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К вопросу о гамма-лазере на ядерных уровнях

В свете новых результатов по получению ядерных изомеров и их использованию в качестве мишеней для ядерных и фотоядерных реакций рассмотрены физические основания для создания гамма-лазера. Рассмотрены некоторые новые возможности увеличения эффективности «накачки» ядерных уровней. В качестве предпочтительных кандидатов для рабочего вещества гамма-лазера предложены определенные изотопы и изомеры. Констатированы широкие возможности новых исследований в области стимулированной эмиссии гамма-квантов.

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On the Question of a Gamma-Ray Laser on Nuclear Levels

Physical grounds of the γ-ray laser creation are discussed in the light of new results on nuclear isomers production and their application as targets for nuclear and photonuclear reactions. Some new possibilities of increasing the efficiency of the nuclear levels pumping are also considered. Definite isotopes and isomers are suggested as favorable candidates for a γ-laser material. Wide perspectives for new research in the field of stimulated γ-emission are assumed.

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

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1. The question of a γ-laser imposes controversial demands:

a) The nuclear electromagnetic output transition should be of the Mössbauer type but not strongly converted. Thus the transition energy must fall within a certain range.

b) The natural width of the level should be comparable or greater than the broadening but low enough to provide a reasonable pumping efficiency during the interval of its lifetime.

c) A sufficiently long lifetime means a decreased matrix element for the transition which conflicts with the requirement for a maximum pumping cross section.

d) Powerful pumping may heat the sample and thus disturb the possibility for recoil-free gamma emission.

So it is clear that the key to the creation of a short-wave laser has been a search for new ideas and fundamental studies as well as a technological build up. As described two years ago [1] it may be possible to fulfil the mentioned demands a) and b) by a delicate choice of an isotope with an optimum nuclear level scheme, c) using high intensity bremsstrahlung pumping and d) by some special solid-state technology of amorphous diamond substrates.

2. Background of modern investigations

Five years ago the presence of giant cross sections was first revealed [2] for the bremsstrahlung induced (γ,γ')-reaction on the high-spin nuclear isomer ^{180}Ta^{m} ($I,K^{\pi}=9,9^{-}$). As of today many other examples of the excitation of levels with both higher and lower spins than the ground state have been studied using bremsstrahlung irradiations. These processes were suggested as an efficient way for γ-laser pumping. High-scale cross sections for pumping have now been observed [3] in many cases and are currently interpreted in terms of the existence in heavy nuclei of special K-mixed intermediate states at excitation energies of about 3 MeV. Besides the laser ideas, this problem of strong K mixing for excited nuclear levels has an independent scientific significance. In addition to the already mentioned investigations of the Dallas-Darmstadt collaboration, it is possible to find the recent papers on K-mixing studies in different nuclear processes [4-12]. The papers cited here touch the K-mixing question in spectroscopic studies of short-lived isomers [4-6] in ^{174}Hf and ^{182}Os, in studies of (n,γ), $(\gamma,2n)$ and $(\alpha,2n\gamma)$ reactions on high-spin nuclei [7-9], in polarization measurements using nuclear resonance fluorescence methods [10], and in selectivity of the level population [11,12] in the γ-cascade of (n,γ)-reaction. The number of experiments on K-mixing is not great despite their importance. Therefore, these studies should be continued using a variety of experimental approaches.

3. Pumping by short powerful bremsstrahlung flashes

Accelerators of electrons with a peak intensity of about a MegaAmpere have become increasingly more common in the last few years. They produce single bremsstrahlung flashes with durations of the order of 50 ns. Such short pulses allow the consideration of nuclear levels with lifetimes in the range of ns. In Fig.1 partial level schemes [13] are presented for ^{161}Dy and ^{169}Tm, favorable candidates for a γ-laser material when short-pulse pumping is used. In particular, the $5/2^{-}$ level in ^{161}Dy with a 29 ns lifetime has a relatively low excitation energy of 25.65 keV and only a small spin difference from the ground state. Thus, it is promising that there will be a high pumping cross section. In addition, the small energy of 25.65 keV for the output transition makes it the best Mössbauer-type transition while the conversion coefficient is somewhat small (about 2). Such properties are close to ideal ones but the lifetime is so short that this places severe limitations on the duration and power of the pumping flash.

