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Experimental Demonstration of the Thin-Film Liquid-Metal Jet as a Charge Stripper

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For high-power heavy ion accelerators, the development of a suitable charge stripper, which can handle intense beams, is essential. This Letter describes the first experimental demonstration of a heavy ion liquid lithium charge stripper. A 10–20 μm thick liquid lithium jet flowing at >50 m/s was formed and confirmed stable when bombarded by various heavy ion beams, while increasing the charge state of the incoming beams to the desired charge state range. This demonstration proved the existing power limitation with the conventional strippers can be overcome by the liquid-metal stripper, opening completely new possibilities in high-power accelerator development.

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Introduction.—The need of high-power heavy ion beam facilities has been increasing more than ever in the 21st century. Areas of applications range from the fundamental science topics such as the properties of atomic nuclei, the origin of elements, and testing laws of nature, to the societal applications and benefits in the areas of medicine, energy, and material science. Linear accelerators have been the most cost effective in achieving high-power beams of heavy ions, specially, with the development of superconducting radio-frequency (SRF) cavities. A prime example of such a high-power heavy ion accelerator is the Facility for Rare Isotope Beams (FRIB) recently completed at Michigan State University (MSU) [1]. Open for users, May 2022, FRIB is poised to become one of world's most powerful rare isotope facilities.

To reduce the cost of the facility and utilize the accelerating voltage most effectively, these accelerators normally include a charge stripper to remove electrons from the ions to increase the charge state of the beams in the beam line at an intermediate energy determined by an optimization of charge state increase at the stripper. The optimization is between stripping too early and not reaching a high charge state and stripping too late and not using all the accelerating voltage effectively. A factor of 2 or more in charge state increase can be typically obtained.

Thin carbon foils have been the traditional charge strippers used in heavy ion accelerators but are limited in power density by the damage they suffer (sublimation and radiation damage) and consequently short lifetimes. Very heavy ions deposit several orders of magnitude more energy than protons per unit length in the stripping media, making a uranium beam most challenging to strip [2]. The best performing heavy ion stripper foils (highly oriented graphite carbon sheets) have been used at the Radioactive Isotope Beam Factory (RIBF) at RIKEN [3]. The reported performance results are for U beams at 50 MeV per nucleon (MeV/u). At this high energy the energy deposition per ion per unit length is about 60% of the energy deposition at FRIB's stripper energy. The demonstrated total numbers of ions traversing the stripper are of the order of 2×10^{18} particles, corresponding to about half a day (12 h) of FRIB full power operation. The beam spot at RIKEN's RIBF rotating disk stripper is 2 mm in diameter, which is twice the diameter of the FRIB's beam spot (1 mm). Thus, if FRIB were to use a similar rotating carbon disk stripper the FRIB running time to achieve the same total fluence would be only about six hours, completely unpractical.

The solution to this issue is the use of a self-replenishing media like a liquid or a gas. Gas strippers achieve a significantly lower charge state than solids and the use of liquids looks more promising [4–9]. The previous liquid stripper works employed vacuum compatible oils as the stripper media, which do not possess the best nuclear physical characteristics for charge stripping and thermophysical properties to tolerate an intense beam irradiation. In addition, their methods of forming the liquid films were not the best methods to effectively remove the thermal input from the beams to the film. A better choice of the stripper media is to use a liquid metal, especially lithium. Some of the advantages of using liquid lithium are its low vapor pressure and high heat capacity.

The use of liquid metal in an accelerator is actually not a new idea, proposed as early as in 1950s [10] and liquid-metal targets have been common for high power light ion accelerators (protons and deuterons). There are mainly two types of liquid-metal targets; one is a target with beam windows and another is a "windowless" target. An example of the former is the mercury target at the Spallation Neutron Source and at the Japan Proton Accelerator Research Complex [11,12] and those of the latter are liquid lithium targets [13–18] including the present work. Particularly worth noting is that the windowless liquid lithium targets, in which flowing liquid lithium is directly exposed to the beam line vacuum (high vacuum pressure of $<1\times10^{-4}$ Pa) with intense beam irradiations, have demonstrated the compatibility of the flowing liquid lithium with vacuum. However, all of the previous lithium targets, except for the one originally proposed by Nolen, flow along a curved wall, which pressurizes the flow to suppress the potential boiling [13–16,18]. For the charge stripping application, the ability to form a free-jet target without a backwall is an important consideration, because high-power beams must pass through the liquid and subsequently damage the wall structure. This is due to the much higher energy deposition per unit length from the heavy ion beams [2]. Thus, the presence of the backwall restricts the applicability of this type of windowless target to charge stripping. As a consequence, no liquid-metal charge strippers have been built before. In addition, the light ion targets are thick (typically order of tens of centimeters) to completely stop the beams while charge strippers are very thin (of the order of micrometers) and only small fluctuation in thickness can be tolerated, making application of the conventional liquid lithium targets to charge stripping challenging.

