

Nuclear Excitation by Free Muon Capture

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(Received 11 March 2022; revised 24 June 2022; accepted 16 August 2022; published 27 September 2022)

Efficient excitation of nuclei via exchange of a real or virtual photon has a fundamental importance for nuclear science and technology development. Here, we present a mechanism of nuclear excitation based on the capture of a free muon into the atomic orbits ($NE\mu C$). The cross section of such a proposed process is evaluated using the Feshbach projection operator formalism and compared to other known excitation phenomena, i.e., photoexcitation and nuclear excitation by electron capture (NEEC), showing up to 10 orders of magnitude increase in cross section. $NE\mu C$ is particularly interesting for MeV excitations that become accessible thanks to the stronger binding of muons to the nucleus. The binding energies of muonic atoms have been calculated introducing a state of the art modification to the Flexible Atomic Code. An analysis of experimental scenarios in the context of modern muon production facilities shows that the effect can be detectable for selected isotopes. The total probability of $NE\mu C$ is predicted to be $P \approx 1 \times 10^{-6}$ per incident muon in a beam-based scenario. Given the high transition energy provided by muons, $NE\mu C$ can have important consequences for isomer feeding and particle-induced fission.

DOI: 10.1103/PhysRevLett.129.142501

Manipulating nuclear transitions is a highly desirable goal due to its implications in the energy sector [1–8]. Long-lived nuclear excitations, formally isomers, have lifetimes that are sometimes comparable to the age of the universe and have a potential to release hundreds of megajoules of energy stored in few cubic centimeters. The former aspect is crucial in designing new energy storage solutions: long duration has been suggested to be the key driver towards a decarbonized future [9]. Unfortunately, an efficient process to excite and control the lifetime of isomers is currently lacking.

Nuclear levels, in general, are not easily accessible: they often have high spin with respect to the ground state, the resonances are very narrow and predominantly in the MeV range, in which no high-intensity monochromatic light sources exist yet. Alternatively, to direct excitation via photon absorption, few other secondary electromagnetic processes exist—such as Coulomb excitation [10], nuclear excitation upon electron capture (NEEC) [11,12] or transition (NEET) [13–15] and excitation upon muon cascade [16]. Here we present an alternative electronucleus excitation mechanism that presents one of the highest excitation cross sections: the nuclear excitation by free muon capture ($NE\mu C$). The high energy transferred to the nucleus expands the range of isotopes suitable for the process and makes $NE\mu C$ relevant for muon-induced fission [17–21] and for the feeding of long-lived isomers [1], as shown in Fig. 1.

It is instructive to compare electronic and muonic electronucleus processes. Excitation upon muon cascade

[22,23] and NEET occur as a result of the transition between bound muonic and electronic orbitals, respectively. The two processes have been experimentally observed [24–28] and are considered to be well established. Here, we shall also mention an excitation delayed with respect to the free muon capture and muonic cascade leading to nuclear transmutation by nuclear orbital muon capture [21], which is of electroweak and not of

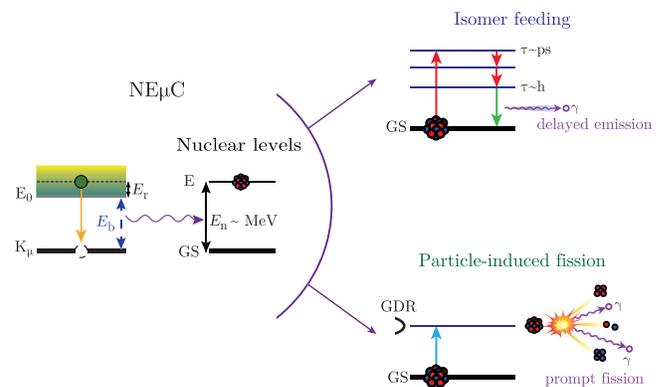


FIG. 1. Nuclear excitation by muon capture ($NE\mu C$): the capture of a free muon leads to a resonant excitation of the nucleus. The excited nucleus can subsequently decay towards lower levels reaching a long-lived state, i.e., isomer feeding. Another possibility is present if the excitation of the nucleus is in resonance with the giant dipole (GDR) or quadrupole resonances (GQR), and the latter is above the fission barrier: the nucleus can undergo a prompt fission induced by $NE\mu C$.

electromagnetic origin and it is thus omitted from most of the quantitative comparison in this Letter.

