Reply to: Possible overestimation of isomer depletion due to contamination

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REPLYING TO S. Guo et al. *Nature* http://doi.org/10.1038/s41586-XXX-XXXX-X (2021)

We appreciate the interest of Guo et al., the points that they raise, and the opportunity that we have to provide additional details that are not included in ref. [1]. This allows us to strengthen our experimental case[1] while, in parallel, recent developments are improving our theoretical understanding of nuclear excitation by electron capture (NEEC), such as the exploration of a substantial increase in predicted NEEC probability when considering capture by an ion in an excited state (S. Gargiulo et al., submitted) or the impact of the momentum distribution of target electrons (J.R. et al., submitted). In the accompanying Comment[2], Guo et al. focus on whether potential background contributions were underestimated in our analysis. As discussed below, these concerns are mostly unwarranted; aside from a small systematic uncertainty that could possibly slightly reduce our reported NEEC excitation probability[1] of P_{exc} = 0.010(3), our original conclusions still stand.

Guo et al.[2] rightly note that the 263-keV isomeric transition (half-life $T_{1/2}$ = 6.85 h) is not expected to be observed in true, prompt coincidence with any γ-rays with energies higher than 1,478 keV. However, the authors appear to argue that the NEEC-indicative 268-keV transition ($T_{1/2}$ = 3.5 ns; short enough to be in prompt coincidence) should behave more like the delayed 263-keV γ-ray—appearing in the gated spectrum solely through chance coincidences—than like the four other prompt ^{93}Mo lines at 123, 203, 770 and 963 keV that could be eliminated from the spectrum through background subtraction. The opposite is true. With a half-life much longer than the 82.47 ns between ATLAS beam pulses, there is no way to correlate between the isomer decays and the specific reaction (or beam pulse) that created the isomer. The essentially uniform time distribution would indeed result in chance coincidences between the 263-keV isomeric transition and γ-rays arising from other reactions. However, this chance-coincidence rate would differ from that of prompt ^{93}Mo γ-rays (including the four aforementioned background lines, as well as the 268-keV γ-ray) that can be produced in two independent reactions within the same coincidence window. The behaviour of the 263-keV peak thus cannot be used as a reliable gauge, as Guo et al.[2] propose, for what to expect for prompt γ transitions.

To estimate the rate of chance coincidences between two unrelated ^{93}Mo decays, we can follow the steps outlined by Guo et al.[2], with some corrected values. Adopting the PACE4 cross-sections, along with our experimental parameters[1] (beam intensity and target thicknesses) and the 90% factor for populating above spin 17/2, gives a ^{93}Mo production rate of ~8.4 kHz. The approximate yield passing through the 268-keV transition is at the low end of their quoted range, ~20%, giving a rate of ~1.7 kHz for the production of the 268-keV γ-ray. The coincidence window of 90 ns (Guo et al. assumed 100 to 1,000 ns) gives a chance-coincidence probability of ~0.03%, well below the NEEC excitation probability[1] of P_{exc} = 1.0(3)%.

We can deduce the chance-coincidence probability more directly from the data, instead of relying solely on the above estimates. Data were recorded using a 2-μs coincidence window, encompassing many beam pulses, with a narrow 90-ns window defined in the offline analysis. For a given reaction occurring within a specific 90-ns window, the probability of a second reaction occurring in either the same window or a similar one associated with a different beam pulse is equivalent. Events were selected for which the 268-keV and 1,478-keV transitions were observed within one 90-ns coincidence window, along with a 685-keV γ-ray that could arrive at any time within the full 2-μs window. The time difference between the 685-keV and 268-keV transitions shows that most events correspond to the 685-keV γ-ray that arrives within the same 90-ns window as the other two γ-rays (prompt coincidence). By taking the ratio of the number of events detected with the 685-keV transition in a separate 90-ns window to their number in prompt coincidence (same 90-ns window), we find that the probability for chance coincidences between two ^{93}Mo decays is 0.08(8)%, consistent with the above estimate using the PACE4 cross-sections. Again, we conclude that such chance coincidences are fairly small by comparison.

