Experimental evidence for temperature effect on energy loss of low-energy D^+ in liquid lithium *

FANG Kai-Hong(方开洪)[1)] ZOU Jian-Xin(邹建新) LIU Dong-Dong(刘东东)
ZHAO Jiang-Tao(赵江涛) WANG Tie-Shan(王铁山)
School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, China

Abstract: In order to investigate the temperature effect on the stopping power of liquid lithium material for keV D^+, the excitation functions of the α-particle yields for the ^{6}Li(d,α)^{4}He reaction in liquid lithium (495–600 K) have been measured for the bombarding energies from 50 to 70 keV by 2.5 keV steps. The observations show that the thick-target α-yield increases statistically as lithium temperature increases. These phenomena revealed that the only possible reason is a temperature effect on the stopping power, i.e., increasing temperature resulting in a lower stopping power. As the lithium temperature increased from 495 to 600 K, the energy loss of deuterons decreased about 6.7% in the energy region of E<70 keV.

Key words: stopping power, temperature effect, liquid Lithium, d-^{6}Li reaction

PACS: 34.50.Bw, 34.90.+q, 25.45.-z **DOI:** 10.1088/1674-1137/39/8/084001

1 Introduction

The energy loss of ions through matter has long been the subject of theoretical and experimental investigation. For higher energies the Bethe theory and Bragg rule accurately predict stopping cross-sections with the aid of experimentally measured mean excitation energy. For lower energies particularly near the Bohr velocity region, the complexity of energy-loss has not been possible to precisely predict on the basis of theory alone, or even from empirical information. The challenge comes from the complexity of the electronic structure of the colliding partners, their relative colliding energy and the charge transfer process [1]. For this reason the study of the simple H^+-Li (proton-lithium) collision system would be of heuristic benefit since the single electron models are applicable due to the outermost electron (2 s) being much more loosely bound than the innermost electrons (1 s) [2]. Therefore, experimental measurements of a simple collision system are still desirable for particular topics, such as physical and chemical state [3], matter temperature [4] and pressure [5].

Since Swann [6] first noted the stopping cross-section would change between the gas and solid phase due to dielectric effects, and Fermi [7] estimated that the solid phase should have lower stopping than gas to the order of several percent by including the dielectric screening correction, the first experimental observation [8] of the physical state effect reported that the gas stopping cross-section for H^+(100 keV) was 20% greater. A summary of the phase effect can be found in Ref. [9]. To estimate the influence of temperature on stopping cross-section, Von Weiszacker theory [10] predicts a strong influence from temperature, and afterwards a few measurements [11] with solid phase, i.e., aluminum and tin as a stopping medium down to liquid helium temperature, gave negative results. In the plasma phase, however, the temperature effect has been confined to consideration of stopping cross-section in the context of Inertial Confinement Fusion [12]. Since the technical difficulties of measuring the stopping cross-section in a liquid medium in the keV energy region are nearly insurmountable, there is no extant experimental work dealing directly with the temperature dependence of stopping power with liquid phase. Rather, in the absence of any suggestions to the contrary, it is generally assumed that there is none and the effect is ignored.

However, Von Weizsacker theory [10] predicts a strong influence of temperature on the stopping power especially for light metals such as Al, Na and Li, i.e., for aluminum the decrease of stopping cross-section is expected to be 6% from 20 to 273 K. Meanwhile, Sabin's estimation [4] shows that the temperature effect should be larger when the projectile velocity is lower. In order to experimentally confirm the temperature influence on the stopping power of a light-nucleus matter for a

Received 8 December 2014
∗ Supported by National Natural Science Foundation of China (11305080, 11275085)
1) E-mail: fangkh@lzu.edu.cn

low-volocity incident projectile, we have chosen Li material as a stopping medium for keV D^+ ions. Meanwhile Li matter has a relatively low melting point and reacts with the incident projectile (D^+), i.e., ^{6}Li(d,α)^{4}He reaction in which the emitted thick-target α-yields relate to the stopping cross section.

