

## The South Korean Laser Isotope Separation Experience

By Mark Gorwitz (1996)

The literature concerning activities in the laser isotope separation area by South Korea has been limited in nature. Most important publications have been published in Korean and have not been translated.

The Laser Spectroscopy Laboratory and the Laboratory for Quantum Optics, Korean Atomic Energy Research Institute (KAERI) located in Taejon are the lead laboratories for research in the laser isotope separation area. Support is provided in the spectroscopy area by the Department of Physics, Korea Advanced Institute of Science and Technology, Taejon. TEA CO<sub>2</sub> laser research is done by the Department of Physics, Sogang University, Seoul. Copper vapor laser research is done by the Department of Physics, Kyungpook National University, Daegu and the Department of Physics, Chonnam National University, Kwangju. Support in the dye laser area has been provided by the Department of Physics, University of Ulsan, Ulsan. Support in the optics area has been provided by the Korea Research Institute of Standards and Science, Taejon.

The early Korean effort during the mid 1970's was directed towards the separation of isotopes of light elements by the multiphoton dissociation process. TEA CO<sub>2</sub> lasers were developed for this purpose.

A 1981 KAERI document contained the following information about early laser isotope separation efforts:

"To achieve a separation of isotopes by multiphoton dissociation method, high power laser is needed. In our laboratory, a photoionized TEA CO<sub>2</sub> laser which has a power of 1 Mega Watt was successfully constructed. In TEA CO<sub>2</sub> laser operation it is important for high power and high efficiency to establish a stable glow discharge uniformly, UV preionization employing a trigger wire was used. The uniformity of discharge between electrodes depended on the capacity of storage capacitor. And the number of initial electrons effective in the main gap discharge depended on the position of a wire. The electrode shape which have a Rogowski profile was an important parameter for the uniform discharge. Optimum performance was obtained with 1:1:8 ratio of CO<sub>2</sub>:N<sub>2</sub>:He at atmospheric pressure. Also a discharge tube (68 cm in length) for a CW ion laser was made of quartz tubes. It consists of a small bored tube (5 mm i.d.), water jacket, gas return path, Brewster windows, and electrodes closed by a molybdenum foil seal. The current-voltage characteristics have been investigated up to current density of 66A/cm<sup>2</sup> for the tube filled with 0.1 torr Xe."

The effects of seeding on the stability and uniformity of the glow discharge in a TEA CO<sub>2</sub> laser and well as the output were

studied. Researchers at Sognag University studied the mechanism of decomposition of various additives and concluded that studying the decomposition products of tri-n-propylamine (current best quality seeding agent) will lead to a more effective seeding gas."

Some details of later TEA CO<sub>2</sub> have been published. The following information was obtained from a 1986 paper: "Our experiment is achieved using a double-discharge TEA CO<sub>2</sub> laser with a hexagonal acrylic box having Brewster windows and a CO<sub>2</sub> cell. A gold-coated total reflector with 10-m radius of curvature and a ZnSe flat half mirror with 80% reflectivity mounted to the end of the cell are set 175 cm apart, forming an optical cavity. The electrodes of brass have a discharge length of 24 cm, a width of 2 cm, and a height of 3 cm. The preionization is achieved using two tungsten trigger wires with 330-pF trigger capacitors. The laser is energized using a two-stage Marx-Bank generator with a 26-nF capacitor per stage, and the voltage to the circuit is set at 18 kV. The laser medium introduced into the amplifier is a mixture of CO<sub>2</sub>, N<sub>2</sub>, and He gas, of which the ratio is 2:1:10."

"The CO<sub>2</sub> cell made of pyrex glass has a total length of 60 cm and a heating length of 35 cm. One end of the cell facing the cavity is blocked by an anti-reflecting-coated ZnSe flat window, and heating is done by a Ni-Cr wire and a transformer. Half of the CO<sub>2</sub> gas flowing in the cell is exchanged per second."

