PHYSICAL REVIEW A VOLUME 33, NUMBER 5 MAY 1986

Reply to comments of Bichsel *et al.* on "Mean excitation energy for the stopping power of metallic aluminum"

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(Received 13 December 1984)

Replies are made to comments made by Bichsel *et al.* on the choice of inner-shell correction, experimental energy-loss-data set used, neglect of range data, and the relevance of current optical oscillator-strength data to the metallic Al I value.

On several points I agree with Bichsel, Inokuti, and Smith:[1] (1) Establishing a definitive Bethe mean excitation energy (I) for metallic Al is important, (2) additional stopping-power measurements above 10 MeV should be made, and (3) there was an elementary error in Ref. 2 that, when corrected, would raise my alternative I value to 150–155 eV. The error in proton mass occurred only in the analysis of experimental data in Ref. 2, i.e., the relativistic factors β and γ. I based my alternative estimate on one set of experimental data[3] (the only set of stopping-power data, as opposed to range data, extending above 10 MeV), and an inner-shell correction based on explicit Born approximation calculations.[4] In addition, in Ref. 2 I suggested that the evidence supporting a metallic Al I value of 166 eV was weak. Bichsel *et al.*[1] argue that my choice of both inner-shell correction and experimental data set are incorrect and reiterate the arguments for $I = 166$ eV.

Inner-shell corrections are shown in Fig. 2 of Ref. 1, and in Sec. II of Ref. 1 it is pointed out that my correction is not of the form $C/Z\alpha 1/E_p$ above 30 MeV. This occurs because in extracting shell corrections from Born approximation calculations I did not fit the results to an asymptotic form. Rather, I set the subshell correction to zero (Table III of Ref. 4) at a high energy, above which the subshell correction fluctuated around zero (100 MeV for the K shell in Al). The fluctuations are a numerical roundoff effect. The subshell stopping powers are calculated to 3 digits. As one goes toward the asymptotic region the subshell correction is on the order of the final digit. These calculations were not designed for asymptotic studies, but expanded calculations for the asymptotic region are underway.

The basic question is the size of my correction as compared to Walske's[5] when both corrections are large, and mine is a factor of 2 larger than Walske's. Walske's correction is based on hydrogenic calculations, with a modification due to Brown,[6] to simulate real atom properties. The shell corrections account for the departure of the subshell stopping power from its asymptotic form due to the details of atomic structure. It is surprising that when the subshell correction is large, hydrogenic and free-electron gas Thomas-Fermi estimates are considered satisfactory. My correction is based on numerical calculations in the plane-wave Born approximation and is subject to computational errors. The structure in my correction near 8 MeV in Fig. 2 of Ref. 1 arises from fluctuations in the $2p$-shell correction (Table III of Ref. 4). This is a minor point, and in developing an I value the L-shell correction was smoothed (Fig. 1 of Ref. 4). The departure of my Z^* values (the coefficient of the log term in the reduced subshell stopping power) from the values of Bethe, Brown, and Walske,[7] $\frac{1}{2}(Z_K + N_K)$, where Z_K (N_K) is the summed subshell optical oscillator strength (subshell occupation number), may indicate a significant numerical error. However, in calculations on the stopping power of the noble gases,[8] both when the Z_K^* values are larger than (Ar and Kr) and in agreement (Xe) with $\frac{1}{2}(Z_K + N_K)$, my K-shell corrections are a factor of 2 larger than Walske's.[5] Further, in calculating ionization generalized oscillator strengths one truncates an infinite sum over angular momentum l (at $l = 12$ in my calculations). The truncation could affect the generalized oscillator strengths at the Bethe ridge. However, such truncation errors should be cumulative when summed over subshells, yet the coefficient of the leading term in my stopping-power expression is $Z = 13$, within numerical error. It is possible that effects of the truncation are present in the shell correction. Studies of the effects of the departure of Z^* values from the Bethe, Brown, and Walske[7] values are underway, but an examination of possible truncation errors is difficult.

