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Lifetime Measurements in Mn I and Mn II

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Abstract

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Radiative lifetimes in neutral and singly-ionized manganese have been measured from beam-foil spectra in the region 2 000–5 000 Å, using 249 keV Mn^{+++} ions from an electromagnetic isotope separator. Several of the transitions have been used in the determination of the Mn abundance in the solar photosphere. Our measured lifetimes for Mn I levels are 2–6 times longer than those inferred by the *gf*-values of Corliss and Bozman. A markedly better agreement exists between beam-foil data and Warner's results for Mn II terms, but even here the beam-foil lifetimes appear to be significantly longer. Our results are consistent with a recently suggested upward revision of the photospheric Mn abundance from log $N_{Mn} = 4.8 - 4.9$ to 5.4.

1. Introduction

The oscillator strengths in the first and second spectra of the transition metals are essential to the determination of the solar and stellar abundances of these important elements which play a sensitive role in models of stellar evolution [1–4].

The complexity of the spectra of these elements has hitherto generally precluded accurate configuration-interaction calculations of *f*-values. Most of the available information is instead based on experimental studies, e.g. measurements of spectral line intensities in emission. Recently important results have also been obtained from lifetime measurements with the beam-foil technique. Such beam-foil experiments and their astrophysical consequences have been expertly reviewed by Smith [5]. An interesting critical evaluation of various methods for lifetime and *f*-value determination in transition metals can be found in a recent article by Blackwell and Collins [6].

The present paper, which discusses beam-foil studies of term lifetimes in Mn I and Mn II, represents a continuation of our beam-foil program for the iron-group elements, initiated in 1971 [7].

2. Summary of previous work

A study of Ref. [8] shows that many investigators have determined Mn I and Mn II *f*-values. For the Mn I resonance multiplet absolute *f*-values were determined already in the 1950's, e.g. by Huldt and Lagerqvist [9] and Bell et al. [10]. Relative oscillator strengths were measured by Allen and Asaad [11] who further normalized their scale using the absolute values given in Ref. [9]. Later Allen [12] also published theoretical *f*-values, based on the *f*-sum rule and the Coulomb approximation. Perhaps the most extensive material, from emission measurements, can be found in the monograph of Corliss and Bozman [13]. However, since these *f*-values [13] were typically 2–3 times higher than the results of previous work, attempts have been made to adjust the Mn I *f*-values to a common scale [14, 15]. More recently Woodgate [16] and Blackwell and Collins [6] determined relative *f*-values for many Mn I lines and normalized the results to Penkin's [17] absolute transition probabilities for the Mn I resonance lines, a well motivated decision in view of the good agreement between Refs. [9] and [17]. As will be discussed further below the thus obtained absolute *f*-values of (particularly) Ref. [16] are markedly lower than those of Refs. [11–13].

Corliss and Bozman [13] also measured many Mn II *f*-values. Here corrections to their scale have been suggested by Warner [18] who also made extensive emission measurements as well as Coulomb-approximation calculations. Warner's corrections imply a reduction of the Corliss and Bozman (CB) line strengths by approximately a factor of 5.

In this context it might be worth adding that the theoretical methods, e.g. the Coulomb approximation, which so far have been applied to the transition metals, have severe limitations in these complex spectra. An interesting comparison is possible for the $4p\ z\ ^7P^0$ term lifetime in Cr I, isoelectronic to Mn II. Here accurate level-crossing [19] and beam-foil [20] measurements have yielded the values 33 and 35 ns, respectively (with 10% uncertainties) whereas Coulomb-approximation calculations result in a markedly different value, 10 ns.

3. Experiment

We accelerated Mn ions in an 83 kV isotope separator. Triply charged Mn^{+++} was used for most measurements, in order to expand the time scale after the foil, reduce the fractional energy loss in the foil and to minimize the downstream divergence of the excited particles. Typical currents of Mn^{+++} were 0.2–0.5 μA and under this bombardment the nominally 4–5 μg/cm² foils usually lasted for 10–15 minutes. Spectra between 2 000 and 5 000 Å were taken with a Heath EUE-700 35 cm monochromator, equipped with an EMI 6256 SA photomultiplier. The photon counts were recorded with an Elscint INS-11 single channel analyzer.

