

Sub-Hertz Optical Transitions in Excited Yb^+

Patrick McMillin^{✉,*}, Hassan Farhat[✉], William Liu[✉], and Wesley C. Campbell[✉]

*Department of Physics and Astronomy, University of California Los Angeles, Los Angeles, California, USA,
Challenge Institute for Quantum Computation, University of California Los Angeles, Los Angeles, California, USA,
and Center for Quantum Science and Engineering, University of California Los Angeles, Los Angeles, California, USA*

 (Received 23 February 2026; revised 13 April 2026; accepted 10 June 2026; published 31 July 2026)

We present the observation of three semiforbidden transitions in singly ionized ytterbium from the metastable $^2F_{7/2}^o$ state. Owing to the long lifetimes of both the upper and lower states involved, these transitions are narrow and complement those already frequently used in this atom for quantum information and searches for new physics. We report the absolute frequencies of these electric quadrupole transitions, their isotope shifts, hyperfine structure, and measurements of the quadrupole transition moments. We find that the spontaneous lifetimes of these excited states are limited by slow magnetic dipole emission to lower-lying, odd-parity states, the lowest of which is accessible by laser excitation from $^2F_{7/2}^o$ and is an attractive candidate for hosting a long-lived qubit.

DOI: [10.1103/tz6r-9wj4](https://doi.org/10.1103/tz6r-9wj4)

The ytterbium atom is a popular platform for precision measurement [1–4] and quantum information science [5,6] due mostly to its combination of ease of control and desirable nuclear properties. Both the neutral and singly ionized charge states have multiple metastable states that furnish them with optical transitions with sub-10 Hz natural linewidths, which are used for optical clocks [7,8] and precision tests of physics beyond the standard model (BSM) [9–11].

In particular, the presence of multiple semiforbidden transitions and an abundance of stable isotopes for ytterbium has allowed isotope-shift determinations with high accuracy. A recent study found a statistically significant King plot nonlinearity in a search for signatures of BSM physics [12]. Although the contributions of higher-order nuclear effects to isotope shift nonlinearities are difficult to calculate theoretically, they can be accounted for by including narrow transitions and isotope pairs in the King plot analysis to isolate potential BSM signals in isotope shift measurements. Additional narrow transitions in the same element, in combination with the requisite number of stable isotopes, can be used to construct a higher-dimensional generalized King plot to eliminate poorly bounded nuclear parameters [13].

Further, additional optically narrow transitions can be used as alternative optical clocks and qubit transitions. The extremely long lifetime (≈ 2 yr, [14]) of the metastable $^2F_{7/2}^o$ state in Yb^+ has been identified as a unique feature that can serve as a second ground state of the ion for quantum information processing. A qubit defined on the zero-field hyperfine clock states ($m_F = 0$) in the $^2F_{7/2}^o$ state

of $^{171}\text{Yb}^+$ has been shown to have excellent state preparation and measurement fidelity [15,16] and could be implemented as part of the “*omg* protocol” for trapped ion quantum information processing [17]. However, a key missing ingredient in the current implementation of the *omg* protocol in Yb^+ is qubit-motion coupling of the qubit defined in the substructure of the metastable electronic state. A narrow transition from the $^2F_{7/2}^o$ state to the dozens of excited metastable states arising from the low-lying, multielectron configuration of $4f^{13}(^2F_{7/2}^o)5d6s$ can be utilized to perform the desired qubit-entangling operation.

