Measurement of the He $1s2s\ ^1S_0$ isotopic shift using a tunable VUV anti-Stokes light source

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We describe a high-resolution, vacuum-ultraviolet spectroscopic technique based on a tunable, narrow-band, VUV, spontaneous anti-Stokes light source. The technique was used to measure the absolute energies of the $1s2s\ ^1S_0$ states of ^{3}He and ^{4}He; the $1s2s\ ^1S_0$ level of ^{3}He is 7.8 ± 0.5 cm^{-1} below that of ^{4}He.

High-resolution, vacuum-ultraviolet (VUV) spectroscopy has traditionally been limited by the lack of bright sources and by the low efficiency and resolution of spectrometers. Many of these difficulties can be reduced by making use of the high brightness, tunability, and narrow linewidth of the recently demonstrated spontaneous anti-Stokes VUV source.[1,2] In this Letter we illustrate such an application by directly measuring the isotopic shift of the He $1s^2\ ^1S_0$–$1s2s\ ^1S_0$ transition at 166277 cm^{-1} (20.6 eV) to a resolution of ± 0.5 cm^{-1} (60 μeV). The technique does not require any VUV optics or detectors.

Figure 1 shows the energy levels pertinent to this experiment. A cw glow discharge in a mixture of He and Ne creates population in the He $2s\ ^1S$ metastable level. A tunable laser pump field of frequency ω_p interacts with this population to produce tunable VUV spontaneous anti-Stokes radiation having a photon energy equal to $\hbar\omega_p$ plus the energy of the metastable level. The pump frequency is tuned until the frequency of the generated anti-Stokes radiation corresponds to the Ne $2p^6\ ^1S_0$–$2p^5\ 7s'\ (1/2)_1{}^0$ transition at 583.7 Å, and the Ne, acting as a narrow-band detector, absorbs the radiation and fluoresces on the $2p^5\ 7s'(1/2)_1{}^0$–$2p^5\ 3p'$-$(3/2)_2$ transition at 4886 Å. The energy of the He $2s\ ^1S$ level is therefore determined by the known energy of the Ne $2p^5\ 7s'$ state and the value $\hbar\omega_p$ that corresponds to its excitation. Separate experiments using ^{3}He and ^{4}He were performed to determine the isotopic shift.

The ~2-μm tunable pump for the anti-Stokes VUV source was obtained from a $LiNbO_3$ temperature-tuned optical parametric oscillator (OPO) pumped by the doubled output of a Q-switched Nd:YAG oscillator–amplifier system. Long-term amplitude stability was achieved by using a feedback signal from a photodiode monitoring the OPO output to adjust the Nd:YAG oscillator Q-switch timing. Typically, the OPO output was 100 μJ in a 10-nsec-long pulse at 29 pulses per second; the average power was stable to better than ±2% for up to 8 h of continuous operation; the measured linewidth was 1.9 cm^{-1}.

The OPO output was double passed through the bore of the glow discharge tube. The focal spot had an area of about 10^{-2} cm^2, yielding a typical pump-power density of 2×10^6 W/cm^2. Two physically identical discharge tubes were used; both were commercial (Spectra-Physics Model C-130) 25-cm-long He–Ne laser plasma tubes with a 1.5-mm bore diameter. A highly regulated 10-mA current source was used to maintain the discharge. The first tube was sealed and contained, according to the manufacturer, a 9:1 mixture of ^{3}He:Ne at a total pressure of 3.4 Torr. The second tube was cut open, and a similar mixture of ^{4}He:Ne was flowed slowly through it. The two tubes could be interchanged quickly and comparison scans made under identical system conditions.

Light from the discharge was filtered and focused into a 1-m spectrometer tuned to the 4886-Å Ne $7s'$–$3p'$ fluorescence line. The output of an RCA 31034 photomultiplier was processed with photon-counting equipment consisting of fast preamplifiers, a pulse-height discriminator, and two counting channels (signal and background), each with a gate generator, a coincidence unit, and a counter. A third counter driven by the laser light pulse was used to determine the counting period, typically 3000 pulses. Following each counting period a minicomputer recorded the data from both channels, reset the counters, increased the OPO crystal temperature by 0.05 K (decreasing photon energy by 0.5 cm^{-1}), and initiated a new counting cycle. Typical

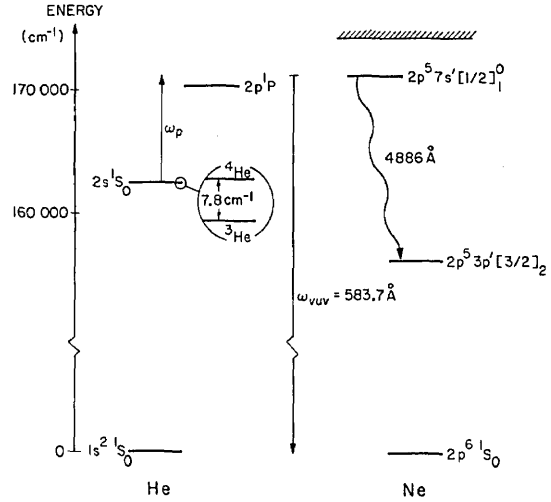

Fig. 1. Partial energy level diagram of He and Ne.

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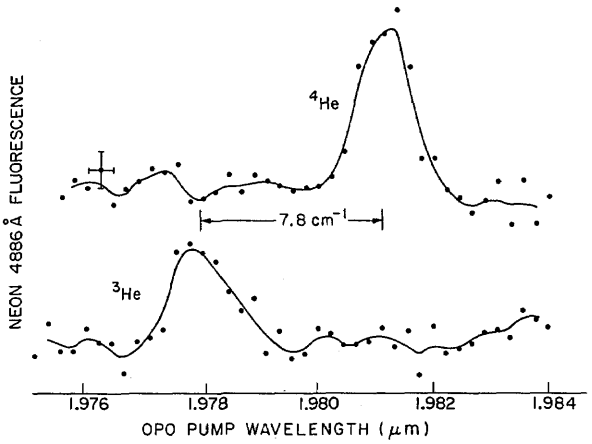

Fig. 2. Observed Ne $2p^5\ 7s'(1/2)_1{}^0$–$2p^5\ 3p'(3/2)_2$ fluorescence at 4886 Å as a function of OPO wavelength for ^{4}He and ^{3}He. In each case the peak corresponds to the emission by He and the absorption by Ne of 583.69 Å anti-Stokes radiation.

scans required 70 min to collect 40 data points covering 20 cm^{-1}.

In the absence of VUV radiation, background fluoresence at 4886 Å is produced primarily by inelastic collisional excitation of the Ne $7s'$ level by the He $2p\ ^1P$ population. This background was measured by gating on the background counting channel 5 μsec before the OPO pulse for an interval of 1.4 μsec, while the signal channel was gated on 20 nsec after the OPO pulse for an interval of 126 nsec. For a counting period of 3000 laser pulses there were typically ~10,000 counts in the signal channel and ~120,000 counts in the background channel. The background channel, however, slightly overestimates the background rate actually present during the signal collection period because the OPO pulse itself photoionizes a portion of the Ne $7s'$ population, temporarily depressing the background fluorescence during the signal collection interval. This reduction was measured to be ~16%. Thus each data point was obtained by multiplying the background channel count by a factor accounting for the ratio of the gate widths and the measured ionization and subtracting the result from the signal channel count.

Figure 2 shows the results from four scans; two scans for each He isotope have been added together and a smoothed curve handdrawn. The uncertainty of the OPO photon energy for each point is less than ± 0.5 cm^{-1}; the vertical error bar represents the uncertainty due to counting statistics. Using the published[3] energy for the Ne $7s'$ state of 171324.0 cm^{-1} and our own calibration of ω_p, we calculate the ^{4}He $2s\ ^1S$ energy to be 166277.3 ± 0.5 cm^{-1}, in agreement with its published value.[4] We find the energy of the ^{3}He $2s\ ^1S$ state to be 7.8 ± 0.5 cm^{-1} lower, consistent with an experimentally inferred value of 8.1 cm^{-1}.[5] The 2.6-cm^{-1} full-width at half-maximum lineshape is consistent with the value estimated by convolving the OPO, the Doppler-broadened He $2s\ ^1S$, and the natural Ne $7s'$ linewidths.

For our experimental conditions we estimate a He $2s\ ^1$S density of about 2×10^{11} cm^{-3} and an induced anti-Stokes A coefficient[1] of 3.8×10^5 sec^{-1}, yielding a production of 1.5×10^8 VUV photons per pulse in the 0.2-cm^3 cell volume. About 20% of the produced VUV light is within the Ne $7s'$ absorption linewidth, and only 80% of that will be absorbed in the ~1.5-mm path length. Hence we estimate a production of 2.4×10^7 Ne $7s'$ atoms per pulse at the peak of the effect. Although the measured signal integrated over the gate width is only about 8% above the background, this corresponds to a laser-induced excitation rate approximately twice that of the He $2p\ ^1P$–Ne $7s'$ collisional rate for the time that the laser is on.

This work represents the first reported direct measurement of the He $2s\ ^1S$ isotopic shift and illustrates the potential of this technique for performing high-resolution VUV spectroscopy without the limitations of traditional VUV apparatus. In this experiment the known Ne $7s'$ target state was used to characterize the He storage states, but in other applications known source properties could be used to study VUV absorber states, such as the inner shell transitions of K or Rb. Extensions to shorter wavelengths using an ionic storage state, such as Li II $1s2s\ ^1S_0$ at 199 Å, should be possible. Finally, we note that the use of a mode-locked pump laser would facilitate time-resolved studies of VUV fluorescence and autoionization.

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