"The laser operates a dominant 10.6-um P-20 and P-18 line in a low pressure of CO<sub>2</sub> cell and a single-line operation of P-20 at more high pressure."

Further information was contained in a 1985 KAERI document:

"Research on laser isotope separation of deuterium using Infrared Multi-Photon Absorption/Dissociation (IR MPA/D) and UV predissociation were reviewed and several kinds of lasers were built for this purpose. A tunable TEA CO<sub>2</sub> laser with power of about 10 MW was assembled and a HF chemical laser with output energy of 300 mJ was built. These lasers are not ready to be used as sources for IR MPA/D experiment yet. The TEA CO<sub>2</sub> laser needs modification for more stable output and higher repetition rate and the HF chemical laser needs improvement for more output energy and tunability. Also a KrF excimer laser was built for UV predissociation experiment, but requires modification for stable output."

Some additional details describing the above lasers are available. The TEA CO<sub>2</sub> laser made in 1983 was reassembled to increase the output power and tune the wavelength. The HF chemical laser was produced for IRMPD of molecules with an -OH structure. And lastly the KrF excimer laser was to be used to pump a dye laser to generate 345 nm radiation for UV predissociation experiments.

Details are also available of hydrogen isotope separation

experiments. Both deuterium and tritium separation were looked at. For deuterium, the literature was reviewed and a state of the art report issued (KAERI/AR-248/84).

The absorption coefficient of  $\text{CDF}_3$  neat and in the presence of  $\text{CHF}_3$  was determined. The effects of added inert gas on  $\text{CDF}_3$  absorption was also studied. Actual photochemical energy requirements for deuterium separation by multi-photon dissociation were calculated and compared to the  $\text{H}_2\text{S}/\text{H}_2\text{O}$  system.

A 1990 paper reports on the selective photodissociation of  $^{32}\text{SF}_6$  by a TEA  $\text{CO}_2$  laser. The following details are provided: The molecular selective photodissociation of  $^{32}\text{SF}_6$  by IRMPD in natural  $\text{SF}_6$  was investigated using a pulsed TEA  $\text{CO}_2$  laser. The TEA  $\text{CO}_2$  laser consisted of 2 stage Marx bank power supply and for uniform discharge, it was designed to use the UV preionization generated by corona discharge along the surface of glass sheets on both electrodes. The laser gave multimode energy of about 1.7 J/pulse, pulse width (FWHM) of less than 100 ns and overall efficiency of about 9.6%. The laser beam (10P(20) line) was focussed into the reaction cell filled with reactant mixture,  $\text{SF}_6/\text{NO}$  or  $\text{SF}_6/\text{H}_2$ ; the  $^{32}\text{SF}_6$  absorbs the laser photons through multiphoton absorption and dissociates, selectively. The study focused on the relative reaction selectivities of  $^{32}\text{SF}_6$  under influence of additives, such as  $\text{NO}$  and  $\text{H}_2$  which were expected to act as a radical scavenger of free radical energy generated from  $\text{SF}_6$  dissociation. The relative reaction selectivity was determined by measuring the unreacted  $^{34}\text{SF}_6/^{32}\text{SF}_6$  ratios using a mass spectrometer."

The following further information was contained in a 1991 KAERI document:

"For IRMPA/D studies, we measured IR fluorescence emitted from vibrationally excited  $\text{DF}^*$  or  $\text{HF}^*$  produced in IRMPD of  $\text{CDF}_3/\text{CHF}_3$  using IR monochromator and InSb IR detector. We could detect the ir fluorescence of  $\text{HF}$  from the sample mixture  $\text{CDF}_3/\text{CHF}_3$  to which the  $\text{CDF}_3$  was selectively irradiated. This means that the molecular selective excitation of  $\text{CDF}_3$  in  $\text{CDF}_3/\text{CHF}_3$  did not give molecular selective dissociation of  $\text{CDF}_3$  because of the fast intermolecular vibrational energy transfer from the excited state  $\text{CDF}_3$  to the ground state  $\text{CHF}_3$ . This technique will play an important role for measuring isotope selectivity in IRMPD of  $\text{CDF}_3/\text{CHF}_3$ . We developed data analysis technique for the laser fluorometer to improve analytical speed and accuracy. We calculated a fluorescence intensity at time zero using two values obtained by intergration of two intervals on the time-resolved fluorescence signal. Applying this method, we could eliminate any interference effects from quenching elements or temperature fluctuations of samples, effectively."

