Measurement of the lifetime of the $1s2p\ ^3P^o$ state in Be III

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Using the collinear beam-laser method, we have measured the spontaneous emission lifetime of the $1s2p\ ^3P_2^o$ level in Be III to be 29.1 ± 0.2 ns, in good agreement with many mutually consistent theoretical values. This resolves the discrepancy between theory and the previous experimental value. We also obtain a value of 0.214 ± 0.002 for the multiplet oscillator strength of the $1s2s\ ^3S \rightarrow 1s2p\ ^3P^o$ transition.

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I. INTRODUCTION

Recent studies of beryllium as a surface material in plasma confinement devices [1,2], including a major experiment at the Joint European Torus (JET), have led to the development of plasma diagnostics employing emission spectroscopy of the beryllium atom in all its ionization states [3]. Be I and Be II provide information about the peripheral regions of the plasma, near the diverters, limiters, and other contact surfaces. Spectral lines in the visible and near uv are most often of interest; unfortunately, however, many prominent lines are unusable because of attenuation in the long quartz fiber coupling lines. These ions are also astrophysically important, playing a role in the determination of chemical abundances in stellar atmospheres [4]. Be III and Be IV yield data from nearly all regions of a confined plasma; they are mostly observed in the extreme uv (xuv). The near uv $1s2p\ ^3P^o \rightarrow 1s2s\ ^3S$ transition is used together with the xuv intercombination line $1s2p\ ^3P^o \rightarrow 1s^2\ ^1S$ to determine the ratio of metastable to ground-state populations in the modeling, to study diffusive imbalance, and as a means of putting the xuv and uv detector calibrations on a common scale. The latter is an important application because of troubling drifts in xuv detector sensitivity.

Vital to all of these plasma diagnostics and astrophysical measurements is a set of precise atomic oscillator strengths for the transitions of interest. Martinson *et al.* [5] recently assembled a comprehensive collection of measured and theoretical lifetimes and oscillator strengths. They pointed out a serious discrepancy, amounting to 7 standard deviations, between the sole experimental value [6] for the lifetime of the $1s2p\ ^3P^o$ level of Be III and several theoretical values that are in good mutual agreement. In the present experiment we have made a measurement of the lifetime of the $1s2p\ ^3P_2^o$ level to an accuracy of 0.7% using the collinear beam-laser method. Because the allowed $E1$ transition to the $1s2s\ ^3S_1$ level is the only one to make a significant contribution to the lifetime, we also obtain an accurate value for the oscillator strength of this transition. Our value is in excellent agreement with all theoretical predictions.

II. EXPERIMENT

In the present work we have used a refined version of the lifetime measurement technique of Ceyzeriat *et al.* [7]; this has been previously described in detail in Ref. [8]. The ions were produced in a modified Danfysik 911A ion source using Be metal as the source feed. A discharge current of ~ 60 mA could be maintained with a potential difference of 400 V. These conditions favored the production of metastable Be^{2+}. The ions were extracted by a 7200-V potential, electrostatically focused and then mass selected by a Wien filter. After being focused by a second einzel lens, the $^9Be^{2+}$ ions were electrostatically deflected through a 5° angle to be collinear and overlapped with the laser beam. A pair of 2-mm-diam apertures, internal to the vacuum and separated by 0.85 m, were used to align the axes of the laser and ion beams. These apertures were rotated out of the beam path during data acquisition to reduce scattered light in the detectors. Typical ion currents recorded through the apparatus were 10 nA.

The laser light at 372 nm was generated by intracavity frequency doubling of a single frequency cw Ti-sapphire ring laser. Typical input power to the apparatus was 20 mW. The stabilized ring laser has a ~ 1-MHz linewidth and its frequency drifts less than 20 MHz per hour. To maintain uniform excitation of the ion beam over the entire range of travel, the laser beam was loosely collimated by a 1.5-m focal length lens before it entered the vacuum. Depending upon source conditions, the observed linewidths were measured to have a kinematically compressed full width at half maximum of between 500 and 800 MHz, corresponding to an energy spread of 5–8 eV per charge. The use of well-regulated power supplies for the anode and acceleration voltages maintained exact resonance of the laser and ion beams for long periods of signal integration.

