PHYSICAL ASPECTS OF THE ISOTOPE SEPARATION BY LASER 
INDUCED SELECTIVE IONIZATION, WITH EMPHASIS 
ON MODEL ANALYSIS

BY SOUBBARAMAYER

DÉPARTEMENT DE PHYSICO-CHIMIE, CEN-SACLAY
91191 GIF-SUR-YVETTE CEDEX, FRANCE

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1 - INTRODUCTION

Basic studies on the process of isotope separation by laser-induced selective ionization have started about fifteen years ago. In the present time, the interest in this process is considerably increased since some countries' decision of funding a substantial R & D program with the ultimate goal of a possible industrial production of reactor grade uranium in the late nineties'. Several hundreds of professionals in the world are now exploring the science of the process, the components technology and the optimal way of industrialization. This review paper is restricted to the physical aspects of the process as they can be understood from the published literature.

The author does not pretend to give an exhaustive bibliography. For an overview on the process, the reader is referred to /1 to 11/. A limited number of other references will be quoted as and when they are used.
2 - PRINCIPLE OF THE PROCESS

2.1 - Spectroscopic identification of isotopic shift and hyperfine structure for uranium atoms

Let me first remind the reader of some elementary features of quantum physics, which can be found in classical textbooks.

When the internal energy of an atom changes, owing to emission or absorption of light, it does not do so continuously but in "quantum" steps. A great wealth of experimental material, chiefly derived from spectroscopy shows that an atom cannot exist in states of continuously varying energy but only in different discrete states of energy, symbolized in Fig. 1, and referred to as "discrete energy levels". These levels and the spacing between them are different for the various chemical elements. The change from one level \( E_i \) to another \( E_j \) is associated with the emission (if \( E_i > E_j \)) or absorption (if \( E_i < E_j \)) of light, whose frequency \( \nu \) is determined by the relation

\[
E_i - E_j = h\nu
\]  

(1)

In Eq. (1), \( h \) is a universal constant known as PLANCK's quantum of action whose value is \( 6.6 \times 10^{-27} \) erg/sec. For memory, I recall that the wavelength \( \lambda \) and the frequency \( \nu \) are related to the light velocity \( C \) (\( C = 3 \times 10^{10} \) cm.s\(^{-1} \)) by

\[
\lambda \nu = C
\]
The usual units of \( v \) are hertz (multiple Gigahertz GHz often used in this topic); \( \lambda \) is expressed, according to authors, in centimeter, angstrom or nanometer (1 cm = 10\(^7\) m = 10\(^8\) Å). Some authors scale the frequency \( v = C \lambda^{-1} \) in cm\(^{-1}\), so that 1 cm\(^{-1}\) means 30 GHz.

Fig. 1 - Discrete energy levels for an atom.

Fig. 2 - Transition of electronic orbitals.

The transition of quantum states is also associated with a jump of electron from one orbit to another. In Fig. 2, the outer electron has jumped to a larger orbit, due to an absorption of light \( h\nu \).

The spectrum of emission or absorption lines is identifiable by highly accurate spectroscopic experiments. Upon what depends the atomic spectrum of lines?
Theory and experiments show that the volume of the nucleus and the structure of the atom (i.e. the number of protons and the number of neutrons) have an influence on the wavelengths and the aspect of the lines. In the case of uranium, the atomic spectrum is extraordinarily rich in lines (about 300,000). Since the uranium 235 (92 protons, 143 neutrons) has a nucleus slightly different from the uranium 238 (92 protons, 146 neutrons), a very fine observation reveals a subtle difference between the lines of the two isotopes. We have reproduced in Fig. 3 a result given by a French team, AVRIL et al. /12/. It represents absorption line of uranium atom in the 5915 Å region. Two important properties are to be pointed out:

- There is a distinct shift between the lines of the two isotopes ($\Delta \nu = 7$ to $10$ GHz).

- While the line of $^{238}\text{U}$ is simple, the line of $^{235}\text{U}$ is multi-component: this is termed HYPERFINE STRUCTURE. The hyperfine structure is the result of a different value of the nuclear spin $I$ ($I = 0$ for $^{238}\text{U}$ and $I = 7/2$ for $^{235}\text{U}$). The number of components of the hyperfine structure is $(2I + 1)$, i.e. 8 for $^{235}\text{U}$.

Isotopic shifts and hyperfine structures for uranium atoms have been investigated in many laboratories in the world and catalogues of lines and isotopic shifts have been constituted.
Fig. 3 - Absorption line of uranium atom near the wavelength $5.915 \text{ Å}$.
2.2 - Selective excitation of $^{235}\text{U}$ atoms by the use of appropriate lasers

The isotopic shift identified preceding suggests that if it is possible to irradiate a mixture of $^{235}\text{U}$ and $^{238}\text{U}$ atoms with a laser light of the very precise resonant frequency of $^{235}\text{U}$ atom, those atoms have a considerably greater chance of becoming excited than the $^{238}\text{U}$ atoms. Such a selective excitation is now feasible with the availability of lasers satisfying the following properties:

- **Tunability**, which permits the production of laser radiation of any frequency from the vacuum ultraviolet to the infrared.

- **High intensity**, sufficient to excite a considerable fraction of atoms. The high intensity of laser radiation allows the stimulation of quantum transitions at a rate higher than the rate of all other competing processes of relaxation and excitation energy transfer.

- **Short controlled duration** of pulse, that can be made shorter than the lifetime of excited atomic states. This makes multi-step excitation possible in times shorter than the relaxation time of any intermediate quantum state.

- **Spatial coherency** of radiation which allows irradiation of extensive volumes.

- **Monochromaticity and temporal coherence** which allow extremely high selectivity. It is now currently possible to obtain line-width inferior to 1 GHz in the visible.
2.3 - Extension of selective excitation to selective ionization of $^{235}$U atoms

The selective excitation is not sufficient to separate the isotopes. Excited atoms, if they undergo no additional physical or chemical reaction, will fall down to the ground state by reemitting the absorbed photon or by dissipating the excess energy in a collision. To follow up the separation procedure, it is essential to transform, in a delay much less than radiative relaxation time ($\lesssim 10^{-7}$ s), the excited atoms to a different physical state which should have a life time at least longer than particle collision time ($\lesssim 10^{-6}$ s) and which should be separable from the surrounding unaffected atoms. A practical method is to carry excited atoms to ionization state by continuing to excite in one or two steps as illustrated in Fig. 4 (ionization potential 6.18 eV).

A first laser excites selectively $^{235}$U atoms while the $^{238}$U atoms remain in ground state. A second laser (in the 2 step scheme) carry the excited atoms $^{235}\text{U}^*$ to ionization state, transforming $^{235}\text{U}^*$ to positive ion $^{235}\text{U}^{+}$ (and an electron). The two processes may be summarized by the two reactions:

$$^{235}\text{U}^0 + h\nu_1 \rightarrow ^{235}\text{U}^*$$

$$^{235}\text{U}^* + h\nu_2 \rightarrow ^{235}\text{U}^{+} + e^-$$

During the two irradiations, the $^{238}$U atoms are not sufficiently affected to get ionized.
Fig. 4 - Three steps and four colours quantum scheme using the 0.076 eV or 620 cm$^{-1}$ metastable level. A two steps scheme was also tested in preliminary experiments.

The 3-step scheme works following the same principle but is a more sophisticated (practically more efficient) version of the process.

Once the $^{235}$U atoms are ionized, they can be deflected by electrical (or electric + magnetic) fields and guided up to a collector located out of the $^{238}$U media, while the neutral $^{238}$U atoms are not sensitive to electrical field.
2.4 - *Summary of the principle of separation by the laser process*

A conceptual picture of the process is schematically represented in Fig. 5 and includes four operations:

- Vaporization of metallic uranium
- Selective excitation
- Extension of excitation to ionization
- Extraction of ions from a neutral background.

3 - MAJOR SYSTEMS

The major systems that comprises a practical module in the AVLIS process are schematically shown Fig. 6. The hardware is divided into a separator system and a laser system, which are, to a great degree, mechanically independent. Uranium is vaporized by a high-voltage electron beam from a crucible containing the metal and the atomic vapor so produced expands upwards in vacuum. The direct flow is irradiated transversely by photoionizing lasers. Tunable laser frequencies are generated in a dye laser system driven by a pump laser system. One choice of pump laser may be copper-vapor laser. Both the pump lasers and dye lasers are configured in master-oscillator power-amplifier (MOPA) chains. The $^{235}\text{U}$ photoions are extracted by electric field and collected on product plates. The remaining vapor, depleted in $^{235}\text{U}$, streams through to the tails collector.
Metallic uranium converted into atomic vapour, contains atoms of $^{238}\text{U} (\circ)$ and $^{235}\text{U} (*)$.

A selective irradiation excites the $^{235}\text{U}$ atoms (●).

One or two more irradiations transform excited $^{235}\text{U}$ atoms to ions $^{235}\text{U}^+$ (■).

An electrical field extracts the ions and guides them up to collecting plate.

*Fig. 5 - Principle of isotope separation by the laser process with atomic vapour. Symbols: ○: atome $^{238}\text{U}$; *: atom $^{235}\text{U}$; ●: excited atom $^{235}\text{U}$; ■: ion $^{235}\text{U}^+$. \*
This rather simple conceptual picture of the process is made considerably more complex when one sets about to do the job in practice. We give here below some more comments on each component of a separation module.

3.1 - Source of uranium atomic vapor

Uranium is vaporized by a high energy linear electron gun. This type of gun has been developed in the literature for selective heat treatment of surface (see for instance /13/, /14/ and /15/). In such a system, electrons are emitted from a long ribbon or filament (Fig. 7) cathod, which, together with a proper beam-former and anodes, is immersed in a weak magnetic field that bends and focuses the linear electron beam into the
center of a water cooled copper crucible where molten uranium is contained in its own frozen skull. In such a system the vapor emanates from the hot line source and expands upward.

![Diagram of linear electron gun](image)

**Fig. 7** - Schematic cross section of a linear electron gun.

We shall come back to this topic in section 4. We do want to emphasize many advantages of this source:

1. It avoids the problem of corrosion of crucible (uranium being its own crucible).
2. It yields very high values of density at the impact zone.
The preceding performance allows a good latitude (10 to 100 cm) for the height over the crucible at which laser beams will pass. It consumes much less energy than joule heating for vapor production.

3.2 - Lasers

As far as the scheme of selective excitation-ionization is concerned, an efficient choice, after an analysis of the physics, reveals to be the three-step lasing process as indicated in Fig. 4. Since the ionization potential of uranium is about 6.2 eV (starting from the ground level) we can use for instance approximately three times 2.1 electron-volts photons. These photons correspond to 5.900 Å or red-orange light. Tunable pulsed dye lasers operating with wavelengths in the red-orange portion of the spectrum are currently the most efficient available. To pump these dye lasers, use of flash lamps has been envisaged in the earlier stage of the design but their efficiency is very low (about 0.2 %). At the present time, the most advanced technology for pumping dye lasers is the Copper vapor laser whose efficiency (about 1 %), power (10 to 200 watts) and repetition rate (5 KHz) are high. The principle of operation of a Copper vapor laser is now well known. Copper atoms are excited to a state of energy level 3.8 eV and undergo transition to low level metastable state of energy 1.4 eV. A detailed scheme of energy levels in a copper laser is represented in Fig. 8. In fact there are two transitions, yielding two colours (green and yellow) of wavelengths 510.5 nm and 576.2 nm. The excitation of copper atoms is obtained by
Fig. 8 - Scheme of energy levels in a copper vapor laser. Two transitions are undergone: the green from the excited state $^2P_{3/2}$ to the metastable state $^2D_{5/2}$ and the yellow from the state $^2P_{1/2}$ to the metastable state $^2D_{3/2}$.

collision with electrons produced by a discharge in a hot tube containing a mixture of copper vapor (partial pressure about 0.3 torr) and a rare gas (neon with a partial pressure 30 torr). A picture of a French copper vapor laser manufactured by the company CILAS-Alcatel has been published in /5/. The schematic section of a copper vapor laser is depicted in Fig. 9. The discharge takes place between two molybden electrodes, in a alumina tube of diameter 2 to 8 cm and length about 1 m or a little bit
more. In fact, rather than pure neon, a mixture of neon and molecular hydrogen yields a better performance, as we shall see in section 4.

![Diagram](image)

**Fig. 9** - Schematic longitudinal section of a copper vapor laser.

- **W**: Water jacket
- **SH**: Thermal shield
- **A**: Alumina
- **E**: Electrodes
- **HV**: High voltage
- **F**: Flow inlet and outlet

The copper vapor laser cannot be used alone for the separation process because it is not tunable, but is perfectly suited to pump dye lasers. A dye laser consists of two major modules, a master oscillator and an amplifier. The former provides the temporal, spatial and spectral waveform to drive the excitation process while the latter generates enough power to illuminate thoroughly the uranium vapor stream.
How many colours are needed to raise $^{235}\text{U}$ atoms contained in the beam from the ground state to the ionized state? In the 3-step scheme, the first 2-steps are excitation transitions and the value of their cross-sections are reasonably high

$$\sigma_{\text{excitation}} \approx 10^{-14} \text{ cm}^2$$  \hspace{1cm} (3)

The problem is more delicate with the last step (ionization). Experiments have shown that the ionization cross-section is several order of magnitude smaller than the excitation cross-section

$$\sigma_{\text{ionization}} \approx 10^{-17} \text{ to } 10^{-18} \text{ cm}^2$$

It follows that the performance of the process requires some other ways of reaching the ionization state. Autoionization of excited atoms is one of these ways.

Autoionization states are bound atomic states whose energy levels relative to the outer valence electron lie above the ionization limit. Usually, above the ionization limit, there is no discrete states and the absorption spectrum is continuous. Nevertheless, for complex atoms like uranium, it was observed (Fig. 10), near the ionization limit, discrete states superposed to continuum states. Some of these discrete states, tend spontaneously towards ionization ($\text{U}^+ + \text{e}$) in a very short time ($10^{-11}\text{s}$) and are termed autoionizing states. Cross-section for the transition from an excited state to autoionizing state was measured (this transition is a resonant one) and the value obtained is

$$\sigma_{\text{autoionization}} \approx 10^{-15} \text{ cm}^2$$  \hspace{1cm} (4)
Fig. 10 - An uranium spectrum between the two first ionization limits (according to COSTE et al. / 16 /).

