

Radiative Lifetimes in Sc I–Sc III

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Received September 15, 1971

Abstract

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Physica Scripta (Sweden) 4, 55–59, 1971.

We have studied the spectra of scandium (600–6 000 Å) with the beam-foil method. In the v.u.v. a few previously unreported Sc III transitions appeared, as well as several well-known Sc III multiplets, whereas most lines in the air region were ascribed to earlier classified Sc I and Sc II transitions. We also measured the mean lifetimes of 20 excited Sc I–Sc III levels and generally found good agreement with previous theoretical calculations and emission measurements.

1. Introduction

Several authors have already determined radiative lifetimes in the iron group (Sc–Zn) with the beam-foil method. For a number of Fe I and Fe II levels Whaling et al. [1–4] and Andersen and Sørensen [5] obtained considerably longer lifetimes than the previously assumed values, which were based on emission measurements [6, 7]. Similar results were recently found also for Cr I [8] and Ti II [9]. The new oscillator strengths for those lines that appear in the spectra of the solar photosphere may drastically change the assumed solar and stellar abundances of these important elements. For example, the Fe I measurements discussed in ref. [2] imply a photospheric iron abundance of $\log N_{\text{Fe}}=7.5$ (on the $\log N_{\text{H}}=12.00$ scale) which is ten times higher than the earlier value of $\log N_{\text{Fe}}=6.51$ [10]. Similar conclusions can probably be drawn from the new Cr and Ti lifetimes [8, 9].

In view of these results we made a beam-foil study of scandium which is the lightest member of the iron group.

2. Experiment

We obtained beams of Sc⁺ and Sc⁺⁺ (80–170 keV) from the isotope separator. Typical intensities were 0.2–0.5 μA through the 5 mm-diam carbon foil. Because of the low particle velocities and moderate intensities we used relatively wide slits and had to be content with 15 Å linewidths in the visible region. The experimental facilities and techniques are described in ref. [11].

Most observed lines were identified with the help of Moore's *Atomic Energy Levels* [12] and the Sc references quoted there. Because of the linewidths the possibilities of blending were always considered.

For the strongest, apparently unblended lines we measured the decay times. The decay curves were analyzed in various ways, described earlier [11, 13]. Most curves showed the presence of longer-lived cascades. In order to reduce these effects and to

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eliminate the background, we differentiated the decay curves [13]. For a more accurate determination of cascade lifetimes we also made numerical integrations of the intensity function, thereby smoothing out statistical fluctuations on the tails. Fig. 1 is a good example of these methods, which were applied to all of our decay curves.

3. Results

3.1. Spectra

Figs. 2 and 3 are two examples of our spectra, taken in the v.u.v. and the visible region, respectively.

Below 2 000 Å all observed lines were ascribed to Sc III transitions (except a number of C I, C II, and H I transitions which often appear as impurities). The Sc III term scheme is based on the work of Russell and Lang [14] who observed transitions from $5s^2S$ and all the $n=4$ terms and also predicted the energies of a few additional terms. The $3d^2D-5f^2F^0$ multiplet at 627 Å was found by Beckman [15], who confirmed the predicted $5f^2F^0$ term value. Holmström [16] has recently identified many Sc III transitions above 2 000 Å.

In our beam-foil spectra the $3d^2D-4p^2P^0$ (1 603, 1 610 Å) and $4p^2P^0-4d^2D$ (1 994, 2 011 Å) transitions were fairly strong. We

