Bethe mean excitation energy for solid aluminum

E. J. McGuire
Sandia National Laboratories, Albuquerque, New Mexico 87185
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A critical analysis is made of the arguments that the Bethe mean excitation energy (I) for solid Al is 163–169 eV. The arguments for this I value are shown to be nondefinitive. Using recent measurements and inner-shell corrections established in the preceding paper, I suggest that the solid-Al I value is in the range of 145–150 eV.

I. INTRODUCTION

Aluminum is widely used as a comparison element in experimental studies of relative proton stopping power.[1] One reason for this is that as a low-Z element Al is amenable to treatment by the Bethe stopping-power theory.[2] That is, the criteria for validity of the Bethe theory are satisfied for protons in the 10–20 MeV regime. One property of solid Al that appears to be well established is the Bethe mean excitation energy I reported to lie in the range of 163–169 eV. Various calculations for atomic Al agree on a value $I \cong 124$ eV for atomic Al.[3,4]

In the course of explicit calculations of atomic Al stopping power using the Born approximation and summing the contribution of excitation and ionization to stopping power,[4] a peculiar feature was observed. The atomic stopping power calculated explicitly was in excellent agreement with measurements in solid Al extending to 18 MeV.[5] This suggested that, perhaps, for solid Al the I value might be the same as for atomic Al.

Several arguments can be made to support the contention that $I = 163-169$ eV is the appropriate Bethe mean excitation energy for solid Al. First, the high-energy measurements (700 MeV) of Barkas and von Friesen[1] indicate $I = 163-169$ eV. Second, the data analysis of Bichsel and Uehling[6] support such an I value. Third, Shiles *et al.*[7] have shown, by integrating over measured oscillator strengths, that $I = 165.7$ eV.

In the following, I point out (1) that the available measurements of I at high energy support an I value of 145–150 eV, (2) that the result of Bichsel and Uehling[6] depend on both the set of measurements used in the analysis and the choice of inner-shell correction, and (3) that while it is agreed that the oscillator strength data used by Shiles *et al.*[7] require a renormalization, their renormalization is incorrect.

II. DISCUSSION AND REANALYSIS

A. Preliminaries

Before examining the arguments mentioned in the Introduction, I present a simple analytical framework. If

$$S_T = (-1/n)\frac{dE}{dx}$$

is a stopping power either measured[5,8] (and therefore, involving small relativistic effects even at 10 MeV) or calculated,[4] then a nonrelativistic energy-dependent mean excitation energy can be defined via

$$\ln I(E_p) = \ln\left[\frac{4M_eE_p}{M_p}\right] - \frac{M_e}{M_p}\frac{E_p}{4\pi a_0^2}\frac{S_T}{Z_e} \quad (1)$$

and a relativistic energy-dependent mean excitation energy via

$$\ln I(E_p) = \ln(2M_ec^2\gamma^2\beta^2) - \beta^2 - \frac{\beta^2 S_T}{4\pi r_0{}^2 m_0c^2 Z_e}\ , \quad (2)$$

where E_p is the proton energy, I is the mean excitation energy, Z_e is the number of electrons in the atom, r_0 is the classical electron radius ($r_0 = e^2/m_0c^2$), and β and γ have their usual meaning. In comparing relativistic and nonrelativistic $I(E_p)$ values, the relativistic values are plotted at

$$\tfrac{1}{2}m_0c^2\beta^2 = \tfrac{1}{2}m_0v^2$$

rather than at $(\gamma-1)m_0c^2$. The measurements of Refs. 5 and 8 and our explicit calculations[4] contain shell corrections. These should become negligible at high energy with the $I(\infty)$ value emerging asymptotically. In addition, the measurements of Refs. 5 and 8 contain $(Z_1)^3$ and $(Z_1)^4$ corrections. These are accounted for using the measured corrections of Ref. 8.

