Modified nuclear level lifetime in hot dense plasmas

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The lifetime variations of a nuclear level in a plasma environment at thermodynamic equilibrium are discussed. Nuclear excitation and decay processes, including radiative processes, nuclear excitation by electron capture and internal conversion, nuclear excitation by electron transition and bound internal conversion are taken into account. The model is able to deal with any number of nuclear level having an influence on the considered level lifetime. Lifetime variations of several orders of magnitude are observed for various levels on different nuclei. The importance of NEET process on the examples of the ^{201}Hg (1.565 keV) and the ^{93}Mo (2424.89 keV) isomer levels is especially pointed out.

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I. INTRODUCTION

In a hot dense plasma, the lifetime of a nuclear level may be significantly altered [1,2] in comparison with the lifetime of an isolated nucleus called in this work the laboratory value. The large number of photons and free electrons modifies the environment in which an excited level of the nucleus naturally decays under laboratory conditions through spontaneous emission and internal conversion (IC), thus modifying its lifetime. Furthermore, new decay modes also appear such as induced photon emission, free electron scattering and bound internal conversion. Moreover, the lifetime is now dependent on excitation processes which can repopulate the excited level, and so increase its lifetime.

These considerations may be of great interest, either in a laser heated plasma [3–6], or in astrophysical plasmas [7–10]. In a previous work [11], we showed that in plasma, the nuclear decay rate of an isomeric level could be greatly enhanced by several orders of magnitude if there exists a nuclear level lying just above the isomer. In the present work, we intend to generalize the concept, by deriving a general expression of the nuclear lifetime of an excited level as a function of the temperature and the density of the plasma.

In the laboratory, an excited level lifetime depends only on the decay rate towards some, up to all, lower levels. In plasma, this decay rate is partially compensated by the excitation rate from the same lower levels. At low temperature, it might be enough to consider only the levels towards which it deexcites. For a higher temperature, higher levels will also matter, at least up to those whose excitation energy is in the same order of magnitude as the temperature. The levels which must be taken into account are those having a significant contribution to these transition rates.

Then, it is very important to involve in the calculation of transition rates every excitation and corresponding deexcitation process [12] which may occur in considered plasmas in order to correctly deduce the lifetime of the involved levels. The main couples of electromagnetic excitation and decay processes are photon absorption and photon emission (both spontaneous and induced), nuclear excitation by electron capture (NEEC), where a free electron is captured on an empty state of an atomic shell, and internal conversion (IC), inelastic electron scattering and superelastic electron scattering, and nuclear excitation by electronic transition (NEET) [13] and bound internal conversion (BIC). We recall here that NEET is a resonant process where an electron on a loosely bound shell decays down to an inner shell. If the energy difference between the two atomic shells matches that of a nuclear transition, the nucleus may be excited. Here, we will neglect all processes of the third order or higher. In particular, we will not elaborate on the electronic bridge process [14]. A nucleus decays by transferring its excitation energy to an electron on an atomic shell which will deexcite by emitting a photon. In cases where it has been experimentally observed, it was shown to be less efficient than direct photon emission [15].

^{201}Hg^{m} is a promising candidate for an experiment designed to show the feasibility of nuclear excitation in a laser created plasma. In Fig. 1 is a simplified level scheme of ^{201}Hg. One sees that the excitation energy is very low: 1.565 keV. Its lifetime has recently been measured [16] to be 81 ± 5 ns. Such a value allows decay well after the particle fluxes following the laser shot have decreased. The ^{201}Hg transition is strongly converted with a multipole mixing ratio $E2/M1$ of $\delta^2 = (2.1 \pm 0.5) \times 10^{-4}$. The matrix elements deduced from the experiment are $B(M1) = 0.00112$ W.u. and $B(E2) = 35.5$ W.u.

