

**Laser isotope separation and proliferation risks**

Werner Fuß

## Laser isotope separation and proliferation risks

Werner Fuß<sup>1,2</sup>

### Abstract

There is an ongoing discussion on the proliferation danger of laser enrichment of uranium by the Silex process. Here this risk is compared to that of other processes, in particular centrifuges. The two methods need a similar size of the plant for a comparable production rate (in separative work units per year) and the time and costs for their construction do not differ much. This conclusion from published material does not depend on technical details of Silex. But enough details are known to allow for additional conclusions: Whereas the selectivity (enrichment factor) in the Silex process seems higher, the energy consumption is probably larger. Due to the laser's repetition rate being insufficient for the molecular beam, the method has probably a low depletion factor; this is a serious disadvantage for cascading for high enrichment such as for bomb uranium, although it may be acceptable for low enrichment without cascading for reactor purposes.

---

<sup>1</sup> The author worked until 2008 in the Max-Planck-Institut für Quantenoptik (MPQ, Garching, Germany), until 1993 on infrared multiphoton excitation and molecular laser isotope separation of light elements, temporally cooperating with the company Uranit in development of a 16  $\mu\text{m}$  laser. He thanks former colleagues of MPQ and Uranit for valuable discussions and information. Address: Weidachstr. 12, 85748 Garching, Germany, e-mail: [w.fuss@mpq.mpg.de](mailto:w.fuss@mpq.mpg.de).

<sup>2</sup> The assessments of the author presented here are not necessarily those of the MPQ.

## 1. Introduction

Laser isotope separation (LIS) has potentially a much higher enrichment factor than most of the conventional methods. This seems attractive, if one can thus in a single step enrich  $^{235}\text{U}$  from natural uranium ( $^{235}\text{U}$  content 0.72 percent,  $^{235}\text{U}/^{238}\text{U}$  ratio  $r_0 = 0.0073$ ) to a concentration  $\geq 3$  percent ( $r_r \geq 0.031$ ) that is required for a light-water reactor. The required enrichment is  $r_r/r_0 = 4.3$ . (A desirable depletion of the waste stream will be discussed below.) On the other hand, a smaller number of steps than in conventional methods would also be needed to obtain bomb-grade uranium ( $\approx 90$  percent  $^{235}\text{U}$ ,  $r_b = 9$ , required enrichment factor  $r_b/r_0 = 1230$ ). The smaller number of steps might imply (1) a small plant that could be easy to hide, (2) a faster throughput, so that verification would have to be too frequent, (3) a fast set up, which might be done unnoticed (see e.g. [1]). Such worries were already discussed in the early times (see e.g. [2]), when LIS of uranium was basically a matter of hopes or promises.

Since the 1970s, many variants of LIS of uranium have been investigated in many countries (for an early review, see [3]). With one exception, all have been abandoned in the nineties due to either technical difficulties or because an economic operation seemed hopeless. The exception is SILEX (Separation of Isotopes by Laser Excitation), which was developed in the nineties in Australia (Silex Systems Ltd.) and is here supposed to be similar to a South African process for light elements (Aerodynamic Separation Process, company Klydon). It was further investigated in cooperation with the US Enrichment Corporation (USEC) which, however, in 2003 decided to focus instead to centrifuges [1]. The Australian company then began a cooperation in 2006 with General Electric (USA) and Hitachi (Japan), set up a pilot plant in Wilmington (North Carolina, USA) and is now preparing to build a production plant at the same location with a capacity of 3 – 6 million SWU (separative work units, or kg uranium SW), after it has been approved by the U.S. Nuclear Regulatory Commission (NRC) in September 2012 [4-6]. (For comparison: A 1-GW light-water reactor needs  $\approx 300$  t SW as a first fill and then 100 t SW every year (p. 70 in [7]).) For carrying out the setup and the enrichment, the two companies founded a subsidiary "Global Laser Enrichment" (GLE), with also a minority share from the Canadian Cameco.

The reported high enrichment factor characteristic for laser isotope separation naturally causes apprehensions that the method may simplify the access to atomic bombs. A report on the recent pertinent discussion on Silex can be found in [4].

Little is known on the technology of Silex; more or less educated guesses on the physical basis are presented in section 10. However, independently of technical details, there is information on the expected capacity (separative work per year) and need of area, time and money (section 11),

features that are crucial for judging the risk. For an assessment, it seemed helpful to compare the performance (i.e., capacity, area, time, money) of Silex with that of a more familiar method, uranium enrichment by centrifuges (section 5 and 11). To judge also the danger of possible future developments, it seems helpful to consider precursor or related technologies (molecular LIS (MLIS) by IR multiphoton dissociation (IRMPD) in section 9, gas-dynamic methods in section 6 and 7). One of the main causes of fear is the potentially high enrichment factor of laser methods. However, this feature is by far not a sufficient premise for an easy access to substantial quantities of highly enriched uranium, as demonstrated by the examples of electromagnetic separation (section 4) and atomic-vapor LIS (AVLIS, section 8); of comparable importance is the depletion factor (section 12), a feature that presents great difficulties with present-day molecular laser isotope separation of heavy elements that require a molecular beam (section 9, 10, 12).

In the following we first consider the motivation for laser isotope separation of uranium (section 2), the basics of separation processes in general (section 3) and a comparison of separation factors of processes of interest (section 4). Non-laser methods are briefly explained in section 5-7, where the centrifuges (section 5) are interesting for the risk comparison and the gas-dynamic methods (section 6, 7) may have technical details in common with Silex. Among the laser methods (section 8-10), AVLIS (section 8) is a method with very high separation factor which was nevertheless abandoned, whereas MLIS by IRMPD (section 9) has the laser, the molecule and other details in common with Silex (section 10). Section 11 compares the released data on size, costs, construction time and capacity of a Silex plant with existing centrifuge facilities. For a risk assessment, section 12 points to the importance of depletion, and section 13 presents some suggestions for verification and export control.

## 2. Why LIS?

*Around 1975* there were two motivations:

(1) A growing demand for enriched uranium asked for preparing new separation capacities. Most of the demand in the Western world had been satisfied since after the World War by the diffusion plants in USA. But due to their huge energy consumption, alternatives were looked for. Centrifuges looked promising but were still in their infancy, so that also further possibilities were investigated such as:

- (a) separation nozzle (Karlsruhe/Germany, attempted demonstration plant in Brazil),
- (b) other gas-dynamic methods (UCOR or vortex-tube process, South Africa),
- (c) chemical exchange processes (mainly in France),
- (d) plasma centrifuge, cyclotron resonance,

(e) various laser methods, thought promising because of high (expected) selectivity and because complications were totally underestimated.

(2) A high selectivity of LIS would allow to extract some of the remaining 0.2 – 0.4 percent of  $^{235}\text{U}$  from depleted uranium (if the costs per separative work unit and the depletion turned out to be acceptable).

The methods (a) – (e) have all been abandoned during the 1990s (in favor of centrifuges) except Silex.

*Nowadays* there are still two motivations:

(1) The use of depleted uranium by Silex is under discussion [6,8,9].

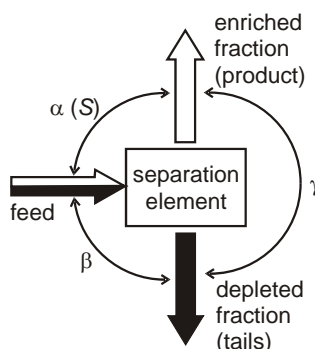
(2) In USA one sees a need for replacement or even expansion of separation capacities, as is indicated by the present imports of enriched uranium from Europe and Russia (47 percent of the US demand [9,10]), by the fact that a new (Urenco) centrifuge plant is working in New Mexico and more such plants are prepared or planned by USEC (new name: Centrus), and because the last diffusion unit was shut down in May 2013 [9]. Furthermore the program "Megatons to Megawatts" ended in 2013, in which Russia sold downblended uranium from dismantled bombs, thus meeting about 37 percent of the US demand for reactor fuel over nearly 20 years [9,10]. (This program is now continued on about half the previous scale: Russia delivers downblended uranium from stocks of bomb uranium [9,10]. There would also be plenty of surplus bomb material in the United States [9].)

### 3. Separation cascades [7]

A separation unit has one feed port with the isotopic mixture (concentration ratio  $r_f$ ), and two outputs, one for the enriched product (ratio  $r_p$ ) and one for the depleted tails (waste) ( $r_t$ ).

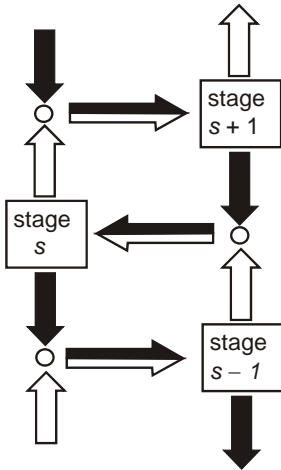
Enrichment, depletion and separation factors ( $\alpha$ ,  $\beta$ ,  $\gamma$ ) are the ratios of these ratios (Figure 1 [7]).

In LIS one also uses the term "selectivity" ( $S$ ): it refers to the physical elementary process for example in one laser pulse, whereas "enrichment factor" refers to material flows in the separation units, so that it would also be influenced by the dead time between laser pulses.



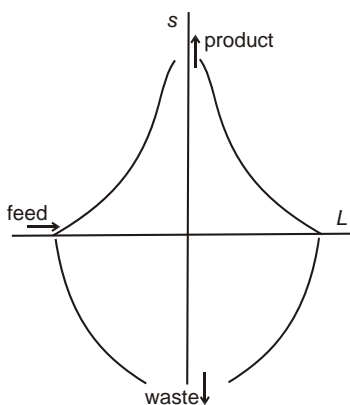
**Figure 1.** Scheme of a separation element with definitions.

