RANGE PARAMETERS OF GOLD IONS IN BORON CALCULATED WITH A FIRST-PRINCIPLES POTENTIAL

V. Kuzmin

Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Joliot-Curie 6, 141980, Dubna, Moscow reg., Russia, +7 49621 64882, V.Kuzmin@jinr.ru

Interatomic potential for the Au-B system have been calculated with density-functional theory (DFT) in order to clarify the origins of discrepancies between available experimental data and the values predicted by Ziegler, Biersack and Littmark (ZBL) theory for range parameters of heavy ions in light targets at energies of about 1-1000 keV. Relativistic effects were taken into account in the evaluations of the potential. Range parameters have been obtained within the framework of the standard transport theory. Good agreement between the calculated projected range and the available experimental data gives use in the range-projection equations the nuclear stopping power, determined with the DFT potential, and the velocity-proportional electronic stopping powers by Lindhard, Scharff and Schiøtt (LSS). Considerable improvement has also been achieved in the description of the projected range straggling. It is concluded that correlations between the nuclear and electronic energy losses can be neglected in the studied energy range.

Introduction

The rapid shrinking of dimensions of interest in technologies using ion beam techniques makes increasing demands on the accuracy of underlying theory. As properties of a modified layer in many respects depend on distributions of energy losses and ranges of stopped ions, it is very important to be able correctly predict these quantities. Based on the binary collision approximation (BCA), the semi-empirical approach developed by Ziegler, Biersack and Littmark (ZBL) [1] is perhaps the most widely used for calculation of stopping powers and ranges of energetic ions in matter. The overall accuracy of stopping power calculations with the newest implementation of the ZBL [2] is of order 5%. For projected range (R_p) and projected range straggling (ΔR_p) an overall agreement of ZBL predictions is of the order of 10% for a wide range of implantation energies and for a large set of ion-target combinations [1]. However, up to 40% higher ranges in comparison with SRIM simulations have been revealed for medium-heavy ions ($29 \le Z \le 83$) implanted into low-Z target materials at energies between 10 and 300 keV [3-7]. Friedland et al. [8] observed excess more than 50% in ranges of gold and lead ions in carbon at energies of about 1-1000 keV. Considerable differences between measured ranges and corresponding results from ZBL calculations also discussed in Refs. [9-10].

These discrepancies were explained in Refs. [5-7] by a correlation between the nuclear and electronic energy losses. In addition to the quasielastic description of atomic scattering [11] it was proposed to transform the universal ZBL potential with the ZBL electronic stopping power for an outgoing part of the path [5-7]. This model allowed to improve a description of a part of experimental results in Ref. [9] Also, the same treatment of a scattering angle was also successfully utilized in Ref. [12] to describe range parameters of bismuth ions in polystyrene in the framework of the transport theory.

It should be noted that the universal ZBL potential is an average of a large number of repulsive potentials, calculated with an approximate method [13]. Therefore, the large discrepancies between the experimental and theoretical values for range parameters can indicate the lack of accuracy of the ZBL potential and/or the ZBL electronic stopping for the ion-target combinations in question.

Previous calculations of range parameters of heavy ions in carbon [14-16] showed that quite a good description of available experimental data [6,8] can be obtained without assumption on a correlation between the nuclear and electronic energy losses. It was concluded that the reasons for the discrepancies were inaccuracies of interatomic potentials and electronic stopping powers of the ZBL theory for considered ion-target combinations.

In order to further study the problem of correct description of interaction of heavy ions with low-Z materials we extend the approach, developed in earlier works [14-16], for boron target. In the present work an interatomic potential for the Au – B system has been calculated with density-functional theory (DFT) and compared with the universal ZBL potential. Relativistic corrections were taken into account in the evaluation of the potential. Range parameters of Au ions in boron have been computed in the framework of the standard transport theory. Coefficients of the range-projection equations have been determined using the first-principles potential. Conclusion on the importance of correlation effects between the nuclear and electronic energy losses are made after a comparison of the calculated range parameters with available experimental data [6].

Interatomic potentials

DFT potential calculations, using the local-spin-density approximation (LSDA) for exchange-correlation functional, were performed with the commonly available quantum chemistry program package GAMESS(US) [17] relying on the finite basis sets approach. In the package, Gaussian-type function basis sets are used to expand molecular orbitals. As conventional problems of quantum chemistry are bounded by consideration of molecular systems near to an equilibrium state, one should take great care regarding the choice of basis sets used in such calculations. The present calculations were performed with recently reported basis sets constructed using the relativistic 3rd order Douglas-Kroll (DK3) transformation [18]. The basis sets were decontracted and augmented with polarization and diffusion functions to improve their flexibility and completeness. The ground states of neutral Au – B diatomic were considered in the DFT calculations.