The NEEC process was claimed to have been observed in a single experiment [12,29–31] and its cross section differs from a theoretical estimate by 9 orders of magnitude [32]. To our knowledge, a muonic analogue of NEEC has never been proposed and in this Letter we investigate this possibility theoretically, underlining key differences with other excitation processes.

Similarly to NEEC and contrary to NEET and muon cascade excitation, $NE\mu C$ describes the capture of a *free* lepton in a corresponding atomic orbital, and is thus not constrained by the restriction of matching the transition energies between *bound* atomic and nuclear levels. Since both NEEC and $NE\mu C$ depend on the interaction between the nuclear and atomic environment, tight muon orbits are expected to provide a higher nuclear excitation cross section than their electronic counterparts. In particular, here we report findings of a $NE\mu C$ integrated cross section up to 1.82×10^5 b eV, that is 5 orders of magnitude higher than any corresponding NEEC cross section reported so far [33–37]. To evaluate the $NE\mu C$ cross section we used the advanced theory based on the Feshbach projection operator formalism developed by A. Pálffy for the NEEC process and presented in Refs. [33] and [38]. In this context, the $NE\mu C$ rate for an electric transition can be written in muonic atomic units as

$$Y_n^{(e)L} = \frac{1}{4\pi\alpha} \frac{4\pi^2\rho_i}{(2L+1)^2} R_0^{-2(L+2)} B\uparrow(EL)(2j_b+1) \times \sum_k |\tilde{R}_{L,k_b,k}|^2 C(j_b L j; 1/2 0 1/2)^2, \quad (1)$$

where $B\uparrow(EL)$ is the reduced transition probability of the L th multipolar transition, $C(j_1 j_2 j; m_1 m_2 m)$ is the Clebsch-Gordan coefficient, j_b and j , are the total angular momentum, while k_b and k are the Dirac angular momentum of the bound and free muon, respectively. $\tilde{R}_{L,k_b,k}$ is the radial integral that depends on the muon bound and free wave functions, which are obtained as solutions of the Dirac equations for a specific atomic configuration using the modified version of the Flexible Atomic Code (FAC) [39].

The $NE\mu C$ cross section can be expressed as

$$\sigma_{NE\mu C} = 2\pi^2 \lambda_\mu^2 Y_n \frac{\frac{\Gamma_r}{2\pi}}{(E - E_r)^2 + \frac{\Gamma_r^2}{4}}, \quad (2)$$

where λ_μ is the free muon wavelength and E_r the resonance energy. The integration of the cross section over the continuum energies, considering that $\Gamma_r \ll E_r$, gives the so-called resonance strength:

$$S_{NE\mu C} = \int \sigma_{NE\mu C}(E) dE = 2\pi^2 \lambda_\mu^2 Y_n. \quad (3)$$

For $NE\mu C$ to be possible the nuclear transition energy (E_n) has to be larger than the muon binding energy (E_b),

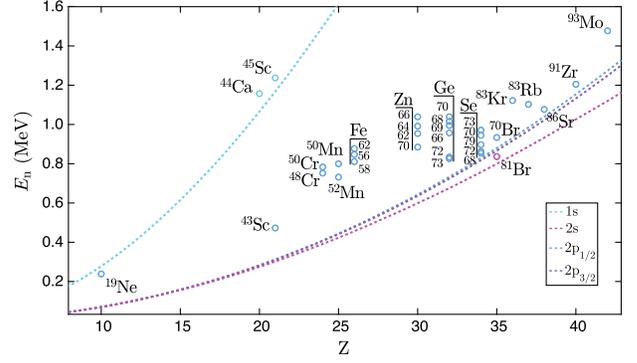


FIG. 2. Isotopes matching the search criteria in case of $E2$ transition. Only the nuclear transitions with a $B\downarrow(E2) > 10$ W.u. have been included in the plot. The color of the markers indicates the closest shell allowing $NE\mu C$. Vertical black lines group several isotopes of the same element.

