PROCEEDINGS OF SCIENCE

Enhanced d(d,p)t cross section in metallic environments

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Understanding the electron screening effect in the laboratory is critical for a correct interpretation of low-energy nuclear reactions in stars. Extensive studies of the electron screening effect in deuterated metals (54 metals) and other environments have been carried out in Bochum in the last 4 years. Experimental results of anomalous enhancements have been interpreted in terms of the Debye plasma model applied to quasi-free metallic electrons. For the d(d,p)t reaction in metallic environments, the variation of hydrogen solubility in the samples as a function of temperature has also been measured, showing an anti-correlation with screening enhancement as expected. Within this model, the deduced number of valence electrons per metallic atom also agrees with the corresponding number from the Hall coefficient. Recently, the expected temperature dependence of the screening potential has been verified together with the expected Z_t scaling (with the metallic host) of the Debye radius. Recently results on the ^{6,7}Li(p,α) reactions in Li insulators, Li metal and PdLi alloys confirm the expected behaviour of the screening effect and further supports the applicability of the Debye model.

International Symposium on Nuclear Astrophysics - Nuclei in the Cosmos - IX
25-30 June 2006
CERN

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PoS(NIC-IX)169

1. Introduction

Thermonuclear reactions between charged particles in stellar plasmas take place at sub-Coulomb energies and their cross sections $\sigma(E)$ drop exponentially with decreasing energy according to the equation $\sigma(E) = E^{-1}S(E)\exp(-2\pi\eta)$, where E is the centre-of-mass energy between the interacting nuclei, $\eta = Z_1 Z_2 e^2/\hbar v$ is the Sommerfeld parameter (in standard notations) and $S(E)$ is the astrophysical S-factor. The exponential factor accounts for the quantum-mechanical probability of tunneling through the Coulomb barrier as seen by bare nuclei. However, both in the stellar plasma and in the laboratory the actual charges are shielded by the presence of the surrounding electrons so that the Coulomb barrier is effectively reduced both in height and radial extension. This in turn leads to a higher cross section for the screened nuclei, $\sigma_s(E)$, than would be the case for bare nuclei, $\sigma_b(E)$. The enhancement factor can be expressed in terms of the electron screening potential U_e as [1]:

$$f_{lab} = \frac{\sigma_s}{\sigma_b} \approx \exp\left(\pi\eta(E)\frac{U_e}{E}\right), \tag{1.1}$$

for $S_b(E+U_e) \approx S_b(E)$ and $U_e/E \leq 0.1$.

In a stellar plasma a similar enhancement factor can be defined which however depends on the temperature and density conditions through the Debye-Hückel radius R_D of the electron cloud surrounding the fully ionized particles (see later). So far, no measurement has been carried out in a plasma to determine the appropriate electron screening enhancement. However, similar effects can be observed in metals as described in the following sections.

Theoretically, the electron screening potential U_e can be calculated, for example, in the adiabatic limit from the difference in atomic binding energies between the compound atom and the projectile plus target atoms of the entrance channel (i.e. 27 eV).

Experimentally, the determination of the screening potential is based on a comparison between the bare (S_b) and the screened (S_s) S-factors. The former is obtained as an extrapolation of experimental data at energies around the Coulomb barrier where the shielding effect can safely be neglected. By contrast, measurements at energies well below the Coulomb barrier provide directly the screened $S_s(E)$-factor. Expressing the enhancement in terms of the S-factor, i.e. $f_{lab} = S_s/S_b$, a fit to these latter data allows for the determination of U_e, as free parameter, according to the eq.1.1.

In recent years an extensive investigation of the electron screening for the d(d,p)t reaction has been carried out in Bochum on deuterated metals, semiconductors and insulators, i.e. 58 elements in total [3], [4] (previous experiments from other groupsother experimental data [5]-[7]). For insulators and semiconductors U_e values in agreement with gas D_2 target experiments $U_e = 25$ eV [9] were found, whereas U_e values typically one order of magnitude larger have been observed for metals.

2. Experimental procedure and data analysis

Details of the equipment, procedures and data analysis can be found in [4]. Briefly, the 100kV accelerator of the Dynamitron Tandem Laboratorium in Bochum at the Ruhr-Universität Bochum provided deuteron beams with currents up to 50-60 μA at the lowest energies (E_d=5 keV). Target preparation involved the cleaning of a fresh material "M" by in-situ Kr sputtering. The deuterated

