

# An Algorithm for Computing Gradients of Functionals in Structural Materials Science

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## Abstract

When describing and modeling the crystal structure of a material, interatomic interaction potentials are used. To solve the problems of parametric identification of potentials, which consists of the selection of potential parameters, the use of various optimization methods has recently become increasingly important. In this case, it becomes necessary to determine the gradients of some quantities characterizing the substance with respect to parameters of potential. Based on the Fast Automatic Differentiation-technique, an algorithm has been built to determine the exact values of the gradients of bulk modulus and shear modulus of the test substance with respect to potential parameters in the case when the total interatomic energy of the atoms' system is determined using the Tersoff's potential.

## 1 Introduction

Recently, optimization problems from the field of structural materials science have become more relevant, among which the well-known problems of identifying the parameters of mathematical models stand out.

Various mathematical models are used to study the atomic structures of materials. Some parameters of these models are unknown. They should be identified from the condition that the calculated properties of the modeled material are close to its properties, which were found experimentally.

When describing and modeling the crystal structure of a material, interatomic interaction potentials are used. To describe the properties of crystals with a covalent bond (for example, carbon, silicon, germanium, etc.), Tersoff's potential is often used ([Tersoff1988]). Structural identification of potentials for a specific crystalline material is one of the important stages of molecular dynamics modeling.

The problem of parametric identification of potentials is often reduced to some optimization problem (see, for example, [Abgaryan2014]), which can be formally written as a problem of minimizing the following functional:

$$f(\xi) = \sum_{i=1}^m \omega_i (y_i(\xi) - \tilde{y}_i)^2 \quad (1)$$

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In: Sergei S. Goncharov, Yuri G. Evtushenko (eds.): Proceedings of the Workshop on Applied Mathematics and Fundamental Computer Science 2021, Omsk, Russia, 24-04-2021, published at <http://ceur-ws.org>

where  $\omega_i$  is the weight coefficient;  $\tilde{y}_i$  is the value of the  $i$ -th material characteristic obtained experimentally, and  $y_i(\xi)$  is the value of the same material characteristic calculated using Tersoff potential determined by the components of the vector  $\xi$  ( $\xi \in R^m$  is a vector to be identified). The solution to the problem is looked for on the set  $X \subseteq R^m$ , which is a parallelepiped. Its boundaries are chosen so that it obviously contained the admissible range of parameters. The number of items in the formula (1) varies depending on the studied material. For the numerical solution of this problem, gradient minimization methods are often used. There exists the need to efficiently calculate the exact value of the cost function gradient with respect to parameters of Tersoff potential.

These derivatives are often calculated (in particular, see [Abgaryan2014]) using the finite difference method. Studies have shown that the finite difference method does not allow the calculation of the gradient of a cost function with acceptable accuracy and requires  $(m + 1)$  times to calculate the value of the function.

One of the terms in the formula (1) is the total energy of the system of atoms. As the interatomic potential energy, the Tersoff potential was chosen. The other two terms in the formula (1) are bulk modulus and shear modulus of a material. In [Albu2016], using the Fast Automatic Differentiation-technique (FAD-technique) (see [Evtushenko1998]), formulas to calculate the exact gradient of the total energy with respect to parameters of Tersoff potential (specific for modeled substance) were received.

In this paper, with the help of FAD-technique, we derived formulas to calculate the gradients of bulk modulus and shear modulus of material with respect to Tersoff parameters with machine precision.

## 2 Formulation of the Problem

The energy  $E$  of a atoms' system is calculated with the help of expression  $E = \sum_{i=1}^I \sum_{\substack{k=1 \\ k \neq i, j}}^I V_{ij}$ , where  $V_{ij}$  is the

interaction potential between atoms marked  $i$  and  $j$  ( $i$ -atom and  $j$ -atom). In present paper the Tersoff potential is used as interaction potential:

$$V_{ij} = f_c(r_{ij}) (V_R(r_{ij}) - b_{ij}V_A(r_{ij})),$$

$$V_{ij}^R = V_R(r_{ij}) = \frac{D_e}{S-1} \exp\left(-\beta\sqrt{2S}(r_{ij} - r_e)\right), \quad V_{ij}^A = V_A(r_{ij}) = \frac{SD_e}{S-1} \exp\left(-\beta\sqrt{\frac{2}{S}}(r_{ij} - r_e)\right),$$

$$f_c(r) = \begin{cases} 1, & r < R - R_{cut}, \\ \frac{1}{2} \left(1 - \sin\left(\frac{\pi(r - R)}{2R_{cut}}\right)\right), & R - R_{cut} < r < R + R_{cut}, \\ 0, & r > R + R_{cut}, \end{cases}$$

$$b_{ij} = (1 + (\gamma\zeta_{ij})^\eta)^{-\frac{1}{2\eta}}, \quad \zeta_{ij} = \sum_{\substack{k=1 \\ k \neq i, j}}^I f_c(r_{ik})g_{ijk}\omega_{ijk}, \quad \omega_{ijk} = \exp(\lambda^3\tau_{ijk}),$$

$$\tau_{ijk} = (r_{ij} - r_{ik})^3, \quad g_{ijk} = 1 + \left(\frac{c}{d}\right)^2 - \frac{c^2}{d^2 + (h - \cos\Theta_{ijk})^2}.$$

Here  $I$  is the number of atoms in considered system;  $r_{ij}$  is the distance between  $i$ -atom and  $j$ -atom;  $\Theta_{ijk}$  is the angle between two vectors, first vector begins at  $i$ -atom and finishes at  $j$ -atom, second vector begins at  $i$ -atom and finishes at  $k$ -atom;  $R$  and  $R_{cut}$  are known parameters, identified from experimental geometric properties of substance. Tersoff Potential depends on ten parameters ( $m = 10$ ), specific to modeled substances:  $D_e, r_e, \beta, S, \eta, \gamma, \lambda, c, d, h$ .