and the free muon energy (E_r) has to match their difference (i.e., $E_r = E_n - E_b$). This condition defines the search for the nuclear transitions that can be excited by the $NE\mu C$ mechanism. In Fig. 2, we plot the muonic binding energies for K and L shells, calculated with the FAC [39] modified for muonic atoms, and nuclear excited levels that satisfy the above criteria with respect to the nuclear ground state for E_r up to 0.4 MeV above the corresponding muonic levels. For the sake of the presentation we only show the $E2$ transitions, which are generally the strongest. The Table I reports the $NE\mu C$ rates and resonance strengths for selected isotopes together with the nuclear transition energy and the required energy of the free muon, including several $E1$ transitions.

Binding energies for muonic atoms are obtained by numerically solving the Dirac equation including the effect of the finite size of the nucleus using the Fermi distribution function with parameters adjusted to reproduce the rms charge radii of Ref. [40]. Vacuum polarization is taken into account using the standard Uehling potential, while self-energy correction is included using the method of Ref. [41]. The nucleus recoil effect is approximated with an effective Hamiltonian term proposed in Ref. [42].

In Table II we compare the $NE\mu C$ and NEEC resonance strengths of a few of the strongest transitions. For all considered cases the $NE\mu C$ is substantially stronger than NEEC. The enhancement found ranges between 5 to 10 orders of magnitude. Table II also offers a comparison with the direct process of photoexcitation. Results show that in the case of an $E1$ transition, as for ^{138}Ba and ^{207}Pb , S_γ , and $S_{NE\mu C}$ are comparable, while for quadrupolar excitations $S_{NE\mu C}$ is substantially larger than S_γ . The choice of comparing $NE\mu C$ with NEEC and direct photoexcitation is due to the fact that all these processes can excite the same generic nuclear level, having as the degree of freedom the energy of the free muon or electron and the energy of the photon. NEET, and excitation upon muon cascade, instead

TABLE I. Resonance strengths for the isotopes highlighted by the search criteria for $E1$ and $E2$ transitions and capture in the K and L shells. Isotopes are ordered with respect to the mass number. $T_{1/2}$ indicates the half-life of the nuclear ground state.

Isotope	$T_{1/2}$	L^{th}	E_n (keV)	nl_j	E_r (keV)	$Y_{\text{NE}\mu\text{C}}$ (1/s)	$S_{\text{NE}\mu\text{C}}$ (b eV)
^{11}Be	13.76 s	$E1$	320.04	$1s_{1/2}$	275.54	1.39×10^{14}	12.09
^{19}Ne	17.22 s	$E2$	238.27	$2p_{3/2}$	168.21	1.16×10^{14}	16.58
^{43}Ca	Stable	$E1$	1394.47	$1s_{1/2}$	329.02	4.95×10^{10}	3.62×10^{-3}
^{44}Ca	Stable	$E2$	1157.02	$1s_{1/2}$	92.20	4.14×10^{12}	1.08
^{45}Sc	Stable	$E2$	1236.70	$1s_{1/2}$	69.08	2.84×10^{12}	0.99
^{48}Cr	21.56 h	$E2$	752.19	$2s_{1/2}$	360.43	4.69×10^{13}	3.13
^{48}Cr	21.56 h	$E2$	752.19	$2p_{1/2}$	342.84	2.63×10^{16}	1.84×10^3
^{48}Cr	21.56 h	$E2$	752.19	$2p_{3/2}$	345.88	5.15×10^{16}	3.58×10^3
^{52}Mn	5.591 d	$E2$	731.66	$2p_{1/2}$	287.05	9.28×10^{15}	771.14
^{68}Se	35.5 s	$E2$	853.75	$2s_{1/2}$	92.94	3.52×10^{14}	90.87
^{68}Se	35.5 s	$E2$	853.75	$2p_{1/2}$	24.54	1.42×10^{17}	1.38×10^5
^{68}Se	35.5 s	$E2$	853.75	$2p_{3/2}$	36.18	2.75×10^{17}	1.82×10^5
^{73}Ge	Stable	$E2$	825.8	$2p_{1/2}$	92.66	3.81×10^{16}	9.85×10^3
^{73}Ge	Stable	$E2$	825.8	$2p_{3/2}$	101.84	7.41×10^{16}	1.75×10^4
^{81}Br	Stable	$E2$	836.8	$2s_{1/2}$	37.22	1.43×10^{14}	91.82
^{86}Sr	Stable	$E2$	1076.7	$2p_{3/2}$	54.77	9.52×10^{15}	4.17×10^3
^{91}Zr	Stable	$E2$	1204.8	$2p_{1/2}$	51.18	1.32×10^{16}	6.19×10^3
^{91}Zr	Stable	$E2$	1204.8	$2p_{3/2}$	72.26	2.56×10^{16}	8.49×10^3
^{93}Mo	4.0×10^3 y	$E2$	1477.2	$2p_{1/2}$	203.4	6.25×10^{16}	7.38×10^3
^{93}Mo	4.0×10^3 y	$E2$	1477.2	$2p_{3/2}$	228.53	1.21×10^{17}	1.27×10^4
^{138}Ba	Stable	$E1$	6244.8	$1s_{1/2}$	44.19	2.69×10^{14}	146
^{202}Hg	Stable	$E1$	4922	$2p_{1/2}$	329.41	6.12×10^{14}	44.66
^{207}Pb	Stable	$E1$	4980.5	$2p_{1/2}$	165.51	3.09×10^{15}	447.50
^{207}Pb	Stable	$E1$	4980.5	$2p_{3/2}$	350.64	5.67×10^{15}	388.75