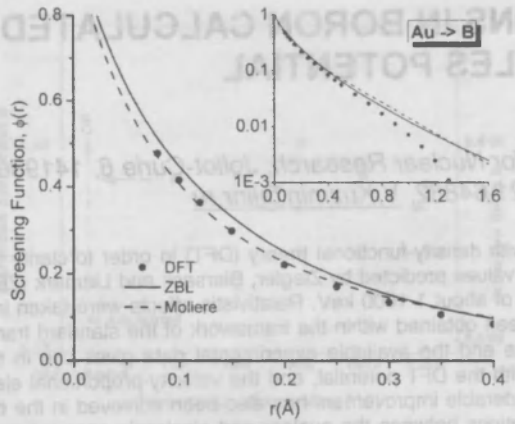

Fig. 1. Screening functions for Au – B diatomic. DFT denotes the density-functional calculations with relativistic corrections included by the DK3 method. Also shown are the ZBL and Molière screening functions. The inset indicate the long range behavior of the screening functions.

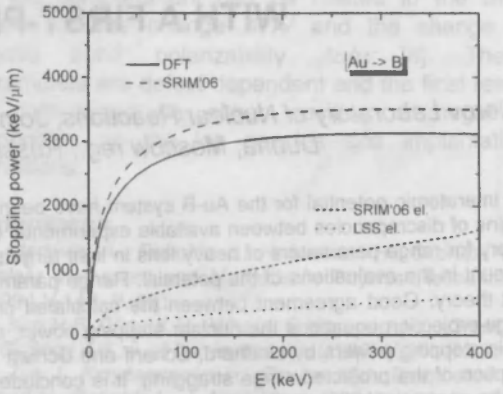

Fig. 2. Nuclear stopping powers of Au ions in B for the DFT potential, and the ZBL potential (SRIM'06). The ZBL (SRIM'06 el.) and LSS electronic stopping powers are given for comparison.

The results of the calculations for the screening function with relativistic corrections included by the DK3 method are shown in Fig. 1. Also given for comparison are those for the ZBL and the Moliere potentials. One can see that for the Au – B system the Moliere potential much better corresponds to the DFT one at interatomic distances of most importance for studied here ion energies of about 10 – 1000 keV. Similar situation was observed before for the Au – C system [14]. It is worth noting that the average of the relative errors squared for the Moliere potential is 237% whereas for the ZBL potential that is only of about 5% [1]. Using the DK3 method at HF level of theory it was found that relativistic corrections decrease screening functions by approximately 1% for r = 0.1 Å, and ~ 4% for r = 1 Bohr. At separation 1.2 Å the value is about 20% too low.

Stopping powers and range parameters

The classical treatment of the scattering problem (see, for example, [19]) has been used to compute the nuclear stopping power. The results of the calculations are depicted in Fig. 2 along with the ZBL nuclear stopping power. Also shown are the ZBL and LSS [20] electronic stopping powers for comparison. At 20 keV the nuclear stopping power determined by the ZBL potential are about 10% larger than DFT counterpart, increasing with energy up to 15% at 300 keV. Seems, this value are not large enough to explain the existing discrepancies in a description of the ranges. Therefore, we also consider a possible alternative to the ZBL electronic stopping power. At 20 keV the ZBL electronic stopping power is about 15% of the ZBL nuclear stopping power, reaching nearly 40% at 300 keV. So, knowing of an accurate value of the electronic stopping power is very important for the energies of interest. Unfortunately, available experimental data relevant to the studied ion-target combinations and energy range are very limited. Choice of well-known electronic stopping power by Lindhard, Scharff and Schiøtt (LSS) [20] for the present calculations is explained an attempt to bring theoretical data in correspondence with experimental one. Fig. 2 demonstrate that the ZBL electronic stopping considerably surpass the LSS one in our case. At 300 keV the ZBL electronic stopping power is approximately twice as large the LSS.

The standard transport theory [20] was employed in order to evaluate projected range and projected range straggling. The general scheme of deducing range-projection equations can be found elsewhere (see, for example, [21] and references therein). Second-order coupled differential range-projection equations were solved by the computer code ID03 [22]. Using second-order equations is crucial in order to obtain correct value for projected range straggling. The second moment of the electronic energy loss was neglected in the present calculations. The values for the projected range and projected range straggling differ from those of SRIM by less than 1% and 3% respectively if the universal ZBL potential and the ZBL electronic stopping power are taken as input quantities for the ID03.

The R_p and ΔR_p values calculated with the LSS electronic stopping power and the nuclear stopping power as determined by the DFT potential were compared to the values of these parameters measured in [6]. The results of the calculations for the Au – B system are presented in Fig. 3 along with those computed with SRIM-2006 computer code and data reported in Ref. [6]. One can see that the calculations using the DFT potential and LSS electronic stopping give quite a good description of the experimental data for the projected ranges of Ref. [6]. Maximum deviation is about 5% (average ~ 4%). The corresponding SRIM-2006 calculations deviate from experimental values by about of 27% (average ~ 24%). Also, as seen in Fig. 3 considerable improvements are observed in the predictions of ΔR_p using the DFT potential: average deviation is about 27% against 48% by SRIM-2006.

