Trapped atomic ions combine the benefits of long storage time and excellent isolation from the environment, providing an attractive physical system for numerous applications in quantum technology [1, 2]. Among the singly-charged ions most commonly investigated, $^{171}\text{Yb}^+$ has found applications in tests of fundamental physics [3, 4], frequency metrology [5, 6], and quantum information processing (QIP) [7, 8, 9]. All of these applications rely on a deep understanding of atomic structure theory, which is guided by and refined through precision measurements [10–13, 14, 15]. However, accurate calculation of the atomic properties for $^{171}\text{Yb}^+$ has proven to be highly challenging due to its large number of valence electrons and complications arising from mixing between electronic configurations [15], making it a particularly interesting system for experimental investigation.

The nuclear spin of $^{171}\text{Yb}^+$ is one half, causing the $^2\text{S}_1/2$ electronic ground state to split into a hyperfine doublet separated by $\sim 12.64$ GHz. Its metastable levels (Fig. 1) include $^2\text{D}_{3/2}$, $^2\text{D}_{5/2}$, and $^2\text{F}_{7/2}$ with lifetimes of 52.7 ms [19], 7.2 ms [20], and $\gtrsim 5.4$ years [21], respectively. The short lifetime of $^2\text{D}_{5/2}$ has made the state less attractive for frequency metrology compared to $^2\text{D}_{3/2}$ [22, 23] and $^2\text{F}_{7/2}$ [5, 25, 26]: experimental measurements on $^2\text{D}_{5/2}$ were last reported more than 20 years ago [27]. Still, measurements with improved precision provide a good benchmark to verify challenging and model-dependent atomic physics calculations. Furthermore, the metastable $^2\text{D}$ levels in $^{171}\text{Yb}^+$ provide extra degrees of freedom for applications in QIP. These finds use in implementing entangling gate operations [28, 29] or to scalably improve qubit measurement fidelities with minimal technical overhead [29].

In this Letter, we report precision measurements on branching ratios, the electric quadrupole (E2) reduced matrix element, the quadratic Zeeman (QZ) coefficient, and the hyperfine splitting of the $^2\text{D}_{5/2}$ state in $^{171}\text{Yb}^+$, with precision up to two orders of magnitude higher than the previous best reported values. Our work focuses on the 411 nm electric quadrupole (E2) $^2\text{S}_{1/2} \leftrightarrow ^2\text{D}_{5/2}$ transition. The $^2\text{D}_{5/2} | F = 3 \rangle$ decays to $^2\text{S}_{1/2} | F = 1 \rangle$ and $^2\text{F}_{7/2} | F = 3, 4 \rangle$ and $^2\text{D}_{5/2} | F = 2 \rangle$ decays to $^2\text{S}_{1/2} | F = 0, 1 \rangle$ and $^2\text{F}_{7/2} | F = 3 \rangle$. A 760 nm laser is used for fast repumping of $^2\text{F}_{7/2}$ to $^2\text{S}_{1/2}$ via $^1\text{D}[3/2]_{3/2}$. Some hyperfine structures and Zeeman sublevels are omitted for clarity.

We report measurements of the branching fraction, hyperfine constant, and second-order Zeeman coefficient of the $^2\text{S}_{1/2}$ level in $^{171}\text{Yb}^+$ with up to two orders-of-magnitude improvement in precision compared to previously reported values. We estimate the electric quadrupole reduced matrix element of the $^2\text{S}_{1/2} \leftrightarrow ^2\text{D}_{5/2}$ transition to be $12.5(4) \times 10^{-20}$. Furthermore, we determine the transition frequency of the $^2\text{F}_{7/2} \leftrightarrow ^1\text{D}[3/2]_{3/2}$ at 760 nm with a $\sim 25$-fold improvement in precision. These measurements provide benchmarks for quantum-many-body atomic-physics calculations and provide valuable data for efforts to improve quantum information processors based on $^{171}\text{Yb}^+$.

FIG. 1. Selected energy levels and transition wavelengths in $^{171}\text{Yb}^+$. Dashed lines show relevant decay channels. Our work focuses on the 411 nm electric quadrupole (E2) $^2\text{S}_{1/2} \leftrightarrow ^2\text{D}_{5/2}$ transition. The $^2\text{D}_{5/2} | F = 3 \rangle$ decays to $^2\text{S}_{1/2} | F = 1 \rangle$ and $^2\text{F}_{7/2} | F = 3, 4 \rangle$ and $^2\text{D}_{5/2} | F = 2 \rangle$ decays to $^2\text{S}_{1/2} | F = 0, 1 \rangle$ and $^2\text{F}_{7/2} | F = 3 \rangle$. A 760 nm laser is used for fast repumping of $^2\text{F}_{7/2}$ to $^2\text{S}_{1/2}$ via $^1\text{D}[3/2]_{3/2}$. Some hyperfine structures and Zeeman sublevels are omitted for clarity.

