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S. A. Karamian, J. J. Carroll*

POSSIBILITY OF COMBINING NUCLEAR LEVEL PUMPING IN PLASMA WITH LASING IN SOLID

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*Department of Physics and Astronomy, Center for Photon-Induced Processes, Youngstown, 44555, Ohio, USA

1. Introduction

The triggered release of isomer energy is very attractive for the production of source emitting powerful pulses of radiation in the gamma-ray range. For example, self-stimulation of an isomeric transition would create a gamma-ray laser, and such a device might also be possible using a triggered release as a precursive step to a lasing transition. There are, however, many problems to be solved since significant barriers are already visible in proposed variants for realizing a gamma-ray laser. The problem of nuclear isomer triggering by X-ray radiation is reviewed recently in Ref. [1]. The list of long-lived nuclear isomeric states has been reduced in Ref. [2] and new candidates for triggered release of energy have been discussed.

In literature, many mechanisms of nuclear excitation by changes in electron configuration were proposed in addition to the resonance photon absorption process. There are, among them, nuclear excitation by electron transition (NEET), nuclear excitation by electron capture (NEEC), laser assisted NEET and electron bridge mechanisms. Some experiments and theoretical analyses performed in this field can be cited, Refs. [3-14], but the exciting progress is not yet reached. So that, the efforts to find the best physical mechanism for nuclear level excitation (triggering) and a new experimental schemes might still be a relevant approach today. The ^{181}Ta, ^{193}Pt, ^{201}Hg, ^{205}Pb, ^{229}Th and ^{235}U nuclides were proposed is Refs. [5,9,13] to be the best candidates for nuclear excitation by X-ray photons with $E_x < 7$ keV in plasma. We discuss some other nuclei, in particular, the ^{242m}Am long-lived isomer ($T_{1/2}$=141 y). They are selected by the parameters of long lifetime (or stable), of high production cross-section (if radioactive) and low multipolarity of the excited transition, as M1, E1 and E2.

The efficiency of energy transfer from atomic to nuclear subsystem is defined not only by the microscopical properties of atomic and nuclear levels, but also by the state of the substance contained the atoms and nuclei. Recently, a new experimental results have

been obtained on the time-scale of the desorption of atoms from the solid surface to the ablation-cloud after powerful fms pulse of the laser light, Ref. [15], and on the consequent relaxation of the solid sample (crystal), Ref. [16].

2. Processes in laser plasma

As is know, the interaction of TW laser pulse with solid matter generates immediately an induced oscillation of electrons with a frequency of laser light. Such collective excitation of electronic subsystem decays then with an energy transfer to the highly-excited states of atoms and to deep ionization of atomic shells. The nuclear subsystem can be also perturbed because of interaction with fast electrons, or with characteristic X-ray photons, bremsstrahlung and thermal spectra of radiation. K- and L-electrons can be ejected from heavy atoms, and the vacancies created serve as a source of characteristic photons with an energy effective for excitation of some specific nuclear states.

The excitation of nuclear levels in ^{201}Hg and ^{181}Ta isotopes under exposure them to the plasma radiation created by short laser pulses was tested in Refs. [3-5], and the perspectiveness of this direction was supported experimentally. The device for laser pumping of nuclear level can be, in principle, small and simple in configuration. Possible scheme is shown in Fig.1. Focused beam of the laser light creates the high-power density spot on the sample surface. The cross-size of the spot can be as small as 100 μ. Excited electrons leave their atoms immediately, and solid matter is transformed to be a dense plasma of ionized atoms. Other radiational processes are developing rapidly. Then, the desorption of atoms to the vacuum and thermal ablation follow. Finally, the hot spot is recrystallized and at the end, the ablation cloud is deposited onto the sample and surrounding surfaces.

