Laser Isotope Enrichment

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The idea of laser isotope enrichment grew from the laser's ability to concentrate its output power in a narrow range of wavelengths. Different isotopes of the same element are very hard or impossible to separate chemically, but the difference in their masses leads to differences in their spectra, which in principle can be used to selectively excite one isotope and isolate it by some photo-induced process.

The first proposal came from the Atomic Energy Commission's (AEC's) Mound Laboratories in Miamisburg, Ohio, which in 1961 began a classified investigation of using lasers to enrich the concentration of fissionable uranium-235. Others independently proposed laser uranium enrichment. A company called Radioptics proposed it to the AEC in 1963 and later unsuccessfully sued the AEC for violating their trade secrets. A French group received a patent in France in 1965, and by the time a U.S. version of the patent issued in 1969 the idea was looking attractive.

The impetus came from the development of the tunable dye laser and the growth of nuclear power. The U.S. depended on the gaseous diffusion process developed during World War II to enrich U-235 concentration to the levels needed for atomic bombs. Gaseous diffusion is energy-intensive, expensive, and raises U-235 concentration only a small amount on each pass. Laser enrichment offered to reduce cost, improve efficiency, and increase recovery of U-235.

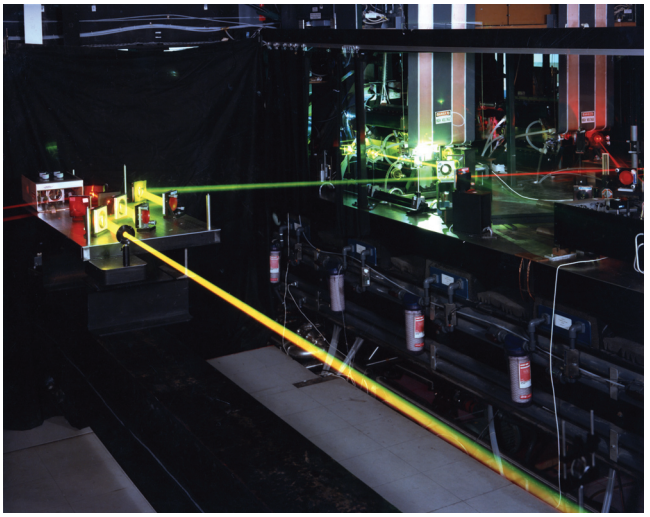
At the Avco-Everett Research Laboratory, Richard Levy and G. Sargent Janes developed a two-step process to enrich U-235. First a dye laser would selectively excite U-235 atoms in uranium vapor, then an ultraviolet laser would ionize the excited U-235 atoms, so they could be collected [1]. (Figure 1 shows the process.) Avco lacked money to develop the technology, so they formed a joint venture with Exxon Nuclear, hoping to build a private uranium enrichment business.

Avco-Everett founder Arthur Kantrowitz initially worried that laser enrichment might open the door to nuclear proliferation. "At first glimpse it seems like it's a garage operation. A garage operation for separating uranium isotopes is a frightening thing," he recalled in a 1985 interview. He imposed special security restrictions but eventually realized "this is not an easy way to make a bomb. It might be an easy way to make 1000 bombs, but it is not a terrorist operation" because of its technical complexity [2].

In 1972 the AEC launched competing laser uranium enrichment projects at its Los Alamos and Livermore laboratories.

John Emmett, director of Livermore's laser program, chose to try selective excitation of U-235 atoms in uranium vapor with the relatively well-developed tunable dye laser. That paralleled the Avco approach but was based on earlier work by Ray Kidder of Livermore. They proposed a two-step process, starting with using visible output of a narrow-band dye laser tuned to excite U-235, then ionizing the excited uranium atoms. In early 1973 Livermore hired three developers of the first continuous-wave dye laser from Eastman Kodak, Ben Snavely, Otis Peterson, and Sam Tuccio, to start and manage the program. "It seemed like an exciting thing to do at the time," Snavely recalled many years later, an opinion echoed by the other two.