In Fig. 1 the results of the nonrelativistic Born calculations[4] are shown as solid circles. Above 40 MeV the calculations lead to an I value of 120–125 eV. The squares are the data of Sørensen and Andersen[5] treated nonrelativistically. The precipitous decline of the squares between 10 and 18 MeV indicates that the nonrelativistic treatment of the data is incorrect. The open circles are the data of Sørensen and Andersen[5] treated relativistically. The large differences between circles and squares arise from the factor β^2 multiplying the measured stopping power, and the fact that we exponentiate calculated quantities to determine $I(E_p)$. The important conclusion is that the measurements of Sørensen and Andersen[5] show no flattening out even at 18 MeV. The measurements are not asymptotic and, based on the data alone, one can say no more than $I(\infty) \leq 160$ eV. However, if one assumes that the difference between I (18 MeV) and $I(\infty)$ in both the calculations and the measurements of Ref. 5 is the nonzero inner-shell correction, then the calculations indicate a reduction of 13 eV; i.e., the experimental data, if extended to higher energies, would reach the asymptotic value

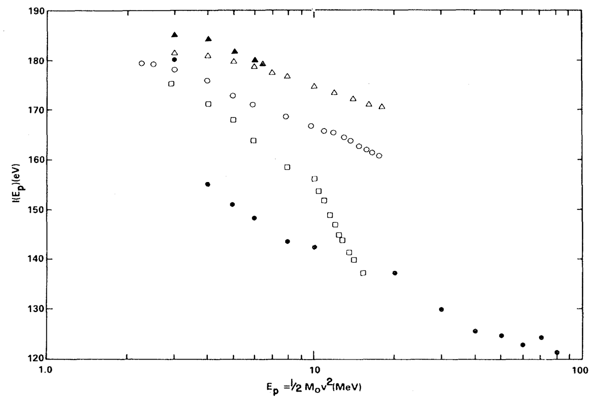

FIG. 1. Solid circles are nonrelativistic $I(E_p)$ values from atomic calculations. The open squares and open circles are $I(E_p)$ values obtained from the data of Ref. 5, via the nonrelativistic and relativistic analyses of Eqs. (1) and (2), respectively. Open triangles are obtained using the parameters of Ref. 6 and Eq. (3). The solid triangles are $I(E_p)$ values obtained from the data of Ref. 8.

$I(\infty) \approx 147$ eV.

On the other hand, the data of Andersen *et al.*,[8] treated relativistically, are shown as solid triangles in Fig. 1. This data set extends to 6.4 MeV only. Using the calculated inner-shell correction of Fig. 1,

$$I(6.4) - I(\infty) = 23 \text{ eV},$$

suggests an $I(\infty)$ for this data set of 156 eV.

Thus the use of two recent and precise (both claim errors $\leq 0.5\%$) sets of measurements leads to significantly different solid Al I values.

B. The high-energy results

There are three relevant and related papers on the stopping power of Al at 700 MeV, by Bakker and Segrè,[9] Mather and Segrè,[10] and Barkas and von Friesen.[1] In the paper by Bakker and Segrè[9] relative stopping powers in various materials are measured. They are made absolute by assuming an Al I value measured before World War II by Wilson,[11] i.e., $I = 150$ eV. The later work of Mather and Segrè[10] presents absolute measurements of I for various materials; two values are given for Al, 145.5 and 150.3 eV. In a later paper, Barkas and von Friesen[1] return to the same topic with better multiple scattering corrections. They make relative stopping-power measurements, and attempt to make them absolute using the then recently published value $I = 163$ eV of Bichsel and Uehling.[6] The conclusion one reaches from examining the work at high energy is that only Refs. 10 and 11 are relevant and they indicate an I value in the 145–150-eV range. References 1 and 9 assume rather than determine an Al I value.