Because in conventional separations methods  $\alpha$  or  $\gamma (= \alpha\beta)$  is not sufficient to carry out the desired enrichment in a single step, cascading is necessary. In such a scheme (Figure 2 [7]) one uses the depleted fraction of stage  $s + 1$  together with the enriched fraction of stage  $s - 1$  to feed stage  $s$ . To avoid generation of mixing entropy in the merging flows, one has to choose  $\alpha = \beta$ , if possible. (Such a cascade is called symmetric.) This is difficult in those laser methods which use atomic or molecular beams: If the repetition rate of a pulsed laser is not sufficient (as often), some part of the flow will remain unirradiated, so that depletion is insufficient.



**Figure 2.** Scheme of a (symmetric) separation cascade.

To obtain the necessary material flow, each stage of the cascade usually consists of many parallel separation elements. In a cascade ending at high enrichment of uranium, the first stage must carry  $\approx 140$  times more material flow than the last stage. A similar difference of throughput is also expected in methods with high separation factors, as long as the number of steps is  $> 1$ . The throughput (and hence the number of parallel separation elements in each stage) as a function of the stage number is shown in Figure 3. The shape of the (smoothened) curve is defined recursively (p. 65 of [7]); the enrichment part is initially exponential.



**Figure 3.** Throughput  $L$  ( $\propto$  number of parallel separation elements per stage) as a function of stage number  $s$ .

It is worth noting that using a method with high separation factor implies savings in the number of separation elements only beginning with the second stage. So, in the initial step LIS would require the same throughput as a conventional method. Conversely, one can profit from this feature: using LIS for the first step facilitates its combination with less selective methods in the subsequent steps. This was, for example, done in the  $^{13}\text{C}$  laser enrichment plant in Kaliningrad (operated 1998 – 2005), where  $\text{CO}_2$  laser induced multiphoton dissociation of  $\text{CHClF}_2$  raises the concentration from 1.1 to near 50 percent (separation factor  $\approx 120$ , similar as in the demonstration [11], which also depleted the material by 30 percent); after conversion of the product ( $\text{C}_2\text{F}_4$ ) to  $\text{CO}_2$ , the high enrichment was then accomplished by centrifuges by cooperation partners [12]. The material flow and number of centrifuges is thus reduced by a factor of 50 compared to a centrifuge-only process.

It is worth pointing here to a characteristic difference of LIS from conventional separation methods: Whereas the latter use their expenditure for all gas components, in LIS the expensive laser photons are spent ideally (i.e., if the selectivity is high enough) only for the rare isotope. This advantage of LIS is obviously lost after a certain pre-enrichment (the quantity of the desired isotope remains the same in all steps of the cascade), so that conventional methods may become more competitive for later steps of an enrichment cascade.

The following is also an instructive example: Compared to an initial  $^{235}\text{U}$  abundance of 0.72 percent, starting from 20 percent (as is now available in Iran) saves already half the number of steps required to reach 90 percent. Because the material flow starting from this intermediate enrichment is also reduced by a factor of 30, one would save a factor of  $\approx 60$  in the number of centrifuges, if bomb uranium is desired. Such a facility would be easy to hide, and the technology is available in Iran. The time required for the actual enrichment would be less than four days [13]. It is hence not clear, why the Iranian government boasts about their knowledge of LIS (which has been investigated there since about 1975 – under the Shah –, with an interruption from 1978 to probably the late 1990s).

If one now liked to compare the necessary size of a laser-based versus centrifuge facility to produce bomb uranium, one could try to estimate the number of separation elements and their throughputs, considering also the relative size of centrifuges versus lasers plus accessories. This seems difficult, also in view of the very limited public knowledge on the Silex process. Instead, in section 11 the relative size and performance of the facilities is taken from open information of GLE for Silex and Urenco for centrifuges.

#### 4. Separation factors in some processes

As already said, for reactor-grade uranium one needs an enrichment factor  $\alpha \geq 4.3$  (or a separation factor  $\gamma \geq 18 = 4.3^2$ , if  $\alpha = \beta$  is chosen) and for bombs  $\alpha \geq 1230$ . A cascade of  $s$  stages can obtain  $\alpha_s = \alpha^s$ . The number of stages required is thus proportional to  $\ln(\alpha)$ . Table 1 compares the separation factors achievable by various methods. It should be noted, however, that for a method requiring more than one step the enrichment factor alone is misleading: As explained in section 12, depletion is of comparable importance.

*Table 1.* Uranium separation factors in different enrichment methods.

method	separation factor $\gamma$	source, comments
diffusion	1.003	p. 76 of [7]
centrifuge (countercurrent)	$\geq 1.5$	p. 95 of [7] (see section 5)
mass spectrometer (electromagnetic separation)	10 ... $\geq 10000$	dependent on space charge
separation nozzle; vortex-tube process	1.015; 1.05	p. 104-5 and 120-1 of [7] (see section 6, 7)
laser: AVLIS	$\leq 20000$	[14]; in practice probably $\ll 1000$ (resonant charge exchange and space charge! Section 8)
laser: MLIS (IRMPD)	$\geq 10$	see text (section 9)
laser: MLIS (Silex)	2 ... 20	various press releases (section 10)

For the centrifuge,  $\gamma$  depends on the design (peripheral velocity, length, countercurrent-to-feed ratio) (section 5; survey of different designs and performances in [13]). In the mass spectrometer, the best separations can only be obtained, if the throughput is very limited (e.g. for analytical purposes, or for separation of most elements in milligram quantities). Higher throughputs and densities in the electromagnetic separator cause space charges; for example, in the 1940-s the "Calutron" (of cyclotron type) needed three stages (after a minor pre-enrichment) to obtain bomb-quality uranium and hence had probably  $\alpha \approx 10$ . (The method is not considered anymore a high proliferation risk. This is a good example that a potentially high enrichment factor is not a sufficient criterion.) A similar density problem is faced in atomic-vapor LIS (AVLIS), although milder, because only the minor isotope is ionized by the laser; some sources suggest that it had



only  $\alpha \approx 10$  under production conditions in the demonstration plant of [14]. With the molecular LIS by infrared multiphoton dissociation (IRMD) of  $\text{UF}_6$  the company Uranit (the German branch of Urenco) obtained in 1990 a separation factor "sufficient for reactor uranium in one step", according to group members in their research team. This implies  $\gamma \geq 10$ .

It is worth mentioning that in the commercial production of reactor uranium by centrifuges, the primary design goal is to optimize the separation capacity (in SWU/a), not the separation factor  $\gamma$  or enrichment factor  $\alpha$ . A high  $\alpha$  is even considered a drawback, as it reduces the flexibility to produce the enrichments desired by individual clients.

## 5. Centrifuges

Because Silex will be compared mainly with centrifuges, it seems appropriate to recall here their basic properties.

In a modern centrifuge with a peripheral velocity around  $v = 750$  m/s (limited by material strength [13]), a  $\text{UF}_6$  layer enriched in the heavier isotope is formed at the wall and one with the lighter fraction slightly more inside. On the axis there is practically vacuum, so that the feed-throughs for the gases do not need gaskets. The radial separation factor contains  $v^2$  and  $\Delta m$  (mass difference) in the exponent [7,13]. A countercurrent (Figure 4), induced thermally or mechanically in the centrifuge, substantially improves the separation factor  $\gamma$ : without throughput by a factor containing the length-to-diameter ratio  $Z/d$  in the exponent; with a countercurrent-to-feed ratio of 4 one reaches  $\gamma = 1.74$  in a centrifuge with  $d = 20$  cm and  $Z = 5$  m with the  $v$  above (dimensions of the Urenco centrifuge TC21) according to the calculations of [13]. (The value  $\gamma = 1.5$  [7] in the table above refers to a less advanced centrifuge. Note also the remark at the end of the previous section.) However, the mechanical resonance frequency of a cylinder is the lower, the longer its length. Therefore the longer centrifuges consist of several cylindrical segments connected by bellows. Instead, Russian centrifuges are very short ( $< 1$  m). It is argued with the robust technique and the long lifetime. On the other hand, also the Urenco centrifuges of around 1985 still work continuously without failure since that time.

An important feature of centrifuges is their low energy consumption. This is because separation is carried out near equilibrium. Losses such as in bearings or from stagnation pressure or in electrical lines can be minimized. By contrast, diffusion and various gas-dynamical methods contain irreversible steps; compression work cannot be recovered.

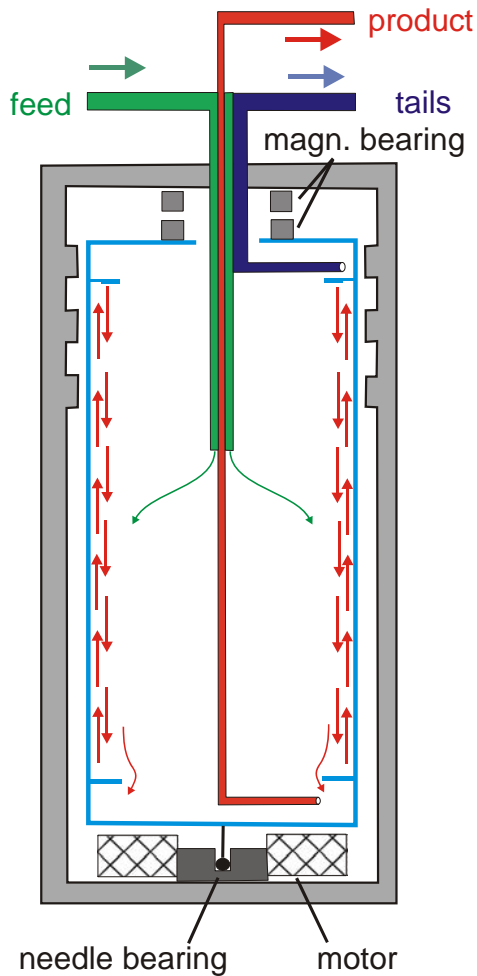


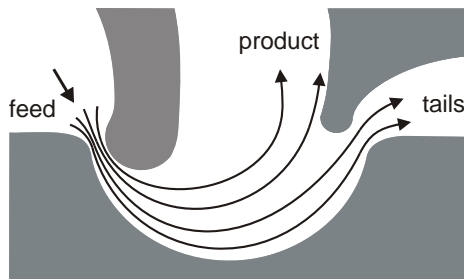
Figure 4. Countercurrent centrifuge.

