

β decays of the heaviest $N = Z - 1$ nuclei and proton instability of ^{97}In

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We report on new or more precise half-lives, β -decay endpoint energies, and β -delayed proton emission branching ratios of ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn . The measured values are consistent with known mirror transitions in lighter $T_z = -1/2$ nuclei, shell-model calculations, and various mass models. In addition to the β -decaying ($9/2^+$) ground state, circumstantial evidence for a short-lived, proton-emitting isomer with spin ($1/2^-$) was found in ^{97}In . Based on the experimental data, a semiempirical theory on proton emission, and shell-model calculations, the proton separation energy of the ^{97}In ground state was determined to be -0.10 ± 0.19 MeV. The existence of the short-lived, proton-unstable ($1/2^-$) isomer in ^{97}In establishes ^{96}Cd as an rp -process waiting point.

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The heaviest $N = Z$ doubly magic ^{100}Sn and atomic nuclei in its vicinity have been actively investigated both theoretically and experimentally [1], because several important topics in nuclear structure and astrophysics converge in this region of the chart of nuclides. Significant efforts have been made to address questions concerning the robustness of the $N = Z = 50$ shells and evolution of single-particle energies [2–8], the effect of proton-neutron (pn) isoscalar/isovector interactions

in heavy $N \sim Z$ nuclei [9,10], and the location of the proton drip line. The most notable results were reported along the $N = Z$ line [11–13], where the production rates of such exotic radioactive isotopes were at the lowest allowed limit. Many of the $N \sim Z$ nuclei are also relevant for the rapid proton capture (rp) process [14] of nucleosynthesis. Their decay properties have been reported in several works in this context [15–18] to determine more precisely the contribution of the rp -process to the observed elemental abundance in the solar system and the galaxy.

The first experimental results on the heaviest $N = Z - 1$ nuclei have emerged in recent years. The even- Z nuclei ^{91}Pd , ^{95}Cd , and ^{99}Sn have been found to be stable against proton

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TABLE I. Implantation counts, parent β -decay correlation fractions, random background correlation rates, β -decay $T_{1/2}$, Q_{EC} , $\log ft$, and $b_{\beta p}$ values of ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn . Theoretical $T_{1/2}$ values for ^{91}Pd , ^{95}Cd , and ^{99}Sn are taken from Ref. [35]. An isomeric state in ^{97}In is hypothesized to emit a proton and become ^{96}Cd , whose decay correlation fraction and the half-life range are listed separately.

Nucleus	Implantation counts	Correlation (%)	Background rate (Hz/nucleus)	$T_{1/2}^{\text{expt}}$ (ms)	$T_{1/2}^{\text{lit}}$ (ms)	$T_{1/2}^{\text{theo}}$ (ms)	Q_{EC} (MeV)	$\log ft$	$b_{\beta p}$ (%)
^{91}Pd	390	70(4)	0.35(2)	32(3)	$>1.5 \mu\text{s}$ [19]	44.5	11.8(22)	3.4(5)	$3.0^{+1.1}_{-0.9}$
^{95}Cd	476	68(3)	0.41(2)	32(3)	29(8) [23] 73^{+53}_{-28} [20]	31.7	10.2(17)	3.1(5)	$4.5^{+1.2}_{-1.0}$
^{97}In	278	50(4)	0.31(2)	36(6)	26^{+47}_{-10} [20]				
$^{97}\text{In}/^{97m}\text{In}$		35(3)/29(2)	0.31(2)	28(5)		1.3–230 μs	10.0(30)	3.0(9)	$1.7^{+1.7}_{-0.8}$
^{99}Sn	77	62(6)	0.32(3)	24(4)	$>200 \text{ ns}$ [20]	20.6	14.7(36)	3.8(7)	$3.9^{+3.4}_{-1.7}$

emission [19,20], through which noticeable reaction flows occur in type-I x-ray bursts and steady-state burning processes [21]. On the other hand, the odd- Z species ^{89}Rh and ^{93}Ag have been shown to be proton unbound [22]. ^{97}In is an interesting case, since its experimental half-life of 26^{+47}_{-10} ms [20] is similar to $T_{1/2} = 29(8)$ ms of ^{95}Cd [23]. If proton emission from ^{97}In is hindered, then the assumption of ^{96}Cd as a waiting-point nucleus in the rp -process must be scrutinized. One possible implication of a proton-stable ^{97}In is the reduction in the population of $A = 96$ isobars, which reduces the contribution from x-ray bursts to the production of ^{96}Ru found in large quantities in the solar system [24].