In this Letter, we demonstrate the presence of additional sub-hertz optical transitions between metastable levels of Yb^+ . We report laser spectroscopy of electric-quadrupole (*E2*) transitions from $^2F_{7/2}^o$ to three J_1K -coupled states, presented in Fig. 1, at red and near-infrared wavelengths with $J \geq 9/2$ that are within the tuning range of the titanium-sapphire laser used for this Letter (see Ref. [18] for a discussion of other potentially metastable levels, including the recently confirmed $^3[3/2]_{5/2}^o$ metastable state at 26759.02 cm^{-1} [23]). Isotope-shift measurements for these transitions are provided, as well as the $^{171}\text{Yb}^+$ hyperfine *A* coefficients of the excited levels. We report the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition moment and radiative lifetime of the excited state showing a sub-hertz optical transition linewidth. Further, we report the upper limits on the *E2* Einstein *A* coefficients of the $^2F_{7/2}^o \leftrightarrow ^3[9/2]_{9/2}^o$ and $^2F_{7/2}^o \leftrightarrow ^1[11/2]_{11/2}^o$ transitions. These results, which demonstrate transitions with linewidths narrower than those involving the 2D_J states in Yb^+ , suggest that metastable Yb^+ may prove useful for applications in clockwork, precision measurement, and quantum information

*Contact author: pmcmi17@physics.ucla.edu

processing. Because of the small $E2$ transition moments, we have found that the spontaneous emission lifetime of the excited states investigated here are limited by magnetic dipole ($M1$) decays to other states with the same electron configuration.

The selected J_1K -coupled excited states in this Letter have $J > 7/2$, odd parity, and the same $4f^{13}$ core-electron configuration as $^2F_{7/2}^o$. Similar to the $^2S_{1/2} \leftrightarrow ^2D_J$ transitions, each $E2$ transition from the $^2F_{7/2}^o$ state to a J_1K -coupled excited state involves a single-electron excitation from the $6s$ to $5d$ subshells. However, unlike transitions originating from the $^2S_{1/2}$ state, these transitions occur between states containing a partially filled $4f$ subshell and can be spin-forbidden for the outer electron pair.

For the spectroscopic identification of these new transitions, single ytterbium ions were trapped in a radio frequency Paul trap with a magnetic field of 4.00(7) G applied to define the quantization axis and destabilize coherent dark states. To initialize the ion in the $^2F_{7/2}^o$ manifold, laser light is applied to transfer the population from the $^2S_{1/2}$ state to the $^2D_{5/2}$ state ($\tau = 7.2$ ms), which favorably leaks to the $^2F_{7/2}^o$ state with a branching fraction of 0.83(3) [24]. The linear polarization and k vector of this light are both set perpendicular to the magnetic field ($\mathbf{k} \perp \mathbf{B}$ and $\hat{\mathbf{e}} \perp \mathbf{B}$) to select for $\Delta m_J = \pm 2$ transitions [25]. Excited state readout is facilitated by a laser tone driving the $^2F_{7/2}^o \leftrightarrow ^1[3/2]_{3/2}^o$ $E2$ transition, which causes the population to preferentially decay back to the $^2S_{1/2}$ ground state. The $^2F_{7/2}^o$ population is measured by observing the presence or lack of laser-induced fluorescence on the strong $^2S_{1/2} \leftrightarrow ^2P_{1/2}^o$ transition.

We implement a pulsed spectroscopy sequence with the spectroscopy laser's frequency controlled by a double-passed acousto-optic modulator to determine the frequency of the transitions. Ion population is first prepared in the $^2F_{7/2}^o$ state by exciting the $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ transition for 200 ms. A 5 ms spectroscopy pulse at variable frequency then drives the population from $^2F_{7/2}^o$ to an excited J_1K -coupled state. Spectroscopy light propagates with $\mathbf{k} \perp \mathbf{B}$ and linear polarization $\hat{\mathbf{e}} \parallel \mathbf{B}$, selecting for $\Delta m_J = \pm 1$ transitions [25]. We then immediately depopulate the $^2F_{7/2}^o$ state by driving the $^2F_{7/2}^o \leftrightarrow ^1[3/2]_{3/2}^o$ transition for 50 ms and then perform state detection with 1 ms of light applied on the $^2S_{1/2} \leftrightarrow ^2P_{1/2}^o$ transition. Ion fluorescence indicates failed transfer out of the $^2F_{7/2}^o$ manifold, while the absence of ion fluorescence indicates successful transfer.

