The South Korean Laser Isotope Separation Experience

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I. Introduction

Mark Gorwitz, Special Contributor to the Institute for Science and International Security (ISIS) Online, writes: "Taken as a whole, the open literature shows the extent of South Korean research in the area of laser isotope separation. Technology learned in one enrichment program has been successfully transferred to another. The undeclared uranium enrichment experiments have been the main beneficiary of such a transfer and are a proliferation concern that deserves to be thoroughly examined by the IAEA."

II. Essay by Mark Gorwitz

"The South Korean Laser Isotope Separation Experience" by Mark Gorwitz

Introduction and Findings:

South Korea has been actively researching molecular and atomic vapor laser based isotope separation techniques since the early 1980s. These efforts have been geared towards supplying industrial, medical and civilian nuclear applications.

In September 2004, a number of news reports appeared concerning experiments conducted by researchers at the Korea Atomic Energy Research Institute (KAERI) using laser isotope separation to enrich and separate uranium isotopes. News sources reported that levels of enrichment averaged about 10% and reached almost 80%. According to the South Korean government, about 200 milligrams of enriched uranium were produced. South Korea reportedly used 3.5 kilograms of uranium as feed for the experiments.

According to a 2001 article written by Lawrence Livermore National Laboratory, an atomic vapor laser isotope separation program (AVLIS) "was regarded as too difficult a technology for a typical proliferating nation in the middle economic rank to utilize. However as technology advances, this will not remain so".(2) The current case of South Korea is illustrative of this. South Korea was able to successfully apply knowledge learned in earlier AVLIS efforts on elements such as gadolinium and ytterbium to an undeclared uranium enrichment program. Nucleonics Week quotes Dr. Jeffrey Eerkens, an expert in laser isotope separation methods, as saying, "on the basis of the experience that KAERI scientists apparently acquired using the laboratory equipment for separation of stable isotopes such as ytterbium or samarium, it would have been very straightforward for KAERI to have enriched a small quantity of uranium using the same equipment. The required frequency for U-235 is different but the technology is the same. If they knew how to enrich ytterbium, then they could enrich uranium".(3).

This report, which surveys the open literature, shows the extent of South Korean research in the area of laser isotope separation. The undeclared uranium enrichment experiments are a proliferation concern that deserve to be examined thoroughly by the IAEA.

Background:

South Korea's multiyear laser isotope separation research programs have involved researchers from KAERI, private industry and academia. The majority of the program's research efforts were clearly aimed at industrial and medical applications. Pertinent scientific research that has been carried out and reported in publicly available reports and the scientific literature was analyzed for this and an earlier report. Efforts in both the molecular and atomic vapor isotope separation schemes were looked at. Sources used were Chemical Abstracts, Engineering Abstracts, Nuclear and Energy Citation Abstracts, INSPEC and the European patent database.

The scientific literature concerning activities in the laser isotope separation area by South Korea has been fairly detailed, and open scientific publications of experiments detailing spectroscopy of uranium vapor date back to the early 1990s. Although most publications have been published in Korean and have not been translated, abstracts are usually available for those publications. A significant number of conference and scientific papers have also appeared in the English-language scientific literature. Lastly, a number of patents dealing specifically with laser isotope separation have been issued to South Korean researchers.

The Laser Spectroscopy Laboratory and the Laboratory for Quantum Optics, both located at the Korean Atomic Energy Research Institute (KAERI) 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, the Department of Physics, Sogang University, the Department of Physics, Chungnam University, and the Department of Physics, Yeungnam University. TEA CO2 laser research is done by the Department of Physics, Sogang University, Seoul. The Department of Physics, Kyungpook National University, Daegu, the Department of Mechanical Engineering, Chosun University, and the Department of Physics, Chonnam National University, Kwangju are carrying out copper vapor laser research. Support in the dye laser area has been provided by the Department of Physics, University of Ulsan, Ulsan. The

Korea Research Institute of Standards and Science, Taejon, has provided support in the optics area.

Molecular Laser Isotope Separation (MLIS) Related Research:

The early South Korean efforts during the mid 1970's were directed towards the separation of isotopes of light elements by the multi-photon dissociation process. TEA CO2 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 multi-photon dissociation method, high power laser is needed. In our laboratory, a photo-ionized TEA CO2 laser which has a power of 1 Mega Watt was successfully constructed. In TEA CO2 laser operation it is important for high power and high efficiency to establish a stable glow discharge uniformly, UV pre-ionization 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 has a Rogowski profile, was an important parameter for the uniform discharge. Optimum performance was obtained with 1:1:8 ratio of CO2:N2: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/cm2 for the tube filled with 0.1 torr Xe."(4)

The effects of seeding on the stability and uniformity of the glow discharge in a TEA CO2 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--propylamine (current best quality seeding agent) will lead to a more effective seeding gas."