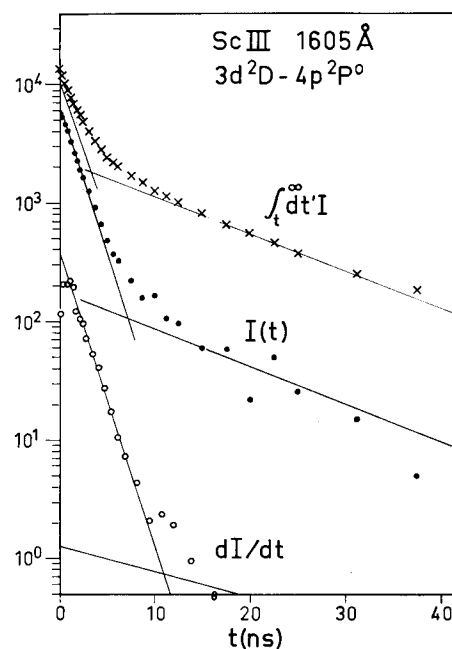


Fig. 1. Curve fits to the measured decay curve, its integral and derivative for the $4p^2P^0$ term of Sc III. All fits contain the same two components, 1.7 and 14 ns, but the admixtures vary in proportion to the relative lifetimes.

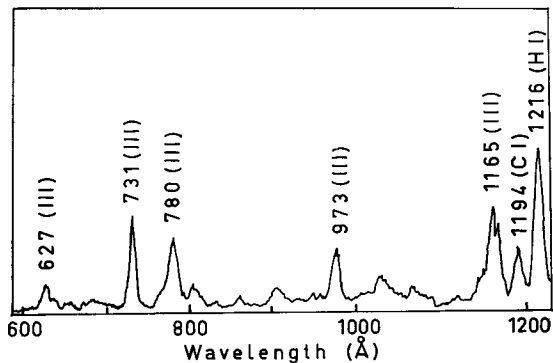


Fig. 2. Beam-foil spectrum of scandium between 600 and 1200 Å, obtained with a 1 m grating monochromator, equipped with a Bendix Channeltron. The beam was $0.4 \mu\text{A Sc}^{++}$ of 166 keV energy. The identified Sc III lines and a few impurity lines have been indicated. The Sc III transitions at 1605 and 2006 Å were about 10 times stronger (on an uncorrected scale) than the lines shown.

also found the previously observed $3d^2D-4f^2F^0$ (731 Å) and $3d^2D-5p^2P^0$ (627 Å) lines. Beckman [15] searched for the $3d^2D-5p^2P^0$ combination but it was obscured by a strong O IV line (779 Å) in her spectra. Fig. 2 shows that the Sc III $3d^2D-5p^2P^0$ (780 Å) and $4s^2S-5p^2P^0$ (973 Å) transitions were present in the beam-foil spectra. We also found the $4p^2P^0-5d^2D$ transition around 1165 Å and some indications of the $4p^2P^0-6s^2S$ multiplet (1152 Å), none of which appeared in Beckman's spectra. Above 2000 Å we observed the $4s^2S-4p^2P^0$ doublet (2699, 2735 Å) and possibly the $4d^2D-4f^2F^0$ transition at 4067 Å. Our detection efficiency was fairly low between 2100 and 2400 Å and we did not observe the $4d^2D-5f^2F^0$ transition at 2115 Å. Fig. 4 displays all the Sc III transitions seen in this experiment.

Above 2000 Å we identified most of our lines with Sc I and Sc II multiplets. The Sc I and Sc II spectra have been extensively studied by Russell and Meggers [17] and the energy level diagrams are displayed in their article and in ref. [18]. Our spectra showed that the $3d4p^2P^0$, z^1D^0 , z^1F^0 , z^3D^0 , and z^3F^0 terms of Sc II were strongly excited at the beam-foil interaction, whereas transitions from z^3P^0 were somewhat less pronounced. Among the even terms $3d4de^3F$ and e^3G were sufficiently populated to permit decay measurements. Transitions from several other Sc II

