Radiative lifetimes of some group II ions by the Hanle effect in a fast-flowing helium afterglow*

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Radiative lifetimes of the first $^2P_{3/2}$ state of Mg II, Ca II, Zn II, Sr II, Cd II, and Ba II are reported as measured by the Hanle effect in a fast-flowing helium afterglow. They are, respectively, 3.65(0.12), 6.61(0.30), 2.4(0.3), 6.64(0.10), 2.86(0.25), 6.78(0.40) in units of 10^{-9} sec. The ions in the afterglow are created by Penning ionization of the neutral metal atoms, thus providing a steady-state, field-free region for observation. Comparisons are made with measurements by other methods, and discrepancies are discussed.

INTRODUCTION

Radiative lifetimes and depolarization cross sections are important parameters of excited states of ions. They serve not only as a test of existing theories but are also useful for astrophysical calculations and for understanding relaxation mechanisms in discharges and afterglows. We have measured radiative lifetimes of the first $^2P_{3/2}$ level of six group II ions by the Hanle[1] effect in a fast-flowing helium afterglow. These are the first measurements on group II ions reported in a flowing afterglow.

Until the Hanle measurements of Smith and Gallagher it was common to have uncertainties on the order of $\pm 30\%$ in the measurements of group II excited-ion lifetimes.[2,3] Some of this earlier work utilized the Hook method[4] and the arc method.[5] In the case of the Hook method, the large uncertainty was mostly due to the difficulty in measuring the ion density absolutely. The accuracy of the Hanle technique, on the other hand, depends only on knowing the relative densities, or working in the limit of very low densities at which collisional and radiation trapping effects vanish.

Gallagher and Smith reduced these large uncertainties to $\pm 5\%$ or less by the Hanle effect in a pulsed argon afterglow for several group II ions. A recent measurement of Kelly *et al.*[6] by the Hanle effect on the Sr II $^2P_{3/2}$ lifetime departed seriously from the value reported by Gallagher and Smith. Kelly reported measuring a very large depolarization cross section with the Sr neutral, and also obtained a lifetime some 15% shorter than Gallagher's value. Kelly's measurements were made by the Hanle effect in a cw discharge of Sr without a noble-gas buffer. This large difference in the Sr II lifetime, together with the lack of high-precision data on most group II ions (other than Gallagher's) led us to remeasure the radiative lifetimes of several group II ions in the $^2P_{3/2}$ state.

APPARATUS

The flowing afterglow is especially well suited for measuring lifetimes and alignment depolarization cross sections by the Hanle effect. It allows for rapid change and easy monitoring of system parameters, such as relative ion density and buffer-gas pressure. Thus, collisional depolarization and coherence narrowing can easily be controlled and their effects observed. Also the material under study can be quickly and easily changed so that lifetimes of many different species can be studied in the same apparatus over a short period of time. Another advantage is that, unlike a beam apparatus or a sealed cell, high-vacuum techniques are not necessary since flow rates are many orders of magnitude greater than outgassing rates.

Since the techniques and apparatus utilized here were reported in detail in a previous paper on Yb,[7] only a brief discussion of a few pertinent features appears here. The flow tube is evacuated by a 540 $ft^3\,min^{-1}$ mechanical pump and a Roots blower. Flow velocities are on the order of 10^4 cm sec^{-1} with background helium pressures in the range of 0.1–1 Torr. Of particular interest is the method of production of the ion ground-state density. A microwave cavity at the entrance to the flow tube is used to produce a discharge in helium. By the time the flow reaches the interaction region, the major constituents are helium metastables and helium neutrals. With a judicious setting of the discharge intensity, a helium metastable density on the order of 10^{11} cm^{-3} can be obtained in the interaction region with a background helium neutral density on the order of 10^{16} cm^{-3} and a helium ion density of about 10^{10} cm^{-3}. The reactant neutral is titrated into the interaction region by a furnace wound with coaxial heater wire. Penning ionization by the helium metastables (He^m) produces copious quantities of the reactant ions in steady state in a field-free region. Typical cross sections for this familiar reaction, $He^m + X \rightarrow He$

$+X^+ + e$, are greater than 10^{-15} cm^2.[8] Here X is the reactant atom. Ion densities as high as 10^{10} cm^{-3} are estimated by optical absorption. This technique for producing ions releases us from concerns over perturbations due to an ionizing electric field, and decreases necessary signal accumulation times over pulsed techniques. Another advantage of this configuration is the *in situ* magnetic field calibration it allows. The He^m atoms are optically pumped, and magnetic resonance is performed with the 10 830 Å light monitored in transmission, thus creating a helium magnetometer.