Recording the number of failed and successful events as a function of laser frequency generates the population remaining in the $^2F_{7/2}^o$ manifold. Detailed linescans for the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition in each stable even isotope of Yb^+ are shown in Fig. 2. The uncertainties of the transition frequencies shown in Table I are on the order of 10s of MHz

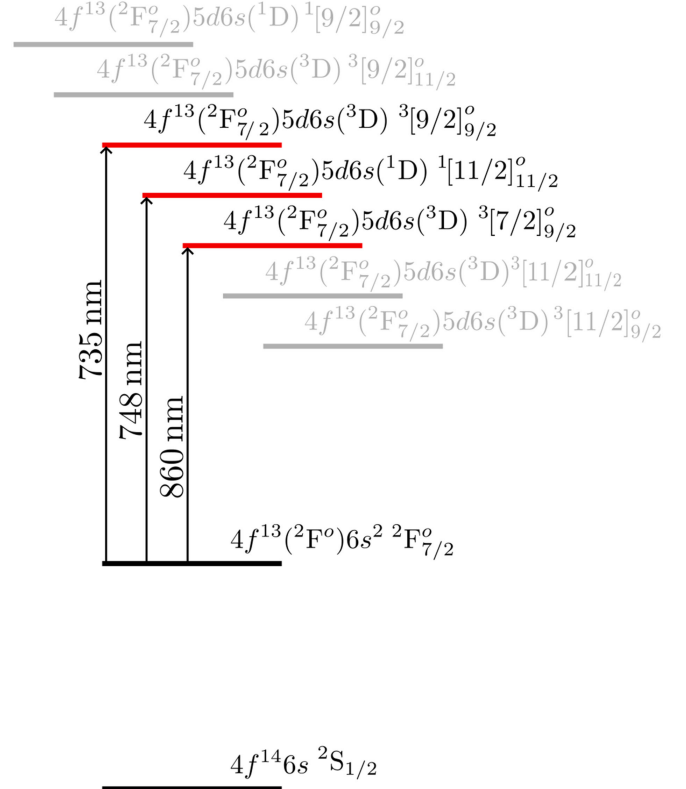


FIG. 1. Level diagram displaying driven transitions from the $^2F_{7/2}^o$ state in Yb^+ . The upper states' electron configuration $4f^{13}(^2F_{7/2}^o)5d6s$ indicates a state where the $4f$ shell has one hole forming a $^2F_{7/2}^o$ core and two additional electrons occupy the $5d$ and $6s$ orbitals. Term symbols that follow the format $^{2S_o+1}[K]_J$ indicate that the state follows the J_1K -coupling scheme, in which the angular momenta of the core couple to that of the outer electrons. The population in each highlighted excited state is read out via depletion of the remaining population in the $^2F_{7/2}^o$ via the $^2F_{7/2}^o \leftrightarrow ^1[3/2]_{3/2}^o$ transition, which then decays back to the ground state.

primarily due to the incoherent population preparation in the Zeeman sublevels of $^2F_{7/2}^o$ state and the laser line-width (≈ 2 MHz).

A King plot for the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$, $^2F_{7/2}^o \leftrightarrow ^1[11/2]_{11/2}^o$, and $^2F_{7/2}^o \leftrightarrow ^3[9/2]_{9/2}^o$ transitions compared to the $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ transition for even isotopes of Yb^+ is presented in Fig. 3. For $^{168}\text{Yb}^+$, only the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition was measured. The King plot shows consistency with a linear dependence, as anticipated at this level of precision if any nonlinearity is similar in scale to previous studies [9,12]; future spectroscopy with high frequency precision may be used to probe isotope-shift nonlinearity. For example, a recent experiment [9] produced a generalized three-dimensional King plot for ytterbium using the $^2S_{1/2} \leftrightarrow ^2D_{3/2}$ and $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ transitions in Yb^+ and the $^1S_0 \leftrightarrow ^3P_0$ in Yb . Variants of this could be constructed