The time scales of different processes are given in Table 1. Relaxation of solid matter after the pulsed release of energy has been studied recently in Ref. [15,16] for the

case of the fms pulses of laser-light impact to the metal and semiconductor samples. It is found that all processes develop rapidly, and the solid is recrystallized within 100 ps. It would be interesting to stress here that absolutely different process of the pulsed energy release in solid is characterized by similar time-scale, Ref. [17,18]. High-energy heavy ion deposits a lot of energy within a restricted volume of nuclear track in solid. The electronic medium is immediately excited, the same as in the case of the laser pulse, and then a consequence of the relaxation processes follow. It was shown experimentally [17,18] that in perfect crystals the recrystallisation is one of the probable final stages, and it is reached again within time-scale of 100 ps. Such similarity is definitely not a casual event, but the confirmation of the general properties of processes in solid after pulsed energy release.

Time-scale of nuclear fluorescence defined by the lifetime of excited level, has been estimated using standard systematics of the strength of nuclear electromagnetic transitions. As is clear in Table 1, the nuclear level decay time is typically longer than the time-scales of all other processes in atomic and solid subsystems. Even recrystallisation of solid may happen before the nuclear fluorescence events. So that, the nuclear radiance takes place in solid surroundings, and this is favourable for lasing.

Such an advantage accomplishes other attractive properties discussed in Refs. [3-5] for the scheme assuming the laser pulses as a tool for pumping of a hypothetical γ-ray laser on nuclear levels. However, it would be necessary to discuss also the disadvantages of the scheme in Fig.1:

1. Some part of excited nuclei must be released to the gas phase due to the ablation of atoms from the exposed surface. As long time as of about 10 ns is required for the condensation of the ablation cloud back to the surrounded solid surfaces. Nuclear levels may be long-lived, but for them, the probability of excitation is lowered because of their narrow width. For levels with optimum lifetime of about 100 ps, the efficiency of radiance in solid is going to be reduced because of the losses of excited nuclei to the gas

phase. The compromise can be found by the choice of the optimum surface density of the power. The rate of ablation can be reduced while the total number of excited nuclei is conserved. The full energy of the laser pulse can be distributed over larger area at the surface.

2. The resonance tuning between nuclear and atomic resonators cannot be perfect even when both frequencies coincide. The width of atomic resonance is typically much larger than the nuclear one because of orders of magnitude different lifetimes of the atomic and nuclear states.

This leads to the low value of the "Q-factor" corresponded to the resonance between nuclear and atomic modes. Such a situation has been confirmed recently in the direct measurements of the NEET process probability in the synchrotron-radiation experiment of Ref. [14]. The nuclear transition $3/2^+ \rightarrow 1/2^+$ with E=77.351 keV in ^{197}Au has been excited by the nuclear conversion of the atomic MI→K transition after K-vacancy ionization. The difference in energy was only 51 eV, and this was comparable with the total width of the K-vacancy state. The probability of NEET was found to be $5 \cdot 10^{-8}$ being pretty close to the widths ratio for nuclear and atomic states, Γ_n/Γ_a. The nuclear transition lifetime is known experimentally (2.76 ns), and the atomic level width can be taken from the systematics of Ref. [19].

The mismatch of widths suppresses not only the NEET probability but also the integrated cross-section for the resonance photon absorption. This fundamental reason reduces the nuclear excitation probability and it can not be cancelled completely. One may discuss only some palliative options capable to soften the restrictions above. The lifetime of atomic transition is normally increased for lower energy of characteristic photons, the same as in the case of nuclear transitions. One may suppose that vacancies in M-shell should be much longer-lived than K- and L-vacancies, and this would be promising to reach comparable magnitudes for the atomic and nuclear widths. In fact, this is not true. For

characteristic energies below 15 keV, the radiative width decreases to be lower the width of the intrinsic conversion of the transition energy within atomic shell through the Auger and Coster-Kronig processes. The total width is covered mostly by two latter processes, and the fluorescence yield of the X-ray photons is going to be $<<1$. Thus, the resonance photon excitation process is suppressed not only due to the widths mismatch, but also directly due to the low yield of characteristic photons.