targets were then produced by bombarding the cleaned sample with a deuteron beam at a given energy until a saturation level was achieved, as measured by the proton yield from the d(d,p)t reaction. The implantation procedure was repeated over the energy range of the planned measurement typically taking about 4 days of running. At the end of the implantation a stoichiometry M_xD was attained at the sample surface and an uniform deuteron distribuition target was produced (i.e. ERDA analysis). The deduced thin-target yield curve, $Y(E_d,\theta)$, was obtained as the difference in the thick-target yield for two adjacent points and divided by the energy step. The result is related to the cross-section $\sigma(E_{eff})$ via $Y(E_d,\theta) = \beta\varepsilon_{eff}(E_d)^{-1}\sigma(E_{eff})$, with the effective energy E_{eff} [1] and the constant β, as measured using a radioactive source. The effective stopping power $\varepsilon_{eff}(E_d)$ for the M_xD target is given by the expression $\varepsilon_{eff}(E_d) = \varepsilon_D(E_d) + x\varepsilon_M(E_d)$; the deduced energy dependence of $\sigma(E_{eff})$ is nearly independent of the stoichiometric ratio x. The resulting cross-section $\sigma(E_{eff})$ was obtained as the weighted average of all runs and plotted in terms of the astrophysical $S(E)$ factor, whereby the absolute scale was obtained by normalization to the bare astrophysical S-factor $S_b(E)$ ([9] and references therin). Relative to this function, the data were fitted with the enhancement factor of eq.1.1, thus allowing for the determination of the screening potential U_e. Numerical values of U_e for each material investigated, as well as their hydrogen solubility $y = 1/x$ as determined from the normalization of data to the bare $S_b(E)$ factor can be found in [4]. As compared to measurements with a gaseous D_2 target, a large screening (of the order $U_e \sim$300 eV) is observed in metals, whereas a small (gaseous) effect is obtained for insulators and semiconductors. Clearly the enhanced cross-section is due to electron effects of the environment of the target deuterons. An explanation of the large enhancement was suggested ([2], [3]) by the plasma screening of Debye applied to the quasi-free metallic electrons. The electron Debye radius around the deuterons in the lattice is given by

$$R_D = \sqrt{\frac{\varepsilon_o kT}{e^2 n_{eff}\rho_a}} = 69\sqrt{\frac{T}{n_{eff}\rho_a}} \quad [m] \tag{2.1}$$

with the temperature T of the free electrons in K, n_{eff} the number of valence electrons per metallic atom, and the atomic density ρ_a in units of atoms/m^3. With the Coulomb energy between two deuterons at R_D set equal to $U_e = U_D$ one arrives at $U_D = 2.09\times10^{-11}\sqrt{\frac{n_{eff}\rho_a}{T}}$ $[eV]$. For $T = 293K$, $\rho_a = 6\times10^{28} atoms/m^{-3}$, and $n_{eff} = 1$ one obtains a radius R_D which is about a factor 10 smaller than the Bohr radius of a hydrogen atom, hence the characteristic values $U_e \sim 300$ eV as observed experimentally.

An exception to the large enhancements observed for metals was found for the metals of group 3 and 4 of the periodic table and the lanthanides, which showed a small screening. At room temperature all these elements are characterized by a relatively high hydrogen solubility $y = 1/x$ that gives the deuterated targets of these metals the properties of insulators. Indeed, for the metals with high U_e values, their solubility was typically small (a few percent) leaving the metallic character of the samples essentially unchanged. It is known that hydrogen solubility [8] in metals decreases with increasing temperature. With the assumption that the temperature-dependent solubility $y(T)$ affects directly n_{eff}, one obtains:

$$U_D(T) = \begin{cases} \frac{1}{4.78\times10^{10}}\sqrt{\frac{[n_{eff}(T)(1-y(T))]\rho_a}{T}} \quad \text{[eV]} & \text{, y(T)} \le 1 \\ 0 & \text{, y(T)} > 1 \end{cases} \tag{2.2}$$

where a temperature dependence of $n_{eff}(T)$ is also taken into account.

In order to verify the expected Debye enhancement, all metals of groups 3 and 4 and the lanthanides have been studied at $T = 200°C$ (for details see [4]), where it was expected that the solubility reduced to a few percent and a large screening became observable. As was observed by Huke et al. (reference therein [11]): "...the higher the deuteron density, the lower the screening value".

2.1 Temperature and ion charge dependence of screening potential

According to the Debye model, lower U_e values should be expected at higher sample temperatures [4], i.e. $U_e(T) \propto T^{-1/2}$. This prediction has been explored for Pt (in the range $T = 20 - 340°C$) and for Co (at $T = 20°C$ and $200°C$), as both metals have a solubility of a few percent (i.e. low deuteron density) at all temperatures investigated. Besides, over the present temperature range, the reported Hall coefficient for Pt increases by about 20% leading to a corresponding decrease in n_{eff} (eq.2.2), which we took into account; there is good agreement between observation and expectation (see tables in [4]). As a consistency test we also studied the insulator C at $T = 200°C$: the solubility decreased from 0.35 ($T = 20°C$) to 0.15, but no enhanced screening was observed, as expected for an insulator, i.e. $n_{eff} = 0$ (eq.2.2). The data represent the first observation of a temperature dependence of a nuclear cross section. The Kr sputtering and the high vacuum (in the 10^{-8}mbar range) guarantee a sufficient clean surface target in the investigated energy range (homogeneous deuteron target by RBS analysis, see [4] for details).