The fundamental drawbacks of the windowless concept are the uncertainties in the geometric stability of the flow due to the hydrodynamic and/or thermal behaviors of the flow. This is because a liquid jet is inherently unstable and, when *in vacuo*, thermodynamically only metastable. Heat deposition to the liquid could induce spontaneous phase transition from liquid to vapor, which is detrimental (evaporation) or catastrophic (boiling).

One of the first attempts to use a free-jet target was published in 1966 [19]. However, due to the severe evaporation or possibility of boiling in the flow with the high-power beam bombardment and its potential consequences, free-jet target systems have not subsequently been developed and demonstrated.

The use of a liquid lithium free jet as a charge stripper in a heavy ion accelerator was first proposed by Nolen in 2000 [20]. In the course of the research and development efforts that followed, FRIB had decided to use a windowless liquid lithium free jet as its baseline charge stripper. For efficient acceleration of the heavy ion beams at FRIB, charge stripping at 10–20 MeV/u is desirable and it was estimated that a target with the density thickness of 0.5–1 mg/cm^2 would be needed. This value is equivalent to a 10–20 μm thick liquid lithium film (liquid lithium density is about 0.5 g/cm^3). The FRIB design power for U beam is 400 kW at 200 MeV/u at the production target. At the stripper the U energy is approximately 17–20 MeV/u. The U ions at 17 MeV/u will deposit about 53 MeV per ion for the 10 μm thick lithium film, or 1.3% of the ion energy. With 5.2×10^{13} ions per second the total power deposited at the stripper is approximately 450 W. For a circular beam spot of 1 mm diameter the power density on the film is 56 MW/cm^3. The conventional stripper made of a thin carbon film would never be able to withstand such a high-power deposition. A discussion on carbon foil lifetimes can be found in [2].

With the target thickness of 10–20 μm and the heat load of 450 W, the concept of the windowless liquid lithium free-jet stripper was investigated, and several experimental demonstrations were performed at Argonne National Laboratory (ANL), and later together with FRIB [21–23].

Demonstration of high power beam deposition.—The investigations at ANL revealed that the hydrodynamic instability could be overcome by flowing the liquid lithium film jet at >50 m/s (corresponding drive pressure of 1 MPa). A roughly 10 μm thick, 1 cm wide, stable lithium film jet was successfully formed *in vacuo* [21,22]. The high flow velocity was not only for the hydrodynamic stability but also necessary to carry away the intense beam power to avoid boiling or excessive vaporization. To demonstrate the adequacy of the lithium film jet to sustain a high-power deposition, a 65 keV proton beam was used. At this energy the protons stopped within the first 1.5 μm of the lithium film. The experiment demonstrated that the velocity of >50 m/s was sufficient to carry away 300 W of the thermal power deposited in the lithium film within a 1 mm diameter beam spot and a thickness of the first 1.5 μm over the total thickness of 10 μm [23]. The estimated peak volumetric heat input from the proton beam was approximately 65 MW/cm^3, more than the FRIB average power density deposition (56 MW/cm^3). Also noted is the 300 W power deposition was more than half of the total FRIB's power deposition of 450 W. This experiment did not include the radiation damage that the heavy ions would have on a solid carbon foil, but with a self-replenishing liquid the lattice damage is not an issue. These demonstrations confirmed the applicability of the windowless liquid lithium free jet as a charge stripper; however, its actual charge state distribution after stripping remained unproven.

