Facility

MAY 7 8 28 AM '79

RECEIVED

Oscillator Strength Trends in Group IVb Homologous Ions*

Myron H. Miller
Department of Mechanical Engineering
United States Naval Academy
Annapolis, Maryland 21402

and

Institute for Physical Science and Technology
. University of Maryland
College Park, Maryland 20742

and

Roger D. Bengtson
Department of Physics
University of Texas at Austin
Austin, Texas 78712

Shock tube data are used to examine systematic f value behavior in prominent visible transition arrays (ns-np, np-(n+1)s, np-nd) for the homologous emitter sequence Si II, Ge II, Sn II and Pb II. Regularities found in these data are compared with trends in lighter elements. Agreements and disparities with theoretical and experimental oscillator strengths from the literature are noted.

(NASA-CR-158688) OSCILLATOR STRENGTH TRENDS IN GROUP IVb HOMOLOGOUS IONS (Naval Academy) 1? p HC A02/MF A01 CSCL 20L

N79-25909

Unclas
G3/76 26424

*Research supported in part by NASA Grant No. NGR44-012-258.

Introduction

Systematic trends in line strengths within homologous sequences, spectral series and isoelectronic sequences are effective aids for assessing theoretical and experimental gf data.[1,2] Owing in part to difficulties in introducing heavier elements into controlled spectroscopic light sources, homologous sequence behavior has been the least extensively investigated of these trends, with examination to date being confined to the first three rows of the periodic table,[1,2] primarily to discriminate between gf value data for some light metals and S I line strengths.[1]

Recent theoretical calculations have predicted systematic behavior in strengths of the dominant visible transition arrays of the group IVb first ions: Si II, Ge II, Sn II, Pb II.[3] Verification of this trend, which spans four rows of the periodic table, is the primary purpose of this paper. Our secondary purpose is to assess the comparative reliability of traditional Coulomb approximations[4] and relativistic, semiempirical calculations[3] in predicting heavy element line strengths. Finally, we examine cases where regularities in homologous sequence help discriminate between various experimental line strengths for the group IVb ions. The subject spectra are accessible to ground-based astronomy and will be prominent under excitation conditions prevailing in type A, O and B stars. The roles of silicon burning in heavy element synthesis, and of lead as the end point of the uranium decay chain, heighten interest in the abundance implications of these data.

Experimentally, the group IVb first ions are particularly suitable for examining line strength trends. The dominant visible transition arrays (sharp, principle and diffuse series) provide excellent signal-to-noise ratios, but are sufficiently Stark effect broadened[5-8] so that peak optical depths remain manageable. Ample interline separations avoid serious blending, even at the high electron densities (4-16 x 10^{16} cm^{-3}) relied upon to establish local thermal equilibrium.[9] Interference from the simultaneously excited neutral spectra is minimal. Undue complexity in theoretical modelling is avoided because the dominant transitions involve no core-equivalent electrons, and the energy level separations are not conducive to configuration mixing.[1]

Identical shock tube apparatus and methods are used to excite, record, and analyze Si II, Ge II, Sn II and Pb II line strengths[7,8,11,12]. Through out, the experiments were conducted in accordance with a program to minimize the usual sources of systematic error. Because the spectroscopic range encompassed the region from the near UV to the near IR, gf values were obtained for all the bright lines in each transition array, thereby permitting results to be tested for conformity with quantum mechanical sum rules.[13]

Experimental

The oscillator strengths in this examination of systematic trends were obtained from the same well-instrumented gas driven shock tube. Initial work[14,15] addressed shock behavior and conformity of the plasma light source to a homogeneous, LTE model. Details of apparatus and procedures are given elsewhere,[7,8,11,12,16,17] the intent here being to review the generic experimental problems and how they were addressed.

The two primary drawbacks to shock tube line strength determinations are: (1) critical sensitivity of emitter densities to errors in measured source temperature, and (2) absolute photographic photometry's proneness to bias, especially when emulsions have limited dynamic range and nonlinear gamma curves.

Sensitive dependence on temperature data arises when emitting levels have excitation potentials much larger than mean thermal energies. In the shock tube (typical mean thermal energy of 1 eV), a repeatable temperature error of 3-4% is translated by the Saha-Boltzmann relations into a 50% error in the density of ionic emitters. A four part strategy minimizes this traditional error source.