Copper vapor laser research has been ongoing since the mid 1980's. This early effort was done at the University of new Mexico. The lead American researcher was J.J. Kim and the lead Korean researcher K. Im was on leave from Chonnam National

University. J.J. Kim later moved to the Center for Research in Electro-Optics and Lasers, University of Central Florida. N. Sung after completing work there on transverse-discharge copper vapor lasers returned to the Korea Advanced Energy Research Institute. The results claimed that the transverse-discharge CVL's could be developed for practical applications.

In a 1987 paper written with J.J. Kim, N. Sung stated that: The copper-vapor laser (CVL) has recently received a great deal of attention as a practical device for applications in dye-laser pumping for isotope separation..." A research contract from Lawrence Livermore provided part of the funding for this effort.

By 1987 researchers at Kyungpook National University had succeeded in developing a 1.5 W, 5 kHz repetition rate laser using xenon as the inert buffering gas.

A 1978 paper provided details of a tunable dye laser pumped by a pulsed N<sub>2</sub> gas laser suitable for spectroscopic experiments. "The dye laser is transversely pumped by the focused 3371 AA emission line of a pulsed N<sub>2</sub> gas laser. The N<sub>2</sub> laser's peak power output is 1 MW at the repetition rate up to 100 pulses/sec. The dye laser output power is 1.36 kW at 2.5 ns pulse width, the spectral bandwidth is  $3.3 \times 10^{-2}$ /AA centered at 5900 AA, the emission wavelength is variable over the visible range by choosing one of seven dyes such as PPO, PBBO, POPOP, 4-methyl umbelliferone, Rhodamine 6G, Rhodamine B and cresyl violet perchlorate. Using the present tuning method, with the holographic grating, the prism and the etalon, the tunable dye laser with a short cavity, 15 cm in length is realized."

A KrF excimer laser for pumping a dye laser was developed in the early 1980's. The 345 nm generated laser radiation was to be used for UV preionization experiments. Problems however arose in assembling the grating required for the dye laser.

Considerable work has also been done on the development of XeCl excimer lasers. Experiments have focused on the development of controllable long pulse lasers using a simple capacitor-discharge circuit. An injection locked laser with a modified-branch conformal unstable resonator was also developed and characterized. Lastly a UV-preionized discharge-pumped laser was developed. The following design details were provided: "A maximum output energy of 96mJ/pulse, pulse duration of 20 ns (FWHM) and beam cross section of  $7 \times 17$  mm<sup>2</sup> have been extracted from the gas mixture of HCl/Xe/He = 0.2/3.1/96.7% in total pressure of 3.5 atm, which was pumped with 26 kV of charging voltage.

Frequency has been controlled in pulsed dye lasers by the use of a volume holographic transmission grating along with a tuning mirror. The purpose of the grating was to maximize energy conversion and to minimize linewidth distortion. A Nd:YAG laser was used to pump a dye cell containing Rhodamine 6G.

"The energy conversion efficiency is greatly improved compared with that of the grazing incidence type, and the linewidth is reduced compared with that of the Hansch type." The Hansch type was used in early Israeli pulse dye lasers.  
Argon ion lasers

Argon ion pumped dye laser research has been by T.S.Kim of the Department of Physics, University of Ulsan in collaboration with the University of Rochester, Rochester, New York. These experiments involved measuring the photon-number fluctuations in a single-mode dye jet laser. Argon ion pumped lasers are known to have been used by many groups in atomic vapor spectroscopic studies. Recent experimental results are described in the Israel section of this report.

