

Stopping Power of Au for Silver Ions at Low Velocities

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Abstract

Energy loss measurements for the slowing down of Ag ions in Au, in the velocity range $1.6v_o < v < 4.4v_o$, where v_o is the Bohr velocity, are presented. The measurements were performed using the Doppler shift technique and also with a new method, where a secondary beam of low velocity heavy ions is produced by elastic scattering of the accelerated beam. The results are compared to the SRIM2000 calculations (www.srim.org) and to recent measurements in this velocity region.

Key words: Stopping Power; Energy Loss

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1 Introduction

The stopping power of solids for heavy ions at low velocities is still poorly known. Since *ab initio* calculations are unable to produce reliable quantitative estimates, most of the predictions in use are of a semiempirical nature. In the case of heavy ions, the large number of combinations of Z_1 , Z_2 , wide range of energies, and sparse experimental data, make even the semiempirical predictions contain, in general, large uncertainties. This is especially true at low velocities (around and below the Bragg peak) due to the additional difficulties in predicting the rapid changes in the charge state of the slowing ion, and to the complicated dependence of the stopping force on the atomic numbers. An accurate knowledge of the stopping force in this low velocity region is necessary in many experimental techniques, like those for ion implantation, surface analysis, etc. In nuclear physics, for instance, the measurement of pico-second

lifetimes of nuclear states is normally performed with the Doppler shift attenuation method (DSAM) [1]. Here the knowledge of the stopping power as a function of the velocity of the recoiling excited nucleus is used to determine the time scale for the nucleus decaying in flight while slowing down in a heavy substrate, usually Au or Pb. Since the velocities acquired by the nucleus come from the kinematics of a nuclear reaction induced by heavy ions, their initial values will be, in most cases, a few percent of the velocity of light. For this velocity region, (i.e. $v < v_e = v_o Z_1^{2/3}$, where v_e is the Thomas-Fermi velocity of the electrons in an atom with atomic number Z_1 and $v_o \simeq c/137$ is the Bohr velocity), one of the most commonly used semiempirical calculations is that developed by Ziegler, Biersack, and Littmark (ZBL) [2,3]. In that work, both the energy losses by inelastic collision of the ion with electrons of the target (electronic stopping power) as well as the more sudden losses by atom-atom elastic collisions, which are important in this velocity region, are taken into account. In this paper, we present experimental data for the stopping of silver ions in gold at velocities below $4.4v_o$, using two different techniques. The results are compared with the predictions of the ZBL parameterization and with other recent measurements in the same velocity region.

2 Measurements and Analysis

The experimental data we present here were obtained with two different techniques. In the first one the Doppler Shift method, already employed in several other measurements [4,5,6], was used. In this technique, the mean energy of Coulomb excited target nuclei ($^{107,109}\text{Ag}$ in the present case) after traversing the stopper foil, is measured by the determination of the Doppler shift in the energy of the gamma rays emitted by the decaying nuclear states. Excited nuclei were produced by collision with heavy projectiles (^{16}O , ^{28}Si) and the nuclei recoiling in a cone centered around 0° were selected by imposing time coincidence between the projectiles scattered near 180° , detected in a Si annular detector, and the gamma rays.

For velocities below $\sim 2v_o$, the use of the Doppler shift technique would produce results with large uncertainties, due to the small difference between shifted and unshifted energies. In order to obtain reliable data for these energies, we developed a new technique which proved to be better than the one used for the preceding measurements. The second set of data was obtained using the secondary heavy ion beam (Ag in the present case) produced by elastic scattering of the primary accelerated beam (^{16}O , ^{28}Si), the scattered particle being detected in kinematic coincidence with the recoil. Using a highly collimated geometry, it is possible to have a well defined secondary beam, both in energy and in direction.