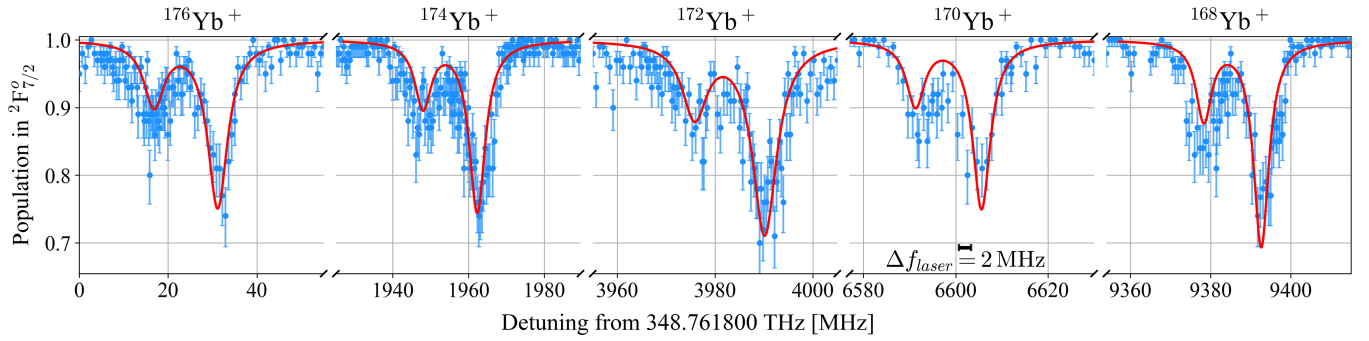


FIG. 2. $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$, $|\Delta m_J| = 1$ transition spectra across all stable even isotopes of Yb^+ . The population distribution across the Zeeman sublevels of the $^2F_{7/2}^o$ state results in the asymmetry between the $\Delta m_J = -1$ (lower frequency) feature and $\Delta m_J = +1$ (higher frequency) feature in each isotope. The linewidth of our spectroscopy laser is represented by the scale bar in the $^{170}\text{Yb}^+$ subplot.

by substituting in one of these new transitions, which may help us to understand the various contributions to the nonlinearity. Alternatively, a generalized four-dimensional King plot is possible with the addition of one more ytterbium isotope and a fourth, narrow transition [13], such as any of the three presented here.

In order to perform hyperfine spectroscopy of the J_1K -coupled states in $^{171}\text{Yb}^+$, we leverage hyperfine selection rules. We simplify the determination of the sign of the excited states' hyperfine coefficients by initially populating only one hyperfine manifold ($F = 3$) in the $^2F_{7/2}^o$ state via the $^2S_{1/2}, F = 0 \rightarrow ^2D_{5/2}, F = 2$ transition. We measured the hyperfine A coefficients of the $^3[7/2]_{9/2}^o$, $^1[11/2]_{11/2}^o$, and $^3[9/2]_{9/2}^o$ states to be 2131(5), 288(3), and 1616 (4) MHz, respectively. Hyperfine splittings were then verified by populating the $F = 4$ manifold in the $^2F_{7/2}^o$ state and driving the available transitions. Figure 4 displays the centroid frequencies of the transitions, along with the hyperfine splittings of the excited states.

The odd-parity excited states investigated here can decay via magnetic dipole ($M1$) transitions to other odd-parity states, potentially causing the population to become stuck in inaccessible states. Because of the imperfect description of these states by the J_1K coupling scheme, second-leading-order contributions to the terms must be considered

when evaluating the $M1$ decay pathways. The $M1$ transition strengths can be calculated by expressing the magnetic dipole operator μ as the sum of operators μ_c and μ_o that act on the core and outer electrons, respectively. The calculated $M1$ lifetimes of the $^3[7/2]_{9/2}^o$, $^1[11/2]_{11/2}^o$, and $^3[9/2]_{9/2}^o$ states are 9.9, 0.41, and 0.24 sec, respectively (see Supplemental Material [18] for details). Many $M1$ decay products are themselves metastable, as an $M1$ decay event frequently leads to “dark” ions, which have taken up to 45 min to re-enter the Doppler cooling cycle. The shorter $M1$ lifetimes of the $^1[11/2]_{11/2}^o$ and $^3[9/2]_{9/2}^o$ states, along with the long $M1$ lifetimes of their decay products, hampered our ability to measure a finite $E2$ moment for these transitions.

