01 Jan 1976

Radiative Lifetimes And Alignment Depolarization Cross Sections For YbI And II By The Hanle Effect In A Flowing Helium System

F. H.K. Rambow

Laird D. Schearer
Missouri University of Science and Technology

Recommended Citation

F. H. Rambow and L. D. Schearer, "Radiative Lifetimes And Alignment Depolarization Cross Sections For YbI And II By The Hanle Effect In A Flowing Helium System," *Physical Review A*, vol. 14, no. 2, pp. 738 - 743, American Physical Society, Jan 1976.
The definitive version is available at https://doi.org/10.1103/PhysRevA.14.738

PHYSICAL REVIEW A VOLUME 14, NUMBER 2 AUGUST 1976

Radiative lifetimes and alignment depolarization cross sections for Yb I and II by the Hanle effect in a flowing helium system*

F. H. K. Rambow and L. D. Schearer
Physics Department, University of Missouri-Rolla, Rolla, Missouri 65401

(Received 29 March 1976)

The radiative lifetimes of the 17992-, 25068-, 28857-, 37414-, 40564-, and 44017-cm^{-1} neutral levels and the 30392-cm^{-1} ion level of Yb have been measured by the Hanle method in a fast-flowing He system. The lifetimes (in units of 10^{-9} sec) were found to be 820(20), 5.12(0.12), 14.4(0.4), 77.4(6.0), 9.32(0.6), 39.1(3.5), and 5.8(0.6), respectively. In a flowing system with He as a buffer gas the alignment depolarization cross sections with Yb were obtained and are reported here for the first time. They are (in units of 10^{-15} cm^2) 5.0(1.0), 5.9(1.0), 5.9(1.2), 17.9(2.0), 28.2(3.0), 5.16(4.0), and 7.2(2.5), respectively.

INTRODUCTION

Lifetime and depolarization cross sections are important parameters of excited states of atoms. We report here radiative lifetimes and collisional depolarization cross sections for six odd levels of neutral ytterbium and one level of singly ionized ytterbium as measured by the Hanle effect[1] in a flowing system with helium as the buffer gas. The electron configuration of one of these neutral levels (37 414 cm^{-1}) has not yet been identified.[2] Although ytterbium was discovered in 1907, relatively little progress was made towards a quantum description of its spectra until recently, due in part to the difficulty of obtaining pure samples of Yb. Since 1950 more efficient light sources have been developed and relatively large quantities of high-purity rare earths have been accumulated as by-products of the purification of thorium and uranium by ion-exchange chromatography.

A useful description of its arc and spark spectra was first given by Meggers and Scribner in 1937.[3] The first useful Zeeman measurements on Yb I were made in the late 1950's. In 1965 Meggers and Corliss published data for some 7300 spectral lines, including Zeeman classification of 1300 lines.[4] At present a quantum description of Yb I is being prepared for publication by J. Tech at the U. S. National Bureau of Standards.[2] However, the electron configuration of several levels remain unidentified.

We found few reliable lifetime measurements and no depolarization cross sections in the literature for Yb. One of the first lifetime measurements on Yb was that of Baumann and Wandel in 1966.[5] They measured radiative lifetimes for the ^1P and ^3P levels of the $4f^{14}6s6p$ configuration of Yb I by the Hanle effect in a beam. These and all subsequent measurements can be broken roughly into three groups. The longest lifetimes were measured by the technique of anomalous dispersion and total absorption.[6] These averaged 20% longer than measurements in the second group using the Hanle technique and taken by Bauman and Wandel, Lange *et al.*,[7] and ourselves. Also agreeing with these data for the neutrals and included in the second group was the technique of delayed coincidence[8] used by Burshtein *et al.*, which gave a value only slightly longer than that obtained via the Hanle effect. However, their value for the ion lifetime, which incidentally is the first such measurement, is about 20% longer than ours. Possible reasons for this will be discussed later. The third group, also using the Hanle method but as a by-product of level crossing hyperfine studies, was that of Budick and Snir.[9,10] Their values obtained for the $6s6p\,^1P_1$ and $4f^{13}5d6s\,^2(3\frac{1}{2},2\frac{1}{2})_1^o$ levels were 100% and 20% longer, respectively, than those in the second group. Their value for the $6s6p\,^3P_1$ was only slightly shorter than that of the second group. These discrepancies can be explained if coherence narrowing and collisional depolarization were not properly taken into account.