A significant advantage of the collinear laser and ion beam geometry is the calibration of the ion velocity. The wave numbers of the $1s2s\ ^3S_1 \rightarrow 1s2p\ ^3P_2^o$ hyperfine components are known from previous work to 1 part in 10^8 [9]. By measuring the Doppler-shifted wave numbers of these hyperfine components, the ion velocity can be calibrated to approximately 1 part in 10^4, yielding a completely negligible contribution to the experimental uncertainty of the lifetime measurement. A Doppler tuning region, comprised of a series of three post-acceleration electrodes (described in detail in Ref. [8]) surrounding the overlapped laser beam and ion beam, was translated on a precision ball slide. A negative bias on these electrodes Doppler shifted the laser frequency into resonance with a single hyperfine component of the $1s2s\ ^3S_1 \rightarrow 1s2p\ ^3P_2^o$ transition. The relative potentials on

the three electrodes were chosen to provide a steep fall of the potential seen by the ions as they left the excitation region.

The decaying ions subsequently passed through a grounded cylinder, which was slotted to permit the observation of spontaneous emission. Decay curves were acquired by translating the Doppler tuning region while an *f*/0.7 lens system focused the spontaneous emission from a fixed location through a field stop and onto a photomultiplier. A spherical mirror was used to double the light collection efficiency of the lens system. Since the $1s2p\ ^3P_2^o$ level decays almost entirely to the metastable $1s2s\ ^3S_1$ level, the wavelength of the laser-induced fluorescence viewed at right angles to the beam differed from that of the laser by only ~ 7.0 Å due to the Doppler shift. Rejection of scattered laser light with a monochromator was not compatible with reasonable collection efficiency. Instead, a series of carefully positioned light baffles and apertures reduced the detected scattered laser light to less than 1000 counts/s and a colored glass filter blocked any observed light from collisions between ions and residual gas.

To subtract the remaining background signal due to scattered laser light and photomultiplier dark current, the negative potentials on the Doppler-tuning region were modulated at a frequency of 1 kHz between ground and their chosen values. Two counting channels, each gated to one phase of the modulation voltage, were used to monitor the fluorescence count rates from the fixed photomultiplier. The decay curves were acquired as the difference of the two count rates when the ions were resonant and nonresonant with the laser frequency.

Most of the data were acquired with the laser resonant with the $1s2s\ ^3S_1,\ F=5/2 \rightarrow 1s2p\ ^3P_2^o,\ F=7/2$ hyperfine component for maximum signal. This component is 8.3 GHz higher than the lowest component of the $J=2$ hyperfine structure [9]. Excitation of ions in the region viewed by the photomultiplier can cause systematic errors in the lifetime measurement. To avoid such errors, the amplitude and the sign of the modulation were chosen so that the laser frequency in the ion rest frame was at least 3.3 GHz *below* resonance with any of the remaining hyperfine components of that fine-structure component during the ground phase of the modulation. The sign is determined by the fact that the velocity distribution has a long tail on the low-energy side but a sharp cutoff on the high-energy side.

To monitor the constancy of the excitation, the modulated fluorescence in the Doppler-tuning region was detected by a small photomultiplier, which translated in vacuum with the postacceleration electrodes. The photocurrent was phase detected by a lock-in amplifier, providing a signal proportional to the rate of ion excitation. This signal was subsequently used to normalize the fluorescence decay curve for small variations in excitation efficiency due to drifts in ion energy or current, laser wavelength or power, and varying overlap of the two beams.

For a typical decay curve, the Doppler-tuning region was translated in steps of 1.27 mm, dwelling for 3 s at each position. Simultaneously with the recording of the two count rates, the normalization signal from the internal photomultiplier, the laser power, and the ion beam current were also recorded. At the beginning of the decay curve, the signal count rates were ~ 4000 counts/s. A total of 36 decay curves were accumulated over several days, each spanning 4.6 lifetimes and beginning approximately 1.5 lifetimes after excitation. In addition to these data, daily measurements of the laboratory frequency of the $2\ ^3S_1 - 2\ ^3P_2^o$ hyperfine components were acquired for calibration of the ion velocity.