The gamma-ray energies were measured with a HP germanium detector placed at the 0° direction, with about 2.3 keV FWHM energy resolution for the ^{60}Co lines. The stopper employed consisted of a rolled Au foil, of 2.02 ± 0.04 mg/cm² thickness, determined by measuring the energy loss of alpha particles from a ^{241}Am source. The uniformity of the foil was measured in two perpendicular directions also using α -particle energy loss. After the thickness determination, about 0.5 mg/cm² natural silver was evaporated onto one side of the foil. Before the measurements, the composite target was kept in vacuum or an argon atmosphere to avoid contamination or oxidation of the surfaces. A thick Ta foil with a thin natural Ag layer evaporated onto one side was used as a beam catcher, positioned near the gamma ray detector. This provided unshifted Ag gamma-ray energies which, recorded simultaneously with the Doppler shifted lines were used as references to measure the energy shifts. The gamma-ray lines from Coulomb excited levels on Au and Ta were also seen in the coincidence spectra as chance events and used for calibration of the energy scale. A typical gamma-ray spectrum in coincidence with the backscattered beam particles is shown in Figure 1. Most of the lines in this spectrum, including those from Ag stopped in Ta, are removed by subtracting the spectrum produced only by chance coincidences. In order to obtain the recoil energy from the gamma-ray energy shifts, we must include a series of corrections due to the angular distribution of the scattered projectiles and of the gamma rays on the detector face, energy dependence of the efficiency of the gamma-ray detector and the relativistic effects in the solid angles viewed from the source reference system. These corrections are described in detail in Ref. [7]. The average energy of the recoiling ions, before entering the stopper material, was determined by careful simulation of the processes involved, including the angular dependence of the Coulomb excitation process, solid angles and target thickness [6].

The experimental energy losses, obtained at four different initial Ag energies, are shown in Table 1. The uncertainties shown in the energy losses are mainly from the centroid determinations of the shifted and unshifted gamma-ray peaks. As the relative uncertainties in the energy loss increase with the inverse of the energy difference between the shifted and unshifted peaks, a quite thick stopper foil had to be employed. In this way, a large fraction of the initial energy is lost in the stopper (about 50%) and the measured $\Delta E/\Delta x$ cannot be related to the stopping power at the average energy. The data analysis was carried out by using a reasonable parameterization for both the electronic (S_e) and the nuclear (S_n) stopping powers in the range of energy of the measurements and relating the experimental values of the stopper thickness Δx , the measured energy loss ΔE and the initial energy E_i by the integral:

$$\Delta x(E_i, \Delta E) = \int_{E_i}^{E_i - \Delta E} \frac{dE}{S_e(E) + S_n(E)}$$

In a similar way, using the fitted S_e , ΔE_{fit} corresponding to the loss in the stopper can be obtained. Since, in the energy region of the measurements, the contribution of S_n to the stopping is less than 5%, the predicted S_n from [2] was used. For S_e , we adopted the same procedure used in [3], where the electronic stopping is obtained by scaling the stopping power of Au for protons with the effective charge of the Ag ions as a function of its velocity. We then introduced multiplying factors in three of the coefficients for the proton curve of these calculations, which were found to be the most influential in the energy region related to the present measurements. The actual function used for protons in [3] including the multipliers F_1 , F_2 and F_3 is:

$$S_e^{Low}(E) = 1.041 E^{0.00405} + F_1 \cdot 5.61 E^{F_2 \cdot 0.439}$$

$$S_e^{High}(E) = \frac{F_3 \cdot 3646}{E^{0.869}} \ln\left(\frac{2878}{E} + 0.0139E\right)$$

with

$$S_e = \frac{S_e^{Low} S_e^{High}}{S_e^{Low} + S_e^{High}}$$

The best values for these multipliers were found in a least-square procedure for Δx as a function of $(E_i, \Delta E)$ in the integral shown above: $F_1 = 0.87$ (3), $F_2 = 1.00$ (1), $F_3 = 1.46$ (8). In Figure 2a we show the experimental values of the energy loss as a function of the initial energy of the Ag ions, and also the prediction of both the adjusted curve (solid line) and the original parameterization of [3] (dashed line). The predicted ΔE 's obtained with the coefficients from [3] are also shown in Table 1.