As for true coincidences with statistical γ-rays, we see no reason to expect a dramatic difference in the behaviour of the prompt 268-keV γ-ray compared to other prompt ^{93}Mo transitions. Placing a double gate on the 1,478-keV transition and one of the prompt lines (268 keV, as well as the 123-keV and 203-keV lines used for determining the *k* background parameter in ref. [1]) produces spectra with no noteworthy differences in the continuum at high energies—see Fig. 1. Whereas any residual counts in the 123-keV and 203-keV peaks might be unidentifiable in figure 2a of ref. [1] because of low statistics after background subtraction, they can be clearly seen (and fitted) in two of the component spectra in external data figure 3 of ref. [1]; this was the basis for determining the *k* value for the background subtraction. The lower intensities of the background lines compared to the 268-keV transition are reflected in the corresponding statistical uncertainties from the fits to the g_1p_2 and b_1p_2 spectra. The values of *k* for each of the four background lines are plotted in blue in the inset to Fig. 1. The weighted average of these *k* values has a statistical uncertainty comparable to those of the 268-keV and 685-keV transitions (in green), even though the individual uncertainties for the background lines are larger. The 268-keV and 685-keV peaks clearly have *k* values that differ from those of the lines attributed to background, even though they should be similarly coincident with statistical γ-rays. We attribute this excess of counts to NEEC.

Comparing the g_1g_2 and b_1g_2 spectra by graphically overlaying them as a means of evaluating the background subtraction can be misleading. Statistical variations by channel make the height of a peak a less reliable gauge of its size than the fitted area. Nevertheless, we follow the example of Guo et al.[2] and similarly plot in Fig. 2 an overlay of the $g_1p_2 = g_1g_2 - g_1b_2$ and

$b_1p_2 = b_1g_2 – b_1b_2$ spectra (including the important background contributions g_1b_2 and b_1b_2, neglected by Guo et al.) that were used to deduce the *k* values in ref. [1], with the latter spectrum scaled by 1.33. The four background lines at 123, 203, 770 and 963 keV would all be oversubtracted if $k = 1.33$ was used. (Here, the peak heights visually suggest the same conclusion as the areas.) The actual ratios of areas (the correct approach) are the *k* values plotted in the inset to Fig. 1. We also note that the 235-keV and 244-keV peaks arising from the strong ^{92}Mo channel, visible in the spectra in Guo et al.[2], would not be eliminated by the simple construction $g_1g_2 – 1.33b_1g_2$; however, the full background subtraction in figure 2 of ref. [1] (and as evident in Fig. 2) does remove this background.

The excitation (NEEC) probability P_{exc} was determined from figure 3 of ref. [1] from the sum of the spectra double-gated on 1,442/241 keV and 686/241 keV, by comparing the 268-keV and 2,475-keV peak areas, corrected for efficiency and internal conversion. Given the successful background subtraction with $k \approx 1$ in the spectra in figure 2 of ref. [1], we adopted $k = 1$ for the spectra of figure 3 as well. We note that the strong 1,734-keV transition that feeds the isomer parallel to the 2,475-keV γ-ray[3] (not shown in ref. [1]) is in true coincidence with the 241-keV and 1,442-keV γ-rays, but not with the one at 686 keV. Thus, we can perform a similar *k* analysis as before, with gates 1 and 2 corresponding to 686 keV and 241 keV, respectively, yielding $k = 1.07(14)$. Furthermore, adjusting *k* between 0.7 and 1.3 for the summed spectra in figure 3 of ref. [1] only changes the deduced value of $P_{exc} = 0.010(3)$ by ~9%—small compared with the uncertainty—so the result is fairly robust.

Guo et al.[2] are concerned by the presence of a 263-keV peak in figure 3b of ref. [1]. They are correct that the high-lying 262-keV transition identified in ref. [3] would be weak and Doppler-shifted, and thus could not be the peak observed in this spectrum. However, it does not arise from the isomer decay either. There is an additional (so far unpublished) 262-keV γ-ray, emitted from stopped ^{93}Mo nuclei, that was identified in our benchmark experiment at the Australian National University (ANU)[1]. This is the peak that appears via the 686/241-keV double gate, and it has the correct size relative to the 278-keV peak, expected from the unpublished level scheme (both peaks are marked with asterisks in figure 3 of ref. [1]). No coincidences in the control reactions were found to interfere with the NEEC signature itself[1].

In summary, our result of P_{exc} = 0.010(3) is largely supported by the suitable background subtraction and analysis techniques described in ref. [1] and clarified here. A potential systematic error arising from chance coincidences may reduce this somewhat (from 0.0096 to 0.0088, quoting additional precision)—a small change relative to the uncertainty of 0.003. NEEC has not been ruled out, nor have any more likely candidate mechanisms been proposed. That said, we do encourage others to explore NEEC using different experimental approaches, as it would be valuable to have independent confirmation of this long-sought phenomenon.