We estimated the doppler broadening—due to the finite acceptance angle of the monochromator and the beam divergence after the foil—to produce linewidths of 2–5 Å over our wavelength range, while the contribution from the instrumental resolution was typically 2 Å (100 μ slits).

The spectral lines were identified using Refs. [21–23] and the original literature quoted therein. We also compared our beam-foil data with sputtering spectra. The latter were obtained by bombarding metallic Mn targets with 83 keV Ar^+ ions (typical

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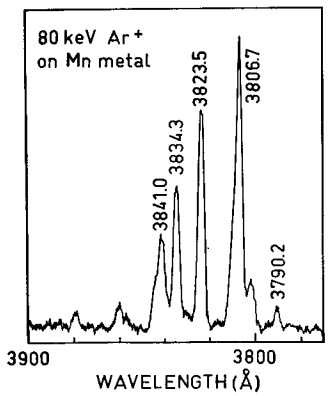

Fig. 1. Part of a sputtering spectrum of Mn, obtained by bombarding a target of solid Mn with 80 keV, 20 μA Ar^+. The lines indicated by wavelength belong to the Mn I a^6D–z^6F^0 multiplet.

beam currents were 20 μA). These spectra consisted mostly of Mn I transitions, and because of the high intensities, work at higher resolution than in the beam-foil case was possible. Fig. 1 displays a partial sputtering spectrum around 3 800 Å showing the fine structure of the Mn I a^6D–z^6F^0 multiplet (we follow the spectroscopic notation of Ref. [23]).

The lifetimes were measured—as usual—by recording the spectral line intensities as a function of the distance from the foil. The beam intensity varied typically by less than 5%. The counting rates at various points were normalized to a preset amount of integrated beam current collected in a Faraday cup. The beam current was kept reasonably low during decay-time measurements, in order to minimize possible changes in the light output, caused by foil damage.

Our experimental time resolution, mostly due to the finite length of the viewed beam segment, was both measured and estimated to be 0.6 ns for 249 keV incoming ions. Thus we expected our detection system to give reliable lifetimes as short as a few ns and as long as about 40 ns, where Rutherford scattering out of the viewing volume becomes troublesome.

After background subtraction most of our curves appeared as single exponentials, but in a few cases corrections for cascading were necessary. The beam velocity after the foil was directly measured, using an 18 cm 90° electrostatic analyzer. We found that 249 keV Mn^{+++} ions typically lost 60 ± 15 keV in our 5 μg/cm² foils, the surface density of which was also determined by measuring the energy loss for protons and comparing the data to the tables of Northcliffe and Schilling [24]. With the foil thickness fairly accurately known we also compared our measured energy loss for Mn ions with the formulae of Lindhard et al. [25], which in our case predict a 12 keV electronic and 22 keV nuclear energy loss. The total theoretical energy loss of 34 keV is somewhat lower than our above-mentioned experimental value, and the discrepancy widens if we adopt the arguments of Hvelplund et al. [26] stating that only a fraction of the nuclear stopping, tabulated by Lindhard et al. [25], should be considered for those ions which after the foil continue in the forward direction. In our decay-curve analysis we have still decided in favour of our directly measured velocity for the foil-emergent Mn ions, and in most cases we have used a velocity of 0.81 ± 0.04 mm/ns ($\beta = 1/370$). At our energy the discrepancy between the measured and calculated energy loss corresponds to a 6% difference in ion velocity, so the use of the calculated loss would have yielded lifetimes 6% shorter than those reported below.

