Microwave spectroscopy of the 1snp^3P_J fine structure of high Rydberg states in ^4He

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The 1snp^3P_J fine structure of high Rydberg states in helium has been measured by microwave spectroscopy of single-photon transitions from 1snp^3S_J levels in pulsed supersonic beams. For states with principal quantum numbers in the range from n = 34 to 36, the J = 0 → 2 and J = 1 → 2 fine structure intervals were both observed. For values of n between 45 and 51 only the larger J = 0 → 2 interval was resolved. The experimental results are in good agreement with theoretical predictions. Detailed characterization of residual uncancelled electric and magnetic fields in the experimental apparatus, and calculations of the Stark and Zeeman structures of the Rydberg states in weak fields, were used to quantify systematic contributions to the uncertainties in the measurements.

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I. INTRODUCTION

High-resolution spectroscopy of the energy-level structure of simple atomic systems containing two leptons – such as the helium atom, positronium atom, or hydrogen molecule – plays an important role in experimental tests of bound-state quantum electrodynamics (QED) [1, 2]. In the case of the helium atom, this has motivated a wide range of studies from the vacuum-ultraviolet to the infrared and microwave regions of the electromagnetic spectrum [3–8]. In particular, since being identified as a sensitive probe of the fine-structure constant, α, [9] the fine structure of the 1s2p^3P term [10] has attracted significant theoretical and experimental interest [e.g., 11–14]. Similarly, precise measurements and calculations of the 1snp^3P_J fine structure, for principal quantum numbers n ≤ 10, have also been performed [17–21].

For higher values of n, the fine structure intervals and the magnitude of the corresponding QED corrections reduce significantly. However, precise measurements of the fine structure of the triplet Rydberg states are essential for a number of recently developed experiments. These include experiments that involve coupling Rydberg helium atoms to chip-based microwave circuits for applications in hybrid cavity quantum electrodynamics and quantum information processing [22–24], and studies of Förster resonance energy transfer in collisions with polar ground-state molecules [25]. In these cases, Rydberg states with n = 30 to 70 and values of the electron orbital angular momentum quantum number 0 ≤ ℓ ≤ (n − 1) [20] are employed. In general these experiments take advantage of: (i) the minimal detrimental effects caused by helium adsorption within the experimental apparatus, particularly in cryogenic environments [27–28], (ii) the opportunity to control the translational motion and trap Rydberg helium atoms using inhomogeneous electric fields [29–33], and (iii) the possibility of efficiently implementing resonance enhanced two-color two-photon Rydberg state excitation schemes using readily available continuous wave (cw) diode lasers.

For triplet states with ℓ > 0, the fine structure that arises as a result of the spin-orbit interaction must be accounted for when precisely determining transition frequencies and transition dipole moments, or when calculating the Stark structure of low-|M_J| sublevels. The fine structure must also be considered when characterizing dephasing and decoherence in microwave transitions between these states, and in implementing microwave dressing schemes to reduce the sensitivity of low-ℓ states to static or radio-frequency electric fields [34].

The fine structure of the 1snp^3P_J levels in helium is typically referred to in terms of ν_{0,2} and ν_{1,2} – the frequencies associated with the intervals between the J = 0 and J = 2 levels, and the J = 1 and J = 2 levels, respectively. These intervals scale with n^3, whereas the sensitivity of the corresponding levels to electric fields increases with n^7. Consequently, for high values of n even small stray electric or magnetic fields can cause Stark or Zeeman shifts that are comparable to the fine structure.

Here we report studies of single-photon 1snp^3S_J → 1snp^3P_J transitions, for values of n between 34 and 51. From the measured spectra ν_{0,2} and ν_{1,2} were both determined for n = 34 to 36. In the spectra recorded for values of n between 45 and 51 the smaller ν_{1,2} interval could not be resolved. In these cases the interval ν_{0,2} ≃ ν_{0,2} was measured. This interval corresponds to the energy difference between the J = 0 level and the average energy of the unresolved J = 1 and J = 2 levels weighted by their multiplicities, 2J + 1.