2.2 Measurements with the elastic scattering method

The experimental set up for the measurements using the elastic scattering technique is shown in Figure 3. The Si detector for the scattered primary

beam (Det1) was positioned at $\theta_{sc} = 60^\circ$ with a total angular aperture of about 2° . For ^{16}O and ^{28}Si primary beams, the corresponding Ag recoil angle (θ_{rec}) was about 55° . The stopper foil was placed in a mobile holder, and could be moved into or away from the secondary beam direction. The energy of the recoiling silver ions was also measured with a Si detector (Det2), with and without the stopper foil intercepting the recoiling Ag ions. The relation of the converted pulse height to the recoil energy was found to be linear in the region of the measurements, even if the effect of the detector's dead layer plays an important role for these low energy particles. For measurements in a larger energy range, non-linear calibration curves may be necessary. The correct kinematic angle for the secondary beam was found by experimentally determining the maximum of the angular distribution of the recoils, measured without the stopper foil.

In order to calibrate the energy scale of the recoils detector, the energies of the recoiling Ag ions were calculated using a Monte Carlo procedure: First, the scattering of the beam is sorted at a random angle (within the scattering detector solid angle) and position in the target. Then the energy loss of the beam up to that point, the recoil energy and angle of the Ag ions are calculated. Finally, the electronic energy loss and the Ag-Ag elastic collisions in the target are followed using the formalism described in [8]. In these calculations, the electronic stopping power and scattering function from [2] were used. With the target thickness usually employed ($\sim 80 \mu\text{g}/\text{cm}^2$), these corrections were rather small and not critically dependent on the accuracy of the stopping power data used in the simulations. We also simulated the dependence of the energy of the Ag ions, after passing the stopper foil, as a function of the average multiple scattering angle (see Figure 3). Since the detector for the recoils cover only a small fraction of the total angular distribution, the measured energy should be smaller than the average including all angles. The simulation has shown that this difference is less than 1% for the worst case (lower energy). A typical spectrum for the recoils detector is shown in Figure 4. Since for the present case the scattering and recoil angles are almost identical, due to the angular spread of the recoils, the reverse solution (with the scattered primary beam coming to this detector and the recoil to the scattering detector) is also seen.

In the measurements using this method, the thickness of the Au stopper, $0.62 \pm 0.02 \text{ mg}/\text{cm}^2$ is much smaller than that of the one employed with the Doppler shift technique. With the smaller stopping power for the lower energies used in the measurements, the energy losses in the stopper are now much smaller, so the differential approach ($\frac{\Delta E}{\Delta x} \simeq \frac{dE}{dx}(\bar{E})$) could be used.

The results of the measurements of the stopping of Ag in Au using this technique are shown in Table 2. In Figure 2b, the experimental data are shown along with the predictions of the ZBL program, using both the original parameter set and also the adjusted parameters obtained for the measurements

using the Doppler shift method, at higher energies. A single experimental point available in the literature [4] is also shown in this Figure.

Since the energies are directly measured in the method using elastic scattering, the results here are of much better quality than those based on the Doppler shift. With a typical beam current (1-2 pA), about 1-2 hour of data collection can give better results than those with the Doppler shift method, where normally 4-6 hours of measurement are necessary for each experimental point. The data for this second method were taken in two different measurements, labeled I and II in Table 2. It should be noted that this method provides a very reliable way to measure the angular distribution of the secondary beam, after traversing the stopper foil, by measuring the yield of recoiling ions as a function of the recoiling angle. These measurements may give important information about the process of multiple elastic scattering, for which almost no experimental data are available in the literature. Measurements of this angular distribution are in progress and will be reported later.