A small excitation energy suggests the use of 30–50 keV X-ray generators or synchrotron radiation for pumping. Also, when one uses a rather long-wave radiation it may be possible to increase the efficiency of the pumping by the Bragg reflection of the radiation. The X-rays are directed into the gap between two parallel monocrystals at the Bragg angle to the surface as shown in Fig.2. Multiple Bragg reflections of the radiation lead to repeated use of the same incident flux for the pumping of the crystal nuclei. Crystals might be specially prepared for this experiment by optical class polishing in the chosen crystallographic plane. The potential for using multiple reflections of X-rays by crystals as well as by multilayer hyperlattice structures should be considered in more detail as a way to achieve a pumping-efficiency gain. The monochromatization of X-rays by the Bragg reflection might be also important.

Returning to Fig.1 one can say that ^{169}Tm is also an interesting isotope. Its $7/2^-$ level has about the same properties as the ^{161}Dy level but a lower pumping cross section can be expected. Another level in ^{169}Tm is more favorable in lifetime but its transition energy is not small enough to expect it to provide good Mössbauer radiation. The level scheme of ^{187}Os shown in Fig.3 is similar to the case of ^{169}Tm and it should also be considered as a candidate for laser emission because of the longer lifetimes of its levels.

It is possible to estimate how many nuclei could be excited by one flash of a MA electron accelerator. As parameters for the accelerator we take those for the existing "Hydramite" machine: E_e=1 MeV, peak current — 0.5 MA, and pulse duration — 50 ns. The conversion efficiency for the electrons to bremsstrahlung is about 10^{-3} and the activation efficiency, A_f [1], is assumed to be about 10^{-29} cm^2 for a level such as that in ^{161}Dy. When a sample of nominally 1 g weight is placed close to the converter, the number of excited nuclei will be about 5×10^7/flash·g. A great gain in the pumping efficiency can be achieved by increasing the electron energy. For instance, at an endpoint energy of E_m=4 MeV the conversion efficiency is 100 times higher than at E_m=1 MeV and the activation efficiency is improved by a factor of 10, so a total gain of 10^3 could be achieved. However, MA accelerators producing electron energies near 4 MeV are not yet technologically feasible.

4. Nuclear isomers as a material for γ-laser

The only isomer abundant in nature is ^{180}Ta^m and its weight samples are readily available. Also, long-lived ^{178}Hf^{m_2} can be produced on the cyclotron beam in microweight quantities according to ref.[14]. They should both have giant cross-sections for the bremsstrahlung induced inelastic scattering. Other isomers are producable in nuclear reactions in a quantity which is individually determined by the lifetime and reaction yield. The isomeric material can be considered as an already pumped one because an isomeric state is by definition an excited nuclear level. It is not easy perhaps but possible in principle to exploit the isomeric material as a pumped medium.

4a. Production of the ^{178}Hf^{m_2} longlived isomer

The unique property of the 31-year lived state in ^{178}Hf with $I,K^\pi=16,16^+$ has been known for decades. It is located at an excitation energy of E^*=2.446 MeV, below the yrast-line and thus is an yrast-trap with the longest known lifetime. Progress in investigations with this isomer became possible two years ago when an efficient method for its production in microweight quantities was developed [14]. The ^{178}Hf^{m_2} production method will be described here in brief as an example of how the task of any isomer production can be solved.

For the production of the isomer it is necessary to choose the producing reaction which is optimum with respect to both the absolute yield and the isomeric-to-ground state ratio as well as to arrange intensive long irradiations. After a series of experiments on different beams from γ-rays up to ^{12}C ions we stopped upon the producing reaction ^{176}Yb(^{4}He,2n)^{178}Hf^{m_2}. The cross-section and excitation function of this reaction were measured and presented in fig.4. The stable isotopes ^{178,177,176}Hf excitation functions were calculated using the statistical model code. So the isomer-to-ground state ratio σ_{m_2}/σ_g could be determined as a function of ^{4}He-ion energy. The known values of σ_{m_2}/σ_g for the ^{178}Hf^{m_2} production are presented in fig.5 and their systematical dependence versus a maximum angular momentum of a projectile is seen. At the case of an α-induced reaction the σ_{m_2}/σ_g value reaches 3–6% meanwile the cross-section is about 5–9 mbarns in an optimum energy range near 28–36 MeV. In the case of a 36 MeV ^{4}He ions taking into account the energy losses we can estimate that the ^{176}Yb$_2$O$_3$ target thickness of about 90 mg/cm^2 is the optimum one.