Another interesting case is ^{93}Mo^{m}, which has a 7 h isomeric state as shown on the simplified level scheme of Fig. 2. It can decay in plasma through an upper level lying 4.8 keV above when the temperature is higher than 200 eV [11]. Adopted value in Weisskopff units for the matrix elements are given in Table I. The unknown matrix element for the indirect transition has been assumed to be equal to the $E2$ transition to the lower level.

This paper is organized as follows. In the first part, we will focus on the enumerated processes in the case of relatively low temperature plasmas. One can note that under such conditions, one can neglect inelastic electron scattering. We describe radiative processes, IC and NEEC along the lines of the model developed in Ref. [11]. The contribution of NEET and BIC processes are dealt with the model exposed in Ref. [13].

In the second part, we will make applications concerning the first excited level ($t_{1/2} = 81$ ns) of the ^{201}Hg located at 1.565 keV, using a two level system. Another application devoted to ^{93}Mo^{m} ($t_{1/2} = 6.85$ h) isomeric state will be

TABLE I. Matrix elements for ^{93}Mo transitions.

Transition	Multipolarity	Matrix element (W.u.)
$17/2^+ \rightarrow 21/2^+$	$E2$	4.48
$17/2^+ \rightarrow 13/2^+$	$E2$	4.48
$21/2^+ \rightarrow 13/2^+$	$E4$	1.431

presented, using a generalized model of a multi level system. In a last part, we will draw some conclusions and perspectives.

II. TRANSITION RATES IN PLASMA

In this paper, we assume that plasmas are at local thermodynamic equilibrium (LTE). The atom description is that of the relativistic average atom model [17,18]. The statistical nature of the electronic spectrum is taken into account using a Gaussian distribution of the electronic configurations around the transition given by the average atom [19–23]. For each excitation process, we must take into account its corresponding deexcitation process, as summed up in Table II.

Each couple of excitation and deexcitation rates must verify the principle of detailed balance:

$$\frac{\lambda_e}{\lambda_d} = \frac{2J_f+1}{2J_i+1} e^{-\frac{\Delta E}{k_B T}}, \tag{1}$$

where k_B is the Boltzmann constant, ΔE the nuclear transition energy and J_i and J_f the spins of lower state i and the upper state f.

These transition rates rule the behavior of the populations N_i and N_f of both states, which will reach asymptotic values given by the Boltzmann law

$$\frac{N_f}{N_i} = \frac{2J_f+1}{2J_i+1} e^{-\frac{\Delta E}{k_B T}}. \tag{2}$$

At thermodynamic equilibrium, the photon population can be described by a Planck distribution, that gives a photon absorption rate

$$\lambda_e^\gamma = \frac{2J_j+1}{2J_i+1} \frac{\ln 2}{T^\gamma_{j\rightarrow i}} \frac{1}{e^{\frac{\Delta E}{k_B T}} - 1} \tag{3}$$

and the total (spontaneous and induced emission) deexcitation rate

$$\lambda_d^\gamma = \frac{\ln 2}{T^\gamma_{j\rightarrow i}} \frac{e^{\frac{\Delta E}{k_B T}}}{e^{\frac{\Delta E}{k_B T}} - 1}, \tag{4}$$

where $T^\gamma_{j\rightarrow i}$ radiative lifetime.

In Fig. 3, we show the radiative transitions rates of ^{201}Hg. The emission rate is constant at low temperature, when only

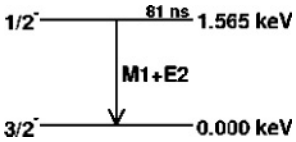

FIG. 1. Simplified level scheme of ^{201}Hg.

TABLE II. Excitation and deexcitation processes.

Excitation		Deexcitation	
λ_e^γ	Photon Absorption	λ_d^γ	Photon emission
$\lambda_e^{\mathrm{NEEC}}$	NEEC	λ_d^{IC}	Internal conversion
$\lambda_e^{\mathrm{NEET}}$	NEET	λ_d^{BIC}	Bound internal conversion

spontaneous emission matters. When the temperature gets higher than the nuclear transition energy, induced emission becomes significant and the emission rate increases. The absorption rate increases monotonously as the photon density increases with temperature.