C. The analysis of Bichsel and Uehling

From the analysis of range and stopping-power data available to them at the time Bichsel and Uehling[6] obtained an effective energy-dependent I value for Al given by

$$\ln I(\text{eV}) = \ln(163 \text{ eV}) + (C_K + C_L)/13 , \tag{3}$$

where C_K was taken from Walske's[12] hydrogenic calculations and C_L was taken to be 1.5 (MeV)/E_p (MeV). $I(E_p)$ obtained from this expression is shown in Fig. 1 as open triangles between 3 and 18 MeV. The $I(E_p)$ values are 3–10 eV higher than those of Sørensen and Andersen[5] and in good agreement with those of Andersen *et al.*[8] However, the $I(E_p)$ values of Bichsel and Uehling show a smaller slope with increasing E_p than do the data of Refs. 5 and 8. In the preceding paper,[13] I present inner-shell corrections obtained from explicit Born calculations. They are larger than those used by Bichsel and Uehling[6] but from 4 to 8 MeV they are in good agreement with those obtained from experiment by Andersen *et al.*[8] The value of $I(\infty)$ obtained by Bichsel and Uehling[6] depends critically on the choice of inner-shell corrections. To show this I use

$$\ln I(\infty) = \ln I(E_p) - (C_K + C_L)/13 \tag{4}$$

with $I(E_p)$ chosen from the relativistic analysis of the data of Sørensen and Andersen.[5] I use the data of Ref. 5 rather than Ref. 8 because the measurements in Ref. 5 extend to higher energy. The authors of Ref. 8 call attention to the discrepancy between the data of Ref. 5 and Ref. 8, but do not resolve the discrepancy.

For $C_K + C_L$, I use both the values used by Bichsel and Uehling,[6] and those I obtained in the preceding paper.[13] The results are shown in Table I. From Table I it is clear that the $I(\infty)$ values obtained with the Bichsel-Uehling[6] choice of $(C_K + C_L)/13$ and the data between 3 and 18 MeV lie in the range of 154–160 eV. On the other hand, while the $I(\infty)$ values obtained with my $(C_K + C_L)/13$

TABLE I. The second column lists the $I(E_p)$ values found from a relativistic analysis of the data of Ref. 5; the third and fifth columns list the summed K- and L-shell corrrections of Refs. 6 and 13, respectively, while the fourth and sixth columns list the Al I values obtained from Eq. (4). The seventh column lists the I value found using the sixth column and the empirical $(Z_1)^3$ and $(Z_1)^4$ corrections of Ref. 8.

E_p (MeV)	$I(E_p)$	$(C_K + C_L)/13$	$I(\infty)$	$(C_K + C_L)/13$	$I(\infty)$	$I(\infty)_c$
3	178.0	0.107	160.0	0.354	125.0	132.6
4	176.0	0.104	158.6	0.227	140.3	147.0
5	173.0	0.098	156.9	0.179	144.7	150.3
6	171.0	0.092	156.0	0.1415	148.4	153.1
8	168.7	0.080	155.7	0.1231	149.2	152.7
18	160.7	0.045	153.6	0.1000	145.4	146.7

values from the preceding paper show greater variation [because the (C_K+C_L) values are larger], above 5 MeV and including the point at 18 MeV the $I(\infty)$ values lie between 145 and 150 eV.

In using the experimental data to determine $I(\infty)$, one is including $(Z_1)^3$ and $(Z_1)^4$ effects (L_1 and L_2, for protons). These corrections enter via

$$\ln[I(\infty)]_c=\ln[I(\infty)]+L_1+L_2 .$$

Using the expressions

$$L_0=4.19\times 10^{15}Ep(\mathrm{MeV})S_T/Z_2 ,$$

$$L_1=2.68\frac{M_p}{M_e}\frac{(Z_2)^{2/3}}{E_p\,(\mathrm{Ry})}\left[1-0.132\ln\left[\frac{M_eE_p(\mathrm{Ry})}{M_p(Z_2)^{2/3}}\right]\right]\times\left[\frac{L_0}{(Z_2)^{1/2}}\right],$$

and

$$L_2=-1.6\frac{M_p}{M_e}\frac{1}{E_p(\mathrm{Ry})} ,$$

where L_1 and L_2 are empirically determined in Ref. 8, I find the $I(\infty)_c$ values given in the last column of Table I. Including the $(Z_1)^3$ and $(Z_1)^4$ effects slightly raises the I values inferred from the data of Ref. 5, but does not change the basic conclusion: $I=145-150$ eV for solid Al.

