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GASES AND FLUIDS

Quasihard-Sphere Model in Simulation of the Processes of Particle Scattering

V. A. Vol'pyas and E. K. Gol'man

St. Petersburg State University of Electrical Engineering, St. Petersburg, 197376 Russia

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Abstract—A model of interatomic potentials of interaction is suggested for static simulation of the processes of elastic scattering of atomic particles by atoms of gas, plasma, and solid. In the developed model, the atomic particle radii, whose magnitude depends on the energy of their relative motion, are internal parameters. The suggested quasihard-sphere model enables one to simulate elastic processes of scattering of atomic particles, using different interatomic potentials of interaction with relatively high rates of statistical simulation characteristic of simulation within the hard-sphere model. The Born–Mayer potential is selected as the interatomic potential of interaction and modified for a wide class of partners in atomic collisions. It is demonstrated that the suggested mathematical model of quasihard spheres describes fairly correctly the processes of elastic scattering of atoms in a gas medium and of displaced atoms in a solid with an almost constant rate of static simulation. © *2000 MAIK "Nauka/Interperiodica".*

INTRODUCTION

In order to perform numerical simulation of the processes of interaction of atomic particles in a gas and in a solid by the Monte Carlo method, one must determine a number of basic stochastic variables which describe adequately the processes of particle scattering. In describing the processes of elastic interaction of atomic particles, the main characteristic is provided by the scattering angle which defines the energy loss and the subsequent behavior of their motion. The scattering angle relates to each other the impact parameter b, the interatomic potential of interaction $U(r)$, and the energy of relative motion of particles E_c. Given a spherically symmetric potential of interaction, the scattering angle Θ in a center-of-mass-system is described by the expression [1]

$$\Theta = \pi - 2b\int_{r_{\min}}^{\infty} \frac{dr/r^2}{[1 - U(r)/E_c - b^2/r^2]^{1/2}}, \qquad (1)$$

where r is the interatomic distance; E_c is the kinetic energy of relative motion of atomic particles in the center-of-mass system for $r \longrightarrow \infty$; and $r_{\min}$ is the shortest distance within which the particles come closer together, which is the root of the radicand in the denominator.

Expression (1) in the analytical form may be integrated only for the hard-sphere potential and for a number of power potentials and their linear combinations. In describing the processes of elastic scattering of atomic particles for more real interatomic potentials of interaction, one restricts oneself to the use of various approximate methods within both classical and quantum-mechanical description [2, 3].

The complexity of the computational procedure during numerical simulation of the processes of particle scattering by the Monte Carlo method is largely defined by the choice of the interatomic potential of interaction. In so doing, one must make a sound compromise between the real physical description of the interaction processes and relative simplicity of the computational procedure during their mathematical simulation. The use of the interatomic potential of hard spheres results in a significant simplification of the calculation procedure, first of all, during statistical simulation of the process of elastic scattering of atomic particles in a gas and in a solid. However, an important disadvantage of this interaction potential is the absence of correlation between the interaction cross section and the energy of relative motion of colliding particles.

In the case of particle interaction in a gas medium in the range of low energy values which do not exceed the respective ionization potentials, the elastic-scattering cross section is of the order of gas-kinetic and little depends on the energy of colliding particles. In this case, the classical hard-sphere scattering is a good approximation; in this energy range, the particle scattering (except for the case of small values of scattering angles) is assumed to be spherically symmetric, and the interaction between colliding particles is defined by their outer electron shells and must be determined for each pair of colliding particles. When the energy of relative motion of colliding particles increases, significant scattering occurs under conditions of considerable overlapping of their electron shells, and the interaction potential is largely defined by the inner electrons whose velocities are much higher than the collision rates of atomic particles. The excitation of outer electrons occurring in the process causes little variation in the

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scattering potential the effective range of whose action corresponds to smaller interatomic distances.

In describing the interaction of atomic particles in a solid, an empirical rule exists according to which the collisions leading to an appreciable scatter occur at distances of the order of half the equilibrium distance between neighboring atoms. In the case of such small distances, one can ignore the long-range attractive forces which define the bonding forces in solids. By the order of magnitude, this range of distances corresponds to the sizes of colliding atomic particles. Based on the same principle is the method of determining, to a first approximation, ionic and atomic radii [4]. Therefore, in a fairly wide range of energies of colliding atomic particles, the classical hard-sphere scattering is a good approximation from the practical standpoint.