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The 13.98(1) GHz/T linear Zeeman shift of the $^2S_{1/2}\leftrightarrow^2D_{5/2}$ stretched states $^{32,33}$. A 369.5 nm laser in conjunction with electro-optic modulators (EOMs) is used for Doppler cooling, state preparation, and detection. Separate 369.5 nm laser beams allow for selective measurements of just the population in $^2S_{1/2}\langle F=1\rangle$ using the “standard detection” (SD), or the entire $^2S_{1/2}$ “manifold detection” (MD). A 935 nm laser continuously repumps population which has decayed into the $^2D_{3/2}$ manifold.

We drive the $^2S_{1/2}\leftrightarrow^2D_{5/2}$ transition using an external-cavity diode laser at 411 nm, which is locked to an ultra-low expansion reference cavity with a free spectral range (FSR) of 1.5 GHz, a finesse of approximately 32,000, and a frequency drift rate of $\sim 320$ mHz/s. An additional, cavity-stabilized 760 nm diode laser configured with a cateye reflector $^{36,37}$ is used to provide efficient repumping from the long-lived $^2F_{7/2}$ via $^4D_{3/2}[3/2]_1/2$, which has a lifetime of $\sim 28.6$ ns and decays primarily to the $^2S_{1/2}$ ground states $^{38,39}$. To maximize repumping efficiency, an EOM driven at 5.258 GHz is used to create an additional laser frequency component, matching the hyperfine splittings of 3.620 GHz $^{38,40}$ and 8.8 GHz $^{32}$ for $^2F_{7/2}$ and $^1D_{3/2}[3/2]_3/2$, respectively. The 5.258 GHz microwave signal used to drive the EOM is tuned by maximizing fluorescence counts on the 369.5 nm detection transition while simultaneously illuminating the ion with 411 nm and 760 nm light. With an intensity of $\sim 0.63 \ W/mm^2$, the typical repumping time from the $^2F_{7/2}$ state to the $^2S_{1/2}$ manifold is $\sim 20$ ms, significantly faster than other repumping channels $^{19,20,30,41}$, such as the 638 nm transition to $^1D[5/2]_5/2$. All laser beams are controlled by acousto-optic modulators driven by direct digital synthesis sources referenced to a rubidium frequency standard.

We begin by performing spectroscopy on the $^2S_{1/2}\leftrightarrow^2D_{5/2}$ optical transition near 411 nm, resolving individual hyperfine and Zeeman levels as shown in Figure 2. The ion is initialized in $^2S_{1/2}\langle F=0\rangle$ by optical pumping and optionally transferred to $^2S_{1/2}\langle F=1, m_F=0\rangle$ through application of a microwave $\pi$-pulse. After applying a 411 nm spectroscopy pulse of fixed duration, state-dependent fluorescence is induced by a 369.5 nm (MD) pulse that distinguishes population in the $^2S_{1/2}$ manifold (“bright”) from that in other levels (“dark”). A 20 ms pulse at 760 nm is applied to repump population decayed to $^2F_{7/2}$ at the beginning of each measurement cycle. The center frequency for each transition is extracted by fitting the spectroscopic peaks to Voigt profiles. We measure a frequency of 729.487 752(178) THz for the $^2S_{1/2}\langle F=0\rangle \leftrightarrow^2D_{5/2}\langle F=2, m_F=0\rangle$ transition where the uncertainty corresponds to $\pm 0.1$ pm. We take this conservative value from our wavemeter’s maximum guaranteed accuracy when the measurement wavelength is more than $\pm 200$ nm away from the calibration wavelength at 632 nm. Our frequency measurement is consistent with previous measurements $^{27}$. We note that a more precise measurement of the $^2S_{1/2}\leftrightarrow^2D_{5/2}$ transition in the $^{172}$Yb$^+$ isotope has recently been reported $^{42}$.

The lifetimes and decay branching ratios of $^2D_{5/2}$ are measured by a sequence that initializes in $^2D_{5/2}\langle F=2\rangle$ or $^2D_{5/2}\langle F=3\rangle$ through multiple sequential $\pi$ pulses to different Zeeman sublevels, followed by an immediate detection used for post selection on events with successful transfer, see Fig. 3(a). Following a variable wait time, two final detection pulses determine the population in the $^2S_{1/2}$ states and shelved manifolds. The lifetime is extracted by fitting exponential decays, weighted by QPN, to time-delayed data (Fig. 3(b,c)), where we take the asymptotic limits of the fit as branching ratios. This is the first $^2D_{5/2}$ lifetime and branching fraction measure-
From the text provided, we observe the following key points:

- The decay process of $^2D_{5/2}$ is discussed, with transitions to different Zeeman sublevels identified.
- The concept of branching ratios and lifetimes is introduced, with values provided for different transitions.
- The role of magnetic fields and optical frequencies in the decay processes is emphasized.
- Tables and figures are used to illustrate the decay channels and branching fractions.