The second couple of processes we are interested in are NEEC [24,25] and IC. Their rates have been obtained in Ref. [11]. Denoting E_b as the bound electron energy, E_r the free electron energy, and ε the dispersion of the electronic transition energy around the average atom value, the NEEC rate is expressed by

$$\lambda_e^{\mathrm{NEEC}} = \frac{2\pi}{\hbar} n(E_r) |\langle \psi_f \varphi_b | H | \psi_i \varphi_r \rangle|^2 (2J_f+1) f_{FD}(E_r) \times [1 f_{FD}(E_b)] \frac{1}{2} \left[\mathrm{erf}\left(\frac{E_r}{\varepsilon\sqrt{2}}\right) - \mathrm{erf}\left(\frac{E_b}{\varepsilon\sqrt{2}}\right) \right] \tag{5}$$

and the IC rate by

$$\lambda_d^{\mathrm{IC}} = \frac{2\pi}{\hbar} n(E_r) |\langle \psi_i \varphi_r | H | \psi_f \varphi_b \rangle|^2 (2J_i+1) f_{FD}(E_b) \times [1 f_{FD}(E_r)] \frac{1}{2} \left[\mathrm{erf}\left(\frac{E_r}{\varepsilon\sqrt{2}}\right) - \mathrm{erf}\left(\frac{E_b}{\varepsilon\sqrt{2}}\right) \right]. \tag{6}$$

In both expressions, n is the free electronic state density as a function of energy, and f_{FD} is the Fermi-Dirac statistics function.

In the case of ^{201}Hg, these rates are shown on Fig. 4 for a density of 10^{-2} g/cm^3 corresponding to the value of the cutoff density in a plasma created by a neodymium laser. At low temperature, the internal conversion rate is very close to its laboratory value, and it decreases when the plasma temperature causes conversion electrons to be ionized. The slope variations near 0.2 keV and 0.6 keV correspond to the ionization of the O and N shells, respectively. The NEEC rate first increases as the atom gets ionized and free electrons are more numerous, and then decreases when their kinetic energy gets higher than the nuclear transition energy.

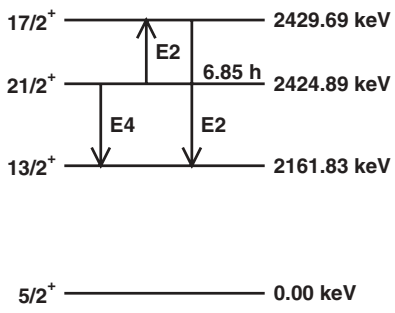

FIG. 2. Simplified level scheme of ^{93}Mo.

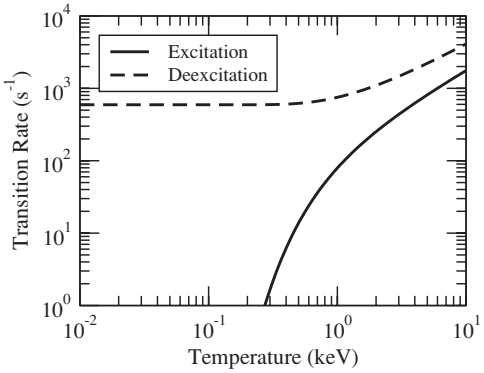

FIG. 3. Radiative transition rates of the 1.565 keV level of ^{201}Hg.

Finally, the NEET and BIC processes have to be considered. The NEET rate has been derived in Ref. [13] and presented for the ^{201}Hg^m in [16]. It is expressed as the product of a probability of the NEET process as a function of time with the creation rate of an atomic configuration with an electron on the upper shell and a hole in the lower shell. At thermodynamic equilibrium, the NEET probability can be assumed to take its asymptotic value and with $\bar{\delta}$ designating the average mismatch between the atomic and nuclear transitions, indexes 1 and 2 of the upper and lower atomic shells, the NEET rate is

$$\lambda_e^{\mathrm{NEET}} = \frac{2\pi}{\hbar}(2J_1+1) f_{FD}(E_1)[1-f_{FD}(E_2)]|R_{12}(\bar{\delta})|^2 \times \frac{e^{-\frac{\bar{\delta}^2}{2\varepsilon^2}}}{\sqrt{2\pi\varepsilon^2}}, \tag{7}$$

where ε is the same dispersion defined in Eq. (5) and R_{12} is the coupling matrix element. It is close to the matrix element for NEEC and its expression is detailed in Ref. [13].