At Los Alamos, Reed Jensen and John Lyman chose to try selective enrichment in UF_6, the compound used in gaseous diffusion, which sublimes at about 55 deg Celsius and is easier to handle than uranium vapor. They found a large isotope shift in a 16-μm absorption band of UF_6 and discovered that ultraviolet photons could photodissociatiate excited UF_6 molecules, precipitating solid UF_5 from the gas phase reaction and releasing free fluorine into the gas. Developing



▲ **Fig. 1.** The Avco-Everett scheme for laser enrichment of uranium required the combination of four laser beams to produce the desired wavelengths to select U-235. (AVCO Research Laboratory, courtesy AIP Emilio Segre Visual Archives, Physics Today Collection.)

▲ **Fig. 2.** Four milligrams of uranium with its U-235 concentration enriched to 3% by a dye laser process at Livermore is visible at the bottom of this test tube—the first time this much uranium was enriched by lasers. 1975 photo from the Lawrence Livermore National Laboratory. (Lawrence Livermore National Laboratory.)

the process would require finding a narrowband 16-μm laser that could generate enough power to dissociate $^{235}UF_6$. Los Alamos chose C. Paul Robinson to be the director of the program to solve all those problems.

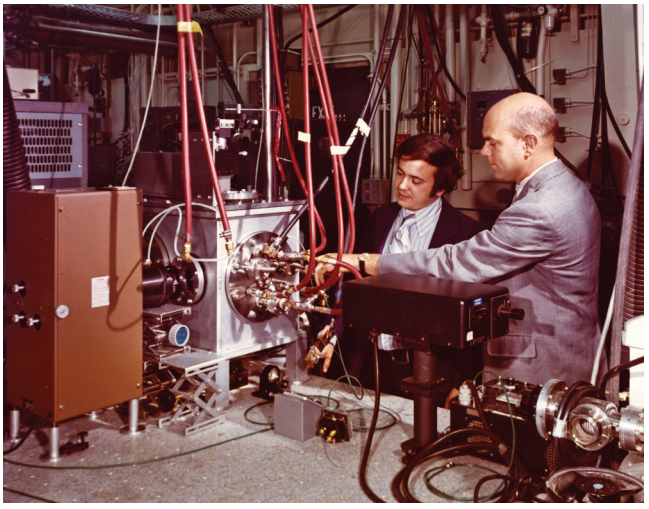
At Livermore, Snavely clashed with Edward Teller and particularly recalled Teller's disapproval of a metal-vapor process that eventually was adopted for the Atomic-Vapor Laser Isotope Separation (AVLIS) program (see Fig. 3). When Snavely told him he expected the process to succeed by the end of September, Teller grumbled, "You mean by the 31st of September?" Snavely ignored him, and Teller pointedly said, "You know September has only 30 days." Snavely then replied, "Yes, I knew that, but I wasn't sure that everybody knew it," and Teller threw him out of his office. Yet Snavely recalled that after he succeeded, Teller made a point of congratulating him when they met at a University of California ceremony.

Livermore was the first to report uranium enrichment in June 1974 at the International Quantum Electronics Conference in San Francisco. They illuminated a beam of hot uranium vapor with a dye laser emitting near 590 nm, selectively exciting U-235 atoms that then were ionized with ultraviolet light from a mercury arc lamp [3]. Figure 2 shows the enriched uranium oxidized to form "yellowcake" visible in the bottom of a test tube. That process would not scale to mass production, but Richard W. Solarz and Jeffery A. Paisner later found a way to coherently pump the selected isotope all the way from the ground state to an autoionization state (Rydberg level), permitting cost-effective isotope separation.

Meanwhile, Los Alamos developed a two-step process in which a 16-μm source first excited vibration of cooled UF_6 molecules containing U-235 and then a 308-nm xenon-chloride laser removed a fluorine atom from the excited UF_6. The resulting UF_5 precipitated as a solid that could be filtered from the gas. Developing the cooling process was a major accomplishment; it required flowing UF_6 diluted with a noble gas through a supersonic nozzle to

cool it while keeping it in the gas state to maintain a narrow line spectrum, and pulsing the gas flow in synchronization with the laser pulses.