Some details of later TEA CO2 research efforts have been published. The following information was obtained from a 1986 paper: "Our experiment is achieved sing a double-discharge TEA CO2 laser with a hexagonal acrylic box having Brewster windows and a CO2 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 pre-ionization 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 CO2, N2, and He gas, of which the ratio is 2:1:10".

"The CO2 cell made of pyrex glass has a total length of 60 cm and a heating length of 35 cm. An anti-reflecting-coated ZnSe flat window blocks one end of the cell facing the cavity, and heating is done by a Ni-Cr wire and a transformer. Half of the CO2 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 CO2 cell and a single-line operation of P-20 at more high pressure".(5)

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 pre-dissociation were reviewed and several kinds of lasers were built for this purpose. A tunable TEA CO2 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 CO2 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 pre-dissociation experiment, but requires modification for stable output".(6)

Some additional details describing the above lasers are available. The TEA CO2 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 pre-dissociation 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).(7)

The absorption coefficient of CDF3 neat and in the presence of CHF3 was determined. The effects of added inert gas on CDF3 absorption was also studied. Actual photochemical energy requirements for deuterium separation by multi-photon dissociation were calculated and compared to the H2S/H2O system.

A 1990 paper reports on the selective photo-dissociation of 32SF6 by a TEA CO2 laser. The following details are provided: The molecular selective photo-dissociation of 32SF6 by IRMPD in natural SF6 was investigated using a pulsed TEA CO2 laser. The TEA CO2 laser consisted of 2 stage Marx bank power supply and for uniform discharge, it was designed to use the UV pre-ionization 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 focused into the reaction cell filled with reactant mixture, SF6/NO or SF6/H2; the 32SF6 absorbs the laser photons through multi-photon absorption and dissociates, selectively. The study focused on the relative reaction selectivities of 32SF6 under influence of additives, such as NO and H2, which were expected to act as a radical scavenger of free radical energy, generated from SF6 dissociation. The relative reaction selectivity was determined by measuring the unreacted 34SF6/32SF6 ratios using a mass spectrometer".(8)

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

"For IRMPA/D studies, we measured IR fluorescence emitted from vibrationally excited DF* or HF* produced in IRMPD of CDF3/CHF3 using an IR monochromator and InSb IR detector. We could detect the ir fluorescence of HF from the sample mixture CDF3 /CHF3 to which the CDF3 was selectively irradiated. This means that the molecular selective excitation of CDF3 in CDF3/CHF3 did not give molecular selective dissociation of CDF3 because of the fast intermolecular vibrational energy transfer from the excited state CDF3 to the ground state CHF3. This technique will play an important role for measuring isotope selectivity in IRMPD of CDF3/CHF3. We developed data analysis technique for the laser fluorometer to improve analytical speed and accuracy. We calculated fluorescence intensity at time zero using two values obtained by integration 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".(9)

Laser Development:

Copper vapor laser research, which is an integral part of any atomic vapor laser isotope program, has been ongoing since the mid 1980's. The early foreign assisted research was done at the University of New Mexico and the lead Korean researcher K. Im was on leave from Chonnam

National University.(10) Another scientist, N. Sung, after completing work at the University of New Mexico on transverse-discharge copper vapor lasers returned to the Korea Advanced Energy Research Institute. The results published on this research claimed that the transverse-discharge copper vapor lasers (CVL) could be developed for practical applications.

In a 1987 paper, N. Sung stated: "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..."(11) A research contract from Lawrence Livermore National Laboratory 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. Researchers at Chonan and Chosun Universities are also involved in the development of copper vapor lasers useful for pumping dye lasers.

A 1978 paper provided details of a tunable dye laser pumped by a pulsed N2 gas laser suitable for spectroscopic experiments: "The dye laser is transversely pumped by the focused 3371 AA emission line of a pulsed N2 gas laser. The N2 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 x10-2/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".(12)

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 predissociation 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*17 mm2 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. And the following conclusion was reached: "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 pumped dye laser research has been conducted 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 in this area have been reported by Israeli researchers.