Following the reasoning which led to this last expression, we can derive the corresponding BIC rate given by

$$\lambda_d^{\mathrm{BIC}} = \frac{2\pi}{\hbar}(2J_2+1) f_{FD}(E_2)[1-f_{FD}(E_1)]|R_{21}(\bar{\delta})|^2 \times \frac{e^{-\frac{\bar{\delta}^2}{2\varepsilon^2}}}{\sqrt{2\pi\varepsilon^2}}. \tag{8}$$

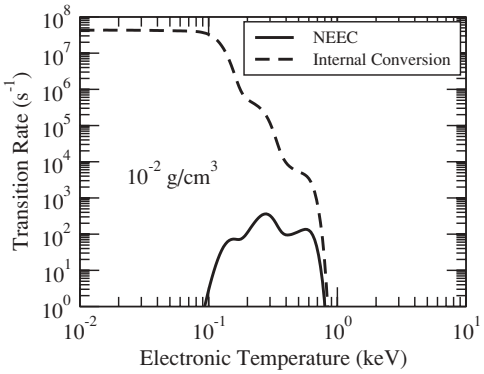

FIG. 4. NEEC and internal conversion rates of the 1.565 keV level of ^{201}Hg.

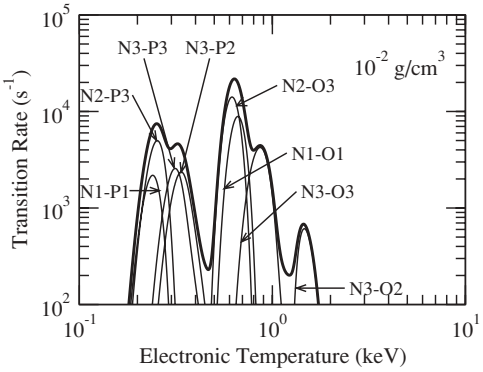

FIG. 5. NEET rates of the 1.565 keV level of ^{201}Hg.

These expressions stand for a given atomic transition which matches the nuclear transition. In practice, we must sum these rates over all atomic transitions which achieve matching conditions somewhere in the explored density temperature domain. The total NEET rate and a sample of the more significant atomic transitions contributions is given in Fig. 5 for a density of 10^{-2} g/cm^3. The total NEET rate is higher than the NEEC and photon absorption rates as the wave functions overlap is better when involving two bound electron wave functions.

The total transition rates are presented in Fig. 6. They include radiative processes, NEEC and IC, and NEET and BIC. Each couple of process dominates within a given temperature range. Below 0.1 keV, IC and NEEC rule the transition rates, with a nearly constant deexcitation rate close to the laboratory measured decay rate of the isomer. Between 0.1 and 2 keV, the resonance structure of NEET and BIC rules the total transition rates. For higher temperatures, inelastic electron scattering could begin to play a part. However, it was not taken into account in this study, and so only radiative transitions govern the transition rates behavior for higher temperatures.