Estimates of the scattering and beam divergence, using the results in Refs. [27] and [28], yielded in our case (249 keV Mn^{+++}, 5 μg/cm² carbon foil) a RMS scattering angle of 7.7°. For longer decay times this scattering will clearly lead to an underestimation of the correct lifetimes and cascade contributions. (Note however, that here we have systematic errors which may partly neutralize each other, since the Rutherford scattering results in underestimated decay times whereas the neglect of cascade contributions usually leads to overestimated ones.)

To determine the extent to which the beam divergence may distort our decay curves, we first made numerical estimates, which showed that no scattering corrections were necessary for downstream distances corresponding to less than 45 ns after the excitation. In most cases, especially for Mn II transitions, the decay curves were in regions of poor signal-to-noise before corrections became significant. For a few Mn I decay curves corrections were still made, however. To check the accuracy of this procedure we directed 166 keV Mn^{++} through a 15 μg/cm² foil, thereby increasing the predicted RMS scattering angle to 16.5°. The decay curves obtained in this case yielded after our numerical corrections the same decay times as the 249 keV, 5 μg/cm² data.

We also accelerated V^{++} ions to 166 keV and measured a few V I and V II lifetimes which had previously been studied by Andersen et al. [29]. For the levels z^3G^0 in V II (3 276 Å) and y^6D^0 in V I (4 112 Å) we obtained the decay times 6.7 and 9.0 ns, respectively, in satisfactory agreement with the corresponding values of 7.3 and 9.2 ns in Ref. [29]. As a final check we also measured a few Cr I lifetimes and noticed good accord with previous work [20].

4. Results

All prominent lines in the beam-foil spectra were identified as tabulated Mn I and Mn II transitions, with the exception of one unidentified line at $4\,000 \pm 2$ Å. Nearly all of the more intense lines listed by Meggers et al. [21] appeared in our spectra. At 249 keV ion energy many Mn II lines were quite strong, decreasing in favour of Mn I transitions at lower energies. Fig. 2, which shows a very line-rich part of a spectrum in the UV, illustrates

Table I. *Radiative lifetimes in Mn II*

Wavelength[a] (Å)	Transition[b]	Mean lifetime of upper term (ns)		
		This work	Other experiments	Theory
2 563	$a^5G - z^5F^0$	4.1 ± 0.4	< 2.2[c]	
2 576	$a^7S_3 - z^7P^0_4$	4.7 ± 0.5		
2 593	$- z^7P^0_3$	4.4 ± 0.4	1.3[c]; 7.3[d]	3.3[e]
2 605	$- z^7P^0_2$	4.4 ± 0.4		
2 625	$a^5G - z^5H^0$	4.4 ± 0.4		
2 680	$a^3G - z^3G^0$	4.6 ± 0.5		
2 708	$a^5G - z^5G^0$	5.3 ± 0.5	1.0[c]; 3.0[d]	3.27[e]
2 879	$a^3G - z^3F^0$	5.0 ± 0.5		
2 886	$a^3G - z^3H^0$	6.5 ± 0.6		
2 933	$a^5S_2 - z^5P^0_1$	(6.8 ± 1.5)[f]		
2 939	$- z^5P^0_2$	5.3 ± 0.5	0.56[c]; 3.1[d]; 3.0[g]	4.2[e]
2 949	$- z^5P^0_3$	5.4 ± 0.5		
3 029	$z^5P^0 - e^5S$	7.0 ± 0.7	1.6[g]	3.3[e]
3 441	$a^5D_4 - z^5P^0_3$	5.3 ± 0.5	0.56[c]; 3.1[d]; 3.0[g]	4.2[e]

[a] The wavelengths given refer to the particular line (within the multiplet) for which we measured the decay constant. In a few cases we did not perform measurements on the most intense member of the given multiplet, in order to avoid blends from other multiplets.
[b] We use the notation of Ref. [23]. Whenever the term lifetime was determined from several lines of the multiplet we also give the J quantum numbers.
[c] Corliss and Bozman [13] emission measurement.
[d] Warner [18] renormalization of the Corliss and Bozman scale.
[e] Warner [18] Coulomb approximation calculation.
[f] Uncertainty introduced mainly by cascades and blends.
[g] Warner [18] emission measurement.