The absolute production yield of about $3\cdot10^8$ isomeric atoms per second was achieved in the irradiations on the external beam of the U-200 cyclotron at FLNR JINR. The maximum intensity of a 36 MeV ^{4}He^{++} beam was restricted on the level of 100 μA by the target instability. A special design of the Yb$_3$O$_3$ target on a cooled substrate was worked out in order to solve the problem of long expositions to the powerful beam. The effective beam time used for the production of the ^{178}Hf^{m_2} isomer in the course of three years totals to about 2500 h. There have been produced more than $1.5\cdot10^{15}$ atoms of the isomer, i.e. about 0.5 μg.

A very important question is the purity of the produced isomeric material both from ballast activities and weight contaminations by widely spread elements. The high enriched isotope ^{176}Yb (99,998%) was produced on the mass-separator at Orsay specially to eliminate the activities of ^{172}Hf and ^{175}Hf in the hafnium fraction. The clean backing materials and chemically purified ^{176}Yb$_2$O$_3$ oxide were used for the cyclotron target preparation. After irradiation by a ^{4}He-ion beam there were observed in the Yb target the following long-lived induced activities: ^{7}Be, ^{22}Na, ^{46}Sc, ^{51}Cr, ^{54}Mn, ^{56}Co, ^{57}Co, ^{58}Co, ^{65}Zn, ^{88}Y, ^{88}Zr, ^{134}Cs, ^{139}Ce. In the chemical isolation of the hafnium fraction all these activities were washed out. It means a complete purification of the hafnium fraction from ballast activities and also from many weight contaminants with different chemical properties.

Finally prepared ^{178}Hf^{m_2} sources and targets for nuclear reactions studies were deposited on a pure Be, C, Al and quartz substrates and analyzed for the impurities by a variety of methods using activation, charged particle scattering and X-ray fluorescence techniques. In the framework of the international collaboration which had been organized to study the ^{178}Hf^{m_2} isomer the nuclear reactions (n,γ), (d,d$'$), (p,t) and the Coulomb excitation by the ^{208}Pb ions were observed successfully [7,15–17] with using these isomeric targets. Also, a hyperfine interaction experiment was accomplished [18] by the method of laser-induced resonance spectroscopy of atomic levels.

4b. Pumping from isomeric state

As has been already mentioned above a giant cross-section for the (γ,γ')-reaction on the ^{180}Ta^m isomeric target was observed [2] five years ago. This means that a variety of levels in ^{180}Ta were excited after the bremsstrahlung capture. Among them a favorable for laser radiation state can be populated, but the concrete one has not been revealed yet due to experimental difficulties. The partial level scheme of the ^{178}Hf is shown in Fig.6.

The ^{178}Hf^{m_2} nucleus is very interesting for the γ-laser problem because one can expect a high cross-section for the pumping by bremsstrahlung for the level with $I,K^\pi=14,14^-$,

E^*=2.574 MeV and $T_{1/2}\doteq 68$ μs as well as for the level with $I,K^\pi=6,6^+$, E^*=1,544 MeV and $T_{1/2}$=78 ns. The output transitions from these levels are too energetic for a stimulated γ-emission although they are convenient for experimental studies of the pumping cross-section. In the framework of the Dubna-Dallas collaboration a first experiment is being prepared to observe the process of pumping short-lived excited nuclear levels starting with an isomeric state.

In general, pumping from a long-lived, high-K isomer possesses significant advantages including an easier population of short-lived isomeric levels with any K values. In addition, there exists a possibility of using a rather soft radiation since the intermediate levels with mixed K should lie close in energy to the initial isomeric state. This could increase the pumping efficiency orders-of-magnitude with respect to the pumping from the ground state.

Up to now we considered the γ-laser as a nuclear analog of ruby laser. One can suggest another scheme — a variant of laser amplifier, i.e. the device in which an external radiation stimulates a coherent emission in the pumped medium. This scheme is analogous to a subcritical nuclear reactor stimulated by an external beam. In both cases the avalanche process is induced by some impact agent.

4c. Stimulated isomeric transition

One can consider the nuclear isomeric sample as a pumped medium and the question is what radiation should be used for the stimulation of the isomeric transition. The X-ray or bremsstrahlung radiation with high enough spectral density is a possibility. Modern MA electron beam accelerators should be tested in such experiments.