Equipment in linac tunnel.—Now the first demonstration liquid lithium charge stripper (LLCS) system has been constructed by FRIB at MSU [24,25]. In 2021, the LLCS system was mated with FRIB's heavy ion linac and subsequently, the first demonstration of the liquid-lithium, thin-film jet as a charge stripper was successfully carried out. This article is the first report discussing the latest experiment and performance of the LLCS at FRIB.

The LLCS system consists of a liquid lithium loop equipped with a high-pressure electro-magnetic (EM) pump, vacuum system, argon (Ar) supply system, and Secondary Containment Vessel (SCV) as shown in Fig. 1(a). A part of the loop is a vacuum chamber (VC), which forms part of the linac beam line. The nozzle in the VC produces a round liquid lithium jet issuing from a 0.5 mm orifice in the nozzle. The round jet subsequently impacts the deflector, forming a willow leaflike liquid

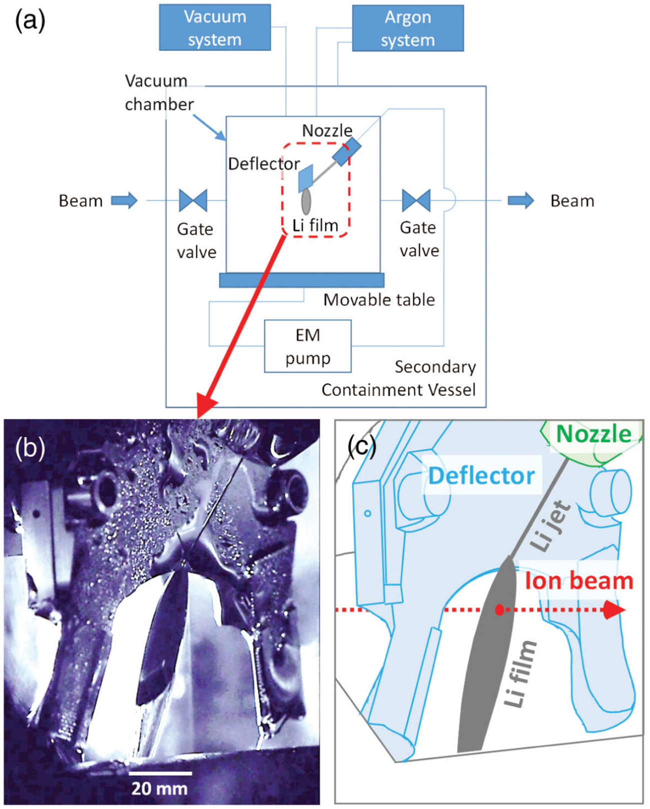

FIG. 1. Schematic of liquid lithium charge stripper system and lithium film. (a) Schematic illustration of the LLCS system, area indicated by red-dot line with more details shown in (b) and (c). (b) Photograph of the liquid lithium film formed in the LLCS vacuum chamber. The extremely smooth surface of the lithium film appeared as a mirror. (c) Illustration of the liquid lithium film with labels for clarity.

lithium film *in vacuo*, which intercepts the beam, as shown in Figs. 1(b) and 1(c). The VC is on a movable table within the SCV so that the stripper can be moved around the beam, while the beam path was maintained stationary.

In the design phase, considerable effort was made for safety. Liquid lithium is known for its high reactivity with air, water, and many other materials. One of the safety measures that have been implemented for operating the LLCS system in the accelerator tunnel is the SCV completely encasing the lithium loop and being filled with Ar during operations.

The lithium was heated by electric heaters installed along the lithium loop at around 220 °C (the melting point of lithium is 180.5 °C) and pressurized at 1.2 MPa by the EM pump.

Xenon ($^{124}\mathrm{Xe}^{26+}$) and argon ($^{36}\mathrm{Ar}^{10+}$) beams from the first linac section (LS1) of the FRIB linac were used for the initial characterization of the liquid lithium film. At the location of the LLCS in the linac, the energies of $^{124}\mathrm{Xe}^{26+}$ and $^{36}\mathrm{Ar}^{10+}$ beams were 17 MeV/u and 20 MeV/u, respectively. The corresponding root-mean-square (rms) beam radius was estimated to be 0.5 mm. These beams were used to measure the temporal stability of the liquid lithium stripper by monitoring the beam energy loss through the stripper over a long period (typically tens of minutes). During the energy loss measurements, the film was moved around the beam to measure the spatial profile of the film. The charge distribution was also measured to evaluate the performance of the LLCS. The thermal performance of the film was measured with an $^{36}\mathrm{Ar}^{10+}$ beam at 17 MeV/u and a peak current of 12 particle μA, the highest peak current allowed within the present accelerator operational envelope. The beam duty cycle was set at 5.4% with the repetition rate of 10 Hz, resulting in the instantaneous peak beam power of 7340 W during each