3 Discussion and conclusions

In this work, we present new data for the stopping power of gold for silver ions for energies in the range 7 - 50 MeV corresponding to velocities between about $1.6v_o$ and $4.4v_o$, where v_o is the Bohr velocity. The data were taken using two different techniques, and excellent agreement between the two data sets was found, in the sense that the parameterized curve obtained for the data set at higher velocity, reproduces quite well the data at lower velocity (see Figure 2). In the velocity region of the present measurements, the mechanisms of energy loss are expected to depend on the details of the electronic density in the periphery of the atom at the crystalline site. This causes an oscillatory dependence of the electronic stopping cross section on the atomic number of the stopper element, making it difficult to extrapolate the data from other media. Thus, failure of the semiempirical formulation is not unusual in this velocity region. This can be observed when comparing experimental data with the predictions from [3]. These calculations are one of the most used and include the oscillatory aspect mentioned above. The trends observed for the present measurements (see Figure 2) can be compared with those for other measurements in the same velocity region. In [9], for example, the data for light ions in several stopper elements show similar results to those of the present work. There is good agreement with the ZBL predictions at lower velocities, the experimental points deviating towards higher values as energy increases. In other measurements, such as the data for Nd in Pb from [10], show stopping power values larger than the ZBL predictions for all measured velocities. On the contrary, the Si in Al data from [11], and Ag in Gd [6], experimental data are below the ZBL predictions. These examples illustrate

the difficulties for prediction of the stopping powers in this velocity region. A much more extensive experimental data base is necessary in order to construct more reliable semiempirical predictions for the low velocity region.

The technique based on the elastic scattering has shown to produce better results than those based on the Doppler shift, and also uses less measurement time. A second and important aspect related to this new technique is that it makes possible the measurement of the angular distribution of the recoils after passing through the stopper foil, allowing the study of the multiple elastic scattering mechanism.

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E_i	v/v_o	ΔE_{exp}	ΔE_{fit}	ΔE_{ZBL}
35.1	3.62	17.3 (6)	17.3	16.6
41.3	3.93	19.4 (5)	19.8	18.6
46.3	4.16	21.6 (4)	21.8	20.0
52.3	4.42	24.4 (6)	24.0	21.6

Table 1

Results from the measurements using the Doppler shift technique. All energies are in MeV . E_i are the Ag initial energy and v/v_o the corresponding velocity relative to v_o . ΔE_{exp} the measured energy loss in the Au foil. ΔE_{fit} and ΔE_{ZBL} are the energy losses obtained with the fitted and original ZBL[3] functions (see text). The energy losses were measured with a stopper foil 2.02 mg/cm² thick.

E_i	Exp.	\overline{E}	v/v_o	$\frac{dE}{dx}_{exp}$	$\frac{dE}{dx}_{fit}^{DS(a)}$	$\frac{dE}{dx}_{ZBL}^{(b)}$
8.4	I	7.20	1.64	3.10 (12)	2.95	3.15
10.3	II	9.14	1.85	3.67 (15)	3.54	3.77
11.8	I	10.3	1.96	4.05 (16)	3.92	4.14
11.9	II	10.6	1.99	4.07 (16)	4.01	4.26
13.3	II	11.9	2.11	4.40 (18)	4.45	4.67
13.9	I	12.2	2.13	4.52 (18)	4.54	4.76
15.9	I	14.0	2.29	5.11 (20)	5.12	5.29

Table 2

Stopping power of Au for Ag ions: results from the experiment using the elastic scattering method. All energies are in MeV and stopping powers in MeV.cm²/mg. The v/v_o corresponding to \overline{E} is also shown. The two series of data are labeled Exp. I and Exp. II. The stopper foil used for both series was 0.62 mg/cm² thick. Most of the uncertainty in the experimental stopping power comes from the stopper thickness. (a) Using the ZBL program [3], but with the fitted coefficients obtained from the Doppler shift data. (b) from [3].