Data availability

Data analysed for the original publication were re-examined for this Reply; no new data were generated.

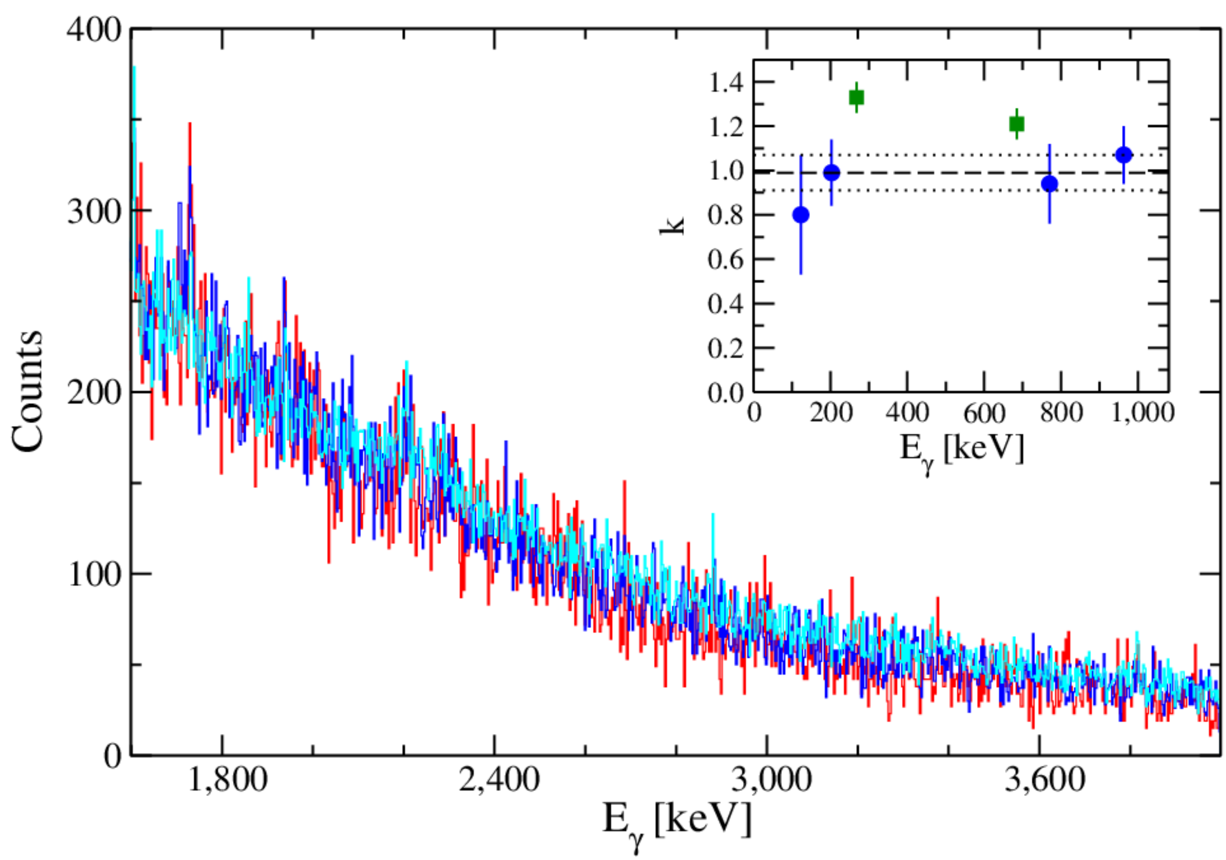

Fig. 1 | Spectra double-gated on 1,478 and 123 keV (red), 203 keV (blue) or 268 keV (cyan). The spectra have been normalized to the latter. Inset, values of *k* derived from the g_1p_2 and b_1p_2

spectra for the 268-keV and 685-keV transitions (green) and four background lines (blue). The weighted average and corresponding uncertainty for the latter transitions are marked by the dashed and dotted lines, respectively. Uncertainties are indicated at 1σ or the 68% confidence level.

