Electron screening in $^1$H($^7$Li,$\alpha$)$^4$He reaction

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Abstract: We have measured the cross section for the $^1$H($^7$Li,$\alpha$)$^4$He reaction at lithium beam energies from 0.3 to 1 MeV. Hydrogen was forced by diffusion into Pd and PdAg alloy foils. A large electron screening effect was observed only at low hydrogen concentrations. A dependence of the screening potential on Hall coefficient of the metallic host could not be established.

Due to Coulomb repulsion the cross section $\sigma$ for charged particle induced nuclear reactions drops rapidly with decreasing beam energy. To separate the strong energy dependence the astrophysical S factor is introduced. The cross section is then written as a function of c.m.s. energy $E$ as

$$\sigma(E) = \frac{S(E)}{E} e^{-2\eta},$$

where $\eta=Z_1Z_2e^2/4\pi\varepsilon_0\hbar\sqrt{2E/\mu}$ is the Sommerfeld parameter, $Z_1$ and $Z_2$ the charge numbers of interacting nuclei and $\mu$ their reduced mass. In this way all nuclear interactions are described by $S(E)$, which usually only slowly varies with energy. It is known that the cross section increases at low energies when the interacting nuclei are not bare, i.e. are in the form of atoms and molecules or in plasma [1]. The enhancement ratio could be written as

$$f(E) = \frac{\sigma(E + U_e)}{\sigma(E)},$$

where $U_e$ is the screening potential energy. It was recently observed by two independent groups that the cross section for fusion of two deuterons increases even more when deuterium is implanted into a metal [2,3]. A similar increase was subsequently observed in other nuclear reactions [4,5]. The cross section increase was attributed to metallic valence electrons, which may come closer to the deuteron and more effectively screen its charge than in a hydrogen atom. However, the size of the screening effect strongly depends on the host material and the reason for this dependence is not known. Raiola et al. [6] have observed a connection between $U_e$ and the Hall coefficient of the metallic host, while Kasagi [2] suggested that $U_e$ depends on deuterium concentration in the metal. To further investigate electron screening in metals we tried to test both hypotheses by studying the effect in Pd and PdAg alloys at different hydrogen concentrations.

To simplify the experiment we employed the inverse kinematics reaction $^1$H($^7$Li,$\alpha$)$^4$He and measured emitted $\alpha$ particles at a backward angle of 150°. In inverse kinematics the reactions occur on average deeper in the target at the same center of mass energy as in normal kinematics. In this way we were less sensitive to surface contamination and could avoid ultra high vacuum conditions necessary in the $^2$H+$^3$H reaction. The $^7$Li beams with energies between 300 and 4300 keV were accelerated by the Tandetron accelerator at Jožef Stefan Institute. To detect $\alpha$ particles we used a 100 µm thick silicon detector with an area of 300 mm² placed 42 mm from the target. The solid angle $\Omega$ embraced by the
detector was about 1% of $4\pi$. The detector was covered by a 3 $\mu$m thick Al foil to prevent scattered beam particles from hitting the detector. The metallic targets were 100 to 150 $\mu$m thick. The summary of all used targets is given in table 1. Hydrogen was absorbed into the metal due to a pressure gradient from the target back side. Typically, a hydrogen pressure of 1.0 bar was applied from the back, while $1\cdot10^{-7}$ mbar was maintained at the front of the target and concentrations varied from about 0.01 to 0.37 protons per metallic atom. Hydrogen concentrations were determined by elastic recoil detection analysis (ERDA), where the target was tilted by 75° and elastically scattered protons measured at 30° with respect to the beam [7]. Polymide (Kapton) was used as a reference for both cross section and ERDA measurements [8]. Since it is known that Kapton loses hydrogen upon irradiation with light ion beams, only $1.7\cdot10^{14}$ $^7$Li ions/cm$^2$ were allowed to hit the target. After reaching this beam dose, the target was changed. Examples of ERDA spectra taken at 4.3 MeV beam energy are shown in fig. 1. Different hydrogen concentrations in various metallic targets were achieved in the following way. The targets were left in hydrogen gas at 1 bar and room temperature for various times, from 24 hours for the high concentration Pd target to less than an hour for the Pd$_{51}$Ag$_{49}$ target. When foils were put into vacuum and pressure was applied from the back side, their hydrogen concentrations rose for a few days and then stabilized at the values listed in Table 1. Only when hydrogen concentration was constant we started measuring $\alpha$-particle yields. After about a week of Li bombardment hydrogen concentrations started to decrease and this was when we stopped measuring. By that time enough Li ions were implanted into the metal so that they could start influencing hydrogen penetration. The low concentration Pd target was used as we received it from the manufacturer without pretreatment. No hydrogen pressure was applied to the back of this foil. Before treatment with H$_2$ gas, all other targets had almost the same hydrogen concentration as the low concentration Pd foil, despite the fact that they were acquired from two different manufacturers. Moreover, when we unloaded hydrogen from the targets by heating to 300 °C in vacuum for several hours, the minimum concentration reached was the one listed in the last row of table 1.