## 6. Gas-dynamic separation: The separation nozzle

This method is briefly described here, because section 10 suggests that Silex uses a modified separation nozzle. Details can be found in [7].

On expansion of (e.g.)  $\text{UF}_6$  through a nozzle, the lighter isotope diffuses slightly faster in a direction transverse to the molecular beam. To improve the separation factor, E.W. Becker (Karlsruhe) guided the molecular beam along a curved wall to profit from the centrifuge effect and diluted the vapor with a light carrier gas ( $\text{H}_2$  with 4 percent of  $\text{UF}_6$ ) to obtain higher velocities. Using 0.25 bar for the feed and an expansion factor of 2, a separation factor of 1.015 was obtained (1.025 for two consecutive nozzles) [7].

With this input pressure  $p$  the optimum radius of curvature of the nozzle wall was  $R \approx 0.1$  mm [7]. Gas-dynamical scaling laws imply [7] that this radius can be increased, if the pressure is lowered according to  $R \propto p^{-1}$ .



**Figure 5.** Separation nozzle.

Because of too high energy consumption (for the compression work that is not recovered), the method was abandoned in the late 1980s, before a demonstration plant in Brazil went into operation.

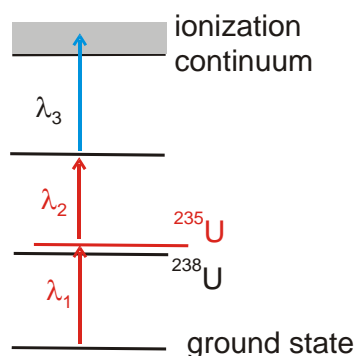
### **7. Gas-dynamic separation: The UCOR (vortex-tube) process**

Also this method may have some gas-dynamic principles in common with the Silex process (section 10) and is therefore briefly mentioned. The public knowledge on this process is very incomplete. The available information is collected in [7]. It was developed in South Africa by UCOR (Uranium Corporation) on the same basic principle as the separation nozzle. But instead of only stimulating a half turn in the curved nozzle, the gas mix ( $H_2$  with 1-2 percent  $UF_6$ ) is blown through a nozzle tangentially to the inner side of a tube wall and subsequently spirals down along this "vortex tube", performing many revolutions. The heavier fraction is enriched near the wall due to the centrifugal forces. As compared to a normal vortex tube according to Ranqué-Hilsch (see Wikipedia), which is used e.g. in workshops for cooling tools, the tube is probably slightly conical and has both fractions exit at the same end of the tube. Separation factors of 1.045 – 1.055 were reported [7]. A special cascading technique ("Helikon") was combined with this method, which allowed to handle the large difference between enrichment and depletion factor.

Also the UCOR (vortex-tube) process has a high energy consumption, even slightly higher than diffusion, due to non-recovered compression work. It was used for enriching to both, reactor-grade and bomb-grade uranium. When the South-African government decided to break out of its isolation, it also joined the Non-Proliferation Treaty in 1991 and abandoned uranium enrichment.

## 8. Laser isotope separation: AVLIS

The atomic-vapor LIS (AVLIS) is related to the electromagnetic isotope separation, with the difference, that only the less abundant isotope is selectively ionized; the positive ions are collected at a cathode. Typically (for variants, see [14,15]), a uranium atomic beam is ionized by three dye-laser wavelengths; two of them ( $\lambda_1$ ,  $\lambda_2$ ) giving rise to the selectivity, because the intermediate resonant level exhibits an isotopic shift, that is due to a volume effect of the nucleus. (The corresponding absorption spectrum to this level is shown in [15].) Due to the ionization method, the space-charge problem is mitigated. Nevertheless, the vapor density must be kept low (order of  $10^{13}$  atoms  $\text{cm}^{-3}$ ) to reduce resonant charge exchange between the isotopes. At small densities, a selectivity of  $>20000$  was demonstrated [14]. One can suppose that, to raise the throughput, one increases the density to a level that still allows for a separation factor of 10-20 and thus an enrichment to reactor grade uranium in a single step. (Actually, the process is full of complications that are not reported here.)



**Figure 6.** Levels involved in AVLIS of uranium: One level with isotopic shift (caused by nuclear volume for orbitals that have a noticeable density there), two selective wavelengths ( $\lambda_1$ ,  $\lambda_2$ ).

A demonstration plant with a planned capacity of 500 t SW/a was set up in Livermore, California. A partial operation succeeded in 1985 [14] (but till the end only for a few days with the planned power). From the photograph in [14] one can estimate that the buildings took an area of  $\approx 1$  ha. The attempt was abandoned in 1999 due to never ending material problems and lack of reliability, mainly caused by the liquid uranium at 2500 K. This happened in spite of the long time and large expenses ( $>2$  billion USD [10]) for the investigations. A French demonstration plant (SILVA), developed and operated over 4 years, has produced at its end (2003) ca. 200 kg of uranium enriched to 2.5 percent [16].

If the method had worked for producing reactor uranium in sizable quantities, it would have been a high risk for proliferation: In an existing plant, one could simply reduce the density and throughput (without public notice) to raise the enrichment factor to a value sufficient to produce bomb-grade uranium in a single step. A low throughput with high enrichment is what is needed

for an atomic bomb, whereas for a reactor a high throughput with low enrichment is necessary. It must be asked, why some atomic powers (USA, Russia, Great Britain, France, China) tried to develop just AVLIS, although they always pointed to the separability of civil and military applications of nuclear fission.

But even after its economic failure, AVLIS must be considered a risk for proliferation: With lower quantities involved, materials and reliability problems might be reduced to a tolerable level. The complexity of the process is fortunately a (limited) barrier.

### 9. Laser isotope separation: MLIS

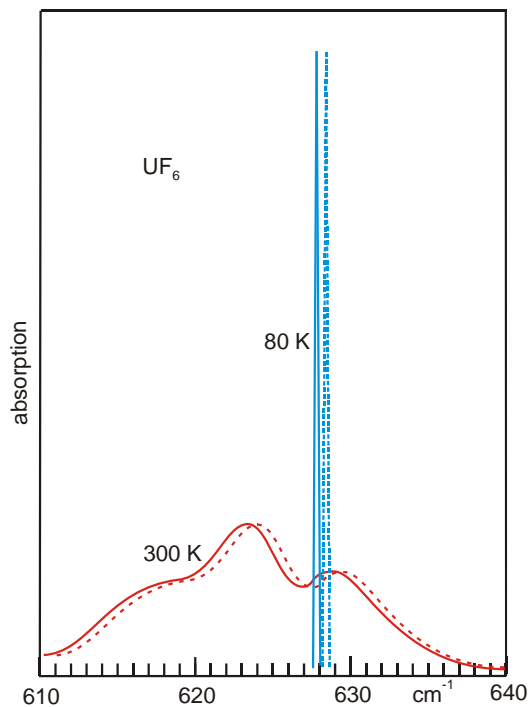
Because Silex is known to use  $\text{UF}_6$  with IR excitation (section 10), molecular LIS techniques should be considered; in particular, MLIS by IRMPD (see below) has the laser and the molecular beam in common with Silex. In the early times till about 1990, mainly two methods were investigated for this molecule:

- (a) Selective vibrational excitation in the infrared ( $\approx 16 \mu\text{m}$ ) and subsequent photodissociation of the vibrationally excited molecules in the long-wavelength wing of a UV band. (The IR excitation broadens and shifts the UV bands.) However, the longest-wavelength band of  $\text{UF}_6$  shows only slow (pre-) dissociation (during which selectivity can be lost by collisions) and has a too broad wing (spectrum in [17,18]), so that the selectivity of IR excitation is not maintained. This method is not promising with  $\text{UF}_6$  [19]. – If one would try two-step UV ionization instead of photodissociation, one would obtain a similarly low selectivity and in addition also suffer from the low densities required (as in AVLIS, section 8).
- (b) Selective vibrational excitation by one to three IR photons (weak lasers fine-tuned to the respective frequencies near  $16 \mu\text{m}$ ) and then multiphoton dissociation (IRMPD) by a stronger laser (pulsed, tens of MW,  $\geq 5 \text{ J/cm}^2$  [19]) at slightly longer wavelength. (The pre-excitation shifts the next absorption steps by anharmonicity. For the mechanism of IRMPD, see [20])

After dissociation, the product  $\text{UF}_5$  is collected as a solid.

The isotope shift ( $0.6 \text{ cm}^{-1}$ ) in the IR spectrum, due to a mass effect, is much smaller than the spectral width at room temperature, that is caused by the rotational structure and by hot bands (absorptions from thermally excited vibrational levels). Therefore one cools the molecule by adiabatic expansion in a molecular beam, using a carrier gas such as nitrogen; for sufficient cooling, one needs an expansion leading to velocities near or above that of sound. (By contrast, cooling is not necessary for light elements due to their larger isotope shifts.) Spectroscopic selectivities around 500 can be obtained already near 80 K (Figure 7, drawn from data of [21], for

a spectrum at lower temperature see [22]). To obtain long absorption path lengths, one uses slit nozzles.