The distance between  $i$ -atom and  $j$ -atom is determined by the formula:

$$r_{ij} = \sqrt{(x_{1i} - x_{1j})^2 + (x_{2i} - x_{2j})^2 + (x_{3i} - x_{3j})^2},$$

where  $x_{1i}, x_{2i}, x_{3i}$  are the Cartesian coordinates of  $i$ -atom. If  $\Theta_{ijk}$  is the angle between two vectors, connecting  $i$ -atom with  $j$ -atom and  $k$ -atom respectively, then

$$\cos\Theta_{ijk} = q_{ijk} = (r_{ij}^2 + r_{ik}^2 - r_{jk}^2)/(2r_{ij}r_{ik}).$$

Let  $a$  be the initial length of the edges of the lattice of atoms;  $\tilde{a} = \alpha a$  ( $\alpha \in R$ ) — length of the edges of the lattice of atoms after deformation;  $\rho = \tilde{a} - a$  — deformation parameter. Then  $\frac{a + \rho}{a} = \left(1 + \frac{\rho}{a}\right) a$ . If  $\bar{r}_k = (x_{k1}, x_{k2}, x_{k3})$  are the coordinates of some lattice atom before deformation and  $\tilde{r}_k = (\tilde{x}_{k1}, \tilde{x}_{k2}, \tilde{x}_{k3})$  are its coordinates after deformation, then  $\tilde{x}_{k1} = \left(1 + \frac{\rho}{a}\right) x_{k1}$ ,  $\tilde{x}_{k2} = \left(1 + \frac{\rho}{a}\right) x_{k2}$ ,  $\tilde{x}_{k3} = \left(1 + \frac{\rho}{a}\right) x_{k3}$ .

Let  $E(\bar{r}_1, \bar{r}_2, \dots, \bar{r}_I)$  is the total energy of atoms system before deformation. Then  $E(\tilde{r}_1, \tilde{r}_2, \dots, \tilde{r}_I) = E\left[\left(1 + \frac{\rho}{a}\right) \bar{r}_1, \left(1 + \frac{\rho}{a}\right) \bar{r}_2, \dots, \left(1 + \frac{\rho}{a}\right) \bar{r}_I\right]$  is the total energy of atoms system after deformation. The bulk modulus and the shear modulus of the material are proportional to  $B(E)$ , that can be calculated by formula:

$$B(E) = \frac{\partial^2}{\partial \rho^2} E \left[ \left(1 + \frac{\rho}{a}\right) \bar{r}_1, \left(1 + \frac{\rho}{a}\right) \bar{r}_2, \dots, \left(1 + \frac{\rho}{a}\right) \bar{r}_I \right] \Big|_{\rho=0}.$$

Therefore, to calculate the gradients of the bulk modulus and shear modulus of the material with respect to Tersoff parameters, it is sufficient to calculate the gradient of the function  $B(E)$  with respect to these parameters.

### 3 Algorithm for Calculating the Gradient of a Function $B(E)$

Let us construct the multistep algorithm to calculate the total energy  $E$  of atoms' system (interaction potential is Tersoff Potential). For compactness further in the study we introduce vectors  $\bar{u}$  and  $\bar{z}$  having the following coordinates:  $\bar{u}^T = [u_1, u_2, \dots, u_{10}]^T$ ,  $\bar{z}^T = [z_1, z_2, \dots, z_{10}]^T$ , where  $u_1 = D_e$ ,  $u_2 = r_e$ ,  $u_3 = \beta$ ,  $u_4 = S$ ,  $u_5 = \eta$ ,  $u_6 = \gamma$ ,  $u_7 = \lambda$ ,  $u_8 = c$ ,  $u_9 = d$ ,  $u_{10} = h$ ;

