^{176}Lu isotope properties as a cosmothermometer

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The ^{176}Lu properties as a cosmochronometer and a cosmothermometer are studied, combining Coulomb excitation with the recoil method. The results confirm that this isotope should be dismissed as a reliable cosmochronometer. On the other hand, this nucleus synthesis takes place, in all probability, at temperatures between 2.35_{10} and $3.6_2 \times 10^8$ K. The assignment of the levels reached leads to some difficulties as to their reduced transition probabilities.

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I. INTRODUCTION

The ^{176}Lu level scheme has been studied in many works, experimentally through (n,γ), (d,p), (p,n), (t,α) reactions [1–5] (and references therein), and theoretically [6,7]. This isotope, whose ground state, $g[I^{\pi}=7^-]$, has a very long half-life ($t_{1/2}=3.78_5\times 10^{10}$ y), is screened for the r process, ^{176}Yb being stable, and is thus synthesized only through slow neutron capture by ^{175}Lu (s process). These characteristics award cosmochronometer and cosmothermometer properties to the ^{176}Lu nucleus [8,9]. This nucleus also possesses an isomeric state, $m[I^{\pi}=1^-]$, with a rather short period ($t_{1/2}=3.635_3$ h). Therefore, the ^{176}Hf synthesis happens only through the ^{176m}Lu production, the long ^{176g}Lu half-life enhancing mainly the ^{177}Lu formation through neutron capture.

In a stellar environment many ^{176}Lu levels can be excited, namely by nuclear resonant fluorescence (NRF). Some of these levels may then be connected to the isomeric state through one or several deexcitation cascades. The levels directly fed act as ''relay'' levels. A quick destruction of the ^{176}Lu isotope may then occur [10,11]. In that case, the use of ^{176}Lu as a cosmochronometer needs a great deal of caution.

The ''relay'' level with the lowest energy would be at 838.640 keV (5^-) and belong to the rotational band built on the $\{\pi 1/2^+[411]\otimes \nu 7/2^-[514]\}_{4^-,K=4}$ state, with a half-life 2.5 ps$\leqslant t_{1/2}\leqslant$300 ps [2], or 10.0 ps$\leqslant t_{1/2}\leqslant$300 ps [3], and a branching ratio to the isomer between 0.022 and 0.055.

We strived to study these two properties of ^{176}Lu with a new method, combining Coulomb excitation and the recoil technique, in order to try to improve the precision on the half-lives of the excited relay levels and to compare our results to those obtained by other methods.

II. ANALYSIS OF EXPERIMENTAL METHODS

A. Nuclear resonant fluorescence

Two techniques can be used for this process (NRF), both making use of a strong photon flux coming either from radioactive sources or from a bremsstrahlung radiation, near an electron accelerator, with an energy between zero and a maximum value. The produced activity is measured off line. However, the smallness of the cross section must be balanced by a very strong photon flux and a thick target. Then, the natural radioactivity of ^{176g}Lu may interfere with the produced radioactivity (detection of the transition deexciting the first 2^+ state of ^{176}Hf, at 88.35 keV) and reduce the accuracy of the method [12–14].

B. Coulomb excitation and recoil method

Through Coulomb excitation, E_i energy projectiles can excite, among others, some relay levels of the target nuclei, at energy E_j^*, liable to decay partly to the isomer. By changing the projectile, or its energy, it is possible to excite a more or less widely spread amount of levels. Here also, the measurement off line will ascertain whether the isomeric state has been reached. To avoid the disadvantages mentioned before, Coulomb excitation is combined with the recoil method [15]. The catcher only collects the nuclei ejected from the target. The natural activity of the latter does not disturb the measurement, the sensitivity of which is then much enhanced.