**Figure 7.** Infrared spectra of  $\text{UF}_6$  (solid and broken lines: the heavier and lighter isotope) at room temperature and at 80 K. The spectra at 300 K show P, Q and R branches. For the 80 K spectra only the Q branches are shown; for  $^{235}\text{UF}_6$  it is in the gap between Q and R branch of the heavier isotope. The absorption cross section at the lower temperature ( $\approx 10^{-16} \text{ cm}^2$ ) is higher than shown.

However, the selectivity for the total process (pre-excitation + IRMPD) is much lower, because the wavelength dependence of multiphoton dissociation is broad and is not limited to the pre-excited molecules. Uranit obtained around 1990 a selectivity  $S$  "sufficient for one-step enrichment to reactor grade", i.e.  $S \geq 10$ . While this seems reasonable, the process was abandoned, (1) because there was still too much need of development and (2) because there was no hope of becoming competitive with centrifuges. Problems were the energy consumption for compression to produce the molecular beam (and other losses), the very small cross section for multiphoton absorption that would require absorption lengths of kilometers, the fact that the solid product  $\text{UF}_5$  is abrasive for the optical elements in the chamber, the small repetition rate of the laser that left large parts of the molecular beam unirradiated (details below), and more. The compression energy for producing the molecular beam and the insufficient laser repetition rate will also be problems for Silex (section 10); also the same laser is used.

The  $16 \mu\text{m}$  are produced by shifting a pulsed  $\text{CO}_2$  laser by the rotational Raman effect in para-hydrogen below 1 bar at liquid-nitrogen temperature (under these conditions, only the  $J = 0$  state is populated and the gain is highest) in a multipass cell (Herriott type, focusing, e.g. 25 passes). To

obtain sufficient gain, the pump (CO<sub>2</sub>) laser must have  $\geq 30$  MW (not MW/cm<sup>2</sup>: steeper focusing to obtain higher intensity and gain is compensated by a shorter gain length due to faster divergence) and focusing must be just below the break-down threshold, so that the process is very sensitive to impurities including dust (which can be a problem with gas circulation, that is needed at high repetition rate). Fine tuning is done by tuning a high-pressure ( $\approx 10$  bar) CO<sub>2</sub> laser. Because such lasers have difficulties to reach the power for the Raman threshold, one injects simultaneously a pulse from a line-tunable TE CO<sub>2</sub> laser (which is anyway needed for IRMPD, but will also be necessary for Silex for the Raman process to work). After obtaining a 16 $\mu$ m wavelength, side-band frequencies can be generated by electrooptic amplitude modulation.

A high repetition rate is necessary in order not to leave unirradiated material in the rapidly moving molecular beam in the time between the laser pulses. For a gas velocity of 200 m s<sup>-1</sup> (still subsonic for N<sub>2</sub> as carrier gas) and an irradiated cross section of 1 cm<sup>2</sup>, one would need 20 kHz. Transverse-electric discharge (TE) CO<sub>2</sub> lasers up to 1 kHz and high-pressure CO<sub>2</sub> lasers up to 300 Hz are commercially available from SDI Lasers (part of PaR, Pretoria) (probably similar from Trinita in St. Petersburg and perhaps others), with which Silex Systems Ltd. collaborates. (A physical obstacle for higher repetition rates are the shock waves, perturbing the optical paths. They are produced in the laser gas by the electrical discharge and in the Raman cell by deposition of laser energy.) This is obviously insufficient. (In AVLIS, a copper-vapor laser pumped dye laser reaches 5–10 kHz without problems.) Moreover, at such a repetition rate and also the high pulse powers, the lifetimes of many components is limited.

Using the criterion that bomb uranium needs a high enrichment factor combined with only modest throughput, whereas reactor uranium demands for a high throughput with modest separation factor, MLIS seems much less dangerous for proliferation than AVLIS: The density in the molecular beam (up to 10<sup>15</sup> cm<sup>-3</sup> of UF<sub>6</sub> molecules) can be higher by two orders of magnitude than with the atomic beam, with a similar beam velocity. Furthermore, the selectivity is limited by intrinsic characteristics of the process to much below the spectroscopic value, whereas in AVLIS the very high spectroscopic selectivity can be put into practice [14] by limiting the throughput, which would be hard to notice by external controls.

## 10. MLIS: Silex

Very few technical details are publicly known about the Silex process. Several press releases reported the following information on Silex (see, e.g., [23] for points 1-4). Independent confirmation of the features 1–3 can be found in the report by J.L. Lyman [24], who assessed the method for USEC and for IAEA on a visit to Silex Systems in 2000–2002. Point 5 is also from an independent source [25]. For the separation factor (point 4), there is only the statement of the

company; however, a value in this range seems credible, because only a substantial improvement over centrifuges can justify the process.

- (1) The method uses  $\text{UF}_6$  in a (nonspecified) carrier gas,
- (2) which is cooled (obviously in a molecular beam by adiabatic expansion through a nozzle; static cooling would cause condensation).
- (3) Irradiation of the light isotope is done by one or several wavelengths near  $16\ \mu\text{m}$ , which is produced by a pulsed  $\text{CO}_2$  laser (fine-tunable, high pressure [24]), Raman-shifted in hydrogen.
- (4) The separation factor (different words were used in the media) is between 2 and 20.
- (5) According to IAEA [25], "the feed, product and tails remain  $\text{UF}_6$ ". This point implies that it is not IRMPD, although common features are the laser and the molecular beam.

This information (points 1 to 5) can be considered established, with some uncertainty about the separation factor (point 4). By contrast, the following six paragraphs present only indirect evidence and the conclusion that Silex may be based on a gas-dynamic process. This physics-based guess of technical features is attempted in order to assess the possibility, that future developments will increase the enrichment factors enough to reduce more drastically the number of steps to obtain bomb uranium.

#### *10.1. Silex probably uses selective inhibition of clustering in a molecular beam and perhaps a centrifuge with stationary wall*

There are indications that the process is related to developments in South Africa: Greenpeace quotes in 2005 a contract between Silex Systems "with a South African company to work on its research and development project" (p. 17 of [26]; direct information not available anymore) and quotes (at the same place) a report [27] that says that due to financial cuts of the South African Atomic Energy Corporation "some researchers (seem to) have been approached by Silex Systems to work on a similar project in Australia". Whereas these researchers might mean some laser specialists from SDI Lasers/Pretoria (Silex Systems already before bought  $\text{CO}_2$  lasers from there and is interested in higher repetition rates), there are indications (see below) that also the LIS processes in the two countries are related and that they exchanged expert knowledge in this field. In the 1990s, SDI split into a laser company (SDI Lasers, now part of PaR/Pretoria) and a company (Klydon: klydon.co.za) investigating isotope separation of light elements such as silicon (as also Silex Systems), saying that it would also work with  $\text{UF}_6$ . They call their method Aerodynamic Separation Process (ASP), saying it is based on a centrifuge with stationary wall.

This reminds one of the South African vortex-tube process or the separation nozzle (section 6-7), only combined with a laser. To increase the separation factor in the centrifugal part from  $\approx 1.02$



(separation nozzle with  $\text{UF}_6$  in  $\text{H}_2$ : section 6) to  $\approx 10$  (Silex), it would be desirable to drastically increase the mass difference between the isotopic molecules. In fact, this is possible, (a) if the molecular beam is operated under conditions, where collisions form clusters of the kind  $(\text{UF}_6)_n$  or  $\text{UF}_6 \cdot \text{G}_m$  with small  $n$  and  $m$  and with G being the carrier gas or a component of it, and (b) if a laser with isotopic selectivity either dissociates clusters containing  $^{235}\text{U}$  or inhibits formation of them by selective excitation (heating) of monomeric  $^{235}\text{UF}_6$  still in the collisional region of the molecular beam. (The clusters form by condensation during adiabatic cooling. At a distance of a few slit widths downstream from the nozzle, collisions terminate due to the decreased density and temperature.) The mass difference (on which the separation factor depends exponentially in a centrifuge) between the non-associated lighter isotope and the clustered heavier isotope would increase from 3 to 355 u (dimer) or 203 u (cluster with G = cyclo- $\text{C}_4\text{F}_8$ , a heavy molecule (200 u) not absorbing at 16  $\mu\text{m}$ ), for instance.

Both methods, selective cluster dissociation and selective inhibition of their formation, were demonstrated by the van den Bergh group [28-31] with  $\text{SF}_6$ , although with a straight beam without centrifuge. The separation factors were small ( $\leq 1.6$  for inhibition of clustering with Ar [30]; for IRMPD of  $\text{SF}_6$  it is easy to obtain values  $> 100$  [32]). But they would probably increase on combination with a stationary-wall centrifuge; the beam must then be subsonic, of course. The IR spectra of the clusters are broad ( $\text{SF}_6$  clusters: [28,29,33],  $\text{UF}_6$  clusters: [34,35]) compared to uranium isotopic shifts, and complications arise due to the presence of different clusters [31]. Hence the selective condensation inhibition will be the only possibility.