For the process, it is much more efficient that the third step carries the excited atoms to autoionizing states.

A final point to consider is the population distribution in the atoms produced by electron beam gun. At the temperature 2,800 K there is only 47 % of the atoms in the ground state level. Another 28 % are in the first low level excited state or the 620 cm$^{-1}$ state. The remaining 25 % are excited to higher energy levels and in fact some are ionized. As a result of this population distribution, for the process to be efficient, lasers must be included in the system to excite atoms both from the ground state and the 620 cm$^{-1}$ state to some higher excited state. Taking these requirements into account, the term scheme of excitation-ionization is shown in Fig. 4.
Thus, while there are three steps, four colours are used to excite approximately 75% of the desired atoms and bring into the first level. How many of them will reach the ionized state? We will see it in section 4 while analyzing the kinetics of the irradiations.

3.3 - Ion extractors

In the scheme of the module represented in Fig. 6 the vapor emanates from the hot line source and expands upward through a series of plates (Fig. 11) which are aligned parallel to the flow and aimed at the line source. As the vapour flows between the plates the four colour laser beam is passed back and forth through the many channels illuminating the vapor and ion-
zing the $^{235}\text{U}$ atoms. The plates are connected to generators and maintained at specified potentials. The electric field in the interspace of electrodes interacts with the plasma created by laser and draws the ions to the electrodes (which play thus also the role of ion collector). The unionized $^{238}\text{U}$ atoms are not affected by the electric field, pass on and are deposited on a tails collector plate where they are condensed.

There are many phenomena causing decline in separation efficiency. Let us quote two of them.

Uranium atoms leaving the crucible may be partly ionized or become ionized by the electron beam. These are unwanted ions and should be eliminated before reaching the irradiation zone (for instance by an appropriate use of electric or magnetic fields).

Another problem arises from the uranium atoms which possess a transverse velocity component leading to precipitation of unwanted atoms onto the ion collecting plates. A way of suppressing these direct flow and scattering is to put small shields in front of the plates which are perpendicular to the flow and which shadow the product plates, as depicted in Fig. 11.
4 - PHYSICS OF THE AVLIS PROCESS

An intensive study of the process physics is to be carried out by any team involved seriously in the AVLIS program because of the strong interplay between the primary science, the technology and engineering, the performance and the system cost. As there is more physics involved here than one can cover extensively in a single talk. I will merely outline some specific physical aspects of the process.

4.1 - Physics of the vaporizer

This section includes the dynamics of electrons in a magnetic and electric field (useful to scale the electron gun) and fluid flow problems met in the vaporizer.

4.1.1 - Model calculation of a magnetically focused linear electron gun

Consider again the schematic cross-section of the linear electron gun represented in Fig. 7. The gun consists of a hot ribbon (or filament) cathode (which may be heated electrically or by another standard rapidly scanning electron gun), a beam former (the cathode and the beam former are held at the same potential), two anodes and the target (uranium crucible). The target is at 270° from the cathode. A magnetic field (for instance created by appropriate coils) is applied normal to the plane of the figure. The electron gun is long, i.e. its dimensions in the direction perpendicular to the plane of the figure
are large compared to its transverse dimensions. This permits a
two dimensional calculation. An electron emitted from the cathode,
under the combined action of electric and magnetic field, drifts
towards the target as shown in the figure 7.

First, the calculation of the electric field $\mathbf{E}$ in the
plane $(X,Y)$, or of the potential function $V(X,Y)$

$$\mathbf{E} = - \nabla V$$

is performed by solving the Laplace equation, ignoring the exist-
tence of space charges, in two dimensional scheme

$$\Delta V = 0 \quad (5)$$

with prescribed values $V_i$ on the contours (beam former, cathode,
anodes and target). This calculation is usually performed with a
computer program by finite element method.

An alternate method of computing the potential field has
been indicated three years ago in my lectures to TSINGHUA univer-
sity students: the method, developed by my colleague C. CORTET,
is mathematically very interesting and is outlined below.

Consider $n$ conductors $C_1, C_2 \ldots, C_n$ in the plane

![Diagram]

held at specified potentials $V_1, V_2 \ldots, V_n$. The electric field
at any point $M$ is created by charge densities $\mu_i(s)$ distributed
on the frontiers $\Gamma_i$ of the conductors. The potential at a point M is given by

$$V(M) = \frac{n}{2} \sum_{i=1}^{n} \mu_1(s) \log PM \, ds + C \quad (6)$$

ds being the infinitesimal length of arc on $\Gamma_i$. Moreover

$$\sum_{i=1}^{n} \int_{\Gamma_i} \mu_1(s) \, ds = 0 \quad (7)$$

(which means that there is no net charge in the domain). The charge densities $\mu_1(s)$ are unknown and the problem is reduced to determine them, consistent with the data of the values of potentials on the frontiers $\Gamma_i$.

For any point M located on the frontier $\Gamma_i^*$, the equation (6) is transformed into

$$V_i = \frac{n}{2} \int_{\Gamma_j} \mu_j(s) \log PM \, ds + C \quad (8)$$

There are n equations ($i = 1, 2, \ldots, n$) like (8) to determine the n functions $\mu_1(s)$. The constant C will be determined by the equation (7). Equation (8) is an integral equation of the first kind. It is well known in Mathematics that the integral equations of the first kind, in spite of their apparent simplicity, is much more complex to solve (the existence of solution is not always possible) than FREDHOLM's equation. In our particular case, numerical solution of (8) is possible. The method consists in discretizing properly. First, any integral may be discretized linearly

$$\int_{a}^{b} f(x) \, dx \approx \frac{b-a}{k} \sum_{k=1}^{k} f(x_k) \quad (9)$$
Now, choose a test function $T(\sigma)$ defined on the contour $\Gamma_i$ of a conductor $C_i$. Multiplying eq. (8) by $T(\sigma)$ and integrating over $d\sigma$ result in

$$V_i \int_{\Gamma_i} T(\sigma) \, d\sigma - C \int_{\Gamma_i} T(\sigma) \, d\sigma = \int_{\Gamma_i} T(\sigma) \, d\sigma \sum_j \int_{\Gamma_j} \mu_j(s) \log PM \, ds$$

Use the notation $PM = r(P,M) = r(s,\sigma)$. With a slight change, equation (10) may be written as follows:

$$V_i \int_{\Gamma_i} T(\sigma) \, d\sigma - C \int_{\Gamma_i} T(\sigma) \, d\sigma = \sum_j T(s) \log r(s,\sigma) \, d\sigma$$

Discretizing the integral $\int_{\Gamma_j} \mu_j(s) \, ds$, as indicated by (9), and introducing in (11), result in:

$$V_i \int_{\Gamma_i} T(\sigma) \, d\sigma - C \int_{\Gamma_i} T(\sigma) \, d\sigma = \sum_{j,k} H_{jk} \mu_j(s_k) \int_{\Gamma_i} T(\sigma) \log r(s_k,\sigma) \, d\sigma$$

In conclusion, each test function $T$ allows to write one linear equation between the discretized unknown quantities $\mu_j(s_k)$ and the constant $C$. We can obtain as many equations as the total number of unknowns by selecting the same number of linearly independent test functions $T$. A convenient choice may be Legendre polynomials, or the powers of $\sigma$ ($1, \sigma, \sigma^2, \ldots, \sigma^n$), etc...

The Laplace equation code, or the integral equation code, yields the equipotentials in the plane $(X,Y)$. An example of com-
Computation is shown in Fig. 12. The computations have been performed in CEA by J. BILLET by the integral method in the case of a potential on the beam former and the cathode -30 kV, the anodes and the target being held at zero potential.

![Exquipotentials in a linear electron gun. A: anode; C: cathode.](image)

The next step is the following: the potential functions $V(X,Y)$, hence the electrical field $\mathbf{E}(X,Y)$, precedingly calculated, is introduced in the electron equation of motion

$$
\mathbf{m}_e \frac{d\mathbf{v}}{dt} = -e (\mathbf{E} + \mathbf{v} \times \mathbf{B})
$$

(13)

to trace the electron trajectories. An example of such electron
trajectories is shown in Fig. 13. It corresponds to the gun of Fig. 12 and the value of magnetic field

\[ B = 100 \text{ Gauss} \]

A parametric study yields the configuration of an electron gun producing satisfactory focusing on the target. Nevertheless, I want to draw your attention to the assumption (neglecting the space charge effect) underlying the preceding calculations. A model including space charge effect should be used to scale a high power electron gun. Reference / 17 / solves this problem. In France, a code has been also developed jointly by the Laboratoire National SATURNE (CEA) and the Company CISI-Ingenierie. An application of this code (named BACCHUS-F) is presented in / 18 / by Ph. RAVIER and J.P. PENICAUD.
In what follows, we will deal with "fluid flow" type problems. A schematic view of the evaporator is displayed in Fig. 14. An uranium ingot is inserted in a crucible cooled on its skin. An energetic electron beam produced by a linear gun is focussed on the upper face of the ingot and deposits almost instantaneously a power density of several kW.cm$^{-2}$. In fact, the power density is not uniform but decreasing from a high value at

![Schematic diagram of the evaporator.](image-url)
the center of the beam to a low value at the edge of the beam. The deposited energy increases considerably the local temperature and results in a local melting of the metal, creating a hot liquid pool around the electron impact zone. Vapor is emitted from the liquid free surface, expands upwards in the vacuum and condenses on a cold plate. Some other phenomena occurring in the evaporator are listed in Fig. 14. Three problems of the category fluid flow can be identified: the flow of the liquid uranium in the pool, the kinetic boundary layer of vapor surmounting the liquid free surface and finally the expansion of the vapor from the outlet of the kinetic layer to the condensation plate. We survey these three topics successively in the following sections.

4.1.2 - Flow of liquid uranium in the molten cavity

For sake of modelization simplicity, the molten region is assumed to be a rectangular cavity (Fig. 15) with the side and bottom walls at the melting temperature of uranium (1405 K). The heating of the liquid from above by the electron beam is modelized by the data, as boundary condition, of a temperature profile on the top free surface (the temperature is higher at the center of the electron beam than at the edge). The flow is driven by two forces:

- A surface force, called thermocapillarity. Along the free surface, the temperature difference of the liquid between the centre of the beam and the edge causes variation in surface tension and this variation causes a fluid motion called thermocapillary flow or MARANGONI flow /19/.
A volume force, well known in the literature as buoyancy or thermogravitation. The temperature field in the liquid volume causes variation in density, hence variation in gravitational force: the resulting fluid motion is the usual buoyancy flow or thermogravitational flow.

The flow pattern and the temperature field in the cavity are obtained by the solution of a model including Navier-Stokes equations, energy equation, state equation of the liquid and boundary conditions not only on the side and bottom walls but
also on the free surface. The following assumptions are incorporated:

- The flow is stationary and laminar. Marangoni flow in our case, as we shall see later in the results, have a good chance to be turbulent. The laminar assumption is at best, a first approximation, in the absence of adequate data on turbulence of hot liquid uranium.

- The BOUSSINESQ approximation is valid for the state equation. More sophisticated investigation should adopt the state equation experimentally determined by GATHERS and al. /20/ for hot liquid uranium.

- The fluid is incompressible. This needs an explanation. The density of the liquid is constant in all the equations except in the gravity term.

- The free surface of the fluid is flat. This is rather a drastic approximation and the least justified. In fact, the cratering of the free surface can be calculated and the problem taking into account the cratering can be solved numerically. The flat free surface approximation is nevertheless adopted, because it permits an elaborate mathematical study. We will come back farther on the cratering of the free surface.

The model is written in the next page. The coordinate axis are indicated in Fig. 15. Notations and their meaning are listed below.
\( x, y \) Space coordinates
\( u, v \) Velocity components
\( T \) Temperature
\( \rho \) Density, \( \rho_o = 17.9 \text{ g.cm}^{-3} \)
\( p \) Pressure
\( g \) Acceleration due to gravity, \( g = 980 \text{ cm.s}^{-2} \)
\( \beta \) Coefficient of volumetric expansion: For liquid uranium
\( \beta = 0.9.10^{-4} \text{ K}^{-1} \)
\( T_o \) Melting temperature of uranium, \( T_o = 1405 \text{ K} \)
\( \mu \) Dynamic viscosity of liquid uranium = 0.06 \text{ g.cm}^{-1} \text{s}^{-1} \)
\( \nu \) Kinematic viscosity, \( \nu = \mu \rho^{-1} = 0.0035 \text{ cm}^2 \text{s}^{-1} \)
\( \chi \) Thermal diffusivity = 0.14 \text{ cm}^2 \text{s}^{-1} \)
\( \sigma \) Surface tension, \( \sigma \text{ dynes.cm}^{-1} = 1.550 - 0.14 T(\text{K}) \)
\( h \) Height of the cavity
\( L \) Width of the cavity.