In plasma, atoms are strongly ionized, the maximum of charge-state distribution corresponds to the values of q=10-30, dependent on the plasma temperature, Ref. [9]. The wide charge-state distribution creates the series of characteristic X-ray energies deviating from standard ones known for single vacancy, and this is helpful for exact tuning of the atomic and nuclear transitions energies. On the other hand, high stage of ionization should be effective to suppress the probability of the Auger and Coster-Kronig processes for soft characteristic transitions with $E_X \leq 10$ keV in strongly ionized heavy atoms. Respectively, the total width of atomic resonance is decreased and the relative probability of radiative processes is increased. Both effects are useful for the nuclear conversion of atomic excitation. Thus, one can anticipate that in plasma surroundings even M-vacancies must be active for the production of characteristic lines and for the resonance excitation of nuclear transition via photon absorption and NEET processes.

In Ref. [4], there was discussed the possibility to suppress the electron conversion of nuclear γ-rays in hot plasma. This effect increases the contribution of the radiative width to the total width of nuclear level (decreasing the conversion coefficient), and it can be useful in some schemes of nuclear level pumping and radiance. In addition, the possibility to use the electron conversion of nuclear radiation for the separation of some specific nuclear states in a multilevel scheme has been proposed in Ref. [4] also.

However at present, we are going to stress that the hot plasma may serve as a tool for the suppression of the nonradiative conversion of the ionization energy within atom.

This way the efficiency of energy transfer from atomic to nuclear mode can be significantly increased.

When nuclear and atomic transitions have the same energy and multipolarity, the hybridization of atomic-nuclear components is possible. Special wave function of the hybride state includes such peculiarities as: beating in amplitude of two modes with repumping of the energy, retardation in the decay time of both modes, nonexponential decay low and so on. As a result, the conversion of energy within the atom may be additionally suppressed, and the nuclear conversion of electron transition is enhanced. The hybridization was discussed in Ref. [20]. Finally, one can conclude that some processes exist and prohibite the resonance energy transfer from atom ionization to nuclear excitation, despite the atomic-nuclear resonance is not very sharp resonance, in origin.

3. Candidates for the atomic-nuclear resonance

Fortunate similarity in energies of atomic and nuclear transitions can be found for some isotopes, they are listed in Table 2. Resonance photon excitation of nuclear level requires only the appropriate value of photon energy, while the multipolarity matching has no significance. Photons can be emitted even by the atom of another element present in the sample as a catalyze admixture. This kind example was proposed in Ref. [21] for the transition in the ^{161}Dy nucleus excited by the Sn K-X ray with appropriate energy. However, higher efficiency must be reached when the characteristic X-ray energy is absorbed by the nucleus within the same atom.

The NEET process appears when both energy and multipolarity of atomic and nuclear transitions are identical. In definition, NEET happens only within individual atom-nucleus system. The examples of probable NEET manifestation are reduced in Table 2, and among them there is the case of ^{197}Au realized recently in the experiment of Ref. [14].

In Table 2 the difference in energy of atomic and nuclear transitions appears to be not very low, in some cases of about 1 keV. But one has to remember that atomic transition

energies E_x are given for the single vacancy atom, and this value has to be changed for each individual ionization state of the ion. In hot plasma, wide distribution of the charge states arises, and the series of atomic terms correspond to them. So that, better conditions for the atomic-nuclear resonance may be created in plasma and tuned by the variation of plasma temperature.

The long-lived ^{242m}Am isomeric state with $I^{\pi}=5^-$ seems to be a nucleus of special interest. Triggering via the E2 transition to 3^- state at 52.9 keV releases the energy stored in a form of the isomeric excitation energy. Saved in the ^{242m}Am sample specific energy is as high as 20 MJ/g. Resonance triggering of ^{242m}Am requires transition energy of (4.30±0.05) keV, and it can be supplied by the atomic transition in Am atom. Even three transitions have the appropriate energy. Photon absorption and NEET mechanisms both are possible, because the multipolarities can be also identical. The quantitative parameters are given in Table 2.

In addition to the unknown strength of the nuclear transition in ^{242}Am, another problem has been visible that is connected with the rather low energy of the atomic transition, of about 4.3 keV. As is known, Auger and Coster-Kronig conversion dominates at such energies, and the efficiency of energy transfer to nuclear subsystem should be reduced. Fortunately, as we discussed above, the hot plasma condition suppresses the rate of Auger and Coster-Kronig processes. So that, even M-vacancy in heavy atom may have noticeable fluorescence yield, and the nuclear conversion of the atomic transition is not suppressed. Thus, the triggering of the ^{242m}Am isomer remains to be one the best possibilities for pulsed release of a clean nuclear energy, and the observation of the ^{242m}Am triggering is one of the challenges of modern experimental physics.