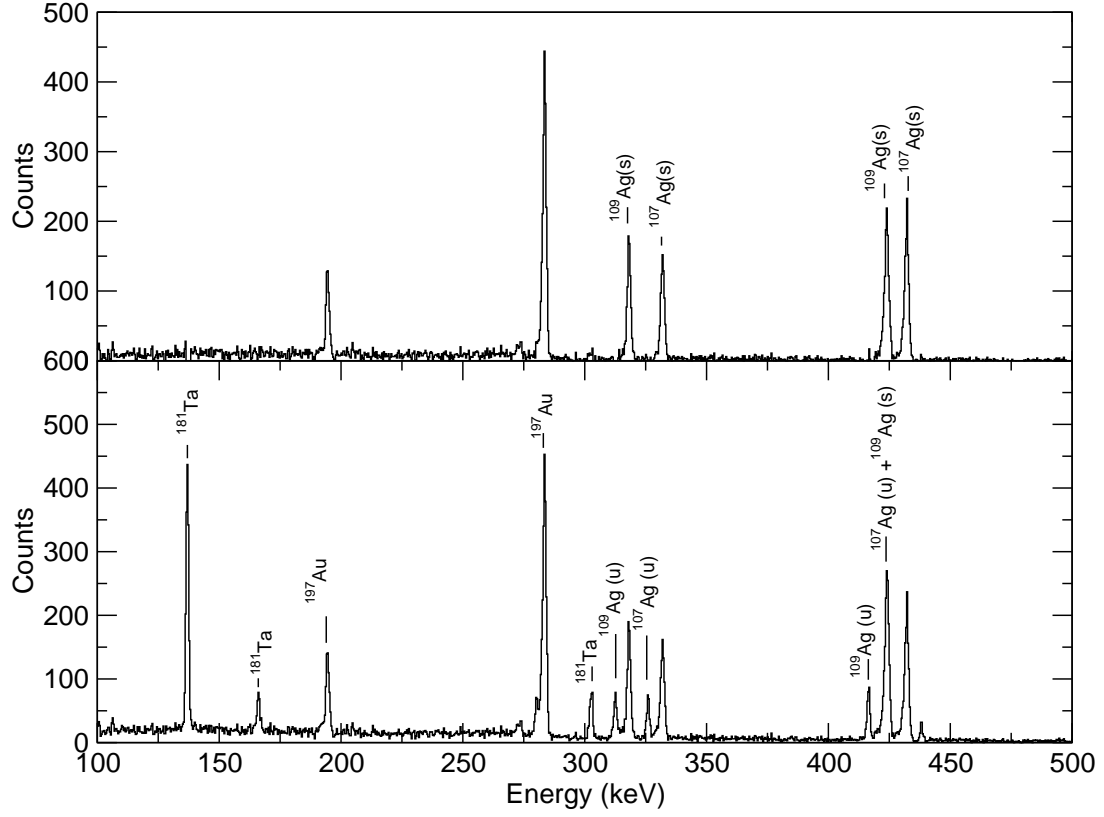


Figure 1. Gamma ray spectrum showing shifted (s) and unshifted (u) peaks in coincidence with backscattered beam particles before (bottom) and after (top) chance subtraction, where only the lines from $^{107,109}\text{Ag}$ and from ^{197}Au remain.

