Nuclear Instruments and Methods in Physics Research B 267 (2009) 2657–2661

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Range parameters of slow gold ions implanted into light targets

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ARTICLE INFO

Article history:
Available online 23 May 2009

PACS:
34.20.Cf
31.15.Ar
31.30.Jv
61.80.Az

Keywords:
Interatomic potentials
Density-functional theory
Stopping power
Range parameters
Heavy ions
Carbon
Silicon carbide

ABSTRACT

Interatomic potentials for Au–C, Au–B, Au–N and Au–Si systems, calculated with density-functional theory (DFT) methods, have been used to evaluate the range parameters of gold in B, Si, BN and SiC films at energies of about 10–400 keV. The potentials have been employed to describe scattering angles of a projectile and to calculate the nuclear stopping powers and the higher moments of the energy, transferred in single collisions. Utilizing these findings the range parameters have been obtained by the standard transport theory and by Monte-Carlo simulations. A velocity proportional electronic stopping was included into the consideration. The approach developed corresponds completely to the standard classical scheme of the calculation of range parameters. Good agreement between the computed range parameters and available experimental data allow us to conclude that correlation effects between the nuclear and electronic stopping can be neglected in the energy range in question. Moreover, it is proven for the first time that the model by Grande, et al. [P.L. Grande, F.C. Zawislak, D. Fink, M. Behar, Nucl. Instr. and Meth. B 61 (1991) 282], which relies on the importance of correlation effects, contains inherent contradictions.

1. Introduction

Nowadays, ion beam implantation is widely used to modify surface properties of solids and even to form new materials. New technological challenges request more and more accurate predictions for stopping, depth distributions of stopped ions and damage induced by the implantation. On the other hand, a comparison of calculated results with experimental data leads to a conclusion concerning the validity and accuracy of approximations used in the underlying theory.

The semi-empirical approach, developed by Ziegler, Biersack and Littmark (ZBL) [1] and embodied in the family of computer codes TRIM/SRIM [2], is perhaps the most widely approach used for the calculation of the stopping powers and ranges of energetic ions in matter owing to its rather high overall accuracy, availability and user-friendly interface. However, up to 40% higher ranges in comparison with results of TRIM simulations have been revealed for medium–heavy ions ($29 \leqslant Z \leqslant 83$) implanted into low-$Z$ target materials at energies between 10 and 400 keV [3–12]. To explain these discrepancies a phenomenological model was developed and reported in detail in [6–8] (hereafter referred to as the GZFB model). In this model, the disagreements were attributed to a correlation between the nuclear and electronic energy losses. The atomic scattering process was considered to be quasi-elastic (see for e.g. [13]). Additionally it was supposed, that after the collision the movement of particles is defined by a substantially changed potential, which corresponds to an excited state of the colliding partners. Subsequently, other scientific groups also applied the GZFB model to describe [14,15] or explain [16,17] experimental data. With a few exceptions (e.g. Xe into C [8] and Yb into $KTiOPO_4$ [15]), adaptation of the GZFB model dramatically improved the description of experimental data for most of varied combinations of heavy ion – light targets, considered in [6–12,14,15], where unmodified TRIM/SRIM calculations significantly underestimated range parameters.

Another explanation of the inconsistency was given in the [18,19]. Friedland with coworkers [19] observed range enhancements of nearly 50% for Au and Pb ions implanted into C at energies of about 600–1000 keV, in a close correspondence with earlier measurements of [18] for Au/C. Authors of these papers ascribed the inconsistency to erroneous ZBL electronic stopping powers [18,19]. In [19] it was stressed that the comparison of experimental results with computations using averaged potentials is an inappropriate method to either confirm or reject the existence of correlation effects.

Our previous papers on the calculation of range parameters of heavy ions in carbon [20] have demonstrated quite a good correspondence between available experimental data and results obtained within the classical schemes [1,21]. It was achieved using first-principles potentials in calculations of the projectile

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doi:10.1016/j.nimb.2009.05.013

scattering angle and also the nuclear stopping powers and higher moments of the energy, transferred in a single collision. A velocity proportional electronic stopping power was taken into account. It followed from these considerations that inaccuracies in the ZBL potential and/or the ZBL electronic stopping were source of the severe disagreement between the experimental and theoretical values of the range parameters [20].

The present paper extends the preceding studies [20] in order to clarify an origin of the discrepancies discussed above. At the beginning the GZFB model is scrutinized. For the first time it is proven that the GZFB model violates the energy conservation law. Then, the previously developed approach [20] is used to calculate the range parameters of gold in B, Si, BN and SiC films at energies of about 10–400 keV. Interatomic potentials for Au–C, Au–B, Au–N and Au–Si systems are calculated with density-functional theory (DFT). Range parameters of Au in B and Si films are evaluated in the framework of the standard transport theory; those for Au in BN and SiC are obtained by Monte-Carlo simulations.