In fact, this method was also suggested by J.W. Eerkens (see [36]) for  $\text{UF}_6$ , although without a centrifuge and therefore small enrichment factor. Importantly, Eerkens with his company began merger negotiations with (the precursor of) Silex Systems, but stopped them because of funding disagreement according to [36]; it seems evident that there must be some overlap between Eerkens' "repression of condensation" and Silex, with the main difference perhaps the use of a centrifuge with stationary wall in Silex. (In a straight beam as recommended in [36], separation would be based on diffusion, which only depends on the square root of relative mass difference. This is confirmed by the small separation factor in the  $\text{SF}_6$  experiments [30]. Another drawback of the straight beam is the very poor cut: The skimmer usually transmits only of the order of 5 percent of the beam [31]. This further decreases the depletion factor, which is anyway poor due to the insufficient repetition rate of the laser.)

As already said, one has to irradiate the beam in a region where collisions still take place (i.e. during the initial expansion near the narrowest part of the nozzle [31]). On the other hand, one must avoid too many collisions, because they would lead to equilibration between the isotopes

and thus would destroy selectivity. As a consequence, there will be a distribution of species: cluster formation will not be complete, some of the  $^{238}\text{U}$  will be present in monomeric form and conversely some of the lighter isotope (after again cooling down by collisions with the carrier gas) will be found as clusters. This is intrinsic to the process. As a result, *the separation process will – also in future – be far from preserving the selectivity of excitation.* This conclusion is probably independent of the exact embodiment of the process.

Technically, one probably blows the  $\text{UF}_6$  strongly diluted in a carrier gas (light, to obtain high velocities to increase the centrifugal force; perhaps not too light to avoid the necessity of unrealistic repetition rates, and perhaps with a heavy additive such as cyclo- $\text{C}_4\text{F}_8$  for cluster formation) through a slit nozzle tangentially to a curved wall, such as done with the separation nozzle (Figure 5, with irradiation perpendicular to the plane of drawing, after the narrowest part of the nozzle). (The vortex tube is probably out of question, because there would be too many collisions during the many revolutions of the gas, so that the isotopically selective excitation would be lost.) The slit nozzle has to be long enough to provide sufficient path length of absorption of the laser. For such a length the laser must be not strongly focused; a realistic focal spot might be  $1\text{ cm}^2$ . (Larger cross sections seem not possible, because the collisional region cannot be easily expanded.) The radius of curvature of the wall (Figure 5) must therefore be larger by two orders of magnitudes than in the conventional separation nozzle, and the operation pressure smaller by the same factor (section 6).

The conclusion that Silex uses a gas-dynamic process with selective suppression of clustering and use of a centrifuge with stationary wall is based on indirect evidence and may be only a guess. It implies (two paragraphs above) that the separation factor is intrinsically limited. By contrast, the following consideration is also valid for other molecular-beam methods, hence only depends on point 2 above, which was considered established.

#### *10.2. The repetition rate is not sufficient for a depletion required for cascading; conceivable solutions*

As already explained in section 9, with the laser beam diameter of  $\approx 1\text{ cm}$  and the molecular beam velocity of  $200\text{ m s}^{-1}$ , a repetition rate of 20 kHz would be desirable (section 9). (Lyman seems to assume that 5 kHz are acceptable [24], perhaps as a trade-off between desirability and feasibility.) In particular the continuously tunable (high-pressure)  $\text{CO}_2$  laser with its  $\leq 300\text{ Hz}$  is far from such a specification (section 9). As a consequence, much material will pass the nozzle without irradiation, and the depletion will be correspondingly small. (Lyman suggested a degrading of the enrichment factor instead [24].) Also the other laser problems mentioned in section 9 apply here. Pulsed  $\text{CO}_2$  lasers (as in IRMPD) are needed for reaching the threshold in the Raman process.

In view of such difficulties, one can consider conceivable technical solutions (next four paragraphs):

To solve the problem with the insufficient laser repetition rate, one could use a pulsed nozzle for the molecular beam. With piezo-electrically driven valves such nozzles can reach repetition frequencies in the 1 kHz range and hence match a feasible pulse rate of the lasers (see [37] for an early design). One can obtain rather short gas pulses ( $\leq 100 \mu\text{s}$ , corresponding to geometrical lengths of  $\leq 2 \text{ cm}$  at a velocity of  $200 \text{ m s}^{-1}$ ). Usually one can only use the center of such a pulse, because its beginning and end do not have the desired temperature and degree of clustering. With one third of a 2-cm gas pulse irradiated, one could ideally reach a depletion factor  $\beta \leq 1.5 = (2/3)^{-1}$  (corresponding to 1/3 of the  $^{235}\text{UF}_6$  clusters dissociated). Typically  $\beta$  will be worse. An optimistic estimate is  $\beta \leq 1.25$  (1/5 of  $^{235}\text{U}$  recovered), if one takes into account that the skimmers do not discriminate the species with perfect efficiency, that there is a distribution of clusters and that the laser does not saturate the transition in the molecules. The main problem of the pulsed nozzles is, however, their service lifetime. Usually they do not make more than  $10^{10}$  pulses, after which the sealing surfaces are worn out. With 1 kHz, this would last just 4 months. If Silex uses this technique, it had to do some development in this respect, also in matching the existing designs to the necessary large sizes (larger openings, slit nozzles).

Of course, a disadvantage of taking 1 kHz instead of 20 is that the throughput of a single stage would decrease by the same factor.

In principle, one could also multiplex the lasers, that is, take  $\geq 10$  lasers instead of one, and combine their beams by a rotating mirror. Of course, this method would add much more complexity to the process. It may not be realistic for an industrial process.

Another remedy of the small depletion could also be, to send the gas mix repeatedly through the separation zone. If this is done in consecutive separation units, this increases the number of stages. If the same stage is used several times, it increases the throughput time by the same factor. Lyman also notes that "multiple stages will probably be required" [24].

An *advantage* of Silex over IRMPD is the higher absorption cross section (estimated  $10^{-16} \text{ cm}^2$  at 80 K, by more than two orders of magnitude higher than for multiphoton dissociation), implied by the resonant excitation by one photon (perhaps few photons, points 3 and 5 above); this opens the chance to absorb a large part of the laser photons. Another advantage is that the product is chemically unchanged (point 5 above); however, the  $\text{UF}_6$  must be separated from the carrier gas, e.g. by cooling, the energy for which is not negligible.

*To summarize:* Silex irradiates the rare isotope of  $\text{UF}_6$ , adiabatically cooled in a molecular beam with a carrier gas, using pulsed  $\text{CO}_2$  lasers (one at high pressure, continuously tunable), Raman-shifted in cryogenic para-hydrogen. (See section 9 for details on the laser.) Realistic repetition rates imply that a large part of the molecular beam remains unirradiated, so that the depletion will be small. (For the case this problem has been solved, see section 12.) Most likely Silex uses selective inhibition of cluster formation (condensation with a component of the carrier gas). Because condensation leads to a distribution of clusters, because the streams with the different masses are not ideally separated and because the skimmers for separation do not work perfectly, the separation factor is intrinsically limited to much below the spectroscopic value. This conclusion seems fairly certain. Because the separation by the mass-dependent diffusion across a straight molecular beam is insufficient, it is here supposed that Silex uses a beam guided by a curved wall, thus profiting from a centrifugal effect.

### **11. Comparison of Silex and centrifuge plants with respect to size and performance**

The comparison in this section is made on the basis of public information from the involved companies. It does not depend on the technical considerations of section 10.

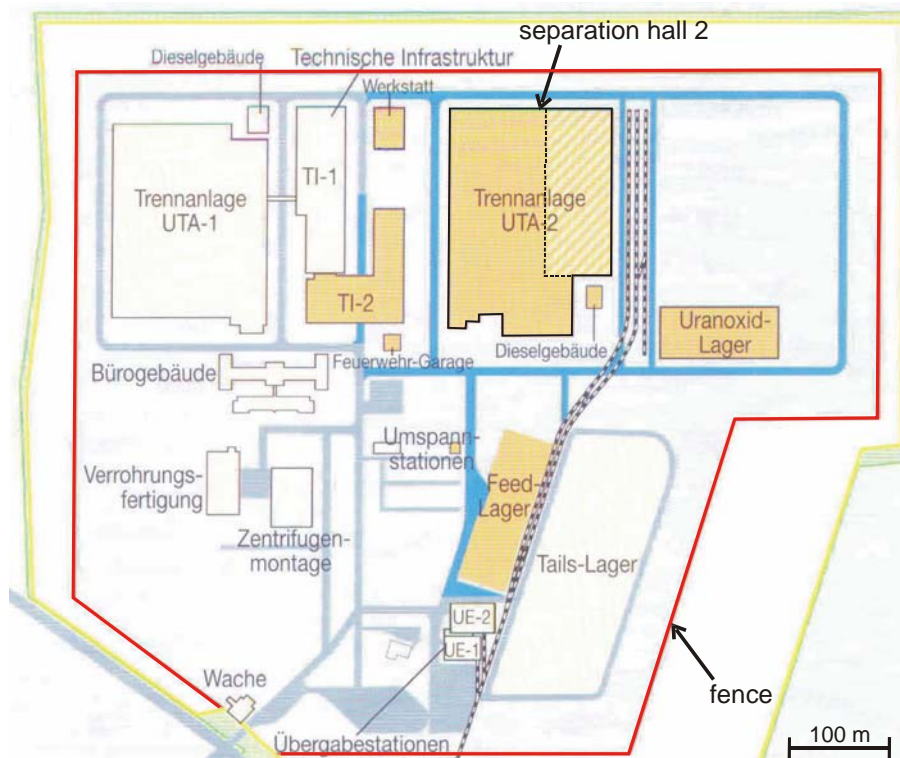
Information on Silex:

- The enrichment plant, planned in Wilmington, will have a capacity of 3500 to 6000 t SW/a [6,38].
- It will take an area of 40 ha [4] with the main building about 5.6 ha [39]. It is said that this is about one quarter of the area required by conventional techniques [40].
- "Low power consumption and capital costs" compared to centrifuges, "enrichment factor 2 to 20 compared to 1.3 with centrifuges" [23]

It is not clear, to which centrifuge facilities these comparisons refer. The standard centrifuge in 1980 in the West had a separation factor of 1.5 [7], and more recent ones 1.74 [13]. (Note also the remark at the end of section 4.) Hence it seems worthwhile to check also the other statements.