This Rapid Communication reports on β -decay $T_{1/2}$, Q_{β} , and $b_{\beta p}$ measurements of ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn , enabling a quantitative description of their roles in the rp -process and tests of the mass and shell-model theories at the proton drip line. The nuclei of interest were produced via fragmentation of a 345-MeV/u ^{124}Xe primary radioactive-isotope (RI) beam on a 740-mg/cm 2 ^9Be target at the RIKEN RI Beam Factory. Isotopes of similar mass-over-charge ratios A/q and atomic number Z were separated at the first stage of the RIKEN projectile fragment separator (BigRIPS) by a $B\rho$ - ΔE - $B\rho$ method with a 3-mm Al wedge degrader, dipole magnets, and slits at the dispersive foci. The filtered beam was identified on an event-by-event basis by $B\rho$ -TOF- ΔE measurements at the later stages of BigRIPS and the ZeroDegree spectrometer [25,26] using position-sensitive parallel-plate avalanche counters [27], plastic scintillators, and a gas-filled ionization chamber [28]. The particle identification plot obtained in this experiment is shown in Fig. 1 of Ref. [22]. The flight time through the separation and identification systems was calculated for each species in its rest frame with LISE++ [29], which ranged from 600 to 630 ns depending on A and Z .

Ion implantation and particle decay measurements took place in the wide-range active silicon strip stopper array for β and ion detection (WAS3ABi) [30]. The nuclei were implanted in one of the three double-sided silicon strip detectors (DSSSDs) of WAS3ABi, each with 1-mm thickness. Each DSSSD was segmented into 60×40 1-mm strips in x and y directions, respectively. For every ion implantation event, its implantation pixel position was determined by evaluating the x -side strip with the minimum time-to-digital converter (TDC) time and the y -side strip with the maximum energy

deposit. In the offline analysis, noise events of WAS3ABi were suppressed by setting a minimum energy threshold of 100 keV per strip. Ten single-sided segmented strip detectors (SSSSDs) were placed farther downstream for Q_{β} measurements. Events accompanying proton emission were separated from positron events by requiring a minimum of 1500 keV energy deposited in a single pixel of a DSSSD as described in Ref. [31].

Decay events were correlated to a previously implanted ion if an energy above 100 keV was registered within one-pixel distance of the implantation position in the same DSSSD. The time correlation window was set to 5 s before and after ion implantation, where the $t < 0$ time events were used to determine the random background correlation rate in the half-life analysis. A maximum likelihood method (MLH) on unbinned data was used to determine the half-life of each nucleus, where the fit function contained the parent, β -daughter, and βp -daughter decay components with half-lives and $b_{\beta p}$ values listed in Ref. [32] and a constant background for random correlations. Only two generations of isotopes were considered in the Bateman equation, as the half-lives of the granddaughter species were comparable or greater than the 5-s MLH evaluation range. Electron capture branching ratios were negligible for the parent nuclei. For ^{97}In the β -daughter component was based on the $(9/2^+)$ ground-state half-life of 1.10(8) s for ^{97}Cd [16]. The Q_{β} values were determined also by the MLH method on the total positron energy spectrum, where the probability density function was derived from GEANT4 simulations of positrons inside WAS3ABi at various trial Q_{β} inputs [33,34]. For the Q_{β} analysis, only the correlation events between 0 to 150 ms were analyzed to maximize the parent-decay component. Q_{β} spectra with $t < 0$ ms and $t > 500$ ms were used to determine background contributions from random correlations and daughter decays.

