Light Polarization And Quantum Interference Effects In Unresolvable Atomic Lines Applied To A Precise Measurement Of The $^{6,7}\text{Li} D_2$ Lines

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Abstract. Precise measurement of the isotope shift of the lithium $2s\rightarrow3s$ and $2s\rightarrow2p$ (D) lines has been proposed [1] and subsequently used [2] as a sensitive probe of nuclear charge radii, including halo nuclei in unstable isotopes. Despite this success, large discrepancies have remained among observations of D lines of the stable isotopes. We find that when the excited state splittings are of order the excited state natural line widths, as is the case for the $D_2$ line of lithium, light-polarization-dependent quantum interference modifies the lineshape and represents an overlooked but significant systematic effect. We present expressions for the corrected lineshapes and demonstrate that they yield consistent line centers for arbitrary polarization of the excitation laser.

In our experiment Doppler free spectra were collected using a tunable single-frequency diode laser referenced to a frequency comb [3]. We analyze spectra of the $^{6,7}\text{Li} D_2$ lines taken at various excitation laser polarizations and show that failure to account for the quantum interference changes the inferred line strengths and shifts the fitted line centers by as much as 1 MHz, a systematic effect large enough to account for discrepancies between previous measurements. We obtain revised values for the absolute transition frequencies of the unresolvable $^{6,7}\text{Li} D_2$ features. Combining these with our previous results for the $D_1$ lines [3], we derive the $^{6,7}\text{Li}$ excited state fine structure intervals, the $2s\rightarrow2p$ isotope shift, the splitting isotope shift and the relative nuclear charge radii [4]. This analysis should also be important for precise spectral measurements in a number of other species including partially resolved $D_2$ lines in hydrogen, lithium, potassium, francium, and singly-ionized beryllium and magnesium.

PACS: 32.10.Fn, 32.30.Jc, 32.70.Jz, 42.50.Gy, 42.62.Ehs, 42.62.Fi, 21.10.Ft

4. K. Pachucki (private communication).