The numerical values given above for liquid uranium are drawn from /21/. In the boundary condition on free surface, \( T_m \) is a data and the function \( f(y) \) is given such that

\[
\forall y \in [0, L] \left\{ \begin{array}{l}
\ f(y) \geq 0 \\
\ \text{Max } f = 1
\end{array} \right.
\]
**EQUATIONS**

- **CONTINUITY**:
  \[
  \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0
  \]  
  \[\text{(14)}\]

- **MOMENTUM**:
  \[
  u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} = g \left[ 1 - \beta (T - T_0) \right] - \frac{1}{\rho_0} \frac{\partial p}{\partial x} + v \Delta u
  \]  
  \[\text{(15)}\]

  \[
  u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} = -\frac{1}{\rho_0} \frac{\partial p}{\partial y} + v \Delta v
  \]  
  \[\text{(16)}\]

- **ENERGY**:
  \[
  u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} = \chi \Delta T
  \]  
  \[\text{(17)}\]

  \[
  \Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}
  \]

- **BOUSSINESQ**:
  \[
  \rho = \rho_0 \left[ 1 - 2 \beta (T - T_0) \right]
  \]  
  \[\text{(18)}\]

**BOUNDARY CONDITIONS**

- **On solid boundaries**:
  \[
  x = h \quad Y = 0 \quad Y = L \quad u = 0 \quad v = 0 \quad T = T_s
  \]  
  \[\text{(19)}\]

- **On free surface**:
  \[
  x = 0
  \]
  \[
  u = 0 \quad \mu \frac{\partial v}{\partial x} = -\frac{d\sigma}{dT} \frac{\partial T}{\partial y}
  \]  
  \[\text{(20)}\]

  \[
  T \cdot T_s = (T_m \cdot T_s) f(Y)
  \]

**LIQUID FLOW MODEL**
In order solve the equations (14) to (20), we proceed to two transformations like in /22/:

- Introduction of the vorticity $\omega$ and streamfunction $\psi$

$$
\frac{\partial u}{\partial y} \quad \frac{\partial v}{\partial x} \quad \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2}
$$

- Scaling of variables:

$$
u = U_0 \frac{h}{L} u^* \quad v = U_0 \frac{h}{L} v^*
$$

Characteristic velocity: $U_0$

$$
U_0 = \frac{\alpha}{\mu L} (T_m - T_o) \frac{h}{L} \frac{1}{\mu L}
$$

$$
x = hX \quad y = LY \quad T^* = \frac{T - T_o}{T_m - T_o}
$$

$$
\psi^* = \frac{\psi}{LU_0} \quad \omega^* = \frac{\omega}{U_0}
$$

The model (14) to (20) is transformed into a non-dimensional model written in the next page, where, like in all problems of fluid mechanics, fundamental non-dimensional numbers are in evidence. In our case the domain of interest is

$\varepsilon < \varepsilon_G$ very small ($10^{-4}$ to $10^{-7}$)

$Pr \ll 1$ (Liquid metals) $Pr = 0.02$ for liquid uranium

$A \sim 1$ ($A = 1$ square cavity)
NON-DIMENSIONAL MODEL

\[ U^* \frac{\partial \omega^*}{\partial X} + V^* \frac{\partial \omega^*}{\partial Y} = \varepsilon_0 \frac{\partial T^*}{\partial Y} + \varepsilon_0 \left[ \frac{\partial^2 \omega^*}{\partial Y^2} + \frac{1}{A^2} \frac{\partial^2 \omega^*}{\partial X^2} \right] \]  
\[ (21) \]

\[ U^* \frac{\partial T^*}{\partial X} + V^* \frac{\partial T^*}{\partial Y} = \frac{\varepsilon_0}{Pr} \left[ \frac{\partial^2 T^*}{\partial Y^2} + \frac{1}{A^2} \frac{\partial^2 T^*}{\partial X^2} \right] \]  
\[ (22) \]

\[ U^* = \frac{\partial \Psi^*}{\partial Y} \quad V^* = -\frac{\partial \Psi^*}{\partial X} \quad \omega^* = \left( \frac{\partial^2 \Psi^*}{\partial Y^2} + \frac{1}{A^2} \frac{\partial^2 \Psi^*}{\partial X^2} \right) \]

- BOUNDARY CONDITIONS: \( X, Y \in [0, 1] \)

\[ \begin{cases} 
Y = 0 & Y = 1 & X = 1 \\
U^* = V^* = \Psi^* = T^* = 0 \\
X = 0 & U^* = 0 & T^* = f(Y) & \omega^* = \frac{\partial T^*}{\partial Y} \\
f(Y) \text{ Specified} 
\end{cases} \]  
\[ (23) \]

4 NON-DIMENSIONAL PARAMETERS

<table>
<thead>
<tr>
<th>( \varepsilon_0 )</th>
<th>( \varepsilon_o )</th>
<th>( Pr )</th>
<th>( A )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_0 = \frac{1}{R_s} )</td>
<td>( \varepsilon_o = \frac{Gr}{R_s} )</td>
<td>( Pr = \frac{\nu}{\chi} )</td>
<td>( A = \frac{h}{L} )</td>
</tr>
</tbody>
</table>

\( R_s = \frac{U_L}{\nu} \) Surface Tension Reynolds Number

\( Gr = g \beta (T_m - T_0) L^2/\nu^2 \) Grashof Number

\( Pr \) Prandtl Number \( \rightarrow \) Liquid

\( A \) Aspect ratio \( \rightarrow \) Geometry of the cavity

\( (R_s, Pr, \text{ Marangoni Number}) \)
The system of equations (21) to (24) has been studied numerically in /22/ in the case

\[ \varepsilon_\gamma \gg \varepsilon_G \]  

(25)

The flow is driven dominantly by thermocapilarity. The authors of /22/ have investigated the solutions in the range of parameters

\[ \varepsilon_G = 10^{-5}, \varepsilon_0 = 2.5 \times 10^{-3} \text{ to } 10^{-4} \]  
(corresponding to \( R_\infty = 400 \text{ to } 10,000 \))

\[ Pr = 0.1 \text{ to } 10 \quad A = 0.2 \text{ to } 5 \]

An example of results is reproduced in Fig. 16. It represents the streamline patterns in a square cavity (only the left half of the

![Streamline patterns for various surface tension Reynolds numbers](attachment:image.png)

Fig. 16 - Streamlines for various surface tension Reynolds numbers \( R_\infty \) (from Ref. /22/).
cavity is shown on account of the symmetry) for $R = 400, 2000$ and $10,000$ respectively. When $R_j$ is low one big cell fills the half of the cavity. When $R_j$ increases, a secondary cell begins to form at the bottom left corner of the cavity and occupies more and more volume. For $R = 10000$ the secondary cell becomes comparable (in volume occupied) to the primary cell at the top of the cavity. It should be pointed out that the primary cells correspond to the strongest values of circulation flux.

The preceding range of parameters, as studied in /22/, does not include the operating domain in the AVLIS evaporator. Here are some possible figures for AVLIS: $h = L = 2$ cm.

<table>
<thead>
<tr>
<th>$T_m$ (K)</th>
<th>2800</th>
<th>3000</th>
<th>3200</th>
<th>3500</th>
</tr>
</thead>
<tbody>
<tr>
<td>$U_c$ (cm.s$^{-1}$)</td>
<td>3266</td>
<td>3733</td>
<td>4200</td>
<td>4900</td>
</tr>
<tr>
<td>$R_c$</td>
<td>$1.9.10^6$</td>
<td>$2.1.10^6$</td>
<td>$2.4.10^6$</td>
<td>$2.8.10^6$</td>
</tr>
<tr>
<td>$Gr$</td>
<td>$0.8.10^8$</td>
<td>$0.9.10^8$</td>
<td>$1.10^8$</td>
<td>$1.2.10^8$</td>
</tr>
<tr>
<td>$\varepsilon_c$</td>
<td>$0.51.10^{-6}$</td>
<td>$0.47.10^{-6}$</td>
<td>$0.41.10^{-6}$</td>
<td>$0.35.10^{-6}$</td>
</tr>
<tr>
<td>$\varepsilon_G$</td>
<td>$2.2.10^{-5}$</td>
<td>$2.10^{-5}$</td>
<td>$1.7.10^{-5}$</td>
<td>$1.5.10^{-5}$</td>
</tr>
</tbody>
</table>

The surface tension REYNOLDS number is 100 times bigger than the highest Reynolds number considered in /22/. Moreover, as far as the importance of the buoyancy as compared to thermocapillarity is concerned, the preceding table shows that we need solution of the system (21) to (24) in the case opposite to (25), that is

$$r_c \ll \varepsilon_G \quad (26)$$

Inequality (26) does not mean that, in this case, the capillary effect is negligible and that the solution is the buoyancy flow.
The smallest parameter $\varepsilon_c$ multiplies the highest derivatives in equations (21) and (22) and this is typically a singular perturbation problem. Capillary effect exists but is concentrated in a thin boundary layer near the top free surface. One method of approach of the problem may consist in dividing the flow field into a boundary layer adjacent to the top free surface and an outer area filling almost the whole cavity, as depicted in Fig. 17, and in determining the flow in each of these areas. The two solutions must be matched in order to form a uniform solution in the whole cavity.

Fig. 17 - Capillary boundary layer and deep core in the cavity.
Using the methods of boundary layers, now-well established, we find the following results:

- **Capillary boundary layer**:

  The scaling of the variables is

  \[
  \begin{align*}
  X &= \varepsilon^n \xi \\
  Y &= Y \\
  \Psi' &= \varepsilon^m \tilde{\Psi} \\
  \omega' &= \varepsilon^p \tilde{\omega} \\
  U' &= \varepsilon^p \tilde{U} \\
  V' &= \varepsilon^q \tilde{V} \\
  T' &= T
  \end{align*}
  \]

  The governing equations of the flow in the capillary boundary layer are:

  \[\begin{align*}
  \tilde{U} \frac{\partial \tilde{\omega}}{\partial \xi} + \tilde{V} \frac{\partial \tilde{\omega}}{\partial Y} &= \frac{1}{A^2} \frac{\partial^2 \tilde{\omega}}{\partial \xi^2} \\
  \tilde{\omega} &= -\frac{1}{A^2} \frac{\partial^2 \tilde{\Psi}}{\partial \xi^2} \\
  \tilde{U} &= \frac{\partial \tilde{\Psi}}{\partial Y} \\
  \tilde{V} &= -\frac{\partial \tilde{\Psi}}{\partial \xi} \\
  \tilde{U} \frac{\partial T^*}{\partial \xi} + \tilde{V} \frac{\partial T^*}{\partial Y} &= \frac{1}{Pr} \frac{\partial^2 T^*}{\partial \xi^2} \frac{1}{A^2}
  \end{align*}\]

- **Boundary Conditions**:

  \[\begin{align*}
  \xi &= 0 & \tilde{U} &= 0 & T^* &= f(Y) & \tilde{\omega} &= \frac{\partial T^*}{\partial Y}
  \end{align*}\]

  The matching condition for \( \xi \to \infty \) will be written later.
Deep core

The scaling of the variables is now:

\[ X = \varepsilon \frac{1}{2} X \quad Y = Y \quad \psi^* = \varepsilon^{1/2} \varepsilon \psi \quad \omega^* = \varepsilon^{1/2} \varepsilon \omega \]

\[ U^* = \varepsilon^{1/2} U \quad v^* = \varepsilon^{1/2} v \quad T^* = T \]

The governing equations of the flow in the deep core are:

\[ U \frac{\partial \omega}{\partial X} + V \frac{\partial \omega}{\partial Y} = \frac{\partial T}{\partial Y} \]

\[ U \frac{\partial T}{\partial X} + V \frac{\partial T}{\partial Y} = 0 \]

\[ U = \varepsilon^{1/2} U \quad v = \varepsilon^{1/2} v \quad \omega = - \frac{\partial \psi}{\partial X} - \left( \frac{\partial^2 \psi}{\partial Y^2} + \frac{1}{A^2} \frac{\partial^2 \psi}{\partial X^2} \right) \]

Boundary conditions:

\[ Y = 0 \quad Y = 1 \quad X = 1 \]

\[ U = V = \psi = T = 0 \]

Matching conditions:

\[ \varepsilon^{2/3} U (\zeta = \infty) = \varepsilon^{1/2} U \quad (X = 0) \]

\[ \varepsilon^{1/3} V (\zeta = \infty) = \varepsilon^{1/2} V \quad (X = 0) \]

\[ T^* (\zeta = \infty) = T \quad (X = 0) \]
In the range of parameters defined by (26) and which might concern the AVLIS evaporator, the singular model (21) to (24) is replaced by the regular system of equations (27) to (31). Unfortunately equations (27) to (31) are non linear and a solution does not seem to be available by standard mathematical methods. Further contributions are needed to solve thoroughly this system.

In order to have a rough estimation of the capillary boundary layer, we look for a solution of equations (27) with boundary conditions (28) but with a somewhat arbitrary assumption that the matching takes place along a 'neutral line' on which the fluid velocity is zero

\[ \tilde{U}(\zeta = \infty) = 0 \quad \text{and} \quad \tilde{V}(\zeta = \infty) = 0 \]  

Using the methods developed by L.G. NAPOLITANO /23/, a class of similar solutions can be found for the system of equations (27), (28) and (32) in the particular case when the interface temperature gradient varies as a power of the surface arc length.

First, attention must be drawn to the fact that in equations (27), (28), (32) the velocity field can be decoupled from the temperature field: the stream function is determined by the following self-consistent system:
\[
\frac{\frac{3^2}{3^2}}{\frac{3^2}{3^2}} - \frac{\frac{3^2}{3^2}}{\frac{3^2}{3^2}} = \frac{1}{2} \frac{3^2}{3^2}
\]

\[
\xi = 0 \quad \frac{3^2}{3^2} = 0 \quad - \frac{1}{2} \frac{3^2}{3^2} = f'(Y)
\] (33)

\[
\xi = \infty \quad \frac{3^2}{3^2} = 0 \quad \frac{3^2}{3^2} = 0
\]

Eq. (33) admits similar solutions of the class

\[
\dot{\varphi} = \frac{1}{A} Y^\alpha \varphi(n) \text{ with } n = \xi / \gamma^\beta
\] (34)

if the temperature gradient on the interface varies as

\[
f'(Y) = C Y^{3\alpha - 2}
\] (35)

and if the constants \( \alpha \) and \( \beta \) are related by

\[
\beta = 1 - \alpha
\] (36)

In eq. (35) the constant \( C \) can be determined by one more information on the interface temperature distribution. For instance, if we assume that the interface temperature is maximum for \( Y = 1/2 \), the constant \( C \) is

\[
C = (3\alpha - 1) 2^{3\alpha - 1}
\] (37)

The function \( \varphi(n) \) in eq. (34) is then solution of the differential equation of fourth order:

\[
\dddot{\varphi} = (3\alpha - 2) \varphi' \varphi''
\]

\[
\eta = 0 \quad \varphi' = 0 \quad \varphi'' = - A^4 (3\alpha - 1) 2^{3\alpha - 1}
\] (38)

\[
\eta = \infty \quad \varphi = 0 \quad \varphi' = 0
\]
For \( a \) and \( A \) specified, eqs. (38) can be solved numerically. An example of computation is presented in the following for values of \( a \) and \( A \) arbitrarily chosen

\[
\begin{align*}
a &= 1 \\
A^2 &= 1/2
\end{align*}
\]

The results are presented in Fig. 18. On the left, Fig. 18(a) represents streamlines in the left half cavity (symmetry is assumed relative to \( Y = 1/2 \)). On the right, Fig. 18(b) represents horizontal velocity profiles. The thickness of the capil-

![Diagram of streamlines and horizontal velocity profiles](image-url)
lary boundary layer scales as the surface tension REYNOLDS number $R_o$ to the power ($-1/3$)

\[
\frac{\xi}{h} = \xi_0 (R_o)^{-1/3}
\]  

(39)

where $\xi_0$ is a constant depending on the aspect ratio $A$ and the parameter $a$, characterizing the interface temperature distribution (i.e. the focalization of the electron beam). For $a = 1$, $A^2 = 0.5$ we have $\xi_0 = 2$.