In Ref. [2], we proposed the ^{242m}Am and ^{177m}Lu isomers as new candidates for triggered release of energy, in addition to earlier discussed Hf isomers. Both nuclides are radioactive and can be produced with high yield in the reactors. ^{242m}Am is normally

accumulated in the used reactor fuel as one of the radioactive products due to the multiple neutron capture process. For the ^{177m}Lu production, one has to expose in neutron flux the special target made of the enriched ^{176}Lu isotope. The lifetime of ^{177m}Lu ($T_{1/2}$=161 d) does not allow to keep this material during the years after production ready for experiments. Thus, the experimental works with ^{177m}Lu can be performed only if some standard or high-flux reactor, as well as the laboratories for chemical isolation and mass-separation of radioactive products are available. Such requirements restrict the choice of institutions capable to develop the experimental program with ^{177m}Lu.

On the other hand, ^{177m}Lu stores 20 times larger specific energy per nucleus, than ^{242m}Am, and being triggered it has to emit this energy in a form of short-wave γ-radiation in the range of 100-200 keV. Because of the advantageous properties of ^{177m}Lu, we try to understand, are there the promising chances for the triggering it by X-ray and synchrotron radiation, or not.

The level scheme of ^{177}Lu is not well developed till now. Two rotational yrast bands with $K^{\pi}=7/2^{+}$ and $9/2^{-}$ are known, and the $19/2^{+}$ and $19/2^{-}$ members of these bands lie at an excitation energy of 1093 and 1073 keV. The isomeric ^{177m}Lu, $K^{\pi}=23/2^{-}$ state has the excitation energy of 970 keV. One may assume a possible triggering via E2 transition with the energy of 103 keV from $23/2^{-}$ to $19/2^{-}$ levels. Unfortunately, this transition would be strongly hindered by K-quantum number, because of $(\Delta K-\lambda)=5$ for it. Despite this, one may discuss that the K-mixing component of the wave-function has a noticeable amplitude for the 6-th level of the $K^{\pi}=9/2^{-}$ band with I=19/2. Comparison with the neighbouring ^{176}Lu looks natural and can be productive, since it is much better studied than ^{177}Lu in the aspect of the level spectroscopy.

The fragment of the ^{176}Lu level scheme is shown in Fig.2. Almost stable ground state has $K^{\pi}=7^{-}$, and the short-lived ($T_{1/2}$=3.7 h) isomer with $K^{\pi}=1^{-}$ lies at 123 keV. So, there are two systems of levels with high and low K-values. They can be coupled only via

some mediating levels possessing the special K-mixing wave-function. And such levels exist, as known after experiments, Refs. [23-25]. In Ref. [23] the 1^- isomer was excited in irradiations of the ^{176}Lu target by the intense radioisotope γ-radiation of ^{60}Co and ^{137}Cs sources. The monoenergetic γ-radiation of ^{137}Cs consists of 662 keV photons, and successful population of the 1^- isomer shows the presence of some K-mixing level at an energy below 662 keV. The spectroscopical identification of this level was not yet successful. However, in Ref. [25] the level with I, $K^{\pi}=5$, 4^- at 838 keV was definitely recognized as a mediating level between K=7 a K=1 levels.

Even more exotic properties have been found in Ref. [24] for the transition from I, $K^{\pi}=12$, 12^+ isomeric state at 1515 keV to I, $K^{\pi}=10$, 4^+ rotational state at 1159 keV. This transition should be strongly hindered by K, $(\Delta K-\lambda)=6$, but in experiment, rather low reduced hindrance factor has been found. So that, the wave-functions of high-K isomeric state and low-K rotational level turn out to be mixed. This is important for the hypothetical triggering of the ^{177m}Lu isomer. If the properties of ^{176}Lu and ^{177}Lu nuclei are resemble, one may expect a successful ^{177m}Lu triggering by synchrotron radiation. The experimental test looks relevant and even intriguing.