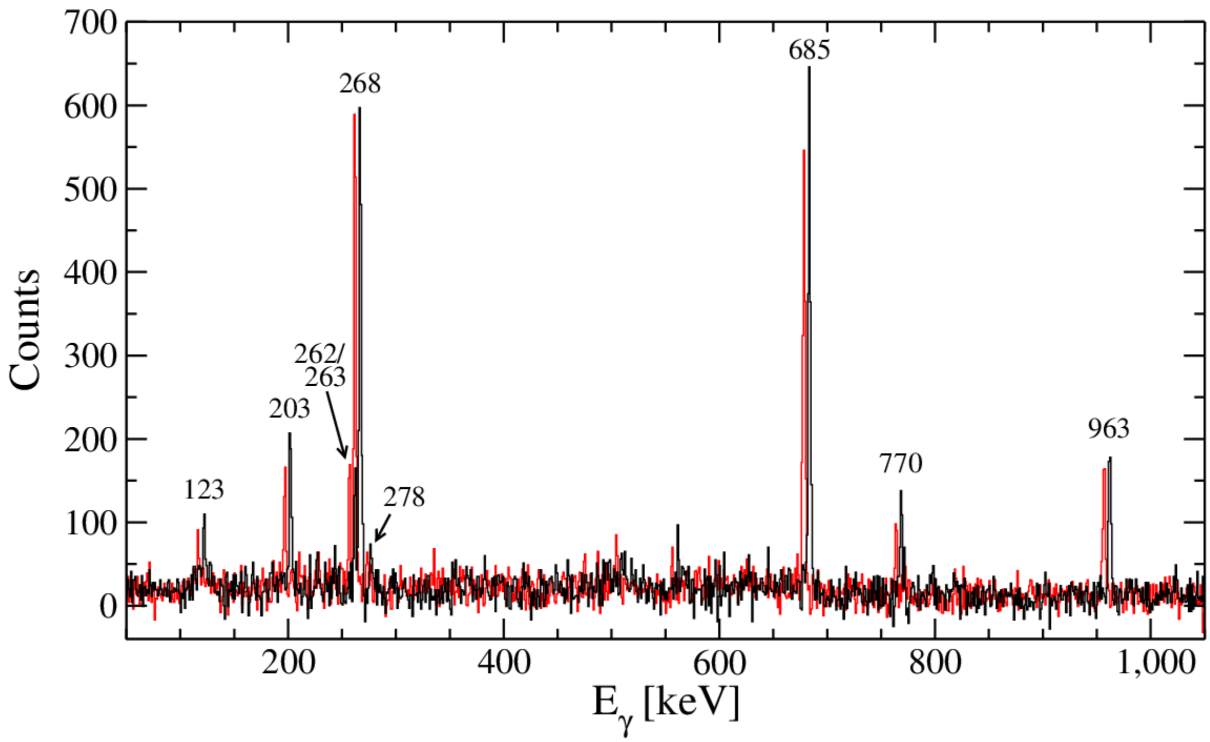

Fig. 2 | Spectra for background subtraction for the double gate on 2,475 keV (γ-ray 1) and 1,478 keV (γ-ray 2). Spectrum $g_1p_2 = g_1g_2 - g_1b_2$ is in red and shifted 5 keV lower for clarity; $b_1p_2 = b_1g_2 - b_1b_2$ is in black and multiplied by 1.33.

1. Chiara, C. J. et al. Isomer depletion as experimental evidence of nuclear excitation by electron capture. *Nature* **554**, 216–218 (2018).

2. Guo, S., Fang, Y., Zhou, X. & Petrache, C. M. Possible overestimation of isomer depletion due to contamination. *Nature* http://doi.org/10.1038/s41586-XXX-XXXX-X (2021).

3. Fukuchi, T. et al. High-spin isomer in ^{93}Mo. *Eur. Phys. J. A* **24**, 249–257 (2005).

Acknowledgements This work was supported by the US Department of Energy (DOE), Office of Science, Office of Nuclear Physics, under contract numbers DE-AC02-06CH11357 and DE-AC02-98CH10886 and grant numbers DE-FG02-97ER41041 and DE-FG02-97ER41033; the National Science Foundation under grant number PHY-1203100; and the Polish National Science Centre under grant number 2017/25/B/ST2/00901. This research used resources of Argonne National Laboratory's ATLAS facility, which is a DOE Office of Science User Facility, and of the Heavy Ion Accelerator Facility at the ANU.

Author contributions All co-authors were members of the collaboration that collected, analysed and interpreted the data reported in the original Letter, which have been further examined here. C.J.C. composed the response, with input from J.J.C. and G.J.L. All co-authors had the opportunity to review the response.

Competing interests The authors declare no competing interests.

Additional information

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