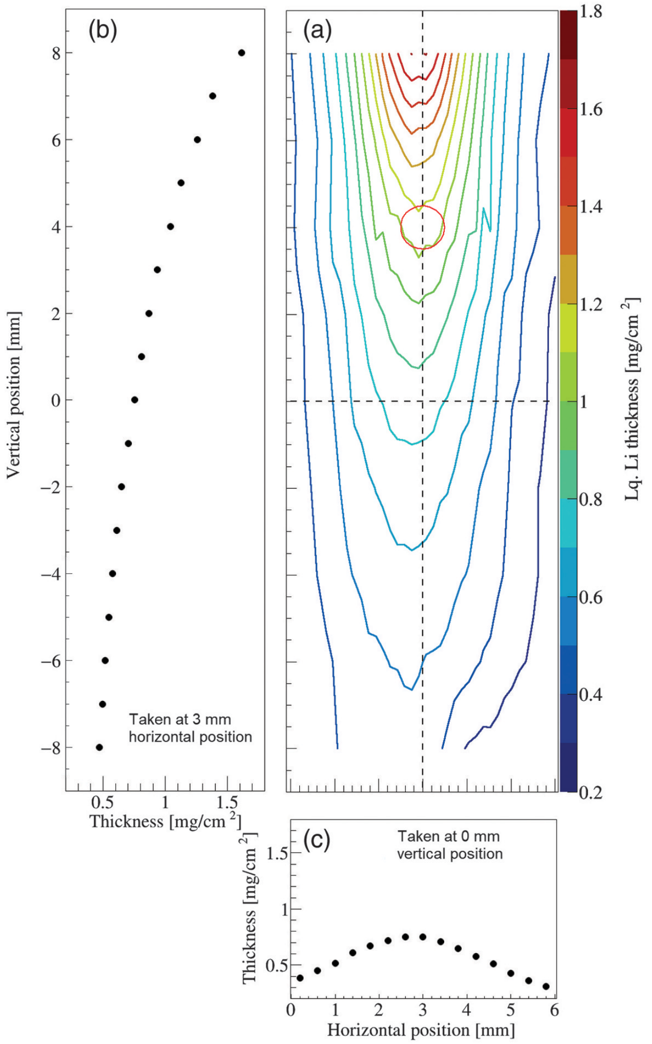

FIG. 2. Spatial profile of the liquid lithium film obtained from the energy loss measurement with 20 MeV/u Ar beam. (a) A two-dimensional profile. Red circle indicates a typical beam location of a 1 mm beam relative to the film. Dashed lines indicate where cross sectional profiles were obtained, which are shown in (b) and (c). (b) Vertical cross sectional thickness profile. (c) Horizontal cross sectional thickness profile.

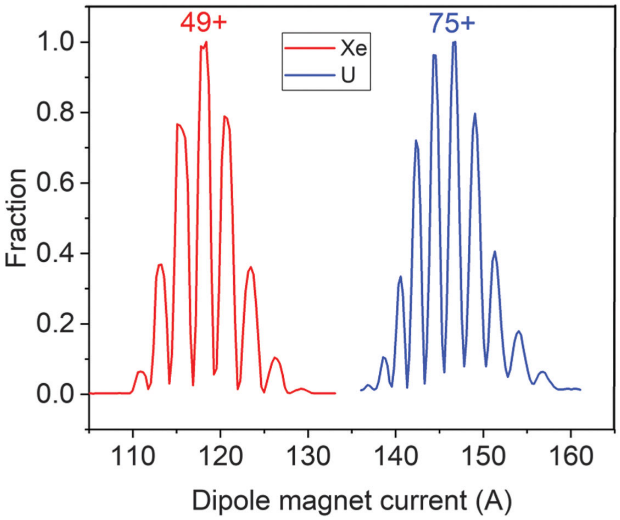

FIG. 3. Xenon (red) and uranium (blue) charge state distributions after the liquid lithium stripper. The thickness is $1.05\ \mathrm{mg/cm^2}$ for the xenon and $1.40\ \mathrm{mg/cm^2}$ for uranium beams.