One or several relay levels may be excited. When there is only one relay level, at energy E_j^*, and working with several projectile energies E_i, the measured value of the cross section $\sigma_m(E_i,E_j^*)$ for producing the isomer allows to estimate the energy E_j^* of the relay level together with the quantity $r_{jm}B(E\lambda)_{gj}$; $B(E\lambda)_{gj}$ is the reduced transition probability of the ground state g towards the j level and $r_{jm}=I_{jm}/I_{\text{total}}$ is the branching ratio of the j level deexcitation to the isomer m, directly or through several cascading transitions. For several relay levels, a set of equations is obtained, namely,

$$\sigma_m(E_i)=\Sigma_j \sigma_m(E_i,E_j^*).$$

Some difficulties are also encountered with this method as

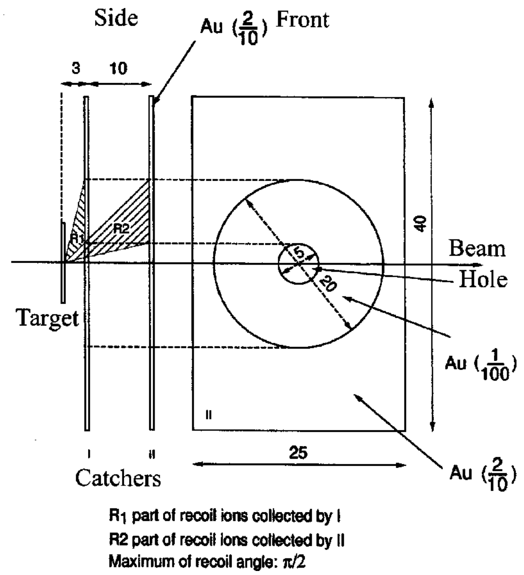

FIG. 1. Simplified scheme of the target and double catcher set-up (front and side view); the recoil ion cones are shown.

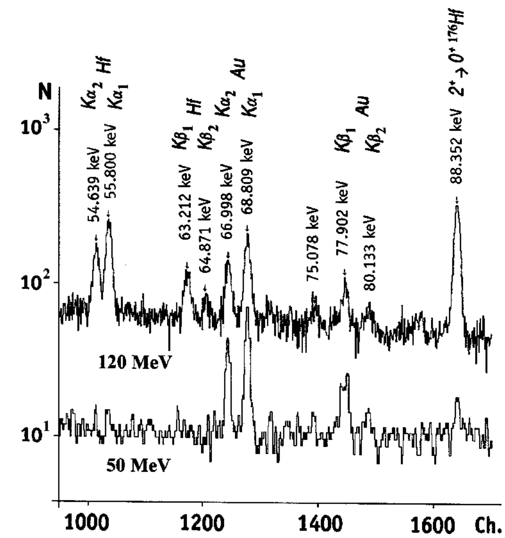

FIG. 2. Eight hours γ spectrum collected by the X detector for the 50 and 120 MeV irradiations.

the escape of the recoil nuclei needs a very thin target and the energy of the level(s) reached can only be determined, as in the NRF case, with a limited accuracy.

III. EXPERIMENTAL TECHNIQUES

The incident ions (^{32}S) are accelerated at sub-Coulomb energies of 50, 60, 70, 80, and 120 MeV (0.8 times the Coulomb barrier) by the Tandem accelerator of IPN (Orsay). The effective diameter of the beam is of the order of 1 mm.

The target, made of 500 μg/cm^2 of lutecium, 99.5% enriched in ^{176}Lu, on a 30 μg/cm^2 carbon foil, is however slowly damaged by the beam impact, making the monitoring difficult, with a Faraday cup for instance. To reduce this effect and distribute evenly the beam impact on it, the target experiences externally driven translation and rotation motions. The catchers are gold disks (0.01 mm thick) with a 20 mm diameter and a 5 mm diameter hole to let the beam through. They are, respectively, situated at 3 and 10 mm from the target. This short distance is due to the large emission angle of recoil nuclei, contrary to what happens when the recoil products follow a fusion reaction (Fig. 1). The catchers are set in a 0.2 mm thick gold frame (width = 25 mm, height = 40 mm) located in a cubic chamber on the beam line. This frame can be easily extracted through a load lock and taken to the detection area.

Another monitoring mode than the Faraday cup is also used, based on the Coulomb excitation of the first two states of the rotational band built on the ^{176}Lu ground state. The intensity of the three γ rays deexciting these levels, with well-known reduced transition probabilities, is measured with a 20% efficiency Ge detector, set at 30 cm from the target and at 55° with respect to the beam direction.

