Frequency Metrology in Quantum Degenerate Helium: Direct Measurement of the \(2\ 3S_1 \rightarrow 2\ 1S_0\) Transition

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Precision spectroscopy of simple atomic systems has refined our understanding of the fundamental laws of quantum physics. In particular, helium spectroscopy has played a crucial role in describing two-electron interactions, determining the fine-structure constant and extracting the size of the helium nucleus. Here we present a measurement of the doubly forbidden 1557-nanometer transition connecting the two metastable states of helium (the lowest energy triplet state \(2\ 3S_1\) and first excited singlet state \(2\ 1S_0\), for which quantum electronic and nuclear size effects are very strong. This transition is weaker by 14 orders of magnitude than the most predominantly measured transition in helium. Ultracold, submicrokelvin, fermionic \(^3\)He and bosonic \(^4\)He atoms are used to obtain a precision of \(8 \times 10^{-12}\), providing a stringent test of two-electron quantum electrodynamic theory and of nuclear few-body theory.

The first observations of helium emission spectra at the end of the 19th century revealed two separate series of lines, associated with orthohelium and parahelium, respectively. In 1926, Heisenberg explained the distinction between these two spectra on the basis of wave mechanics, electron spin, and the Pauli exclusion principle (1). The spectrum of orthohelium arises from triplet states for which the electron spins are parallel, whereas in parahelium the electron spins are antiparallel, forming singlet states (Fig. 1). From the lowest state of orthohelium, the \(1S_0\) state (denoted \(2\ 1S_0\)), only excitations to triplet states have been observed. Orthohelium transitions from the \(2\ 3S_1\) state and associated studies of the \(n=2\) \(P_{0,1,2}\) \((n=2,3)\) fine-structure splittings (2–7) have enabled tests of quantum electrodynamics (QED) (8, 9), as well as a determination of the fine-structure constant (5, 10). In the singlet spectrum of helium (parahelium), electric-dipole transitions from the \(1\ S_0\) ground state (7) and from the metastable \(2\ 1S_0\) state (12, 13) have also provided tests of high-precision QED calculations. All these frequency metrology studies have been performed using either atomic beams or gas discharges. However, helium in the metastable \(2\ 3S_1\) state (\(\text{He}^+\), lifetime \(8 \times 10^7\) s) can be laser-cooled and trapped, which allows much longer interaction times for excitation of weak transitions. \(\text{He}^+\) atoms have been cooled to \(\mu\)-kelvin temperatures, which revealed quantum statistical effects of bunching and antibunching (14) and allowed quantum degeneracy to be achieved for both the bosonic isotope \(^4\)He (15, 16) and the fermionic isotope \(^3\)He (17).

Here we observe an orthohelium-parahelium transition, specifically, the 1557-nm transition between the metastable \(2\ 3S_1\) and \(2\ 1S_0\) states (Fig. 1), both in \(^4\)He and \(^3\)He. This transition is an excellent testing ground for fundamental theory of atomic structure. Because of a large electron density at the nucleus, the energy of \(S\) states is the most sensitive to QED and to nuclear size effects (8). For the \(2\ 3S_1\) and \(2\ 1S_0\) metastable states, QED terms contribute 4 and 3 GHz respectively, to a total binding energy of 10 GHz (8, 9). The present accuracy in the QED calculations is 2 MHz, based on an estimate of non-evaluated higher-order terms. Many of these terms are common between the isotopes. Therefore, in the calculation of the isotope shift (i.e., the difference between the transition frequencies for \(^4\)He and \(^3\)He), mass-independent terms cancel, and the uncertainty is reduced to the sub-kHz level (18). As the finite nuclear charge radius shifts the \(2\ 1S_0\) state by 2.6 MHz and the \(2\ 3S_0\) state by 2.0 MHz, an accurate isotope-shift measurement allows a sensitive determination of the difference in the mean charge radius of the \(\alpha\) particle and of the \(3\)He nucleus, which provides a stringent test of nuclear charge radius calculations and experiments (19).