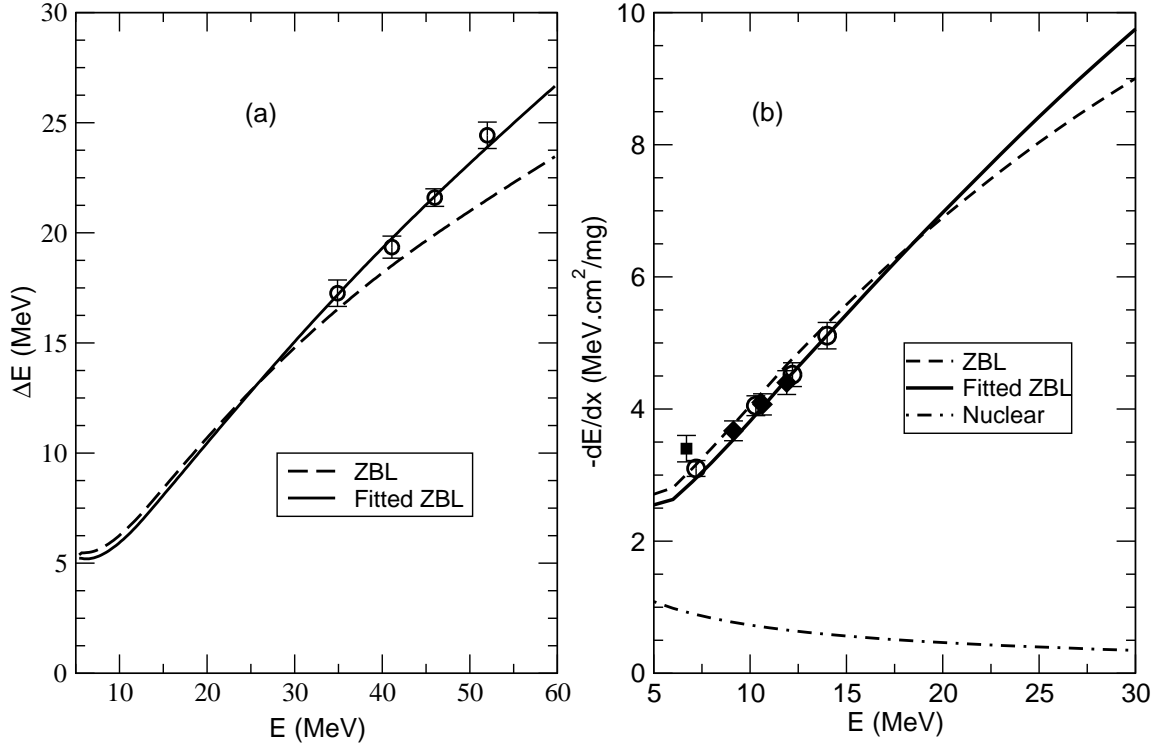


Figure 2. (a) Energy loss in the Au target, measured with the Doppler shift method, as a function of the initial Ag energy. The solid curve corresponds to the fit of the ZBL parameterization to the experimental points. (b) Circles and diamonds: data obtained with the elastic scattering technique, labeled Exp. I and Exp. II in table 2 respectively. Square: datum from [4]. These data are compared to the fitted function (see text) and to the ZBL predictions [3].

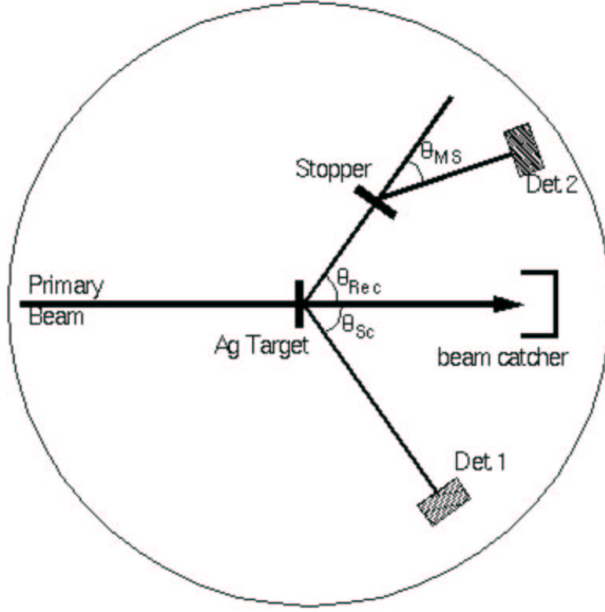


Figure 3. Geometry employed for the elastic scattering technique. The angle θ_{sc} is fixed at 60° and θ_{rec} is taken from kinematics. θ_{ms} can be varied in order to measure the angular distribution after multiple scattering. For the energy-loss measurements, the $\theta_{ms} = 0$ can be precisely found by measuring the maximum of the distribution, without the stopper foil.