III. LIFETIME OF AN EXCITED LEVEL IN PLASMA

As pointed out in introduction, the usual notion of an excited level lifetime needs to be more carefully considered in plasma. Under laboratory conditions, the excited level can only be depopulated by deexcitation, and its lifetime is solely dependent on the deexcitation processes. In plasma, the excited level

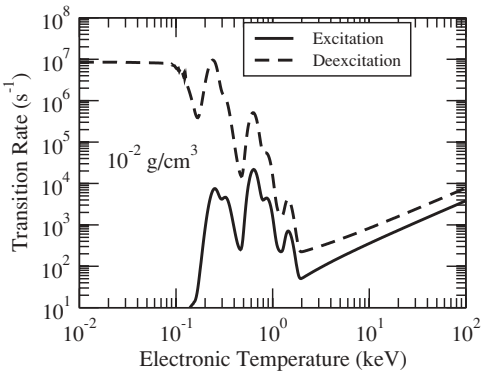

FIG. 6. Total transition rates of the 1.565 keV level of ^{201}Hg.

can also be repopulated by excitation from the ground state, which has two significant consequences. First, the deexcitation characteristic time is longer, and second, the system of both levels will finally reach an equilibrium state where both levels are populated according to the Boltzmann ratio. Therefore, the usual notion of a nuclear lifetime associated with only the decay rate of this level no longer holds. We need to replace it with the characteristic time of evolution of a system taking into account all involved levels.

A. Two level system and $^{201}Hg^m$ lifetime

In order to test the influence of the repopulation process, we will first consider here a system with a ground level and a single excited level. This system is relevant when the energies of the higher levels are much higher than the plasma temperature, such as $^{201}Hg^m$.

As in plasma, both excitation and deexcitation of the excited level occur, the decay of the population of the excited level towards the ground level is then ruled by the differential system

$$\begin{cases} \frac{dN_i}{dt} = -\lambda_{if} N_i + \lambda_{fi} N_f, \\ \frac{dN_f}{dt} = +\lambda_{if} N_i - \lambda_{fi} N_f, \end{cases} \tag{9}$$

where λ_{if} is the excitation rate and λ_{fi} the deexcitation rate.

If the thermodynamic conditions are stationary, the transition rates are constant and this system is easily solved. The populations as a function of time are given by

$$\begin{cases} N_i(t) = \frac{\lambda_{fi} + [\lambda_{if} N_i(t_0) - \lambda_{fi} N_f(t_0)] e^{-(\lambda_{if}+\lambda_{fi})(t-t_0)}}{\lambda_{if}+\lambda_{fi}}, \\ N_f(t) = \frac{\lambda_{if} - [\lambda_{if} N_i(t_0) - \lambda_{fi} N_f(t_0)] e^{-(\lambda_{if}+\lambda_{fi})(t-t_0)}}{\lambda_{if}+\lambda_{fi}}. \end{cases} \tag{10}$$

Both populations will reach an equilibrium state after a characteristic time given by

$$\tau = \frac{\ln 2}{\lambda_{if} + \lambda_{fi}}. \tag{11}$$

This equilibration time is the characteristic lifetime of the excited level. If the excitation rate is neglected, it reduces to the usual lifetime as observed in laboratory.

When the equilibrium is reached, the ratio of the populations can be easily calculated by zeroing the differential terms in the system (9), which is in agreement with the Boltzmann law in Eq. (2).

Figure 7 shows the lifetime of the two level system of the ^{201}Hg as a function of temperature for a density of 10^{-2} g/cm^3. The solid and dotted lines correspond to the lifetime obtained with and without including the NEET/BIC processes, respectively. Up to a temperature of 100 eV, the $^{201}Hg^m$ lifetime is a constant equal to the laboratory value. Between 100 eV and 2 keV, NEET and BIC rule its behavior, with the resonance structure observed on their transition rates. Inside this interval, the lifetime increases by more than four orders of magnitude, from 81 ns to more than a millisecond.

Compared to the calculation without the NEET/BIC processes, the lifetime is very structured and it may be reduced, for some temperatures, by more than two orders of magnitude. This can have a dramatic consequence on a laser driven plasma experiment, where created isomers could decay down to the ground state faster than expected. For temperatures higher than 2 keV, all electron shells involved in NEET and NEEC are fully ionized, and the isomer lifetime is then ruled solely by radiative transitions.