The natural widths of the long-lived isomeric states such as $^{180}Ta^m$ and $^{178}Hf^{m_2}$ are too small and the efficiency of the γ-ray stimulation by the bremsstrahlung spectrum is not high. Short-lived states are more favorable for the γ-laser emission. That is in contradiction with an achievable quantity of isomeric nuclei. The duration of the production runs on the accelerators is usually not longer than one month. Hence, the lifetime of the radioactive nucleus should be not shorter than 10 d, otherwise the number of produced nuclei will be restricted on the equilibrium level: $N_{equ} \sim T_{1/2}$, by the radioactive decay.

The isomer $^{177}Lu^m$ ($I^\pi=23/2^-$, $T_{1/2}$=160 d) should be considered as a favorable example. The milligram quantities the $^{177}Lu^m$ can be produced in neutron irradiations of the ^{176}Lu gram specimen on a powerful reactor. The isomeric transition with an energy E_γ=116 keV has moderate Mössbauer properties but the conversion coefficient is high enough. It is important to mention now that the limitations for the conversion coefficient are not valid in the case of the stimulated isomeric transition. The emission will be induced by short pulses which are much shorter than the electron conversion half-life, and the last process is not compatible.

Thus, a variety of isomers with low transition energies $\leq$50 keV can be considered as possible candidates for a stimulated depopulation. Also, there is a possibility to consider the synchrotron radiation as an impact agent. The synchrotron radiation is coherent with narrow angular distribution, so its quality is much better with respect to the bremsstrahlung. Attempts to observe the γ-ray emission stimulated by the synchrotron radiation were already described in literature [19,20]. One can assume wide perspectives for future research on the stimulated emission of γ-rays.

5. Pumping by the heavy-ion induced Coulomb excitation

The advantage of a heavy-ion beam as a pumping agent lies in the localizaton of pumped nuclei in a small volume as is preferable for the stimulated emission. The application of heavy ions in the variant of short flashes is restricted by the maximum achievable current in modern accelerators as well as by heat-deposition limitations. With this in mind we will consider the excitation of levels with lifetimes of the order of seconds. In Fig.3 the level scheme is presented for the ^{73}Ge nucleus. The $1/2^-$ level having a lifetime of 0.5 s has a low excitation energy of 66.73 keV and decays by a weakly converted transition with good Mössbauer properties.

Let us estimate the efficiency for pumping by the Coulomb excitation for the isomeric level in ^{73}Ge. The Coulomb excitation of the 8^- isomer in ^{178}Hf was observed recently in ref.[21] with good enough cross-section. The ^{132}Xe ion with an energy of 550 MeV just below the Coulomb barrier for the nuclear interaction is a powerful projectile for this type of excitation. One can expect that a complete set of levels of any isotope below 2 MeV will be excited by Xe ions in multiple Coulomb excitation processes. The level can be fed by the decay from upper levels even if it has an individually small cross section for excitation. This may be the case for the $1/2^-$ isomeric level in ^{73}Ge. To estimate the excitation cross section we assume an effective $B(E2\uparrow)$ of about one single-particle Weisskopff unit, a moderately low estimation for the excitation of an eventual level. So, using the first order perturbation theory one can immediately evaluate the cross-section to be $\sigma_{C.E.} \approx 0.4$ b. This estimation gives only a realistic scale of the value not an accurate quantity. The cross-section for the Coulomb excitation can be measured easily for the $1/2^-$ level in ^{73}Ge, but at this time no such measurements have been made and we must use the estimated cross section. Energy losses for the Xe ions in matter are large. A drastic decrease of the cross section accompanies a decrease in ion energy so the average cross section value should be about $\bar{\sigma}_{C.E.} \approx 0.2$ b for a layer of Ge having a 5 μm thickness.

A flux of about 10^{12} Xe ions/s per cm^2 cross area of the beam is a reasonably good intensity for a modern heavy-ion accelerator. So after a 1 s exposure of an isotopically enriched ^{73}Ge to the such a beam about 5×10^6 nuclei will have been pumped within a 5×10^{-4} cm^3 volume on the surface of the ^{73}Ge sample. This number of atoms is one order-of-magnitude lower than that estimated for the bremsstrahlung pumping using the "Hydramite" accelerator. However, the weight concentration of excited nuclei is 40 times more in the case of the Coulomb excitation. A surface localization of the pumped nuclei satisfies easier the demand that the thickness of the emitting layer is to be about one absorption length for the radiation. The scheme of heavy-ion pumping and radiation emission is shown in Fig.7. The Ge is a favorable material because it exists in the form of perfect monocrystals having a high Debye temperature (743 K).