2 Experiment

The experiment was performed using a low-energy high-current ion beam generator at the Research Center for Electron Photon Science, details of which were reported in Ref. [13] In this work, we measured the thick-target yields of α-particles emitted from the ^{6}Li(d,α)^{4}He reaction from 50 to 70 keV occurring in liquid Li medium. The measurements were performed in different temperature regions, i.e., 495±13, 532±6, 576±8 and 600±4 K.

Two Si surface barrier detectors (300 μm in thickness and 450 mm^2 in area) were employed to detect the charged particles, with a total solid angle ($\Delta\Omega/4\pi$) of 5.0%. The detectors were placed at 125° with respect to the beam direction. A thin Al foil (5 μm thick) was placed in front of the detector to prevent scattered particles from hitting the detector directly. The detector holder, made of Al, was cooled by water at 5° Celsius to avoid being heated up by thermal emissions from the target. The liquid ^{6}Li target (enrichment of 95%) was in an open container which was placed horizontally at the center of the chamber with pressure about 9×10^{-4} Pa.

As an active element, lithium is easily covered with compounds such as LiD, thus much attention should be paid to the cleanness of the target surface during the bombardment. The deterioration can be easily known for the liquid lithium phase, since the monitoring data of the radiation thermometer which was arranged to monitor the temperature of the target surface should fluctuate anomalously when contamination spots exist, while the yield of protons from the D(d,p)T reaction would increase strongly. Thus, during the measurements we kept the D/Li atomic ratio less than 0.02%. When the ratio exceeded the upper-limit value, the target surface was shaved by the scraper in the chamber. We kept the same beam power (500 mW) during the experiment, in order to keep the target conditions as similar as possible. Limited by the melting and boiling point of lithium, the present work observed the ^{6}Li(d,α)^{4}He reaction in liquid Li from 495 to 600 K.

3 Results and discussion

The thick-target charged particle spectrum measured at 70 keV is illustrated in Fig. 1, where four peaks are seen: protons (p) from the D(d,p)t reaction, protons (p_1 and p) from the ^{6}Li(d,p_1/p)^{7}Li reactions, and α-particles from the ^{6}Li(d,α)^{4}He reaction as indicated. The continuous events below 1000 channels originate from the ^{7}Li(d,α)n^{4}He reaction due to the ^{7}Li (5%) in the enriched target. The key observation object (E_α=10.9 MeV) is clearly identified, while the peak area of protons from the monitoring D(d,p)t reaction indicates the concentration of deposited deuterons during the bombardment, which was kept as $x<0.02\%$ (LiD_x) in the measurement

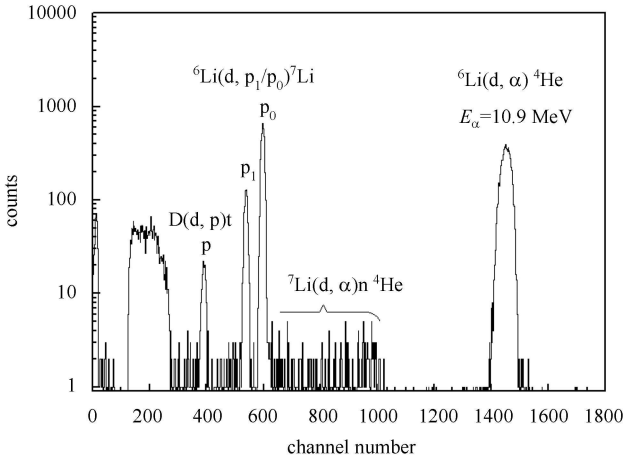

Fig. 1. Charged particle spectrum obtained by the bombardment of 70 keV D^+ on liquid lithium.