Ti:Sapphire lasers suitable for spectroscopic studies 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."(13)

Recent work has also been reported on the development of an extended-cavity violet diode laser that has been used to perform spectroscopic studies on samarium.(14) Earlier experiments done on samarium had used a commercial single-mode tunable diode laser. Similar experiments were also done on ytterbium and gadolinium.

Atomic Vapor Laser Isotope Separation (AVLIS) Related Research:

South Korea has had a very active program in the atomic vapor laser isotope separation area for many years. Both American and Russian researchers have provided scientific support to the South Korean program. American support has been in the area of laser development and Russian support has been in the AVLIS area. This AVLIS program has mainly 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, and has focused on the metals erbium, gadolinium, samarium, and ytterbium. This work has been a partial collaborative effort with the General Physics Institute, Moscow. Support has been provided by Russian researchers in both the theoretical and experimental areas.

Arisawa, a leading Japanese laser isotope researcher (JAERI) 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 photo-ionization of magnesium atoms. The abstract presented the following information: "A spectroscopic study of photoelectrons arising from non-resonant multi-photon 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 1010-13 W/cm2 are used for single and double ionization. The emphasis is placed on the photoelectron spectra and 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".(15)

The effects of the AC-Stark shifts on the selective resonant ionization of both lithium (Li) and Strontium (Sr) were studied. The abstract of a 1993 paper reads: "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 examined the optimum conditions for isotope separation."(16)

An April 1991 report entitled "Development of nuclear fuel: Development of laser spectroscopic technology in nuclear industry"(17), details multi-step photo-ionization experiments on mercury and the development of a computer code (MCDF) for calculating the transition probabilities of mercury atoms.

he May 1992 progress report states that the following research efforts were carried out that year: "multi-step photo-ionization spectroscopy of mercury atom carried out by 3-color 3-step ionization scheme, selective photo-ionization using polarization spectroscopy, design and construction of an ion separator chamber and theoretical study for spectroscopic parameters of mercury".(18)

The January 1993 progress report states "that laser atomic spectroscopic study on actinium element has been performed in many areas of spectroscopy." The report goes on to further state "In spectroscopic experiment, first and second ionized states for actinium element are identified and the most efficient scheme for actinium element is identified. In addition, the corrosion problem for filament material due to the heating of the actinium element has been studied".(19)

Another 1993 report entitled "Magnetic field effect on selective photo-ionization", states "the magnetic field effects on selective photo-ionization of the atoms of the lanthanides have been examined in a point of view of the enhancement of the efficiency of selective photo-ionization".(20)

In 1994 details on the optogalvanic spectroscopy of uranium, thorium and rubidium using diode lasers were published. Using commercially available sources, detailed experimental results were later published for erbium, gadolinium, lanthanum, lutetium, and samarium.(21) Further optogalvanic results were presented for uranium in a 1999 paper.(22) A reading of this and other papers shows that research in the optogalvanic spectroscopy area has been ongoing since the early 1990s. Brazilian researchers have openly reported similar research as part of their uranium laser isotope separation program. In a 1999 review of the Brazilian uranium laser isotope separation, it was stated that the optogalvanic effect "is a powerful and inexpensive technique for investigation of atomic and molecular species, and is particularly useful in the spectroscopy of refractory elements, like uranium"(23). Brazilian researchers using optogalvanic spectroscopy were able to measure two photon absorption assignments, lifetimes of uranium excited states and superelastic relaxation rates. South Korean scientists were able to obtain similar information from their experiments.

Argon/hollow cathode and sealed tube sources as sources of uranium vapor for spectroscopic experiments later gave way to resistively heated sources. A high temperature oven was later developed to generate vapor of the element being studied.

Hollow cathode sources were another means of measuring multi-photon absorption spectroscopy in uranium vapor used by KAERI. These experiments used commercial argon buffer gas sources obtained from Cathodeon, Ltd. Collisions between the buffer gas and the metallic vapor can cause problems in observing certain excited transition states. However, by careful choice of experimental technique, they were able to obtain useful information on photo-ionization and lifetime states of uranium and other elements such as samarium and ytterbium.

Another paper presented details of numerical calculations. The following abstract was given: "The authors present results of numerical calculations obtained through solving integral-differential equations for the electron density matrices sigma11 and sigma22 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 6Li and 7Li, and that the validity is rather limited in the cases with small isotope shift as in 5p/2/1/S of 88Sr and 90Sr". These calculations were performed on a CRAY C90 YMP supercomputer.