To summarize, the time-scale of different processed after pulsed release of energy are compared, and the typical time of the nuclear fluorescence appears to be longer the atomic and solid relaxation processes. The possibility of nuclear radiance in solid surroundings after the sample recrystallisation is promising for lasing of nuclear γ-rays. The nuclear excitation in hot dense plasma via the resonance between atomic and nuclear transitions also possesses some attractive properties. Among them, the suppression of Auger and Coster-Kronig conversion of the ionization energy, shift of the characteristic energies of the atomic levels, dependent on a charge state, and possible hybridization of the atomic-nuclear wave functions. The discussed processes may be useful to increase the efficiency of nuclear excitation by laser pulse. The nuclear states-candidates for the

Table 1. Time-scales of different processes induced by powerful laser pulse in solid.

	Process	Time-scale, sec.
1.	Electron-gas excitation	$\sim 10^{-15}$
2.	X-ray emission past ionization	$\sim 10^{-16}$-10^{-15}
3.	Ablation due to superthreshold power:	
	a) Fast desorption;	$\sim 2 \cdot 10^{-14}$
	b) Temperature mechanism	10^{-12}-10^{-11}
4.	Recrystallisation of melted layer	10^{-11}-10^{-10}
5.	Nuclear fluorescence:	with account conversion
	a) Collective E2 at E_γ>50 keV;	$<10^{-10}$
	b) Single-particle E1 at E_γ>50 keV	$<2 \cdot 10^{-10}$
6.	Condensation of the ablation cloud	$\sim 10^{-8}$

Table 2. Resonance between nuclear and atomic transitions and possible mechanism of excitation: PE – photoexcitation, NEET – nuclear excitation by electron transition.

Nuclide	$T_{1/2}$	Nuclear Transition	E^*, keV	Electron Transition	E_x, keV	Discussed in Ref.	Mechanism
^{98}Tc	$4.2 \cdot 10^6$ y	$6^+ \rightarrow 5^+$ M1 and E2	21.8	$N_I \rightarrow K$ M1	20.98	[20]	PE and NEET
				$N_{IV} \rightarrow K$ E2	21.04		NEET and PE
^{103}Ru	39.3 d	$3/2^+ \rightarrow 5/2^+$ M1 and E2	2.81	$M_{IV} \rightarrow L_I$ E2	2.94	[20]	PE and NEET
				$M_{II} \rightarrow L_I$ E1	2.74		PE
				$M_I \rightarrow L_I$ M1	2.64		NEET and PE
^{161}Dy	Stable	$5/2^+ \rightarrow 5/2^-$ E1	25.652	Sn $L_{III} \rightarrow K$ E1	25.28	[21]	PE
^{181}Ta	Stable	$7/2^+ \rightarrow 9/2^-$ E1	6.238	$M_I \rightarrow L_{III}$ E1	7.17	[4]	PE and NEET
^{189}Os	Stable	$3/2^- \rightarrow 5/2^-$ M1 and E2	69.537	$M_I \rightarrow K$ M1	70.82	[22]	PE and NEET
^{197}Au	Stable	$3/2^+ \rightarrow 1/2^+$ M1 and E2	77.351	$M_I \rightarrow K$ M1	77.300	[14]	NEET and PE
^{235}U	$7 \cdot 10^8$ y	$7/2^- \rightarrow 1/2^+$ E3	0.0768	Plasma Radiation	Continuos spectrum	[9]	PE
^{242m}Am	141 y	$5^- \rightarrow 3^-$ E2	4.30	$L_{III} \rightarrow L_{II}$ M1 and E2	4.44	[20]	PE and NEET
				$N_{II} \rightarrow M_{II}$ M1	4.30		PE
				$N_{VII} \rightarrow M_{III}$ E2	4.24		NEET and PE