This article is structured as follows: Sec. [II] provides an overview of the calculations employed to determine the intervals between the 1snp^3P_J levels and the effects of weak magnetic and electric fields on this fine structure. Sec. [III] contains a description of the experimental apparatus, the techniques used to prepare, probe and detect the high Rydberg states, and the methods used to characterize and minimize stray electric and magnetic fields. The results of the measurements of the fine-structure of the 1snp^3P_J Rydberg levels are presented in Sec. [IV].
Sec. I a discussion of the systematic uncertainties in the measurements is provided and conclusions are drawn.

II. CALCULATIONS

A. Rydberg energy level structure

In the absence of external electric or magnetic fields the Hamiltonian, \( \hat{H}_0 \), associated with a single excited Rydberg electron in the helium atom is diagonal in an \( |nLSJM\rangle \) basis, where \( n \) is the principal quantum number, \( L \equiv \ell \) is the total electron orbital angular momentum quantum number, \( S \) is the total electron spin quantum number, \( J \) is the total angular momentum quantum number, and \( M_J \) is the projection of \( \vec{J} \) onto the laboratory quantization axis. The binding energy, \( W(n, \delta) \), of each eigenstate is given by

\[
W(n, \delta) = - \frac{R_{\text{He}}}{n^2} \frac{\mu_{\text{He}}}{(n-\delta)^2},
\]

where \( h \) is the Planck constant, \( c \) is the speed of light in vacuum, \( \delta \) is the quantum defect, and \( \mu_{\text{He}} \) is the Rydberg constant for helium corrected for the reduced mass, \( \mu_{\text{He}} = m_e M_{\text{He}}^+ / (M_{\text{He}}^+ + m_e) \), where \( m_e \) is the electron mass and \( M_{\text{He}}^+ \) is the mass of the He\(^{+}\) ion core. The value of the quantum defect depends on the quantum numbers \( n \), \( L \), \( S \), and \( J \), and can be determined using the recursive Ritz expansion \[35\]

\[
\delta(n) = c_0 + \frac{c_2}{(n-\delta)^2} + \frac{c_4}{(n-\delta)^4} + \cdots,
\]

where the terms in curved (curly) brackets represent Wigner 3J (6J) symbols. As in Ref. [39],

\[
g'_L = \sqrt{\frac{(2L+1)(L+1)}{6}} g_L,
\]

and

\[
g'_S = \sqrt{\frac{(2S+1)(S+1)}{6}} g_S.
\]

To calculate the energy level structure in the presence of a magnetic field the eigenvalues of the complete matrix containing the zero-field and Zeeman elements are then determined. The intensity of electric dipole transitions between pairs of Zeeman sublevels can be calculated by summing and squaring the contributions from each component of each \( \langle n', L', S', J', M'_J | e \hat{\vec{\gamma}} | n, L, S, J, M_J \rangle \) electric dipole transition moment, weighted by the corresponding elements of the initial and final state eigenvectors [40].

To evaluate the effects of weak magnetic fields on the 1s\(^3\)S\(_1\) and 1snp\(^3\)P\(_J\) levels, the quantum defects in Ref. [40] were employed. Figure 1 shows the calculated energy level structure of the 1s45p\(^3\)P\(_J\) term for magnetic fields up to 50 \( \mu \)T. In this figure, both the large, \( \nu_{0,2} \), and small, \( \nu_{1,2} \), fine structure intervals can be identified in low fields. As the magnetic field increases the smaller interval becomes obscured by the Zeeman splitting of the 1s45p\(^3\)P\(_1\) and 1s45p\(^3\)P\(_2\) levels. The 1s45p\(^3\)P\(_1\) (\( M_J = -1 \)) and 1s45p\(^3\)P\(_2\) (\( M_J = +2 \)) sublevels cross in a magnetic field of \( B_z = 3 \) \( \mu \)T. Similar calculations indicate that the equivalent crossings occur at 6 \( \mu \)T and

where the values of \( c_i \) for each set of \( L \), \( S \), and \( J \) are obtained by fitting Eq. [2] to precise calculations \[36\] or measurements \[37, 38\] of the corresponding Rydberg series. Higher order corrections to Eq. [1] have been reported \[39\] but their contributions are below the spectral resolution of the experimental apparatus used here.

