls. An autoalignment system maintains the pointing accuracy for the beams to be amplified in Venus and the relative timing of all CVLs is under computer servo control. The complete system provides tunable power for use in enrichment experiments and in other evaluations of the AVLIS process performance.

This laser system typically runs 13 h/day, 5 days/week, and has accumulated greater than 2000 CVL h/week of operation. In a recent series of tests we operated the system in an intensive manner. It was on 18 to 19 h/day and available to deliver power 14 to 15 h/day. In one elapsed time period of 92 h, the system, even though it was not operated around the clock, was in a system-ready mode for more than 50 h, or 55% of the time.

AVLIS Program Development

Construction of several additional laser electro-optical systems is planned for the intermediate time period between now and a full production facility. The two largest systems are the Special Isotope-Separation Laboratory (SISL) system, expected to become operational at LLNL in the 1984-85 time period, and the Development Demonstration Module (DDM) system to be built in Oak Ridge, TN, by Union Carbide Corporation and LLNL for operation in 1988. An important purpose of the laser system within the DDM is to prototype the subsequent production system. The SISL laser system, expected ultimately to supply tunable output power at the kilowatt level, is shown in Fig. 8. The SISL building is presently under construction.

Fig. 8. Artist's concept of the new AVLIS office/laboratory complex (SISL) now under construction.