Figure 2 shows the α yields (solid dots) measured from 70 to 50 keV with unit counts per μC of the implanted deuteron. For a certain energy (the individual plots in Fig. 2), the yields obtained at four different temperatures (495±13, 532±6, 576±8 and 600±4 K) increase linearly with temperature, where the tendency is estimated by a dashed line for guidance. The thick-target α-yield ($Y_\alpha^{\rm thick}(E_{\rm d})$) is the integral of the cross section and the inverse of stopping power over the range of incident projectile, which can be expressed as follows

$$Y_\alpha^{\rm thick}(E_{\rm d},T) = 2N_{\rm d}\times0.95\rho_{\rm Li}(T)\frac{\Delta\Omega_{\rm lab}(T)}{4\pi} \times\int_0^{E_{\rm d}}\frac{{\rm d}\Omega_{\rm cm}}{{\rm d}\Omega_{\rm lab}}\sigma(E_{\rm cm},T)[\rho_{\rm Li}(T) \times S(E_{\rm d},T)]^{-1}{\rm d}E. \quad (1)$$

where $E_{\rm d}$ is beam energy in the lab system, $N_{\rm d}$ is the number of projectiles, $\rho_{\rm Li}(T)$ is the temperature-dependent number density of the target Li, $\Delta\Omega_{\rm lab}(T)$ is the solid angle subtended by the detectors (this term also shows a slight T-dependence due to the volume expansion [14] of the Li target as temperature increases), ${\rm d}\Omega_{\rm cm}/{\rm d}\Omega_{\rm lab}$ is the solid angle ratio of the c.m. system to the lab system, and $\sigma(E_{\rm cm},T)$ is the cross section [13] of the ^{6}Li(d,α)^{4}He reaction with incident energy ($E_{\rm cm}$) in the c.m. system, which might be affected by environmental temperature [15], i.e., $\sigma_{\rm screen}(E) = f[E,U_{\rm s}(T)]\times\sigma_{\rm bare}(E)$ where $f[E, U_{\rm s}(T)]$ is an enhance-

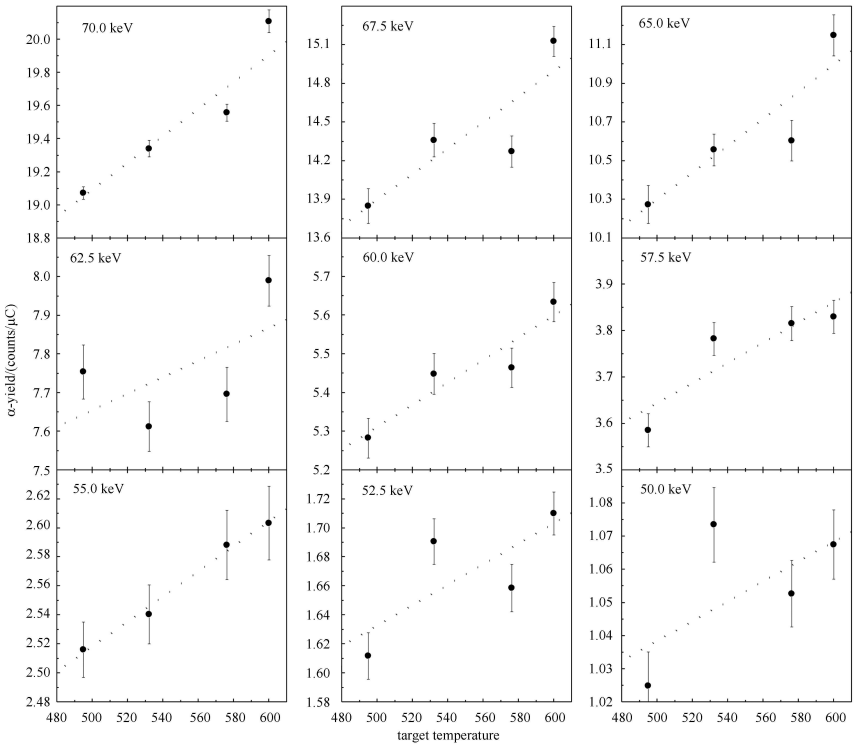

Fig. 2. α-yield from the ^{6}Li(d,α)^{4}He reaction. In each plot, the four dots are the measured values in the different temperatures (495, 532, 576 and 600 K). Nine incident energies were investigated from 70 to 50 keV. The dashed line in each plot shows the increase of the α-yield with temperature.

ment factor and $U_s(T)$ is the screening energy, $\rho_{Li}\times S(E_d,T)=dE/dx$ is the stopping power of lithium for incident deuterons, where $S(E,T)$ is the stopping cross-section. Since the effect of T-dependent $\rho_{Li}(T)$ on thick-target yields is eliminated in the calculation, as seen in Eq. (1), the residual reasons which influence the thick-target yields must originate from the solid angle ($\Delta\Omega_{lab}(T)$), nuclear reaction cross section ($\sigma(E_{cm},T)$) and stopping cross section ($S(E,T)$).