(1) Hydrogen Balmer lines are used as an internal f value standard for transspecies relative intensity measurements.[7,8,11,12] The populations as functions of temperature are quite similar for H_β and the subject ion lines, so that a few percent error in measured temperature induces only minor error into measured gf values. Lack of demixing of plasma constituents in the shock tube make it possible to relate the hydrogen and group IVb abundances through the fixed ratios of the test gas SiH_4, $Si(CH_3)_4$, GeH_4, $Sn(CH_3)_4$ and $Pb(C_2H_5)_4$ constituents.

(2) In each experiment, temperatures are measured redundantly. A black body temperature is found by photoelectrically recording

the emission and absorption of[18,19] optically thick H_α, temporarily backlighted by a bright flashlamp. Electron densities, measured via H_β halfwidths,[20,21] together with pressures recorded by fast transducers, are inverted to find a Saha equilibrium temperature. The excitation temperatures of NeI λ5852Å and H_β are obtained by photoelectrically monitoring the integrated intensity of these lines. In a typical experimental run, these four independent temperatures group within ±5% of their mean.

(3) The range of source conditions is deliberately varied ($10,000^\circ K < T < 13,000^\circ K$) to facilitate testing for possible temperature-induced bias.

(4) Carbon is a constituent in most of the test gases. Measurement of the CI λ5052Å oscillator strength, which is comparatively well known,[17,22] serves to gage accuracy of simultaneous ion line strength determinations.

Among the steps taken to reduce repeatable error in time-resolved photographic phontometry are:

(1) Photographic recording is used only to measure relative line intensities: the question of absolute photometric repeatability

therefore does not arise. Line strengths were measured relative to H_{β}, whose gf value is known. Routine compensation for line's finite optical depths relies on simultaneously recorded absolute multichannel photoelectric data.

(2) In each experiment the same emission lines are recorded by two spectrographs. These view the plasma along similar optical paths, but have different resolutions and use different emulsion types. One spectrograph's instrument profile is small compared to ionic line widths, and the well-resolved line profiles it records are fitted to appropriate Voigt shapes.[23] The second instrument's slits essentially integrate ionic lines: these profiles are read with a planimeter. Relative line intensities obtained these two ways agree satisfactorily when exposures are adequate.

(3) A program of photometric quality control resulted in approximately one half of the data being rejected. Criteria for this culling included:

(a) Whether photographic sampling times fell within the quiescent region behind first-reflected shock waves (this was ascertained from photoelectric monitors positioned at the photographic focal plane).

(b) Attainment cf desirable signal-to-noise (grain) ratios and registry of the exposures upon the optimal portion of the emulsion's response range.

(c) Whether the peaks of the brightest ion lines (generally, ns-np) had optical depths exceeding 0.5 (beyond this, compensation for radiative trapping was deemed unreliable).

Precision in relating the integrated energies in two line profiles was estimated at typically 25-30% per experiment. After rejecting data from the photometrically inferior shots, the relative strength of each ionic line was measured in 10-40 experiments. Statistical analysis of random error in resultant gf values yielded uncertainties in conformity with these estimates-- i.e., approximately 5-10%, depending on line strength, wavelength and freedom from interference. Overall accuracy, including provision for possible repeatable error, was typically 25% for the absolute gf values of the more prominent ionic lines. Multiplet average f values are based on shock tube results for 2-3 individual lines per multiplet and are felt to be reliable to $\pm$20% (90% confidence limits).

A gross gage of the reliability of this data set is provided by the f sum rules for one electron system.[13] Because the preponderance of radiative channels into and out of the np levels involve a single optically active electron orbiting a closed s-shell, one expects approximate conformity with these sum rules. Table 1 compares shock tube results with the f sums expected from the Wigner-Kirkwood (WK) and Thomas-Reichie-Kuhn (TRK) sum rules. Although Coulomb and asymptotic approximations are used to augment the shock tube data, experimental findings are seen to comprise the majority of each sum. In cases where the shock tube data has provided the strengths of two-electron transitions to or from np levels, these f values are included in the sums: otherwise, they are neglected. The ns-np partial sums are difference measurements, ns-np transitions having opposite sign from (n + 1)s - np transitions. The experimental (with Coulomb approximation augmentation) sums agree within tolerance with the WK and TRK expectation values without exception.