2. Critical review of the GZFB model

In the GZFB model the interaction potential was supposed to change substantially after the collision. This excited potential V_{exc} was defined by the expression [8]:

$$V_{exc}(r) = \lambda V_{ZBL}(r) + Q(b) \quad \text{with} \quad \lambda = 1 - Q/V_{ZBL}(r_0), \tag{1}$$

where $V_{ZBL}(r)$ is the ZBL universal potential and r_o is the apsis of the collision. The local electronic energy loss $Q(b)$, dependent on the impact parameter b through the apsis r_o, was taken in the form:

$$Q(b) = S_e(2\pi)^{-1}\left(\frac{\alpha}{a_u}\right)^2 \exp\left(-\alpha\frac{r_0}{a_u}\right), \tag{2}$$

where S_e is the ZBL electronic cross section, a_u is the universal screening length and α was taken to be 0.5. It was asserted that this approximation for V_{exc}, Eq. (1), fulfills some basic requirements. One of the requirements reads: "with $r \to \infty$, V_{exc} goes to the right asymptotic value $Q(b)$" [8]. Let us consider a consequence of this statement in more detail. In the elastic collision the scattering angle θ in the center-of-mass (CM) system is given by (see for example [13,22]):

$$\theta = \pi - 2\int_{r_o}^{\infty} \frac{bdr}{r^2(1 - V(r)/E_c - b^2/r^2)^{1/2}}, \tag{3}$$

where $V(r)$ is the interatomic interaction potential, and E_c is the total energy of colliding particles in the CM system. E_c is related to the initial kinetic energy of a projectile in the laboratory system, E, by $E_c = (m/m_1)E$, where the reduced mass m is defined as $m = m_1m_2/(m_1 + m_2)$, where m_1 and m_2 are the projectile and target masses, respectively. It worth mentioning that Eq. (3) is readily deduced from the law of conservation of energy (see for example [22]). Moreover, Eq. (3) reflects the isotropy of time, providing equal contributions to the total scattering angle θ from both incoming and outgoing path in the elastic collision. In the GZFB model the scattering angle is also calculated with Eq. (3), however, the outgoing path is determined by V_{exc} instead V_{ZBL}. Considering θ as a functional of V, $\theta\{V\}$, the total scattering angle, Ξ, within the GZFB model can be written as: $\Xi = 0.5\,\theta\{V_{ZBL}\} + 0.5\,\theta\{V_{exc}\}$ This dependence implies that the system loses the energy Q at r_o. The total energy is a constant before and after this point. If we write the energy conservation law in a cylindrical frame (CM system), we obtain the following expression for an arbitrary point on the outgoing path:

$$E_c - Q = \frac{mv_r^2}{2} + \frac{M^2}{2mr^2} + V_{exc}(r), \tag{4}$$

where v_r is the radial velocity and M is the angular momentum. M is constant for a central-force potential. Therefore, in the limit $r \to \infty$ Eq. (4) reduces to:

$$E_c - Q = \frac{mv_\infty^2}{2} + Q, \tag{5}$$

or

$$\frac{mv_\infty^2}{2} = E_c - 2Q, \tag{6}$$

where v_∞ is the velocity at infinity. The term in the left side of Eq. (6) gives the total energy of the system after the collision, when particles are at infinite distance from each other, and interaction is absent. E_c is initial energy in the system. Thus the system has lost amount of energy twice as much as it was defined by the statement of the problem. The evident origin of the above contradiction is the incorrect asymptotic behavior of the V_{exc}. Any potential in Eq. (3) must go to zero at infinity. This statement has a very simple qualitative explanation: as the force operating on a particle is equal to the negative of the derivative of the potential, any constant potential cannot contribute to the scattering angle of this particle.

The reasons for the increasing of the projected range in the GZFB model are: (a) treatment of the atomic scattering process as the quasi-elastic one, that decreases the total stopping power; (b) the use of the erroneous formula, Eq. (1), that underestimates the scattering angle and so additionally decreases the nuclear stopping power; (c) the choice the value of 0.5 for the constant α in Eq. (2) (compare with the value 0.3 in the original paper by Oen and Robinson [23]) also lowers the electronic stopping power.

3. Interatomic potentials

In order to obtain accurate interatomic potentials for Au–C, Au–B, Au–N and Au–Si systems, density-functional theory (DFT) calculations were performed using commonly available quantum chemistry program package GAMESS(US) [24] which realizes the finite basis sets approach. In the package, Gaussian-type function basis sets are used to expand atomic/molecular orbitals. In the present work, a 3rd order Douglas–Kroll–Hess (DKH) [25–28] approximation has been used to take into account relativistic corrections. DKH provides highly accurate results, using ordinary (non-relativistic) basis sets. Basis sets of [29], optimized with DKH, were used in the calculations. The basis sets were uncontracted and augmented with polarization and diffuse functions to improve their flexibility and completeness. All the calculations were done on electronic states with the lowest spin multiplicity. The nuclear stopping powers as given by the DFT potentials, were found not sensitive to a particular choice of the exchange-correlation electron energy functional in the studied energy range. Details of the calculations can be found in [20].

In Fig. 1 normalized differences between the DFT and ZBL potentials for Au–B and Au–Si diatomics as a function of interatomic separation are plotted. Also shown for comparison are those between the DFT and Moliére potentials. One can see that for the Au–Si diatomic the ZBL potential corresponds to the DFT potential within 7% for $r \leqslant r_0$ ($r_0 = 0.529$ Å, Bohr radius). In this interval the difference between the DFT and ZBL potentials for Au–B system exceeds 15%, considerably increasing with interatomic distances. Similar results were found for Au-C system before [20].

4. Stopping powers and range parameters

The classical treatment of the elastic scattering problem (see e.g. [1]) has been used in the present calculations. The atomic scattering angle is described by Eq. (3). The nuclear stopping powers given by the standard formula (see e.g. 1), have been calculated with the DFT and ZBL potentials. The results of these computations