The maximum value of the horizontal velocity is at the coordinates $(\xi = 0, Y = 1/2)$ and scales as the surface tension REYNOLDS number $R_o$ to the power (2/3)

\[
V_{\text{max}} = \frac{\nu}{L} R_o^{2/3} \varphi_o'
\]

(40)

where $\varphi_o'$ is a constant depending on $A$ and $a$. For $a = 1$ and $A^2 = 0.5$ we have $\varphi_o' = 1$.

As an illustration consider the case $h = 1 \text{ cm}$, $L = 1.4 \text{ cm}$, $a = 1$ we have plotted in Fig. 19 the maximum horizontal velocity on the interface and the thickness of the capillary layer versus the maximum interface temperature. The capillary layer is very thin: the thickness is a few percent of the pool depth. The maximum horizontal velocity is around $30 \text{ cm.s}^{-1}$. The REYNOLDS number of the flow in the capillary layer is about $10^4$. These results suggest that the flow in the capillary layer might become turbulent.
Fig. 19 - Maximum horizontal velocity on the interface and thickness of the capillary layer.

4.1.3 - Kinetic vapor layer above the interface

This section is due to a joint work by F. BOUJOT, N. PETOUX and SOUBBARAMAYER. A communication is presented to this workshop by one of the authors. The methods and the main results are summarized below.

Starting from the melt surface, the vapor is set in motion of expansion upwards in the vacuum. Within a very thin layer over the melt, the vapor flow undergoes a drastic "reorganization". The investigation of this layer (called kinetic or
Knudsen layer) is usually carried out in the literature by the kinetic theory of evaporation and condensation. For our problem, we will use a simplified model called the "two-surface problem" and depicted in Fig. 20:

![Diagram of the two-surface problem](image)

The vapour is emitted from one plate at temperature $T_a$ and condenses on the other plate at temperature $T_b$ ($T_b < T_a$). $P_{a,b}, \rho_{a,b}$ denote pressure, density of the saturated vapour at temperature $T_{a,b}$. $I_a$ is the mean free path in the reference state $(T_a, P_a)$ and $L$ is the inter-plate distance. The Knudsen number $K_n = I_a/L$ is small.

The behaviour of the monoatomic gas is described by the BGK equation in the one-dimensional case. On each plate, the gas molecules leaving the condensed phase have the Maxwellian distribution corresponding to the saturated gas at the surface temperature.
In the case of small deviation from saturated equilibrium state and small KNUDSEN number, the stationary BGK equation can be linearized and solved analytically / 27 / . The uranium vapour pressure law is written : \( P_{\text{sat}} = a \exp \left( -\frac{C}{T} \right) \), \( a = 0.833 \times 10^{12} \) CGS, \( C = 6.033 \times 10^{4} \) K, then even for a small \( T_{a} - T_{b} , P_{a} - P_{b} \) may be large. Therefore the linear approximation is irrelevant for our problem and we study numerically the fully non-linear BGK equation.

The one-dimensional BGK equation is written :

\[
\frac{\partial f}{\partial t} + \frac{\partial f}{\partial x} = A_{\text{col}} \cdot \varepsilon (f_{e} - f) \tag{41}
\]

with :

\[
f_{e} = \frac{\rho}{(2\pi RT)^{3/2}} \exp \left[ -\frac{(\xi_{1} - u_{1})^{2} + \xi_{2}^{2} + \xi_{3}^{2}}{2RT} \right],
\]

\[
\rho = \int f \, d\xi_{1} \, d\xi_{2} \, d\xi_{3} , \quad u_{1} = \frac{1}{\rho} \int \xi_{1} \, f \, d\xi_{1} \, d\xi_{2} \, d\xi_{3} ,
\]

\[
T = \frac{1}{3R_{e}} \int \left[ (\xi_{1} - u_{1})^{2} + \xi_{2}^{2} + \xi_{3}^{2} \right] f \, d\xi_{1} \, d\xi_{2} \, d\xi_{3} ,
\]

where \( f \) denotes the velocity distribution function, \( \xi_{1} \) - the molecular velocity, \( \rho \) - the density, \( u_{1} \) - the velocity and \( T \) - the temperature. \( A_{\text{col}} \cdot \rho \) is the collision frequency and we take \( R = \text{Boltzmann constant} / \text{molecular mass} \).

We put : \( \xi_{1}^{2} = \xi_{1}^{2} + \xi_{2}^{2} + \xi_{3}^{2} \).

We have the following boundary conditions :
\[ f = \frac{\rho_c}{(2\pi RT_a)^{3/2}} \exp\left(-\frac{\xi_1^2}{2RT_a}\right), \text{ for } \xi_1 > 0 \text{ in } X = 0 \]  
\[ f = \frac{\rho_b}{(2\pi RT_b)^{3/2}} \exp\left(-\frac{\xi_1^2}{2RT_b}\right), \text{ for } \xi_1 < 0 \text{ in } X = L \]

We choose the following initial conditions:

\[ f = \frac{\rho_a}{(2\pi RT_a)^{3/2}} \exp\left(-\frac{\xi_1^2}{2RT_a}\right), \text{ for } \xi_1 > 0 \]  
\[ f = \frac{\rho_b}{(2\pi RT_b)^{3/2}} \exp\left(-\frac{\xi_1^2}{2RT_b}\right), \text{ for } \xi_1 < 0. \]

We transform equations (41), (42) and (43) by CHU / 28 / averaging method. The method consists in multiplying eq. (41) successively by 1 and \( \xi_2^2 + \xi_3^2 \) and then integrating the two equations over the whole space of \( \xi_2 \) and \( \xi_3 \). Instead of one unknown function \( f \) we have now two functions \( g \) and \( h \) but two variables were eliminated. We obtain the following system:

- Fundamental equations:

\[ \frac{\partial}{\partial t} \left( \begin{array}{c} g \\ h \end{array} \right) + \frac{\partial}{\partial x} \left( \begin{array}{c} g \\ h \end{array} \right) = \frac{2}{\sqrt{\pi}} \left( \begin{array}{c} g_e - g \\ h_e - h \end{array} \right) \]  
\[ \left( \begin{array}{c} g_e \\ h_e \end{array} \right) = \frac{1}{\sqrt{\pi}} \frac{1}{\bar{\tau}^{1/2}} \left( \begin{array}{c} 1 \\ \bar{\tau} \end{array} \right) \exp \left[ - (\xi - \bar{u}_1)^2 \frac{1}{\bar{\tau}^2} \right] \]

\[ \bar{\tau} = \int g \, d\xi, \quad \bar{u}_1 = \frac{1}{\bar{\tau}} \int \xi \, g \, d\xi \]
\[ \bar{T} = \frac{2}{3} \bar{\tau}^{-1} \left[ \int (\xi - \bar{u}_1)^2 \, g \, d\xi + \int h \, d\xi \right] \]
- Boundary conditions:

\[
\begin{align*}
\begin{pmatrix} g \\ h \end{pmatrix} &= \frac{1}{\sqrt{\pi}} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \exp(-\zeta^2), \quad \text{for } \zeta > 0 \text{ in } \bar{x} = 0 \\
\begin{pmatrix} g \\ h \end{pmatrix} &= \frac{1}{\sqrt{\pi}} \frac{T_b^{-1/2}}{\bar{T}_b} \begin{pmatrix} 1 \\ T_b \end{pmatrix} \exp\left[-\zeta^2 \frac{T_b^{-1}}{T_b}\right], \\
& \quad \text{for } \zeta < 0 \text{ in } \bar{x} = \bar{L}.
\end{align*}
\]

- Initial conditions (\(t = 0\))

\[
\begin{align*}
\begin{pmatrix} g \\ h \end{pmatrix} &= \frac{1}{\sqrt{\pi}} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \exp(-\zeta^2), \quad \text{for } \zeta > 0, \\
\begin{pmatrix} g \\ h \end{pmatrix} &= \frac{1}{\sqrt{\pi}} \frac{\bar{T}_b}{\bar{T}_b^{1/2}} \begin{pmatrix} 1 \\ T_b \end{pmatrix} \exp\left[-\zeta^2 \frac{T_b^{-1}}{T_b}\right], \quad \text{for } \zeta < 0.
\end{align*}
\]

- Notations:

\[
g = \left(2RT_a\right)^{1/2} c_a^{-1} \int \int f \, d\xi_2 \, d\xi_3,
\]

\[
h = \left(2RT_a\right)^{-1/2} c_a^{-1} \int \int \left(\xi_2^2 + \xi_3^2\right) f \, d\xi_2 \, d\xi_3,
\]

\[
\bar{t} = \left(2RT_a\right)^{1/2} l_a^{-1} \, t, \quad \bar{x} = l_a^{-1} \, x, \quad \zeta = \left(2RT_a\right)^{-1/2} \, \xi_1,
\]

\[
\bar{\rho} = c_a^{-1} \, c, \quad \bar{T} = T_a^{-1} \, T, \quad \bar{u}_1 = \left(2RT_a\right)^{-1/2} \, u_1,
\]

\[
\bar{T}_b = c_a^{-1} \, c_b, \quad \bar{T}_b = T_a^{-1} \, T_b \text{ and } \bar{L} = l_a^{-1} \, L \text{ are given constants}.
\]
The integro-differential system (44) to (46) has been solved numerically by the finite difference method /29/. We present here the results of computations for one case, chosen as an example of illustration:

\[ \bar{L} = 100 \quad \bar{T}_b = 0.6 \quad \bar{v}_b = 17 \times 10^{-6} \]

The results are presented in Figures 21, 22, 23 and 24. In Fig. 21 we have plotted the spatial profiles of the temperature at respectively the time

\[ \bar{\epsilon} = 0, 50, 100, 250, 400 \text{ and } 1000. \]

Computations have been carried out up to \( \bar{\epsilon} = 2000 \) but the results do not change for \( \bar{\epsilon} > 1000 \) : so the profile at \( \bar{\epsilon} = 1000 \) is the steady state. This profile points out that the temperature undergoes a discontinuity at the crossing of the interface. When there is a net mass transport across the interface, the vapor is not in thermodynamic equilibrium with the liquid in contact: The temperatures of the vapor and liquid are different and the pressure of the vapor is different from the saturation pressure. The steady state profile shows also a very strong decrease from the hot plate along a short distance (about 20 mean free paths): This is the kinetic or KNUDSEN layer. Then the profile is very slowly decreasing on a long distance and ends by a strong variation near the cold plate. Similar results on the pressure, density and velocity are depicted respectively in Figures 22, 23 and 24.
From the preceding investigation of the KNUDSEN layer we retain two important results:

- The vapor close to the melt surface presents a deviation from the thermoaynamic equilibrium and our computations yield the maximum amplitude of this deviation:

\[
\varepsilon_T = 0.16 \quad \varepsilon_P = 0.46 \quad \varepsilon_\theta = 0.36 \quad (47)
\]

The normalized velocity of the vapor close to the interface is

\[
\bar{u}_1 = 0.35 \quad (48)
\]

The dimensional mass flux \( m \) of the vapor is

\[
m = 0.83 \, c_a \left( \frac{R}{2 \, \pi \, T_a} \right)^{1/2} \quad (48)
\]

the coefficient 0.83 showing that 17% of the atoms leaving the melt are backscattered.

All these results are in agreement with those found in /30/.

- The thickness of the KNUDSEN layer is about 20 mean free paths. Within this layer the vapor flow undergoes a drastic reorganization. In particular the velocity distribution function evolves from a single sided distribution into one with an isotropic random-velocity component superimposed on a bulk drift velocity. Moreover, the vapor undergoes an expansion including a cooling and an acceleration. At the outlet of the kinetic layer the normalized macroscopic characteristics of the vapor are

\[
\bar{T} = \frac{2}{3} \quad \bar{c} = \frac{1}{3} \quad \bar{P} = \frac{2}{9} \quad \text{Mach} = 1 \quad (50)
\]
The results (50) will enter as boundary conditions in the model of the supersonic vapor beam which starts from the outlet of KNUDSEN layer and flows up to the condensation surface.

Equations (47) to (49) have an important application to the derivation of the interface curvature. The momentum conservation law across the interface, projected on the normal (Fig. 25), yields the YOUNG-LAPLACE equation augmented with vapor thrust term:

\[
\frac{c}{R} = P_{\text{liq}} - P_{\text{vap}} + \frac{\gamma_{\text{vap}}}{\gamma_{\text{liq}}} \left( u_{\text{vap}} - u_{\text{liq}} \right) - \gamma_{\text{liq}} gh \cos \phi \tag{51}
\]

\[m (\text{normal}) \]

\[\phi\]

\[\text{Vapor - Interface} \]

\[\text{Liquid} \]

\[\ell (\text{tangent}) \]

**Fig. 25 - General shape of the interface.**

In eq. (51) the meaning of the terms is as follows:

- \(1/R\) curvature of the interface
- \(\gamma\) surface tension
- \(P_{\text{liq}}\) liquid pressure = saturation pressure \(P_a\) at the temperature \(T_a\) of the liquid (Fig. 20)
\[ P_{\text{vap}} \] vapor pressure close to the interface. \( (P_{\text{liq}} - P_{\text{vap}}) = 0.46 \text{ Pa} \) according to (47)

\[ \mu_{\text{vap}} u_{\text{vap}} \] mass flux \( m \) of the vapor, eq. (49)

\[ u_{\text{vap}} \] vapor velocity = 0.35 \( (2RT_a)^{1/2} \), eq. (48)

\[ u_{\text{liq}} \] normal velocity of the liquid \( u_{\text{liq}} \ll u_{\text{vap}} \)

Eq. (51) is a second order non linear differential equation determining the shape of the interface and can be integrated numerically. As illustration, a computation has been carried out and the results are represented on Fig. 26 where the shapes of the interface are drawn for three profiles of the temperature on the interface. For low values of maximum temperature, the interface is very near to a flat profile, but for higher values of heating, the cratering of the interface becomes significantly deep: the flow in the cavity should be investigated by taking into account the curved boundary.

