Gargiulo *et al.* Reply: The authors of the preceding Comment [1] have brought forth various aspects related to the nuclear excitation by free muon capture (NEμC) process, which was presented in Ref. [2]. To address any potential misunderstandings and ensure clarity, we will provide detailed responses to each of the points raised, offering further context and clarifications.

In the second paragraph of the Comment, the authors argue that only the s wave would be dominant in the scattering process and that this would be in contrast with the selection rules for the chosen transitions. However, this argument overlooks a key aspect of the theoretical derivation. The expression of the radial integral—appearing in Eq. (1) of Ref. [2]—is obtained by expanding the continuum wave function of the captured muon in partial waves, following the derivation of the nuclear excitation by electron capture (NEEC) process [3–6]. This expansion is valid at all energies and decomposes the wave into its constituent components, possessing a given orbital angular momentum l and total angular momentum $j = |\kappa| - 1/2$. Only the wave functions of the free muon satisfying the selection rules imposed by the spin and parity changes of the transition will then be used to compute the radial integral. Thus, no conflicts arise between the selection rules and the multipolar transitions presented in Table I.

Another point raised refers to the assumption that all incident muons fulfill the energy criteria for the capture. It is worth noting that the resulting probability P does not depend on the number of muons satisfying the energy-matching condition. In fact, the muon flux enters only in the determination of the total number of excited nuclei $N^{\mathrm{exc}}_{\mathrm{NE}\mu\mathrm{C}} = P\phi_\mu$. This value is mentioned only a single time: "resulting approximately in ten to one thousand nuclear excitations per second" to provide a crude estimate for a possible experiment, factoring in planned upgrades at specific facilities. When muons are slowed down in a target material, they experience energy losses in discrete steps. The total amount of muons that satisfy the energy-matching conditions will depend on the initial energy of the muons, the shell in which it is captured, the lifetime (energy width) of the excited nuclear level, and the material considered. Hence, the value $N^{\mathrm{exc}}_{\mathrm{NE}\mu\mathrm{C}}$ can be simply reevaluated by multiplying P by the flux that meets these conditions from time to time in the particular experimental scenario considered. This being said, the assumption that all the "particles" reach resonance is widely used in this context [7,8] as a first-order approximation.

The Comment also addresses the issue of separating muon and nuclear degrees of freedom in our analysis. Although this represents an approximation, this separation has been widely used to treat the radiationless excitation upon muon cascade (NEμT) [9] and was found to be in reasonable agreement with experimental measurements [10]. Hence, we adopted a similar assumption as a starting point for our initial investigation into this new process. It is important to note that the relevance of dynamic mixing on the final cross-section values has not been quantified. Nonetheless, future models could benefit from incorporating it for better accuracy.

The Comment also argues that our Letter ignores other dominant capture paths. This criticism, however, appears to overlook the context provided in Ref. [2]. In our study, we have estimated a probability of $P \sim 10^{-6}$ for the NEμC process in $^{93\mathrm{m}}$Mo. This low probability clearly indicates that NEμC is not the predominant mechanism following a given muon capture. Moreover, it is crucial to emphasize that when studying nuclear excitation processes (NEμC in our case) the purpose is not comparing them with other, more probable, atomic processes like Auger or radiative emissions (as mentioned in the Comment), but rather with those processes that could potentially excite the nucleus. Regarding this aspect, a direct comparison between NEμC and nuclear excitation upon muon cascade (NEμT) between the same nuclear levels is not always feasible. The two processes are generally not in direct competition due to a fundamental distinction. NEμT requires the energy matching between the nuclear excitation and the transition energy between two bound orbitals; this condition might be encountered in a small subset of isotopes given the selection criteria. On the other hand, NEμC has broader applicability across all isotopes due to an additional degree of freedom: the kinetic energy of the free particle that can be used to ensure the energy matching. Consequently, our study has focused on comparing NEμC with other processes that share this feature, such as NEEC and direct photoexcitation, allowing comparisons between the same nuclear levels. Moreover, NEEC has been a reference for our study, owing to its similarities with NEμC and the current scientific efforts directed toward its understanding and experimental verification. Yet, when the two processes (NEμC and NEμT) entered in direct competition, i.e., such as in our case study involving ^{238}U muon-induced fission, we have offered a comparison between them. Additionally, we have compared the prompt fission resulting from these two processes with the delayed fission initiated by the electroweak (orbital) muon capture. In this circumstance, our findings revealed that the orbital muon capture and NEμT were significantly more efficient than NEμC at inducing fission in ^{238}U.

While our Letter discusses the possibility of using NEμC to activate or deplete isomers (long-lived nuclear excitations), this occurs only as a prospect. Indeed, all the nuclear levels presented in Table I of Ref. [2] have half-lives shorter than 1 ns, which falls below the formal definition of isomers requiring $T_{1/2} \geq 10$ ns [11]. It could not have been otherwise since these levels are connected to the ground state by low-order multipolar transitions ($E1$ or $E2$). Direct isomer excitation from the ground state can, however, occur considering other higher-order multipolar transitions. In our outlook, we clarified that the population

of the isomer, as shown in Fig. 1, cannot happen directly from the ground state to the isomer, but may proceed to it through subsequent decays upon the initial excitation from the ground state.

Once a nuclear level is excited, the cascade toward the ground state is not controllable, and no pathway can be externally chosen to specifically populate the isomeric state. Nonetheless, this observation does not preclude the potential of identifying a partial-level scheme where NEμC could be effectively used to activate or deplete an isomeric state, albeit with a certain branching ratio.

Finally, the authors of the Comment highlighted that excited nuclei are likely to be destroyed by nuclear orbital muon capture. While we acknowledge the significance of this aspect, it is important to mention that orbital muon capture occurs with a certain characteristic delay (∼100 ns for heavy nuclei) compared to the prompt excitation of the nucleus by NEμC. Therefore, the experimental observation of NEμC can be designed using time-resolved techniques. Additionally, for light isotopes ($Z < 15$) the total muon capture rate gradually becomes negligible [12], while nuclear excitation by NEμC remains feasible. The list of isotopes that can be excited through NEμC can be expanded by considering capture in higher orbitals and higher multipolar transitions.

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