$$\begin{aligned} z_1 &= \left\{ z_1^{ijk} = \sqrt{(x_{1i} - x_{1k})^2 + (x_{2i} - x_{2k})^2 + (x_{3i} - x_{3k})^2} \right\} \equiv F(1, Z_1, U_1), \\ z_2 &= \left\{ z_2^{ijk} = \sqrt{(x_{1j} - x_{1k})^2 + (x_{2j} - x_{2k})^2 + (x_{3j} - x_{3k})^2} \right\} \equiv F(2, Z_2, U_2), \\ z_3 &= \left\{ z_3^{ijk} = q_{ijk} = \frac{(z_{13}^{ij})^2 + (z_1^{ijk})^2 - (z_2^{ijk})^2}{2z_1^{ijk} z_{13}^{ij}} \right\} \equiv F(3, Z_3, U_3), \\ z_4 &= \left\{ z_4^{ijk} = f_c(z_1^{ijk}) \right\} \equiv F(4, Z_4, U_4), \\ z_5 &= \left\{ z_5^{ijk} = g_{ijk} = 1 + \left(\frac{u_8}{u_9}\right)^2 - \frac{(u_8)^2}{(u_9)^2 + (u_{10} - z_3^{ijk})^2} \right\} \equiv F(5, Z_5, U_5), \\ z_6 &= \left\{ z_6^{ijk} = \tau_{ijk} = (z_{13}^{ij} - z_1^{ijk})^3 \right\} \equiv F(6, Z_6, U_6), \\ z_7 &= \left\{ z_7^{ijk} = \omega_{ijk} = \exp((u_7)^3 z_6^{ijk}) \right\} \equiv F(7, Z_7, U_7), \\ z_8 &= \left\{ z_8^{ijk} = f_c(r_{ik}) g_{ijk} \omega_{ijk} = z_4^{ijk} z_5^{ijk} z_7^{ijk} \right\} \equiv F(8, Z_8, U_8), \\ z_9 &= \left\{ z_9^{ij} = \zeta_{ij} = \sum_{k=1; k \neq i, j}^I z_8^{ijk} \right\} \equiv F(9, Z_9, U_9), \\ z_{10} &= \left\{ z_{10}^{ij} = \gamma \zeta_{ij} = u_6 z_9^{ij} \right\} \equiv F(10, Z_{10}, U_{10}), \\ z_{11} &= \left\{ z_{11}^{ij} = (\gamma \zeta_{ij})^\eta = (z_{10})^{u_5} \right\} \equiv F(11, Z_{11}, U_{11}), \\ z_{12} &= \left\{ z_{12}^{ij} = b_{ij} = (1 + z_{11}^{ij})^{-\frac{1}{2u_5}} \right\} \equiv F(12, Z_{12}, U_{12}), \\ z_{13} &= \left\{ z_{13}^{ij} = \sqrt{(x_{1i} - x_{1j})^2 + (x_{2i} - x_{2j})^2 + (x_{3i} - x_{3j})^2} \right\} \equiv F(13, Z_{13}, U_{13}), \\ z_{14} &= \left\{ z_{14}^{ij} = V_{ij}^R = \frac{u_1}{u_4 - 1} \exp\left(-u_3 \sqrt{2u_4} (z_{13}^{ij} - u_2)\right) \right\} \equiv F(14, Z_{14}, U_{14}), \\ z_{15} &= \left\{ z_{15}^{ij} = V_{ij}^A = \frac{u_1 u_4}{u_4 - 1} \exp\left(-u_3 \sqrt{\frac{2}{u_4}} (z_{13}^{ij} - u_2)\right) \right\} \equiv F(15, Z_{15}, U_{15}), \\ z_{16} &= \left\{ z_{16}^{ij} = f_c(z_{13}^{ij}) \right\} \equiv F(16, Z_{16}, U_{16}), \\ z_{17} &= \left\{ z_{17}^{ij} = V_{ij} = z_{16}^{ij} (z_{14}^{ij} - z_{12}^{ij} z_{15}^{ij}) \right\} \equiv F(17, Z_{17}, U_{17}), \end{aligned}$$

$$(i = \overline{1, I}, \quad j = \overline{1, I}, \quad j \neq i, \quad k = \overline{1, I}, \quad k \neq i, j).$$

The energy  $E$  of the atoms' system with the help of new variables may be rewritten as follows:  $E(z(\bar{u})) = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I z_{17}^{ij}(\bar{u})$ . Variables  $z_1, z_2, \dots, z_{17}$  (the phase variables) are determined by the specified above multistep

algorithm  $z_l = F(l, Z_l, U_l)$ , ( $l = 17$ ), where  $Z_l$  is the set of elements  $z_n$  in the right part of the equation  $z_l = F(l, Z_l, U_l)$ , and  $U_l$  is the set of elements  $u_n$  that appear in the right side of this equation. Note that each component  $z_l$  depends on a number of other components ( $z_l^{ij}$  or  $z_l^{ijk}$ ).

Let us introduce also the following designations:  $\tilde{z}_1, \tilde{z}_2, \dots, \tilde{z}_{17}$  and  $\tilde{\tilde{z}}_1, \tilde{\tilde{z}}_2, \dots, \tilde{\tilde{z}}_{17}$ , where