B. Magnetic field effects

In the presence of a weak magnetic field, \( \vec{B} = (0, 0, B_z) \), the total Hamiltonian for the Rydberg system is

\[
\hat{H} = \hat{H}_0 + \hat{H}_Z.
\]

The first-order perturbation by the magnetic field is

\[
\hat{H}_Z = \mu_B (g_L \vec{L} + g_S \vec{S}) \cdot \vec{B} = \mu_B g_L B_z L_z + \mu_B g_S B_z S_z,
\]

where \( \mu_B \) is the Bohr magneton, \( g_s \approx 2.002319 \) is the electron spin \( g \)-factor, and \( g_L = 1 - m_e / M_{\text{He}}^+ \) is the electron orbital \( g \)-factor. To first order in \( \alpha \), the matrix elements associated with \( \hat{H}_Z \) are \[39\]
with the radial integrals, \( \langle n' L' | r | n L \rangle \), calculated using the Numerov method [41]. For each value of \( n \) of interest calculations were performed using three separate basis sets with \( | M_J | = 0, 1 \) or 2, and all of the corresponding Rydberg states within the range from \( n = 5 \) to \( n + 5 \). A Stark map depicting the energy level structure of the 1s36p \(^3\)P term in fields up to 120 mV cm\(^{-1}\) is displayed in Fig. (3a). For electric fields of \( \sim 50 \) mV cm\(^{-1}\) the fine-structure components all exhibit similar Stark shifts of \( \sim 320 \) kHz. In fields of 10 mV cm\(^{-1}\) this shift is 13 kHz and the corresponding changes in the intervals \( \nu_{0,2} \) and \( \nu_{1,2} \) between the levels are \( \pm 0.2 \) kHz and \( \pm 0.4 \) kHz, respectively. In the case of the 1s35p \(^3\)P term, the Stark shifts of the individual levels in a field of 10 mV cm\(^{-1}\) are \( \sim 250 \) kHz and the changes in \( \nu_{0,2} \) and \( \nu_{1,2} \) are \( \pm 2 \) kHz and \( \pm 5 \) kHz, respectively.

### III. EXPERIMENTAL TECHNIQUES

#### A. Rydberg helium spectroscopy

A schematic diagram of the apparatus used in the experiments reported here is displayed in Fig. 2. Pulsed supersonic beams of helium atoms in the metastable 1s2s \(^3\)S\(_1\) level were generated in an electric discharge at the exit of a pulsed valve operated at a repetition rate of 50 Hz. To maximise shot-to-shot stability, the discharge was seeded with electrons that emanated from a heated tungsten filament located \( \sim 20 \) mm downstream from the exit of the valve [42]. The beam passed through a 2-mm-diameter skimmer after which stray ions produced in the discharge were removed by the electric field generated between a pair of parallel plate electrodes (region I. in Fig. 3). The beam then proceeded to the photoexcitation region of the apparatus (region II. in Fig. 3), which was formed by a further two parallel 70 \( \times \) 70 mm copper electrodes separated by 8.3 mm. Between these electrodes, co-propagating cw ultraviolet (\( \lambda = 388.9751 \) nm) and infrared (\( \lambda \sim 788 \) nm) laser beams were focused to intersect the collimated atomic beam. To excite a spatially localized bunch of atoms to Rydberg states, a pulsed electric field of \( \sim 1 \) V cm\(^{-1}\) was applied in the excitation region for a period of 3 \( \mu \)s. The lasers were tuned to drive 1s2s \(^3\)S\(_1\) \( \rightarrow \) 1s3p \(^3\)P\(_1\) \( \rightarrow \) 1sns \(^3\)S\(_1\) transitions in this field.

After laser photoexcitation spectroscopy of single-photon 1sns \(^3\)S\(_1\) \( \rightarrow \) 1snp \(^3\)P\(_J\) transitions was performed using a pulsed source of microwave radiation. The microwave pulses had a typical duration of 5 \( \mu \)s and entered the vacuum chamber 1 \( \mu \)s after the atoms were excited to the Rydberg levels. Population transfer between the Rydberg levels by the microwave radiation was identified by

### C. Electric field effects

The effect that an electric field, \( \vec{F} = (0, 0, F_z) \), has on the Rydberg levels is determined by calculating the eigenvalues of the Hamiltonian

\[
\hat{H} = \hat{H}_0 + \hat{H}_S,
\]

where,

\[
\hat{H}_S = eF_z r \cos \theta.
\]