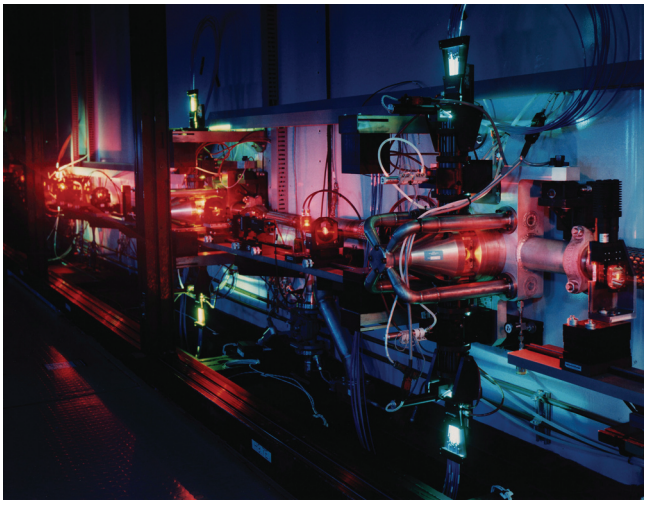
Los Alamos first demonstrated enrichment in 1976, but the details were kept classified until 1978, when the news was released to *Laser Focus* in a remarkably roundabout way. A reporter first visited the lab, but researchers who showed him around the lab told him nothing about uranium enrichment results. A few days after his visit, a university researcher phoned the author to suggest that he call Los Alamos and ask, "Have you enriched macroscopic quantities of uranium?" The author did, and it was as if he had said "open sesame." Los Alamos officials were delighted to answer "yes" and provide details on their two-step process [4]. Evidently security had authorized the disclosure only in response to those exact words.



▲ **Fig. 3.** Benjamin Snavely (right) and Sam Tuccio examine the laser system used to enrich U-235 concentration in hot uranium vapor at Livermore. (Lawrence Livermore National Laboratory.)

By the late 1970s, uranium enrichment was a major research program. The two competing government programs consumed a total of about a hundred million dollars a year. Jersey Nuclear-Avco Isotopes continued its atomic uranium enrichment research, spending a total of over $70 million before shutting it down in 1981 after the government refused to fund a demonstration plant [5].

The laser community tended to see selective laser excitation as the big challenge and focused its attention on the lasers. Livermore had the more straightforward problem, and built a bank of high-power copper-vapor lasers to pump large dye lasers for its AVLIS program. By 1982, Livermore had a master oscillator/power amplifier (MOPA) array of copper-vapor lasers emitting 7 kW, pumping a dye-laser MOPA array emitting 2.5 kW day in and day out (see Fig. 4). Los Alamos needed to develop a 16-μm source, which it achieved by Raman-shifting the output of carbon-dioxide lasers. Although details of that technology were kept under security wraps, Los Alamos was able to generate the required power and linewidth with efficiency considered reasonable at the time. The heart of that system was a hydrogen-fluoride optical parametric oscillator, developed by George Arnold and Robert Wenzel. That oscillator was originally used to perfect the spectroscopic data and was subsequently used as the seed source for the Raman-shifted carbon-dioxide laser amplifier.



▲ **Fig. 4.** Copper-vapor pumped dye lasers scaled for uranium enrichment at Livermore. Most of the green light from the copper lasers was tightly confined so it could efficiently pump dye lasers, which emitted red-orange light tuned to three absorption lines of U-235 vapor. (Lawrence Livermore National Laboratory.)

Little mentioned at the time was a parallel, classified program aimed at purifying plutonium for use in nuclear weapons. Fissionable plutonium-239 is produced by irradiating U-238 with neutrons in a special reactor. However, some U-238 atoms absorb a second neutron, producing Pu-240, which

fissions spontaneously so only low levels can be tolerated in nuclear weapons. The "special isotope separation" program launched in 1975 was intended to produce essentially pure plutonium-239. It remained small for a few years, reaching only about $5 million in 1980, but funding jumped in 1981, and the Reagan Administration boosted the budget to $76 million in 1983 in a plan to assemble more than 14,000 additional nuclear warheads in the next decade. Livermore and Los Alamos each had their own plutonium projects, based on adapting their preferred processes for use with plutonium.