The target is irradiated during about two periods of the isomer for each chosen energy; the catchers are then removed and placed in front of a thin LeGe detector, with a 150 eV resolution at 6 keV. This detector is set in a 10 cm thick cylindrical iron shielding so that the remaining background is very low. A succession of eight measurements, of one hour each, is made to follow the decay of the ^{176}Hf X rays and 88.35 keV γ ray. These spectra are then summed up for analysis.

A simulation, taking into account the various parameters of the recoil ion kinematics, has been performed in order to determine the collection and detection yield for the different energies of the ^{32}S incident ions. The total yield seems to remain constant within better than 15% for the various beam energies E_i.

IV. EXPERIMENTAL RESULTS

The spectra associated to the 50 and 120 MeV irradiations are displayed in Fig. 2. These data (γ and X ray energies, half-life, conversion coefficients, . . .) show, beyond doubt, that the ^{176}Lu isomeric level (1^-) is actually reached.

The formula used to extract the cross section value, monitored on the ^{176}Lu Coulomb excitation is [16]

$$\sigma_m^{\exp}=\frac{\lambda t_i N^c_{\gamma 88}(1+\alpha_{T88})(1-N^c_\gamma \tau_\gamma)\epsilon_{\gamma 184}(\sigma_{8^-}+\zeta\sigma_{9^-})}{\beta(\%)(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})e^{-\lambda t_m}(1-N^c_X\tau_X)\epsilon_{\gamma 88}N^c_{\gamma 184}(1+\alpha_{T184})}, \quad (1)$$

TABLE I. Average experimental cross-section values for the production of the isomer at different beam energies.

E_i (MeV)	50	60	70	80	120
$\overline{\sigma_m^{\rm exp}}$ (mb)	0.0027_9	0.0294_{20}	0.113_9	0.373_{31}	6.31_{35}

where λ is the radioactive constant of the isomeric level, t_i, t_c, and t_m the irradiation, counting, and manipulating times, $N^c_{\gamma 88}$ the number of 88.35 keV γ transitions counted by the X (LeGe) detector, α_{T88} the total conversion coefficient of the 88.35 keV transition, $1-N^c_X\tau_X$ and $1-N^c_\gamma\tau_\gamma$ the dead time corrections, $\epsilon_{\gamma 184}$ and $\epsilon_{\gamma 88}$ the efficiencies of the monitoring detector at 184 keV and of the X detector at 88 keV, $\beta(\%)$ the percentage of the isomer decay to the 2^+ level of ^{176}Hf, $N^c_{\gamma 184}$ the number of 184 keV γ rays, between the ^{176}Lu 8^- and 7^- states, detected by the monitor, σ_{8^-}, σ_{9^-}, ζ, and α_{T184} the excitation cross sections of the 8^- and 9^- levels, the intensity of the deexcitation of the 9^- level to the 8^- one with respect to its total deexcitation and the total conversion coefficient of the 184 keV transition.

The cross sections σ_{8^-} and σ_{9^-} are computed with the Winther and de Boer program [17]. The values of the reduced transition probabilities, branching ratios, quadrupole moments, etc., are either taken from the tables [18] when these quantities have been measured, or calculated from the rotational model otherwise.

For beam energies between 50 and 120 MeV, nine independent measurements of the cross section corresponding to the production of the isomer have been performed. The average values $\overline{\sigma_m^{\rm exp}}$ for each incident energy E_i, which will be used hereafter, are listed in Table I. A simple calculation shows that a beam energy of 40 MeV would lead to a $N^c_{\gamma 88}$ value near 5, ruling out such a measurement.

V. ANALYSIS OF THE RESULTS

A. Coulomb excitation

The $E2$ excitations only will be considered here. The perturbation theory may be used to calculate Coulomb excitation for low excitation probabilities [19]. The results are yet approximate and a correction ϵ_{ij} must be added to the value of the $f_{E2}(\xi_{ij})$ function used in this calculation. It is deduced from the Winther and de Boer program [17]; it depends on E_i and E_j^* and can reach about 20% at most. The quantity

$$f'_{ij}=(1+\epsilon_{ij})f_{E2}(\xi_{ij}) \tag{2}$$

will be used hereafter.