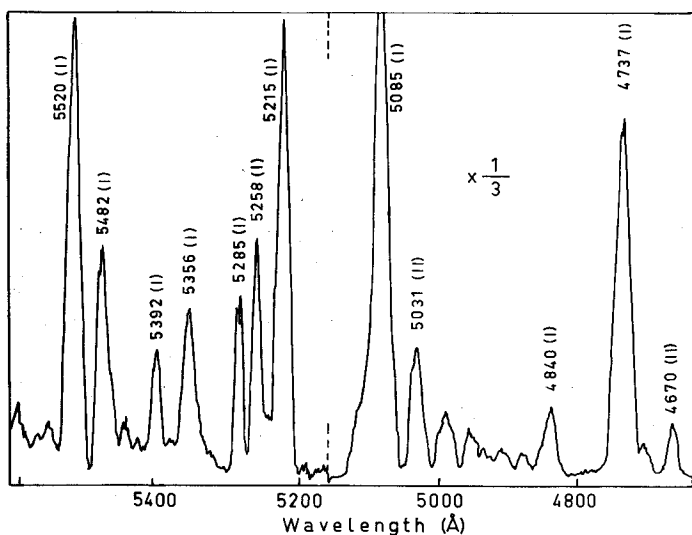


Fig. 3. Beam-foil spectrum of scandium between 4600 and 5600 Å, observed with a 0.25 m Jarrell-Ash monochromator and an EMI 6256 photomultiplier. The energy was 83 keV Sc^+ (0.2 μA). The strongest Sc I and Sc II transitions have been indicated. Note that the gain is reduced between 4600 and 5200 Å.

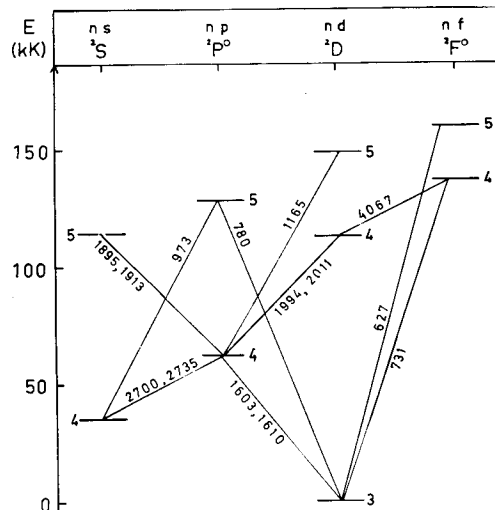


Fig. 4. Partial energy level diagram for Sc III, showing all the transitions observed in this experiment.

terms were also present in the spectra, but of considerably lower intensity. However, we found a multiplet with the strongest component at 2065 Å, which we classify as the $3d4p^2D^0-4p^2f^3P$ transition. Russell and Meggers [17] observed the $4s4p^2P^0-4p^2f^3P$ combination (2667–2701 Å) but were not able to find other transitions from $4p^2f^3P$, which are below 2500 Å. Also several Sc I levels below $40\,000 \text{ cm}^{-1}$ were strongly excited, in particular $3d4s4p^2D^0$ and y^2F^0 , $3d^24p^2G^0$, z^2G^0 , y^4D^0 , and y^4F^0 .

A large fraction of all the multiplets listed by Russell and Meggers [17] appeared in our spectra and, inversely, only a few of our weak lines remain unidentified. An observed line at $3715 \pm 3 \text{ Å}$ coincides with an unidentified line at 3717.10 Å, listed by Meggers et al. [19]. We also saw a few weak, unknown lines below 2000 Å, one of which $876 \pm 2 \text{ Å}$ is close in wavelength to some of Beckman's [15] unclassified lines.

3.2. Lifetimes

The results of our decay measurements are given in Table I, together with previous experimental and theoretical lifetimes. In parentheses after our beam-foil lifetimes we include the cascade decay times and the replenishment ratio, which is a measure of the amount of cascading [20]. In a number of cases we have measured the decays of two components of the same multiplet, and the lifetimes of a few Sc II and Sc III terms have been determined from two branches. The results agree within the estimated uncertainties.

Sc III. Previous information about Sc III lifetimes is limited to the work of Weiss [21] who used Hartree-Fock wavefunctions for calculations of the $4s^2S-4p^2P^0$ and $3d^2D-4p^2P^0$ multiplet strengths. However, additional information exists for Ca II and K I, isoelectronic to Sc III.