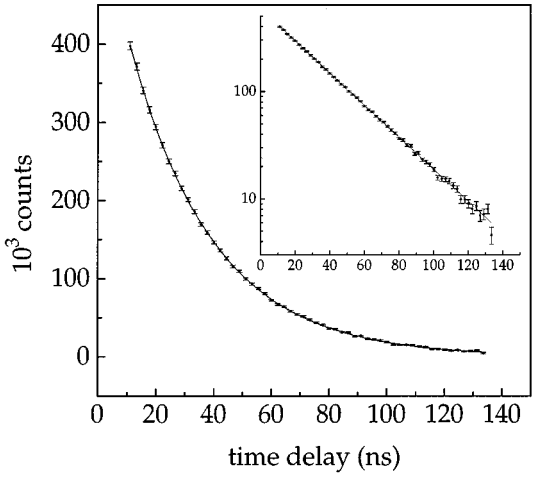

FIG. 1. Least-squares fit to summed and normalized fluorescence decay curves for the $1s2p\ ^3P_2^o$ state of Be III. The abscissa is measured relative to the first channel of data. The lifetime was determined to be 29.1(2) ns and the background was -253 ± 335 counts. The χ^2 per degree of freedom for the fit was 0.73. The inset is a semilogarithmic plot of the same data with the fitted background subtracted.

III. RESULTS

The 36 individual measurements were then summed and analyzed using a weighted nonlinear least-squares fitting routine to determine the lifetime, an amplitude, and any residual background not eliminated by the digital gating. The weights were determined from uncertainty estimates from counting statistics and the effect of the normalization procedure. Since the decay curves are the difference of two count rates (one the sum of signal and background, the other only background) the uncertainty for each data point was estimated as the square root of the sum of total counts in both counting channels. The relative uncertainty introduced by normalization was estimated as the ratio of the standard deviation of the first ten channels to the mean of all channels of the normalization signal and was added in quadrature to the relative statistical uncertainty for each channel. The aggregate decay curve and exponential fit are shown in Fig. 1. The background for the aggregate data was determined to be -252 ± 335 counts, consistent with zero. The fit to the summed decay curves produced a lifetime of 29.1(2) ns and a χ^2 per degree of freedom, χ^2_{reduced}, of 0.73, which suggests that the experimental errors have been conservatively estimated.

As an internal check of data consistency, the normalized decay curves were analyzed individually, producing a weighted average lifetime of 29.1(1) ns , which is in excellent agreement with the aggregate figure. A histogram of the distribution of these individual measurements is shown in Fig. 2. The aggregate data were also refit without any nor-

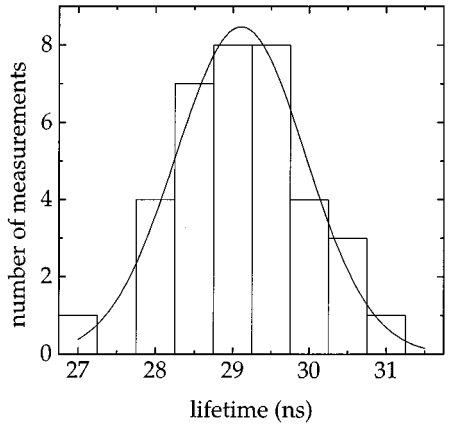

FIG. 2. Histogram of individual measurements of the lifetime of the $1s2p\ ^3P_2^o$ state of Be III. Each of the normalized decay curves was individually analyzed to determine a decay constant. The weighted average of all 36 measurements was 29.1(1) ns, in excellent agreement with the lifetime from the fit to the aggregate decay curve.