In the hard-sphere model, the shortest distance within which two colliding particles come closer together, $r_{\min}$, for any values of the impact parameter b is always equal to the sum of radii of atomic particles and does not depend on the energy of their relative motion. This results in a considerable limitation, within the hard-sphere model, of the possibility of static simulation in a wide range of energy of colliding particles. Attempts were made previously [5–7] at describing the process of elastic scattering of atomic particles using the procedure of fitting the approximate potential of interaction to the real one for some distance between the atomic particles which makes the most contribution to the particle scattering. However, almost all of the derived approximate potentials of interaction are little valid for high impact parameters which lead to overestimated values of transmitted energy under conditions of elastic collision of atomic particles. The procedure of simulation of the processes of elastic scattering of atomic particles, suggested by us, restricts the range of high impact parameters at thermal energies of atomic particles by their gas-kinetic sizes. For high values of the energy of collision of atomic particles, the range of high impact parameters corresponds only to very small values of scattering angles and transmitted energy and plays no important part.

APPROXIMATION OF INTERATOMIC INTERACTION POTENTIAL

A combination of the simplicity of computational procedure in using the hard-sphere potential with the correctness of physical description of the processes of interaction of atomic particles may be accomplished by using the interatomic potential of quasihard spheres (QHS). We will treat in more detail the form and procedure of using the interatomic potential of interaction of QHS. A number of test potentials applicable to atoms of various elements and containing fitting parameters may be used as real interatomic potentials of interaction. Most convenient from the standpoint of mathematical application is the Born–Mayer interatomic potential [8]

$$U_{B-M}(r) = A_{B-M}(Z_1Z_2)^{3/4}\exp[-r/b_{B-M}], \qquad (2)$$

where Z_1 and Z_2 are ordinal (atomic) numbers of colliding atomic particles, and A_{B-M} and b_{B-M} are constants determined for each pair of colliding atomic particles. This purely exponential interatomic potential of interaction enables one to analytically express the shortest distance $r_{\min}$ within which two colliding atomic particles come closer together from the equation

$$U_{B-M}(r_{\min}) = E_c, \qquad (3)$$

where E_c is the energy of relative motion in the center-of-mass system of two colliding atomic particles; the solution of this latter equation has the form

$$r_{\min}(E_c) = -b_{B-M}\ln\frac{E_c}{A_{B-M}(Z_1Z_2)^{3/4}} \qquad (4)$$

and, in the case of central collision ($b = 0$), defines the minimum distance between two atomic particles at the point of stopping during infinite motion of the incident particle. The use of other, more complex real interatomic potentials, whose solution relative to $r_{\min}$ does not permit an analytical solution for different values of the collision energy, is possible; however, it complicates the computational procedure of simulation and renders it less flexible as regards the adaptation to variations of conditions of real physical experiment.

If the value of the shortest distance $r_{\min}$ within which the atomic particles come closer together is identified with the sum of the radii of hard spheres at the point of contact (Fig. 1), one can use the hard-sphere model to determine the microscopic cross section for elastic scattering and the free path. In so doing, the sum of the hard-sphere radii is a variable quantity and varies as a function of the energy of relative motion of colliding atomic particles; from this standpoint, colliding atomic particles may be regarded as quasihard spheres.

The interatomic potential of quasihard spheres may be determined in the form

$$U_{\text{qhs}}(r) = \begin{cases}\infty & \text{for } r < r_{\text{hs}} = r_{\min}(E_c)\\ 0 & \text{for } r > r_{\text{hs}} = r_{\min}(E_c),\end{cases} \qquad (5)$$

where $r_{\min}(E_c)$ is the solution of equation (3) and, for the Born–Mayer interatomic potential (2), is defined by expression (4).