With regard to the choice of experimental data sets in Ref. 2, the data of Sorensen and Andersen[3] are the only stopping-power data for Al extending above 10 MeV. When one compares plane-wave Born approximation calculations with experiment, one wants data extending to as high an energy as possible. The later results of Andersen, Bak, Knudsen, and Nielson[9] were motivated by disagreements between the Sorensen and Andersen[3] results and other data. The later results of Andersen *et al.*[9] were lower than those of Ref. 3, indicating a systematic error in one of the two sets, or over optimistic error bars. Andersen *et al.*[9] indicate no preference for one set of data over the other. A more extensive discussion of the data can be found in Refs. 10–13. If the data of Ref. 3 are rejected, then my estimate is raised to $I = 155$–160 eV.

I did not consider data sets obtained from range measurements. As I was merely questioning the established $I = 166$ eV value, rather than attempting to establish a new "definitive" value, one set of precise data seemed sufficient. However, since the authors of Ref. 1 introduce range data, it seems useful to discuss it. With the range data of Bloembergen and van Heerden[14] shown in Table I of Ref. 1, a strong case is made in Ref. 1 for $I = 166$ eV. However, the data of Ref. 14 show some internal inconsistency, i.e., in several instances higher-energy protons have shorter range. To broaden the comparison, I include in my own Table I Al proton ranges obtained from the recent compilation of Janni,[15] by a linear fit, and two measured values near 100 MeV obtained by Portner and Moore.[16] Janni's results are obtained from a best fit of the Bethe formula to an extensive

TABLE I. The ranges in columns 2, 5, and 7 are from Table I of Ref. 1. The range values in column 3 are from Ref. 15, and percent differences are taken with respect to these smooth values. The last two rows are from Ref. 16 and are included to show the variability of precise measurements.

E (MeV)	R_{ex} (gm/cm^2)	R_J (gm/cm^2)	$\frac{R_{\mathrm{ex}}-R_J}{R_{\mathrm{ex}}}$ (%)	R_{th} (gm/cm^2)	$\frac{R_{\mathrm{th}}-R_J}{R_J}$ (%)	R_M (gm/cm^2)	$\frac{R_M-R_J}{R_J}$ (%)
11.82	0.2271	0.2277	−0.3	0.2271	−0.3	0.2271	−0.3
14.91	0.3438	0.3401	1.1	0.3442	1.2	0.3430	0.9
17.84	0.4684	0.4668	0.3	0.4690	0.5	0.4666	−0.04
34.96	1.547	1.537	0.7	1.549	0.8	1.531	−0.4
37.16	1.757	1.719	2.2	1.726	0.4	1.706	−0.8
39.66	1.928	1.925	0.2	1.939	0.7	1.915	−0.5
42.57	2.198	2.188	0.5	2.199	0.5	2.171	−0.8
44.86	2.402	2.397	0.2	2.414	0.7	2.382	−0.6
47.67	2.634	2.675	−1.5	2.690	0.6	2.653	−0.8
52.08	3.189	3.130	1.7	3.149	0.6	3.104	−0.8
52.33	3.188	3.157	1.0	3.176	0.6	3.130	−0.9
56.68	3.687	3.638	1.3	3.661	0.6	3.607	−0.9
56.96	3.784	3.670	3.1	3.693	0.6	3.638	−0.9
61.79	4.258	4.241	0.4	4.268	0.6	4.207	−0.9
62.10	4.280	4.279	0.02	4.306	0.6	4.241	−0.9
65.78	4.756	4.737	0.4	4.770	0.7	4.697	−0.8
66.10	4.786	4.779	0.1	4.812	0.7	4.738	−0.9
72.94	5.773	5.692	1.4	5.730	0.7	5.641	−0.9
73.05	5.714	5.707	0.1	5.745	0.7	5.656	−0.9
75.70	6.141	6.077	1.1	6.120	0.7	6.024	−0.8
75.84	6.078	6.097	−0.3	6.140	0.7	6.044	−0.9
99.58	9.768	9.863	−1.0				
99.88	9.840	9.914	−0.7				