malization, producing a 5% increase in the lifetime compared to that with normalization, but also yielding a nonzero background and a large χ^2_{reduced} value of 5.5. Normalizing the decay curve only to fluctuations in the recorded laser power and ion current signals increased the lifetime by 1.9% and yielded a χ^2_{reduced} of 0.83. The actual normalization signal should in principle yield a better estimate of systematic variations during the data acquisition since records of laser power and ion current fluctuations cannot reveal subtle changes in excitation probability during scans of the Doppler-tuning region due to varying overlap of the two beams. This is evident in the better statistical fit to the normalized aggregate decay curve using the actual normalization signal. The decay curve was also systematically analyzed by "tail fitting" only data beyond a given channel. Figure 3 shows the variation of the lifetime as the initial

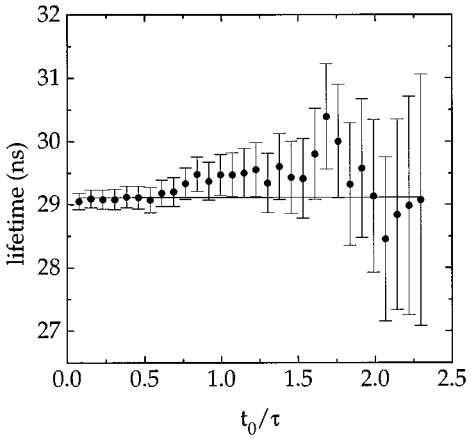

FIG. 3. Tail fitting analysis of the lifetime τ of the $1s2p\ ^3P_2^o$ state of Be III. The quantity t_0/τ is the number of lifetimes of data at the beginning of the decay curve that were not included in the tail fitting analysis. The fitting procedure included parameters for the background, amplitude, and decay constant. The error bars at each point represent the statistical uncertainty in the fit to the lifetime of the remaining exponential tail.

channel of the tail fitting procedure is increased. No significant systematic trends in the lifetime were observed as the first 2.5 lifetimes of data were ignored.

Several other random and systematic errors may affect the experiment. As was previously mentioned, the uncertainty in conversion from distance to time is negligible due to the collinear geometry. Collisions with background gas can reduce the observed lifetime. The cross section for collisional quenching of the excited state of Be^{2+} with the residual gas in the vacuum is not known; however, using 1×10^{-14} cm^2 as a very conservative upper limit on this cross section, the mean free path at our background pressure of 4×10^{-7} Torr is 76 m. For an ion velocity of 5.71×10^5 m/s, the collisional quenching rate is 7.5×10^3 s^{-1}, which represents a negligible correction of $+0.006$ ns to the observed lifetime. Lengthening of the observed lifetime due to radiation trapping is negligible at an ion density of $\sim5.2\times10^4$ cm^{-3}. Other systematic errors due to quantum beats or the Stark effect are also completely negligible in the field-free region maintained by the grounded flight tube and the external Helmholtz coils.

Our lifetime value of 29.1 ± 0.2 ns is in strong disagreement with the only previous measurement, in which Andersen, Jessen, and Sørensen [6] obtained a lifetime of 22.3 ± 1.0 ns using the beam-foil method. Although a cascade analysis was carried out in that work for some transitions, none was performed for that particular lifetime. Martinson *et al.* [5] have speculated that the observed value might have been shortened by beam divergence due to Rutherford scattering in the foil.

We can directly obtain the oscillator strength of the $1s2s\ ^3S_1\rightarrow1s2p\ ^3P_2^o$ transition observed because the other decay branches from $1s2p\ ^3P_2^o$ are completely insignificant. The next in importance to this allowed $E1$ branch is an $M2$ decay to the $1s^2\ ^1S_0$ ground state, with a transition probability of 6.33×10^2 s^{-1} [16]. There has been a long history of theoretical work on oscillator strengths in Be III. The earliest precision calculation was Weiss' [10] 1967 variational calculation with 50-term Hylleraas-type functions, which explicitly depend on the interelectronic coordinate, r_{12}, and thus include correlation. Dalgarno [11] also carried out a variational calculation. In 1971, Schiff, Pekeris, and Accad [12] used 364-term Hylleraas expansions to achieve an accuracy of "one, or occasionally two, in the last digit quoted," amounting to better than 1 part in 10^4 in the case of the $1s2s\ ^3S\rightarrow1s2p\ ^3P^o$ transition. The rather conservative criteria for choosing the number of digits were that (1) results using the length and velocity formulations of the $E1$ matrix element had to agree to one or two in the last place retained, and (2) the results of the largest expansion had to agree similarly with the extrapolated value. This calculation is generally regarded as the "benchmark" against which others are measured.