Figure 1 shows the β -decay time distributions and positron energy spectra, as well as the extracted half-life and Q_{β} values of ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn . The $T_{1/2}$ values are either new or more precise than literature values (see Fig. 2), and they agree well with the predictions given in Ref. [35]. The half-lives and large Q_{β} values of these $T_z = -1/2$ nuclei are consistent with the hypothesis of mixed ground-state to ground-state Fermi and Gamow-Teller decays of $T'_z = -T_z$ mirror nuclei, where the isobaric analog states are easily accessible due to the large β -decay energy window. With this assumption the binding energy

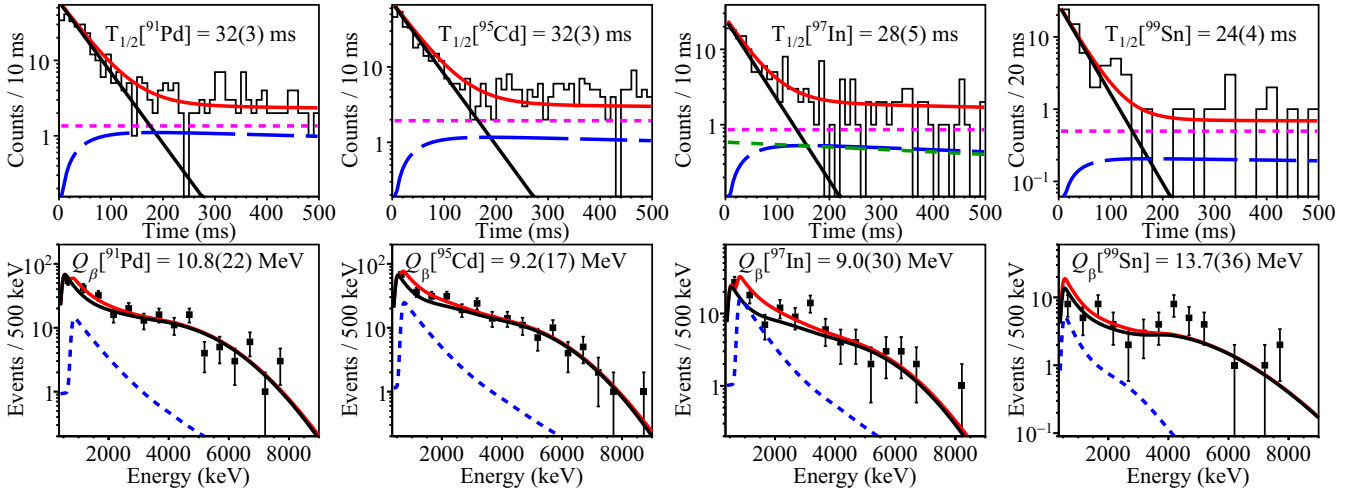


FIG. 1. Top row: β -decay time distributions and half-lives of ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn . The full fit function (red solid line) is composed of the parent decay (solid black), β -decay daughter (long dashed blue), and background (short dashed magenta) components. A fraction of ^{97}In is postulated to be populated in an isomeric state undergoing proton emission within the 600- μs dead time of WAS3ABi, and its decay component is drawn as a green dashed line. Bottom row: experimental positron energy distributions of the four nuclei compared with simulated Q_β probability density functions (red line) containing the parent decay (solid black) and β -decay daughter (blue dashed line) components.

difference Q_{EC} between the parent and the daughter nucleus was calculated as $Q_{EC} = Q_\beta + 2m_e$. In addition, βp emission branching ratios $b_{\beta p}$ were determined for the first time based on the number of single-pixel events with $\Delta E > 1500$ keV. The small $b_{\beta p}$ values are consistent with the current type-I x-ray burst rp -process reaction flow calculations which involve ^{91}Pd , ^{95}Cd , and ^{99}Sn with negligible $b_{\beta p}$ [36]. Aside from the βp events, all of the remaining β -decay branch was assumed to populate the ground states of the daughter nuclei for $\log ft$ calculations. The results are given in Table I, where the $\log ft$ values are consistent with other decays of $T_z = -1/2$ nuclei [37]. No β -delayed γ -ray transitions were observed in ^{91}Pd , ^{95}Cd , and ^{99}Sn .