The cross section for the isomer production through the E_j^* relay level is

$$\sigma_m(E_i,E_j^*)=r_{jm}\sigma_{C.E.}(E_i,E_j^*) \tag{3}$$

and for several relay levels

$$\sigma_m(E_i)=\Sigma_j r_{jm}\sigma_{C.E.}(E_i,E_j^*). \tag{4}$$

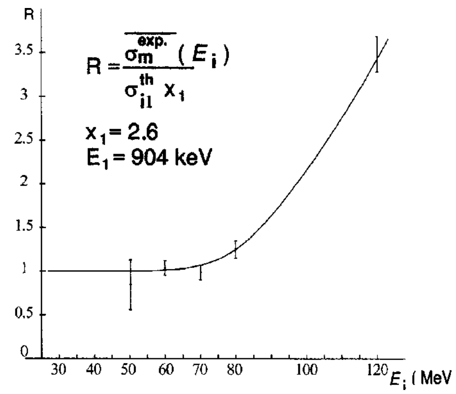

FIG. 3. Variation of $R=\overline{\sigma_m^{\rm exp}}(E_i)/\sigma_{i1}^{\rm th}x_1$ with E_i for $x_1=2.6$ e^2 fm^4 and $E_1^*=904$ keV. The effect of the higher energy levels is visible for $E_i>70$ MeV.

The magnetic excitations are strongly hindered with respect to the electric ones. Electric dipolar excitations would only lead to levels with an angular momentum $I\geqslant 6^+$, needing at least three transitions to reach the isomer.

For calculation convenience, we set

$$x_j=r_{jm}B(E2)_{gj}/B(E2)_{\rm th}$$

and

$$\sigma_{ij}^{\rm th}=C_{E2}[E_i-(1+A_1/A_2)E_j^*]B(E2)_{\rm th}f'_{ij}\ \ {\rm b},$$

$B(E2)_{\rm th}$ being an arbitrary value chosen equal to 6 e^2 fm^4 and C_{E2} a constant [19].

Equations (3) and (4) can then be written

$$\sigma_m(E_i,E_j^*)=\sigma_{ij}^{\rm th}x_j \tag{5}$$

and

$$\sigma_m(E_i)=\Sigma_j\sigma_{ij}^{\rm th}x_j\,. \tag{6}$$

1. Excitation of a single relay level

Through the 50, 60, and 70 MeV measurements only, one is likely to excite mainly the relay level with the lowest energy, or, at most, some relay levels very close to this lowest energy. Hereafter, the expression ''group of relay levels'' will be used if it is impossible to identify a single relay level from several ones very close in energy. For a single group of relay levels, characterized by x_1 and E_1^*, the minimization of the χ^2 value related to the three nonlinear equations

$$\sigma_{i1}^{\rm th}x_1=\overline{\sigma_m^{\rm exp}}(E_i)\ \ \text{with}\ \ E_i=50,\ 60,\ 70\ \text{MeV},$$

where the $\overline{\sigma_m^{\rm exp}}$ values and the corresponding errors (rms) are the weighted mean values of Table I, leads to the following result:

$$x_1=2.6_9,\quad E_1^*=904_{50},\quad \chi^2=0.6.$$

Using these data, in Fig. 3 are plotted the values of the ratio

$$R=\frac{\overline{\sigma_m^{\rm exp}}(E_i)}{\sigma_{i1}^{\rm th}x_1}. \quad (7)$$

This ratio should stay equal to unity, within errors, if there is only one group of relay levels around 900 keV. It is obvious from Fig. 3 that other relay levels interfere at 80, and even more at 120 MeV. On the contrary, the contribution of the high energy relay levels is likely to be negligible at 50 and 60 MeV, and within the errors (8%) at 70 MeV. Taking these levels into account will only induce a slight change in the x_1 and E_1^* values already obtained.