In the \( | n L S J M_J \rangle \) basis the corresponding matrix elements can be expressed as [41].

\[
\langle n' L' | r | n L \rangle \propto \sum_{M_L = M_J + M_S} \left[ (-1)^{L' - L - 2S + 2M_J} \sqrt{(2J + 1)(2J' + 1)} \left( \begin{array}{ccc} L & S & J' \\ M_L & M_J - M_L & -M_J \end{array} \right) \right] \times \left( \begin{array}{ccc} L' & 1 & L \\ -M'_L & 0 & M_L \end{array} \right),
\]

strates the level to which stray magnetic fields must be reduced in order for the small fine-structure intervals to be resolved.
FIG. 2. (a) Calculated Stark structure of the 1s36p 3P term in helium. The vertical axis is displayed with respect to the energy of the J = 2 level in zero electric field. The calculated interval, ν_{0,2}, between the 1s36p 3P_0 and 1s36p 3P_2 levels is shown in panel (b), while the smaller interval, ν_{1,2}, between the 1s36p 3P_1 and 1s36p 3P_2 levels is shown in panel (c). The three curves in panel (b) represent the intervals between the 1s36p 3P_0 sublevel with M_J = 0 and the 1s36p 3P_2 sublevels with |M_J| = 0, 1 and 2. In panel (c) the three continuous (dashed) curves represent the intervals between the 1s36p 3P_1 sublevel with M_J = 0 ((M_J) = 1) and the 1s36p 3P_2 sublevels with |M_J| = 0, 1 and 2.

state-selective ramped-electric-field ionization between a third electrode pair (region III, in Fig. 3). The resulting electrons were then accelerated onto a microchannel plate (MCP) detector. The time-dependent ionization electric field was configured to ensure that the signals corresponding to field ionization of the initial and final Rydberg states were clearly distinguishable.

B. Magnetic field control and characterization

The magnetic field in the excitation region of the apparatus was controlled using three pairs of parallel coils wound directly onto the outside of the vacuum chamber and operated in a Helmholtz configuration. Each coil was 160 mm in diameter and the individual pairs were aligned with the x, y, or z axes.

To minimize stray magnetic fields in region II (see Fig. 4, microwave spectra of the 1s45s 3S_1 → 1s45p 3P_J transitions were recorded as the currents applied to the coils were systematically varied. Two examples of such spectra are shown in Fig. 4. For the minimum field configuration (upper spectrum in Fig. 4), the 176 kHz fine-structure interval between the J = 1 and J = 2 levels of the 3P term could not be resolved. However, increasing or decreasing the currents gave rise to observable Zeeman splittings, as can be seen in the lower spectrum in Fig. 4. The vertical bars overlaid with the recorded spectra represent the results of calculations of the relative intensities of single-photon electric-dipole transitions between the 3 Zeeman-split sublevels of the 3S_1 level and the 9 Zeeman-split sublevels of the 3P term. The dashed curves represent calculated spectra for magnetic fields of 4 μT and 25 μT, as indicated, which were obtained by convolving the corresponding sets of calculated transition frequencies and intensities with Gaussian functions with full-widths-at-half-maximum (FWHM) of Δν = 350 kHz. The calculated transition frequencies have been shifted by −170 kHz to bring them into line with the measured spectra. This shift to the absolute 1s45s 3S_1 → 1s45p 3P_J transition frequencies is attributed to a combination of effects, including Stark and Doppler shifts. For reference, a stray electric field of 10 mV cm⁻¹ (see Sec. III C) is calculated to cause a shift in this transition frequency of 40 kHz.

From the coil geometry and the currents applied to generate the cancellation magnetic field the magnitude of the stray field present when the coils were switched off was inferred to be ∼ 50 μT. This is approximately equal to the magnitude of the Earth’s magnetic field in London (UK), at the time the experiments were performed,
of $|\vec{B}| \approx 4 \mu$T. From the experimental resolution and calculated Zeeman shift, we estimate that this field was successfully cancelled in the measurement region to $\lesssim 4 \mu$T (compare the experimental data and the results of the calculations in the upper panel of Fig. 5).