The decay properties the four nuclei are summarized in Table I. The initial analysis of the decay curve fit yielded a β -decay correlation percentage of 50(4)% for ^{97}In , much lower

than the expected value of 66(4)% from a linear interpolation of the values obtained from ^{91}Pd , ^{95}Cd , and ^{99}Sn . These percentages were determined by dividing the integral of the parent β -decay fit components by the number of implanted ions which have not decayed by βp events. Regarding ^{97}In , we propose the existence of an isomeric state ^{97m}In which has decayed within the 600- μs dead time of WAS3ABi after implantation. Based on the discovery of the odd- Z proton emitters ^{89}Rh and ^{93}Ag [22], ^{97m}In was assumed to decay into ^{96}Cd by $1p$ emission. Therefore an additional β -decay component of ^{96}Cd [$T_{1/2} = 0.93(6)$ s from weighted average of Refs. [10,15,20,23], green dashed line in Fig. 1] was included in the half-life analysis of ^{97}In . With this alternative hypothesis the combined β -decay correlation was 64(4)%, consistent with the expected value. The $b_{\beta p}$ value for ^{97}In in Table I is attributed to its ground state. Taking the 2σ -low value as the initial sample size, the upper limit on the half-life of ^{97m}In was derived by solving the exponential decay equation with an elapsed time of 600 μs ; the final sample size was assumed to be 3.57, which is the 2σ upper limit of zero observations in Poisson statistics [40]. The resultant upper limit was 230 μs . The lower limit on the half-life of the isomer was calculated by assuming a 2σ reduction of ^{97m}In in counts during the 600-ns flight through the separator, which yielded 1.3 μs . The $T_{1/2}$ limits of the isomer are shown in Fig. 2.

The existence of the two states in ^{97}In was investigated with a semiempirical theory of proton emission [41], which relates the partial $T_{1/2}$ of a state to its emitted proton energy Q_p and the angular momentum l . Below the $Z = 50$ shell, an unpaired proton may be emitted from either the $p_{1/2}$ orbital ($l = 1$) or the $g_{9/2}$ orbital ($l = 4$), corresponding to the $1/2^-$ and $9/2^+$ states. The proton-emitting state in ^{97}In is likely to be $1/2^-$ due to its lower centrifugal barrier and higher energy relative to the $9/2^+$ state. The energy of the ^{97}In $1/2^-$ state was calculated with multiple sets of shell-model (SM)

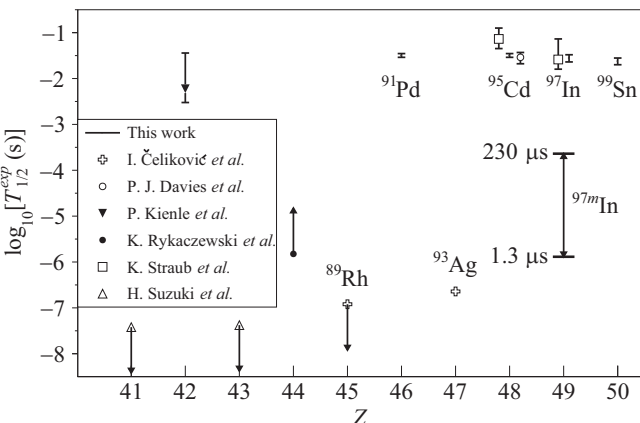


FIG. 2. Experimental half-lives of $T_z = -1/2$ nuclei for $40 < Z \leq 50$. Values obtained outside of this work are taken from Refs. [19,20,22,23,38,39].

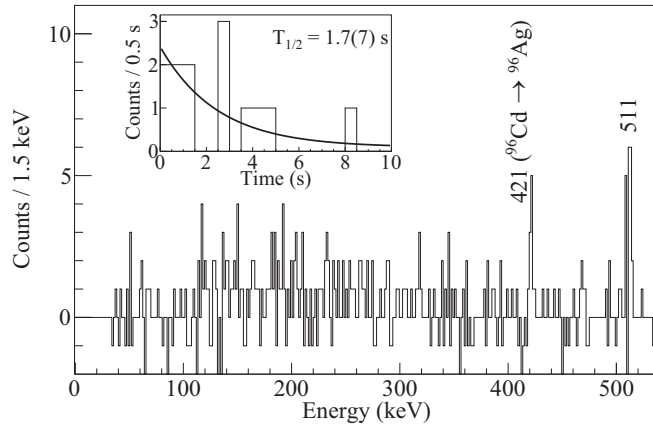


FIG. 3. γ -ray spectrum following β decays between 0 and 3 s after ^{97}In implantation. Decay events preceding the implantation of the isotope were used for background subtraction. The peak at 421 keV is known to originate from the β decay of ^{96}Cd to ^{96}Ag [10,23], where the presence of ^{96}Cd can be explained by the proton emission from ^{97m}In . The inset shows the β -decay time distribution gated by the 421-keV peak, and the corresponding half-life which is consistent with the $T_{1/2}$ of ^{96}Cd .