The condensed matter state of the sample is also very important for the stimulated emission in addition to nuclear properties. Thus, it is necessary to discuss the behavior of the Ge sample under heavy-ion beam. The macroscopic heating of the sample by the 100 W/cm^2 beam during a 1 s pulse could be small enough if a thin (0.1 mm) Ge plate is fixed by heat conductive glue on a cooled holder. The nontrivial question is what damage of the monocrystalline structure is expected. Fortunately this was studied experimentally [21] about 5 years ago and anomalously low radiation damage was revealed in Ge crystals exposed to Xe ions. This new result was interpreted in terms of the perfect recrystallization of each track produced by each Xe ion in the Ge. This process is developed on a ps time scale, so the Ge crystal conserves its perfect structure. For the stimulated γ-emission it is preferable to have a perfect lattice of pumped nuclei because the emission along a crystallographic direction may have a higher probability.

In principle Coulomb excitation cross sections can be two orders-of-magnitude higher than the estimate but normally only for collective-rotational states which have lifetimes lower than 2 ns. The approach to such short lived levels is not available at the moment.

6. Ultraviolet laser on nuclear levels of ^{229}Th

The isotope ^{229}Th ($I^\pi=5/2^+$, $T_{1/2}=7.34\cdot10^3$y) is available in a weight quantity as a daughter of ^{233}U. Its first excited state ($I^\pi=3/2^+$) has an unique low excitation energy $E^*\doteq(4.5\pm1)$ eV according to measurements [23]. The lifetime of the $3/2^+$ level is estimated to be about 20 h due to an electromagnetic decay. Internal electron conversion is impossible because of the small energy of radiation, however, a nonradiative transfer of the energy on the atomic shell excitation is possible. The probability of such a process can be high when the energies of atomic and nuclear transitions are coinciding. Otherwise, it should be of the same order of magnitude as the probability of the ultraviolet radiation absorption by the atom. The shortening of the level lifetime and decrease of the radiation yield due to a nonradiative transition are non easy to calculate with a good accuracy.

Such a level with an energy of an ultraviolet range $E^*=4.5$ eV can be pumped at temperatures of about 10^4K, i.e. using a regular technique applied for the atomic level pumping. The cross-section of the pumping can be different but its scale is determined by the radiation wavelength both for the atomic and nuclear levels. So, the medium with an inverse population of nuclear levels in ^{229}Th can be created and laser radiation should follow that. No principle restrictions are visible at the moment. Being created, this laser will be not a γ-laser but only ultraviolet one, however, it has a principle significance because it will be a first laser on the nuclear levels.

Summary

One can assume that the great difficulties on the way to a γ-ray laser realization can be overcome after extensive works in such directions as using of isomeric materials, pumping of nuclear levels by short bremsstrahlung flashes or by synchrotron radiation or even by heavy-ion beam and also application of a precision crystallographic technology and Bragg-reflections of X-rays for their monochromatization and pumping-efficiency multiplication. Some physical recommendations and suggestions are considered in the present paper. The key idea is, perhaps, not found yet but it can arise when experimental studies will be performed. At least the creation of the ultraviolet laser on first excited level (4 eV) in the ^{229}Th nucleus looks pretty realistic from a physical point of view (not touching upon the technical details).

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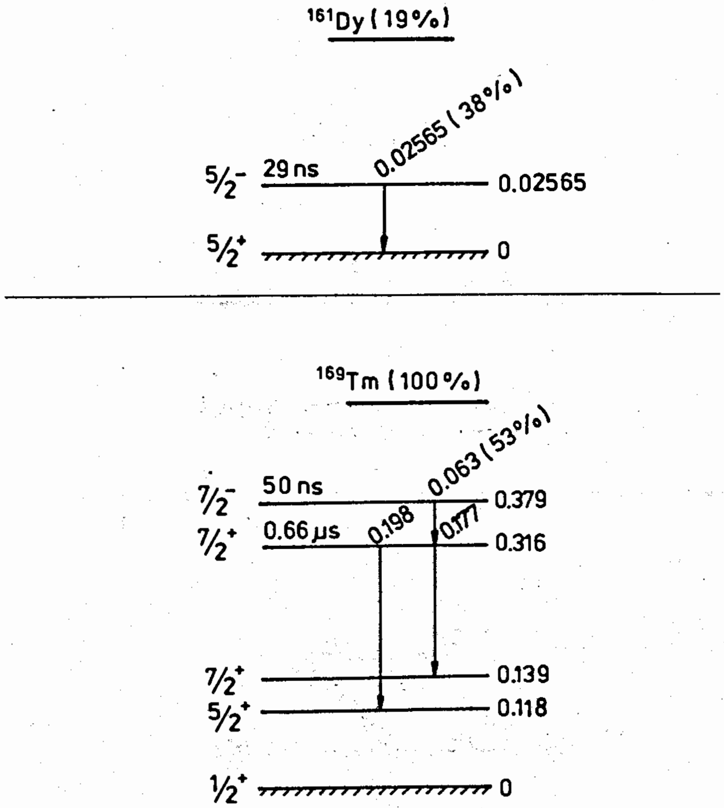