The phenomenon of expansion and contraction will change the surface location of the liquid Li target and modify the distance from the beam-spot to the detector, thus the solid angle $\Delta\Omega_{lab}(T)$ is a function of temperature. To estimate the effect of $\Delta\Omega_{lab}(T)$ change on thick-target α-yields, the related geometric parameters of the Li target and detectors as well as the density change of lithium $\rho_{Li}(T)$ [14] were involved, finding that the influence is about 0.2% as Li temperature increases from 495 to 600 K. In that sense the effect due to the change of solid angle is negligible.

Raiola [15] measured the D(d,p)T reaction in Platinum (Pt) metal with the Pt temperature changing from 20° to 340° Celsius, and found the T-dependence of cross section to be simply explained by the screening effect of quasi-free electron in Pt metal by using the plasma screening of the Debye model, which leads to $U_s\sim T^{-1/2}$. Since the increasing temperature tends to weaken the ability of electrons to gather around and screen positive charges [16], the cross section (and thick-target α-yield) should decrease as the environmental temperature increases. Therefore, the influence from $\sigma(T)$ gives a negative correlation with the observed results. Simply assuming the screening energy (i.e., $U_s\sim$700 eV at T=495 K [13, 15]) follows the Debye model, the thick-target α-yield which is the most likely to be affected in the low energy region, i.e., 50 keV, would decrease less than 1.8% as Li temperature increases from 495 to 600 K.

So far, we can confirm that the increasing α-yield must be mainly caused by the smaller stopping power as temperature increases, as seen in Eq. (1). We can make a simple assumption that the stopping power has the same energy dependence in shape as SRIM [17] but that the absolute value depends on the temperature, i.e., $S(E,T)=S_{SRIM}(E)\times A(T)$. The values of $A(T)$ were searched for which give the minimum value of χ^2, the Chi-square Test, defined as

$$\chi^2=\sum\frac{[Y_\alpha^{exp}(E_d)-Y_\alpha^{thick}(E_d,T)]^2}{[\Delta Y_\alpha^{exp}(E_d)]^2}, \qquad (2)$$

where Y_α^{exp} and ΔY_α^{exp} are the measured thick-target α-yields and corresponding statistical errors which have been plotted in Fig. 2. The deduced results show that

the stopping cross section decreased about 6.7% as Li temperature increased from 495 to 600 K, where the errors ($\Delta A(T)$) in $A(T)$ were estimated by $\chi^2_{\rm min}\pm1$, e.g., in the process of searching for the best value of A, one may find the value A^* which leads to $\chi^2=\chi^2_{\rm min}+1$, then $\Delta A(T)$ is defined by (A^*-A). The deduced values of $A(T)$ for stopping correction are plotted as the dots in Fig. 3, with the straight line given as a guide. As extrapolated in Fig. 3, the correction factor seems to be 1.0 as the temperature approaches the Li boiling point (∼620 K, in the vacuum chamber). It means that the stopping power approaches the values estimated by SRIM as the Li material comes near the lithium vapor state, however, the five-parameter empirical formula [17] obtained by fitting the experimental values mainly originated from the solid-film lithium (∼10^{18} atom/cm^2), i.e., Ref. [2].