Our measurements on the 1605 Å multiplet ($3d^2D-4p^2P^0$) gave a lifetime of $1.7 \pm 0.2 \text{ ns}$ for the $4p^2P^0$ term, while the lines at 2699 and 2735 Å ($4s^2S-4p^2P^0$) had the decay times $2.0 \pm 0.3 \text{ ns}$ and $2.3 \pm 0.4 \text{ ns}$ respectively. The error bars overlap but we believe that 1.7 ns is the best estimate of the $4p^2P^0$ lifetime. The v.u.v. transitions were more intense in our spectra and, furthermore, the region around 2700 Å might be blended with Sc I lines. Weiss's [21] theoretical values are 1.27 ns (dipole length approximation) and 1.66 ns (dipole velocity approximation). This work agrees very well with the velocity form. It is interesting to note the similarity with Ca II, where the calculated $4p^2P^0$

Table I. Radiative lifetimes in Sc I–Sc III

Spectrum	Wave-length (Å)	Transition	Lifetime of upper level (ns)			
			This work ^a	Other experiments	Theory	
Sc I	3 912	$3d4s^2a^2D - 3d4s4py^2F^0$	7.8 ± 0.8 (31; 0.19)	4.7^b	3.4^c	
	4 021	$3d4s^2a^2D - 3d4s4py^2D^0$	8.0 ± 0.8 (30; 0.11)	4.1^b	4.4^c	
	5 007	$3d^24sb^2D - 3d^24pw^2D^0$	6.1 ± 0.6 (23; 0.07)	5.3^b		
	5 215	$3d^24sa^2G - 3d^24py^2G^0$	14.2 ± 1.5	4.9^b		
	5 519	$3d^24sa^2F - 3d^24pz^2G^0$	11.7 ± 1 (48; 0.13)	14^b	$15^c; 14^d$	
	4 737	$3d^24sa^4F - 3d^24py^4D^0$	8.5 ± 0.8 (30; 0.12)	8.3^b		
	5 085	$3d^24sa^4F - 3d^24py^4F^0$	12.3 ± 1	7.5^b	$10^{c,d}$	
Sc II	3 354	$3d4sa^1D - 3d4pz^1F^0$	6.5 ± 0.6 (39; 0.03)	$4.2^b; 3.4^e; 9.4^f$	4.1^g	
	4 670	$3d^2b^1D - 3d4pz^1F^0$	7.2 ± 0.7 (41; 0.05)			
	4 247	$3d4sa^1D - 3d4pz^1D^0$	7.8 ± 0.8 (25; 0.05)	$10^b; 8.7^e; 17^f$	9.1^g	
	5 031	$3d^2b^1D - 3d4pz^1P^0$	5.5 ± 0.5 (17; 0.22)			
	2 065	$3d4pz^3D^0 - 4p^2f^3P$	1.6 ± 0.3 (9; 0.25)	$3.8^b; 2.0^e$	3.4^g	
	2 824	$3d4pz^3D^0 - 3d4de^3F$	5.4 ± 0.5 (13; 0.09)			
	3 057	$3d4pz^3F^0 - 3d4de^3G$	3.4 ± 0.3 (27; 0.12)	2.3^b	2.6^g	
	3 575	$3d4sa^3D - 3d4pz^3D^0$	5.6 ± 0.6 (37; 0.04)	$5.4^b; 5.0^e; 2.8^f$	5.3^g	
	4 318	$3d^2a^3F - 3d4pz^3D^0$	6.5 ± 0.6 (47; 0.05)			
	3 622	$3d4sa^3D - 3d4pz^3F^0$	5.1 ± 0.5 (17; 0.12)	$6.9^b; 5.8^e; 1.6^f$	6.1^g	
	4 390	$3d^2a^3F - 3d4pz^3F^0$	6.0 ± 0.6 (17; 0.12)			
	Sc III	627	$3d^2D - 5f^2F^0$	2.7 ± 0.7 (12; 0.25)		
731		$3d^2D - 4f^2F^0$	3.5 ± 0.8 (30; 0.18) ^h			
4 067		$4d^2D - 4f^2F^0$	4.2 ± 0.8 (23; 0.30) ^h			
780		$3d^2D - 5p^2P^0$	3.6 ± 0.4 (11; 0.07)			
973		$4s^2S - 5p^2P^0$	3.3 ± 0.8 (11; 0.26)			
1 165		$4p^2P^0 - 5d^2D$	2.4 ± 0.3 (14; 0.13)			
1 605		$3d^2D - 4p^2P^0$	1.7 ± 0.2 (14; 0.04)	1.27^i		
2 711		$4s^2S - 4p^2P^0$	2.0 ± 0.3 (12; 0.12)			
1 907		$4p^2P^0 - 5s^2S$	1.4 ± 0.2 (10; 0.09)	1.66^k		
1 995		$4p^2P^0 - 4d^2D$	1.2 ± 0.2 (5.5; 0.14)			
2 010			1.2 ± 0.2 (6.8; 0.15)			