In this experiment we were particularly careful to account for these effects. The data reported here have been corrected for collisional broadening and coherence narrowing. Our measurements of collisional depolarization cross sections are, to our knowledge, the first reported. Furthermore, the fluctuation of a data point over several measurements averaged <2% for the neutral atom due to the large signal-to-noise ratio. The error limits we have set in Table I represent maximum confidence limits but do not take into account hyperfine effects. Scatter of the data lies within these limits in all cases. The influence of nuclear spin is considered separately.

APPARATUS

The Hanle effect in a flowing system is particularly suited to measurements of lifetimes and depolarization cross sections. It allows for rapid change and easy monitoring of system parameters, such as ytterbium density and buffer-gas pressure. Thus collisional depolarization and radiation trapping can be easily controlled and their effects observed. Another advantage is that material under study can be quickly changed and, unlike a beam apparatus or sealed cells, high-vacuum techniques are not necessary, since flow rates are many orders of magnitude greater than outgassing rates.

The flow tube is welded from aluminum conduit and is evacuated by a mechanical forepump and roots blower combination rated at 540 ft^3/min. With helium admitted at the entrance of the flow tube, flow velocities on the order of 10^3–10^4 cm/sec were obtained with background He pressures in the range of 0.01–1 Torr. The Yb vapor is titrated into the interaction region by an oven wound with coaxial heater wire. The resonance light source is a flowing hollow cathode lamp with a water-cooled anode and air-cooled Pyrex jacket. Resonance emission from the lamp was monitored by a 1-m Jarrell Ash monochromator having a resolution of 0.05 Å. No evidence for broadening or self-reversal of the line profile was observed within this limit.

A schematic diagram of the apparatus is shown in Fig. 1. Light from the flowing hollow cathode is focused by a fused quartz lens through a quartz window into the flow region just above the oven. Magnetic field coils in a Helmholtz configuration are mounted coaxially on the flow tube. Three more sets of Helmholtz coils cancel the Earth's field in the scattering region to less than 10 mG residual field. Light scattered perpendicular to both the incident light direction and the magnetic field is collected and passed through a $\frac{1}{4}$-m Jarrel Ash monochromator with 1000-μm slits to a photomultiplier.

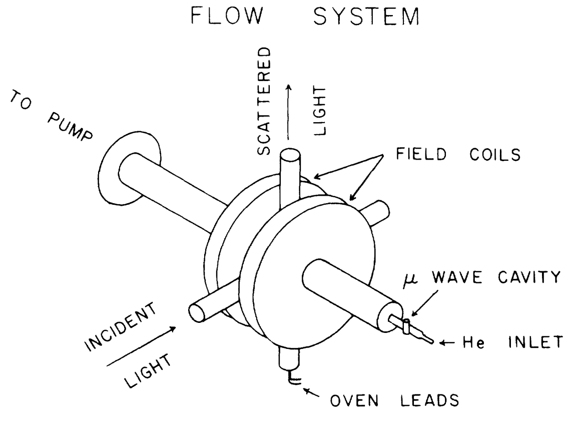

FIG. 1. Schematic diagram of apparatus.

The magnetic field is swept at 18 Hz by a triangular current waveform. The current waveform is shaped by a feedback network that holds the sweep linear to better than 0.5% over the entire waveform. Linearity can be monitored by two methods. One is the helium magnetometer, which is discussed below, and the other is a probe coil inserted into the flow tube. When the voltage induced in this coil is integrated it provides an accurate picture of the magnetic field inside the flow tube. Linear field sweeps of over 100 G peak to peak were obtained.

One of the advantages of performing this experiment with a flowing He system is the *in situ* field calibration it allows. A microwave discharge at the flow inlet produces He metastables which drift down the tube and are optically pumped by light from a flowing He lamp focused just above the oven.[11] A small rf coil provides the oscillating magnetic field to induce the Zeeman transitions. The He light is monitored in transmission by a Polaroid infrared polarizer filter in conjunction with an infrared-sensitive photodiode. This provides not only a calibration of the field but also a check on field sweep linearity. Linearity is defined by measuring the spacing between resonances in gauss for a series of radio frequencies, i.e., 10, 20, . . . , 50 MHz, and dividing the maximum difference in spacing by the average spacing.