Ti:Sapphire lasers suitable for spectroscopic have been developed by KAERI. The following details have been provided concerning this laser: "We have constructed a self-seeded Ti:sapphire laser oscillator by using a dual-cavity configuration that consists of a Littman configuration cavity and a partially reflecting feedback mirror. This configuration can be decomposed with two kinds of cavity, a grazing-incidence cavity and a standing-wave cavity. The former behaves as an injection seeder and the latter as a slave oscillator. This Ti:sapphire laser system is capable of delivering a continuously tunable laser pulse with a narrow linewidth. Injection at the laser emission region of the free-running Ti:sapphire laser resulted in essentially complete energy extraction."

South Korea has a very active program in the laser isotope separation area. Scientific support has been provided by both American and Russian researchers. American support has been in the area of laser development and Russian support has been in the AVLIS area. This AVLIS program aimed at separating isotopes of the lanthanide elements for use as burnable poisons in nuclear reactors has been under development for a number of years.

Arisawa stated "that tunable lasers for AVLIS application require high repetition rate, high average power, high reliability and high stability. Ti:sapphire laser or F-center laser could be candidates, but each needs a good pumping source and nonlinear crystal for converting the wavelength from fundamental IR wavelength to visible one at high efficiency. A direct application of the diode laser as a tunable source might be promising in the future, if wavelength range coverage, average power and price are satisfied."

Results were published in 1990 on selective photoionization of magnesium atoms. The abstract presented the following information: "A spectroscopic study of photoelectrons arising from nonresonant multiphoton ionization of magnesium atoms in a high intensity laser field is performed experimentally. Both the 532 nm and 1064 nm excitations in the intensity region of  $10^{10-13}$  W/cm<sup>2</sup> are used for single and double ionization. The emphasis is

placed on the photoelectron spectra and their their variations with laser wavelength, intensity, and polarization. Also, the ionization process of doubly charged ions which can be produced either by a stepwise process or by the simultaneous removal of two electrons is discussed."

The effects of the AC-Stark shifts on the selective resonant ionization of both Li and Sr was studied. The abstract of a 1993 paper reads as follows: "The authors show that the ionization rate and the isotope selectivity become sensitive functions of the wavelength and intensity of the laser due to the AC-Stark shifts of the energy levels involved in the two-photon resonant three-photon ionization of Li and Sr atoms. They also examine the optimum conditions for isotope separation."

A paper contained in the same journal presented details of numerical calculations. The following abstract was given: " The authors present results of numerical calculations obtained through solving integro-diferential equations for the electron density matrices  $\sigma_{11}$  and  $\sigma_{22}$  and the ionization rate  $P$  in two-photon resonant three-photon ionization of Li and Sr, as well as the selectivity  $S$  for isotope separation. They also compare their results with the predictions from quasi-stationary solutions based on the rate approximation. Their numerical results for  $P$  and  $S$  show that the quasi-stationary solutions are valid at very high and very low laser intensities when the isotope shift is large as in the case with  $4s$  of  ${}^6\text{Li}$  and  ${}^7\text{Li}$ , and that the validity is rather limited in the cases with small isotope shift as in  $5p_{3/2,1/2}$  of  ${}^{88}\text{Sr}$  and  ${}^{90}\text{Sr}$ ."

These calculations were performed on a CRAY C90 YMP supercomputer.

The most important work in the area of atomic vapor laser isotope separation (AVLIS) has involved the rare earth metals especially ytterbium. This work carried out at the Korea Atomic Energy Research Institute has been a partial collaborative effort with the General Physics Institute, Moscow. Support has been in both the theoretical and experimental areas.