The text is rich with scientific notation and references to theoretical predictions. The discussion involves the use of magnetic fields and optical frequencies to study the decay of nuclear states in ions, which is a fundamental aspect of atomic physics.

### Table 1: Branching ratios and lifetimes for the $^2D_{5/2}$ state

| Decay to $^2S_{1/2}$ | Decay to $^2F_{7/2}$ | Lifetime (ms) |
|----------------------|----------------------|---------------|
| This work (exp.) $^{171}$Yb$^+$ $|F = 3\rangle$ | 17.6(4)% | 82.4(4)% | 7.1(4) |
| This work (exp.) $^{171}$Yb$^+$ $|F = 2\rangle$ | 18.4(4)% | 81.6(4)% | 7.4(4) |
| Taylor et al. [20] (1997 exp.) $^{172}$Yb$^+$ | 17(4)% | 83(3)% | 7.2(3) |
| Yu et al. [19] (2000 exp.) $^{174}$Yb$^+$ | - | - | 7.0(4) |
| Fawcett et al. [11] (1991 calc.) Yb II | 19.7% | 80.3% | 5.74 |

### Figure 3: Lifetime and branching ratio measurements

(a) Measurement pulse sequence similar to Fig. 2(a), with an added variable wait time $t$, surrounded by MD and SD pulses for post selection and measurement of branching to the two $^2S_{1/2}$ levels, respectively. A series of $n = 5$ π pulses on the accessible 411 nm transitions to different Zeeman sublevels maximizes the shelving success to $^2D_{5/2}$ $|F = 2\rangle$. Due to the similar Zeeman splitting in $^2S_{1/2}$ $|F = 1\rangle$ and $^2D_{5/2}$ $|F = 3\rangle$ manifolds, a similar $n = 3$ procedure is used to prepare $^2D_{5/2}$ $|F = 3\rangle$ from $^2S_{1/2}$ $|F = 1, m_F = 0\rangle$.

(b) State decay curves as a function of wait time after initialization of the target state. The populations $P(0)$ and $P(1)$ (or) refer to the two hyperfine levels in the $^2S_{1/2}$ ground state manifold, while $P(\text{shelf})$ refers to the shelved population in $^2D_{5/2}$ $|F = 3\rangle$. (c) Overview of the decay channels and branching fractions measured in this work.
field perpendicular to the quantization axis oscillating at the trapping radio frequency is measured to be less than 10 nT, having a negligible impact on our measurements.

The quadratic Zeeman coefficient of the ground state transition is

$$QZ_S = \frac{1}{2} \frac{g_J S - g_I J}{} \frac{2B^2}{h^2 \Delta_{HF,S}} \tag{2}$$

where $\Delta_{HF,S} = 12.642\,812\,118\,466$ GHz is the hyperfine splitting of in the $^2S_{1/2}$ ground state and $g_{J,S}$ is Landé $g$-factor for $^2S_{1/2}$. The quadratic Zeeman coefficient of $^2D_{5/2} | F = 3, m_F = 0 \rangle$ with respect to the $^2S_{1/2} | F = 0 \rangle$ ground state is

$$QZ_{D,F=3} = \frac{1}{4} \frac{g_{J,D} - g_{I} J}{} \frac{2B^2}{h^2 \Delta_{HF,D}} - \left( -\frac{1}{4} \frac{g_{J,S} - g_{I} J}{} \frac{2B^2}{h^2 \Delta_{HF,S}} \right). \tag{3}$$

A recent theory calculation of $g$-factor anomalies in $^{171}$Yb$^+$ gives $g_{J,S} = 2.0031(3)$ and $g_{J,D} = 1.20051(2)$ where the uncertainties are the estimated maximum error of the calculation [16]. Using these values, we evaluate the QZ coefficients to $QZ_S = 0.03110(1)$ Hz/µT$^2$ and $QZ_{D,F=3} = -0.35606(1)$ Hz/µT$^2$, yielding a ratio of the quadratic Zeeman coefficient of -11.448(2), which does not agree with our measurement in Fig. 4. This could indicate an effect not captured by Eqs. (2) and (3), or a discrepancy in the $g_J$ values above. Spectroscopic measurement values of $g_{J,S}$ and $g_{J,D}$ were given by Meggers as 1.998 and 2.102 [27] without specifying uncertainties. Assuming an uncertainty of 0.002 for both values, we evaluate the QZ coefficients and ratio to be $QZ_S = 0.03094(4)$ Hz/µT$^2$, $QZ_{D,F=3} = -0.3571(12)$ Hz/µT$^2$ and -11.54(4), which also does not agree with our observations. Our measurements notwithstanding, we note that Meggers’ ground state $g$-factor value of 1.998 implies a $g$-factor anomaly correction of $-4 \times 10^{-3}$ which is of opposite sign to the $\delta g = +79.8 \times 10^{-3}$ reported in Ref. [16]. This suggests potential issues in previous $g$-factor measurements in Yb$^+$.