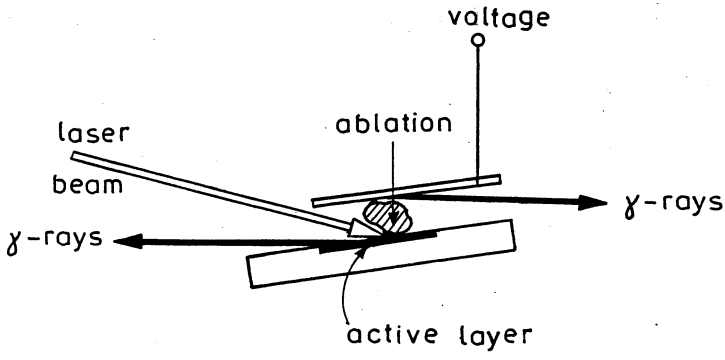

Fig.1. Schematic illustration of the nuclear level pumping by laser pulse.

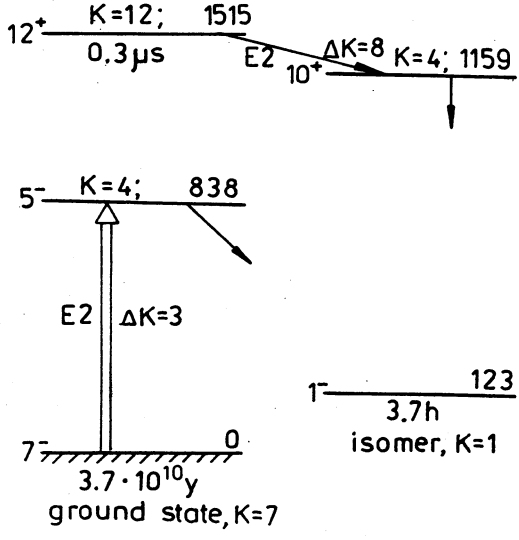

Fig.2. Mediating levels in ^{176}Lu, as can be deduced from Refs. [24,25]. Energies of levels are given in keV.

pumping by atomic transitions are characterized. Among them, there are also the nuclear isomers-candidates for triggered release of the stored nuclear energy.

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Карамян С. А., Кэрролл Дж. Дж.
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О возможности совмещения накачки ядерного уровня
в плазме с лазерным высвечиванием в твердом теле

Обсуждаются возможные схемы использования ядерных изомеров для накопления и высвобождения «чистой» ядерной энергии. В благоприятных случаях резонанс между атомным и ядерным переходами может проявляться в форме гибридизации атомно-ядерного возбуждения. Рассмотрены ядерные уровни — кандидаты на стимуляцию высвечивания посредством атомного перехода. В горячей плазме, генерируемой короткими, мощными лазерными импульсами света, возникает множество ионизационных состояний и возбужденных конфигураций атомных оболочек. В условиях горячей плазмы нерадиационная конверсия энергии ионизации внутри атома может быть подавлена. Сравнивается временная шкала различных процессов в ядерной, атомной подсистемах и в конденсированной среде. Процессы быстрой ионизации в твердом теле, эмиссии рентгеновской радиации в плазме, плавления и рекристаллизации образца могут предшествовать ядерной флуоресценции. Временная шкала (короче 0,1 нс) делает такую последовательность перспективной для группового возбуждения короткоживущих мод в ядерной подсистеме.

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Karamian S. A., Carroll J. J.
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Possibility of Combining Nuclear Level Pumping
in Plasma with Lasing in Solid

Nuclear isomers can be used for the storage and release of «clean» nuclear energy, and the visible schemes are discussed. Resonance between the atomic and nuclear transitions may be manifested in a form of the hybridization of atomic-nuclear excitation at the appropriate case. The nuclear levels — candidates for triggering via atomic transitions are described. A variety of the ionization states and atomic-shell configurations arizes in hot plasma generated by the short powerful pulse of laser light. The nonradiative conversion of the ionization energy within atom can be suppressed in the hot-plasma surroundings. Time-scales of different processes in nuclear, atomic and condensed-matter subsystems are compared. The processes of fast ionization in solid, X-ray radiance in plasma, sample melting and recrystallisation may precede nuclear fluorescence. Time-scale shorter 0.1 ns makes this sequence promising for the group excitation of short-lived modes in nuclear subsystem.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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