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 photo-ionization 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 > or = 100 is achieved only for low average laser intensities, I < or = 10 mW/cm2. The excitation of ions produced in photo-ionization 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".(24)

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, 4f13(2F7/2)6s26p3/2 (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 2œ 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 were 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 Ytterbium (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 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 photo-ionization".(25)

In 1996, results were presented on the two-photon selective photo-ionization of ytterbium. Both ion yields and selectivity were reported for 168Y. They claimed "that the selectivity for 168Y was increased higher than 20000 when the laser was blue-detuned to the most efficient position".(26) For the coherent excitation this experiment used a commercial single-mode dye laser (Lumonics, HyperDYE SLM) pumped by a frequency-doubled Nd:YAG laser (Lumonics, HY-750). Ionization was done using a broadband dye laser (Lumonics, HD-500) pumped by a frequency-tripled Nd:YAG laser. Ytterbium vapor was generated from a natural source using resistive heating.

The effect of laser intensity on the selective three-step photo-ionization of 168Yb has also been studied and it was noted that a noticeable change in isotope abundance of 168Yb occurred when the laser intensity was varied.

A later paper added the following comment: "Some scheme of Yb has the possibility to yield high efficiency in selective ionization of an isotope".(27) Using a diode-pumped solid-state laser and three-color dye lasers 20 mg of 25.8% enriched 168Yb was produced.

Further detailed information on the spectroscopy of atomic vapors is contained in a series of April 1998 reports. The first report focuses on studies of the "thermodynamic properties of metallic atoms in both gas and beam states". The second report focuses on ytterbium and presents details on experiments on fluorescence in dense atomic vapor, single color two-photon resonant three-photon ionization and the production of a high-temperature oven and its spectroscopic application. A third report provides information on the hyperfine coupling constants. And the in last report of the April series detailed studies were reported on both the flux and velocity of resistively heated atomic beam sources "in order to control the population of a specific state so that we can increase the efficiency of photo-ionization". These are some of the most important parameters involved in laser isotope separation.(28)

Similar two and three-color photo-ionization experiments have been reported for erbium, gadolinium and samarium in a series of scientific papers. Additional details of experimental work involving samarium were presented in an April 2000 report.(29) Using a high temperature oven, KAERI scientists were able to generate samarium vapor having an atomic density of 8x1014 atoms/cm3.

A very recent review on the Indian uranium laser isotope separation program states "that after establishing that uranium isotopes can be selectively ionized, we had to wait for some time to plan an isotope collection experiment. It was not possible to carry out the experiment unless a major improvement was made regarding uranium vapor generation".(30) Any large-scale enrichment program would use an e-beam source for heating and vaporizing material and this is not beyond the current capability of South Korea. KAERI scientists have been issued two patents in the area of e-beam heating in recent years.

At least three patents dealing specifically with laser isotope separation have been issued to scientists from the Korea Atomic Energy Research Institute (KAERI). One patent deals with feedback control methods, another deals with isotope separation of lanthanum or actinium by diode laser, and the third deals with an isotope separation scheme for thallium. These patents serve to illustrate the level of competence that South Korea has achieved in the area of laser isotope separation.(31)

Conclusion:

Taken as a whole, the open literature shows the extent of South Korean research in the area of laser

isotope separation. Technology learned in one enrichment program has been successfully transferred to another. The undeclared uranium enrichment experiments have been the main beneficiary of such a transfer and are a proliferation concern that deserves to be thoroughly examined by the IAEA.

III. Notes by Mark Gorwitz

1. This review was adopted from a 1996 report entitled: Second Tier Nuclear Nations: Laser Isotope Separation Programs, written by the author. Further scientific references may be found in the Open Source Scientific References bibliography associated with the current article.

2. For proliferation concerns over laser isotope separation see: S.A. Erickson, Nuclear Proliferation Using Laser Isotope Separation - Verification Options, UCRL-JC-145343, October 2001

3. Nucleonics Week, September 13, 2004, South Korea's KAERI optics lab used dye lasers to separate U-235

4. D.W. Suh, Apparatus for the Isotope Separation, KAERI/RR-256/80

5. Chil-Min Kim, Hot Band Effect of the Vibrational Transitions Lines in a Tunable CO2 Laser, New Physics (Korean Physical Society), Vol. 26, p101-6, 1986