^a When cascading occurs we give the cascade lifetime (in ns) and replenishment ratio in parenthesis after the main lifetime.

^b Corliss and Bozman [6] emission measurement.

^c Goldberg, Müller and Aller [28] Coulomb approximation.

^d Goldberg, Müller and Aller [28] f-sum rule.

^e Warner [25] emission measurement.

^f Aller [27] astrophysical f-value.

^g Warner [25] Coulomb approximation.

^h See discussion in the text.

ⁱ Weiss [21] SCF calculation, dipole length approximation.

^k Weiss [21] SCF calculation, dipole velocity approximation.

lifetimes are 5.52 ns (length) and 7.25 ns (velocity), respectively [21], while the beam-foil measurement of Andersen et al. [22] yielded 7.5 ± 0.5 ns. A detailed discussion about these two theoretical approaches has been given by Crossley [23] who recommends the length approximation for low and the velocity form for high transition energies.

Our $5s^2S$ lifetime, 1.4 ± 0.2 ns, corresponds to $f=0.13 \pm 0.02$ for the $4p^2P^0-5s^2S$ transition. For Ca II the experimental value 0.16 ± 0.02 agrees with the theoretical estimate, 0.173 [24]. In this sequence the f -values are expected to decrease with increasing Z towards the hydrogenic value, 0.0529 [24]. Our $4d^2D$ lifetime implies a Sc III $4p^2P^0-4d^2D$ oscillator strength of 0.84 ± 0.14 , which is comparable to the Ca II experimental value 0.91 ± 0.06 [22]. The beam-foil lifetime for the $5d^2D$ term, 2.4 ± 0.3 ns, can only give an upper limit of 0.14 for the $4p^2P^0-5d^2D$ oscillator strength, because the upper level can also decay to $5p^2P^0$ and $4f^2F^0$. An interesting discrepancy exists for this line in Ca II, where theory recommends the value 0.122 [24] and the experimental result is 0.23 ± 0.04 [22].

The experimental $4f^2F^0$ decay time (Table I) would correspond to an upper limit of 0.03 for the $3d^2D-4f^2F^0$ oscillator strength, which is substantially lower than the theoretical values in K I and Ca II [24]. It is probable, however, that the Sc III $4f^2F^0$ term decays faster than observed by us and that our mean value, 3.8 ns, only represents the lifetime of a cascading level, most

likely $5g^2G$. This is further supported by the fact that we got a shorter decay time for $5f^2F^0$ than for $4f^2F^0$, while the lifetimes in a spectral series should increase with n in these simple spectra. It would have been very difficult to determine decay times shorter than 1 ns in the present experiment. Measurements at higher velocities are therefore needed to fully clarify this point.