Table II. *Radiative lifetimes in Mn I*

Wavelength[a] (Å)	Transition[b]	Mean lifetime of upper term (ns) This work	Other experiments	Theory
3 806	$a^6D_{9/2}$–$z\,^6F^0_{11/2}$	17±4	8.7[c]; 3.3[d]; 4.3[e]; 18[f]	9.3[g]; 8.1[h];
3 823	$a^6D_{7/2}$–$z\,^6F^0_{9/2}$	18±4		6.9[i]
4 049	$a^6D_{5/2}$–$z\,^6D^0_{3/2}$	11.6±1.5	8.9[c]; 2.7[d]; 4.5[e]; 12[f];	11[g]; 9.8[h]
4 083	$a^6D_{5/2}$–$z\,^6D^0_{7/2}$	10.4±1.5	8.2[k]	
4 456	z^6P^0 –e^6D	11±2	5.2[d]; 20.5[f]	
4 626	a^2H –z^2I^0	16±3	10[d]; 28[f]	
4 764	a^4F –z^4F^0	15±3	17[c]; 6.0[d]; 8.1[e]; 20[f]	14[g]

[a, b] See Table I.
[c] Allen and Asaad [11], emission measurement.
[d] Corliss and Bozman [13] emission measurement.
[e] Hefterlin and Gearhart [15] critical compilation and emission measurement.
[f] Woodgate [16] emission measurement.
[g] Allen [12] calculation based on the *f*-sum rule.
[h] Allen [12] Coulomb approximation calculation.
[i] Stewart and Rotenberg [30] Thomas-Fermi calculation.
[k] Blackwell and Collins [6] absorption measurement.

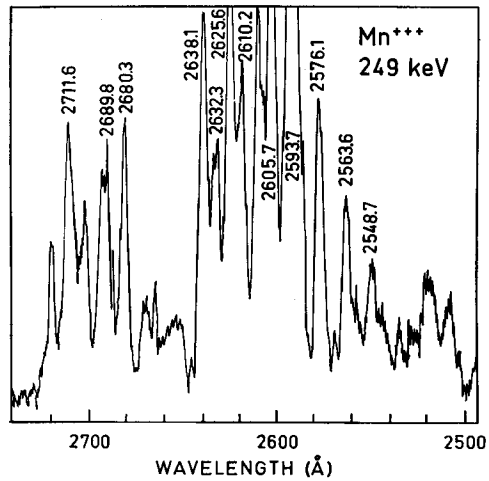

Fig. 2. Beam-foil spectrum of Mn, 2 500–2 740 Å, obtained by directing a beam of Mn^{+++}, 249 keV, 0.2 μA through a carbon foil. The indicated lines belong to Mn II multiplets.

clearly the possibilities of blending of various Mn multiplets at our available resolution. In view of this blending we abstained from decay measurements for several relatively strong spectral lines. In a few cases it was possible to measure the term lifetimes from weaker components of the corresponding multiplets.

The results of our lifetime measurements are listed in Table I (Mn II) and Table II (Mn I). These tables also include previously measured or calculated values. Most previous authors have presented their results in the form of *gf*-values for individual lines or multiplet oscillator strengths, $g_t f$, i.e. the sum of all *gf*-values within the multiplet. To facilitate comparison with our measured lifetime data, we have chosen to convert oscillator strengths into lifetimes according to the relation

$$\tau^{-1} = \frac{6.67 \cdot 10^{15}}{(\bar{\lambda})^2} \frac{g_t f}{g_A} \qquad (1)$$

where $g_A = (2S+1)(2L+1)$ is the statistical weight of the upper term and $\bar{\lambda}$ the weighted mean wavelength (in Å) of the multiplet. (Whenever the excited term decays into more than one final term, we have of course first added the individual decay probabilities before computing the term lifetime.) Since there may be deviations from pure *LS* coupling, as well as appreciable term splitting, the various *J*-states of the excited term need not have identical radiative lifetimes. However, a check of previous work shows that the differences are seldom larger than 15–20%.