All ERDA measurements showed a constant depth profile of hydrogen down to a depth to which these measurements are sensitive. The only deviation from this behavior was a peak on the surface of the foils. It was present in all foils, but since the number of atoms in this peak was relatively small, the surface peak only became clearly visible when hydrogen concentration in the bulk was low. The width of the peak in ERDA spectra coincides with the measurement resolution and therefore, the peak corresponds to a very thin layer at the surface. 4.5-$10^{16}$ atoms/cm$^2$ would correspond to 6.6 nm of a Pd crystal. The origin of the surface peak is unclear to us, since all foils had a shiny metallic surface. Unlike the bulk concentration, the surface peak remained constant at all times and the number of atoms in this peak was similar in all foils. We could not remove the surface peak by heating the foils to 300 °C in vacuum. The energy loss of lithium in 4.5-$10^{16}$ hydrogen atoms/cm$^2$ is 850 eV at 1 MeV beam energy and 500 eV at the lowest beam energy [9]. Since this is less than the uncertainty of the beam energy, the energy loss in the surface peak was neglected in our calculations.

During all measurements we continuously monitored Rutherford backscattered spectra of Li ions in a second silicon detector (RBS detector), positioned at a scattering angle of
150°. Contrary to the $\alpha$-particle detector the RBS detector was not covered with an Al foil and it detected lithium ions backscattered on heavier metallic nuclei. This was done to check the beam position at the target. These spectra were also used to check the continuous beam dose measurement with a mesh current integrator [10]. All measurements were performed at room temperature (300 K). The target temperature rose by at most 2 K at the highest beam energies due to the beam heating. Such a temperature difference is too small to significantly change hydrogen concentration in the foils. We also repeatedly measured $\alpha$-particle yields at 1 MeV beam energy, to check the hydrogen concentration in the target.

**Table 1** List of targets, their hydrogen contents per metallic atom, Hall coefficients [11], number of hydrogen atoms on the surface and fitted screening potential energies.

<table>
<thead>
<tr>
<th>Target</th>
<th>thickness [µm]</th>
<th>H concentration</th>
<th>Hall coefficient [$\cdot 10^{-11}$ m$^3$/As]</th>
<th>Surface peak [$\cdot 10^{15}$ atoms/cm$^2$]</th>
<th>$U_e$ [keV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kapton</td>
<td>50</td>
<td>C$<em>{22}$H$</em>{10}$N$_2$O$_5$</td>
<td>-</td>
<td>-</td>
<td>&lt;0.6</td>
</tr>
<tr>
<td>Pd</td>
<td>100</td>
<td>0.37(1)</td>
<td>-7.7</td>
<td>-</td>
<td>&lt;0.3</td>
</tr>
<tr>
<td>Pd$<em>{77}$Ag$</em>{23}$</td>
<td>125</td>
<td>0.149(5)</td>
<td>-20</td>
<td>45(5)</td>
<td>&lt;0.7</td>
</tr>
<tr>
<td>Pd$<em>{55}$Ag$</em>{25}$</td>
<td>150</td>
<td>0.023(1)</td>
<td>-21</td>
<td>20(1)</td>
<td>2.2±1.1</td>
</tr>
<tr>
<td>Pd$<em>{51}$Ag$</em>{49}$</td>
<td>100</td>
<td>0.0155(5)</td>
<td>-35</td>
<td>23(1)</td>
<td>4.1±1.4</td>
</tr>
<tr>
<td>Pd</td>
<td>100</td>
<td>0.0058(3)</td>
<td>-7.7</td>
<td>20(1)</td>
<td>&lt;10</td>
</tr>
</tbody>
</table>
Figure 1: ERDA spectra of a) Kapton b) high concentration Pd foil and c) low concentration Pd foil as a function of detected proton energy. The beam dose in c) was ten times the one in a) and b). The calculations drawn with solid lines were done with the SIMNRA code [12] using cross sections from ref. [13]. The perpendicular distances \( d \) from the surface are drawn with dashed lines.
Sample $\alpha$-particle spectra taken at a) 340 keV Li energy in Kapton and b) 1 MeV Li energy in high concentration Pd foil.