C. Electric field control and characterization

The two electrodes that demarcate region II in Fig. 3 were used to control the electric field in the excitation region. However, these electrodes are themselves a source of stray electric fields, which arise from adsorbates, and patch and contact potentials [14, 45]. To cancel contributions from these fields the electric potentials applied to the electrodes were adjusted to minimise the Stark shift of the two-photon $1s55s^3S_1 \rightarrow 1s56s^3S_1$ transition. The dependence of the measured transition frequencies, obtained by fitting Gaussian functions to the experimental data, on the offset potential applied to the upper electrode is displayed in Fig. 5. A quadratic function was fit to this data (continuous blue curve) to determine the optimal potential to minimize the stray electric field in the $y$ direction. In this process, the $1s55s^3S_1 \rightarrow 1s56s^3S_1$ transition frequency in the minimum achievable field was measured to be $2\nu = 39112992 \pm 2$ kHz.

The contributions to the stray electric field in the $x$ and $z$ directions cannot be cancelled with the electrode configuration employed. However, the magnitude of the residual uncancelled stray electric field could be determined from the difference between the minimum-field transition frequency and the zero-field transition frequency.

To experimentally determine the zero-field $1s55s^3S_1 \rightarrow 1s56s^3S_1$ transition frequency the technique recently reported by Lee, Nunkaw and Gallagher [16] was employed. This involved recording single-photon microwave spectra of the $1s55s^3S_1 \rightarrow 1s56s^3S_1$ interval in a range of applied electric fields. As the fields were reduced toward zero, the intensity of this electric-dipole-forbidden single-photon transition reduces, as can be seen in Fig. 6. In weak fields the spectral intensity of the transition decreases approximately linearly with the magnitude of the electric field. Therefore, the measured transition frequency can be extrapolated to zero intensity to obtain the zero-field transition frequency of $39113003 \pm 19$ kHz. The $11$ kHz difference between this and the minimum-field transition frequency (measured by two-photon spectroscopy) suggests that the magnitude of the residual electric field in the apparatus during the measurements was $9 \pm 7$ mV cm$^{-1}$. This estimate was made using the measured relative static electric dipole polarizability of the $1s55s^3S_1$ and $1s56s^3S_1$ states, as determined from the data in Fig. 5.

The $1s55s^3S_1 \rightarrow 1s56s^3S_1$ transition frequency calculated using the quantum defects in Ref. [56] is $\nu_{calc} = 39112998.17 \pm 0.04$ kHz. The $6$ kHz difference between this and the measured two-photon transition frequency implies that the residual uncancelled stray electric field was $7 \pm 1$ mV cm$^{-1}$. This estimate of the stray field obtained by comparison of the experimental data with the theoretical predictions is in agreement with that determined purely by experimental means.

IV. RESULTS

Having minimised the stray electric and magnetic fields in the experimental apparatus and determined the mag-
The calculated frequencies and relative intensities of the transitions to the $J = 0, 1,$ and 2 levels are represented by the vertical grey bars. The measured values of $\nu_{0,2}$ and $\nu_{1,2}$ were extracted by fitting the sum of three independent Gaussian functions to each spectrum. The results are presented in Table II alongside the theoretically predicted fine-structure intervals.

In general, there is a good quantitative agreement between the experimentally measured and calculated fine-structure intervals for $n = 34$ to 36. The fine-structure intervals decrease with $n^{-3}$ and it becomes more difficult to resolve $\nu_{1,2}$ for higher values of $n$. This interval was not resolved for values of $n$ between 45 and 51 (see, e.g., upper spectrum in Fig. 4). However, the $\nu_{0,1,2}$ interval between the spectral intensity weighted average position of the $J = 1$ and $J = 2$ levels, and the $J = 0$ level could be measured. These measured intervals together with the corresponding theoretical predictions are also included in Table II.