5.4 ms period. The peak power loss in the lithium film ($0.6\ \mathrm{mg/cm^2}, dE/dx = 405\ \mathrm{keV}/\mu\mathrm{m}$) was 50 W.

The estimated volumetric power deposition in the lithium during the high-power test reached $6.2\ \mathrm{MW/cm^3}$, or 11% of the FRIB full power operation value ($56\ \mathrm{MW/cm^3}$).

Since it took approximately 20 μs for the flowing lithium at 50 m/s to completely cross the beam spot of 1 mm, it was considered that the longest time constant of any thermal and fluid dynamic responses of the lithium flow was 20 μs. Thus the 5.4 ms long beam, which was 270 times longer than the longest time constant, may be considered well representing a continuous beam.

Subsequently, the LS1 accelerated FRIB's first uranium ($^{238}\mathrm{U}^{36+}$) beam and traversed the liquid lithium film. At the LLCS, the energy was ranging from 17 to 20 MeV/u. The rms beam radius on the film was estimated to be 0.5 mm. The charge distribution was measured to evaluate the performance of the LLCS.

Results.—The beam energy loss measurements confirm that the liquid lithium film is very stable since the Xe beam energy variation was 17 keV/u (peak-to-peak value) with the standard deviation of 3 keV/u over a period of 2.5 h, which corresponds to the 9×10^{-4} peak-to-peak fluctuation of the beam energy. Comprehensive computer simulations indicated that this level of beam energy changes would not introduce any issue with the following acceleration. At the same time, it was noticed that the energy fluctuations are correlated with the slight variation of the vacuum chamber temperature. Thus, further optimization of the LLCS temperature control parameters is expected to reduce such energy fluctuations. The spatial distribution of the energy loss measurements shows that the thickness of the film decreased with the distance from the impact point of the deflector (see Fig. 2), which is consistent with the previous measurement [21]. The charge state distributions obtained with 17 MeV/u xenon and uranium at different locations on the lithium film are shown in Fig. 3. The unit of thickness here is $\mathrm{mg/cm^2}$. The conversion of the energy loss to the film thickness in $\mathrm{mg/cm^2}$ was calculated with the SRIM code [26]. The charge state distributions of the 20 MeV/u uranium beam were measured in the liquid lithium and a carbon foil at 1 $\mathrm{mg/cm^2}$ thickness. It was found that the average charge states are 73.7 and 76.9 for the liquid lithium and carbon, respectively.

Conclusions and impacts.—The two major issues with the traditional carbon foil strippers were sublimation and radiation damage by high-power heavy ion beams. The replenishing liquid lithium free jet solves the radiation damage to the lattice and the previously reported high-power proton beam demonstration performed at ANL, proved that the free jet was stable at high-power deposition. It was experimentally confirmed that the windowless liquid lithium thin free jet could be used as a charge stripper. The hydrodynamic stability of the liquid lithium thin free jet with a thickness of 10–20 μm, flowing at 50 m/s in the high vacuum environment, was proven effective even with U beams to match the post stripper accelerator section. The charge stripping characteristics of the lithium film were obtained, producing charge state distributions with only slightly lower peak charge state than an equivalent carbon foil. With the demonstration of stripping heavy ion beams, additional application potentials may exist. For example, by optimizing the lithium film thickness and controlling the film location, high-power pulsed proton facilities could use this method for H^- stripping [27]. It was shown for the first time the technical limitations associated with the conventional solid stripper technologies can be overcome by employing the concept of the windowless liquid-metal free-jet stripper, opening new possibilities in high-power accelerator development.

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[1] J. Wei *et al.*, Int. J. Mod. Phys. E **28**, 1930003 (2019).
[2] J. A. Nolen and F. Marti, Rev. Accel. Sci. Techol. **06**, 221 (2013).

[3] H. Hasebe, H. Okuno, A. Tatami, M. Tachibana, M. Murakami, H. Kuboki, H. Imao, N. Fukunishi, M. Kase, and O. Kamigaito, in *Proceedings of the 28th World Conference of the International Nuclear Target Development Society (INTDS2016)* (AIP Publishing, Melville, 2018), 030004.