Sample $\alpha$-particle spectra taken at beam energy of 1 MeV in Pd and at 340 keV in Kapton targets are shown in fig. 2. Measured thick target $\alpha$-particle yields are shown in figs. 3 and 4 together with yields calculated as follows. A textbook definition of reaction cross section for a thin target is

\[ N_\alpha = 2\Omega WN_{\alpha} \frac{\rho N_{\alpha} x}{M} \sigma, \]

where $N_\alpha$ is the number of detected $\alpha$ particles, W their angular distribution, $N_{\text{Li}}$ the number of Li ions, $N_A$ Avogadro's number and $\rho$, $x$ and $M$ the density, thickness and molar mass of the target. The factor of 2 comes from two equivalent $\alpha$ particles produced in the reaction. $\rho N_A x/M$ represents the number of hydrogen atoms in Kapton or metallic
target in an area hit by the beam. For the surface peak this factor is given in column 5 of table 1. The contribution of the surface peak to the $\alpha$-particle yield was evaluated using the above equation. However, the contribution of the hydrogen distributed below the surface of the metal had to be calculated by transforming eq. 1.3 into differential form and integrating over energies from the beam energy $E_0$ to 0.

$$N_\alpha = 2N_{Li} \frac{\rho N_A}{M} \int_{E_0}^{0} \rho \frac{\sigma(E)}{dE_{Li}} dE_{Li}.$$  \hspace{1cm} (1.4)

Stopping power $dE_{Li}/dx$ was calculated using SRIM [13], except for Kapton, where it was taken from ref. [14]. The astrophysical S factor was taken from ref. [4] as

$$S(E) = 0.055 + 0.21E - 0.31E^2[MeV \cdot b],$$  \hspace{1cm} (1.5)

where $E$ is in MeV. The $\alpha$-particle angular distribution was taken from ref. [15]. Electron screening effect was taken into account by replacing $\sigma(E)$ with $\sigma(E+U_e)$. Therefore, the bare nuclei cross section was taken from ref. [4] and the only free parameter in the fit was $U_e$. The $U_e$ resulting from one parameter least squares fits to the data are summarized in table 1. The yields measured at 1 MeV were not included in the fits, because $S(E)$ was only determined up to the c.m.s. energy of 83.3 keV [4], which corresponds to Li beam energy of 0.7 MeV. As can be seen from the table and from fig. 3, the measured yields in Kapton as well as high concentration Pd and Pd$^{77}$Ag$^{23}$ foils can be well described with calculations for bare nuclei. The screening potential energy in these targets is too small to be stated statistically different from zero by our measurements. This is consistent with normal atomic screening observed in ref. [4] for the insulator Li$_2$WO$_4$, where $U_e=0.185\pm0.15$ keV was measured.
Figure 3 Lithium beam energy (in laboratory frame) dependence of thick target yields for Kapton, Pd and Pd\textsubscript{77}Ag\textsubscript{23} targets. Solid lines represent calculations for bare nucleons in the respective targets.

On the other hand, the Pd\textsubscript{75}Ag\textsubscript{25} and Pd\textsubscript{51}Ag\textsubscript{49} foils exhibit a large screening effect characteristic for conductors [2-4]. As shown in fig. 4, \(U_e=2.2\pm1.1\) and \(4.1\pm1.4\) keV were fitted for these two targets, respectively. This is in fair agreement with the value of 3.8±0.3 keV measured for the Pd\textsubscript{99}Li\textsubscript{1} alloy in ref. [4]. To better illustrate the difference between small and large electron screening we plot yield enhancement factors in figs. 5 and 6. These are ratios between measured and calculated yields for bare nuclei. The calculated enhancement factors for the Pd\textsubscript{75}Ag\textsubscript{25} and Pd\textsubscript{51}Ag\textsubscript{49} foils are also included in fig. 6. To analyze the \(U_e\) fits in detail we list in table 2 \(\chi^2\) for every measured point included in the fit and for both targets showing a large screening effect.
Table 2 List of $\chi^2$ for each measured beam energy for both $U_e=0$ and $U_e$ at the minimum $\chi^2$ and for both targets with nonvanishing $U_e$.