Fig.1. Fragments of level scheme for ^{161}Dy and ^{169}Tm isotopes. The natural abundance of isotopes and energy and yield of γ-ray for the isomeric transition are given.

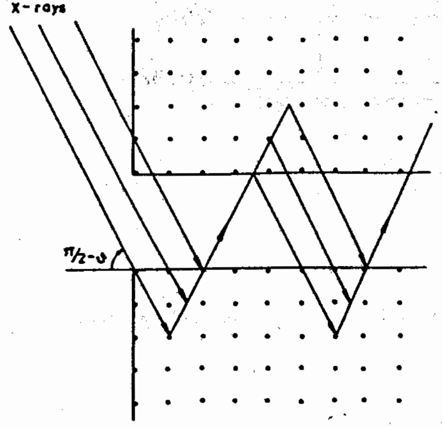

Fig.2. Schematic picture of a multiple Bragg-reflection by parallel crystal layers; ϑ is a Bragg angle.

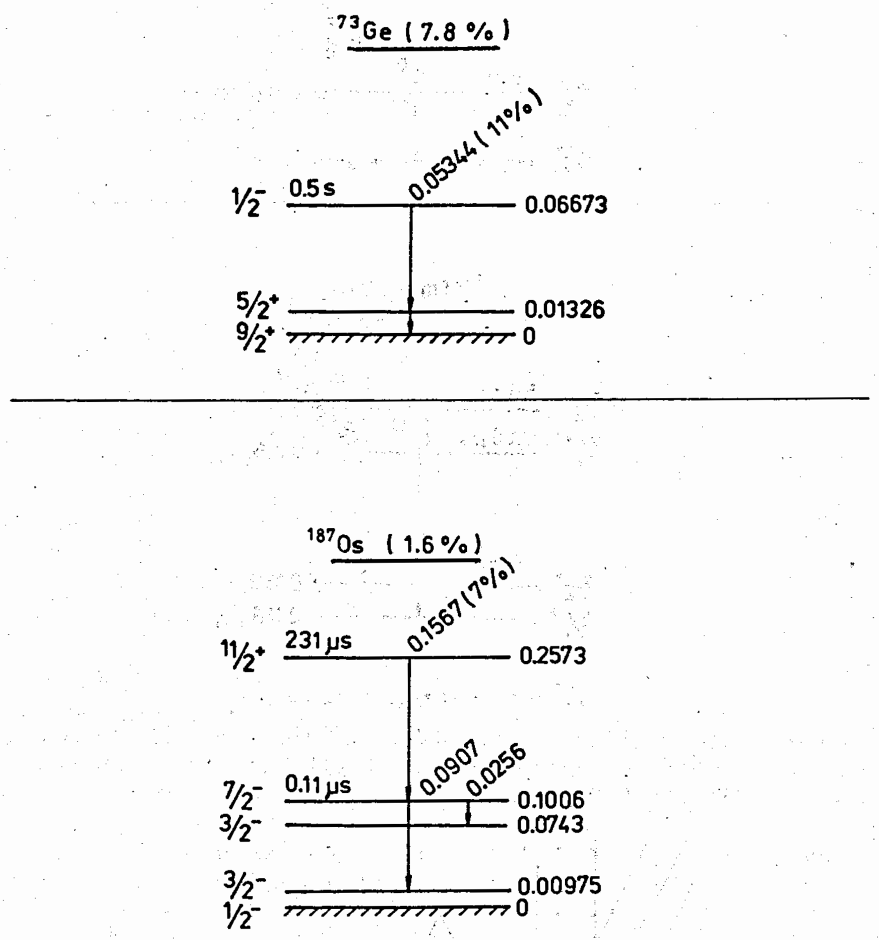

Fig.3. Same as in fig.1 for ^{73}Ge and ^{187}Os isotopes.