We measured the $E2$ Einstein A coefficient of the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition using our laser beam parameters and the measured transfer rate. Transfer rate experiments are identical to frequency determination experiments in their sequence structure. However, the spectroscopy laser frequency remains fixed while the pulse time is varied, as shown in Fig. 6. The remaining population follows simple rate equations as the spectroscopy laser linewidth is much larger than the transition linewidth.

Results are fitted to a two-level system of rate equations representing the population in the $^2F_{7/2}^o$ state (N_F) and the population in the excited $^3[7/2]_{9/2}^o$ state (N_B),

$$\begin{aligned}\dot{N}_F(t) &= A_{E2}N_B(t) + B\rho(\omega)[N_B(t) - fN_F(t)] \\ \dot{N}_B(t) &= -\dot{N}_F(t),\end{aligned}$$

where $\rho(\omega)$ is the energy spectral density of the applied laser light and f is the fraction of population addressed by the laser (see Supplemental Material [18] for details). By applying this model to the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition, we measure the $E2$ Einstein A coefficient to be $A_{E2} = 6(3) \times 10^{-3} \text{ s}^{-1}$ and the corresponding reduced matrix element (reduced to J) to be $1.6(9) e a_0^2$. Using this reduced matrix element as an upper bound for the $^2F_{7/2}^o \leftrightarrow ^3[9/2]_{9/2}^o$

TABLE I. Absolute frequency measurements of the newly observed transitions from $^2F_{7/2}^o$ to each indicated excited state. The 30 MHz uncertainty corresponds to the maximum energy shift between occupied Zeeman sublevels given our applied magnetic field.

Isotope	$^3[7/2]_{9/2}^o$ (THz)	$^1[11/2]_{11/2}^o$ (THz)	$^3[9/2]_{9/2}^o$ (THz)
168	348.77119(3)	Not measured	Not measured
170	348.76841(3)	400.71232(3)	407.73232(3)
172	348.76579(3)	400.70937(3)	407.72974(3)
174	348.76376(3)	400.70708(3)	407.72774(3)
176	348.76183(3)	400.70490(3)	407.72583(3)

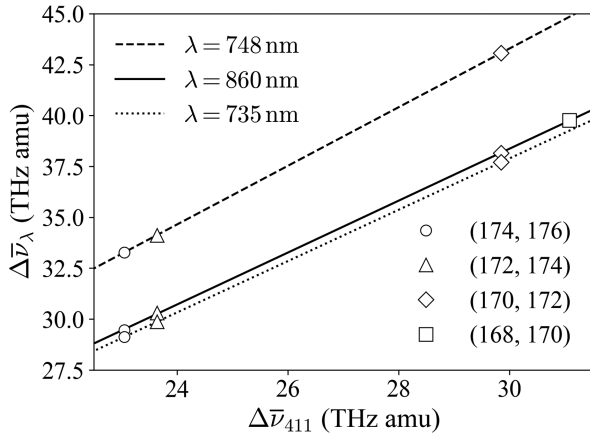


FIG. 3. King plot comparing the modified isotope shifts of the driven transitions against those of the $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ measured by Door *et al.* [26]. Note that only the $^2F_{7/2}^o \leftrightarrow ^3[7/2]_{9/2}^o$ transition was measured for $^{168}\text{Yb}^+$.

transition, we find an estimate for the $E2$ Einstein A coefficient to be $\approx 10 \times 10^{-3} \text{ s}^{-1}$. For the $^2F_{7/2}^o \leftrightarrow ^1[11/2]_{11/2}^o$ transition, we estimate an $E2$ Einstein A coefficient of $\approx 1 \text{ s}^{-1}$. The reported $E2$ transition rates show that the radiative lifetimes of the excited states are