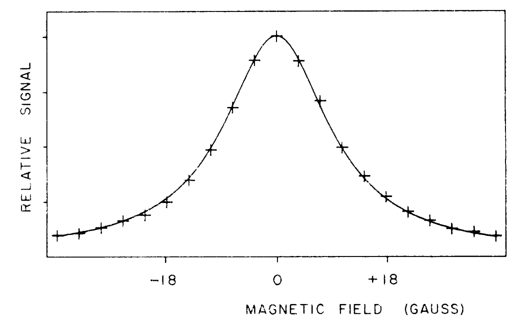

FIG. 1. Computer fit to Lorentzian of the $^2P_{3/2}$ Sr II Hanle signal.

EXPERIMENTAL TECHNIQUE

Briefly, the Hanle effect is a well used zero-field level crossing technique, in which optical resonance radiation is used to coherently excite atoms in the presence of a magnetic field. The resulting fluorescence is plotted as a function of the magnetic field, and from this plot the lifetime can be obtained as a function of the g factor. Measurement methods were described in detail in Ref. 7. They will be only briefly reiterated here. The geometry and polarization used is such that the Hanle signal reduces to a Lorentzian, $(1+X^2)^{-1}$ where $x = 2g\mu H\tau/h$. Here g is the Lande g factor, μ the Bohr magneton, H the magnetic field, τ the lifetime, and h Plank's constant reduced.

A unique complication arises with the ion Hanle signal. Since the ions interact with the magnetic field, the ion density is a slowly varying smooth function of the magnetic field. Also, the Penning reaction heavily populates the $^2P_{3/2}$ ion level. The intensity from the resulting transition, $^2P_{3/2}-^2S_{1/2}$, obscures the direct observation of the Hanle signal on an oscilloscope, and shows the same ion-density field dependence. Field-dependent intensity variations of the ion emission in the absence of the optical resonance excitation were subtracted from the Hanle signals on alternate sweeps of the magnetic field. From this information, the ion field dependence was removed from the Hanle signal.

The corrected Hanle signal was then computer fitted to a Lorentzian by a nonlinear least-squares technique. A typical result of the corrected signal fit to a Lorentzian is shown in Fig. 1. An attempt to fit the uncorrected signal to a Lorentzian generally resulted in a very poor fit and an error of as much as 15% in the width.

It is well known that at moderate densities, coherent multiple photon scattering narrows the measured Hanle-effect linewidth.[9] Measurements were taken over an order-of-magnitude relative ion density to look for coherence narrowing. In the ion density region of our experiment, less than 10^{10} cm^{-3}, coherence narrowing was not observed within experimental error. After coherence narrowing was determined to be negligible, attention was turned to collisional depolarization.[10] The collisional depolarization effect can be described by the equation

$$1/\tau = 1/\tau_0 + n\sigma v ,$$

where τ_0 is the true radiative lifetime, n is the helium density, σ is the alignment depolarization cross section, and v is the relative collisional velocity. When the collisional depolarization term $n\sigma v$ is not very much smaller than the radiative decay constant $1/\tau_0$, the collisional term must be eliminated by extrapolation to zero helium density. Figure 2 shows such a plot for the Sr II $^2P_{3/2}$.

Since Ba and Cd have a relatively high percentage of odd isotopes in natural abundance, it was deemed necessary to calculate their effect on the Hanle signal linewidth. The calculation was carried out

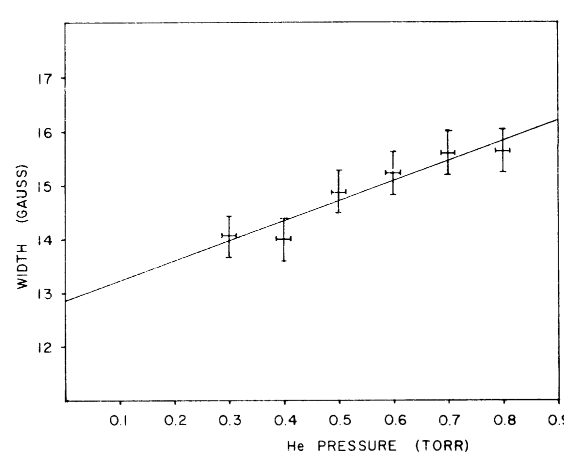

FIG. 2. Extrapolation to zero helium pressure. Data shown is for Sr II $^2P_{3/2}$ Hanle signal.