The criterion of validity of interatomic potential of interaction of quasihard spheres $U_{\text{qhs}}(r)$ may be formulated as follows:

$$\Delta r = r'_{\text{hs}} - r_{\text{hs}} < r_{\text{hs}} = r_{\min}(E_c), \qquad (6)$$

where r'_{hs} is the coordinate of intersection of the tangent (derivative of the real interatomic potential of

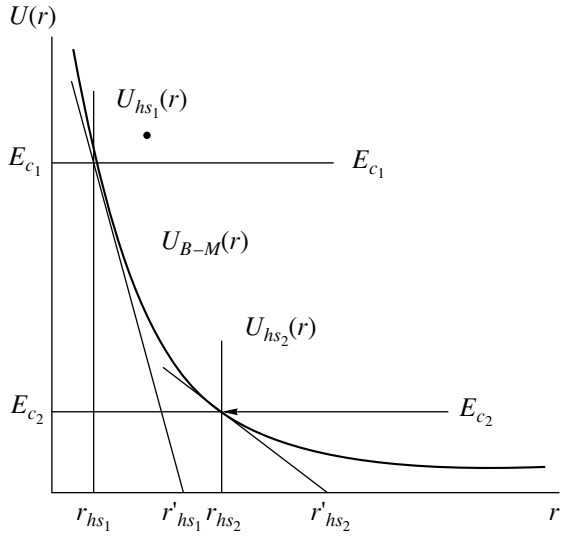

Fig. 1. Quasihard-sphere approximation for the potential $U_{B-M}(r)$ (central collision, $b = 0$).

interaction) drawn from the point $U_{\text{qhs}}(r) = E_c$ with the axis of interatomic distance r (Fig. 1).

The use of the interatomic potential of interaction of quasihard spheres $U_{\text{qhs}}(r)$ will be correct in case conditions (6) is valid or the equivalent relation depending on the collision energy E_c,

$$\frac{r}{\Delta r} = r\frac{|dU(r)/dr|}{U(r)} = r\frac{d\ln U(r)}{dr}\bigg|_{r = r_{\text{hs}}} > 1. \qquad (7)$$

If this criterion is generalized to the region of off-center ($b \neq 0$) collisions, it will take the form

$$\frac{r}{\Delta r} = r\frac{|dU(r)/dr|}{U(r)} > 1 - b^2/r^2. \qquad (8)$$

If this criterion of interatomic potential of interaction of quasihard spheres (7) is applied to the real Born–Mayer interaction potential (2), we will derive the range of validity of the respective of quasihard-sphere interaction potential,

$$r(E_c) > b_{B-M} \approx 0.219 \text{ E}. \qquad (9)$$

One can see from condition (9) that the potential of interaction of quasihard spheres with the Born–Mayer interatomic potential is well valid in the range of great interatomic distances to which correspond both low values of the collision energy and high values of the impact parameter in a wide range of the collision energy of atomic particles. For the exponential Born–Mayer interaction potential (2), this is associated with the fact that, as the interatomic distance r increases (Fig. 1), the quantity $\Delta r = r'_{\text{hs}} - r_{\text{hs}}$ increases slower than the quantity $r_{\text{hs}} = r_{\min}(E_c)$, and their correlation ever better satisfies the criterion of validity (6) of the quasihard-sphere interaction potential.

In order to use the Born–Mayer interatomic potential in the quasihard-sphere model, we modified it using the results of Abrahamson [9] for a wide class of collision partners with the ordinal numbers $Z_{1,2} = 2\ldots80$. In so doing, the constants A_{B-M} and b_{B-M} in the Born–Mayer interatomic potential (2) were represented as the functions $A_{B-M}(Z_1, Z_2)$ and $b_{B-M}(Z_1, Z_2)$ and approximated using the results of [9, 10] with the power functions by the method of least squares. The approximation results are given in Fig. 2. The obtained coefficients of the modified Born–Mayer interatomic potential (2) have the form

$$\begin{aligned} A^{ap}_{B-M}(Z_1, Z_2) &= 95.863(Z_1Z_2)^{0.7383}, \text{ eV}, \\ b^{ap}_{B-M}(Z_1, Z_2) &= 0.122(Z_1^{0.0387} + Z_2^{0.0387}), \text{ Å}. \end{aligned} \qquad (10)$$

The maximum relative error of approximation of the coefficients $A^{ap}_{B-M}(Z_1, Z_2)$ and $b^{ap}_{B-M}(Z_1, Z_2)$, corresponding to the collision of the lightest atomic particles, does not exceed 8% and, in the case of heavy atomic particles, decreases to 3%. In so doing, the modified Born–Mayer interatomic potential of interaction of atomic particles with the ordinal numbers Z_1 and Z_2 assumes the form

$$\begin{aligned} \tilde{U}_{B-M} &= 95.863(Z_1Z_2)^{0.7383} \\ &\times \exp\left[-\frac{r}{0.122(Z_1^{0.0387} + Z_2^{0.0387})}\right], \end{aligned} \qquad (11)$$

where $\tilde{U}_{B-M}$ and r are in eV and Å, respectively.