This same flowing system was used for Hanle measurements on the ion. With the microwave-excited source of helium metastables (He^m) at the inlet to the flow tube, Penning ionization of Yb by He^m produces copious quantities of ions in steady state in a field-free region. Typical cross sections for Penning ionization are on the order of 10^{-15} cm^2. Resulting ion densities as high as 10^{10} cm^{-3} are estimated by optical absorption. This technique releases one from concerns over perturbations due to an ionizing electric field and decreases necessary signal accumulation time over pulsed techniques.

EXPERIMENTAL TECHNIQUE

Briefly, the Hanle method is a well-known zero-field level-crossing technique in which resonance radiation is used to coherently excite atoms in the presence of a magnetic field. The resonance fluorescence is observed as a function of magnetic field and from this resulting plot the decay constant is obtained as a function of the *g* factor. We have chosen the common geometry and polarization

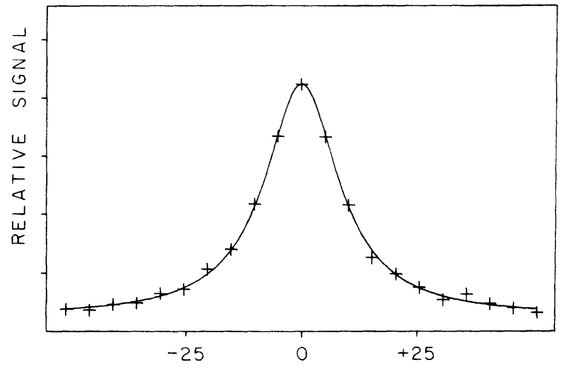

FIG. 2. Yb II Hanle signal fit with Lorentzian. The solid line is the computer fit.

in which the signal due to fluorescence reduces to a Lorentzian,

$$S=[1+(2g\mu H\tau/\hbar)^2]^{-1}. \tag{1}$$

Here g is the Landé g factor, μ the Bohr magneton, H the field in gauss, τ the lifetime in seconds, and $\hbar$ Planck's constant reduced.

The decay constant obtained from this curve is then related to the radiative lifetime by

$$\Gamma^{(L)}=\Gamma-\sum_i \alpha_i^{(L)}x_i\Gamma_i+\gamma_L, \tag{2}$$

with the parameters as given by Saloman and Happer.[12] The radiative lifetime τ_0 is just $1/\Gamma$. In the limit of low Yb density which characterizes our work, x_i, the reabsorption probability, reduces to K_iN and the expression becomes[13]

$$\frac{1}{\tau_{\mathrm{eff}}}=\frac{1}{\tau_0}-\frac{1}{\tau_0}\sum_i\frac{\tau_0}{\tau_i}\alpha_iK_iN+n\sigma\bar{V}. \tag{3}$$

The first term on the right-hand side is the radiative lifetime, the middle term is due to radiation trapping, and the last term accounts for collisional depolarization. The coefficients α_i and K_i depend on the states involved. The sum is over i branches, with τ_0/τ_i the branching ratio for the ith branch. N is the Yb density. In the last term n is the helium density, σ the alignment depolarization cross section, and $\bar{V}$ the average relative collisional velocity.

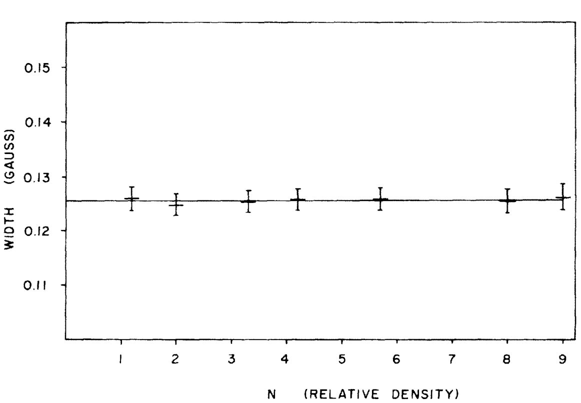

FIG. 3. Linewidth vs Yb density for the $4f^{14}6s6p\ {}^3P_1^o$ level. The horizontal scale is in units of scattered fluorescent intensity, which is linearly related to the Yb density.