In our experiment, the incident energy was much lower and overlapped the Bohr velocity region; considering Weizsacker [10] and Sabin's estimations [4], our observation appears to be acceptable. Nagy [12] investigated the stopping of a finite-temperature electron gas for a slow proton at the reduced temperature $\Theta(k_{\rm B}T/\varepsilon_{\rm F})$ ranging from 0 to 10. For the present temperature region ($\Theta\ll1$), the calculated result predicts that the stopping power decreases almost linearly as temperature increases, however the decrease rate is negligibly small: less than 0.1% for the temperature from 495 to 600 K. From the experimental point of view, Arnau [18] reported that the phase effect on stopping power is as much as 50% between gaseous and solid zinc targets for 50 keV/u protons. Based on our observation, we can safely conclude that the experimental evidence of a temperature effect on energy loss has been found by using the keV-energy deuteron transporting in liquid lithium medium. However a more sophisticated theory is required. Also, the accessory experiments to further validate this observation are highly desired in our future through a variety of ways, i.e., elastic scattering, charge transfer cross section, temperature dependence of sputtering yield. For instance, Gay et al. [19] observed that the secondary-electron yield from carbon foil bombarded by a 175 keV α-particle decreased about 5.5% as the foil temperature increased from 495 to 600 K.

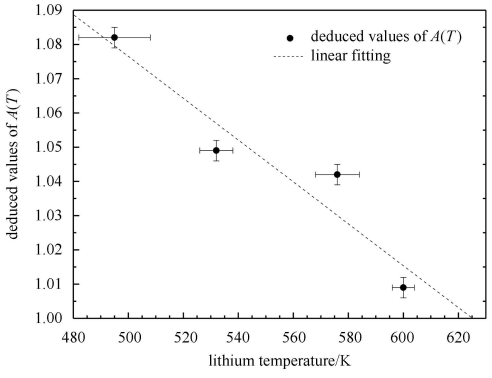

Fig. 3. Correction factor ($A(T)$) for stopping power as a function of lithium temperature. The dots are deduced values from Eq. (2), the straight line is a linear fit for guidance.

4 Conclusion

We conclude that experimental evidence of a temperature effect on energy loss has been found by using keV-energy deuterons transporting in a liquid lithium medium. As the Li temperature increased from 495 to 600 K, the energy loss of deuterons decreased about 6.7% in the energy region of E <70 keV. However, a more sophisticated theory is required. Also, supplementary experiments to further validate this observation are highly desirable and a variety of methods are considered for future work e.g., elastic scattering, charge transfer cross section, and temperature dependence of sputtering yield.

The authors thank the Research Center for Electron Photon Science in Tohoku University, Japan, for their hospitality during the experiments.

References

1 Ziegler J F, CHU W K, FENG J S Y. Appl. Phys. Lett., 1975, **27**: 387
2 Eppacher C, Muino R D, Semrad D et al. Nucl. Instrum. Methods B, 1995, **96**: 639
3 Bauer P, Kastner F, Arnau A et al. Phys. Rev. Lett., 1992, **69**: 1137
4 Sabin J R, Oddershede J, Paidarova I. Nucl. Instrum. Methods B, 1994, **93**: 161
5 Cruz S A. Nucl. Instrum. Methods B, 2004, **222**: 411
6 Swann W F G. J. Franklin Inst., 1938, **226**: 598
7 Fermi E. Phys. Rev., 1940, **57**: 485
8 Sautter C A, Zimmerman E J. Phys. Rev. A, 1965, **140**: 490
9 Thwaites D I. Nucl. Instrum. Methods B, 1985, **12**: 84
10 von Weiszacker C F. Annalen der Physik, 1933, **409**: 869
11 Gerritsen A N. Physica XII, 1946, **5**: 311
12 Nagy I, Arnau A, Echenique P M et al. Phys. Rev. A, 1991, **43**: 6038
13 FANG K H, WANG T S, Yonemura H et al. J. Phys. Soc. Jpn., 2011, **80**: 084201
14 Shimizu Y, Mizuno A, Masaki T et al. Phys. Chem. Chem. Phys., 2002, **18**: 4431
15 Raiola F, Burchard B, Fulop Z et al. J. Phys. G: Nucl. Part. Phys., 2005, **31**: 1141
16 Debye P. Polar Molecules. New York: Dover Publications, 1929
17 Ziegler J F, Biersack J P. code SRIM. http://www.srim.org
18 Arnau A, Bauer P, Kastner F et al. Phys. Rev. B, 1994, **49**: 6470
19 Gay T J, Berry H G. Phys. Rev. A, 1979, **19**: 952