It reflects favorably upon the reliability of relative photographic photometry to note that the relative strengths of lines in transition arrays agreed satisfactorily with J-file sum rules[24] applied to the four spectra.[7,8,11,12]

Results and Discussion

Multiplet f values of group IVb ions from the shock tube and the literature are plotted in Figure 1 as functions of principle quantum number n for the visible members of principle, sharp and diffuse series. The shock tube data are connected by solid lines. The dashed lines represent best-fit linear regressions to the shock tube data: these have been extrapolated to C II (n = 3) to afford comparison with self-consistent field calculations by Weiss.[25] Deviations of the shock tube f values from the best-fit lines can be attributed to experimental error. The goodness of fit, with correlation coefficients for ns-np, np-(n + 1)s and np-nd being 0.75, 0.99 and 0.53, respectively, argues against the positive slopes of the trend lines being accidental. The mild positive slopes (m = 0.127, 0.053 and 0.056, respectively, for ns-np, np-(n+1)s and np-nd) resemble the trends noted by Wiese, et. al[1] in ns-np arrays for light element homologous sequences, where the corresponding slopes were 0.06 for the Li I, Na I and K I sequence and 0.08 for the Be I, Mg I and Ca I sequence.

Gradual increase of f with increasing n is to be expected heuristically since the f sum rules should be satisfied at each step in a homologous sequence,[1, 13] barring undue mixing, cancellation or core effects. As noted earlier, the subject transition arrays are strong and well isolated, so that cancellation and mixing should have little disruptive influence on the smoothness of the trends. Core effects are likewise expected to be minimal as compared with resonance transitions. Homologous trends may be less clearly evident for both the lower and higher members of these series because the resonance lines will be more sensitive to the core and the more highly excited states tend to be more susceptible to mixing and cancellation effects.

Theoretical comparison data consist mainly of the relativistic, semi-empirical calculations by Migdalek,[3] and Coulomb approximations computed by the authors using tabulated integrals of radial wave functions.[4] Of these, the relativistic predictions more closely approximate the experimental trends' slopes. Applying linear regressions to both sets of predictions, one finds that the ratio of slopes, $M_{\text{shock tube}}/M_{\text{predicted}}$ is $\cong 0.9$ and $\cong 0.5$ for Migdalek's calculations of the ns-np and np-(n-1)s sequences, respectively, and $\cong 0.5$ and $\cong 0.2$ for the corresponding Coulomb approximation ratios. However, inspection of Figure 1 shows that relativistic computations systematically overestimate, the mean of $f_{\text{relativistic}}/f_{\text{shock tube}}$ by approximately

1.15 which factor marginally exceeds experimental tolerance. Heavy element lifetime predictions using similar relativistic approaches tend to somewhat underestimate experimental determinations.[27] The Coulomb approximation f values scatter on both the high and low sides of the shock tube trend lines, with the result that the average for $f_{Coulomb}/f_{shock\ tube}$ is approximately 0.98.

Efforts to excite the C II spectrum to useful brightness in the shock tube were unsuccessful, so that no direct comparison can be made with the self-consistent field predictions of Weiss.[25] An interesting feature of Figure 1 is that the extrapolated best fit trend lines to shock tube data pass close to these SCF predictions for all three transition arrays.

The trend lines assist in unraveling the confusing pattern of agreements and discrepancies when comparison is made with other experiments. Shulze-Gulde[28] measured Si II relative line strengths with a controlled arc, scaling absolute values to fit Coulomb approximations. Subsequently, Berry, et. al[29] measured Si II radiative lifetimes. Normalizing the arc data to these lifetimes yields the absolute f values shown in Figure 1 as solid squares. Reducing the absolute values of these data by a factor of 1.6 brings them into satisfactory agreement with shock tube results and associated trend lines, suggesting that the lifetimes rather than the arc determinations of relative line strengths are the source of the discrepancy. The comparison values for Ge II were derived by Andersen, et. al[27] by combining lifetimes measured by the beam foil technique with theoretical branching ratios. Agreement is evident for ns-np transitions, and marginally, for the np-(n + 1)s case, but for np-nd, the lifetime-derived data is a factor of two smaller than the shock tube f value and associated homologous sequence trend line. However, oscillator strengths for 4p-4d, obtained by the same lifetime methods, are noted to fall smoothly onto trend lines for the Ge I isoelectronic sequence.[27] The two Sn II multiplet oscillator strengths denoted in the figure by solid triangles were measured in emission using a constricted arc.[30] For np-nd, agreement is close, but in ns-np, the arc data exceeds shock tube f values and trend lines by a factor of approximately 1.6. This latter marks the sole discrepancy out of the four Sn II multiplets measured in common by the two experiments.[7]

Conclusions

Systematic trends in homologous IVb ion line strengths are clearly discernable over four of the deeper rows in the periodic table for transition arrays involving relatively highly excited states. The utility of these trends for assessing experimental and theoretical gf values of astrophysical interest has been illustrated for three series (principle, sharp and diffuse) whose lines fall in spectral regions accessible to ground-based astronomy.