The solution of equation (3) for this modified Born–Mayer interatomic potential (11) has the form

$$\begin{aligned} r_{\min}(E_C) &= -0.122(Z_1^{0.0387} + Z_2^{0.0387}) \\ &\times \ln\frac{E_C}{95.863(Z_1Z_2)^{0.7383}E}. \end{aligned} \qquad (12)$$

The criterion of validity (7) of the potential of interaction of quasihard spheres with the derived modified Born–Mayer potential (11) takes the form

$$r(E_c) > 0.122(Z_1^{0.0387} + Z_2^{0.0387})E. \qquad (13)$$

Within the obtained quasihard-sphere model, the microscopic cross section for elastic interaction of atomic particles depends on the energy E_C of their relative motion,

$$\sigma_{\text{qhs}} = \pi r^2_{\min}(E_C) \qquad (14)$$

and, accordingly, the free path λ_{qhs} of atomic particles in a gas medium or in a solid is

$$\lambda_{\text{qhs}} = \frac{1}{N\pi r^2_{\min}(E_C)}, \qquad (15)$$

where N is the concentration of atomic particles in a scattering medium.

If, in statistical simulation, the ratio between b^2 and the quantity $r^2_{\min}(E_c)$ is equated to a random number ξ, the expression for the scattering angle ϑ in the laboratory coordinate system assumes the form

$$\vartheta = \arctan\frac{2[\xi(1-\xi)]^{1/2}}{2\xi - 1 + M_1/M_2}. \quad (16)$$

In determining the impact parameter b in the form

$$b = r_{\min}(E_c)\xi^{1/2}, \quad (17)$$

we take into account the increase in the probability of high (in absolute magnitude) values of the impact parameter b, which is due to the fact that an incident atomic particle arrives by chance at different points of the area of the microscopic cross section for scattering.

Given in Fig. 3 by way of example are the results of calculations of free path (15) of Cu atoms under conditions of elastic scattering from Ar atoms, obtained within the quasihard-sphere model with the modified Born–Mayer interatomic potential (11) and normalized to the respective value of free path in the hard-sphere model, as a function of the energy of their relative motion.

The calculation results indicate that the microscopic cross section of elastic scattering of atomic particles $\sigma_{qhs} \sim 1/\lambda_{qhs}$ increases, as the energy of their relative motion decreases, to reach the value of gas-kinetic cross section at thermal collision energies. As the collision energy increases, the elastic scattering cross section decreases to a value restricted by the criterion of validity (7) of the quasihard-sphere model. The maximum collision energy corresponding to the limit of the criterion of validity of the quasihard-sphere model (13) using the modified Born–Mayer interatomic potential (11), under conditions of elastic scattering of Cu atoms from Ar atoms is restricted to ~5 keV. This energy range of interaction of atomic particles is of interest from the standpoint of numerous applied problems of the physics of plasma, gas discharge, and solid.