$$\begin{aligned} \tilde{z}_n &= \left\{ \tilde{z}_n^{ijk} : \tilde{z}_n^{ijk} = \frac{\partial z_n^{ijk}(\tilde{r}_{ij}, \tilde{r}_{ik}, \tilde{r}_{jk})}{\partial \rho} \Big|_{\rho=0} = \frac{\partial z_n^{ijk} \left( (1 + \frac{\rho}{a}) r_{ij}, (1 + \frac{\rho}{a}) r_{ik}, (1 + \frac{\rho}{a}) r_{jk} \right)}{\partial \rho} \Big|_{\rho=0} \right\}, \quad n = \overline{1, 8}, \\ \tilde{z}_n &= \left\{ \tilde{z}_n^{ij} : \tilde{z}_n^{ij} = \frac{\partial z_n^{ij}(\tilde{r}_{ij})}{\partial \rho} \Big|_{\rho=0} = \frac{\partial z_n^{ij} \left( (1 + \frac{\rho}{a}) r_{ij} \right)}{\partial \rho} \Big|_{\rho=0} \right\}, \quad n = \overline{9, 17}, \\ \tilde{\tilde{z}}_n &= \left\{ \tilde{\tilde{z}}_n^{ijk} : \tilde{\tilde{z}}_n^{ijk} = \frac{\partial^2 z_n^{ijk}(\tilde{r}_{ij}, \tilde{r}_{ik}, \tilde{r}_{jk})}{\partial \rho^2} \Big|_{\rho=0} = \frac{\partial^2 z_n^{ijk} \left( (1 + \frac{\rho}{a}) r_{ij}, (1 + \frac{\rho}{a}) r_{ik}, (1 + \frac{\rho}{a}) r_{jk} \right)}{\partial \rho^2} \Big|_{\rho=0} \right\}, \quad n = \overline{1, 8}, \\ \tilde{\tilde{z}}_n &= \left\{ \tilde{\tilde{z}}_n^{ij} : \tilde{\tilde{z}}_n^{ij} = \frac{\partial^2 z_n^{ij}(\tilde{r}_{ij})}{\partial \rho^2} \Big|_{\rho=0} = \frac{\partial^2 z_n^{ij} \left( (1 + \frac{\rho}{a}) r_{ij} \right)}{\partial \rho^2} \Big|_{\rho=0} \right\}, \quad n = \overline{9, 17}, \end{aligned}$$

$$(i = \overline{1, I}, \quad j = \overline{1, I}, \quad j \neq i, \quad k = \overline{1, I}, \quad k \neq i, j).$$

The above values are calculated by the formulas:

$$\begin{aligned} \tilde{z}_1^{ijk} &= r_{ik}/a; & \tilde{z}_2^{ijk} &= r_{jk}/a; & \tilde{z}_3^{ijk} &= 0; & \tilde{z}_4^{ijk} &= \frac{\partial f_c \left( (1 + \frac{\rho}{a}) r_{ik} \right)}{\partial \rho} \Big|_{\rho=0}; \\ \tilde{z}_5^{ijk} &= 0; & \tilde{z}_6^{ijk} &= 3z_6^{ijk}/a; & \tilde{z}_7^{ijk} &= 3z_6^{ijk} z_7^{ijk} (u_7)^3/a; \\ \tilde{z}_8^{ijk} &= z_5^{ijk} (z_4^{ijk} z_7^{ijk} + z_4^{ijk} \tilde{z}_7^{ijk}); & \tilde{z}_9^{ij} &= \sum_{\substack{k=1 \\ k \neq i, j}}^I \tilde{z}_8^{ijk}; & \tilde{z}_{10}^{ij} &= \tilde{z}_9^{ij} u_6; \\ \tilde{z}_{11}^{ij} &= \tilde{z}_{10}^{ij} u_5 (z_{10}^{ij})^{u_5-1}; & \tilde{z}_{12}^{ij} &= -\frac{1}{2} u_5 \tilde{z}_{11}^{ij} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-1}; & \tilde{z}_{13}^{ij} &= z_{13}^{ij}/a; \\ \tilde{z}_{14}^{ij} &= -\frac{u_3 \sqrt{2u_4} z_{13}^{ij} z_{14}^{ij}}{a}; & \tilde{z}_{15}^{ij} &= -\frac{u_3 \sqrt{2/u_4} z_{13}^{ij} z_{15}^{ij}}{a}; & \tilde{z}_{16}^{ij} &= \frac{\partial f_c \left( (1 + \frac{\rho}{a}) r_{ij} \right)}{\partial \rho} \Big|_{\rho=0}; \\ \tilde{z}_{17}^{ij} &= \tilde{z}_{16}^{ij} z_{14}^{ij} - \tilde{z}_{16}^{ij} z_{12}^{ij} z_{15}^{ij} + \tilde{z}_{14}^{ij} z_{16}^{ij} - \tilde{z}_{12}^{ij} z_{16}^{ij} z_{15}^{ij} - \tilde{z}_{15}^{ij} z_{16}^{ij} z_{12}^{ij}; \\ \tilde{z}_1^{ijk} &= \tilde{z}_2^{ijk} = \tilde{z}_3^{ijk} = \tilde{z}_5^{ijk} = 0; & \tilde{z}_4^{ijk} &= \frac{\partial^2 f_c \left( (1 + \frac{\rho}{a}) r_{ik} \right)}{\partial \rho^2} \Big|_{\rho=0}; \\ \tilde{z}_6^{ijk} &= 6z_6^{ijk}/a^2; & \tilde{z}_7^{ijk} &= \frac{3}{a^2} z_6^{ijk} z_7^{ijk} (u_7)^3 (3z_6^{ijk} (u_7)^3 + 2); \\ \tilde{\tilde{z}}_8^{ijk} &= z_5^{ijk} (\tilde{z}_4^{ijk} z_7^{ijk} + 2\tilde{z}_4^{ijk} \tilde{z}_7^{ijk} + z_4^{ijk} \tilde{\tilde{z}}_7^{ijk}); & \tilde{\tilde{z}}_9^{ij} &= \sum_{\substack{k=1 \\ k \neq i, j}}^I \tilde{\tilde{z}}_8^{ijk}; \end{aligned}$$

