Investigation of the ratio of proton-stopping cross sections in Ag and Au

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Knowledge of the ratio of stopping cross sections may help in determining best values from measurements. The case of Ag and Au is discussed, where theoretical considerations show that this ratio always has a value smaller than 1. This is confirmed experimentally for proton energies larger than 70 keV, in contradiction to all published stopping-power tabulations.

In all processes of material analysis and material modification by ion beams, the stopping cross section ϵ is an indispensable quantity. It is defined by

$$\epsilon=-\frac{dE_1}{Ndx}, \tag{1}$$

where E_1 is the projectile's energy, N is the atomic density of the target, and x is the path length in the target. However, the determination of reliable stopping data from measurements, e.g., for protons and He ions, is complicated by the large scatter of data. This applies mainly to low projectile velocities. So best values have been often obtained introducing certain constraints: the stopping cross section should vary smoothly[1,2] with the atomic number Z_2—for want of a quantitative description of the so-called Z_2 oscillations—or proton and helium stopping values should be related by target-independent effective charges of the projectiles.[3]

We will now focus on slow protons in metals, where the stopping is to a large extent due to the nonlocalized target electrons.[4] In this case the stopping properties of the elements with similar band structure should be compared to each other. As an example we will discuss the stopping cross section of Ag and Au. For these noble metals, the Fermi level is found in the middle of the 5s and 6s band, respectively, and the outermost d bands are well below the Fermi level but still within the corresponding s bands.[5]

We now consider the ratio of the stopping cross sections at equal velocity

$$R=\frac{\epsilon_{\mathrm{Ag}}}{\epsilon_{\mathrm{Au}}}. \tag{2}$$

For high velocities R is much smaller than 1. This is most easily seen when the Bethe-Bloch equation is used for ϵ, with an effective number of electrons and hence an effective ionization potential to account for shell corrections.[6] This effective number, which might be defined as the number of electrons slower than the projectile, is larger for Au than for Ag.

For lower velocities all commonly used stopping power tabulations claim that R exceeds the value 1: the crossing of the value 1 is predicted for $E_1=125$ keV (Ref. 1), for $E_1=78$ keV (Ref. 3), for $E_1=66$ keV (Ref. 2), and for $E_1=73$ keV (Ref. 7). In contradiction to that it is very unlikely, from simple considerations, that R exceeds 1 for any proton energy: For very small energies the projectile can only interact with electrons very close to the Fermi surface;[8] any interaction with bound electrons is reduced due to the threshold effect.[9] For these energies we can

TABLE I. Quantities relevant to the description of the stopping due to the nonlocalized electrons in Ag and Au.

	Ag	Au	Units
Coefficient of linear term in low-temperature specific heat[a]	0.6470	0.6988	mJ/mol K^2
Density of states at the Fermi level	0.064 65	0.070 59	a.u.
Density of low-energy model plasma	0.008 773	0.011 42	a.u.
One-electron radius r_s	3.0079	2.7547	a.u.
Atomic density	0.008 666	0.008 750	a.u.
Range of d band relative to Fermi level[b]	-7 to -4	-7 to -2	eV
Plasmon energy[c]	23	24	eV
Density of high-energy model plasma	0.056 851	0.061 902	a.u.

[a]Reference 11.
[b]Reference 13.
[c]Reference 15.

therefore use a model of a free-electron plasma with the same density of states at the Fermi surface. This quantity can be derived[10] from the term linear in temperature of the specific heat at low temperatures. Both processes, the energy loss of slow projectiles and the heating, describe the transfer of small amounts of energy to a dense plasma. In Table I we cite the results of Ref. 11 and the deduced densities of our model plasmas together with the corresponding one-electron radii. From density functional calculations[12] we find that by using these plasma densities the stopping power $S=-dE_1/dx$ of Au exceeds that of Ag by 8%. As the atomic densities differ only by 1% (see Table I) we get for the lowest energies $R=0.93$.

With increasing energy the projectile interacts more and more with electrons at deeper levels, i.e., with electrons of the d band. From Table I it can be seen that the d electrons of Au start to contribute to the stopping process at about half the projectile energy compared to the d electrons of Ag, so $\epsilon_{Ag}<\epsilon_{Au}$ will still be valid. For even higher energies the response of the nonlocalized target electrons might again be described by a free-electron plasma.[14] Here the density has to be adjusted to give the same plasma frequency as found in electron energy-loss measurements.[15] The density is now higher than for the low-energy model (Table I) since the d electrons also take part, to some extent, in plasma oscillations induced by the swift projectile. But we find again a larger density for Au and hence a larger stopping cross section, at least as far as the plasma contribution is considered. The contributions from the cores will enhance this difference mainly due to the loosely bound $4f$ electrons of Au [binding energy about 85 eV (Ref. 16)]. As a check we have measured R in the energy range from 70 to 500 keV. We have employed the method introduced by Andersen *et al.*,[17] where both targets are alternately exposed to the beam for the same amount of time. The ratio R is determined from the relative heights of the backscattering spectra (see, e.g., Ref. 18). Here only the backscattering cross section has to be know, which was calculated[19] for screened atoms from the scattering integral. The result is displayed in Fig. 1.

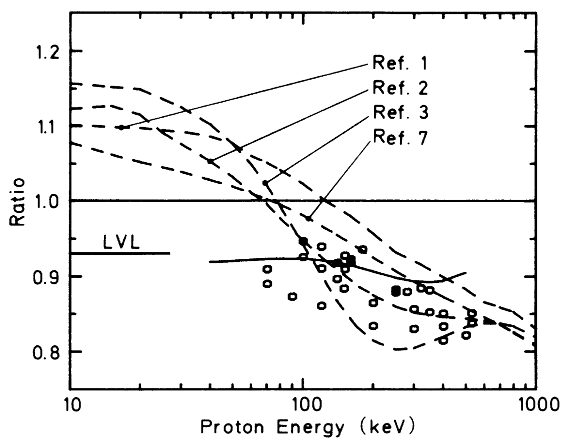

FIG. 1. Ratio of stopping cross sections of Ag and Au. ○, this work (error 5%); solid line, Ref. 20 (error 4%); dashed lines, tabulations (Refs. 1–3 and 7). LVL, low velocity limit (see text).

In spite of the limited range and the 5% scatter of the data, this measurement disproves the trend of R found from tabulations, which are shown as dashed lines. Together with the result of absolute measurements[20] (solid line in Fig. 1), our measurements are also in agreement with the estimated low velocity limit of $R=0.93$.

It is interesting that a value of R smaller than 1 was found also in almost all papers,[21–25] where ϵ values have been published for both elements, and which formed, among others, the basis for the tabulations. (Only in the early work by Bätzner[26] does R approach 1 at $E_1=8$ keV.) The reason why this fact is not reproduced in the tables, is that for Au there exists additional low-energy data,[27,28] which lowered the fitted curve of Au. Using R as a constraint these data would have also brought down the stopping cross sections of Ag to—in our opinion[20]—more realistic values.

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