A good source for comparison is the Urenco facility in Gronau (Germany), which has been expanded between 2005 and 2012 by an additional capacity of 2700 t SW/a. The areas can be taken from the plan submitted to apply for a permit [41] (Figure 8).

The total facility with a capacity of 4500 t SW/a has about 47 ha (within the fence). The new separation hall (2700 t SW /a) has  $\approx 3.5$  ha; only 2.5 ha were needed, however, with the advanced centrifuge type TC-21. Hence the Silex plants are by no means smaller than a centrifuge facility of comparable capacity. Also the other way around: with a comparable size, the new method is not faster or has a faster throughput (in SWU/a).



**Figure 8.** Ureco uranium separation plant in Gronau, with the new separation hall (UTA-2). The figure is from a permit application of Ureco Deutschland (with kind permission), with enhancements and explanations added. (The actually erected building is slightly modified.) In UTA-2 (3.5 ha), only the darker shaded (2.5 ha) area is filled with centrifuges.

It is also not faster to set up: The Silex plant in Wilmington is expected to produce the first quantities of enriched uranium within one year (still in 2014) and to reach full scale around 2020 [10,16] (or according to [42], beginning of construction in 2014 and full production in 2018). (According to a recent press release (July 2014), GLE now slows the pace, due to decreased enrichment demand on the market [43].) The construction of the Gronau extension began in 2005 and its full operation was planned [44] for 2012 (and also achieved). A similar set-up time is expected for the new French centrifuge plant (Georges Besse 2), which started operation in April 2011 and will reach full capacity (7500 t SW/a) in 2016 [16], or the Ureco centrifuges in New Mexico (5700 t SW/a in 2015, after beginning production in 2011, with costs for the setup of 1.5 billion USD [9,16]).

The Silex plant is expected to cost  $\geq 1$  billion USD [9,23,42]. This seems cheaper per SWU/a (by roughly a factor of 2) than the (smaller) extension built in Gronau with costs of 800 million Eur [41] (but similar to the Ureco plant in New Mexico [9]). But firstly, experience with large projects has shown that the costs must later often be corrected to the higher side. Second, the capital costs do not only depend on the costs for the setup but also on the time, after which it

must be replaced (or after which the repair costs reach the same order of magnitude). It is hardly conceivable that the lasers reach an operation lifetime of the order of 30 years such as centrifuges.

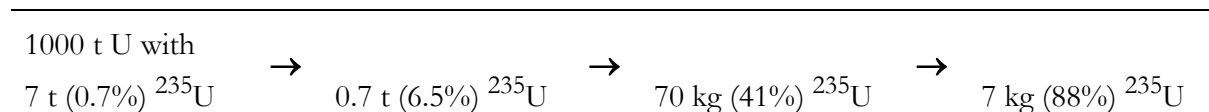
Concerning the power consumption, centrifuges seem to have reached a point, where further reductions make no sense: According to coworkers of Uranit, they consume less than the associated process systems (such as UF<sub>6</sub> handling) and service buildings such as the workshop, laboratories and offices. In view of the pumping for the molecular beams, one can actually expect that Silex consumes more energy. It is also worth noting that marketing for laser methods often calculates with the energy costs (e.g. in [14]). However, laser photons are much more expensive: They are dominated by the costs for the laser compared to the number of photons emitted, before the laser must be replaced; the energy from the power supply of the laser plays practically no role (see e.g. [11,45]).

## 12. Assessment of the danger of proliferation

Besides the higher enrichment factor, the main worries refer to the (claimed) compactness and velocity of the process and short set-up time, also to its attractiveness due to the (claimed) lower capital and operation costs [4]. But according to the preceding section, all these features except the enrichment factor are comparable to centrifuge enrichment. *The claims must hence be interpreted as exaggerations of marketing.*

For obtaining bomb-grade uranium, the high enrichment factor seems most attractive, no doubt, because it would require only few stages. How would a corresponding facility look like? The first stage would enrich, for example, by a factor of ten to a concentration of  $\approx 7$  percent. But the tails would not be much depleted, because the insufficient repetition rate leaves much of the uranium unprocessed. To demonstrate the importance of depletion in cascading, let us assume as an example that one can extract about one tenth of the <sup>235</sup>U. This is much better than imposed by the insufficient repetition rate of the laser ( $\approx 300$  Hz or in future perhaps around 1kHz for the high-pressure CO<sub>2</sub> laser compared to the desirable 20 kHz) with a continuous molecular beam and perhaps by a factor of 2 worse than with a pulsed beam (if one assumes that the problems with pulsed nozzles have been solved, see section 10). In production of reactor uranium, a small depletion may not be a major disadvantage, as long as uranium with natural isotope abundance is cheap. For obtaining higher enrichment, one could now add two similar stages, each by a factor of  $\approx 10$  smaller than the preceding one, due to the smaller throughput. The end product would have a concentration near 88 percent (section 1). However, each step would again leave 90 percent of the available <sup>235</sup>U behind, which now hurts more than in the first step. As the scheme explains, one would also have to start from a correspondingly larger quantity of natural uranium:

5000 tons to produce 35 kg of bomb uranium for one bomb. If this quantity is to be made within one year, it would mean a facility of a comparable size as that planned in Wilmington.



*Scheme.* Effect of three stages with enrichment factor of 10 and depletion factor of 1.1; the latter means that only 10 percent of the <sup>235</sup>U is extracted in each stage.

It would be more rational to continue from the second stage by a method with better depletion factor such as centrifuges, and arrange them as a symmetric cascade (with depletion factor = enrichment factor, section 3), as is done in the case of carbon isotope separation (section 3). Such a combination would not waste so much of the pre-enriched <sup>235</sup>U. In the given example, one needs a number of centrifuges just as usually on beginning the enrichment with reactor uranium (with 6.5 percent in the example). But as the first stage in this example would mean about the same size and expenses with Silex as with centrifuges, one can ask why to use a laser technology at all with all its uncertainties and needs for development.

Suppose there is – or will be developed – a method to improve the poor depletion factor, perhaps by further development of the pulsed nozzles: what would change? The scheme above would then not apply, and the total facility to make bomb uranium could be much smaller. The first stage would have about the same size as with centrifuges (section 11). We can assume that the same statement will apply to the rest of the stages, although their sizes will not matter much, because they are anyway smaller than the first. It will also not make a big difference, whether one uses a pure laser method or a combination with centrifuges or the latter alone. Also the costs and performance will be comparable (section 11). Of course, one can also ask in this case, whether it would be attractive to replace all or part of the centrifuges by a laser method. However, this is a question of assessment, as is shown by the decision of GLE in favor of Silex for reactor uranium. Furthermore, the decisions of proliferators may be determined by other constraints.

In the case of a good depletion factor, a facility to make uranium for just one or a few bombs per year will be quite small, just as with centrifuges: For making 50 kg of 90 percent <sup>235</sup>U from natural uranium, one needs 10–15 t SW. A capacity to make this within a year, is about 300 times smaller than the facility planned in Wilmington (3000–6000 t SW). Such a plant can be easily hidden, as demonstrated by centrifuges in Fordow near Qom (Iran) and in North Korea, which were not detected by technical means but by information transfer.

A proliferation risk assessment ("Kerr report") of Silex was commissioned by General Electric-Hitachi, but not released (2010). Only three proliferation experts had insight. Their statements

and some of the Kerr report are discussed in [4]. The Kerr report said that Silex is no more likely to spread globally than today's centrifuge enrichment approach. This is supported in this and the preceding section by the comparison of size, throughput and costs on the basis of published information from the companies. The comparison even disfavors Silex, if the (probably) poor depletion factor cannot be improved. Also some technical challenges of Silex are illustrated by the example of laser problems in section 9-10. (For other statements of the Kerr report, see [4]. Note also that USEC has abandoned Silex in 2003.) Nevertheless, in spite of challenges and the unfinished development of Silex, it may seem attractive to interested states due to its better enrichment factor than with centrifuges, with the similar size, performance and costs presenting no obstacle.

It would be desirable, if the proliferation risk assessment were not a matter of private initiative but would be commissioned to professionals from IAEA – and/or (in USA) also from NRC or possibly DoE – with full access to information.

One may also ask whether future developments may crucially simplify Silex. In fact, if this process is based on one- or few-photon excitation (as to infer from points 5 and 3 in section 10, which were considered certain), a continuous-wave laser would be more suitable than a pulsed one. If a continuously tunable continuous-wave 16  $\mu\text{m}$  laser can be developed with a power of  $\geq 5$  W (estimated to be needed for saturating the transition in  $\text{UF}_6$ ), one will not suffer anymore from the too low repetition rate or the difficulties with the Raman process. Experimental versions of quantum cascade lasers have already reached around 1 W, though at shorter wavelengths; commercial versions are in the range of 10 mW. It seems possible to develop quantum cascade lasers with the right wavelength and bandwidth and a little more power than at present. A physical barrier for power upscaling is the waste heat that is released in a microscopic volume in this type of laser. Perhaps part of the upscaling can be replaced by assembling many such lasers in an array. It can be supposed that  $>10$  years are needed for such a development; otherwise GLE would not have decided in favor of the Raman-type lasers. An alternative could be free-electron lasers. But it is a question whether to consider them as simplifications, because basically they are accelerators.