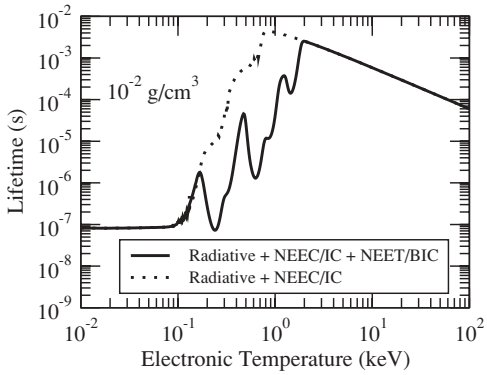

FIG. 7. Nuclear lifetime of the isomeric level of ^{201}Hg.

The lifetime variations can be more readily observed on Fig. 8 where the evolution of a population of the $^{201}Hg^m$ isomer as a function of time is shown for a fixed temperature of 635 eV. The population decays down nearly 1000 times faster when NEET is taken into account. In both cases, the isomeric population converges towards a unique asymptotic value as it only depends on the temperature according to the Boltzmann law (2).

In Fig. 9, the evolution takes into account all processes at different temperatures. The three highest temperatures correspond to either side and to the peak of the second resonance at 485 eV of the lifetime shown in Fig. 7. The asymptotic values increase as a function of temperature, while the lifetimes are not monotonous, thus creating a crossing point between the two last temperatures. These results will be useful tools to design the extraction of the isomers obtained in a laser created plasma.

B. Multilevel system: application to $^{93}Mo^m$

In this part, we generalize the two level models to a multi level system. This will allow dealing with the indirect deexcitation process, where at least three different levels are involved, such as $^{93}Mo^m$. In a general case with n levels

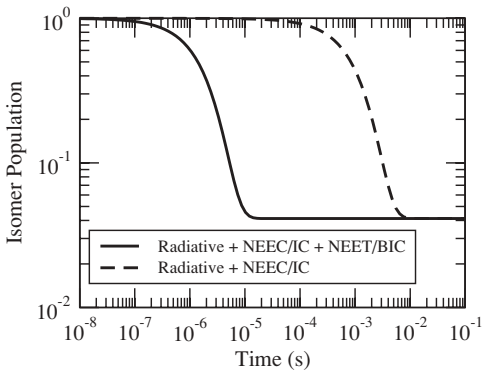

FIG. 8. $^{201}Hg^m$ isomer population evolution in a plasma at 635 eV.

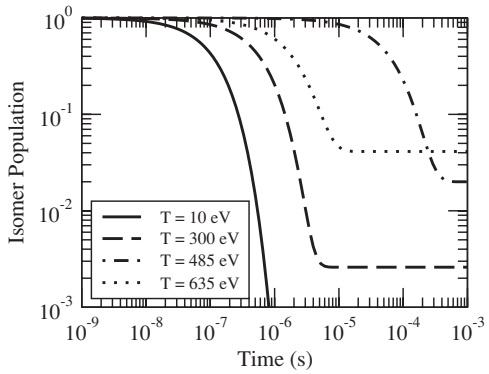

FIG. 9. $^{201}Hg^{m}$ isomer population evolution in plasma for several temperatures.

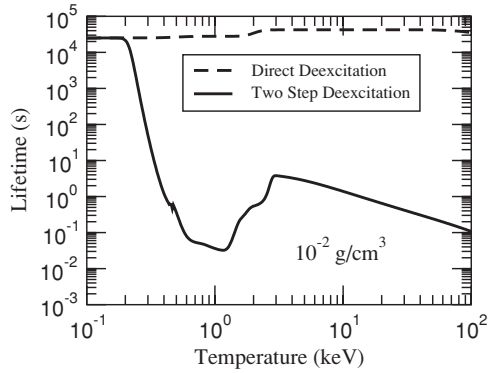

FIG. 11. Nuclear lifetime of the isomeric level of ^{93}Mo.