TABLE I. Radiative lifetimes of some group II $^2P_3$ ions.

Element	Configuration	τ (10^{-9} sec) This work	σ (10^{-15} cm^2) This work	τ (10^{-9} sec) Other work
Mg	$3p$	3.65(0.12)	5.65(2.8)	3.67(0.18) [a]
Ca	$4p$	6.61(0.30)	10(2.8)	6.72(0.2) [a]
Zn	$4p$	2.4(0.3)	...	3.0(0.3) [b] 3.1(0.4) [c]
Sr	$5p$	6.64(0.19)	9.5(1.6)	6.53(0.20) [a] 5.63(0.17) [d]
Cd	$5p$	2.86(0.25)	...	2.6(0.2) [e] 3.4(0.4) [b] 3.4(0.7) [c]
Ba	$6p$	6.78(0.40)	19(6)	6.27(0.25) [a] 7.0(0.6) [f]

[a] Lifetime by Hanle effect. References 2 and 3.
[b] Lifetime by beam-foil technique. T. Anderson and G. Sorensen, J. Quant. Spectrosc. Radiat. Transfer 13, 369 (1972).
[c] Lifetime by phase-shift technique. S. R. Bauman and W. H. Smith, J. Opt. Soc. Am. 60, 345 (1970).
[d] Lifetime by Hanle effect. Reference 6.
[e] Lifetime by the Hanle effect. Reference 13.
[f] Lifetime by Hanle effect. H. Bucka, J. Eichler, and G. V. Oppen, Z. Naturforsch. 21, 654 (1966).

in the same manner as described in detail in Ref. 7. The effect is estimated to be no more than a 2% broadening for Ba and 3% broadening for Cd. Isotope effects in the other materials will be insignificant because of the low natural abundance.[3] We have chosen not to incorporate the hyperfine effect into the lifetimes, but rather state it as an added uncertainty.

EXPERIMENTAL RESULTS

Table I is a compilation of our results along with those of other authors. Our data represent the first lifetime measurements in a flowing helium afterglow on group II ions. Here, by a single technique in a single apparatus, we present radiative lifetimes for six group II ions in the $^2P_{3/2}$ state. Any systematic error would therefore be expected to appear in all measurements. Error bars given in the data are always greater than rms scatter and should at least partially compensate for any systematic error.

Our data are in extremely good agreement with that of Gallagher and Smith. Interestingly enough, the only significant disagreement with their data is the Ba II lifetime, which is closer to the value of Bucka[11] *et al*. There is significant disagreement between Kelly's value for the Sr II lifetime, and that of Gallagher and our own. Our greatest source of experimental error was from the ion background field dependence, as described previously. Had we not corrected for this effect, our measured lifetime would have been about 5.7 nsec,[12] very near Kelly's 5.63. Unfortunately, we could not test Kelly's claim of high depolarization cross section from collisions with neutral Sr since our neutral density were so low ($\leq 10^{12}$ cm^{-3}). His value for the depolarization cross section of 8.4×10^{-13} cm^2, even though large, would have an insignificant effect on our measured lifetime.

Our lifetimes for Zn II and Cd II $^2P_{3/2}$ levels are consistently 15% shorter than measurements by beam-foil and phase-shift techniques. Hamel and Barrat[13] have measured the lifetime of Cd II by the Hanle effect in a dc discharge with He as the buffer gas. Our lifetimes agree with theirs to within experimental error. They also report a depolarization cross section with He of 4.6×10^{-15} cm^2. At the low helium pressure of our experiment, this effect would be very small compared to the error bars. For the same reason, the depolarization cross section for Zn was not measured. From the other measurements it is expected to be $\leq 10^{-14}$ cm^2, and again would have negligible effect on our lifetime measurement, which was taken at about 0.2-Torr He compared to the 1–4 Torr range of Ref. 13.

The depolarization cross sections reported here have large experimental errors because the pressure range covered was too small for some of the small width/Torr slopes encountered when plotting width versus helium pressure. The values obtained here, however, do fall within the range predicted for ion-atom collisions.[3]

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