Mn II

Radiative lifetimes of one septet, five quintet and three triplet terms in Mn II were measured. In two cases the multiplets were resolved and all components measured, and in one case the decay curve was followed in two separate branches.

We separately measured the three components of the Mn II resonance transition a^7S–z^7P^0, obtaining an average z^7P^0 lifetime of 4.5±0.4 ns. The individual decay curves are shown in Fig. 3, and the prominence of the corresponding lines (2 576, 2 593, and 2 605 Å) in beam-foil spectra can be seen in Fig. 2. The principal cascade feeding of z^7P^0 is expected from e^7D (2 428, 2 438, and 2 452 Å) and e^7S (2 762 and 2 796 Å). However, these lines were not significant in the spectra, and correspondingly, the z^7P^0 decay curves exhibited only one exponential over more than 30 ns. Our z^7P^0 lifetime is 3.5 times longer than the original Corliss and Bozman value [13] but markedly shorter than Warner's [18] renormalized CB value. The agreement is best, although not entirely satisfying with Warner's calculated oscillator strength. In Section 2 we have already compared theory and experiment in the analogous Cr I case. We can further note that the beam-foil experiment of Cocke et al. [20] yielded a Cr I z^7P^0 lifetime that was *shorter* than the CB value—the ratio of beam-foil to CB lifetimes is 0.78, when a misprint in Ref. [20] is corrected.

We have also measured both the allowed decay branches from the z^5P^0 term including all three components of the a^5S–z^5P^0 multiplet (2 933, 2 939, and 2 949 Å), as well as the strongest line of the a^5D–z^5P^0 multiplet (3 442 Å). All decay curves, except that of the 2 933 Å line yielded results close to 5.3±0.5 ns, which is 10 times as long as the value obtained by summing the CB probabilities. Also here, considerably better agreement with Warner's results can be noted. Although our decay curves showed single-exponential character—within the uncertainties—we searched the spectra for principal cascades. Unfortunately, the z^5P^0–e^5D transitions (2 578–2 590 Å) were completely masked by the much stronger a^7S–z^7P^0 transitions. However, the z^5P^0–e^5S lines (3 029, 3 039, and 3 056 Å) appeared in the spectra and decay measurements were also possible. Comparisons of the relative

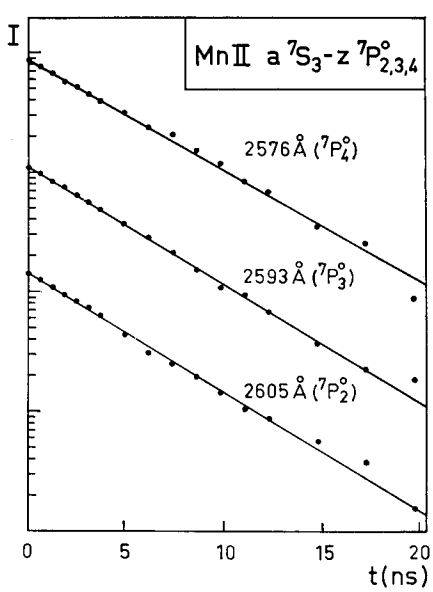

Fig. 3. Decay curves for the Mn II a^7S–z^7P^0 multiplet. All three *J*-states of the z^7P^0 term decay with very similar lifetimes, the average value being 4.5 ns.

intensities of the z^5P^0–e^5S and a^5S–z^5P^0 multiplets further enabled us to study the J-dependence of cascade effects. The cascade transitions from e^5S_2 to the levels $z^5P_2^0$ and $z^5P_3^0$ appeared namely less intense in our spectra than the decays of these $^5P^0$ states to a^5S_2, whereas the $z^5P_1^0$–e^5S_2 line (3 029 Å) was found to be stronger than the a^5S_2–$z^5P_1^0$ branch (2 933 Å). Thus the $z^5P_1^0$ state has the strongest cascade influence, which probably explains the inconsistency in the 2 933 Å decay time (Table I).