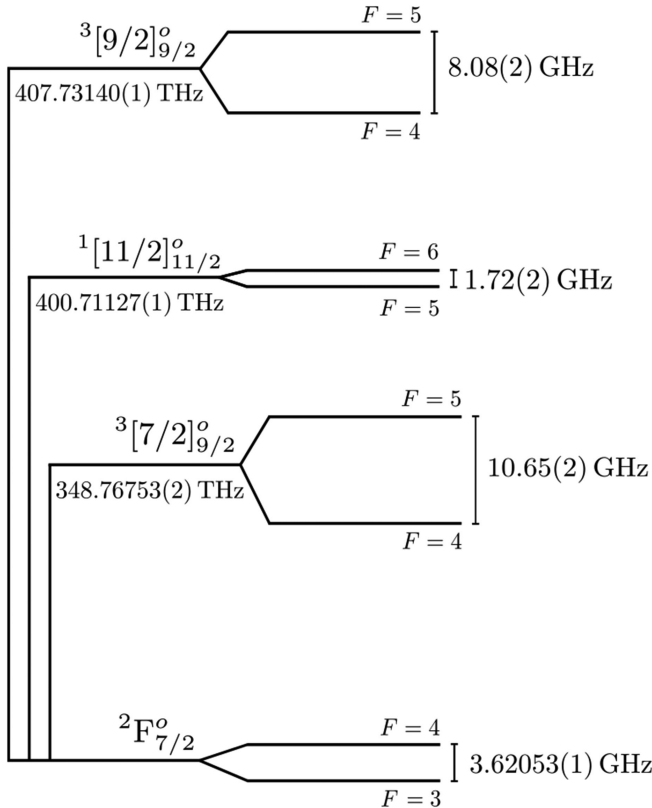


FIG. 4. Hyperfine structure of $^{171}\text{Yb}^+$ of the excited state and the $^2F_{7/2}$. The centroid frequency of each transition is presented along with the hyperfine splitting of each excited state.

limited by $M1$ decays to other J_1K -coupled states. The measured transition matrix element is an order of magnitude smaller than that of the $^2S_{1/2} \leftrightarrow ^2D_{3/2}$ and $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ transitions ($10.1 ea_0^2$ and $12.5 ea_0^2$, respectively [24,27]). Because of the change in electron configuration in these transitions, we expect the $M1$ coupling between the $^2F_{7/2}^o$ and excited J_1K -coupled states to be much smaller than the contributions of the $E2$ coupling.

The main limitation in the presented measurements is the lack of single Zeeman state preparation in the $^2F_{7/2}^o$ state, which can be overcome in the spinless isotopes by applying an additional laser tone on the $^2F_{7/2}^o \leftrightarrow ^1[3/2]_{3/2}^o$ transition. If the light is applied during state preparation of the $^2F_{7/2}^o$ state such that $\Delta m_J = \pm 2$ transitions are not allowed (i.e., $\mathbf{k} \perp \mathbf{B}$, $\hat{\mathbf{e}} \parallel \mathbf{B}$), the population will be stranded in the $m_J = \pm 7/2$ sublevels in the $^2F_{7/2}^o$ state. In $^{171}\text{Yb}^+$, heralded state preparation can be performed by applying resonant microwaves in conjunction with highly manifold-selective laser tones to prepare the $m_F = 0$ state with high fidelity [15]. For preparing the m -type qubit in $^{171}\text{Yb}^+$, this step is already necessary when populating the $^2F_{7/2}^o$ incoherently.

Frequent decays from the excited J_1K -coupled states may necessitate the use of additional repump tones for some applications to ensure that optical cycling can be recovered after an $M1$ decay event. Utilizing a metastable state to perform qubit-motion coupling may be more attractive than a two-photon, $E1$ -mediated transition due to the high number of spontaneous emission channels available to the excited even-parity (J_1, J_2)-coupled states