*To conclude*, the present form of Silex is not suitable for cascading and high enrichment, due to the low depletion factor, a consequence of the laser repetition rate that is insufficient for the molecular beam. In principle, the problem could be solved by using centrifuges for the second and following stages of the cascade. But then one could ask, why not to use them also for the first stage, with no drawback in performance etc.. This question is justified in the same way even if future research and development can solve the problem with the repetition rate.



### 13. Verification and export controls

As just said, detection of a small plant for making bomb uranium on the basis of Silex (as with centrifuges) is probably not detectable from distance by purely technical means. However, one can verify the activity of a declared facility, if on-site inspections are possible (see [24]): As with other methods, one can check the balance of feed, product and tails uranium or  $\text{UF}_6$ , including the degrees of enrichment and depletion. Environmental samples (dust) containing uranium of nonnatural isotopic abundance are also an indication.

A laser which electrical discharges around 1 kJ in  $\approx 1 \mu\text{s}$  thousand times a second is loud and produces electrical interference in the MHz range. Both, the acoustic and electrical noise may be detected even outside the building, even with some limited shielding. The Raman converter is a hydrogen-filled gas tank, 2-4 m long and  $\approx 0.5$  m in diameter (with optical ports and internal mirrors), cooled with liquid nitrogen. The irradiation vessel must provide an absorption path length for the laser beam of one to several meters with a gas flow transverse to it. The two output streams must have a provision (a cold trap, can be cooled by refrigerator liquid) to separate the  $\text{UF}_6$  from the carrier gas (at the end of the cascade, if there is more than one stage); to provide sufficient residence time (because the separation requires diffusion), the trap will be rather long (or consist of multiple traps), probably larger than the irradiation cell. The carrier gas will be pumped back from there to the input vessel, where added  $\text{UF}_6$  evaporates and is allowed to mix; also this vessel will be large to provide residence time, because diffusion is again necessary, and/or it has a provision for thermally inducing gas convection.

An indication is also the combination of safety measures against laser radiation, high voltage ( $\geq 10$  kV), radioactivity and chemical hazards ( $\text{UF}_6$ , hydrogen-air explosions).

To slow down developments in suspected countries, it may help to control the export of special equipments (in addition to the chemicals needed in the  $\text{UF}_6$  preparation, handling, regeneration etc.). If the lasers are imported as complete units: Pulsed  $\text{CO}_2$  lasers (with pulsed transverse discharge, typically near atmospheric pressure) with repetition rates  $\geq 500$  Hz, high power ( $\geq 1$  J/pulse) and good beam quality are rarely used for other purposes; surface modification or purification are exceptions, where beam quality is usually not needed, however. High-pressure ( $\approx 10$  bar)  $\text{CO}_2$  lasers are practically only used in research. The lasers need fast high-power ( $\sim 50$  MW) high-voltage ( $> 10$  kV) switches (thyratrons) and magnetic compressors for the electric pulses.

A characteristic equipment is optics with a coating for reflection or transmission of  $16 \mu\text{m}$ , some of which must even resist corrosion by  $\text{UF}_6$  and chemicals regenerating it from deposits (i.e., by

highly acidic and oxidizing/fluorinating agents). The Raman cell needs mirrors with high reflection at both, 16 and 10  $\mu\text{m}$ , which must resist cryogenic temperatures. This cell also needs a gas circulator with bearings working at such temperatures (hence no oil) without producing any dust or other contamination.

The Raman cell should be operated with para-hydrogen. Export control of it and catalysts to produce or regenerate it may also be helpful. Its practically only other use is, that liquid hydrogen (which is mainly used in research) evaporates less rapidly, if its ortho-form (with its lowest rotational state  $J=1$  that has more energy) is converted to the para-form (allowing  $J=0$ ) before. On the other hand, such a catalyst is only a minor technological barrier.

If there were any development of an alternative 16  $\mu\text{m}$  laser with high-power continuous-wave radiation (quantum cascade lasers, free-electron lasers, considered in section 12), it would certainly take place in a high-technology country (i.e. in USA specially for use with Silex) and could then also be subjected to export control.

#### 14. Conclusion

Although the marketing of Silex claims that it needs less energy, has a smaller footprint and has lower capital costs than centrifuges (also according to a recent information from the company [6]), a comparison of published plans of the Silex plant with existing centrifuge enrichment facilities shows that the sizes are similar (as also stated by D. Kerr [40], who had access to non-public data) and the capital costs are comparable. The claimed reduction of electrical power demand is not only doubtful in view of the compression work needed for the molecular beam, but also makes no sense in view of the power consumed by centrifuges, that is already below that of accompanying processes ( $\text{UF}_6$  handling etc.) and buildings such as offices, workshops and laboratories. Also the throughput (or time needed for enrichment) is similar for the two methods, and the time to set up an enrichment plant is also comparable.

Although the claimed advantage of Silex related to these features has caused many worries about a proliferation risk, the more important characteristic is the high enrichment factor (2 – 20 as stated by the company). An intermediate value ( $\approx 10$ ) is sufficient to obtain reactor grade uranium in a single step. But for bomb grade, cascading is needed also by Silex. It is here pointed out that for this purpose a good depletion is also necessary. It is doubted that this can be obtained by Silex, because the insufficient laser repetition rate leaves much of the molecular beam unirradiated; this problem might be mitigated (though not fully solved), if there was sufficient progress in development of reliable high-repetition rate pulsed nozzles for molecular beams. (A high-power tunable continuous-wave laser at 16  $\mu\text{m}$ , that would solve such problems, is not in sight for the next 10 years.) A better alternative may then be, to use centrifuges in the second and

following stages. Then, of course, the question is, why not to use centrifuges also for the first step, which would not increase the time and effort and would avoid the complications with lasers and the use of additional high technology. Nevertheless, there is room for personal assessment whether to prefer centrifuges or Silex for high enrichment of uranium.

All these conclusions are only based on publicly available information. They do not depend on the exact physical basis of Silex. Nevertheless, indirect evidence was presented in section 10 that it uses a gas-dynamic process with selective suppression of clustering and use of a centrifuge with stationary wall. The most important inference of this guess was that on the intrinsically limited separation factor (i.e., smaller than the spectroscopic selectivity), also in future developments. But even this expectation does probably not depend on the exact embodiment of the process. Also other suppositions (such as the conceivable development of a continuous-wave 16- $\mu\text{m}$  laser) only concern the future. As to the probably poor depletion factor, two scenarios are presented in section 12 (proliferation danger): (1) If it is in fact so poor, cascading (for high enrichment) makes no sense, (2) If the problem has been solved, a high-enrichment plant would have about the size, production rate etc. as with centrifuges.

To summarize, the proliferation risk of Silex is not higher but also not much lower than for centrifuges. For the latter the risk is well known [46]. Both cannot be reliably detected from distance. Suggestions are made, however, how to safeguard on site a declared or detected Silex facility (as already in [24]), and candidates for export control are listed.

It is also reminded of the proliferation risk of AVLIS: Whereas this technique has been abandoned for industrial production of reactor uranium due to material problems with high-temperature metallic uranium, these difficulties may be much mitigated with smaller quantities and throughput, which would be sufficient for a bomb. At the same time, the smaller throughput would raise the enrichment factor to a value that would be sufficient to obtain bomb-grade uranium in a single step.