The Debye radius scales inversely with the nuclear charge Z_t of the target atoms [1], $R_D \propto (Z_t(Z_t+1))^{-1/2}$, and thus $U_D \propto (Z_t(Z_t+1))^{1/2}$. To verify this dependence, we have reinvestigated the electron screening in the $^7Li(p,\alpha)\alpha$ reaction. Previous studies led to an atomic screening potential energy $U_A = 300 \pm 160$ eV [10] consistent with the adiabatic limit (175 eV [12]). For the $^7Li(p,\alpha)\alpha$ reaction with $n_{eff}(Li) = 0.8 \pm 0.2$ at $T = 20°C$ one expects $U_D = 720 \pm 180$ eV for Li metal and therefore $U_e = U_A + U_D = 1020 \pm 240$ eV, assuming a linear addition of both acceleration mechanisms. If an alloy such as $PdLi_x$ is used with a few percent Li admixture x (essentially maintaining the metallic character of Pd), one has $n_{eff}(Pd) = 6.3 \pm 1.2$ [3] and thus $U_D = 3200 \pm 310$ eV leading to the prediction $U_e = U_A + U_D = 3500 \pm 350$ eV. Kasagi et al. [7] performed studies in a $PdLi_x$ alloy (x = 5-7%) finding $U_e = 1500 \pm 310$ eV, but no explanation of this observation was given. In order to test the predictions of the Debye model, the electron screening for the $^{6,7}Li(p,\alpha)$ reactions has been studied for the different environments: LiF, Li_2O and Li_2WO_4 insulators, a Li metal, and two $PdLi_x$ alloys (details in [13]). The results of the 5 samples leads to $U_e = 400 \pm 240$, 320 ± 240, 190 ± 130, 1430 ± 110, and 3630 ± 360 eV for LiF, Li_2O, Li_2WO_4, Li, and $PdLi_{1\%}$, respectively. For the alloy $PdLi_{0.01\%}$ we find $U_e = 3020 \pm 660$ eV, consistent with the above value for $PdLi_{1\%}$. The results indicate that the metallic character of Pd remained essentially unchanged by the small Li content: weighted average $U_e = 3490 \pm 320$ eV. The insulators led to a weighted average $U_e = U_A = 250 \pm 100$ eV in agreement with previous work [10] and the atomic adiabatic limit. The observed value for the Li metal gives a Debye enhancement $U_D = U_e \sim U_A = 1180 \pm 150$ eV and thus $n_{eff}(Li) = 2.1 \pm 0.5$, in fair agreement with the value from the Hall coefficient (confirmations also from the other studied samples [4]). Similar results have been also obtained for the $^6Li(p,\alpha)^3He$ reaction (values given in [13]) and further confirm the applicability of the Debye model in describing the observed enhancements.

3. Discussion

As shown by Czerski et al. [11], the Debye-Hückel screening is applicable only for large temperature ($k_BT > E_F$) for which the electron degeneration vanishes and the Maxwell-Boltzmann statistics can be used. For low temperatures ($k_BT<E_F$) or correspondingly low projectile energies, the strong-screening limit should be applied and no temperature dependence should be observed. Besides, the dielectric function theory, presented to explain the high electron screening effect in the six deuterated target analyzed [11], supports a weak target material dependence of the screening energy as the Czeski's experimental data as showed. The theory provides, however, absolute values being by a factor of 2 smaller than the experimental ones (fig.1 in [11]). In despite of it, our extensive experimental work on the enhanced electron screening in deuterated metals can be explained quantitatively by the "Debye model" applied to the quasi-free metallic electrons. Besides, the present data for the electron screening in the $^{6,7}Li(p,\alpha)$ reactions for different environments give a consistent picture: (i) as suggested previously [10] the present data demonstrate the isotopic independence of the electron screening effect; (ii) for the Li_2WO_4 insulator the atomic electron cloud leads to one acceleration mechanism, while the Li metal and the $PdLi_x$ alloys exhibit an additional acceleration mechanism due to the quasi-free metallic electrons at the Debye radius. In comparison to the data in the d(d,p)t reaction for metals [4], the screening potential energy scales with the charge Z_t of the target nucleus, as expected from the Debye model. Previous studies of the reactions $^9Be(p,\alpha)^6Li$ and $^9Be(p,d)^8Be$ using a metallic Be target led to a high screening potential energy $U_e = 900 \pm 50$ eV [14], which was not understood at that time, but the Z_t scaling of the Debye model explain the high screening effect.

Clearly, an improved theory is highly desirable to explain why the simple Debye model, with the own theoretical limitations of applicability, appears to work so well. Without such a theory, one may consider the Debye model as a powerful parameterization of our data.

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