The aims of the other theoretical work in this area were largely to apply simpler methods that could produce extensive data over the whole isoelectronic sequence and especially for states of high principal quantum number n. The $n=2$ levels (usually in helium) were mainly used to check the accuracy of a new calculation with reference to the full variational method. Sanders and Scherr [13] in fact achieved accuracy similar to that of Schiff, Pekeris, and Accad with

TABLE I. Summary of calculated and measured multiplet absorption oscillator strengths f of the $1s2s\ ^3S \rightarrow 1s2p\ ^3P^o$ transition in Be III. The methods are explained in the text. The length form is listed for the theoretical values, as it is generally the most accurate. Values with an asterisk have been calculated by us from lifetimes published in the corresponding reference.

Author(s)	Reference	Method	f
		Theory	
Weiss	[10]	Variational	0.2131
Cohen and Kelly	[14]	Frozen core	0.222
Sanders and Scherr	[13]	Variation-perturbation	0.213 137
Schiff *et al.*	[12]	Variational	0.213 14
Dalgarno	[11]	Variational	0.213
Cohen and McEachran	[15]	Frozen core	0.221
Lin *et al.*	[16]	RRPA	0.223
Laughlin	[17]	Model potential	0.2137
Fernley *et al.*	[18]	Close coupling	0.2136
Theodosiou	[19]	Coulomb	0.213*
Johnson *et al.*	[20]	RMBPT	0.2134
		Experiment	
Andersen *et al.*	[6]	Beam-foil	0.28(1)*
This work		Beam-laser	0.214(2)

the variation-perturbation method of Hylleraas, in which the $(Zr_{12})^{-1}$ term in the Hamiltonian is handled by a perturbation expansion in powers of Z^{-1}, with the equation for each order being solved variationally. Cohen and co-workers [14,15] applied analytical orbital wave functions in the frozen-core approximation, achieving only $\sim 5\%$ accuracy. Lin, Johnson, and Dalgarno [16] used the relativistic random-phase approximation (RRPA), with the main interest focused on $E1$-forbidden decays at high Z. It is interesting to note that their inaccurate Be III oscillator strength was the one chosen by Summers *et al.* [3] to model JET results.

Laughlin [17] employed a model potential approach to approximate the effect of the $1s$ electron on the active electron, adjusting the parameters of the potential to fit the experimental energy levels. Fernley, Taylor, and Seaton [18], in a long series of papers to provide extensive theoretical data for astrophysical opacity calculations, used the close-coupling method with dipole and quadrupole polarization of the $1s$ ''target'' functions allowed. Theodosiou [19] used a numerical Coulomb approximation with a Hartree-Slater potential, adjusted to fit experimental energy levels. Very recently, Johnson, Plante, and Sapirstein [20] have applied relativistic many-body perturbation theory (RMBPT) to the helium isoelectronic sequence.

We have summarized the totality of theoretical and experimental results in Table I. To present a uniform set of quantities for comparison, we have consistently listed the *multiplet* absorption oscillator strength used by most authors (even when giving the f value for a single fine-structure component, viz. Ref. [16]). We can calculate the multiplet f value to sufficient accuracy simply by using the 9/3 degeneracy ratio appropriate to the entire multiplet (using the mean wavelength, 3722.65 Å, calculated with the weighted average of the energy levels) [21].

IV. CONCLUSIONS

It is clear that our new result resolves the discrepancy between the existing experimental lifetime and the many mutually consistent theoretical values. The accuracy of 0.7% we have achieved is typical of modern beam-laser measurements. With considerable effort, this could be improved to perhaps 0.2%, but it would be extremely difficult to approach the accuracy of the best variational calculations.

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