$$\begin{aligned}
\tilde{z}_{10}^{ij} &= \tilde{z}_9^{ij} u_6; & \tilde{z}_{11}^{ij} &= u_5(u_5 - 1)(z_{10}^{ij})^2 (z_{10}^{ij})^{u_5-2} + u_5(\tilde{z}_{10}^{ij})(z_{10}^{ij})^{u_5-1}; \\
\tilde{z}_{12}^{ij} &= \frac{1 + 2u_5}{4u_5} (\tilde{z}_{11}^{ij})^2 (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-2} - \frac{1}{2u_5} \tilde{z}_{11}^{ij} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-1}; & \tilde{z}_{13}^{ij} &= 0; \\
\tilde{z}_{14}^{ij} &= \frac{2(u_3)^2 u_4 (z_{13}^{ij})^2 z_{14}^{ij}}{a^2}; & \tilde{z}_{15}^{ij} &= \frac{2(u_3)^2 (z_{13}^{ij})^2 z_{15}^{ij}}{a^2 u_4}; & \tilde{z}_{16}^{ij} &= \left. \frac{\partial^2 f_c \left( \left(1 + \frac{\rho}{a}\right) r_{ij} \right)}{\partial \rho^2} \right|_{\rho=0}; \\
\tilde{z}_{17}^{ij} &= \tilde{z}_{16}^{ij} z_{14}^{ij} + 2\tilde{z}_{16}^{ij} \tilde{z}_{14}^{ij} - \tilde{z}_{16}^{ij} z_{12}^{ij} z_{15}^{ij} - 2\tilde{z}_{16}^{ij} \tilde{z}_{12}^{ij} z_{15}^{ij} - 2\tilde{z}_{16}^{ij} z_{12}^{ij} \tilde{z}_{15}^{ij} + \\
&\quad + \tilde{z}_{14}^{ij} z_{16}^{ij} - \tilde{z}_{12}^{ij} z_{16}^{ij} z_{15}^{ij} - 2\tilde{z}_{15}^{ij} z_{16}^{ij} \tilde{z}_{12}^{ij} - \tilde{z}_{15}^{ij} z_{16}^{ij} z_{12}^{ij}.
\end{aligned}$$

To compute the second derivative of a function  $f_c(r)$  there is a need for smoothing this function. It is proposed to replace the function  $f_c(r)$  as follows:

$$f_c(r) = \begin{cases} 0, & r \geq R + R_{cut}, \\ 1, & r \leq R - R_{cut}, \\ C \cdot (f_*)^{\varphi(r)}, & R \leq r < R + R_{cut}, \\ C \cdot (2f_* - (f_*))^{\psi(r)}, & R - R_{cut} < r \leq R, \end{cases}$$

where  $C = \frac{1}{2f_*}$ ,  $f_* = \exp(-\frac{3}{2})$ ,  $\varphi(r) = \frac{R_{cut}^2}{(r - R - R_{cut})^2}$ ,  $\psi(r) = \frac{R_{cut}^2}{(r - R + R_{cut})^2}$ .

Thus,  $B(E)$  is calculated by the formula  $B(E) = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \tilde{z}_{17}^{ij}(\bar{u})$ , where the variables  $z_1, z_2, \dots, z_{17}$ ,

$\tilde{z}_1, \tilde{z}_2, \dots, \tilde{z}_{17}$ ,  $\tilde{\tilde{z}}_1, \tilde{\tilde{z}}_2, \dots, \tilde{\tilde{z}}_{17}$  are determined by mentioned above multistep algorithm.

According to the formulas of FAD-technique, the conjugate variables corresponding to the phase variables  $z_1, z_2, \dots, z_{17}$ ,  $\tilde{z}_1, \tilde{z}_2, \dots, \tilde{z}_{17}$ ,  $\tilde{\tilde{z}}_1, \tilde{\tilde{z}}_2, \dots, \tilde{\tilde{z}}_{17}$  are determined by the formulas:

$$\begin{aligned}
p_5^{ijk} &= (\tilde{z}_4^{ijk} z_7^{ijk} + 2\tilde{z}_4^{ijk} \tilde{z}_7^{ijk} + z_4^{ijk} \tilde{\tilde{z}}_7^{ijk}) p_8^{ijk} + (z_4^{ijk} z_7^{ijk} + z_4^{ijk} \tilde{z}_7^{ijk}) p_8^{ijk} + z_4^{ijk} z_7^{ijk} p_8^{ijk}; \\
p_7^{ijk} &= \tilde{z}_4^{ijk} z_5^{ijk} \tilde{\tilde{z}}_8^{ijk} + \tilde{z}_4^{ijk} z_5^{ijk} p_8^{ijk} + z_4^{ijk} z_5^{ijk} p_8^{ijk} + \frac{3}{a^2} z_6^{ijk} (u_7)^3 (3z_6^{ijk} (u_7)^3 + 2) \tilde{z}_7^{ijk} + \frac{3}{a} z_6^{ijk} (u_7)^3 \tilde{z}_7^{ijk}; \\
p_8^{ijk} &= p_9^{ij}; & p_9^{ij} &= u_6 p_{10}^{ij}; & p_{10}^{ij} &= \left[ u_5(u_5 - 1)(u_5 - 2)(z_{10}^{ij})^{u_5-3} (\tilde{z}_{10}^{ij})^2 + u_5(u_5 - 1)(z_{10}^{ij})^{u_5-2} \tilde{z}_{10}^{ij} \right] \tilde{z}_{11}^{ij} + \\
&\quad + u_5(u_5 - 1)(z_{10}^{ij})^{u_5-2} \tilde{z}_{10}^{ij} \tilde{z}_{11}^{ij} + u_5(z_{10}^{ij})^{u_5-1} p_{11}^{ij}; \\
p_{11}^{ij} &= -\frac{(1 + 4u_5)(1 + 2u_5)}{8(u_5)^3} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-3} (z_{11}^{ij})^2 \tilde{z}_{12}^{ij} + \frac{(1 + 2u_5)}{4(u_5)^2} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-2} \times \\
&\quad \times \tilde{z}_{11}^{ij} \tilde{z}_{12}^{ij} + \frac{(1 + 2u_5)}{4(u_5)^2} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-2} \tilde{z}_{11}^{ij} \tilde{z}_{12}^{ij} - \frac{1}{2u_5} (1 + z_{11}^{ij})^{-\frac{1}{2u_5}-1} p_{12}^{ij}; \\
p_{12}^{ij} &= (-\tilde{z}_{16}^{ij} z_{15}^{ij} - 2\tilde{z}_{15}^{ij} \tilde{z}_{16}^{ij} - z_{16}^{ij} \tilde{\tilde{z}}_{15}^{ij}) \tilde{z}_{17}^{ij}; & p_{14}^{ij} &= \tilde{z}_{16}^{ij} \tilde{z}_{17}^{ij} + \frac{2(u_3)^2 u_4 (z_{13}^{ij})^2 \tilde{z}_{14}^{ij}}{a^2} p_{14}^{ij} - u_3 \sqrt{2u_4} \frac{z_{13}^{ij}}{a} \tilde{z}_{14}^{ij}; \\
p_{15}^{ij} &= (-\tilde{z}_{16}^{ij} z_{12}^{ij} - 2\tilde{z}_{12}^{ij} \tilde{z}_{16}^{ij} - z_{16}^{ij} \tilde{\tilde{z}}_{12}^{ij}) \tilde{z}_{17}^{ij} + \frac{2(u_3)^2 (z_{13}^{ij})^2 \tilde{z}_{14}^{ij}}{a^2 u_4} p_{15}^{ij} - \sqrt{2/u_4} \frac{u_3 z_{13}^{ij}}{a} \tilde{z}_{15}^{ij}; \\
p_{16}^{ij} &= (\tilde{z}_{14}^{ij} - \tilde{z}_{12}^{ij} z_{15}^{ij} - 2\tilde{z}_{12}^{ij} \tilde{z}_{15}^{ij} - z_{12}^{ij} \tilde{\tilde{z}}_{15}^{ij}) \tilde{z}_{17}^{ij}; & p_{17}^{ij} &= 0; \\
\tilde{p}_7^{ijk} &= 2\tilde{z}_4^{ijk} z_5^{ijk} \tilde{\tilde{z}}_8^{ijk} + z_4^{ijk} z_5^{ijk} \tilde{p}_8^{ijk}; & \tilde{p}_8^{ijk} &= \tilde{p}_9^{ij}; & \tilde{p}_9^{ij} &= u_6 \tilde{p}_{10}^{ij}; \\
\tilde{p}_{10}^{ij} &= 2u_5(u_5 - 1)(z_{10}^{ij})^{u_5-2} \tilde{z}_{11}^{ij} p_{11}^{ij} + u_5(z_{10}^{ij})^{u_5-1} \tilde{z}_{10}^{ij} \tilde{p}_{11}^{ij};
\end{aligned}$$

$$\tilde{p}_{11}^{ij} = \frac{(1+2u_5)}{2(u_5)^2} (1+z_{11}^{ij})^{-1/(2u_5)-2} \tilde{z}_{11}^{ij} \tilde{z}_{12}^{ij} - \frac{(1+z_{11}^{ij})^{-1/(2u_5)-1}}{2u_5} \tilde{p}_{12}^{ij}; \quad \tilde{p}_{12}^{ij} = -2(\tilde{z}_{16}^{ij} z_{15}^{ij} + \tilde{z}_{15}^{ij} z_{16}^{ij}) \tilde{p}_{17}^{ij};$$

$$\tilde{p}_{14}^{ij} = 2z_{16}^{ij} \tilde{z}_{17}^{ij}; \quad \tilde{p}_{15}^{ij} = -2(\tilde{z}_{16}^{ij} z_{12}^{ij} + \tilde{z}_{12}^{ij} z_{16}^{ij}) \tilde{p}_{17}^{ij}; \quad \tilde{p}_{16}^{ij} = 2(\tilde{z}_{14}^{ij} - \tilde{z}_{12}^{ij} z_{15}^{ij} - \tilde{z}_{15}^{ij} z_{12}^{ij}) \tilde{p}_{17}^{ij}; \quad \tilde{p}_{17}^{ij} = 0;$$

$$\tilde{p}_7^{ijk} = z_4^{ijk} z_5^{ijk} \tilde{p}_8^{ijk}; \quad \tilde{p}_8^{ijk} = \tilde{p}_9^{ij}; \quad \tilde{p}_9^{ij} = u_6 \tilde{p}_{10}^{ij}; \quad \tilde{p}_{10}^{ij} = u_5 (z_{10}^{ij})^{u_5-1} \tilde{p}_{11}^{ij};$$

$$\tilde{p}_{11}^{ij} = -\frac{(1+z_{11}^{ij})^{-1/(2u_5)-1}}{2u_5} \tilde{p}_{12}^{ij}; \quad \tilde{p}_{12}^{ij} = -z_{15}^{ij} z_{16}^{ij} \tilde{p}_{17}^{ij};$$

$$\tilde{p}_{14}^{ij} = z_{16}^{ij} \tilde{z}_{17}^{ij}; \quad \tilde{p}_{15}^{ij} = -z_{12}^{ij} z_{16}^{ij} \tilde{z}_{17}^{ij}; \quad \tilde{p}_{16}^{ij} = (z_{14}^{ij} - z_{12}^{ij} z_{15}^{ij}) \tilde{z}_{17}^{ij}; \quad \tilde{p}_{17}^{ij} = 1;$$