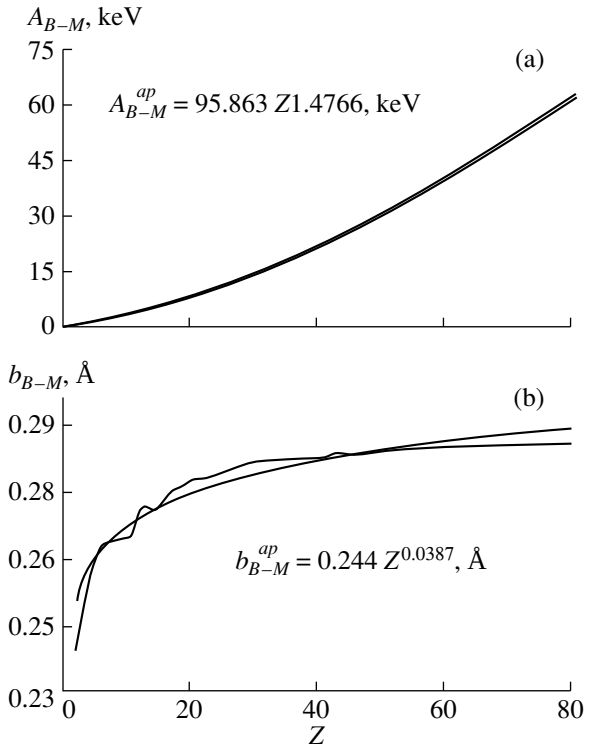

Fig. 2. Approximation of coefficients in the modified Born–Mayer interatomic potential (11): (a) $A^{ap}_{B-M}(Z_1, Z_2)$, (b) $b^{ap}_{B-M}(Z_1, Z_2)$.

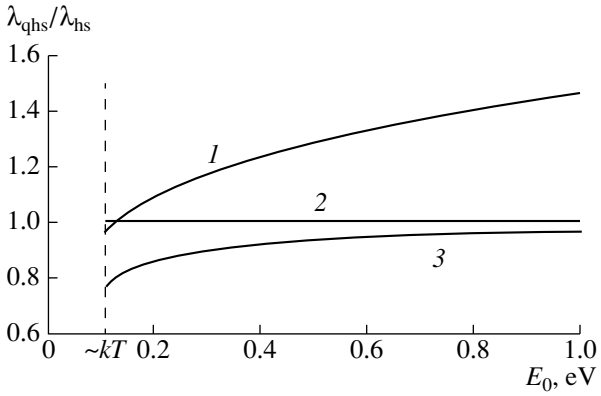

Fig. 3. The free path of Cu atoms upon elastic scattering from Ar atoms as a function of the energy of their relative motion: (*1*) qhs, (*2*) hs, (*3*) [13].

DISCUSSION

Previous attempts have been made [11–14] to introduce into the hard-sphere model the dependence of the interaction cross section on the energy of colliding particles. However, as is seen in Fig. 3 (curve *3*), the most rigorous energy dependence of the elastic-scattering cross section, proposed in [13], which includes the Maxwellian velocity distribution of gas atoms, is little valid in the entire range of energy of colliding particles and reaches the value of gas-kinetic interaction cross section at high energies of colliding particles, which is incorrect.

The application of more real interatomic potentials of interaction results in the necessity of using mathematical procedures which introduce some determinism into the random process of scattering. For example, the use in [14] of the scattering "6–12" Lennard–Jones potential by the procedure of linearization of the scattering angle Θ in the center-of-mass from the impact parameter b enabled one to estimate the maximum

interaction cross section from the condition $\Theta = 0$ at $b = b_{max}$. However, the linear approximation in this case implies that the scattering angle Θ is estimated with inadequate accuracy with the value of the impact parameter b close to maximum. This results in a considerable underestimation of the contribution of scattering in the case of glancing collision and, accordingly, the value of the interaction cross section.

In order to check the degree of the fit of the quasihard-sphere model to the description of processes of elastic scattering of atomic particles involving the use of real interatomic potentials, the results of simulation of processes of transport of atomic particles in a gas were compared to those in a solid. The real interatomic potential of interaction was provided by the modified Firsov potential [15],

$$U(r) = \frac{z_1 z_2 e^2}{r}\psi\left(-\frac{r}{a_f}\right), \qquad (18)$$

where $a_f = 0.8853a_0/(Z_1^{1/2} + Z_2^{1/2})^{2/3}$ is Firsov's screening parameter (a_0 is the Bohr radius) with Nikulin's screening function [16] obtained by approximate solution of the Thomas–Fermi equation using the variation principle,

$$\psi_c(r) = [a\exp(-\alpha/a_f r) + b\exp(\beta/a_f r)]^2, \qquad (19)$$

where $a = 0.7111$, $b = 0.2889$, $\alpha = 0.175$, and $\beta = 1.6625$.

In order to perform numerical integration in expression (1) when determining the scattering angle Θ with interaction potential (18), we developed a mathematical procedure according to which the integration interval $[r_{min}, \infty]$ in expression (1) is divided into three regions.

