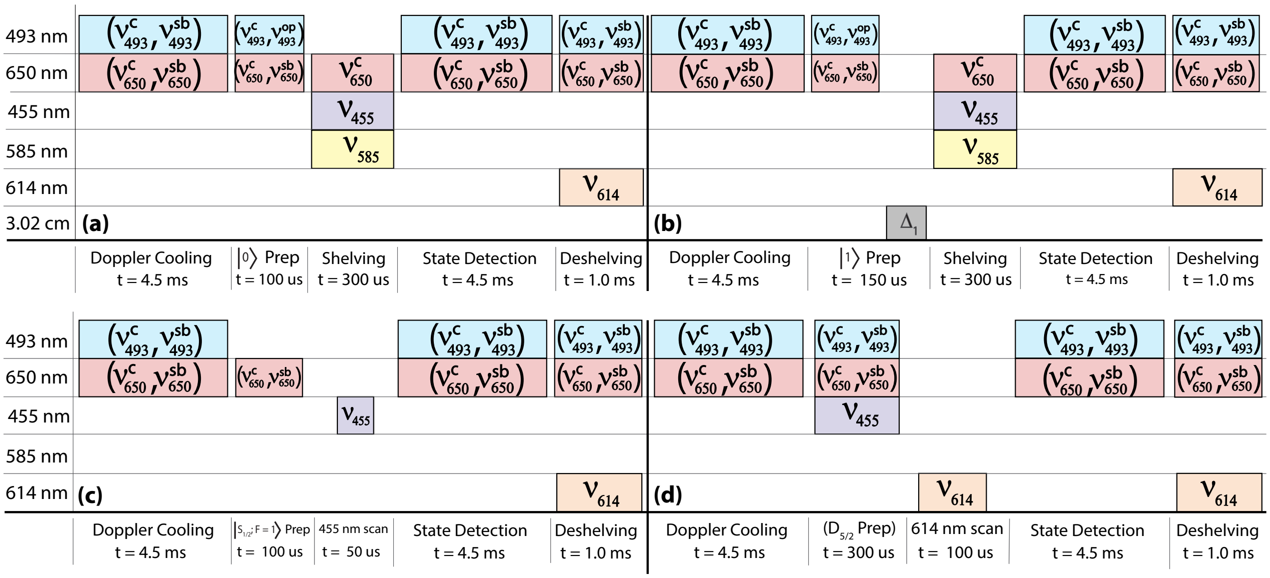
Supplementary notes

^{133}Ba ($\tau_{1/2} \approx 10.5$yr) is manufactured via neutron activation of ^{132}Ba. A 1 mCi sample purchased from Eckert & Ziegler contained an estimated isotopic content of a few percent ^{133}Ba and the remainder primarily ^{132}Ba.



Supplementary Figure 1. Experimental sequences for hyperfine qubit SPAM and ^{133}Ba^{+} spectroscopy. (a) SPAM sequence of $|0\rangle$ state. All lasers are linearly polarized $\approx 45°$ from the magnetic field direction except ν_{455}, linearly polarized along the magnetic field (π-light). (b) SPAM of the $|1\rangle$ state is identical to SPAM of the $|0\rangle$ state with the addition of microwaves near 9.925 GHz after $|0\rangle$ state initialization. (c) Sequence for measuring ^{2}P$_{3/2}$ hyperfine splitting and ^{2}P$_{3/2} \leftrightarrow {}^2S_{1/2}$ isotope shift. (d) Sequence for measuring ^{2}D$_{5/2}$ hyperfine splitting and ^{2}P$_{3/2} \leftrightarrow {}^2D_{5/2}$ isotope shift.

References

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2. Knab, H., Schupp, M. & Werth, G. Precision spectroscopy on trapped radioactive ions: Ground-state hyperfine splittings of ^{133}ba^{+} and ^{131}ba^{+}. *Eur. Lett.* **4**, 1361 (1987).

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Supplementary Table 1. $^{133}Ba^{+}$ hyperfine splittings and isotope shifts relative to $^{138}Ba^{+}$. For reported isotope shifts of the *i*-th electronic transition, defined as $\delta\nu^{i} \equiv \nu^{i}_{133} - \nu^{i}_{138}$, centroid frequencies ($\nu^{i}_{133}$) are calculated from hyperfine measurements and compared to corresponding transitions in $^{138}Ba^{+}$. Bolded values are measurements from this work. $\delta\nu^{585}$ is calculated using previous measurements[1] and results from this work. Hyperfine splittings include a 20 MHz systematic uncertainty due to unresolved Zeeman structure. Isotope shifts include a 28 MHz systematic uncertainty due to unresolved Zeeman structure and wavemeter drift.

Hyperfine Splitting	Frequency (MHz)
$^{2}S_{1/2}$	9925.45355459(10)[2]
$^{2}P_{1/2}$	1810(11)[3]
$^{2}P_{3/2}$	**623 (20)**
$^{2}D_{3/2}$	937 (20)[1]
$^{2}D_{5/2}$	**83 (20)**
Isotope Shift	Frequency (MHz)
$\delta\nu^{493}$	373(4)[3]
$\delta\nu^{455}$	**358(28)**
$\delta\nu^{650}$	198(20)[1]
$\delta\nu^{614}$	**216(28)**
$\delta\nu^{585}$	183(35)