## References

- [1] J. Boureston, C.D. Ferguson, Separation anxiety, *Bull. Atomic Scientists* 61 (2005) 14-18.
- [2] A.S. Krass, Laser enrichment of uranium: The proliferation connection, *Science* 196 (1977) 721-731.
- [3] R.V. Ambartzumian, V.S. Letokhov, in C.B. Moore (Ed.), *Chemical and biochemical applications of lasers*. Academic Press, New York, 1977, p. 167-316.
- [4] E.M. Grossman, Closely held report discounts proliferation risk of lasers for making nuclear fuel, *Global Security Newswire* ([gsn.nti.org/gsn](http://gsn.nti.org/gsn)) (24 May 2012) [www.nti.org/gsn/article/closely-held-report-discounts-proliferation-risk-lasers-making-nuclear-fuel/](http://www.nti.org/gsn/article/closely-held-report-discounts-proliferation-risk-lasers-making-nuclear-fuel/).
- [5] E.M. Grossman, Laser enrichment approval stands at Nuclear Agency deadline, *Global Security Newswire* ([gsn.nti.org/gsn](http://gsn.nti.org/gsn)) (17 Jan. 2013) <http://www.nti.org/gsn/issues/2013-01-17/>.
- [6] [silex.com.au/downloads/asxannouncements/uranium-enrichment-update-4-april-2013](http://silex.com.au/downloads/asxannouncements/uranium-enrichment-update-4-april-2013), accessed May 2013.
- [7] W. Ehrfeld, U. Ehrfeld, in *Gmelin-Inst. f. Anorganische Chemie* (Ed.), *Gmelin Handbuch der Anorganischen Chemie*. Springer, Berlin, 1980, p. 57-149.
- [8] OpticsOrg Newsdesk, 'Turning point' for laser uranium enrichment - US Department of Energy and Global Laser Enrichment begin negotiating over Kentucky stockpile, (2013 Dec 3), <http://optics.org/news/4/12/4> accessed April 2014.
- [9] World Nuclear Association, US Nuclear Fuel Cycle, (2015), <http://www.world-nuclear.org/info/Country-Profiles/Countries-T-Z/USA--Nuclear-Fuel-Cycle/>, accessed Feb. 2015.
- [10] J.M. Hylko, SILEX process promises third-generation uranium enrichment technology for U.S., (2013), <http://www.powermag.com/silex-process-promises-third-generation-uranium-enrichment-technology-for-u-s/>, accessed April 2014.
- [11] W. Fuß, J. Göthel, M.M. Ivanenko, W.E. Schmid, P. Hering, K.-L. Kompa, K. Witte, Macroscopic isotope separation of  $^{13}\text{C}$  by a  $\text{CO}_2$  laser, *Isotopenpraxis Environmental Health* 30 (1994) 199-203.
- [12] V.Y. Baranov, A.P. Dyad'kin, D.D. Maluta, V.A. Kuzmenko, S.V. Pigulskiy, V.S. Mezhevov, V.S. Letokhov, V.B. Laptev, E.A. Ryabov, I.V. Yarovoi, V.B. Zarin, A.S. Podoryashy, Production of carbon isotopes by laser separation, *Proc. SPIE* 4165 (2000) 314-323.
- [13] A. Glaser, Characteristics of the gas centrifuge for uranium enrichment and their relevance for nuclear weapon proliferation, *Science and Global Security* 16 (2008) 1-25.
- [14] J.A. Paisner, Atomic vapor laser isotope separation, *Appl. Phys. B* 46 (1988) 253-260.
- [15] C. Schwab, A.J. Damião, C.A.B. Silveira, J.W. Neri, M.G. Destro, N.A.S. Rodrigues, R. Riva, Laser techniques applied to isotope separation of uranium, *Progress in Nuclear Energy* 33 (1998) 217-264.
- [16] World Nuclear Association, Uranium enrichment, (2013), [world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Uranium-Enrichment/#.UZzwNUr-uSo](http://world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Uranium-Enrichment/#.UZzwNUr-uSo), accessed May 2013.
- [17] S. De Silvestri, O. Svelto, F. Zaraga, Laser study of the photophysical properties of  $\text{UF}_6$ , *Appl. Phys. B* 21 (1980) 1-7.
- [18] W.B. Lewis, L.B. Asprey, L.H. Jones, R.S. McDowell, S.W. Rabideau, A.H. Zeltmann, R.T. Paine, Electronic and vibronic states of uranium hexafluoride in the gas and in the solid phase at very low temperatures, *J. Chem. Phys.* 65 (1976) 2707-2714.
- [19] J. Horsley, P. Rabinowitz, A. Stein, D. Cox, R. Brickman, A. Kaldor, Laser chemistry experiments with  $\text{UF}_6$ , *IEEE JQE* 16 (1980) 412 - 419
- [20] W. Fuß, K.-L. Kompa, The importance of spectroscopy for infrared multiphoton excitation, *Progr. Quantum El.* 7 (1981) 117-151.
- [21] R.J. Jensen, C.P. Robinson, Progress in laser enrichment in Los Alamos, Los Alamos National Laboratory, report number LA-UR-76 1652 (1976),
- [22] M. Takami, T. Oyama, T. Watanabe, S. Namba, R. Nakane, Cold jet infrared absorption spectroscopy: The  $\nu_3$  band of  $\text{UF}_6$ , *Jap. J. App. Phys.* 23 (1984) L88-L90.
- [23] [nextbigfuture.com](http://nextbigfuture.com), (2011), [nextbigfuture.com/2011/08/ge-silex-laser-uranium-enrichment.html](http://nextbigfuture.com/2011/08/ge-silex-laser-uranium-enrichment.html), accessed March 2013.
- [24] J.L. Lyman, Enrichment separative capacity for SILEX, Los Alamos, Los Alamos National Laboratory, report number LA-UR-05-3786 (2005), [www.fas.org/sfp/othergov/doe/lanl/docs4/silex.pdf](http://www.fas.org/sfp/othergov/doe/lanl/docs4/silex.pdf).
- [25] D. Rousseau, J. Lepingwell, Isotopic separation by laser based technologies: safeguard related aspects, report number IAEA-CN-184/262 (2010), [http://www.iaea.org/Public/root/SG\\_SYMPOSIUM/Documents/AbstractsRepository/262.pdf](http://www.iaea.org/Public/root/SG_SYMPOSIUM/Documents/AbstractsRepository/262.pdf).

- [26] F. Barnaby, Secrets, lies and uranium enrichment: The classified Silex project at Lucas Heights, [greenpeace.de/fileadmin/gpd/user\\_upload/themen/frieden/silex\\_report.pdf](http://greenpeace.de/fileadmin/gpd/user_upload/themen/frieden/silex_report.pdf), accessed May 2013.
- [27] M. Barletta, C. Bowen, K. Jamison, G. Kampani, Nuclear and missile related trade and developments for selected countries, *The Non-proliferation Review* (November 1997-February 1998) 150, [cns.miis.edu/npr/pdfs/db53.pdf](http://cns.miis.edu/npr/pdfs/db53.pdf)
- [28] J.M. Philippoz, J.M. Zellweger, H. van den Bergh, Infrared vibrational predissociation of van der Waals clusters: Applications to isotope separation, *J. Phys. Chem.* 88 (1984) 3936-3938.
- [29] J.M. Philippoz, J.M. Zellweger, H. van den Bergh, Infrared vibrational predissociation spectroscopy of  $(\text{SF}_6)_m \text{Ar}_n$  clusters: Implications for effective laser isotope separation, *Surface Science* 156 (1985) 701-711.
- [30] J.M. Zellweger, J.M. Philippoz, P. Melinon, R. Monot, H. van den Bergh, Isotopically selective condensation and infrared-laser-assisted gas-dynamic isotope separation, *Phys. Rev. Lett.* 52 (1984) 522-525.
- [31] H. Van den Bergh, Laser assisted aerodynamic isotope separation, *Laser und Optoelektronik* 3 (1985) 263-273.
- [32] W. Fuß, T.P. Cotter, Energy and pressure-dependence of  $\text{CO}_2$ -laser induced dissociation of sulfur-hexafluoride, *Appl. Phys.* 12 (1977) 265-276.
- [33] J. Geraedts, S. Stolte, J. Reuss, Vibrational predissociation of  $\text{SF}_6$  dimers and trimers, *Z. Physik A* 304 (1982) 167-175.
- [34] T.A. Beu, J. Onoe, T. Taketsugu, Calculations of structure and IR spectrum for small  $\text{UF}_6$  clusters, *J. Chem. Phys.* 106 (1997) 5910-5919.
- [35] S. Tanimura, Y. Okada, S. Takeuchi, FTIR spectroscopy of  $\text{UF}_6$  clustering in a supersonic Laval nozzle, *J. Phys. Chem.* 100 (1996) 2842-2848.
- [36] J.W. Eerkens, J. Kim, Isotope separation by selective laser-assisted repression of condensation in supersonic free jets, *AIChE Journal* 2010 (2010) 2331-2337.
- [37] D. Proch, T. Trickl, A high-intensity multi-purpose piezoelectric molecular beam source, *Rev. Sci. Instr.* 60 (1989) 713-716.
- [38] [trustandverify.wordpress.com](http://trustandverify.wordpress.com), (2010), [trustandverify.wordpress.com/2010/08/06/continued-silex-concerns](http://trustandverify.wordpress.com/2010/08/06/continued-silex-concerns), accessed March 2013.
- [39] [nuclear-news.net](http://nuclear-news.net), (2011), [nuclear-news.net/2011/08/22/silex-laser-uranium-enrichment-opening-opportunities-for-nuclear-terrorism/](http://nuclear-news.net/2011/08/22/silex-laser-uranium-enrichment-opening-opportunities-for-nuclear-terrorism/), accessed May 2013.
- [40] S. Weinberger, Laser plant offers cheap way to make nuclear fuel, *Nature* 487 (2012) 16-17, [nature.com/news/laser-plant-offers-cheap-way-to-make-nuclear-fuel-1.10945](http://nature.com/news/laser-plant-offers-cheap-way-to-make-nuclear-fuel-1.10945).
- [41] Urenco Deutschland, Urananreicherungsanlage Gronau: Kurzbeschreibung des Endausbaus und der voraussichtlichen Auswirkungen auf die Umgebung, [umweltfairaendern.de/wp-content/uploads/2013/01/URENCO-Kurzbeschreibung-Endausbau-voraussichtliche-AuswirkungenUmgebungDez2002.pdf](http://umweltfairaendern.de/wp-content/uploads/2013/01/URENCO-Kurzbeschreibung-Endausbau-voraussichtliche-AuswirkungenUmgebungDez2002.pdf), accessed March 2013.
- [42] B. Bellemare, Nuclear Enrichment Facility to be Constructed in Wilmington, N.C., (2012), [utilipoint.com/2012/09/nuclear-enrichment-facility-constructed-in-nc/](http://utilipoint.com/2012/09/nuclear-enrichment-facility-constructed-in-nc/), accessed May 2013.
- [43] M. Peach, GLE consolidates laser uranium separation as market bites, [optics.org](http://optics.org), (2014), <http://optics.org/news/5/7/48>, accessed Jan. 2015.
- [44] Urenco Deutschland, Enriching the future, [www.urengo.com/Download.aspx?fileid=49](http://www.urengo.com/Download.aspx?fileid=49), accessed March 2013.
- [45] W. Fuß, Survey on laser isotope separation, in *International Symposium on Isotope Separation and Chemical Exchange Uranium Enrichment*, Tokyo, 1990, ed. Y. Fujii, T. Ishida, K. Takeuchi, *Bulletin of the Research Laboratory for Nuclear Reactors, Special Issue 1* (1992) 38-48.
- [46] H.G. Wood, G. A. R.S. Kemp, The gas centrifuge and nuclear weapons proliferation, *Physics Today* 61 (Sept. 2008) 40-45.