Conclusions

In this paper the interatomic potential for the Au – B system have been calculated with DFT including relativistic corrections by DK3 method. The

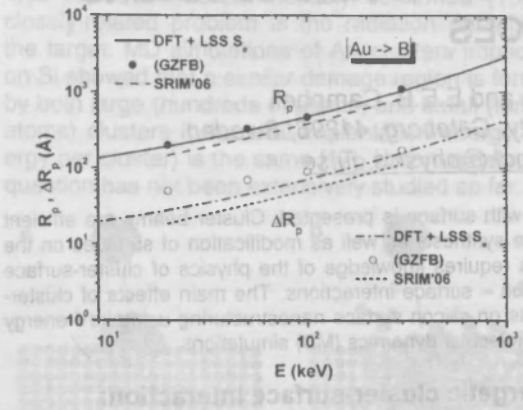

Fig. 3. Ranges parameters *Rp* and Δ*Rp* of Au ions in B as calculated by the DFT potential with the LSS electronic stopping and by ZBL (SRIM'06). The calculations are compared to the experimental data of GZFB [6].

relativistic effects lead to notable decrease of potentials at large distances. The use in the range-projection equations of the nuclear stopping power, determined with the DFT potential, and the LSS electronic stopping power leads to close correspondence between predicted values of the range parameters and experimental data of Ref. [6]. Considerable improvement has also been achieved in the description of the projected range straggling. The present result is an additional evidence of quite a good accuracy of the LSS theory in a description of the electronic stopping of low energy heavy ions. Finally, it is concluded that correlations between nuclear and electronic energy losses can be neglected in the studied energy range.

References

1. *Ziegler J.F., Biersack J.P., Littmark U.* The stopping and range of ions in solids. – N.Y.: Pergamon, 1985.
2. *Ziegler J.F., Biersack J.P.* SRIM-2006.01 computer code (cf. http://www.srim.org).
3. *Grande P.L., Fichtner P.F.P., Behar M., Zawislak F.C.* // Nucl. Instr. and Meth. – 1988. – B33. – P.122.
4. *Grande P.L., Behar M., Biersack J.P., Zawislak F.C.* // Nucl. Instr. and Meth. – 1990. – B45. – P.689.
5. *Behar M., Grande P.L., Wagner de Oliveira R., Biersack J.P.* // Nucl. Instr. and Meth. – 1991. - B59/60. – P.1.
6. *Grande P.L., Zawislak F.C., Fink D., and Behar M.* // Nucl. Instr. and Meth. -1991. - B61. – P.282.
7. *Behar M., Fichtner P.F.P., Grande P.L., Zawislak F.C.* // Mat. Sci. and Eng. – 1995. - R15. – P.1.
8. *Friedland E., Kalbitzer S., Hayes M., Klatt Ch., Konac G., Langpape Ch.* // Nucl. Instr. and Meth. – 1998. - B136-138. – P.147.
9. *Wang K.-M., Lu F., Meng M.-Q., Shi B.-R., Shen D.-Y., Liu Y.-G., Fink D.* // Nucl. Instr. and Meth. – 1998. - B145. – P.271.
10. *Wang K.-M., Shi B.-R., Cue N., Shen D.-Y., Chen F., Wang X.-L., Lu F* // Nucl. Instr. and Meth. – 2004. – B225. – P.503.
11. *Robinson M.T., Torrens I.M.* // Phys. Rev. – 1974. - B9. – P.5008.
12. *Liang J.H.* // Nucl. Instr. and Meth. – 1997. - B132. – P.29.
13. *Gombas P.* Die Statistische Theorie des Atoms und ihre Anwendungen. – Berlin: Shpringer, 1949.
14. *Kuzmin V.* // Nucl. Instr. and Meth. – 2006. – B249. – P.13.
15. *Kuzmin V.* // Surf. Coat. Technol. – 2007. - doi:10.1016/j.surfcoat.2006.10.053 (in press).
16. *Kuzmin V.* // Nucl. Instr. and Meth. – 2007. – B256. – P.105.
17. *Schmidt M.W. et al.* // J. Comp. Chem. – 1993. – 14. – P.1347. (cf. http://www.msg.ameslab.gov/GAMESS/GAMESS.html).
18. *Tsuchiya T., Abe M., Nakajima T., Hirao K.* // J. Chem. Phys. – 2001. – 115. – P.4463.
19. *Wilson W.D., Haggmark L.G., Biersack J.P.* // Phys. Rev. – 1977. - B15. – P.2458.
20. *Lindhard J., Scharff M., and Schiøtt K.* // Mat. Fys. Medd. Dan. Vid. Selsk. – 1963. – 33. no.14.
21. *Littmark U. and Ziegler J.F.* // Phys. Rev. – 1981. - A23. – P.64.
22. *Kuzmin V.* ID03 computer code (unpublished).