We conclude by summarizing in Fig. 27 the results of the two preceding sections on the structure of the flow. Within the liquid pool, the flow is splitted into capillary cells and gravitational cells. MARANGONI cells are located in a very thin layer below the interface while the buoyancy flow covers the deeper part of the cavity. Two important phenomena occur at the interface crossing: the cratering and the deviation from thermodynamic saturation equilibrium for the vapor. A very thin KNUDSEN layer surmounts the interface: inside this layer the vapor flow undergoes an intense reorganization with significant change in macroscopic properties.
Fig. 26 - Shape of the interface for three profiles of surface temperature.
Though restricted to a thin layer, the MARANGONI flow will play an important role in the efficiency decline of evaporation as we deal with surface evaporation. One method of improving the vaporization efficiency might be the use of ZERO MARANGONI alloy /31/. Such alloys are studied in the published literature for many chemical elements but I have not found any reference for uranium.

4.1.4 - Expansion of the vapor beam

In the schema of the evaporator depicted in Fig. 14, the laser irradiation zone is to be located at a distance of several
tens of centimeters over the melt surface. We need to know, in the irradiation zone, the vapor characteristics: density, axial and transverse velocities and temperature.

The simplest approach consists in investigating a two-dimensional beam in the plane (x,y) emanating from a line source MOM' (Fig. 28) and expanding upwards into the vacuum. E. KLEIN and Y. STEPHAN /32/ have published the bidimensional version of a Monte Carlo program that they have worked out by using the direct simulation method developed by G.A. BIRD /33/ which describes molecular or transition flows in a way consistent with BOLTZMANN equation. The brief description of the model and the numerical examples presented in this section are due to the courtesy of E. KLEIN.

Fig. 28 - 2D beam emanating from a line source.
The Monte Carlo-Bird method simulates the gas by several tens of thousands of molecules (say 20 to 100 thousands). The velocity components and position coordinates of the simulated molecules are stored in the computer and modified with time as the molecules are concurrently followed through representative collisions and boundary interactions in simulated physical space. The flow chart of Fig. 29 is applicable to the computer programs for almost all applications.

Advantage may be taken of flow symmetries in order to reduce the number of position coordinates which need to be stored for each molecule. Even when the flow is one or two-dimensional however the three velocity components in a Cartesian reference system are stored for each molecule, the reason being that collisions are always calculated as three-dimensional phenomena.

The simulated region of physical space is divided into a network of cells, which consist of small regions with specified boundaries. The dimensions $\Delta r$ of the cell must be such that the change in flow properties across each cell is small, that is:

$$\Delta r \ll \lambda \quad \text{(condition 1)}$$

Time is advanced in discrete steps of magnitude $\Delta t_m$, such that $\Delta t_m$ is small compared with the mean collision time per molecule $\Delta t_p$:

$$\Delta t_m \ll \Delta t_p \quad \text{(condition 2)}$$
Fig. 29 - Schematic flow chart for applications of the direct simulation Monte Carlo method.

$t$ is the current time, $n_s$ is the sample index, $n_{\text{max}}$ is the total number of sampling and $T(n_s) - T(n_s - 1)$ is the duration of the sampling $n_s$. 
A third condition must also be respected when choosing
the typical cell dimension $\Delta r$ and the time interval $\Delta t_m$. Their
magnitudes must be such that their ratio is not large in compa-
rison with the speed $C_s$ of propagation of acoustic disturbances,
that is:

$$\frac{\Delta r}{\Delta t_m} \lesssim C_s \quad \text{(condition 3)}$$

The reason for this condition is that it prevents a
possible spurious signal from being propagated with speed $\frac{\Delta r}{\Delta t_m}$. However this requires an improbable sequence of collisions
affecting molecules near the boundaries of successive cells, and
it has been found that this ratio must be several times the
speed of sound before the results are distorted. So condition 3
is less stringent than conditions 1 and 2.

The discrete representations of both time and physical
space are necessary to get information on the macroscopic pro-
perties of the flow, which are actually defined as averages
over the microscopic quantities within each cell. However in
the direct simulation Monte Carlo method the discrete represen-
tation of the velocity space is not required.

Condition 2 allows the molecular motion and the inter-
molecular collisions to be uncoupled over the small time interval
$\Delta t_m$ by repetition of the following procedure:

1) All the molecules are moved through distances appro-
priate to their velocity components and $\Delta t_m$:

$$\vec{r}_{n+1} = \vec{r}_n + \vec{v}_n \Delta t_m$$
Appropriate action is taken if the molecule crosses boundaries representing solid surfaces, lines or surfaces of symmetry, or the outer boundary of the flow. New molecules are generated at boundaries across which there is an inward flux.

2) A representative set of collisions appropriate to $\Delta t_m$ is computed among the molecules in each cell. The intermolecular collisions are assumed to be binary, instantaneous and are treated consistently with collision dynamics. The choice of the colliding molecules in a cell does not depend on their relative positions, as permitted by condition 1. The precollision velocity components of the colliding molecules are replaced by the post-collision values. Thus a collision is simply equivalent to a "jump" in the velocity space, while the positions of the colliding molecules remain unchanged.

The mean collision time in a cell ($\Delta t_c$) depends on the shape of the interaction potential and on the local density. Each cell is provided by a time counter which is advanced at each collision by an amount equal to $\Delta t_c$. This incrementation ensures that the appropriate number of collisions is treated over the time interval $\Delta t_m$. The repetition of this step-by-step time process may lead to a steady state. A sampling time can then be defined, during which the microscopic quantities are stored in order to calculate the macroscopic properties of the flow. Let $N$ be the number of molecules which have been in a cell during the sampling time interval, and $(u,v,w)$ the components of the molecular velocity $\vec{c}$. The flow field properties are calculated from the sums of $N$, $u$, $v$, $u^2$, $v^2$ and $w^2$. For example the
velocities in the x and y directions are simply \( \overline{u} = \frac{\xi}{N} u \) and \( \overline{v} = \frac{\xi}{N} v \) respectively. The temperature based on the u components of velocity is given as \( T_\infty \left( \frac{\overline{u}^2}{N} - \overline{u}^2 \right) = (\frac{\xi}{N} u)^2 - (\overline{u})^2 \).

In what follows, we present and comment the results of some illustrative computations. The first computations are pertinent to the simple case of free molecular flow. The results being symmetric about the vertical axis, only the left quarter plane \( y=0 \) is represented in the figures 30 and 31. In the case of Fig. 30 the length of the line source is \( MM' = 2 \text{ cm} \). The density of the vapor is \( \rho_0 \) (flat profile) at the source. The contour plots of the density \( \rho/\rho_0 \) are represented in the domain \( 0 < x < 2.5 \text{ cm} \) and \( 0 < y < 10 \text{ cm} \). The boundary \( x = 2.5 \text{ cm} \) is supposed to be absorbing. Note that at a height of 10 cm over the line source, the density has decreased by a factor 25.

**Fig. 31** : The configuration is different. The beam is collimated by two plane channels located at \( x = 1 \text{ cm} \) (the distance between the channels is equal to the length of the line source). The height of the channels is 6 cm. The contour plots of the density \( \rho/\rho_0 \) are represented in the domain \( 0 \leq x \leq 1 \text{ cm} \) and \( 0 \leq y \leq 10 \text{ cm} \). The boundary \( x = 1 \text{ cm} \) is absorbing in the portion \( 6 \leq y \leq 10 \text{ cm} \) and diffuse reflecting in the solid part \( 0 \leq y \leq 6 \text{ cm} \). The contour plots are more flat than in Fig. 30.

A second type of computations, including the collision effects, is carried out with the following data at the root of the beam (i.e. at the exit of the kinetic layer) :
Fig. 30 - Contour plots of density in a free molecular 2D-expansion of a beam emanating from a line source.
Fig. 31 - Contour plots of density in free molecular 2D-expansion of a beam emanating from a line source and collimated by plane channels.
- Length of the line source \( MM' = 0.5 \text{ cm} \)
- Temperature (assumed to be uniform) \( 2960 \text{ K} \)
- Density \( 2.9 \times 10^{15} \text{ at.cm}^{-3} \)
- Mach number \( 1 \)

The velocity corresponding to Mach 1 is \( 410 \text{ m.s}^{-1} \). The mean free path at the exit of kinetic layer is \( 0.063 \text{ cm} \). This value is small compared to dimensions of the evaporator and, consequently, the effects of collisions are important. The gas flow is simulated with 50,000 particles. The results of computations are represented in Fig. 32. The angular distribution of the velocity at the emission is plotted in Fig. 32(a). The decay of the average temperature is represented in Fig. 32(b). In a distance of 7 cm the temperature has decreased from 2960 K to 650 K. The density decay along the axis of symmetry is plotted in Fig. 32(c): within a distance of 7 cm the density has decreased by a factor 20. Finally the acceleration of the vapor is depicted in Fig. 32(d) where we have plotted the vertical velocity on the axis of symmetry versus the vertical distance. An asymptotic value of \( 680 \text{ m.s}^{-1} \) is reached within a short distance from the melt (approximately 6 to 7 cm). On the same figure is also plotted the vapor velocity when the collision effects are deleted. It can be clearly seen that the collisions contribute significantly to the acceleration.

In conclusion of the section 4.1 one question should be raised: what is the accuracy of the model analysis of the vaporizer in comparison of the experimental results? Very few
Fig. 32 - 2D expansion of a beam: computations by taking into account the collisions.

(a) Angular distribution of the velocity.
(b) Decay of the average temperature versus the vertical distance.
(c) Density decay along the vertical axis.
(d) Vertical velocity versus the vertical distance.
informations are published in the literature about this matter, but they nevertheless permit to formulate three observations:

a) The evaporation efficiency is low: only a few percentage of the power of the impinging electrons is used to produce the vapor. The rest is lost by many mechanisms: backscattering of electrons, thermal radiation, heat conduction in the crucible and heat loss by convective cells in the liquid pool. The latter phenomena include thermocapillary and buoyancy flows, but the possible flow instability and turbulence in MARANGONI motion, which are not included in the analysis, will enhance the heat loss and deteriorate the evaporation efficiency.

b) The vapor velocity in the beam, far from the melt is reported in the literature to be about 1.000 m.s\(^{-1}\), while the beam computation presented in the section 4.1.4 yields a much lower value of 680 m.s\(^{-1}\). Two explanations are given in the literature for this over acceleration of the vapor:

- Heating of the vapor in the kinetic layer, by collision with the energetic electrons.
- Collisions of metastable uranium atoms with ground state atoms with transfer of energy, during the expansion of the beam.

The discussion of these statements implicates a more detailed investigation of the kinetic layer and the beam respectively.
c) The proportion of the metastable atoms in the irradiation area is lower than the thermal population expected from the BOLTZMANN law. This might be a consequence of desexcitation of metastable atoms by collisions mentioned previously, or due to the fact that the vapor in the beam is not in thermodynamic equilibrium.

To my knowledge, these three discrepancies are still open to further investigations.

Finally, I want also to point out that a very important aspect of the vaporizer is not included in this section: the collection of the depleted stream.

4.2 - Physics of the laser system

As specified in section 3, the laser system comprises two different types of lasers: copper vapor lasers (CVL) and dye lasers, the former serving to pump the latter. The physical analysis is usually devoted to the kinetics of the CVL, the light-vapor interaction and the laser beam propagation, with the ultimate objective of getting information on the scaling laws of the CVL, the photoionization fraction of uranium and the photoionization performance of the whole module. This section is divided into two subsections.

4.2.1 - Kinetics of copper vapor lasers

The understanding of the physics involved in a copper vapor laser is progressing since twenty years both through
theoretical and experimental contributions. The main physical phenomena involved in one period of the CVL (we mean by one period the inverse of the repetition rate: for instance for a 5 KHz CVL, the period is 200 microseconds) may be classified in two phases: the discharge phase and the afterglow phase. The duration of the discharge phase is very short (a few hundreds of nanoseconds) while the afterglow covers the quasitotality of the period. We give below a brief description of each of the two phases. The description concerns the stationary state, reached by the CVL after many pulses and in which the final and initial states of one period are equal: such an initial state is composed of not only atoms of copper and buffer gas but also of a residual ternary plasma (electrons, copper ions and rare gas ions).

- **Discharge phase**

  . The electric current impulsed by the switch on of the thyratron penetrates in the medium and deposits power density (this deposition is spatially inhomogenous).

  . This power density generates an additional ionization and heats the electrons up to a few electron-volts.

  . The energetic electrons collide with copper atoms and raise them to higher excited levels, particularly to \(^2\)P and \(^2\)D levels.

  . If the \(^2\)P level atoms are sufficiently enough relatively to \(^2\)D level atoms, the population inversion occurs and there is emission of green and yellow lights.
-. After glow phase

This phase is characterized by three important phenomena, due to collisions particle-particle and particle-wall:

- Cooling of electrons and buffer gas.
- Relaxation of copper metastable levels.
- Recombination of electrons and ions.

Both the two phases have an acute influence on the lasing characteristics of a C.V.L.

The various phenomena inventoried precedingly are investigated in the literature both by model analysis and experiments. We outline below each of the two approaches.

The model analysis, which was developed and published by KUSHNER and WARNER /34 to 37/, takes into account the time and radial dependence of the kinetic, thermodynamic and electric variables. First, the authors define the species whose population evolution has to be calculated. Figure 33 shows the energy levels of copper and neon. Seven levels for the copper atom and four levels for the neon are used. The collisions driving the kinetics of the population are listed in Fig. 34. Data of cross sections for all the types of collisions are drawn from the literature. As an illustration, the copper-electron impact excitation rates are represented in Fig. 35. These rates are calculated by averaging the cross sections over a Maxwellian electron distribution with temperature $T_e$. 
Fig. 33 - Atomic species included in the model and energy level diagrams. (Note the change in vertical scales).

The equations forming the basis of the model describe the penetration of the electric field, the evolution of the temperatures (of electrons and of the gas) the particle kinetics and the photon kinetics. The system is completed by appropriate boundary conditions (the self consistency is maintained by including a detailed electric discharge circuit).
collisions

1-electron-atom.