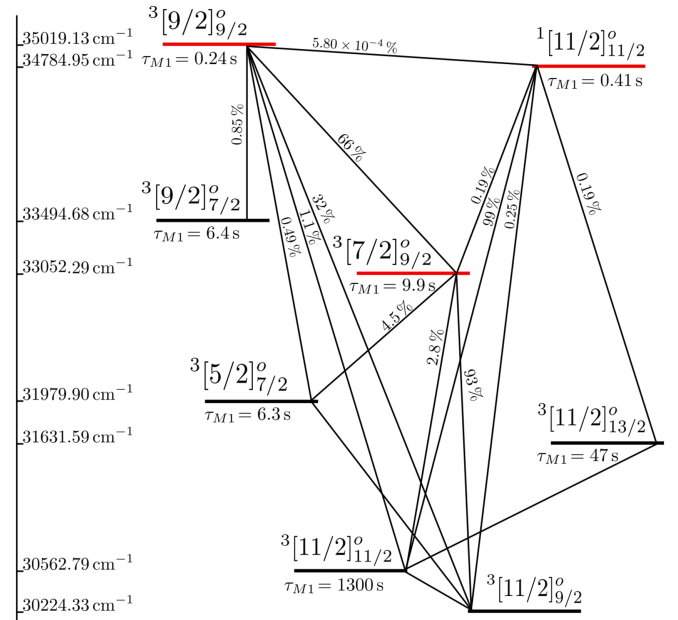


FIG. 5. $M1$ decay pathways from $^3[7/2]_{9/2}^o$, $^1[11/2]_{11/2}^o$, and $^3[9/2]_{9/2}^o$. The presented $M1$ lifetimes and branching ratios are theoretically calculated. The energy levels are sourced from [28].

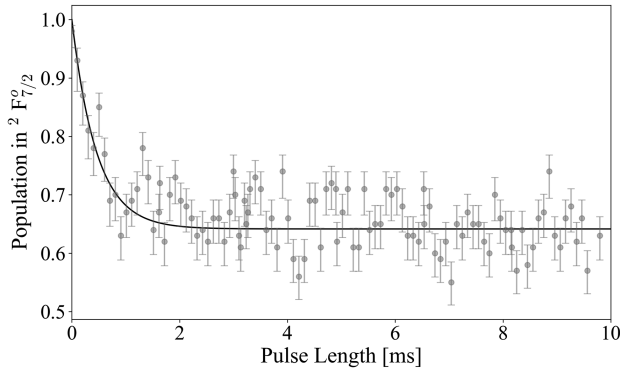


FIG. 6. Population remaining in the $2F_{7/2}^o$ state as a function of spectroscopy laser illumination time. The data are fit to simple rate equations, allowing for extraction of the transition moment when combined with our laser parameters.

in Yb^+ . The $M1$ decay pathways from the higher-lying J_1K states may prove to have large qubit-subspace leakage errors. Therefore, the $2F_{7/2}^o \leftrightarrow 3[11/2]_{11/2}^o$ and $2F_{7/2}^o \leftrightarrow 3[11/2]_{9/2}^o$ transitions may be particularly attractive candidates for implementing qubit-motion coupling owing to the readily available laser sources and the extremely long $M1$ lifetimes of these two low-lying excited states, which are depicted as the two lowest levels in Fig. 5.

In order to further reduce the systematics caused by higher-order standard model terms in isotope shift spectroscopy, more transitions must be added to the King-plot analysis in conjunction with additional isotope pairs. Yb has five stable spinless isotopes, but it also has many long-lived spinless radioactive isotopes. In particular ^{164}Yb , ^{166}Yb , and ^{178}Yb have half-lives of 76 min, 57 h, and 74 min, respectively [29], which are sufficient for laser spectroscopy. The fact that single ions can be isolated and trapped means that atomic sources for these radioactive isotopes can remain at low activity [30] and be efficiently utilized [31].

Acknowledgments—The authors acknowledge Michael Köhl and Vladan Vuletić for helpful discussions. This work was supported by ARO No. W911NF-24-S-0004, NSF No. PHY-2207985 and No. OMA-2016245, and the Gordon and Betty Moore Foundation DOI: 10.37807/GBMF11566.

Data availability—The data that support the findings of this article are publicly available at Ref. [32].

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