All data were taken by accumulating signal vs magnetic field with a Fabri-Tek 1060 signal averager. The resulting curve was plotted on an x-y recorder. All plots were subsequently digitized by a Tektronix Graphics Tablet and the full width at half-height was extracted by a nonlinear least-squares fit to a Lorentzian. A typical fit is shown in Fig. 2. In order to obtain the radiative lifetime a double extrapolation was used. For each helium pressure a set of line widths vs Yb density was extrapolated to zero Yb density by a least-squares fit to a straight line, thus leaving the data independent of Yb density. A typical extrapolation of the linewidth to zero Yb density is shown in Fig. 3. The relative density is obtained from the intensity of the resonance fluorescence. A number of width measurements were taken at each Yb density to reduce random error. The peak-to-peak scatter among the widths at each density was seldom over 4% and on the average was under 2%. Finally, the width vs helium pressure data was plotted and least-squares fitted by a straight line. From the intercept and slope of this line the radiative lifetime and alignment depolarization cross section were deduced. The plot of width vs helium pressure for the $4f^{14}6s6p\,{}^3P_1$ level is shown in Fig. 4 as an example.

The next consideration was that of hyperfine effects. Ytterbium has a 30% natural abundance of odd isotope, with 14% spin-$\frac{1}{2}$ and 16% spin-$\frac{5}{2}$. Even with a 30% abundance one would expect the effect on lifetime measurements to be a few per-

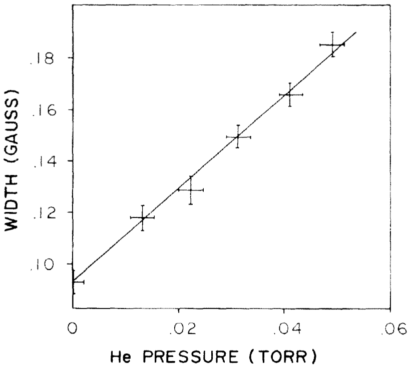

FIG. 4. Width vs helium pressure plot for the $4f^{14}6s6p\ {}^3P_1^o$ levels.

TABLE I. Experimental results.

				Present data		Other data
Line (Å)	Term	Configuration	g_J	σ (10^{-15} cm^2)	τ (nsec)	τ (nsec)
				Yb I		
5556	$^3P_1^o$	$4f^{14}6s6p$	1.48	5.0(1.0)	820(20)	827(40) [a] 760(80) [b] 850(80) [c] 980(70) [d]
3988	$^1P_1^o$	$4f^{14}6s6p$	1.035	5.9(1.0)	5.12(0.12)	5.5(0.25) [a] 12 [f] 5.63(0.25) [e] 6.4(0.2) [d]
3464	$(3\frac{1}{2},3\frac{1}{2})_1^o$	$4f^{13}5d6s^2$	1.26	5.9(1.2)	14.4(0.5)	17 [b] 14.3(0.9) [e] 18.3 [d]
2672	$(?)_1^o$	?	1.02	17.9(2.0)	77.4(6.0)	57.3(4) [e] 93 [d]
2464	$^1P_1^o$	$4f^{14}6s7p$	1.01	28.2(3.0)	9.3(0.6)	10.4(1.5) [e] 12.4 [d]
2272	$^1P_1^o$	$4f^{14}6s8p$	1.00	51.6(4.0)	39.1(3.5)	
				Yb II		
3290	$^2P_{3/2}^o$	$4f^{14}6p$	1.333	7.2(2.5)	5.8(0.6)	7.3(0.6) [c]

[a] M. Baumann and G. Wandel, Ref. 5.
[b] B. Budick and J. Snir, Ref. 9.
[c] M. L. Burshtein *et al.*, Ref. 8.
[d] V. A. Komarovskii and N. P. Penkin, Ref. 6.
[e] W. Lange *et al.*, Ref. 7.
[f] B. Budick and J. Snir, Ref. 10.

cent at most.[14] A careful analysis of the hyperfine structure supports this claim. Assuming a flat lamp profile, it was found for our method of analyzing the Hanle curves the lifetimes could be 3% and 5% longer for neutrals and ions, respectively, than uncorrected results would suggest. The details of these calculations are given in the Appendix.