- Excitation
- Ionization
- Elastic
- Superelastic
- Radiative Recombination
- Collisonal Recombination

\[
\begin{align*}
\text{e} + \text{M} &\rightarrow \text{e} + \text{M'} + \text{e} - \Delta \varepsilon \\
\text{e} + \text{M} &\rightarrow \text{e} + \text{M'} + \text{e} + \text{e} - \Delta \varepsilon \\
\text{e} + \text{M} &\rightarrow \text{e} + \text{M} - \Delta \varepsilon \\
\text{e} + \text{M'} &\rightarrow \text{M} + \text{e} + \Delta \varepsilon \\
e + \text{M} + \text{e} &\rightarrow \text{M}^{**} \\
e + \text{e} + \text{M} + \text{M} &\rightarrow \text{e} + \Delta \varepsilon
\end{align*}
\]

2-atom-atom.

- Penning ionization
- Charge exchange
- Deactivation

\[
\begin{align*}
\text{Ne}^m + \text{Cu}^+ &\rightarrow \text{Cu}^+ + \text{Ne} + \text{e} + \Delta \varepsilon \\
\text{Ne}^+ + \text{Cu}^+ &\rightarrow \text{Cu}^+ + \text{Ne} \\
\text{M}' + \text{M} + \text{M} &\rightarrow \text{M}
\end{align*}
\]

3-particle-wall.

Deactivation

\[
\begin{align*}
\text{M}' &\rightarrow \text{M} \\
\text{M} &\rightarrow \text{M'} + \text{e} + \Delta \varepsilon
\end{align*}
\]

Recombination

\[
\begin{align*}
\text{M'} &\rightarrow \text{M} + \text{e} + \Delta \varepsilon
\end{align*}
\]

Fig. 34 - Three classes of collisions involved in the population kinetics of a CVL.
Fig. 35 - Copper electron impact excitation rates used, by M.J. Kushner and B.E. Warner.

Penetration of the electric field

We begin with Maxwell's electromagnetic equations:

\[ \nabla \times \vec{E} + \frac{\partial \vec{B}}{\partial t} = 0 \]

\[ \nabla \times \vec{B} = \frac{1}{c^2} \frac{\partial \vec{E}}{\partial t} + 4\pi \vec{J} \]

\[ \vec{J} = \sigma \vec{E} \]
By taking the curl of the first equation and substituting into the second equation, we obtain a diffusion like equation for the longitudinal electric field

\[
\frac{\partial^2}{\partial x^2} E - 4 \frac{\partial}{\partial t} \left( \zeta E \right) = 0
\]  

(52)

where \( \zeta \) is the electrical conductivity of the plasma. In obtaining eq. (52), we assumed the electrical neutrality (\( \nabla \cdot \mathbf{E} = 0 \)) and deleted the displacement current term \( \frac{1}{c^2} \frac{\partial \mathbf{E}}{\partial t} \).

-. Electron temperature equation

This equation follows:

\[
\frac{3}{2} \frac{1}{\tau} \left( \frac{3}{2} n_e k T_e \right) = \frac{1}{2} E^2 - \sum_{ij} r_{ij} N_i n_e \varepsilon_{ij} + \sum_{ij} r_{ij} N_i n_e \varepsilon_{ij} + \sum_i N_m N_i r_{p_{ij}} - \frac{3}{2} n_e k \frac{2m_i}{M_i}
\]

\[
\times (T_e - T_g) \frac{1}{m} + \frac{1}{e} (u_e \frac{3}{2} n_e k T_e)
\]

\[
\times (T_e - T_g) \frac{1}{m} + \frac{1}{e} (u_e \frac{3}{2} n_e k T_e)
\]

(53)

The RHS of eq. (53) includes seven terms. The first term is the Joule heating. The second term is for energy loss as a result of the Jth inelastic collision with species \( i \). The third term is for increase in energy due to superelastic collisions. The fourth term accounts for the energy gained as a result of Penning ionization. The fifth term represents the energy exchange with gas atoms and ions due to elastic collisions. The sixth term is for the change in electron temperature as a
result of convection. Electron transport in the radial direction is assumed to be limited by ambipolar diffusion, so the convective velocity \( u_e \) is given by

\[
\begin{align*}
  u_e &= - \frac{kT_G}{n_e} \left[ \sum_i \frac{\nabla n_i}{m_i \nu_i} + \sum_i \frac{n_i}{m_i \nu_i} \cdot \frac{T_e}{T_G} \frac{\nabla n_e}{n_e} \right] \\
\end{align*}
\]  

(54)

where the sums are over all the ionic species. Finally, in eq. (53) the seventh term is the change in electron temperature resulting from conduction.

---

### Gas temperature equation

The gas (neon + copper vapor) temperature equations is given by

\[
\frac{\partial}{\partial t} \left( \frac{3}{2} k \frac{N T_G}{T_e} \right) = \sum_i \frac{3}{2} n_e \frac{\nabla}{M_j} (T_e - T_G) \cdot \nabla + \nabla \cdot \nabla (T_G, \nabla T_G) 
\]  

(55)

The first terms of the RHS of eq. (55) is a heating term due to collisions with electrons. The second term is for conduction, which is the only heat-loss process for the gas.

---

### Charged particle density equations

The governing equation for the electron density is

\[
\frac{\partial n_e}{\partial t} = \sum_i \Gamma_i n_e N_i - \sum_i \Gamma_i n_e (r + n_e r_{cr}) \\
+ \sum_i \Gamma_m N_i r_p - \nabla \cdot (u_e n_e) 
\]  

(56)
In eq. (56) the terms are for ionization of species $i$ with rate $r_i^I$, radiative recombination with ion $i$ with rate $r_r$, collisional radiative recombination with ion $i$ with rate $r_{crr}$, Penning ionization of species $i$ with rate $r_p$, and convective transport of electrons with velocity $u_e$ given by eq. (54).

For the ions, an equation analogous to eq. (56) is valid with a different expression for the convective transport velocity $u_i$:

$$u_i = -\frac{kT_G}{m_1} \frac{\nabla n_i}{n_i} - \frac{kT_e}{m_1} \frac{\nabla n_e}{n_e}$$

Neutral particle density equations

The local density of excited species is determined primarily by the local electron-atom collision rates. This is because diffusion is very slow at these relatively high pressure discharges, e.g., the diffusion loss rate for neutral species is about 50/s, so during one discharge cycle (200 μs at 5 KHz) the loss of excited-state atoms by diffusion accounts for less than 1% of the total change in density. The time rate of change in density of neutral species $i$ is given by

$$\frac{\partial N_i}{\partial t} = \sum_{j} (r_{ej}^{ji} N_j - r_{ej}^{ij} N_i) n_e + \sum_{j,k} r_{H}^{kj} N_j N_k N_i - \sum_{j,k} r_{H}^{kji} N_j N_k$$

In eq. (58) $r_{ej}^{ji}$ is the rate for electron-impact excitation between species $j$ and $i$. The analogous rate for excitation between species $j$ and $i$ by collision with heavy body $k$ is $r_{H}^{kji}$.
Photons kinetics

There are two optical equations corresponding respectively to the green and the yellow transitions. The time rate of change in cavity intensity is

$$\frac{dI}{dt} = h\nu c BI \left( [Cu^+] - \frac{g_3}{g_2} [Cu_m^+] \right) \frac{l_a}{l_c} - \sigma I \left( (1 - R)/l_c + \gamma \right) + h\nu c A [Cu^+] a.$$  (59)

The first term of (59) is for stimulated emission and absorption. $l_a$ is the length of the heated region and $l_c$ is the length of the optical cavity. The second term represents losses due to output coupling with mirror reflectivity $R$ and distributed losses $\gamma$. The last term is a spontaneous emission startup term where $A$ is the Einstein coefficient, $[Cu^+]$ is the density of the upper level, and $a$ is a geometrical factor. $[Cu_m^+]$ is the density of the lower level. $g_3/g_2$ is the ratio of statistical weights of the upper and lower level of transition. The constant $B$ is the cross section of the stimulated emission. The quantities $B$, $g_3/g_2$ and $A$ are different for the green and the yellow transitions.

The set of equations (52) to (59) forms the basis of the CVL model. Appropriate boundary conditions should be joined to the model and those conditions depend upon the electrical circuit. A typical discharge circuit considered by Kushner and al. is represented in Fig. 36. The authors have carried out and published an impressive parametrical study using the model. A similar model, with some modifications, is working...
Fig. 36 - Laser discharge circuit.

also in France. We present herebelow two applications just to illustrate what can be done with the model.

The first example is the simulation of a discharge when all the input data are specified. The aim is not to optimize a laser but to understand the physical phenomena occurring during the discharge phase and during the afterglow phase.

Fig. 37 is pertinent to the discharge phase. The penetration of the electric field is depicted in Fig. 37(a). The penetration takes place from the periphery towards the centre. A skin effect is observed and will affect the power deposition. The populating of the medium by electrons is represented in Fig. 37(b). Note the residual population at the initial time. Due to the skin effect, the periphery is ionized preferentially. The electrons so generated are heated by Joule effect as it can
Fig. 37 - Discharge phase.
be seen in Fig. 37(c). The heating starts to take place preferentially at the periphery, but very rapidly the electron temperature gets a smooth profile much more hot in the centre than near the wall. Fig. 37(d) shows how, locally, the medium gets populated by the upper levels $^2P_{3/2}$ and lower levels $^2D_{5/2}$ of the green transition. During some tens of nanoseconds the curve of $^2P_{3/2}$ population passes over the curve of $^2D_{5/2}$ population and renders possible the population inversion. In fact, the inversion is more favourable if one takes into account the ratio of statistical weights of the levels. The green light intensity is plotted in the lower diagram. The figure 37(d) corresponds to the radial position $r/R = 0.9$.

The afterglow phase is illustrated in Fig. 38. The cooling of the medium is represented in Fig. 38(a) for the electrons and in Fig. 38(b) for the gas. The medium is more hot in the centre than at the boundary. Fig. 38(c) represents the relaxation of the copper metastable levels. The recombination of charged particles is illustrated in Fig. 38(d). The relaxation of the metastables is very slow.

The second example of use of the CVL model concerns the scaling of large bore lasers. The method is to look for optimum conditions of operation for a specified diameter. The results of computations of Kusshner is reproduced in the TABLE (Fig. 39(a)). The CVL power versus the diameter is plotted in Fig. 39(b) where the experimental results are also plotted.
Fig. 38 - Afterglow phase.
### TABLE Nominal operating parameters for 6–8-cm large-bore CVLs.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>6(cm)</th>
<th>7(cm)</th>
<th>8(cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrode spacing (cm)</td>
<td>122</td>
<td>122</td>
<td>152</td>
</tr>
<tr>
<td>Active length (cm)</td>
<td>90</td>
<td>90</td>
<td>122</td>
</tr>
<tr>
<td>Operating temperature (°C)</td>
<td>1500</td>
<td>1450</td>
<td>1450</td>
</tr>
<tr>
<td>Neon pressure (Torr)</td>
<td>32</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>Copper partial pressure (Torr)</td>
<td>0.3</td>
<td>0.15</td>
<td>0.15</td>
</tr>
<tr>
<td>Nominal repetition rate (kHz)</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>Charge voltage (kV)</td>
<td>14</td>
<td>16.3</td>
<td>18.5</td>
</tr>
<tr>
<td>Storage capacitance (nF)</td>
<td>13</td>
<td>12</td>
<td>10.5</td>
</tr>
<tr>
<td>Input power density (W/cm³)</td>
<td>1.35</td>
<td>1.57</td>
<td>1.18</td>
</tr>
<tr>
<td>Peak discharge current (A)</td>
<td>1300</td>
<td>1600</td>
<td>2100</td>
</tr>
<tr>
<td>Average total power (W)</td>
<td>60</td>
<td>80</td>
<td>100</td>
</tr>
<tr>
<td>Code prediction</td>
<td>66</td>
<td>88</td>
<td>115</td>
</tr>
<tr>
<td>Light-pulse length (ns)</td>
<td>62</td>
<td>70</td>
<td>85</td>
</tr>
<tr>
<td>Code prediction</td>
<td>57</td>
<td>62</td>
<td>75</td>
</tr>
</tbody>
</table>

**Fig. 39 - Large bore CVLs.**
The experimental investigations aim to understand the dominant phenomena driving the performance of a CVL and to improve the output power. Recently, the effects of molecular gas impurities in the neon buffer gas on the output characteristics of a CVL have been investigated by Z.G. Huang and al. / 38 /. Experiments were carried out with several impurity gases including \( \text{H}_2, \text{H}_2\text{O}, \text{CH}_4, \) air and \( \text{N}_2 \) and yielded two main results summarized in Fig. 40:

![Fig. 40 - Influence of molecular gases on the output characteristics of a Copper Vapor Laser. Ref. / 38 /.](image)

**Fig. 40(a)**: The output power is plotted versus the pressure of the impurity for various species. Molecules with hydrogen atoms increase the laser output power, whereas nitrogen causes a
decrease. The improvement of the power is 50% compared to the case of pure neon.

Fig. 40(b): The radial distribution of the laser intensity is represented for various hydrogen pressures. Curve (1) without hydrogen, (2) hydrogen pressure = 0.43 torr, (3) 0.57 torr, (4) 0.80 torr, (5) 1.10 torr. Without hydrogen, the radial profile shows a skin effect with a dip in the center. As the hydrogen pressure increases the skin effect disappears gradually. The trend continues even after the optimum pressure was exceeded.

Many other experiments are reported in the literature on the direct measurements of population. In 1979, MILLER /39/ measured the decay of metastables with a dye laser tuned to the green transition and concluded that the deactivation is due to particle diffusion towards the wall. In 1980, SMILANSKI /40/ published hook diagnostic measurements for the upper $^2P$ and lower $^2D$ populations. This method was also used by TENENBAUM et al. /41/ but only to the measurement of the upper laser levels $^2P_{1/2}$ and $^2P_{3/2}$. In 1982, KUSHNER /36/ used also the hook diagnostic measurements for the green laser level populations to compare with the predicted results with his code (Fig. 41) and concluded to a good agreement.

Russian publications focus the attention on the measurement of metastable populations during the afterglow phase. KEL'MAN et al. /42/ have used the method of resonance absorption of the 510.6 nm line to determine the density of $^2D_{5/2}$ levels for various neon pressures. They found (Fig. 42) that
Fig. 41 - Comparison of calculated (a) and measured (b) green laser level populations according to KUSHNER /36/.