(including the ground state) labeled i with a population given by N_i, the evolution of the populations is deduced from the system

$$\frac{dN_i}{dt} = -\sum_{\substack{j=1 \\ j\neq i}}^{n} \lambda_{ij} N_i + \sum_{\substack{j=1 \\ j\neq i}}^{n} \lambda_{ji} N_j. \tag{12}$$

As in the case of the two level system, under stationary conditions, a matrix solution of this system can easily be found. The lifetime of the system is deduced from the eigenvalues of the matrix formed from the transition rates. This matrix has n negative eigenvalues, one of them obviously zero. The characteristic time of the evolution of populations is given by the highest nonzero eigenvalue λ though the relation

$$\tau = -\frac{\ln 2}{\lambda}. \tag{13}$$

This characteristic time is the lifetime of the system, and so is the lifetime of every level in it. It is no longer possible to define individual lifetime for each excited level, as the population of a given level cannot reach an equilibrium value while the others still have not.

In the case of $^{93}Mo^{m}$, the excitation of this upper level occurs through a low energy nuclear transition which allows NEET transitions. Between 400 eV and 3 keV, NEET is the predominant excitation process, as shown in Fig. 10. Inside this interval, the excitation rate is more than 100 times higher than the combination of the other processes.

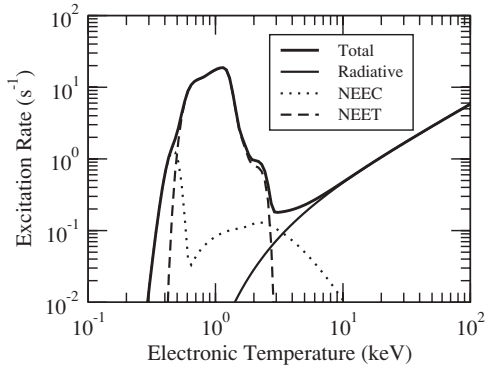

FIG. 10. Excitation rates from the isomeric level of ^{93}Mo to the upper intermediate level.

The lifetime evaluation must take into account this level in addition to the lower level towards which it naturally decays. As can be seen in Fig. 11, the influence of this upper level is preponderant for temperatures higher than 200 eV. The dashed curve shows the lifetime for direct deexcitation only, which is more or less a constant as no internal conversion is possible with such a high energy transition. The solid curve shows that the actual lifetime is decreased by around five orders of magnitude, as almost all of the deexcitation occurs through the upper level.

IV. CONCLUSIONS

We presented in this work, the calculation of the characteristic time of a multi nuclear level system in plasma. Various electromagnetic processes are taken into account: photon absorption and photon emission (both spontaneous and induced), nuclear excitation by electron capture (NEEC) and its inverse process, internal conversion (IC), nuclear excitation by electron transition (NEET), and bound internal conversion (BIC). We showed that the usual definition of the nuclear lifetime as the time during which half of the nuclei decays, does not hold in plasma. We calculated the characteristic lifetime in a thermodynamic equilibrium plasma for two isomeric levels: the $^{201}Hg^{m}$ and the $^{93}Mo^{m}$ located at 1.565 keV and 2424.9 keV, respectively.

The lifetime of the $^{201}Hg^{m}$ isomer in a plasma was obtained. It rises from 81 ns, for a low plasma temperature, to several milliseconds at higher temperature with a very structured behavior mainly due to the NEET/BIC effect. We also calculated the decay of the $^{93}Mo^{m}$ as a function of the plasma temperature and show the tremendous variation of its lifetime.

Experimental evidence of the large variations of the lifetime in plasma has to be observed. To perform such an experiment, we first need to create the isomer before observing its decay in plasma. An ultra-short laser pulse can produce energetic particles which can be used to create a significant amount of isomers in a plasma target. For instance, the $^{93}Mo^{m}$ could be produced by $^{93}Nb(p,n)$ reactions where protons are generated by a subpicosecond laser [26]. Then, the Nb

target, containing some $^{93}Mo^{m}$ isomers, could be heated *in situ* by a nanosecond laser. The advent of the new generation of intense lasers [27] opens the opportunity to reach this goal.

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