Sc II. Detailed information about Sc II lifetimes and oscillator strengths is already available in the literature. Corliss and Bozman [6] derived many Sc II transition probabilities from emission line intensities. Warner [25] made extensive remeasurements of the Sc II f -values, using a high voltage plasma light source, and he also calculated the line strengths with the Coulomb approximation [26]. Astrophysical f -values, extracted from the curve of growth for Sirius, have been quoted by Aller [27]. Our results will be compared to those given in refs. [6, 25, 27].

We measured the decay times of all three singlet $3d4p$ levels. The decay curve for $3d4pz^1P^0$ (5 031 Å) showed pronounced cascade effects, but the fastest component, 5.5 ns, appeared reproducibly. Table I shows that this value lies between Warner's [25] measured and calculated lifetimes. Aller's astrophysical value is really an upper limit, since only two of three branches (to $4s^2a^1S$ and $3d4sa^1D$) were included in ref. [27]. Our decay curves for the $3d4pz^1D^0$ term (4 247 Å) indicated but slight cascading, and the beam-foil lifetime, 7.8 ± 0.8 ns is in good accord with refs. [6] and [25]. Our spectra also showed the intercombina-

tion line $3d4sa^3D-3d4pz^1D^0$ around 3 840 Å, for which we obtained a decay time of 4.7 ns, however. This is difficult to reconcile with the 4 247 Å measurement, unless some unknown blend is present. For intensity reasons we are convinced that the value 7.8 ns is preferable. As a contrast, the decay curves of the 4 670 and 3 354 Å lines, both of which arise from the $3d4pz^1F^0$ term, had nearly identical shapes and yielded a mean lifetime of 6.8 ± 0.6 ns. This value is 50% longer than those of refs. [6] and [25] but it is still shorter than the astrophysical value [27].

We also measured four lifetimes in the triplet system. The $3d4pz^3D^0$ and z^3F^0 lifetimes were both determined from two decay modes with consistent results. The two mean values are in excellent accord with the work of Corliss and Bozman [6] and Warner [25], while the astrophysical lifetimes [27] tend to be too short here. For the $3d4de^3F$ and e^3G terms the beam-foil lifetimes are approximately 50% longer than the values suggested in refs. [6] and [25] (see Table I). We might question if the agreement between beam-foil results and emission data worsens with increasing excitation energy, but there is not yet sufficient evidence for conclusions. However, for Fe I, Whaling et al. [2] found very pronounced effects in this direction. The lifetime of the highest known Sc II term, $4p^2f^3P$ was not previously known, but Corliss and Bozman [6] found that the $4s4py^3P^0-4p^2f^3P$ branch had a probability of 1.8×10^8 s⁻¹, whereas the sum of transition probabilities from $4p^2f^3P$ is 6.3×10^8 s⁻¹, according to this work.

It should be clear from this discussion and Table I that, except for a few minor discrepancies, the agreement between this work and previous f -value determinations for Sc II is very good. Warner [10] points out that the theoretical f -values for Sc II should be very accurate, because of the relative simplicity of the spectrum. This seems indeed to be the case.

Sc I. We determined the lifetimes of five doublet and two quartet terms of Sc I (Table I). Comparison is possible with the emission measurements of Corliss and Bozman [6] and the calculations of Goldberg et al. [28], who applied the Coulomb approximation and the f -sum rules. Reference [28] also quotes a few additional Sc I f -value determinations, which will not be considered here, because they are in disaccord with the remaining data.