Fig. 42 - Dependences of the population of the $4s^2 \, ^2D_{5/2}$ metastable state of copper on the discharge axis on the time from the end of the current pulse, plotted for different neon pressures. Ref. /42/.
at neon pressures exceeding 10 KPa the metastable populations, after an initial rapid decay, increase over 5-10 μsec following the current pulse and finally decay after 10 μs. At lower pressures the metastable populations decrease monotonically. The same method of resonant absorption was used by ISAEV et al. /43, 44/ to study the spatial kinetics of the decay of metastable atoms, resolved over the radius of a CVL. Fig. 43 gives the radial profiles of the metastables obtained by the authors at different moments of time during the interpulse interval. A transformation is observed in the profiles from one with a minimum to one with a maximum on the discharge axis. Moreover, the authors studying the kinetics, point out that the decay of the population is characterized by two characteristic times, one of a few microseconds and the other a few tens of microseconds.

In the Russian literature, there is also /45/ a measurement of the electron population by interferometry.

4.2.2 - **Light-vapor interaction**

The inputs, the equations and the outputs of the model for the study of light-atom interaction is summarized in Fig. 44 drawn from Ref. /1, 4/.

Depending on the relative importance of stimulated and spontaneous processes, the interaction can be described by using either the SCHRODINGER equation or the simplified "rate-equations" system. We say a few words about each of the two approaches.
Fig. 43 - Radial profiles of metastables populations of the $^{2}\text{D}_{3/2}$ (continuous curves) and $^{2}\text{D}_{5/2}$ (dashed curves) levels at different instants during the interpulse interval; neon pressure 300 Torr; $W = 1.3$ (a), 1.5 (b), 2 (c) and 1.8 kW (d); $T_w = 1450$ (a), 1470 (b), 1600 (c) and 1550°C (d); laser output power $P_L = 1.5$ (a,b), 5.2 (c) and 4.8 W (d). Ref. / 43, 44 /.
### Inputs

<table>
<thead>
<tr>
<th>Spectroscopy</th>
<th>Laser Characteristics</th>
<th>Vapor/Separator Characteristics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured</td>
<td>Measured</td>
<td>Measured</td>
</tr>
<tr>
<td>- Level positions</td>
<td>- Temporal pulse shapes</td>
<td>- Vapor distribution</td>
</tr>
<tr>
<td>- Quantum nos.</td>
<td>- Laser beam profile</td>
<td>- Velocity distribution</td>
</tr>
<tr>
<td>- g-factors</td>
<td>- Specified</td>
<td>- Electric and magnetic fields</td>
</tr>
<tr>
<td>- HFS constants</td>
<td>- No. of lasers</td>
<td>- Specified</td>
</tr>
<tr>
<td>- Isotope shifts</td>
<td>- Detunings from atomic resonances</td>
<td>- Separator geometry</td>
</tr>
<tr>
<td>- Transition cross-sections (Dipole moments)</td>
<td>- Energy/pulse</td>
<td>- Optical system</td>
</tr>
<tr>
<td>- Level lifetimes</td>
<td>- Polarizations</td>
<td></td>
</tr>
<tr>
<td></td>
<td>- Propagation directions</td>
<td></td>
</tr>
<tr>
<td></td>
<td>- Temporal phases</td>
<td></td>
</tr>
<tr>
<td></td>
<td>- Pulse timing</td>
<td></td>
</tr>
</tbody>
</table>

### Physical models

- Schrödinger's Equation for laser - atomic vapor interaction
- Maxwell's Equations for laser propagation

### Outputs

- Time and velocity dependences of level populations
- Photonsinization fraction
- Atomic polarizations
- Module photomission performance

Fig. 44 - Modeling of laser - atomic vapor interaction.
First, the rate equation approach is outlined with the simplest scheme, i.e. a two step scheme, though more complex schemes with 3 or 4 colours are more realistic. Nevertheless the analysis of a two step scheme gives an idea of the kinetics of the process and serves as a basis for the calculation of more complicated schemes. Fig. 45 gives an overview of a general spectroscopic scheme of the selective two step atomic photoionization showing also some competing processes.

**Fig. 45**

The following notation is used:

- $U_i^+$ Population at the level $i$ (expressed in atoms/cm$^3$).
- $\sigma_{i,i+1}$ Absorption cross section for the transition $i \rightarrow i+1$ (the cross section of stimulated emission $\sigma_{i+1,i}$ is supposed to be equal to $\sigma_{i,i+1}$) – is expressed in cm$^2$.
- $I_{i,i+1}$ Flux of photons (expressed in photons/cm$^2$/s) of the laser carrying atoms from the level $i$ to the level $i+1$. 
Relaxation time of the atoms excited at the level $i$. 

$t$, time variable; $Z$, space variable.

The rate equations are:

\[ \frac{\partial U_0}{\partial t} = -I_{01} \sigma_{01} (U_0 - U_1) \]

\[ \frac{\partial U_1}{\partial t} = I_{01} \sigma_{01} (U_0 - U_1) - I_{12} \sigma_{12} U_1 - \frac{U_1}{\tau_1} \]

\[ \frac{\partial U_2^+}{\partial t} = I_{12} \sigma_{12} U_1 \]

\[ \frac{\partial I_{01}}{\partial Z} = -I_{01} \sigma_{01} (U_0 - U_1) \]

\[ \frac{\partial I_{12}}{\partial Z} = -\sigma_{12} I_{12} U_1 \]

The initial and boundary conditions are:

\[ t = 0 \quad U_0 = U_0^0 \text{ specified} \quad U_1 = 0 \quad U_2^+ = 0 \]

\[ Z = 0 \quad I_{i,i+1} = I_{i,i+1}^0(t) \text{ specified function of } t \]

The function $I_{i,i+1}^0(t)$ represents the time profile of the laser pulse. For instance, a "rectangular pulse" is convenient for calculations.
Equations (60) and (61) can be easily solved numerically with a computer.

An explicit formula can be given for that solution in the approximate case of "optically thin layer", i.e. the case where the length of propagation of the laser beam is sufficiently small (practically a few meters) to neglect the variation of all the quantities with the spatial coordinate $Z$. Moreover we delete the relaxation term $U_1/\tau_1$. The fluxes $I_{01}$ and $I_{12}$ are rectangular type and synchronized (both are switched on at $t = 0$).

The first three equations of the set (60) are simple linear differential equations with constant coefficients. The rate of production of the ions $U_2^+$ is:

$$\frac{U_2^+}{U_0} = 1 - \frac{a_1 + a_2}{2a_2} \exp\left[-(a_1 - a_2) t\right] + \frac{a_1 - a_2}{2a_2} \exp\left[-(a_1 + a_2) t\right]$$

where $a_1$ and $a_2$ are given by the equations:

$$a_1 = I_{01} \sigma_{01} \left(1 + \frac{I_{12} \sigma_{12}}{2I_{01} \sigma_{01}}\right) ; \quad a_2 = I_{01} \sigma_{01} \left[1 + \left(\frac{I_{12} \sigma_{12}}{2I_{01} \sigma_{01}}\right)^2\right]^{1/2}$$

In order to have a quantitative idea, consider an example of two commercially standard dye lasers which give

$$I_{01} = I_{12} = 5 \times 10^{23} \text{ photons/cm}^2/\text{s}$$

Pulse duration 10 nanoseconds.

and the possible values of cross sections:

$$\sigma_{01} = 10^{-14} \text{ cm}^2 \quad \quad \sigma_{12} = 10^{-15} \text{ cm}^2$$
The solution can be written

\[
\frac{U_2^+}{U_0^-} = 1 - 1.024 \, e^{-0.244 \, T} + 0.0243 \, e^{-10.25 \, T}
\]

\(T\) being the time in nanoseconds. The time rate ionization is represented by the diagram:

At the end of the pulse, 90% of the atoms in ground state are selectively ionized. In this scheme, as only 47% of atoms are in ground level, this means that approximately 40% of the \(^{235}\text{U}\) atoms can be at most expected to be ionized. This is rather poor and this is the main reason why, in the scheme of Fig. 4 one additional laser colour is used to excite also atoms in 620 cm\(^{-1}\) level. It is an easy exercise to compute the kinetics in the case of Fig. 4 (3 steps and four colours). A good performance would yield about 50% of \(^{235}\text{U}\) atoms selectively ionized.

We go on now to outline the second approach: the following summary is drawn from Ref. /7/.
The SCHRODINGER equation is used to describe details of time dependent behaviour in which coherence of the radiation-atom interaction dominates the dynamic. This question is discussed by ACKERHALT and SHORE / 46 / who compare the limits of validity of both rate equations and BLOCH equations in multi-photon ionization.

The atom important parameters to be considered are:

1/\tau : the spontaneous emission rate.
R : the ionization rate.
1/T_{c} : the collisional dephasing rate.

The important parameters for coherence are:

- \Omega = \mu E/h, the RABI frequency.
- \mu = (Debye units) is the dipole momentum value.
- and \ E = (V.cm^{-1}) the electrical field associated with the light intensity (E \propto \sqrt{I}).
- \Delta is the detuning frequency between the atom and the laser; for simplicity, ACKERHALT first considers a two level atom model:

\begin{align*}
(a) \quad \dot{\rho}_{11} & = \frac{1}{2} i \Omega (\rho_{12} - \rho_{21}) + \left( \frac{1}{\tau} \right) \rho_{22} \\
(b) \quad \dot{\rho}_{22} & = - \frac{1}{2} i \Omega (\rho_{12} - \rho_{21}) - \left( \frac{1}{\tau} \right) \rho_{22} - R \cdot \rho_{22} \\
(c) \quad \dot{\rho}_{12} & = - i \Delta \rho_{12} - \frac{1}{2} i \Omega (\rho_{22} - \rho_{11}) \left( \frac{1}{2} \tau \right) \rho_{12} \\
& \quad - \frac{1}{2} R \cdot \rho_{12} - \left( \frac{1}{2} T_{c} \right) \rho_{12}
\end{align*}
From the previous set of equations, it can be remarked that the spontaneous emission terms are those containing \( \rho_{11} \), \( \rho_{22} \), \( \rho_{12} \), whereas collisional dephasing only affects the equation (c) by the intermediate of the term \( \frac{1}{2T_1} \cdot \rho_{12} \).

Two simplifications can be made:

1 - one is to consider that the laser intensities are high enough to neglect the spontaneous emission \( (1/\tau = 0) \);  
2 - using the WILCOX and LAMB procedure which consist to annul the off-diagonal term \( \rho_{12} \) in equation (c), one gets:

\[
\rho_{12} = \frac{i\Delta}{2} \frac{\frac{1}{2} R + \frac{1}{2T_1} - i\Delta}{\left(\frac{1}{2} R + \frac{1}{2T_1}\right)^2 + \Delta^2} \cdot (\rho_{22} - \rho_{11})
\]

This expression of \( \rho_{12} \) used in equations (a) and (b) gives the rate equations:

\[
\rho_{11} = R_S (\rho_{22} - \rho_{11})
\]
\[
\rho_{22} = -R_S (\rho_{22} - \rho_{11}) - R \cdot \rho_{22}
\]

with \( R_S = \frac{\frac{1}{4} \Delta \left(\frac{1}{2} R + \frac{1}{2T_1}\right)}{\left(\frac{1}{2} R + \frac{1}{2T_1}\right)^2 + \Delta^2} \),

being the stimulated emission rate.
The actual "n" level system is computer calculated. As expected in the conditions where the spontaneous phenomena dominates the coherent processes, the rate equations are entirely valid.

A totally coherent multi-steps excitation process could be achieved using "m" pulses.

- "m" pulses area "A" are such that:

\[ A = \frac{\mu}{h} \int_{-\infty}^{\infty} E(t) \, dt = m \]

In such conditions, instead of reaching an equipartition of the population between a pair of levels, the totality of the atoms taken in the lower state can be brought in the upper state (Fig. 46). From the practical point of view, a "m" pulse traveling across a resonant vapour has a tendency to be compressed as a 2 m pulse. In such a case the vapour medium is totally transparent for the 2 m pulse. It is the so-called "soliton" formation.

Since the AVLIS quantum scheme uses up to 4 colours, it seems unrealistic to preserve the coherence of four m pulses all along the optical path which is evaluated as equal to several tens of meters.

The physics of the laser system includes also a very important problem of laser beam propagation which is not at all reviewed here.
Fig. 46 - Schematic representation of the atomic population versus time ($\Omega^{-1}$) (intermediate level). Full line: Rabi flopping as a solution of Bloch equations. Dotted line: solution to rate equations.

4.3 - Physics of ion extraction

This section includes two subsections.

4.3.1 - Time and space restrictions imposed by charge exchange

Once the vapor has been selectively ionized by an appropriate scheme of irradiation (for instance three steps and four colors in order to use the uranium atoms in ground level and the level 620 cm$^{-1}$) the situation is the following:
A "long" volume, which is the zone of interaction between the vapor beam and the laser beam, contains a mixture of electrons, ions and neutral atoms. The mixture is electrically neutral. The ions are the $^{235}\text{U}^+$ ions but there may be also a few amount of the undesired $^{238}\text{U}^+$ ions generated either by the electron gun source either by the lasers themselves whose selectivity is not perfect. The neutral atoms are mainly $^{238}\text{U}^0$ atoms but also a very few of $^{235}\text{U}^0$ atoms which were not selectivity ionized. In order to have a quantitative idea about a typical composition of the mixture, consider the natural uranium with a density of $10^{13}$ atoms/cm$^3$ in the zone of irradiation and assume that lasers ionize selectively 50% of $^{235}\text{U}$ atoms. Assume also that undesired $^{238}\text{U}^+$ ions are of the same order as $^{235}\text{U}^+$ ions. The composition of the mixture is the following:

<table>
<thead>
<tr>
<th>Species</th>
<th>Density</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrons</td>
<td>0.70 $10^{11}$ cm$^{-3}$</td>
</tr>
<tr>
<td>Ions $^{235}\text{U}^+$</td>
<td>0.35 $10^{11}$ cm$^{-3}$</td>
</tr>
<tr>
<td>Ions $^{238}\text{U}^+$</td>
<td>0.35 $10^{11}$ cm$^{-3}$</td>
</tr>
<tr>
<td>Neutrals $^{238}\text{U}^0$</td>
<td>0.9895 $10^{13}$ cm$^{-3}$</td>
</tr>
<tr>
<td>Neutrals $^{235}\text{U}^0$</td>
<td>0.35 $10^{11}$ cm$^{-2}$</td>
</tr>
</tbody>
</table>

Roughly, the process generates an uranium plasma, of density $10^{11}$ charged particles/cm$^3$ at the energy 0.2 eV, immersed in a neutral medium of uranium atoms hundred times more dense. The plasma contains desired $^{235}\text{U}^+$ ions while the neutral surrounding is made of an overwhelming majority of undesired $^{238}\text{U}^0$ atoms. The problem is to take out the ions from the neutral environment and to deposit them on a practical collector.
The method of ion extraction, very briefly sketched in section 3 consists in submitting the whole volume of the irradiation zone (Fig. 47) to an electric field created by two plane electrodes maintained at a controllable difference of potential. The electric field does not at all affect the neutral atoms but interacts with charged particles (electrons and ions) by inducing a dynamics. The aim of this section is to describe quantitatively this charged particle dynamics.