needs more strict conditions to take place, making a comparison for the same levels unsuitable.

Similarly to the NEEC case, the $\text{NE}\mu\text{C}$ cross section is greatly enhanced if the resonance is met at low kinetic energy, given the λ^2 prefactor in Eq. (2). In this respect it is important to inspect the precision of atomic orbital calculations. Considering that the FAC has never been used before to compute muonic binding energies, we compare in Fig. 3 the values obtained using the FAC with the state-of-the-art theoretical calculations for muonic atoms presented in Ref. [43], in the case of ^{40}Zr , ^{147}Sm , and ^{209}Bi . The

overall standard deviations between the differences in the binding energies range from 0.36 keV for ^{40}Zr to 0.87 keV for ^{209}Bi . Much of these discrepancies can be attributed to the self-energy correction included in the present Letter and omitted in Ref. [43]. The agreement between FAC and Ref. [43] improves significantly for the M shell, as the self-energy term becomes negligible. This assesses FAC as a valuable tool for the calculations of binding energies in muonic atoms (more detailed comparison is available in the Supplemental Material [44], which contains Ref. [45]).

 TABLE II. Comparison between $\text{NE}\mu\text{C}$, NEEC, and direct photoexcitation for the same nuclear transition for several isotopes. The apex i indicates a bare nucleus configuration while n the one for a neutral atom. Integrated cross sections are expressed in b eV, while E_n in keV.

Isotope	E_n	nl_j	$S_{\text{NE}\mu\text{C}}^i$	S_{NEEC}^i	S_γ	$S_{\text{NE}\mu\text{C}}^n$
^{52}Mn	731.66	$2p_{1/2}$	771.14	6.83×10^{-7}	0.58	764.73
^{68}Se	853.75	$2p_{3/2}$	1.82×10^5	1.44×10^{-5}	4.29	1.62×10^5
^{73}Ge	825.8	$2p_{1/2}$	9.85×10^3	4.35×10^{-6}	1.34	9.45×10^3
^{93}Mo	1477.2	$2p_{3/2}$	1.27×10^4	8.91×10^{-6}	5.00	1.24×10^4
^{138}Ba	6244.8	$1s_{1/2}$	146	1.57×10^{-2}	164.74	120.66
^{207}Pb	4980.5	$2p_{1/2}$	447.5	6.52×10^{-2}	713.96	432.6