data base of energy-loss measurements, including the data of Ref. 3, using Walske inner-shell corrections, and with an Al I value of 160.1 eV. Janni's ranges (called pathlengths in Ref. 15) are consistently lower than those of Ref. 14 and higher than those of Ref. 16. When I compare the calculated ranges of Table I of Ref. 1 with Janni's ranges I find the percent differences listed in columns 7 and 9, i.e., R_{th} is consistently higher by 0.6% and R_M is consistently lower by -0.8%. This occurs even though R_{th} uses $I=166$ eV, R_M uses $I=150$ eV, and the Janni results use $I=160.1$ eV. My conclusion is that range data are not a sensitive function of choice of I and corrections, and, in consequence, energy-loss measurements are to be preferred for the determination of I. A comparison of the data in Refs. 14 and 16 suggests that range measurements have the same problems as energy-loss measurements; different precise experiments differ by considerably more than their error bars.

To integrate over optical oscillator strengths and conclude $I=166.\pm 5.$ eV, requires that $\ln(I)$ be determined to an accuracy of better than 0.6% (comparable to the accuracy required in energy-loss or range measurements; there is no free lunch). Shiles, Sasaki, Inokuti, and Smith[17] argue that for metallic Al, with an adjusted set of photophysics data, they achieve better than 0.5% consistency in Kramers-Kronig relations, though they make no such accuracy claims for the experimental data used. With the adjusted set of photophysics data they obtain $I=165.6(7)$ eV (modified to "166 eV with an uncertainty of a few eV" in Ref. 1). Unfortunately, the summed oscillator strength in the adjusted calculation is 14.08, rather than 13. This is a difference of 8%, and would produce an error in $\ln(I)$ and a larger error in I. The error in I will depend critically on the location of the error in the oscillator-strength distribution. To correct for this 8% difference Shiles *et al.*[17] modify the optical oscillator strength between 72 and 500 eV with the criteria "By simultaneously requiring that both the f-sum rule be satisfied and that the mean excitation energy agree with experiment."[18] The modified I value becomes 165.7(1) eV. This calculation is then taken as independent confirmation of $I=166$ eV for metallic Al. In their modified calculation Shiles *et al.*[17] have assumed $I=166$ eV and modified the oscillator-strength distribution to satisfy the assumption. In determining an I value from summed optical oscillator strengths one wants optical oscillator strengths that do not bias the calculation. In Ref. 19 the adjusted data of Shiles *et al.*[17] are shown along with the experimental data of Gahwiller and Brown[20] and Hagemann, Gudat, and Kunz,[21] in the photon energy range 75–250 eV. Near 95 eV the adjusted data of Shiles *et al.*[17] are lower than the experimental data, while between 100 and 150 eV they are consistently higher. To see the effect on I of the adjustment of the experimental data, the Shiles *et al.*[17] result, $\ln[I(\mathrm{eV})]=71.9428/14.08$, was recalculated with the experimental data between 75 and 150 eV. With the data of Ref. 20 the I value is 158.2 eV, while with the Ref. 21 data it is 159.0 eV. Given the errors in the photophysics data base, I doubt that these I values are any more relevant in determining a metallic Al I value than those presented in Ref. 17.