A unique complication arises with the ion Hanle signal. The Penning reaction which produces our ground-state ion density heavily populates the $4f^{14}6p$ level. The resulting radiation at 3290 Å is much greater than the Hanle signal and obscures the direct observation on the oscilloscope. Since the ions interact with the applied magnetic field, the ion density is a slowly varying function of field. In order to unscramble the Hanle signal a procedure similar to that of Smith and Gallagher[15] was employed, in which two sets of curves are taken. One set is taken with the polarizer parallel to the field and gives only the ion dependence on magnetic field. The other is taken with the polarizer perpendicular to the field and gives a composite of the Hanle signal with the ion-density field dependence. The result of this correction fitted to a Lorentzian is shown in Fig. 2.

RESULTS

Our data represent the first lifetime measurements made on ytterbium in a flowing system. We were able to extend the number of level lifetimes measured (the $4f^{14}6s8p$ radiating at 2272 Å) and to improve upon the accuracy of existing measurements. As a by-product of lifetime measurements in a flowing system we also obtained the first alignment depolarization cross sections for ytterbium with helium.

The experimental results are presented in Table I. Lifetimes are compared to those measured by other authors. It should be noted that the lifetimes of Komarovskii and Penkin[6] are not direct measurements but are calculated from oscillator strengths obtained by anomalous dispersion and total absorption. These measurements average some 20% longer than ours. Once this 20% adjustment is made the agreement is excellent, implying a systematic error in one technique or the other. The large value obtained for the $6s6p\,^1P_1^o$

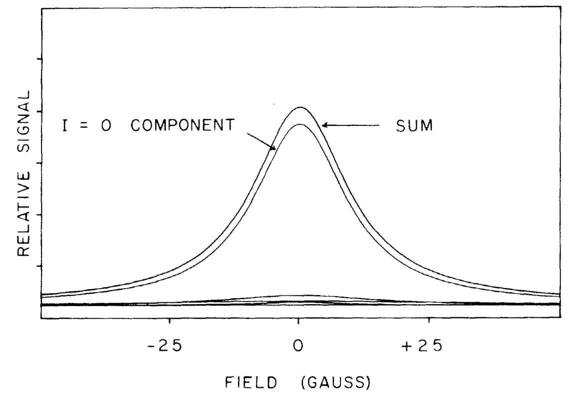

FIG. 5. Components of the Hanle signal for $J=1$ Yb I levels.

lifetime by other workers using the Hanle technique can probably be explained by coherence narrowing, in view of this level's large oscillator strength. Our excellent agreement with other Hanle effect and delayed coincidence measurements of the $6s6p\,^3P_1$ level lifetime, which has an oscillator strength 100 times smaller than the singlet level, supports this claim. This gives us confidence in our lifetime measurement of the $6s8p\,^1P_1^o$ level reported here for the first time and in the lifetime of the $6p\,^2P_{3/2}^o$ ion level.

The lifetimes given in Table I do not contain a correction for hyperfine effects. As discussed in the Appendix, these could result in as much as a 3% increase in neutral lifetime and a 5% increase in the $6p$ ion level lifetime. If this correction is included, our value for the Yb II $^2P_{3/2}^o$ level becomes 6.09 ± 0.6 nsec, which agrees with the value of Burshtein *et al.* at the limits of experimental error. We chose not to incorporate the hyperfine effect into the lifetime because of our lack of knowledge of the lamp profile, but rather state it as an added uncertainty.

Alignment depolarization cross sections are presented without comment save the qualitative observation that for higher nl states σ is larger. No attempt was made to bring the uncertainties for the cross sections in line with those of the lifetimes. The larger experimental errors are due mostly to lack of data over a wide enough pressure range for some of the small width/Torr slopes encountered when plotting width vs helium pressure.

APPENDIX

Using the standard vector coupling coefficients of Condon and Shortley[16] and assuming a broadband "white light"[17] source we obtained the following expression for the signal from the $J=0$ to $J=1$ transitions in Yb with nuclear spins $I=0$, $\frac{1}{2}$, and $\frac{5}{2}$:

$$
\begin{aligned}
S = 1 &- 0.808(\cos^2\alpha\sin^2\alpha' + \cos^2\alpha'\sin^2\alpha) \\
&+ 0.808\cos^2\alpha\cos^2\alpha' \\
&- \sin^2\alpha\sin^2\alpha'\left(\frac{0.7}{1+x^2} + \frac{0.06}{1+(0.667x)^2} + \frac{0.001}{1+(0.4x)^2}\right. \\
&\qquad\left. + \frac{0.026}{1+(0.267x)^2} + \frac{0.021}{1+(1.2x)^2}\right), \qquad (4)
\end{aligned}
$$