The uncertainties stated in Table II are those associated with fitting the microwave spectra and do not in-
TABLE I. Fine structure intervals between the 1snp $^3P_J$ levels in helium. The differences between the measured and calculated values, $\Delta \nu$, are given as a fraction of the combined uncertainty, $\sigma$, associated with fitting each pair of spectral features.

| $n$ | calc. (kHz) | exp. (kHz) | $\Delta \nu/\sigma$ |
|-----|-------------|-------------|---------------------|
| $\nu_{1,2}$ | 34 | 408.73 | 397 | 0.9 |
| | 35 | 374.61 | 366 | 1.1 |
| | 36 | 344.19 | 340 | 0.8 |
| $\nu_{0,2}$ | 34 | 5437.87 | 5387 | 4.2 |
| | 35 | 4984.00 | 5014 | -1.2 |
| | 36 | 4579.27 | 4643 | -2.9 |
| $\nu_{0,\overline{2}}$ | 45 | 2275.66 | 2243 | 2.1 |
| | 46 | 2130.22 | 2150 | -0.9 |
| | 47 | 1996.92 | 1971 | 0.8 |
| | 48 | 1874.51 | 1817 | 2.8 |
| | 49 | 1761.90 | 1813 | -2.9 |
| | 50 | 1658.14 | 1646 | 0.4 |
| | 51 | 1562.36 | 1476 | 1.9 |

V. CONCLUSIONS

We have performed microwave spectroscopy of the 1snp $^3P_J$ fine-structure in high Rydberg states of helium. The smaller, $\nu_{1,2}$, and larger, $\nu_{0,2}$, fine-structure intervals were both measured for $n = 34, 35, 36$. For the more highly excited states studied in the range from $n = 45$ to 51 only a doublet, corresponding to the $\nu_{0,\overline{2}}$ interval, was resolved. For all of the states studied the measured intervals agree with the theoretical predictions obtained using the quantum defects reported in Ref. [12]. The typical full-widths-at-half-maximum of the measured spectral features associated with the 1sns $^3S_1 \rightarrow 1snp$ $^3P_J$ transitions was ~350 kHz, and the uncertainty in determining the transition frequencies ranged from ±15 to ±40 kHz.

The measurement precision in the experiments was limited by the presence of stray magnetic and electric fields. The uncancelled electric field could be reduced in future measurements by careful preparation of the microwave radiation. Presently, stray magnetic fields contribute more significantly to the experimental uncertainty than stray electric fields. Calculations performed at $n = 45$ indicate that a fifty-fold reduction in the mag-

FIG. 8. (a) Measured values of the fine structure interval $\nu_{0,\overline{2}}$ for values of $n$ between 34 and 51 (points). The continuous blue curve represents the function $\nu = an^{-3}$ fit to the experimental data such that $a = 2.076(9) \times 10^5$ kHz. The dashed black curve, which is indistinguishable from the fit, indicates the calculated values for the same interval. (b) The differences, $\Delta \nu$, between the measured and calculated data in panel (a).
mitude of the stray magnetic field would be required to bring the uncontrolled Zeeman shifts down to the order of 1 kHz. This corresponds to fields of $|\vec{B}| \approx 100$ nT. A combination of magnetic shielding and active field cancellation could be used to realize magnetic fields $< 10$ nT. If this were achieved then the spectral resolution of the microwave source and the finite interaction time of the atoms with the microwave field would set the limit on the overall uncertainty. In this case, colder beams of atoms and longer microwave pulses would be required to make further improvements.

The results presented here validate the theoretical predictions of the $1s^2p^3P_J$ fine structure in Rydberg states of helium with high principal quantum numbers. Improvements in the control of stray electric and magnetic fields in the experimental apparatus are expected to reduce the systematic uncertainties in the measured fine structure intervals to the level of 1 kHz. The extreme sensitivity of high Rydberg states to stray electric and magnetic fields make it challenging to perform direct measurements that test bound state QED calculations. However, as demonstrated, this sensitivity can be exploited for electrometry and magnetometry to allow accurate characterization of the fields. The methods used to accurately characterise these fields in the work reported here could also be implemented for field characterization and cancellation in precision measurements of the $1s^2p^3P_J$ and $1s^3p^3P_J$ fine structure. Our measurements of the Rydberg fine structure are of direct importance to hybrid cavity QED experiments involving helium atoms in triplet Rydberg states. They also provide important information which is required in the refinement of experiments to study resonant energy transfer in collisions of ammonia molecules with helium atoms in triplet Rydberg states.

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