Although public statements stressed progress in selective laser excitation of U-235, both labs faced problems in producing a final product. The fundamental problem with both programs was that chemical and physical reactions after the successful laser-induced chemistry or ionization quickly scrambled the isotopes, making it difficult to collect the initially enriched U-235 or isotopically purified plutonium. In the Molecular Laser Isotope Separation (MLIS) program, the pentafluoride molecule could easily steal a fluorine atom from another hexafluoride molecule before it condensed on the collector. In the AVLIS case, the laser-generated ion could steal an electron during the plasma extraction process and be lost from the enriched stream.

Those problems did not deter the Department of Energy's (DOE's) support for laser enrichment, and in 1982, DOE picked the Livermore atomic-vapor approach for uranium and shuttered the molecular separation program at Los Alamos. As would be expected in such decisions, both scientific and political considerations affected the final outcome.

However, a slowdown in nuclear power development after the 1979 Three Mile Island reactor accident reduced concerns about supplies of enriched uranium. As fears of oil shortages eased, new technology for producing reactor fuel became a lower priority. DOE delayed its decision to build a pilot AVLIS uranium plant at Livermore until 1985. The main rationale was economic: DOE calculated that AVLIS could produce separative work units (SWUs), a measure of uranium enrichment, for as little as $25, compared to $70 to $80 for gaseous diffusion. The plan called for phasing out gaseous diffusion except for highly enriched uranium, which the Livermore approach was not configured to produce.

Livermore began operating a pilot-sized laser and separator system in 1986 and spent several years refining the technology before they were able to operate full-sized equipment for tens of hours (see Fig. 5). They demonstrated plutonium enrichment first in the early 1990s, with uranium enrichment and scaling to larger scales to follow.

▲ **Fig. 5.** One of three units for separation of U-235 in Livermore's pilot plant for laser isotope separation. (Lawrence Livermore National Laboratory.)

By this point two external developments affected the need for laser isotope enrichment. The end of the Cold War stopped the build-up of the U.S. nuclear arsenal and eliminated the pressure to purify plutonium for new nuclear warheads. It also made surplus highly enriched uranium from the Russian arsenal available for down-blending into reactor fuel at prices well below freshly enriched uranium.

The 1992 transfer of DOE's enrichment program to the United States Enrichment Corporation put Livermore's program on standby until July 1994. Livermore completed its uranium-enrichment pilot plant in the fall of 1997, and it processed several thousand kilograms in a series of runs involving 24-hour operation of copper-vapor pumped dye lasers spread over 1.5 years. During that time, they also demonstrated doubled-neodymium pumping of

the dye lasers for future pumping in a production facility. But U.S. Enrichment halted those tests in June 1999, citing low prices for enriched uranium and high internal expenses for other work [6]. Those cuts also stopped plutonium enrichment. The motivation for continuing the laser program also was hurt by the continuing successes of the centrifuge programs that had been ongoing worldwide. All told, Livermore's quarter century of laser isotope separation development had cost more than $2 billion.

By then, molecular laser isotope enrichment had been revived by two Australians, Michael Goldsworthy and Horst Struve, who in 1990 began developing a process they called SILEX for Separation of Isotopes by Laser EXcitation. Like the Los Alamos process, SILEX is based on cooling UF_6 so resonances for molecules containing U-235 and U-238 are clearly separated and the molecules are concentrated in the ground state. Excitation with a 16-μm laser source selectively excites molecules containing U-235, producing a product stream enriched in U-235 and a "tails" stream depleted in U-235 but richer in U-238. Details are classified, but the main differences from the old Los Alamos process are thought to be in extraction of the laser-excited U-235 fraction of the material. In the information about this process there has been no hint of the laser-induced chemistry or ionization that initiated the isotope scrambling that plagued the earlier programs.

U.S. Enrichment supported Goldworthy and Struve's work from 1996 to 2002, and after that funding stopped, they formed a public company called Silex Systems Ltd. in Australia. Silex eventually licensed a joint venture of General Electric and Hitachi called GE Hitachi Nuclear Energy to use the process. After a few years of study, GE Hitachi Nuclear applied for a license to build a pilot plant in North Carolina, which the Nuclear Regulatory Commission approved in 2012. The plan is controversial, and the final outcome remains to be seen, but after a near-death experience, laser uranium enrichment is clinging tenuously to life.

Acknowledgment

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