**Fig. 1.** Relevant energy levels, transition wavelengths, and state lifetimes of helium. The magnetic-dipole transition connecting the \(2\ 3S_1\) state and the \(2\ 1S_0\) state has a wavelength of 1557 nm and an Einstein A coefficient of \(9.1 \times 10^{-8}\) \(s^{-1}\). A focused 1557-nm laser also constitutes a trap for ultracold atoms in the \(2\ 3S_1\) state because it is red detuned from the \(2\ 1S_0 \rightarrow 2\ 3P_1\) transitions. As the 1557-nm laser light is blue detuned from the \(2\ 1S_0 \rightarrow 2\ 3P_1\) transition, atoms in the \(2\ 1S_0\) state are antitrapped.
The natural linewidth of the $2^3S_1 \rightarrow 2^1S_0$ transition is 8 Hz, determined by the 20-ns lifetime of the $2^1S_0$ state, which relaxes via two-photon decay to the ground state (Fig. 1). This transition is 200,000 times as narrow as the natural linewidth of the $2^3P$ state, which is most prominently used for spectroscopy in helium. The Einstein A coefficient for the $2^3S_1 \rightarrow 2^3S_0$ magnetic-dipole transition is $\sim 10^{-7}$ s$^{-1}$ (20, 21), smaller by 14 orders of magnitude than for the electric-dipole transitions from $2^1S_1$ to $2^3P_{0,1,2}$ states, which indicates that excitation requires high power and/or long interaction times.

The experiment described here was performed using an apparatus designed for the production of quantum degenerate gases of helium (17, 22). Briefly, the metastable $2^3S_1$ state is populated by electron impact in an electric discharge. The atomic beam is collimated, slowed, and trapped by using standard laser cooling and trapping techniques on the $2^3S_1 \rightarrow 2^3P_2$ transition at 1083 nm. The atoms, optically pumped to $m_j = +1$, are then transferred to an Ioffe-Pritchard-type magnetic trap. $^4$He* atoms are evaporation-cooled toward Bose-Einstein condensation by stimulating radio-frequency (RF) transitions to untrapped states. For $^3$He* (in the $F = 3/2$ hyperfine state), quantum degeneracy is reached by sympathetic cooling with $^4$He*. Either one or both of the two isotopes are transferred into a crossed-beam optical dipole trap. This trap consists of two focused 1557-nm laser beams, intersecting at their foci, as shown in Fig. 2. We transfer up to $10^6$ atoms to this optical trap.

After loading the optical trap, the atoms are illuminated by a separate beam for spectroscopy, which is derived from the same laser as the optical trap beam, but is switched and frequency-shifted by a 40-MHz acousto-optic modulator. A heterodyne signal is set up between the 1557-nm laser and a mode of a femtosecond frequency-comb laser to deduce the absolute frequency of the spectroscopy laser. The frequency comb is based on a mode-locked erbium-doped fiber laser, for which both the repetition rate and the carrier-envelope offset frequency are referenced to a global positioning system-controlled Rubidium clock (23).

After a certain interaction time (typically 1 to 6 s), both the spectroscopy beam and the trap beam are switched off, which allows the atoms to fall because of gravity. The high internal energy of $^4$He* (20 eV above the $1^1S_0$ ground state) allows for efficient detection on a microchannel plate (MCP) detector (Fig. 2). The MCP signal reflects both the number of atoms and their temperature. In the case of $^4$He, the signal has a bimodal character that results from the combination of Bose-condensed atoms and thermal atoms (Fig. 3A); a fit to this signal provides the number of condensed atoms (23). Because the excited state is antitrapped, the trap is depleted when the spectroscopy beam is resonant with the atomic transition. By deducing the remaining number of $2^3S_1$ atoms for various laser frequencies, the atomic resonance frequency is determined from a Gaussian fit to the data (Fig. 3B). The observed linewidth is largely due to the 75-kHz laser linewidth.

Several systematic shifts in the transition frequency are taken into account (23). The largest shift is due to the Zeeman effect. The measured transition, $2^3S_1 (m_j = +1) \rightarrow 2^1S_0 (m_f = 0)$ for $^4$He, and $2^3S_1 (F = 3/2, m_F = +3/2) \rightarrow 2^3S_0 (F = 1/2, m_F = +1/2)$ for $^3$He, is shifted from resonance predominantly by Earth’s magnetic field. The size of the shift is deduced by measuring the resonance frequency of RF spin-flip transitions between the $2^3S_1$ magnetic substates. An additional shift is caused by the momentum transfer from a 1557-nm photon to an atom. In the case of $^4$He, the high density of the condensate could potentially cause a mean-field shift (24). However, by performing the experiment with reduced atomic density, no shift is observed.

The second-largest systematic frequency perturbation is due to the AC Stark shift associated with the intense 1557-nm light that induces the dipole trap: The specific energy state of the trapping potential for an atom determines the AC Stark shift for that atom. For $^4$He, only excitations of atoms condensed in the ground state of the dipole trap are taken into account in determining the transition frequency. As the trap depth depends linearly on laser intensity, measuring the resonance frequency for a range of applied laser powers allows an extrapolation to zero laser intensity. In contrast, $^3$He atoms, because of their fermionic nature, are distributed throughout the energy states of the dipole trap, and as a result, the resonance frequency depends little on the laser intensity.