2. Excitation of several groups of relay levels.

To solve this problem one must consider the system (6) as an integral equation and inverse it to deduce the x_j values. Such a computation has been performed [16]. It shows that, within the accuracy of our measurements, there is no relay level lower than 835 keV and that the lowest energy group of relay levels corresponds to the values

$$x_1=2.2_9 \quad \text{and} \quad E_1^*=885_{50}\ \text{keV}.$$

B. Comparison with previous results

It is possible to connect the

$$835\ \text{keV}<E_j^*<935\ \text{keV} \quad \text{and}$$

$$\Sigma_j r_{jm}B(E2)_{gj}=13_5\ \ e^2\ \ \text{fm}^4$$

solution to the 838.640, 870.003, and 921.472 keV levels mentioned in the literature. The second one can be identified with a γ vibration level observed, in Coulomb excitation, around 870 keV [1]. This 5^- level, strongly bound to the ground state, would be due to the coupling $\{\pi 7/2^+[404]\otimes\nu 7/2^-[514]\}_{7^-,K=7}\otimes 2^+$. It might deexcite to the state of configuration $\{\pi 7/2^+[404]\otimes\nu 3/2^-[512]\}_{5^-,K=5}$ by a rather low energy unobserved transition, this last level deexciting to the isomer through several cascades. However, according to [1], the 870.003 level does not feed the isomer.

The 5^- level at 838.640 keV is interpreted as collective and built on the $\{\pi 1/2^+[411]\otimes\nu 7/2^-[514]\}_{4^-,K=4}$ state, at 722.921 keV, from the (t,α) reaction results [5]; the same character holds for the 921.472 keV one built on the $\{\pi 7/2^+[404]\otimes\nu 1/2^-[510]\}_{4^-,K=4}$ at 788.219 keV, seen in (d,p) reactions. Both levels have very similar deexcitation modes (same r_{jm} values). In this case there is a K interdiction $(n=|\Delta K-\lambda|=1)$, and also an overlap interdiction $\Psi(\pi 1/2^+[411])\rightarrow\Psi(\pi 7/2^+[404])$, inconsistent with the observed results [16].

VI. DISCUSSION

A. Electromagnetic transitions

Let us first consider the case of a single known level at 838.640 keV. As

$$B(E2)_{jg}=15/11B(E2)_{gj},$$

we get

$$r_{jm}B(E2)_{jg}=18_7\ \ e^2\ \text{fm}^4.$$

The r_{jm} parameter depends on the deexcitation intensities of the j level to the isomeric and ground states. According to [2], r_{jm} lies in between 0.069 and 0.117, i.e. $\overline{r_{jm}}=0.093_{24}$, whereas in Ref. [4] $r_{jm}=0.17_3$, the expected values of the conversion coefficients α_T being used in both cases. An average value $r_{jm}=0.14_5$ is taken and, similarly, $r_{jg}=0.74_7$ for the 838.640 keV transition. So, one obtains

$$t_{1/2j}=6.0^{+10.0}_{-4.0}\times 10^{-12}\ \ \text{s}.$$

The Ref. [2] $(2.5\times 10^{-12}\ \text{s}\leqslant t_{1/2j}\leqslant 300\times 10^{-12}\ \text{s})$ and [3] $(10^{-11}\ \text{s}\leqslant t_{1/2j}\leqslant 300\times 10^{-12}\ \text{s})$ results are consistent with ours.

The following conclusions are reached: either the 838.640 keV (or 921.472 keV) level has the 6 ps half-life, in disagreement with the assumed configurations [16]; or a third level exits, which could be the collective level at 870.003 keV with a short half-life, but it should then deexcite to the isomer.

If several levels are involved, $r_{jm}B(E2)_{gj}$ is an average and it is difficult to give a precise $t_{1/2j}$ value for each of them.

B. ^{176}Lu as a cosmothermometer

If, beside the g and m levels, one or several other ones are located between them and the relay level(s), it is possible to solve the system of differential equations $dn_r/dt=f(\lambda_{rs},\lambda_{sr},E_{rs},T)$, with the Ref. [20] notations, and calculate the branching factor

$$f_n=\frac{\Sigma n_i\lambda_n^{(i)}}{\Sigma_i n_i[\lambda_n^{(i)}+\lambda_\beta^{(i)}]}. \quad (8)$$

As the typical conditions for the s process, $\lambda_\beta^{(g)}\ll\lambda_n^{(g)}$ and $\lambda_n^{(m)}\ll\lambda_\beta^{(m)}$, are fulfilled one finds, for a single relay level j and a population in equilibrium between isomeric and ground states, an expression similar to (26) in [2].