The transition probability for the z^5P^0–e^5S multiplet was not reported by Corliss and Bozman. However, our measured lifetime is markedly longer than Warner's [18] measured and calculated values. In the quintet system we also determined the z^5F^0, z^5G^0, and z^5H^0 lifetimes. Our z^5F^0 lifetime exceeds the CB value by at least a factor of 2. The discrepancy may be larger because there are no measured f-values for the a^5D–z^5F^0 multiplet around 1 915 Å. The situation for the z^5G^0 term also follows the "usual" pattern, the beam-foil lifetime being considerably longer than previous values. The z^5G^0 level may also decay to a^5F but no probabilities have been measured or calculated for this particular branch. No previous information can be found for the z^5H^0 lifetime.

Our results for the triplet terms, z^3F^0, z^3G^0, and z^3H^0, can only partially be compared with Refs. [13] and [18], since the latter have not included all allowed decay channels of these terms. For example, the z^3F^0 term combines both with a^3F and a^3G, of which only the a^3G–z^3F^0 branch (2 879–2 898 Å) has been studied in Refs. [13] and [18]. Both the z^3G^0 and z^3H^0 terms de-excite to the a^3G, b^3G, and a^3H levels, and here again only part of the transition probabilities which determine the term lifetimes have been measured earlier. In the absence of branching-ratio information it is thus difficult to make detailed comparisons between beam-foil and emission data for the triplet system, but we note that also for the Cr I triplets the beam-foil lifetimes [20] are 2.4–7 times longer than the CB values.

Mn I

Our results for Mn I transitions are presented in Table II. Here the experimental uncertainties are larger than those quoted for Mn II (cf. Table I). This is explained by the fact that the Mn I lines were comparatively weak at 249 keV. Attempts to obtain more accurate lifetimes by reducing the energy were not very successful because of the above-mentioned difficulties with scattering and energy loss.

The lifetimes of both the z^6D^0 and z^6F^0 terms were obtained by following the decay of two multiplet members. Our mean z^6D^0 lifetime 11.0 ± 1.5 ns is in clear disagreement with the CB value of 2.7 ns but agrees very well with Allen's [12] theoretical values, the emission measurements of Allen and Asaad [11] and Woodgate [16] and the recent absorption experiment of Blackwell and Collins [6]. The situation is similar in the case of z^6F^0 where particularly good agreement with Woodgate's results can be noted. However, for this term all other authors suggest markedly lower lifetimes. Besides Ref. [12] there exists an additional theoretical value for the z^6F^0 lifetime. The Thomas-Fermi calculations of Stewart and Rotenberg [30] predict namely a $z^6F_{11/2}^0$ decay time of 6.9 ns. In this particular case—in contrast to several other examples discussed in Ref. [30]—the Coulomb approximation and the f-sum rule seem to result in somewhat better agreement with the experimental results.

In view of the excellent agreement between this work and Woodgate's results [16] for the z^6D^0 and z^6F^0 terms it first appears somewhat disturbing to note differing results in the case of the e^6D term. However, Ref. [16] measured only the z^6P^0–e^6D branch, while there are also decays from e^6D to y^6P^0, which shorten the experimental lifetime. By combining Woodgate's results for the z^6P^0–e^6D multiplet with the f-values of Hefferlin and Gearhart [15] for the y^6P^0–e^6D branch (8 670–8 740 Å) we get an e^6D lifetime of 13 ns, in much better agreement with the beam-foil result (Table II). Table II further shows that also for the z^4F^0 term there is satisfactory agreement between beam-foil and Refs. [11], [12], and [16]. The 25% difference between this work and Woodgate's [16] result can probably be reduced by considering the decays of the z^4F^0 term to a^4F and a^4G, which branches enhance the spontaneous decay rate of the z^4F^0 term.