Finally, in Ref. 2 I suggested that all of the data sets in the photophysics data base be examined for the effects of oxygen contamination. This includes data at $h\nu > 160$ eV. In Ref. 2 I examined possible effects at $h\nu < 160$ eV, but did not suggest that this exhausted possible corrections. Of course, reducing oscillator strength for $h\nu < 160$ eV will raise I, but corrections for $h\nu > 160$ eV, reducing oscillator strength, will lower I. The experimental photophysics data base for metallic Al leads to a summed oscillator strength of 14.08. To reduce this to 13.0 requires either a careful reexamination of past experiments, or new experiments. For example, a comparison of photon energy cubed, multiplied by a calculated atomic Al photoionization cross section, is in good agreement with recent measurements of Henke *et al.*[22] for photon energies between 100 and 400 eV, but at higher energies the former saturates while the latter continues to rise. This suggests the onset of oxygen K-shell photoionization in the measurements and the possibility of estimating the magnitude of the oxygen contamination. With an estimate of the oxygen contamination, one can correct the data between 100 and 400 eV, as well as above 400 eV. Lastly, with regard to the "profound difference" between the metallic and atomic Al photoabsorption spectrum, the absence of a minimum in the experimental metallic Al photoabsorption spectrum is consistent with either no metallic Al minimum and no oxygen contamination, or an Al minimum masked by oxygen contamination absorption. This is an interesting question having to do with the persistence of such features of atomic photoabsorption as Cooper minima in solid environments. As an alternative to band-structure arguments, one may ask whether the metallic Al valence electron photoionization cross section is identically zero at photon energies well above that at the atomic Cooper minimum; if it is not zero, does it depart significantly from the atomic calculation at high energy; if it does not, does it show residual Cooper minimum behavior at low energy? This question can be addressed experimentally by valence photoelectron spectroscopy on clean samples.

This work was supported by the U. S. Department of Energy.

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[1] H. Bichsel, M. Inokuti, and D. Y. Smith, preceding paper, Phys. Rev. A **33**, 3567 (1986).

[2] E. J. McGuire, Phys. Rev. A **28**, 53 (1983).

[3] H. Sorensen and H. H. Andersen, Phys. Rev. B **8**, 1854 (1973).

[4] E. J. McGuire, Phys. Rev. A **28**, 49 (1983).

[5] M. C. Walske, Phys. Rev. **88**, 1283 (1952).

[6] L. M. Brown, Phys. Rev. **79**, 297 (1950).

[7] H. A. Bethe, L. M. Brown, and M. C. Walske, Phys. Rev. **79**, 413 (1950).

[8] E. J. McGuire, Phys. Rev. A **28**, 2096 (1983).

[9] H. H. Andersen, J. F. Bak, H. Knudsen, and B. R. Nielson, Phys. Rev. A **16**, 1929 (1977).

[10] R. Ishiwari, N. Shiomi, S. Shirai, and U. Uemura, Phys. Lett. **48A**, 96 (1974).

[11] R. Ishiwari, N. Shiomi, and S. Shirai, Phys. Lett. **51A**, 53 (1975).

[12] H. H. Andersen, Phys. Lett. **56A**, 443 (1976).

[13] R. Ishiwari, N. Shiomi, and N. Sakamoto, Phys. Lett. **75A**, 112 (1979).

[14] N. Bloembergen and P. J. van Heerden, Phys. Rev. **83**, 561 (1951).

[15] J. F. Janni, At. Data Nucl. Data Tables **27**, 147 (1982).

[16] P. M. Portner and R. B. Moore, Can. J. Phys. **43**, 1904 (1965).

[17] E. Shiles, T. Sasaki, M. Inokuti, and D. Y. Smith, Phys. Rev. B **22**, 1612 (1980).

[18] Reference 17, p. 1621.

[19] *Optical Properties of Metals*, edited by J. H. Weaver, C. Krafka, D. W. Lynch, and E. E. Koch, Physics Data, Vol. 18-2 (Fachinformationszentrum, Karlsruhe, Federal Republic of Germany, 1981).

[20] C. Gahwiller and F. C. Brown, Phys. Rev. B **2**, 1918 (1970).

[21] H. J. Hagemann, W. Gudat, and C. Kunz, J. Opt. Soc. Am. **65**, 742 (1976).

[22] B. L. Henke, P. Lee, T. J. Tanaka, R. L. Shimabukuro, and B. K. Fujikawa, At. Data Nucl. Data Tables **27**, 1 (1982).