REFERENCES

1. W.L. Wiese, M.W. Smith and B.M. Glennon, Atomic Transition Probabilities, Natl. Bur. Stds. Res. Ser. 4 (U.S. GPO, Washington, D.C., 1969 , Vol. II.

2. M.W. Smith and W.L. Wiese, Astrophys. J. Suppl. Ser. No. 196, 23, 103 (1971).

3. J. Migdalek, J. Quant. Spectrosc. Radiat. Transfer 16, 265 (1976).

4. G. Oertel and L.P. Shomo, Astrophys. J. Suppl. 16, 175 (1968).

5. A. Lesage, S. Sahal-Brechot and M.H. Miller, Phys. Rev. 16, 1617 (1977).

6. W.W. Jones and M.H. Miller, Phys. Rev. 10, 1803 (1974).

7. M.H. Miller, R.S. Roig and R.D. Bengtson, "Experimental Transition Probabilities and Stark Broadening Parameters for Neutral and Singly-Ioned Tin," submitted to Phys. Rev., December, 1978.

8. M.H. Miller, R.D. Bengtson and J. Lindsey, "Line Strengths and Stark Effect Broadening Parameters for Neutral and Singly-Ionited Lead," manuscript in preparation.

9. H.R. Griem, Phys. Rev. 131, 1170 (1963).

10. C.E. Moore, Atomic Energy Levels, Natl. Bur. Stds. Circular 467 (U.S. G.P.O., Washington, D.C., 1952), Vols., I, II, III.

11. M.H. Miller, R.D. Bengtson and R.A. Roig, "Absolute Oscillator Strengths for Si I and Si II," manuscript in preparation.

12. M.H. Miller and R.A. Roig, Phys. Rev. 7, 1208 (1973).

13. H.A. Bethe and E.E. Salpeter, Quantum Mechanics of One-and-Two Electron Atoms (Springer, Berlin, 1957).

14. M.H. Miller, University of Maryland Tech. Note BN 550, 1968 (unpublished).

15. R.D. Bengtson, M.H. Miller, D.W. Koopman and T.D. Wilkerson, Phys. Fluids 13, 372 (1970).

16. R.D. Bengtson, M.H. Miller, D.W. Koopman and T.D. Wilkerson, Phys. Rev. 3, 16 (1971).

17. M.H. Miller, T.D. Wilkerson, R.A. Roig and R.D. Bengtson, Phys. Rev. 9, 2312 (1974).

18. W.R.S. Garton, W.H. Parkinson and E.M. Reeves, Proc. Phys. Soc. Lond. 88, 771 (1966).

19. M.H. Miller and R.D. Bengtson, J. Quant. Spectr. Radiat. Transfer 9, 1573 (1969).

20. P. Kepple and H.R. Griem, Phys. Rev. 173, 317 (1968).

21. W.L. Wiese, D.E. Kelleher and D.R. Paquette, Phys. Rev. 6, 1132 (1972).

22. W.L. Wiese, M.W. Smith and B.M. Glennon, Atomic Transition Probabilities, Natl. Bur. Stds. Res. Ser. 4 (U.S. GPO, Washington, D.C., 1966), Vol. I.

23. J.T. Davies and J.M. Vaughan, Astrophys. J. 137, 1302 (1963).

24. E.U. Condon and G.H. Shortley, The Theory of Atomic Spectra (Cambridge, England, 1935).

25. A.W. Weiss, Private communication (1964): cited in ref. 22.

26. D.R. Bates and A. Damgaard, Phil. Trans. R. Soc. London, A, 242, 101 (1949).

27. T. Andersen and A. Lingard, J. Phys. B: Atom. Molec. Phys. 10, 2359 (1977).

28. E. Schulz-Gulde, J. Quant. Spectrosc. Radiat. Transfer, 9, 13 (1969).

29. H.G. Berry, J. Bromander, L.J. Gurtis and R. Buchta, Physica Scripta 3, 125 (1971).

30. T. Wujec and S. Weniger, J. Quant. Spectrosc. Radiat. Transfer 18, 509 (1977).

Figure Captions

Figure 1. Transition array oscillator strengths displayed as functions of principle quantum number. Solid lines connect shock tube results. Regressions of shock tube data, extended to n = 3 for comparision with CII predictions, are given as dashed lines.