6. C.J. Kim, Development of Applied Optical Techniques Using Lasers, KAERI/RR-437/84

7. No Author Listed, Isotope Separation of Deuterium Using Lasers, KAERI/AR-284/84

8. Jang-Soo Shin, Molecular Selective Photodissociation of 32SF6 by Pulsed TEA CO2 Laser, Korean Applied Physics, Vol. 3, p60-6, 1990

9. C.J. Kim, Development of Applied Optical Techniques Using Lasers, KAERI/RR-437/84

10. K. Im, Transverse-Discharge Copper-Vapor Laser, IEEE Journal of Quantum Electronics, Vol. QE-21, p1747-8, 1985

11. N. Sung, Stimulated Emission in Optically Pumped Atomic-Copper Vapor, Optics Letters, Vol. 12, p885-7, 1987

12. Hwan Suh, Tunable Dye Laser Excited by a Pulsed Nitrogen Gas Laser, New Physics (Korean Physical Society), Vol. 18, p8-14, 1978

13. Do-Kyeong Ko, Self-Seeding in a Dual-Cavity-Type T-Sapphire Laser Oscillator, Optics Letters, Vol. 20, p710-2, 1995

14. Jae Ihn Kim, Frequency-Stabilized High-Power Violet Laser Diode with a Ytterbium Hollow-Cathode Lamp, Optics Letters, Vol. 28, p245-7, 2003

15. Dalwoo Kim, Laser Intensity and Polarization Effects on the Multiphoton Ionization of Magnesium Atoms, Journal of the Korean Physical Society, Vol. 23, p191-8, 1990

16. Wang Young Oh, Effect of AC-Stark Shifts on the Selective Resonant Ionizations, New (Korean Physical Society), Vol. 33, p24-30, 1993

17. J.M. Lee, Development of Nuclear Fuel. Development of Laser Spectroscopic Technology in Nuclear Industry, KAERI/RR-1007/91

18. J.M. Lee, Development of Laser Spectroscopic Technology in Nuclear Industry, KAERI/RR-111--92

19. H.K. Cha, Study on Laser Atomic Spectroscopy, KAERI/RR-1166/92

20. J.M. Lee, Magnetic Field Effect on Selective Photoionization, KAERI/TR-367/93

21. S.C. Lee, Optogalvanic Spectroscopy of U, Th, and Rb Using Diode Lasers, Journal of the Korean Chemical Society, Vol. 38, p34, 1994

22. E.C. Jung, Specific Behaviors of Dynamic Optogalvanic Signals of an Argon Hollow Cathode Discharge, Optics Communications, Vol. 161, p149-55, 1999

23. C. Schwab, etal., Laser Techniques Applied to Isotope Separation of Uranium, Progress in Nuclear Energy, Vol. 33, p217-64, 1998

24. Jongmin Lee, Selectivity of Photoion Extraction for Isotopes of Rare-Earth Elements, Laser Physics, Vol. 4, p1132-8, 1994

25. Jong-Hoon Yi, Autoionization States of the Ytterbium Atom by Three-Photon Polarization Spectroscopy, Physical Review A, Vol. 51, p3053-7, 1995

26. Hyunmin Park, Selective Photoionization of the Ytterbium Atom by Coherent Two-Photon Excitation, Physical Review A, Vol. 53, p1751-55, 1996

27. Hyunmin Park, Isotope Separation of Yb-168 Stable Isotope for Low-Energy Gamma-Ray Sources, Undated report

28. Study on Thermodynamic Properties of Metallic Vapor, KAERI-CM-197/97 The Spectroscopy in the Atomic Vapor, KAERI-CM-207/97 The Development of Rydberg Filed Ionization Method and Its Application to the Infinitesimal Quantity Analysis, KAERI-CM-208/97 Physical and Chemical Studies of Atomic Beams, KAERI-CM-213/97

29. Development of Atomic Spectroscopy Technologies - The Characteristics of Laser Beam Propogation in Resonant and Near-Resonant Atomic Media, KAERI-CM-385/98

30. P. Ramakoteswara Rao, Laser Isotope Separation of Uranium, Current Science, Vol. 85, p615-33, 2003

31. Cha Hyeing Gi, Real-Time Feedback Control Method of Dye Laser Wavelengths Using Mass Composition Signal Generated From Laser Isotope Separation Process, KR2003041656, 2003 Lee Jong Hun, Isotope Separation Device of Lanthanum or Actinium by Diode Laser, KR2003051485, 2003 Jeong Do-Young, Method for Isotope Separation

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