parameters in the $\pi\nu(p_{1/2}, g_{9/2})$ model space: interactions derived with isospin-asymmetric fits by Gross-Frenkel (GF) [3], and isospin-symmetric interactions by Serduke, Lawson and Gloeckner (SLGT0) [2]. The latter was used to predict the half-lives and binding energies of these nuclei [35]. In addition, the “r3g” interaction [42,43] in the $\pi\nu(f_{5/2}, p, g_{9/2})$ model space has been used frequently to calculate the properties of $N \sim Z$ nuclei with a proper mass scaling between ^{56}Ni and ^{100}Sn . GF and SLGT0 calculations report $E(1/2^-) - E(9/2^+) = 783$ and 568 keV, respectively, while the r3g interaction predicts the corresponding excitation energy of 473 keV. The average value of 0.61(18) MeV is consistent with all of the experimentally known $1/2^-$ state energies of odd-mass $^{103-111}\text{In}$ [44–48]. Therefore we propose a $J^\pi = (1/2^-)$ assignment to the proton-emitting state and the $J^\pi = (9/2^+)$ assignment to the β -decaying state. The isomeric ratio of ^{97m}In was 44(3)%, based on the correlated β -decay ratio of ^{96}Cd to ^{97}In . This value is within a theoretical range of 20–50% calculated from the sharp cutoff model in conjunction with the abrasion-ablation model [49,50], which was used to reproduce some of the experimental isomeric ratios in this region [51].

In the β -delayed γ -ray energy spectrum following ^{97}In implantations shown in Fig. 3, a clear peak with 9(4) counts at 421 keV is visible. This line corresponds to the $(1^+) \rightarrow (2^+)$ transition energy in ^{96}Ag following the β decay of the ground state of ^{96}Cd [10,23]. The β -decay half-life associated with this γ ray was 1.7(7) s, consistent with the $T_{1/2}$ of ^{96}Cd mentioned earlier in the text. Based on the detection efficiency of EURICA, $\approx 100\%$ β -decay branch of ^{96}Cd yielding the 421-keV γ ray and the overall β -decay correlation efficiency of 64(4)% for ^{97}In , the intensity of the 421-keV peak was 122^{+74}_{-52} . This value agrees with the number of ^{97}In ions implanted in WAS3ABi, scaled by the 44(3)% isomeric ratio. These results provide strong evidence for a proton-emitting ^{97m}In .

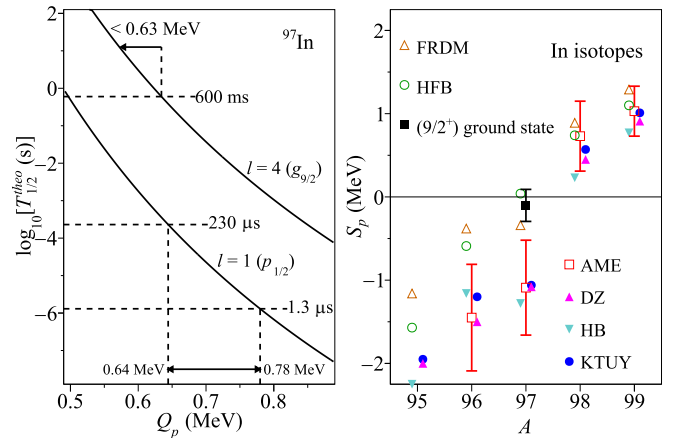


FIG. 4. Left: $T_{1/2}^{\text{theo}}$ as a function of emitted proton energy Q_p and angular momentum l , with derivations given in Ref. [41]. Right: experimental S_p value of the ground state of ^{97}In and isotopic S_p trends of neighboring In isotopes from different mass models [35,53–57].