In the first region $r_{min} < r < (r_{min} + \delta)$, the integrand in the denominator of expression (1) $f(r) = 1 - U(r)/E_c - b^2/r^2$ is expanded by its Taylor series expansion,

$$\begin{aligned} f(r) &= f(r_{min}) + f'(r_{min})(r - r_{min}) \\ &\quad + f''(r_{min})(r - r_{min})^2/2 + \ldots \\ &\approx (r - r_{min})(f'(r_{min}) + f''(r_{min})(r - r_{min})/2). \end{aligned} \qquad (20)$$

The relative error of these transformations is estimated at

$$\Delta = \frac{f'''(r_{min})}{f'(r_{min})} - \frac{(r - r_{min})^2}{6}.$$

At $\delta = 0.01$ ($r = r_{min} + 0.01$), the value of Δ does not exceed $\sim 10^{-4}$.

The upper limit Q of the second region $(r_{min} + \delta) \le r \le Q$. This is the value of r at which the second term $U(r)/E_c$ of the function $f(r)$ becomes small and, in what follows, may be ignored. We took $U(r)/E_c < 10^{-6}$ as the smallness criterion.

In the third region $Q < r < \infty$, the integral in expression (1) has an analytical solution in the form

$$\int_Q^\infty \frac{dr/r^2}{[1 - U(r)/E_c - b^2/r^2]^{1/2}} = \frac{1}{b}\arcsin\left(\frac{b}{Q}\right). \qquad (21)$$

It is almost impossible to perform numerical integration in expression (1) directly in the process of simulation, because this extends considerably the time of static simulation. Therefore, the values of scattering angles $\Theta(b, E_c)$ were first calculated for a wide range of values of the energy E_c of relative motion of colliding atomic particles, with the range of values of the impact parameter b from 0 to b_{max} corresponding to each one of those energy values. The value of the maximum impact parameter b_{max} for each value of the collision energy E_c was determined from the condition $\Theta(b = b_{max}) \approx 0$. The calculation results were used to compile a two-dimensional array of values of the scattering angles $\Theta(b, E_c)$, between the elements of which a spline approximation was performed. This two-dimensional array $\Theta(b, E_c)$, calculated for concrete collision partners, was used in static simulation to determine the value of the scattering angle Θ in each collision event.

In order to compare the results of statistical simulations involving the interatomic quasihard-sphere potential (5), the modified Born–Mayer interaction potential (11), and the real potential of interaction (18), the processes of transport of Cu, Y, and Ba atoms in a medium of Ar and O_2 were calculated.

An analysis of results of statistical simulation reveals that both models agree in the low-energy region and differ slightly when the collision energy increases. This may be due to the fact that modified Firsov's potential (18) with Nikulin's screening function (19) in the case of in the case of large internuclear distances decreases slower than the true interaction potential. Moreover, the modified Born–Mayer interaction potential (11), employed by us in the quasihard-sphere model, is more valid in the case of large internuclear distances which are characterized by interactions between the outer electron shells of colliding atomic particles. The quasihard-sphere model with modified Born–Mayer interaction potential adapts itself better to various combinations of collision partners, because it contains parameters characteristic of concrete pairs of colliding atomic particles.

It is almost impossible to derive the universal interatomic potential of interaction in an analytical form, which could be used in application to a wide class of problems in statistical simulation: even in describing the processes of scattering of particles in a gas medium due to the differences in the electron structure of colliding atoms the real interatomic potential of interaction is not monotonic, and oscillations due to the shell structure of colliding atoms must show up in elastic-scattering cross sections. In addition, the assumptions made in

deriving almost all of the known interatomic potentials of interaction used to describe the processes of scattering of particles in a solid were obtained within the statistical theory of scattering and based on the principles of binarity and independence of collisions characteristic of a gas medium, which is incorrect.

Within the obtained quasihard-sphere model (5) using the modified Born–Mayer interatomic potential (11), calculations were performed of the transport coefficients of atoms in a gas medium at different values of gas pressure up to pressures at which the diffusion motion of atoms being scattered is predominating. Also calculated were the values of ion sputtering in describing the processes of displacement of atoms in a solid and their motion toward the surface. The calculation results obtained for a wide class of collision partners have demonstrated that the suggested model of quasihard-sphere model using the modified Born–Mayer interatomic potential enables one to fairly correctly simulate the processes of elastic scattering of atomic particles at high rates of statistical simulation.

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