[4] J. R. Alonso and B. T. Leeman, IEEE Trans. Nucl. Sci. **26**, 3718 (1979).

[5] D. F. Burch, J. G. Cramer, and P. B. Cramer, Annual Report 1976, Nuclear Physics Laboratory, University of Washington, 1976.

[6] J. G. Cramer, D. F. Burch, R. Rodenberg, and P. B. Cramer, Nucl. Instrum. Methods **185**, 29 (1981).

[7] B. T. Leemann, P. Merrill, H. K. Syversrud, R. Wada, and R. B. Yourd, IEEE Trans. Nucl. Sci. **28**, 2794 (1981).

[8] H. Akiyoshi, T. Chiba, H. Hasebe, M. Kase, and Y. Yano, in *Proceedings of the 13th Symposium on Accelerator Science and Technology, Suita, Osaka, Japan, October 2001* (Particle Accelerator Society of Japan, Tokyo, 2001).

[9] H. Ryuto, T. Chiba, H. Hasebe, M. Kase, and Y. Yano, Jpn. J. Appl. Phys. **43**, 7753 (2004).

[10] R. H. Ball, in Minutes of Meeting of MTA Target Committee Held June 11, 1951, *UCRL-1348 Technology—Material Testing Accelerator* (University of California, Berkeley, 1951).

[11] D. E. Winder, in *Proceedings of the 12th International Particle Accelerator Conference (IPAC), Brazil* (JACoW, Geneva, 2021).

[12] H. Takada *et al.*, Quantum Beam Sci. **1**, 8 (2017).

[13] R. E. Peterson and J. A. Hassberger, Fusion Material Irradiation Test facility initial target design, HEDL SA-2019, Hanford Engineering Development Laboratory, 1979.

[14] R. Kolowith, J. D. Berg, and W. C. Miller, Experimental Lithium System final report, HEDL-TME 84-29, Hanford Engineering Development Laboratory, 1985.

[15] H. Kondo, T. Kanemura, T. Furukawa, Y. Hirakawa, E. Wakai, and J. Knaster, Nucl. Fusion **57**, 066008 (2017).

[16] A. Ibarra *et al.*, Nucl. Fusion **58**, 105002 (2018).

[17] J. A. Nolen, C. B. Reed, V. J. Novick, J. R. Specht, J. M. Bogaty, P. Plotkin, and Y. Momozaki, Rev. Sci. Instrum. **76**, 073501 (2005).

[18] S. Halfon *et al.*, Rev. Sci. Instrum. **84**, 123507 (2013).

[19] J. A. R. Griffith and E. J. Burge, Rev. Sci. Instrum. **37**, 147 (1966).

[20] J. A. Nolen *et al.*, Argonne Physics Division 2000 Annual Report, ANL-01/19, Argonne National Laboratory, 2001.

[21] Y. Momozaki, J. A. Nolen, C. B. Reed, V. J. Novick, and J. R. Specht, J. Instrum. **4**, P04005 (2009).

[22] C. B. Reed, J. Nolen, Y. Momozaki, J. Specht, D. Chojnowski, and R. Lanham, ANL/NE-11/01, Argonne National Laboratory, 2011.

[23] Y. Momozaki, C. B. Reed, J. A. Nolen, J. R. Specht, D. B. Chojnowski, J. S. Song, F. Marti, P. Guetschow, and J. Sherman, J. Radioanal. Nucl. Chem. **305**, 843 (2015).

[24] F. Marti, P. Guetschow, J. A. Nolen, Y. Momozaki, and C. B. Reed, in *Proceedings of the 13th International Conference on Heavy Ion Accelerator Technology, Yokohama, Japan* (JACoW, Geneva, 2015).

[25] T. Kanemura, J. Gao, M. LaVere, R. Madendorp, F. Marti, and Y. Momozaki, in *Proceedings of the North American Particle Accelerator Conference, Michigan, USA* (JACoW, Geneva, 2019).

[26] J. F. Ziegler, M. D. Ziegler, and J. P. Biersack, Nucl. Instrum. Methods Phys. Res., Sect. B **268**, 1818 (2010).

[27] J. Wei, Rev. Mod. Phys. **75**, 1383 (2003).