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**Fig. 2.** Experimental setup. A small fraction of the 1557-nm laser light is split off and coupled via a fiber-optic link to be referenced to a fiber-based frequency comb. A heterodyne signal is monitored on a fast photodiode (PD) to determine the absolute frequency of the 1557-nm laser. The remaining light is divided into the trap beam and the spectroscopy beam. A crossed-beam dipole trap configuration is realized by focusing both the incident and returning trap beam (with orthogonal linear polarizations) to a waist of $\sim 85 \mu m$ at the center of the magnetic trap (represented by the green coils) under a relative angle of 19 degrees, trapping atoms at the intersection. The spectroscopy beam is frequency shifted by a 40-MHz acousto-optical modulator (AOM), overlapped with the returning trap beam and absorbed by a thermopile power meter (PM). A microchannel plate detector is positioned underneath the trap for temperature and atom number determination.

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**Fig. 3.** (A) Bimodal time-of-flight distribution observed when $^4$He* atoms are detected on the MCP detector ~186 ms after the trapping laser light is turned off. The MCP signal is fit to determine the number of Bose-condensed atoms. (B) The percentage of Bose-condensed $^4$He atoms remaining in the $2^3S_1$ state as a function of applied laser frequency (relative to the fitted center frequency $f_0$). The line is a fit of a Gaussian to the data. We measure linewidths varying from 75 to 130 kHz depending on the trap depth and on the isotope.
the measured AC Stark shift does not equal the trap depth (as is the case with \(^{3}\)He) but is reduced due to the density of states within the dipole trap. A nonlinear shift can then potentially arise at high laser intensities, where the larger trap depths allow for higher temperatures. To minimize this effect, \(^{3}\)He atoms are sympathetically cooled to the quantum degenerate regime to populate predominantly the lowest energy states of the trapping potential. Over the course of several months, 20 independent extrapolations were obtained (as shown in Fig. 4) to deduce an absolute frequency of the 2 \(^{3}\)S\(_1\) \(\rightarrow\) 2 \(^{3}\)S\(_0\) transition for \(^{4}\)He of \(f_{\Delta} = 192,510,702,145.6(1.8) \text{kHz}\) and for \(^{3}\)He \((F = 3/2 \rightarrow F = 1/2)\) of \(f_{\Delta} = 192,504,914,426.4(1.5) \text{kHz}\), where the one-standard deviation error in parentheses includes all statistical and systematic uncertainties.

For both isotopes, our result agrees with QED calculations of the ionization energies of the two metastable states (9, 23). The present experimental error in the transition frequency is smaller by three orders of magnitude than estimates of non-evaluated higher-order terms in state-of-the-art QED calculations and presents a significant challenge for groups involved in atomic structure theory.

An indirect value of the energy difference between the 2 \(^{3}\)S\(_1\) and the 2 \(^{3}\)S\(_0\) states can be obtained from the literature (only for \(^{4}\)He) by combining experimental transition frequencies from various systematic sources, e.g., AC Stark shift and Zeeman shift (23). The frequencies quoted on the \(y\) axes are the weighted averages of the data points. Their associated uncertainties (with the uncertainties due to the frequency comb calibration and the mean-field shift added in quadrature) are \(\pm 1.8 \text{kHz}\) for \(^{4}\)He and \(\pm 1.5 \text{kHz}\) for \(^{3}\)He, represented by the green bands.

**Fig. 4.** Measured transition frequencies for \(^{4}\)He (A) and for \(^{3}\)He (B). The error bar on each data point includes contributions from various systematic sources, e.g., AC Stark shift and Zeeman shift (23). The frequencies quoted on the \(y\) axes are the weighted averages of the data points. Their associated uncertainties (with the uncertainties due to the frequency comb calibration and the mean-field shift added in quadrature) are \(\pm 1.8 \text{kHz}\) for \(^{4}\)He and \(\pm 1.5 \text{kHz}\) for \(^{3}\)He, represented by the green bands.

after two-photon decay) opens up the possibility of performing two-photon spectroscopy on the 2 \(^{3}\)S\(_0\) \(\leftrightarrow\) 1 \(^{1}\)S\(_0\) transition (17, 31), where QED and nuclear size effects are strongest.

**References and Notes**

23. Methods and calculations are further detailed in supporting material at Science Online.