The adjoint variables are calculate in the following order:  $\tilde{p}_{17}^{ij}, \tilde{p}_{16}^{ij}, \dots, \tilde{p}_7, \tilde{p}_{17}^{ij}, \dots, \tilde{p}_7, \tilde{p}_{17}^{ij}, \dots, \tilde{p}_5^{ij}$ . Those adjoint variables, which formulas for calculation aren't provided above, aren't used for calculation of the components of the gradient.

According to the formulas of FAD-technique, the partial derivatives of a complex function  $\Omega(\bar{u}) = B(E(\bar{u})) = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \tilde{z}_{17}^{ij}(\bar{u})$  with respect to independent variables  $u_m$ , ( $m = \overline{1, 10}$ ) are determined by the relations:

$$\frac{\partial \Omega}{\partial u_1} = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \frac{z_{14}^{ij}}{u_1} p_{14}^{ij} + \frac{z_{15}^{ij}}{u_1} p_{15}^{ij} \right);$$

$$\frac{\partial \Omega}{\partial u_2} = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( z_{14}^{ij} u_3 \sqrt{2u_4} p_{14}^{ij} + z_{15}^{ij} u_3 \sqrt{2/u_4} p_{15}^{ij} \right);$$

$$\begin{aligned} \frac{\partial \Omega}{\partial u_3} = & \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( z_{14}^{ij} \left( -\sqrt{2u_4} (z_{13}^{ij} - u_2) \right) p_{14}^{ij} + z_{15}^{ij} \left( -\sqrt{2/u_4} (z_{13}^{ij} - u_2) \right) p_{15}^{ij} \right) + \\ & + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \frac{4u_3 u_4 (z_{13}^{ij})^2 z_{14}^{ij} \tilde{z}_{17}^{ij}}{a^2} p_{14}^{ij} - \frac{\sqrt{2u_4} z_{13}^{ij} z_{14}^{ij} \tilde{z}_{17}^{ij}}{a} p_{14}^{ij} \right) + \\ & + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \frac{4u_3 (z_{13}^{ij})^2 z_{15}^{ij} \tilde{z}_{17}^{ij}}{a^2 u_4} p_{15}^{ij} - \frac{z_{13}^{ij} z_{15}^{ij}}{a} \sqrt{2/u_4} p_{15}^{ij} \right); \end{aligned}$$

$$\begin{aligned} \frac{\partial \Omega}{\partial u_4} = & \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( -\frac{z_{14}^{ij}}{u_4 - 1} - 0.5 u_3 \sqrt{2/u_4} (z_{13}^{ij} - u_2) z_{14}^{ij} \right) p_{14}^{ij} \right) + \\ & + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( -\frac{z_{15}^{ij}}{u_4 (u_4 - 1)} + 0.5 (u_3/u_4) \sqrt{2/u_4} (z_{13}^{ij} - u_2) z_{15}^{ij} \right) p_{15}^{ij} \right) + \end{aligned}$$