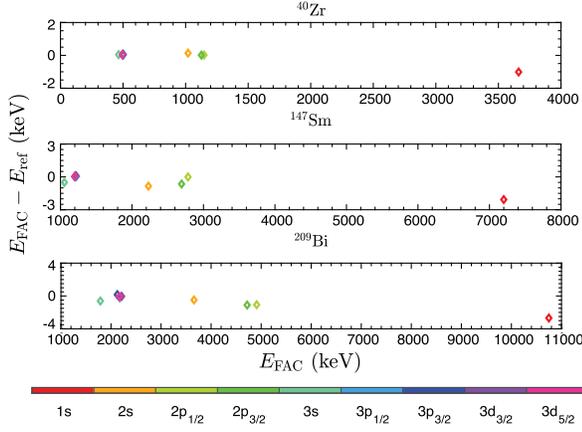


FIG. 3. Accuracy assessment of the FAC. Muonic binding energies computed with the FAC (E_{FAC}) are compared with those of Ref. [43] (E_{ref}). The color of the marker indicates the muonic state.

Another important difference with respect to NEEC is the absence of the high ionization state requirement. In the case of muons, the muonic inner shells are always available for capture and cannot be filled with electrons even for neutral atoms. The presence of electrons in the atomic environment will screen the muonic levels, making the muons less bound by up to few tens of keV, depending on the number of electrons in the shells [43,46,47]. This means that the resonance strengths evaluated for bare nuclei in Table I will be only slightly affected by the electronic charge state of the capturing ion. Thus, NE μ C allows for a capture in the 1s shell of an entirely filled atom. For this reason, we evaluated the NE μ C resonance strengths for the isotopes with the highest $S_{\text{NE}\mu\text{C}}$ of Table I, also in case of a neutral electronic configuration (see Supplemental Materials [44] for further details). Results are shown in Table II. Here we notice, as expected, that the $S_{\text{NE}\mu\text{C}}^i$ and $S_{\text{NE}\mu\text{C}}^n$ are very close to each other, with a slight difference due to the different resonance energy of the neutral case induced by the electron screening. Screening by an arbitrary electronic configuration has been included in the FAC by solving the Dirac equations of both the muon and electrons self-consistently via iteration.

From the experimental point of view, the possibility of capture in neutral atoms can be tremendously useful and could offer an interesting perspective lifting the stringent experimental requirements for NEEC. Indeed, as NEEC simultaneously requires a high ionization state and high density of resonant electrons, the parallel realization of both poses experimental challenges. Lifting the ionization requirement for NE μ C simplifies the experimental scenario. For example, in a beam-based setup NE μ C can be observed by sending a muon beam into a solid target. Analogously to NEEC, the NE μ C probability can be written as [32,48]

$$P = \sum_{\alpha_r} n_i S_{\text{NE}\mu\text{C}}^{\alpha_r} \frac{1}{-(dE_{\mu}/dx)|_{E_r}}, \quad (4)$$

where α_r represents the available capture channels, n_i is the density of atoms, and $-(dE_{\mu}/dx)|_{E_r}$ is the muon stopping power at the resonance energy. The number of excited nuclei per second, assuming a continuous muon beam with a flux ϕ_{μ} (1/s), is given by