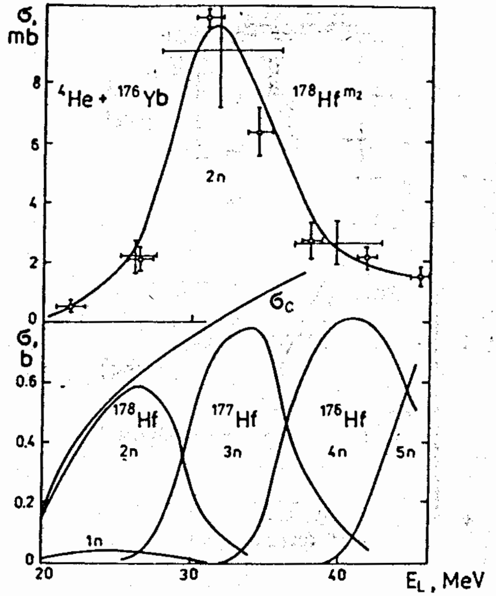

Fig.4. Excitation functions for ^{178}Hf^{m_2} isomer (measured) and ^{178,177,176}Hf stable isotopes (calculated) produced in the nuclear reaction ^{176}Yb(^{4}He,xn).

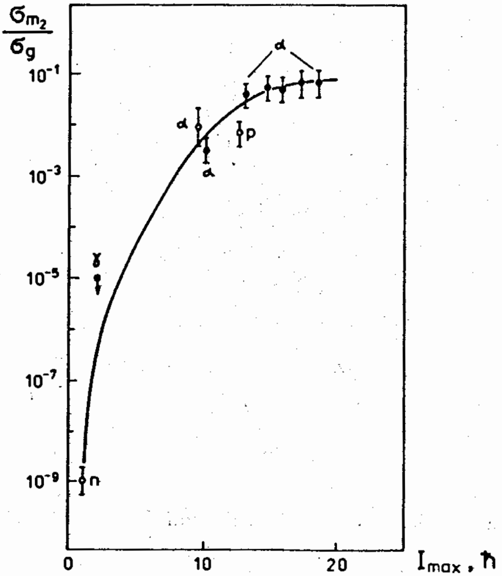

Fig.5. Isomeric to ground state ratio σ_{m_2}/σ_g as a function of the incident particle maximum angular momentum I_{max}. Closed circles show the measured values, opened ones are taken from literature.

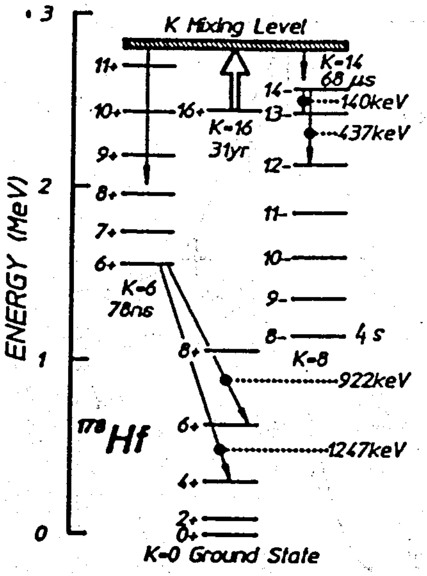

Fig.6. Partial level scheme of the ^{178}Hf nucleus.

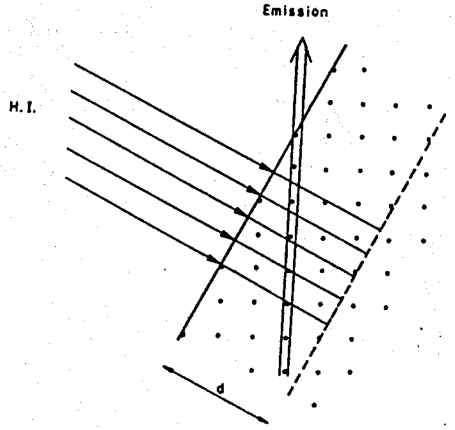

Fig.7. The scheme of the pumping and γ-ray emission in a crystal excited by a heavy-ion beam; d is a pumping thickness.

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