![Diagram of ion extraction](image)

**Fig. 47**

The motion of neutral atoms, which is not altered by the electric field, is approximately that of the beam expansion and can be decoupled from the motion of charged particles. In what follows, we investigate the motion of electrons and ions...
by assuming that neutrals are in rest. The presence of neutrals plays, nevertheless, a very important role through the collision phenomena, specially the resonant charge transfer collision between uranium ions and uranium neutral atoms. During a charge transfer collision between a positive ion \( A^+ \) and a neutral atom \( B^0 \), an electron is transferred from \( B^0 \) to \( A^+ \) according to the reaction

\[
A^+ + B^0 \rightarrow A^0 + B^+ .
\]

This reaction is resonant when the ion and the neutral belong to the same species (this is the case of isotopes). In our case, the charge transfer reaction

\[
^{235}U^+ + ^{238}U^0 \rightarrow ^{235}U^0 + ^{238}U^+
\]

is detrimental to the separation effect. So, it should be carefully taken into account in the modelisation of ion extraction.

The cross sections of resonant charge transfer \( \sigma_{CT} \) for several species have been widely investigated in the literature for several species, by direct measurements or by theoretical calculations. The values of \( \sigma_{CT} \) depends on the species but also on the relative velocity of the colliding particles. For uranium atoms, the calculated \( \sigma_{CT} \) versus relative velocity \( V \) is plotted in Fig. 48 according to three references. The results are not very different and can be summarized by an approximate semi-empirical formula

\[
\sqrt{\sigma_{CT}} = a - b \log_{10} V \quad (62)
\]

with \( a = 30 \), \( b = 3 \), \( V \) in cm.s\(^{-1} \) and \( \sigma_{CT} \) in \( \AA^2 \).
In our case, a reasonable value is

\[ \sigma_{\text{CT}} \approx 2 \times 10^{-14} \text{ cm}^2 \]  \hspace{1cm} (63)

From the cross-section, we can easily derive the frequency of charge transfer collision \( \nu_{\text{CT}} \)

\[ \nu_{\text{CT}} = \sigma_{\text{CT}} n_0 V \]  \hspace{1cm} (64)

where \( n_0 \) is the density of neutral atoms. The charge transfer collisions result in constraints on the extraction time and the
extraction distance. A rough estimation of these two constraints can be made as follows:

- **Limitation of extraction time.** Consider the volume in which lasers have generated ions $^{235}\text{U}^+$. If these ions undergo no motion, their density $n_5^+$ decay according to the law

$$\frac{dn_5^+}{dt} = - v_\text{CT} n_5^+$$

The extraction time $\tau_{\text{extr.}}$ should be

$$\tau_{\text{extr.}} \leq \frac{1}{v_\text{CT}}$$

For $v_\text{CT} = 2 \times 10^{-14}$ cm, $n_0 = 10^{13}$ and $V = 10^5$ cm.s$^{-1}$ this yields

$$\tau_{\text{extr.}} \leq 50 \text{ microseconds.}$$

- **Limitation of extraction distance.** If ions $^{235}\text{U}^+$ move across neutrals along a distance $\ell$, the decay of density is roughly

$$n_5^+ \sim \exp (- v_\text{CT} n_0 \ell)$$

The constraint on the extraction distance $\ell$ is therefore

$$\ell \leq \frac{1}{v_\text{CT} n_0}$$

For $v_\text{CT} = 2 \times 10^{-14}$ cm$^2$, $n_0 = 10^{13}$ atoms.cm$^{-3}$ the results is

$$\ell \leq 5 \text{ cm.}$$
4.3.2 - Extraction model

The early works dealing with this problem studied the combined action of crossed electric and magnetic fields /50/ on ion extraction. The authors of the preceding reference have also built a particle simulation code of a two dimensional plasma /51/ to study the problem. More recent topics don't include a magnetic field and theoretically and electric field is sufficient to meet the rather severe time and space restrictions mentioned in section 4.3.1. The resulting extraction scheme is depicted in Fig. 47. An idealized one-dimensional modelization of the situation shown in that figure is represented in Fig. 49. A slab plasma is suddenly created between two parallel plates. The problem is to follow the evolution of the plasma when a potential difference is established between the plates. Collisions will be neglected.

![Diagram of extraction model](image)

**Fig. 49** - One dimensional modelization of the extraction problem.
F.F. CHEN / 52 / has treated an almost similar problem by neglecting the spatial dependence and by using the BOHM sheath criterion / 53 /. The author has pointed out that the plasma decay proceeds in two distinct stages. In the first stage the sheath is moving very fast and reaches a stagnation state. The second stage is a slow decay of the plasma on a different time scale. If the plasma volume is large enough, the motion will be much slower in the second stage than in the first stage and the density decay exponentially with the time constant \( \tau \) given by CHEN:

\[
\tau = \frac{2D}{3} \left( \frac{kT_e}{M} \right)^{-1/2} - \frac{4}{3} \left( \frac{kT_e}{M} \right)^{-3/4} \frac{2}{(eM)^{1/4}} \frac{1}{\left(5.4 \pi n_o^{1/2} \right)^{3/4}} V^{3/4}
\]

where \( k \) is the Boltzmann constant, \( T_e \) the electron temperature, \( M \) the ion mass, \( n_o \) the initial density of the plasma and \( V \) the potential difference. As an illustration, consider the example of uranium with the following constants:

\[
M = 4 \times 10^{-22} \text{ g} \quad D = 4 \text{ cm} \quad T_e = 0.2 \text{ eV} \quad n_o = 10^{11} \text{ cm}^{-3}
\]

The plasma density decay is represented in Fig. 50. This figure shows that 90% of the plasma is extracted in 220 \( \mu \)s with zero potential and in 60 \( \mu \)s with a potential of 3000 Volt.

A much more elaborated model is published by C. GAUTHEREAU and Y. STEPHAN / 54 / and takes into account also the spatial evolution of the plasma. The authors use a fluid model to represent the ions and a thermal equilibrium model to represent the background electrons. The system of equations follows:
- Continuity:

\[ \frac{\partial n_i}{\partial t} + \frac{\partial}{\partial x} \left( n_i u_i \right) = 0 \]  

(65)

- Momentum:

\[ \frac{\partial u_i}{\partial t} + u_i \frac{\partial u_i}{\partial x} = -\frac{e}{M} \frac{\partial \phi}{\partial x} - \frac{kT_i}{M} \frac{1}{n_i} \frac{\partial n_i}{\partial x} \]  

(66)

- POISSON's equations:

\[ \frac{\partial^2 \phi}{\partial x^2} = \frac{e}{\epsilon_0} \left( n_e - n_i \right) \]  

(67)
Electron isothermal equilibrium statement:

\[ n_e = a(t) \exp \left( \frac{e \varphi}{kT_e} \right) \quad (68) \]

Total number of electrons between the 2 plates:

\[ N_e(t) = \int_0^D n_e \, dx \quad (69) \]

Flux of electrons per unit area of the 2 plates:

\[ \frac{dN_e}{dt} = -\frac{1}{4} \left[ n_e(o,t) - n_e(D,t) \right] \frac{8 kT_e^{1/2}}{\pi m_e} \quad (70) \]

In the above system, the following nomenclature is used:

Variables:
- \( t \): time
- \( x \): spatial coordinate, \( x \in [0, D] \)

Functions to determine:
- \( n_i \): ion density
- \( u_i \): ion velocity
- \( n_e \): electron density
- \( \varphi \): electric potential
- \( a \): multiplication factor in eq. (68) - \( a \) is a function of the time \( t \)
- \( N_e \): total number of electrons
. Constants (data of the problem)

- e electron charge
- \( \varepsilon_0 \) dielectric constant
- M mass of the ion
- \( m_e \) mass of the electron
- k Boltzmann constant
- \( T_i \) temperature of the ions
- \( T_e \) temperature of the electrons
- D distance between the plates

**Boundary conditions**

The plates are assumed to be perfectly absorbing. The potentials of the plates are specified:

\[
\varphi (0, t) = 0 \quad (71)
\]
\[
\varphi (D, t) = V \quad (72)
\]

\( V \) may be a constant or a function of time.

**Initial conditions**:

For \( t = 0 \), the following functions are specified:

- Ion density \( n_i (x, 0) \) (73)
- Velocity of ions \( u_i = 0 \) (74)
- Total number of electrons \( N_e (0) \) (75)
The model of ion extraction is composed of equations (65) to (75). The system of equations (65) to (68) has been treated by M. WIDNER and al. /55/, by assuming a constant, to investigate ion acoustic wave excitation and ion sheath evolution near a negatively biased electrode. The originality of the present model lies in the equations (69) and (70) which take into account the finiteness of the total number of charged particles initially generated. The model (65) to (75) has been solved numerically by using the finite element method and details on numerical methods are given in /54/. We present herebelow a numerical example developped in /54/ by the authors.

First, all the quantities are non-dimensionalized by dividing by appropriate scales: the inverse of the ion plasma pulsation $\omega_{pi}$ (calculated with the maximum initial density) for time scaling, the DEBYE length for length scaling, the maximum initial ion density for density scaling, the BOHM velocity $(kT_e/M)^{1/2}$ for velocity scaling, and the quantity $(kT_e/e)$ for electric potential scaling. The non-dimensionalized variables and functions are designated by the symbols with a bar over: $\bar{t}$, $\bar{x}$, $\bar{n}_i$, $\bar{n}_e$, $\bar{u}_i$, $\bar{\varphi}$, $\bar{u}$ and $\bar{N_e}$.

The numerical example developped by GAUTHEREAU corresponds to a Gaussian initial density profile and an alternating voltage established between the two plates:
\[ n_1(x, o) = \exp \left[ -\frac{1}{2} \frac{a^2}{\bar{x}^2} (\bar{x} - \bar{D})^2 \right] \]

\[ \varphi(o, t) = 0 \text{ for all } t > 0 \]

\[ \varphi(D, t) = -\varphi_{\text{max}} \cos \left( \frac{2\pi t}{T} \right) \] (76)

\[ D = 250 \quad a = 50 \quad \varphi_{\text{max}} = 100 \]

2 two values for \( T \): 2- and 8-

No initial velocity \( \bar{u}_1(x, o) = 0 \)

\[ T_1/T_e = 1 \]

The results of the computations are presented in the figures 54 to 57. The figures 51 to 56 are drawn in the case \( T = 2^- \) whereas the figures 57 to 61 pertain to the run \( t = 8^- \).

Fig. 51, shows the evolution of the electric potential during half a period. We can observe that the electric field vanishes everywhere except in the neighbourhood of the plates, in two non neutral regions that we will call the sheaths.

Fig. 52, shows the sheath displacement during the same time. The electrons move in phase with the external voltage while the ion density profile remains fixed on this time scale. It is possible to verify that the sheath thickness adapts itself to follow Child-Langmuir law. Electrons are allowed to leave the system each half a period on one side, when the potential difference between the plates reaches its maximum.
Figures 53 and 54 show the evolution of the electric potential and electron density on a larger time scale: the sheath thickness increases as plasma density decreases. To summarize, there is a rapid oscillation due to the external frequency combined with a slow progressive motion due to plasma extraction.

The ion density profile remains symmetric (Fig. 55), the voltage frequency being too high for ions to respond. Outside the sheaths, the ion density and flux profiles (Fig. 56) can be matched with an analytical homogeneous quasineutral solution describing isothermal plasma expansion into vacuum.

Figures 57 and 58 show the evolution of the ion flux on both plates. The first spike and the modulation are due to the external voltage. The average value profiles are equal to what can be obtained by thermal expansion into the vacuum.

Figures 59 and 60 show the evolution of the electron flux on the two plates. Electrons flow out by burst, whereas ions leave the system continuously. This is due to the fact that electrons vibrate and reach the plates every half a period.

Fig. 61 shows the decay of the total number of electrons and ions. There are always slightly more ions than electrons in the system because of the lower inertia of the latter. The plasma is practically extracted in a time approximately equal to $t_{\text{extr}} \sim 180$. 
Figure 51: EVOLUTION OF THE ELECTRIC POTENTIAL PROFILE DURING HALF A PERIOD

Figure 52: EVOLUTION OF THE ELECTRON AND ION DENSITY PROFILES DURING HALF A PERIOD
Figure 53 ELECTRIC POTENTIAL PROFILES

Figure 54 ELECTRON DENSITY PROFILES
Figure 55 ION DENSITY PROFILES

Figure 56 ION FLUX PROFILES
Fig. 57 - Ion flux on the negative plate.

Fig. 58 - Ion flux on the positive plate.
Fig. 59 - Electron flux on the negative plate.

Fig. 60 - Electron flux on the positive plate.
Fig. 61 - Decay of the total population of the electron and ion.
The extraction time found by this model is less than what can be computed by CHEN. For instance, in the case

\[ D = 4 \text{ cm} \quad T_e = T_i = 0.2 \text{ eV} \quad n_e(t=0) = 4 \times 10^{11} \text{ cm}^{-2} \]

and a constant voltage of \( V = -3 \text{ KV} \), the extraction time by the present model is 40 microseconds whereas CHEN's formula yields 60 microseconds.

5 - CONCLUSION

Finally what to say as conclusion about the laser process of separation?

. Scientifically, it is a fascination.

. Technologically it is a challenge.

Economically? I don't know! This is not my Job.
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