$$N_{\text{NE}\mu\text{C}}^{\text{exc}} = P\phi_{\mu}. \quad (5)$$

If we limit ourselves to solid targets with stable or long-lived ground states, essential for practical experiments, ^{73}Ge and ^{93}Mo are the most promising isotopes. The stopping power calculated with GEANT4 [49] is of $dE_{\mu}/dx \simeq -501$ MeV/cm and -607 MeV/cm at the resonant energies of 101.84 keV and 228.53 keV, respectively. Considering the capture only in the $2p_{3/2}$ channel the resulting probabilities are $P = 1.54 \times 10^{-6}$ and $P = 1.39 \times 10^{-6}$, respectively. Remarkably, these theoretical probabilities are 5 orders of magnitude larger than those theoretically estimated for the ^{93}Mo isomer depletion through NEEC [32,50], although considering different excitation levels. If we expand the calculations to short-lived isotopes, e.g., ^{68}Se , the single channel excitation probability reaches $P = 1.02 \times 10^{-5}$. In practical terms, for efficient excitation, the initial energy of the incident muons is irrelevant provided that it is above the resonance energy E_r . Indeed, while traveling in the stopping medium, muons will experience a loss of energy due to subsequent collisions, guaranteeing that the resonant energy E_r will be achieved during the slow down process in the target. The precise depth at which it occurs depends on the incident muon energy. In a realistic setup, high-flux muon beams will have a substantial spread of incident muon energies, which will result in a distribution of resonance depth. This effect might reduce the efficiency of NE μ C detection if the spread in depths exceeds the transmission depth of the nuclear emitted gamma photons. Considering 1 MeV energy spread of the muon beam at energies above E_r , the thickness of the target assuring resonance for all the particles is of several tens of microns (i.e., 50 μm for ^{68}Se). Depending on the target and the energy of the gamma photons involved in the transition, the gamma attenuation will be only up to 50% [51].

Currently, the brightest μ^- beam facilities at PSI (Villigen, Switzerland) and MuSIC (Osaka, Japan) are able to deliver a continuous flux of 10^7 muons per second [52]. Planned upgrades would make it feasible in the next years to have fluxes up to 10^8 and 10^9 muons per second, resulting approximately in ten to one thousand nuclear excitations per second. Furthermore, an increase in the excitation cross section is expected if the wave function of the muon is engineered [53], i.e., considering muon vortex

beams [54], as recently suggested for NEEC [8,37,55]. This modification of the wave function could make unfavourable transitions with higher multipolarity more likely to happen.

Given the high energy of nuclear transitions involved in $NE\mu C$ and its increased efficiency compared to direct photoexcitation at higher multiplicities, $NE\mu C$ can be the most suitable process for isomer feeding. In this case the feeding, as shown in Fig. 1, will not happen directly to the isomer state, but arriving to it through subsequent decays upon the initial excitation from the ground state. This is, for example, the case of the energy level schemes of ^{113}In and ^{87}Sr .

Typically, at energies of tens of MeV above the ground state, the density of the excitation states is so high that they overlap in a broad energy range, giving rise to the so-called giant resonances. Excitation of these resonances, independently from the particular excitation mechanism, can lead to fission if the resonance is above the fission barrier. Prompt fission of the nucleus has been achieved under muon excitation and attributed to the muon cascade in ^{238}U [21]. Yet, the possibility of the $NE\mu C$ has not been considered despite it could provide substantially better energy overlap given that one has an additional degree of freedom, that is the energy of the free lepton. Indeed, fission induced by muonic transitions is governed by the energy difference between two muonic bound states, while in the case of $NE\mu C$ the resonance condition is satisfied throughout the whole width of the giant resonance, that can be several MeV wide. To estimate the contribution of the $NE\mu C$ process to the muon induced fission, we calculated the fission cross section induced by $NE\mu C$ for ^{238}U from the photofission process [56,57] and reported it in the Supplemental Material [44]. Integrating the cross section with the energy dependent stopping power retrieved from Ref. [58] (further details are available in the Supplemental Material [44]), provides us with a final fission probability of $\sim 4.30 \times 10^{-5}$ per incident muon. This probability is still small if compared with prompt fission induced by muon cascade ($\sim 10^{-3}$) and delayed fission induced by muon capture ($\sim 10^{-2} - 10^{-1}$) [59]. Nevertheless, for lighter isotopes the muon cascade eventually becomes nonresonant with the giant resonances, while $NE\mu C$ is theoretically always possible.

Most remarkably, the $NE\mu C$ has the highest chance to be observed than the NEEC process in which disagreement between experiment and theory is of 9 orders of magnitude. Measuring the $NE\mu C$ rates and comparing to the estimates provided in the presented Letter will hopefully help to resolve the contradiction and establish the origins of the extremely high experimentally measured NEEC probability.

S. G. and F. C. acknowledge support from Google Inc. I. M. acknowledges support from the SMART-electron

project that has received funding from the European Union's Horizon 2020 Research and Innovation Program under Grant Agreement No. 964591. The authors would like to thank Giovanni dal Maso for the useful insights about muonic beam facilities.

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