The hyperfine splitting of $^2D_{5/2}$ can be deduced from the difference of the transition frequencies between $^2S_{1/2} | F = 1, m_F = 0 \rangle$ and $^2D_{5/2} | F = 2, m_F = 0 \rangle$ as well as $^2D_{5/2} | F = 3, m_F = 0 \rangle$. Electric quadrupole transition selection rules forbid the $^2S_{1/2} | F = 1, m_F = 0 \rangle \leftrightarrow ^2D_{5/2} | F = 2, m_F = 0 \rangle$ transition to be directly driven; therefore we infer its value from measurements of $^2S_{1/2} | F = 1, m_F = 0 \rangle \leftrightarrow ^2D_{5/2} | F = 2, m_F = \pm 1 \rangle$. The frequency of the 411 nm laser is stabilized to the same cavity mode throughout this measurement in order to avoid uncertainties from wavemeter’s accuracy or cavity’s FSR in the determination of hyperfine splitting. All transition frequencies are corrected for cavity drift over the measurement duration of ~4 hours and QZ shifts at the B-field of 441.26(2) µT [45]. From our measurements we extract a hyperfine splitting of $\Delta_{HF,D} = -190.104(3)$ MHz and, correspondingly, a hyperfine constant of $A_{D_{5/2}} = -63.368(1)$ MHz. This measurement is more than two orders of magnitude more precise than the best previously reported value of $-191(2)$ MHz [27], and limited by the precision of the QZ coefficients and fitting errors. Table I summarizes both measured and theoretical values for $A_{D_{5/2}}$, illustrating the difficulty of performing exact calculations for $^{171}$Yb$^+$.

As a final measurement, we determine the $^2D_{7/2} \leftrightarrow ^2D_{3/2}$ transition near 760 nm, used to repump population to $^2S_{1/2}$. We first prepare population in $^2D_{5/2} | F = 2, m_F = 0 \rangle$ using a 411 nm laser pulse, followed by a 10 ms wait time to allow population decay to $^2F_{7/2}$. A subsequent brief application of the 369.5 nm MD beam allows us to remove instances of $^2S_{1/2}$ decay from the data in post processing. A 760 nm laser pulse

\begin{table}[h]
\begin{tabular}{|c|c|}
\hline
Reference & $A_{D_{5/2}}$ (MHz) \\
\hline
This work & (2020 experiment) -63.368(1) \\
Roberts et al. [27] & (1999 experiment) -63.6(7) \\
Nandy et al. [17] & (2014 calculation) -69(6) \\
Porseev et al. [15] & (2012 calculation) -96 \\
Sahoo et al. [13] & (2011 calculation) -48(15) \\
Itano [12] & (2006 calculation) -12.58 \\
\hline
\end{tabular}
\caption{Comparison of measurements and calculations of the $^2D_{5/2}$ hyperfine constant.}
\end{table}
is then applied to induce a transition to the short-lived \(^{1}D[3/2]_{3/2}\) level, which primarily decays to \(^{2}S_1/2\). Successful repump events then yield fluorescence under application of the 369.5 nm MD light. The measured center frequencies are

\[
2^{2}F_{7/2} | J = 3 \rangle \leftrightarrow 1^{1}D[3/2]_{3/2} | F = 1 \rangle : 394.424\,943(20)\text{THz},
\]

\[
2^{2}F_{7/2} | J = 4 \rangle \leftrightarrow 1^{1}D[3/2]_{3/2} | F = 2 \rangle : 394.430\,203(16)\text{THz},
\]

respectively. Here the uncertainties are dominated by line broadening due to our inability to prepare the ion in a specific \(^2F_{7/2}\) Zeeman level as well as the wavemeter’s specified absolute accuracy, which is 10 MHz within ±200 nm of the calibration wavelength of 632 nm. These measurements improve precision by 25-fold over previously reported values [32].

In conclusion, we report measurements of the hyperfine splitting, branching ratios, and quadratic Zeeman coefficient of \(^2D_{5/2}\) in \(^{171}\text{Yb}^+\) with improved precision and investigate the 760 nm transition to efficiently depopulate the \(^2F_{7/2}\) state. These new results can be used to benchmark and verify quantum many-body calculations in \(^{171}\text{Yb}^+\), e.g., in support of studies of PNC physics and the related nuclear anapole moment [13,16,18]. Furthermore, these results provide the necessary characterizations required for improving the measurement fidelity of \(^{171}\text{Yb}^+\) hyperfine qubits as detailed in our separate manuscript Ref. [29].

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