5. Summary and discussion

We have presented a number of experimental lifetimes in Mn I and Mn II, extracted from beam-foil decay curves. The Mn II lifetimes are 3–9 times longer than the CB values, whereas better agreement is found with Warner's data. Even here, the beam-foil lifetimes are approximately 50% longer, however. In the case of Mn I the ratio τ (beam-foil)/τ(CB) varies between 1.6 and 6, but here our measurements tend to give firm support to Woodgate's f-value scale.

Several of the Mn I and Mn II transitions discussed in this paper have been observed in the solar photosphere. Accurate knowledge of the corresponding oscillator strengths is therefore essential for determinations of the solar abundance of manganese. As early as 1948, Unsöld [31] derived a photospheric Mn abundance of log $N_{Mn} = 5.46$ (on the log $N_H = 12.00$ scale), by combining measured equivalent widths for Mn I lines with theoretical oscillator strengths, based on the f-sum rule. Using Allen and Asaad's [11] experimental Mn I f-values Goldberg et al. [32] obtained an abundance of log $N_{Mn} = 4.90$. In a later analysis Müller and Mutschlecner [33] employed the CB f-values, obtaining a slightly lower abundance value, log $N_{Mn} = 4.80$. On the basis of his improved Mn II transition probabilities Warner [34] derived log $N_{Mn} = 4.88$, in good agreement with Refs. [32, 33]. From their new f-values for the Mn I intercombination multiplet a^6S–z^8P^0 and recently measured equivalent widths, Blackwell et al. [35] found evidence for a substantially higher photospheric abundance, log $N_{Mn} = 5.42$. It is interesting to note that this latest value is fairly close to the coronal abundance of log $N_{Mn} = 5.7$, obtained by Pottasch [36] from a study of forbidden Mn transitions in the solar corona.

The excellent agreement between the present investigation and Woodgate's [16] results makes clear that the photospheric Mn abundances based on most of the previous f-value scales, in particular that of Corliss and Bozman [13] must be revised. For the Mn I lines considered by Müller and Mutschlecner [33] the average difference between the oscillator strengths of Refs. [13] and [16] is a factor of 4 (0.60 dex). If we combine the equivalent widths of Ref. [33] and the f-values of Ref. [16] we obtain a photospheric Mn abundance of log $N_{Mn} = 5.4$, in excellent agreement with the value given in Ref. [35].

As already mentioned, the disagreement between beam-foil data and Warner's Mn II f-values [18] is typically 50% and it may therefore appear difficult to modify the abundance 4.88, presented by Warner [34] towards a substantially better agreement with the value 5.42 of Blackwell et al. [35]. However, Warner's value is largely derived from the Mn II a^5D–z^5P^0 multiplet (3 441–3 497 Å), the total oscillator strength, $g_t f$ of which was in Ref. [18] found to be 4.0 (renormalization of the CB scale) or 0.048 (Coulomb approximation). In Ref. [34] the revised CB values were used for the abundance determination. However, Woodgate [16]

has also investigated this particular Mn II multiplet and normalized his measured intensities using the Mn I data. The results so obtained are considerably lower than Warner's revised CB *f*-values. For the lines used in the solar abundance determination the difference is on the average a factor of 3 (0.50 dex). Our beam-oil lifetime also supports a reduction of the a^5D–z^5P^0 multiplet strength. By combining Woodgate's values for the a^5D–z^5P^0 branch with Warner's [18] results for the a^5S–z^5P^0 multiplet it is namely possible to reduce the discrepancy between beam-foil and emission data for the z^5P^0 lifetime to a considerable extent.

There is thus already strong support for an increase of the photospheric Mn abundance, as derived from Mn II transitions, from log $N_{Mn}=4.88$ to 5.4, identical to the value based on Mn I lines. However, an accurate measurement of the relative intensities of the a^5S–z^5P^0 and a^5D–z^5P^0 multiplets, so as to extract *f*-values from the measured z^5P^0 lifetime, is expected to provide even more conclusive information.

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