■ Berry, et. al, Ref. 29

▲ Wujec, et. al, Ref. 30

▬ Anderson, et. al, Ref. 27

○ Migdalek, Ref. 3

△ Coulomb approximation, Ref. 4

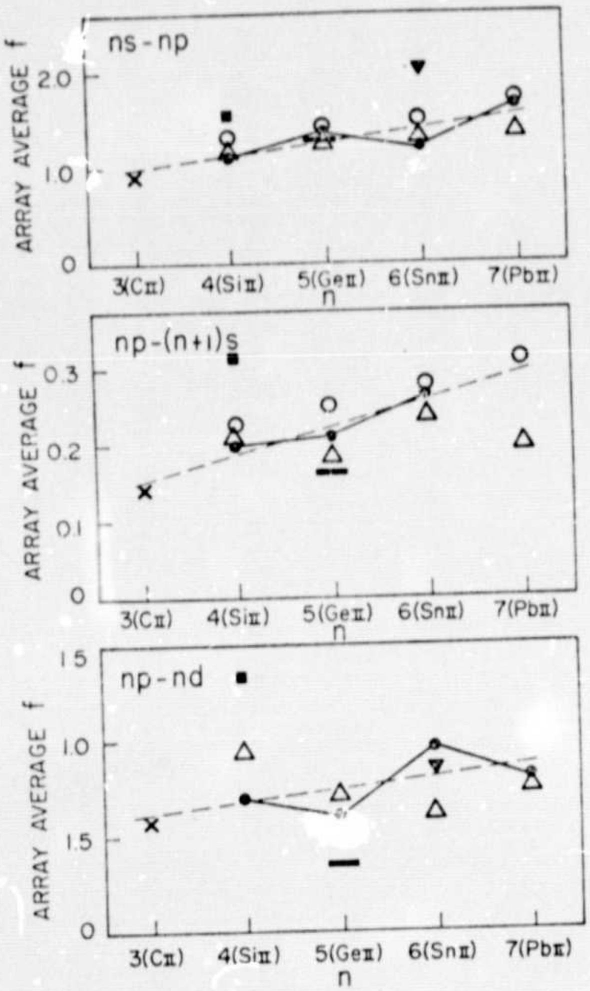
ns - np
ARRAY AVERAGE f
2.0
1.0
0
3(CII)
4(SiII)
5(GeII)
6(SnII)
7(PbII)
n
np-(n+1)s
ARRAY AVERAGE f
0.3
0.2
0.1
0
3(CII)
4(SiII)
5(GeII)
6(SnII)
7(PbII)
n
np-nd
ARRAY AVERAGE f
1.5
1.0
1.5
0
3(CII)
4(SiII)
5(GeII)
6(SnII)
7(PbII)
n

Table 1. Oscillator Strengths Compared with Sum Rules for Single Optically Active Electron.

Ion	n	$f_{exper.}$ (ns-np)	$f_{asymptotic}$	Partial f sum $\sum_m f(np-md)$	Wigner-Kirkwood Sum Rule[13]
Si II	4	0.69	0.46	1.15	1.11
Ge II	5	0.62	0.57	1.19	1.11
Sn II	6	0.98	0.39	1.37	1.11
Pb II	7	0.73	0.38	1.11	1.11

Ion	n	$\sum f_{exper.}$ (ns-np) + (np-[n+1]s)	$f_{asymptotic}$*	Partial f sum $\sum^*_m f(ms-np)$	Wigner-Kirkwood Sum Rule[13]
Si II	4	-0.37 + 0.20 = -0.17	0.00	-0.17[a]	-0.11
Ge II	5	-0.47 + 0.21 = -0.26	+0.04	-0.22	-0.11
Sn II	6	-0.43 + 0.26 = -0.17	-0.12	-0.29[a]	-0.11
Pb II	7	-0.53 + 0.31[b] = -0.21	+0.05	-0.16	-0.11

Ion	n	Total f sum for np: $\sum^*_m f(ms-np) + \sum^*_q f(np-qd)$	Experimental Fraction	Thomas-Reiche-Kuhn Sum Rule[13]
Si II	4	-0.17 + 1.15 = 0.98	69%	1.00
Ge II	5	-0.22 + 1.19 = 0.97	68%	1.00
Sn II	6	-0.29 + 1.37 = 1.08	79%	1.00
Pb II	7	-0.16 + 1.11 = 0.95	55%	1.00

*Shock tube data complemented by Coulomb approximations[4] for $n + 1 \le m < n + 2$ for d levels and $n + 2 \le m \le n + 3$ for s levels. For higher levels and continuum, scaled hydrogenic asymptotic approximations[13] are used.

[a] Includes measured two-electron transitions out of np.

[b] Relativistic calculation by Migdalek.[3]