$$\begin{aligned}
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( -\frac{u_3}{2a} \sqrt{2/u_4} z_{13}^{ij} z_{14}^{ij} \right) \tilde{p}_{14}^{ij} + \left( \frac{2(u_3)^2 (z_{13}^{ij})^2}{a^2} z_{14}^{ij} \right) \tilde{p}_{14}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( \frac{u_3}{2au_4} \sqrt{2/u_4} z_{13}^{ij} z_{15}^{ij} \right) \tilde{p}_{14}^{ij} - \left( \frac{2(u_3)^2 (z_{13}^{ij})^2}{a^2 (u_4)^2} z_{15}^{ij} \right) \tilde{p}_{15}^{ij} \right); \\
\frac{\partial \Omega}{\partial u_5} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( (z_{10}^{ij})^{u_5} \ln(z_{10}^{ij}) p_{11}^{ij} + \frac{(1+z_{11}^{ij})^{-1/(2u_5)}}{2(u_5)^2} \ln(1+z_{11}^{ij}) p_{12}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( (z_{10}^{ij})^{u_5-1} \tilde{z}_{10}^{ij} + u_5 (z_{10}^{ij})^{u_5-1} \tilde{z}_{10}^{ij} \ln(z_{10}^{ij}) \right) \tilde{p}_{11}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( (2u_5-1)(z_{10}^{ij})^{u_5-2} + (u_5^2-u_5)(z_{10}^{ij})^{u_5-2} \ln(z_{10}^{ij}) \right) (z_{10}^{ij})^2 \tilde{p}_{12}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( (z_{10}^{ij})^{u_5-1} + u_5 (z_{10}^{ij})^{u_5-1} \ln(z_{10}^{ij}) \right) \tilde{z}_{10}^{ij} \tilde{p}_{11}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( \frac{(1+z_{11}^{ij})^{-1/(2u_5)-1}}{2(u_5)^2} - \frac{\ln(1+z_{11}^{ij})}{4(u_5)^3} (1+z_{11}^{ij})^{-1/(2u_5)-1} \right) \tilde{z}_{11}^{ij} \tilde{p}_{12}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( -\frac{(1+u_5)}{2(u_5)^3} (1+z_{11}^{ij})^{-1/(2u_5)-2} \right) \tilde{z}_{11}^{ij} \tilde{p}_{12}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( \frac{(1+2u_5) \ln(1+z_{11}^{ij})}{8(u_5)^4} (1+z_{11}^{ij})^{-1/(2u_5)-2} \right) \tilde{z}_{11}^{ij} \tilde{p}_{12}^{ij} \right) + \\
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( \left( \frac{(1+z_{11}^{ij})^{-1/(2u_5)-1}}{2(u_5)^2} - \frac{\ln(1+z_{11}^{ij})}{4(u_5)^3} (1+z_{11}^{ij})^{-1/(2u_5)-1} \right) \tilde{z}_{11}^{ij} \tilde{p}_{12}^{ij} \right); \\
\frac{\partial \Omega}{\partial u_6} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \left( z_9^{ij} p_{10}^{ij} + \tilde{z}_9^{ij} \tilde{p}_{10}^{ij} + \tilde{z}_9^{ij} \tilde{p}_{10}^{ij} \right); \\
\frac{\partial \Omega}{\partial u_7} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \sum_{\substack{k=1 \\ k \neq i, j}}^I \left( 3z_6^{ijk} z_7^{ijk} (u_7)^2 p_7^{ijk} + \frac{9}{a} z_6^{ijk} z_7^{ijk} (u_7)^2 \tilde{p}_7^{ijk} \right) +
\end{aligned}$$

$$\begin{aligned}
& + \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \sum_{\substack{k=1 \\ k \neq i,j}}^I \left( \left( \frac{3z_6^{ijk} z_7^{ijk}}{a^2} \left( 18z_6^{ijk} (u_7)^5 + 6(u_7)^2 \right) \right) p_7^{ijk} \right); \\
\frac{\partial \Omega}{\partial u_8} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \sum_{\substack{k=1 \\ k \neq i,j}}^I \left( \left( \frac{2u_8}{(u_9)^2} - \frac{2u_8}{(u_9)^2 + (u_{10} - z_3^{ijk})^2} \right) p_5^{ijk} \right); \\
\frac{\partial \Omega}{\partial u_9} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \sum_{\substack{k=1 \\ k \neq i,j}}^I \left( \left( \frac{-2(u_8)^2}{(u_9)^3} + \frac{2(u_8)^2 u_9}{((u_9)^2 + (u_{10} - z_3^{ijk})^2)} \right) p_5^{ijk} \right); \\
\frac{\partial \Omega}{\partial u_{10}} & = \sum_{i=1}^I \sum_{\substack{j=1 \\ j \neq i}}^I \sum_{\substack{k=1 \\ k \neq i,j}}^I \left( \left( \frac{2(u_8)^2 (u_{10} - z_3^{ijk})}{((u_9)^2 + (u_{10} - z_3^{ijk})^2)^2} \right) p_5^{ijk} \right).
\end{aligned}$$

The received formulas for calculation of the gradient of function  $B(E(u))$  outwardly are represented quite difficult and bulky. Therefore, there is a natural question: whether to use simpler approaches, for example, finite difference method, to calculate the gradient functions  $B(E(u))$ .

In [Albu2016] the comparison of function gradients calculated by the finite differences and by using Fast Automatic Differentiation formulas was presented. The results of comparison are the following:

- 1) when computing the gradient of complicated function using finite differences, one must conduct researches related to the choice of suitable increments of each parameter;
- 2) for different parameters, the researches must be carried out independently;
- 3) for the same parameter, the researches must be carried out if its value changed;
- 4) to calculate the gradient of complicated function using finite differences one must  $(m + 1)$  times calculate the value of function itself.

In contrary to it, the FAD-technique enables us to calculate gradients of any complicated function with the machine accuracy for arbitrary parameters. The machine time that is needed to calculate the gradient does not exceed three times of calculation of the function itself.

### 3.0.1 Acknowledgements

The research was supported by Russian Science Foundation (project No. 21-71- 30005).

## References

- [Tersoff1988] J. Tersoff. Empirical interatomic potential for silicon with improved elastic properties. *Phys. Rev. B.*, 38:9902–9905, 1988.
- [Abgaryan2014] K.K. Abgaryan and M.A. Posypkin. Optimization methods as applied to parametric identification of interatomic potentials. *Comput. Math. Math. Phys.*, 54(12):1929–1935, 2014.
- [Albu2016] A.F. Albu. Application of the Fast Automatic Differentiation to the Computation of the Gradient of the Tersoff Potential. *Informacionnye tekhnologii i vychislitel'nye sistemy*, 1:43–49, 2016. [in Russian]
- [Evtushenko1998] Y.G. Evtushenko. Computation of Exact Gradients in Distributed Dynamic Systems. *Optimizat. Methods and Software*, 9:45–75, 1998.