It should be noted that the $\lambda_j r_{jm}(1-r_{jm})$ factor is directly deduced from the $r_{jm}B(E2)_{gj}$ measurement. It is possible to give a rather precise value for this term [identical to $\tau_1 V(1-V)$] in [2]:

$$\lambda_1 r_{1m}(1-r_{1m})=12_6^{10}\times 10^9\ \ \text{s}^{-1}$$

for one (several) level(s) at 885_{50} keV.

The shape of the curve giving f_n looks very much like that of Fig. 5 in reference [2]. For the maximum values of the B and λ_n parameters, the curves of [2] and ours are identical beyond 3.3×10^8 K, whereas for the lower values of the same parameters there is a shift of barely 0.1×10^8 K towards the low temperatures. The more crucial errors are those on B, E_j^*, and λ_n. The necessary temperature for the s process is situated, in our case, between a minimum of $2.35_{10}\times 10^8$ K and a maximum of $3.6_2\times 10^8$ K (or $4.4_2\times 10^8$ K), according to the f_n experimental

TABLE II. Effective ^{176}Lu half-life calculated for some temperature values.

$T(10^8$ K)	$t_{1/2(\mathrm{eff})}$ ^{176}Lu (y)				
	Ref. [4]	Ref. [3]		Present work	
		Minimum	Maximum	Minimum	Maximum
1.5	27.822	$0.266\,12\times10^{11}$	$0.375\,79\times10^{11}$	$0.109\,61\times10^{11}$	$0.377\,89\times10^{11}$
2.0	2.5854	8125.5	$0.580\,20\times10^{6}$	1498.6	$0.181\,58\times10^{7}$
2.5	0.62171	1.1035	35.066	0.713 92	35.852

value: 0.31 (or 0.57_{10}) [2]. In this region of temperature, the variation is 0.035×10^8 K for $\Delta E_j^*=10$ keV.

C. ^{176}Lu as a cosmochronometer

The ^{176}Lu effective half-life can be written [4]

$$\lambda_{\mathrm{eff}}(T)=\frac{\sum_j g_j\lambda_j e^{-(E_j^*/kT)}}{\sum_j g_j e^{-(E_j^*/kT)}}. \tag{9}$$

In Table II are listed the ^{176}Lu $t_{1/2(\mathrm{eff})}$ values calculated for some T values. In the minimum and maximum columns are given the extreme values due to the errors on λ_1 $r_{1m}(1-r_{1m})$ and E_1^*. The agreement between the Ref. [2] results (7.0×10^7 s$^{-1}<\lambda_1 r_{1m}(1-r_{1m})<2.0\times10^{10}$ s^{-1}, $E_1^*=838.640$ keV), or those of Ref. [3] (7.0×10^7 s$^{-1}<\lambda_1 r_{1m}(1-r_{1m})<5.0\times10^9$ s^{-1}) and ours is excellent.

It can be seen that, in this range of temperatures, there are large variations of the $t_{1/2(\mathrm{eff})}$ values as well as differences between Ref. [4] and the others. This makes it even more difficult to predict reliably, for a given star, the ^{176}Lu amount likely to be ejected in the interstellar medium and limits strongly the use of this nucleus as a cosmochronometer.

VII. CONCLUSION

By means of Coulomb excitation combined with the recoil method, it is possible to determine the excitation probability of one or several levels at 885_{50} keV, which could be identified with the 838.640 keV level metionned by various authors. From the deexcitation mode of this level, one can deduce a rather precise value of the $\lambda_j r_{jm}(1-r_{jm})$ term which is a member of the equation giving the f_n evolution with temperature. We so find a temperature for the s process between $2.35_{10}\times10^8$ K and 3.6_2 or $4.4_2\times10^8$ K. The agreement between our results and those of Refs. [2,3] is very good.

There is, however, a contradiction between the experimental results and the theoretical predictions concerning the half-lives of the 838.640 and 921.472 keV levels. Other configurations than those deduced from the transfer experiments [1,5] are suggested in Ref. [16].

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