It is clear from the above that the $I(\infty)$ value for Al depends critically on inner-shell corrections when data below 20 MeV are used in the analysis, and on the choice of experimental data sets. While the above analysis is not conclusive, the trend found from the data of Sørensen and Andersen[5] indicates that $I(\infty)$ is lower than $157-159$ eV, and my analysis suggests $I(\infty)$ is in the $145-150$ eV range.

Finally, the use of Walske's hydrogenic K-shell correction in nonhydrogenic systems is an assumption which will be examined in a later paper studying inner-shell corrections as a function of degree of ionization.[14]

D. The oscillator strength integration of Shiles *et al.*

Shiles *et al.*[7] have obtained an I value of 165.7 for Al by integrating experimental $df/d\epsilon$ values over the entire spectrum. However, they point out (1) that the experimental data contain errors, (2) that oxygen contamination is likely to be a problem, and (3) that the raw data for $df/d\epsilon$ integrated over the spectrum lead to a value of 14.08 rather than 13.00. As a consequence of point (3), it was necessary for Shiles *et al.*[7] to renormalize the experimental data by reducing $df/d\epsilon$ at and above the L-edge threshold energy. This renormalization was justified by reference to atomic calculations. That is, the sum of atomic $3s$ and $3p$ oscillator strengths is 3.12, the sum of $2s$ and $2p$ oscillator strengths is 8.33, and the summed $1s$ oscillator strength is 1.54. The experimental oscillator strength integrated from the infrared to the L-shell photoionization threshold (assumed by Shiles *et al.*[7] to exhaust the summed $3s$ and $3p$ oscillator strengths) is 3.1(1), from the L-shell threshold to the K-shell threshold (assumed by Shiles *et al.*[7] to exhaust the summed $2s$ and $2p$ oscillator strengths) the experimental oscillator is 9.3(5), and from the K-shell threshold to infinity is 1.6(1). Shiles *et al.*[7] argue that the agreement of the summed $3s$ and $3p$ atomic oscillator strength (3.1) with the integrated oscillator strength up to the L edge, and the disagreement between the summed $2s$ and $2p$ atomic oscillator strength (8.33) and the integrated experimental oscillator strength between the L and K edges [9.3(5)], indicates that the measurements between the K and L edges are too high. This is the basis for their renormalization.

I suggest that the renormalization of Shiles *et al.*[7] is incorrect. Both the $3s$ and $3p$ atomic Al photoionization cross sections have minima between 10 and 50 eV. As a result the $3s$ and $3p$ continuum oscillator strength is concentrated at low and high energies. Our calculations indicate that 20% of the atomic $3s$ and $3p$ oscillator strength (0.62 of 3.20) is at energies higher than the Al L edge. Since

$$8.33+0.62=8.95$$

is within error bounds of the experimental oscillator strength integrated between the L and K edges [9.3(5)], the main argument for the renormalization of Shiles *et al.*[7] is dubious.

Alternatively, using the same arguments as above, I suggest that 0.62 of the summed experimental oscillator strength up to the L edge is excessive, and probably arises from oxygen contamination. My calculations on photoabsorption in atomic oxygen show that 6.2 of the summed atomic oxygen oscillator (8.0) strength lies at energies below 72 eV (the L edge in solid Al). Thus I conclude that some or all of the experimental data used by Shiles *et al.*[7] for energy less than the Al L-shell threshold contained a contribution to the oscillator strength from oxygen contamination—which, when averaged over the 72-eV range, is effectively a 10% oxygen contamination. Correcting a variety of experimental data sets for an oxygen contamination unspecified in each set is a difficult problem, and one which may limit the approach of Shiles *et al.*[7] to gaseous systems.

III. CONCLUSIONS

From the above I conclude that there is no evidence for an I value in solid Al of $163-169$ eV, other than the analysis of Bichsel and Uehling.[6] However, their analysis depends critically on the choice of shell corrections. Using shell corrections obtained in the preceding paper,[13] which are in agreement with the recent measurements of Ref. 8, and the measurements of Sørensen and Andersen,[5] I find an I value for solid Al of $145-150$ eV. This is in agreement with the limited evidence from high-energy measurements.

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