with

$$x = 2g_J\mu H\tau/\hbar.$$

Here α and α' are the angles the polarization vector makes with the magnetic field direction for incident and reflected beams, respectively. In this experiment we used a polarizer on the detector for all wavelengths above 2672 Å and no polarizers for 2672 Å and below. Upon analyzing the broadening produced with and without the polarizer we found the difference to be completely negligible.

To determine the extent of broadening, the sum of the Lorentzian was compared to a simple Lorentzian ($I=0$ case) by the least-squares-fit program. Also, each Lorentzian component, together with the sum, was plotted and is shown in Fig. 5. It becomes apparent that the very broad components approximate a constant background and thus have little effect on the width of the sum except far into the wings. Using the least-squares fit we found approximately to 2–3% increase in the full width at half-height over the $I=0$ case, which implies that the lifetime is 2–3% longer than the uncorrected results would suggest.

An identical procedure was followed for the ion. The signal as a function of polarization angle was found to be

$$
\begin{aligned}
S = 1 &- 0.470\,(\cos^2\alpha\sin^2\alpha' + \cos^2\alpha'\sin^2\alpha) + 0.470\cos^2\alpha\cos^2\alpha' \\
&- \sin^2\alpha\sin^{-2}\alpha'\left(\frac{0.42}{1+x^2} + \frac{0.019}{1+(1.25)^2} + \frac{0.043}{1+x^2} + \frac{0.0003}{1+x^2} + \frac{0.005}{1+(0.083x)^2} + \frac{0.001}{1+(0.292x)^2} + \frac{0.017}{1+(0.375x)^2}\right). \qquad (5)
\end{aligned}
$$

The effect on measured lifetime in this case is 3–5%, again depending upon how far into the wings the least-squares fit was made.

*Research supported by Office of Naval Research under Contract No. ONR-N00014-69-A-0141-0004.

[1] W. Hanle, Z. Phys. 30, 93 (1924).

[2] J. Tech, J. Res. Natl. Bur. Stand (U.S.) (to be published).

[3] W. F. Meggers and B. F. Scribner, J. Res. Natl. Bur. Stand. (U.S.) 19, 651 (1937).

[4] W. F. Meggers and C. H. Corliss, J. Res. Natl. Bur. Stand. (U.S.) A70, 63 (1966).

[5] M. Baumann and G. Wandel, Phys. Lett. 22, 283 (1966).

[6] V. A. Komarovskii and N. P. Penkin, Opt. Spektrosk. 26, 882 (1969) [Opt. Spectrosc. 26, 483 (1969)].

[7] W. Lange, J. Luther, and A. Steudel, Summaries of the Second European Group on Atomic Spectroscopy Conference, Hanover, 1970 (unpublished).

[8] M. L. Burshtein, Ya. F. Verolainen, V. A. Komarovskii, A. L. Osherovich, and N. P. Penkin, Opt. Spektrosk. 37, 617 (1974) [Opt. Spectrosc. 37, 351 (1974)].

[9] B. Budick and J. Snir, Phys. Rev. A 1, 545 (1970).

[10] B. Budick and J. Snir, Bull. Am. Phys. Soc. 12, 186 (1967).

[11] F. D. Colgrove and P. A. Franken, Phys. Rev. 119, 680 (1960).

[12] W. Happer and E. B. Saloman, Phys. Rev. 160, 23 (1967).

[13] W. W. Smith and A. Gallagher, Phys. Rev. 145, 26 (1966).

[14] See L. G. Williams and D. R. Crosley, Phys. Rev. A 9, 622 (1974); see also A. Gallagher, Phys. Rev. 157, 24 (1966), for a discussion of hyperfine effects.

[15] W. W. Smith and A. Gallagher, Phys. Rev. 145, 26 (1966).

[16] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge U. P., London, 1970), Chap. 3.

[17] One might expect our "white light" source to be a good approximation. The lamp pressure was on the order of 50 Torr with a current of 3 A. Thus Stark and pressure broadening could be expected to contribute significantly to the emission-line width.