Based on the nonobservation of proton events within the 150-ms decay correlation window, a theoretical partial half-life $T_{1/2}^{\text{theo}} > 600$ ms was determined for $1p$ emission from the $(9/2^+)$ state. Following the derivations outlined in Ref. [41], the theoretical half-lives were converted to Q_p values (see the left plot of Fig. 4). The half-life limits on the $(1/2^-)$ isomer corresponded to a Q_p value ranging between 0.64 and 0.78 MeV, while the lower half-life limit on the $(9/2^+)$ ground state resulted in $Q_p < 0.63$ MeV. The Q_p values could also be obtained from WKB estimates [52], which differ by approximately 0.03 MeV. The smaller Q_p value of the $(9/2^+)$ state compared to that of the $(1/2^-)$ isomer is consistent with the energy ordering of the two states predicted by the SM.

The excitation energy of the $(1/2^-)$ isomer was combined with the isomer’s Q_p value to estimate the ground state’s S_p , which yielded $S_p(9/2^+) = -0.10(19)$ MeV. This result is plotted on the right-side plot of Fig. 4, which includes an extrapolated value from the Atomic Mass Evaluations 2016 (AME) [53] and theoretical mass calculations for the neighboring In isotopes: by Duflo and Zuker (DZ) [54]; from a finite-range droplet model (FRDM) [55]; by Herndl and Brown (HB) [35]; by the Hartree-Fock-Bogoliubov (HFB) model [56]; and by Koura, Tachibana, Uno, and Yamada (KTUY) [57]. A larger discrepancy among different models is seen at $A \leq 97$. The S_p value agrees well with FRDM and HFB calculations, which suggest the ground state of ^{97}In to be nearly stable against proton emission. On the other hand, AME’s mass extrapolations and predictions by DZ, HB, and KTUY mass models converge near $S_p \approx -1.1$ MeV which would lead to $1p$ emission with a partial half-life of ~ 200 ns, similar to ^{93}Ag [22]. The experimental observation of a β -decay branch refutes this low S_p for ^{97}In . Both HFB and FRDM predict more binding in odd- Z isotopes compared to the other models and reflect stronger shell-closure and pn -pairing effects toward ^{100}Sn . Specifically, the experimental S_p values of ^{89}Rh , ^{93}Ag , and ^{97}In best agree with the HFB.

The identification of a proton-unbound $(1/2^-)$ isomer in ^{97}In supports the designation of ^{96}Cd as a waiting-point nucleus

of the rp -process path. Despite the existence of the β -decaying ground state in ^{97}In with a larger spectroscopic factor, proton capture by ^{96}Cd would likely populate the $(1/2^-)$ isomer as the Coulomb barrier penetration rate is proportional to $e^{-l(l+1)}$. Thus the rp -process reaction flow through ^{97}In will be minimal.

In conclusion, the heaviest bound $N = Z - 1$ nuclei ^{91}Pd , ^{95}Cd , ^{97}In , and ^{99}Sn were produced and their β -decay properties were studied at the RIKEN Nishina Center. New and more precise half-life and β -decay endpoint measurements of these nuclei were consistent with the mixed Fermi/Gamow-Teller decays of lighter $T_z = -1/2$ nuclei. The measured values are also consistent with various mass- and shell-model predictions assuming robust $N = Z = 50$ shell closures in ^{100}Sn . In ^{97}In , we report a proton-unbound isomer with spin $(1/2^-)$ and $1.3 < T_{1/2}^{\text{expt}} (\mu\text{s}) < 230$ with a signature of the 421-keV γ ray from the β decay of the proton daughter ^{96}Cd . The proton separation energy of the ground state of ^{97}In was determined from the combination of experimental half-life analysis, a semiempirical theory on proton emission, and shell-model calculations. The resulting S_p value of $-0.10(19)$ MeV is much larger than that of ^{89}Rh and ^{93}Ag , explaining the apparent proton stability of ^{97}In . Despite the proton stability of the $(9/2^+)$ ground state of ^{97}In , the proton instability

of the $(1/2^-)$ establishes ^{96}Cd as the rp -process waiting point.

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