The transitions from $3d4s4py^2D^0$ (3 996–4 047 Å) and $3d4s4py^2F^0$ (3 907–3 933 Å) were prominent in our spectra and the decay curves were reproducible. The results 8.0 ± 0.8 ns (y^2D^0) and 7.8 ± 0.8 ns (y^2F^0) should therefore be quite reliable. Table I shows that both these values differ by approximately a factor of two from the lifetimes obtained in refs. [6] and [28]. Besides combining with the ground state, the y^2D^0 and y^2F^0 terms can also decay to higher levels, for example $3d^24sb^2D$ and a^2F , but these transitions have not been observed. The $3d^24pw^2D^0$ lifetime was determined from transitions to $3d^24sb^2D$ (4 991–5 021 Å) and here we note fairly good agreement between this work and the Corliss and Bozman value, 5.3 ns. The latter accounts only for the strongest lines in the multiplets from w^2D^0 and is therefore an upper limit, however. As Table I shows, our $3d^24pz^2G^0$ lifetime agrees fairly well with previous results, whereas a substantial discrepancy is present for the higher-lying $3d^24py^2G^0$ term, because our experiment implies a three times longer lifetime. No calculated value exists for the y^2G^0 term.

Our lifetime for the $3d^24py^4D^0$ agrees quite well with the Corliss and Bozman value [6]. However, the latter only accounts for transitions to $3d^24sa^4F$. Russell and Meggers [17] observed weak intercombination lines to the $3d4s^2a^2D$ ground state (3 061–3 073), and there should also be allowed transitions from y^4D^0

to $3d^24sa^4P$ but the latter (in the 6 800 Å range) have not been observed. The beam-foil result for the $3d^24py^4F^0$ level is consistent with previous work, in particular with the value calculated by Goldberg et al. [28].

In summary, although the agreement between this and previous results is here poorer than in the Sc II case, no drastic differences can be noted.

4. Conclusion

The present beam-foil experiment has shown that the previously determined Sc I and Sc II lifetimes are quite reliable. The solar and stellar abundances of scandium, based on observations of the appropriate Sc I and Sc II multiplets, should therefore be fairly accurate. In 1948 Unsöld [29] obtained a solar scandium abundance of $\log N_{\text{Sc}}=3.33$, which was based on calculations of Sc I transition probabilities with the f -sum rule. This value was reduced by a factor of 3 by Goldberg et al. [28] who found a photospheric abundance of $\log N_{\text{Sc}}=2.82$. As mentioned above they calculated the strengths of 23 Sc I lines with the Coulomb approximation and the f -sum rule. Essentially the same value, $\log N_{\text{Sc}}=2.80$ was suggested by Aller [30], who had access to the Corliss and Bozman Sc I oscillator strengths. Warner [10] used 45 Sc II lines for his analysis of the photospheric Sc abundance. The appropriate f -values originated from ref. [25]. From a very smooth curve of growth the abundance $\log N_{\text{Sc}}=3.04$ was derived. The abundance based on Sc II lines is thus approximately 60% higher than the two preceding values, based on Sc I lines. As already mentioned, the present work tends to confirm the f -values of astrophysically important Sc II transitions, whereas it indicates, that some of the earlier used Sc I f -values are too high. A decrease in the Sc I line strengths would correspondingly increase the estimated photospheric Sc I abundance. Our experiment seems therefore to support the abundance found by Warner [10]. It is interesting to note that the chromospheric Sc abundance may be ten times higher. Pecker and Pottasch [31] observed four Sc II multiplets in the spectra of the chromosphere and obtained a value of $\log N_{\text{Sc}}=4.08$. They used Warner's f -values [25] for these Sc II transitions ($a^2D-z^3F^0$, $a^2D-z^3D^0$, $a^3F-z^3D^0$, $a^1D-z^1D^0$) but also questioned if this f -value scale might be too high. As Table I shows, there is very good agreement between our work and ref. [10] for these transitions, however.

Finally it may be interesting to note that several of the Sc I and Sc II multiplets discussed here also have been observed in stellar spectra. Merrill's monograph [32] gives many references.

Acknowledgements

We are grateful to Professor M. Siegbahn for his support of this work. We are also very much indebted to Professor B. Edlén and Dr J. O. Ekberg for valuable advice and to Dr J.-O. Holmström for sending us his Sc III results prior to publication.

This work was supported by the Swedish Natural Science Research Council (NFR).

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