Information on the selectivity of photo ion extraction for isotopes of the rare earth elements has been published. The abstract presented the following details: "An analysis of selectivity of three-stage photoionization for isotopes of rare-earth atoms with (1-2) GHz isotopic shifts in the absence of polarization effects is performed. Because of field broadening, a sufficiently high selectivity  $\eta > \text{or} = 100$  is achieved only for low average laser intensities,  $I < \text{or} = 10 \text{ mW/cm}^2$ . The excitation of ions produced in photoionization of atoms by an electric field is investigated. The dependence of the selectivity on ion and gas densities, as well as on parameters of the external field, is calculated."

Experimental details were presented in 1995 on actual three-

photon polarization spectroscopy of ytterbium vapor. The General Physics Institute, Moscow was thanked for useful discussions.

The following experimental details were provided: The first (Lambda Physik FL3002/E) and second (Lumonics HD-300) dye lasers were pumped by the second harmonic of a Nd:YAG laser (Lumonics HY750). The wavelength of dye laser 1 was 555.648 nm, and that of dye laser 2 was 581.067 nm. Ytterbium atoms are excited to the intermediate state,  $4f^{13}(^2F_{7/2})6s^26p_{3/2}$  ( $J=2$ ) by these two dye laser pulses. Atoms in the intermediate state are excited to the autoionizing state by the third laser (Lumonics HD-300), which was pumped by the third harmonic of the Nd:YAG laser. the wavelength of the third laser was scanned from 430 to 660 nm using the dye Rhodamine 640, 610, and 590 and Coumarine 540A, 500, 480, 460, and 440."

"The laser have pulse durations of about 8 ns and are pulsed at the rate of 10 Hz. The laser pulses which are used for the excitation of intermediate states arrived at the chamber simultaneously. Approximately 8 ns later, the third laser pulse arrived at the chamber, to avoid the two-photon process. All lasers were incident to the chamber, with an angle of less than  $2^\circ$  between them."

"The linear polarization of lasers was improved by a Glan polarizer placed at the exit of each laser. Circular polarizations of the first and second lasers were made by lambda/4 waveplates. For the third laser, we used lambda/2 or lambda/4 Fresnel rhomb phase retarders. Each polarizer was placed in front of the chamber."

"The linewidth of the exciting laser was 5 GHz, and those of the other lasers were 3 GHz. The wavelengths of the third laser was calibrated by recording the optogalvanic signals from an Ar-Yb hollow cathode lamp (Cathodeon) simultaneously with the ion signal. The air wavelengths of the Ar lines listed in the NBS table were converted into vacuum wavelengths before evaluating the energy levels of the autoionizing states."

"An atomic vapor of Yb was generated by heating pure Yb metal (99.9%) in a tantalum oven. Atoms were collected by two circular apertures placed before the electrodes of a time-of-flight mass spectrometer (TOF MS). 1 us later, atoms were ionized by three dye lasers, and the ions were extracted by applying a voltage pulse of 200-V/cm amplitude to the electrode for 10 us to reduce the perturbation due to a dc electric field. The extracted ions were analyzed by TOF MS (flight length: 1.5 m). Because we used natural Yb, a mass analyzer was needed for angular momentum identification. The ion signals were integrated by a boxcar (Stanford SR245) and stored in a computer."

"When an autoionizing state has a broad linewidth, the peak positions and the intensity of the ion signals are affected by the variation of the dye laser energy. Thus the laser energy was

also recorded as a function of wavelength, and ion signals normalized by the laser energy were used for analysis. The intensity of the third laser was attenuated with a neutral density filter enough to avoid depletion broadening of the line profile."

"As a result of this experiment, 17 autoionizing states were found in the investigated energy range from 50400 to 58000  $\text{cm}^{-1}$ . The line profiles of the autoionizing states were nearly symmetric, and some states had very narrow linewidths comparable to those states observed by Bekov. The results of angular momentum identification required revision of electronic configuration assignments of the states investigated by Borisov. We propose that some autoionizing states with large excitation cross sections be used for efficient photionization."

A later paper added the following comment: "Some scheme of Yb has the possibility to yield high efficiency in selective ionization of an isotope."



## Section 10: South Korean Laser Isotope Separation Work

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