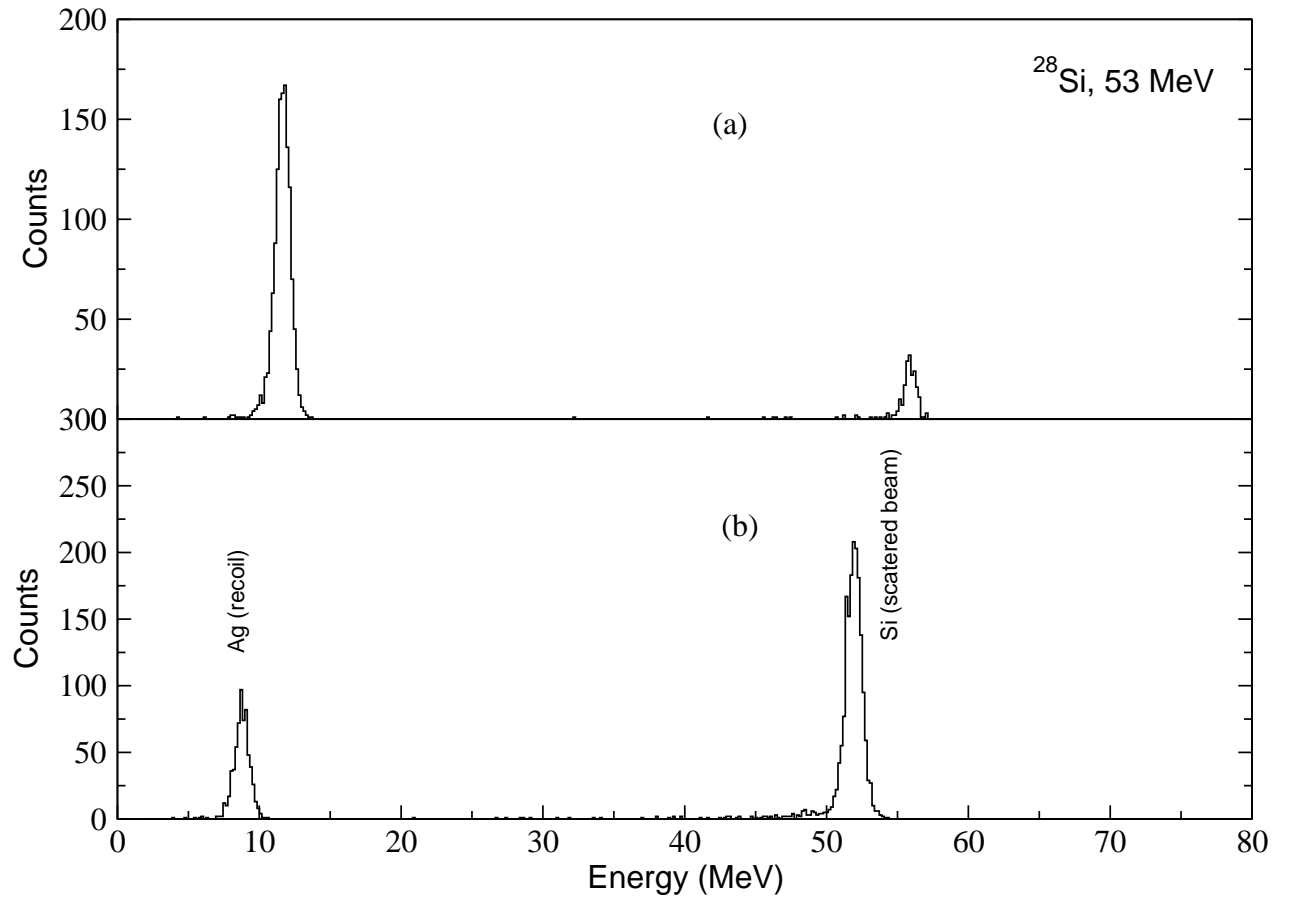


Figure 4. Typical spectra for the energy of the particles in the recoil detector with out (a) and with (b) the stopper foil. Since the recoil and scattering angles are close, due to angular straggling the scattering peak is also seen in the recoil detector (see text).