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>Pd$<em>{75}$Ag$</em>{25}$ target</th>
<th>Pd$<em>{51}$Ag$</em>{49}$ target</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\chi^2(U_e=0)$</td>
<td>$\chi^2(U_e=2.2$ keV)</td>
</tr>
<tr>
<td>850</td>
<td>5.84</td>
<td>2.43</td>
</tr>
<tr>
<td>646</td>
<td>0.04</td>
<td>0.02</td>
</tr>
<tr>
<td>545</td>
<td>0.78</td>
<td>3.10</td>
</tr>
<tr>
<td>443</td>
<td>0.71</td>
<td>0.07</td>
</tr>
<tr>
<td>392</td>
<td>0.06</td>
<td>0.05</td>
</tr>
<tr>
<td>341</td>
<td>4.70</td>
<td>3.98</td>
</tr>
<tr>
<td>sum</td>
<td>12.12</td>
<td>9.65</td>
</tr>
</tbody>
</table>

It is clear that without the lowest energy points a large $U_e$ value would not be strongly supported. However, we found no reason to exclude the lowest energies from our fits. We found improper background subtraction as the only possible way to systematically push the lowest energy points upwards. Therefore, we thoroughly investigated background in the 5 to 10 MeV range in our $\alpha$-particle detector. By measuring background in our vacuum chamber over several months we found that it was very stable and that we made the background subtraction correctly. Moreover, both nonvanishing $U_e$ are at least 2 standard deviations ($\sigma$) above zero. When measurements have a gaussian distribution only 4.5% of them fall outside the 2$\sigma$ limit. Therefore, we could say that our measurements support the two nonvanishing $U_e$ at the 95% confidence limit. The uncertainties of $U_e$ are the ones given by the least squares method, since all uncertainties arising from uncertain hydrogen concentration or background were taken into account in $\alpha$-particle yields.

Large electron screening effect is not evident in the low concentration Pd target. This is most likely because it is masked by the relatively large number of hydrogen atoms at the surface of the target. Our calculation shows that the surface contribution to the yield at the lowest measured energy is more than a factor of five larger than that of the hydrogen in the bulk of this foil. At the same time the large error bar from the $U_e$ fit shown in table 1 would still allow large screening.
Figure 4 Lithium beam energy (in laboratory frame) dependence of the thick target yield for a) the Pd$_{75}$Ag$_{25}$ and b) Pd$_{51}$Ag$_{49}$ targets. Solid lines represents the calculations for bare nuclei while for the dashed lines the screening potentials of 2.2 and 4.1 keV were taken into account in a) and b), respectively.
Figure 5 Yield enhancement factors for Kapton, high concentration Pd and Pd$_{77}$Ag$_{23}$ foils. As in figs. 3 and 4 the energy scale corresponds to the energy of the lithium beam in the laboratory frame.

Our results lead us to several conclusions. Firstly, they show that the large electron screening effect does not occur on metallic surface but it does so only inside the metal. The $^1$H($^7$Li,$^\alpha$)$^4$He reaction in inverse kinematics happens on average much deeper in the metal compared to previously studied reactions [2-4], but anyway our results qualitatively agree with previous measurements. However, we have shown that the effect depends on the concentration of hydrogen in the metal. Large screening is only visible at concentrations of a few percent, while at higher concentrations the effect disappears. This is consistent with the suggestion by Kasagi [2] who observed the largest effect in PdO at the lowest hydrogen concentration. Our screening potentials of 2.2±1.1 and 4.1±1.4 keV obtained at 2.3 and 1.6% hydrogen concentrations, respectively, compare well with $U_e$=3.8±0.3 keV [4] measured at 1% and $U_e$=1.5±0.3 keV [2] at 7% lithium content in PdLi$_x$ alloys. This further corroborates the concentration dependence of the electron screening in metals.
In the presence of concentration dependence it is difficult to talk about the dependence of the screening potential on Hall coefficient $R_H$. According to Rolfs [3], $U_e$ should be inversely proportional to the absolute value of $R_H$. Our measurements could not confirm this claim. We chose Pd and PdAg alloys since their crystal structures and electron densities are very similar, yet $R_H$ varies by almost a factor of five. We expected the largest screening in Pd and smallest in Pd$_{51}$Ag$_{49}$ alloy, but we observed just the opposite. However, it must be said here that it is known in PdAg alloys Ag blocks octahedral sites for hydrogen occupation and, therefore, hydrogen may even in an alloy feel only the Pd neighbors. It should also be mentioned here that Raiola et al. [6] have measured a different value of the Hall coefficient in Pd than previously accepted [11,16].

References: