Application of the Fast Automatic Differentiation to the Computation of the Gradient of the Energy of Atoms’ System with Respect to Tersoff Parameters*

Alla Albu and Vladimir Zubov

Abstract— Gradient optimization methods are often used to tackle problems of computer modeling of materials’ crystal structures. This raises the need to determine the exact value of the gradient of the Tersoff potential using specific parameters of this potential for the modeled substance. Based on the technique of the Fast Automatic Differentiation the formulas that allow the calculation of the exact value of the above-mentioned gradient were derived.

Keywords—Tersoff potential; gradient; Fast Automatic Differentiation

I. INTRODUCTION

For modeling solid atomic structures the Tersoff Potential is often used ([1]). The Tersoff Potential depends on ten parameters specific to the modeling material. These parameters are usually unknown and they should be identified as the solution of the inverse problem. In [2] one possible optimization problem was considered. It consists of minimizing the following cost function

\[ f(\xi) = \sum_{i=1}^{n} w_i (y_i(\xi) - \bar{y}_i)^2, \]

where \( w_i \) is the weight factor; \( \bar{y}_i \) 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 the Tersoff Potential with \( \xi \) parameters (\( \xi \in \mathbb{R}^m \) are vector parameters to be identified). The solution of this problem is looked for on the set \( X \subseteq \mathbb{R}^m \), which is a parallelepiped. A required set of parameters has to provide the minimum deviation of the calculated characteristics of the material from the known experimental values. For numerical solution of this problem the gradient minimization methods are often used. One of the terms in formula (1) is the total energy of the system of atoms. There exists the need to calculate efficiently the exact gradient of the total energy with respect to parameters of the Tersoff Potential.

This gradient is often calculated (see, for example, [2]) using the finite difference method. Studies have shown that gradient difference method does not allow to calculate the gradient of the energy of atoms’ system with respect to Tersoff parameters with acceptable accuracy and requires \((m + 1)\) times to calculate the value of the function.

In this paper, we build a multistep algorithm to calculate the value of total atoms’ system energy in the case where this energy is determined by Tersoff Potential and a multistep algorithm to calculate the conjugate variables, by which the value of the above-mentioned gradient is determined with machine precision on the basis of the Fast Automatic Differentiation methodology.

II. ALGORITHM FOR CALCULATION THE TOTAL ENERGY OF ATOMS’ SYSTEM AND THE ADJOINT VARIABLES

Let \( \vec{r}_k = (x_{k1}, x_{k2}, x_{k3}) \) be the coordinates of some lattice atom. As to total energy \( E(\vec{r}_1, \vec{r}_2, ..., \vec{r}_l) \) it is calculated with the help of expression

\[ E(\vec{r}_1, \vec{r}_2, ..., \vec{r}_l) = \sum_{i=1}^{l} \sum_{j=1}^{l} V_{ij}, \]

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:

\[ V_{ij} = f_c(r_{ij}) (V_R(r_{ij}) - b_{ij} V_A(r_{ij})), \]

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

\[ V_R(r_{ij}) = \frac{D_c}{S - 1} \exp \left( -\beta \sqrt{S} (r_{ij} - r_e) \right), \]

\[ V_A(r_{ij}) = \frac{SD_c}{S - 1} \exp \left( -\beta \sqrt{S} (r_{ij} - r_e) \right), \]

\[ b_{ij} = \left( 1 + (y_{ij})^2 \right)^{\frac{1}{2n}}, \]

\[ y_{ij} = \sum_{k=1}^{l} f_c(r_{ik}) g_{ijk} \alpha_{ijk}, \]

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\[ \omega_{ijk} = \exp(\lambda r_{ijk}), \quad \tau_{ijk} = (r_{ij} - r_{ik})^2; \]
\[ g_{ijk} = g(\theta_{ijk}) = 1 + \left( \frac{c_1}{d} \right)^2 - \frac{c_2}{d^2} + (h - \cos \theta_{ijk})^2. \]

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 the substance. Tersoff Potential depends on these ten parameters \( (m=10) \), specific to modeled substances: \( D_e, \quad r_e, \quad \beta, \quad S, \quad \eta, \quad \gamma, \quad \lambda, \quad c, \quad d, \quad h \).

For using the Fast Automatic Differentiation to calculate the gradient of cost function, we construct the multistep algorithm to determine the total energy \( E \) of atoms' system (interaction potential is Tersoff Potential). The distance between \( i \)-atom and \( j \)-atom is determined by the formula
\[ r_{ij} = \sqrt{(x_{i1} - x_{j1})^2 + (x_{i2} - x_{j2})^2 + (x_{i3} - x_{j3})^2}, \quad (i \neq j), \]
where \( x_{i1}, x_{i2}, x_{i3} \) 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
\[ q_{ijk} = \cos(\theta_{ijk}) = \frac{r_{ij}^2 + r_{ik}^2 - r_{jk}^2}{2r_{ij}r_{ik}}. \]

For compactness further in the study we introduce vectors \( \bar{u} \) and \( \bar{z} \) having the following coordinates:
\[ \bar{u}^T = [u_1, u_2, \ldots, u_{10}]^T, \quad \bar{z}^T = [z_1, z_2, \ldots, z_{17}]^T, \]
where \( u_1 = D_e, \quad u_2 = r_e, \quad u_3 = \beta, \quad u_4 = S, \quad u_5 = \eta, \quad u_6 = \gamma, \quad u_7 = \lambda, \quad u_8 = c, \quad u_9 = d, \quad u_{10} = h; \]
\[ z_1 = \left\{ z_{ijk}^{1} = \sqrt{(x_{i1} - x_{j1})^2 + (x_{i2} - x_{j2})^2 + (x_{i3} - x_{j3})^2} \right\}; \]
\[ z_2 = \left\{ z_{ijk}^{2} = \sqrt{(x_{i1} - x_{j1})^2 + (x_{i2} - x_{j2})^2 + (x_{i3} - x_{j3})^2} \right\}; \]
\[ z_3 = \left\{ z_{ijk}^{3} = q_{ijk} = \frac{(z_{ijk}^{1})^2 + (z_{ijk}^{2})^2 - (z_{ijk}^{2})^2}{2z_{ijk}^{1}z_{ijk}^{2}} \right\}; \]
\[ z_4 = \left\{ z_{ijk}^{4} = f_z(z_{1i}) \right\}; \]
\[ z_5 = \left\{ z_{ijk}^{5} = g_{ijk} = 1 + \frac{(u_1)^2}{u_9} - \frac{(u_2)^2}{u_8} + \frac{(u_3)^2}{u_9} + \frac{(u_5)^2}{u_2} \right\}; \]
\[ z_6 = \left\{ z_{ijk}^{6} = \tau_{ijk} = (z_{1i} - z_{ijk}^{5})^3 \right\}; \]
\[ z_7 = \left\{ z_{ijk}^{7} = \omega_{ijk} = \exp((u_7)^3z_{ijk}^{6}) \right\}; \]
\[ z_8 = \left\{ z_{ijk}^{8} = f_c(r_{ik})g_{ijk} = z_{ijk}^{4} - z_{ijk}^{5}z_{ijk}^{6} \right\}; \]
\[ z_9 = \left\{ z_{ijk}^{9} = z_{ijk}^{7} = \sum_{k=1}^{l}z_{ijk}^{8} \right\}; \]
\[ z_{10} = \left\{ z_{ijk}^{10} = u_6z_{ijk}^{9} \right\}; \]
\[ z_{11} = \left\{ z_{ijk}^{11} = (\gamma z_{ijk}^{7})^{\eta} = (z_{ijk}^{9})^{\eta} \right\}; \]
\[ z_{12} = \left\{ z_{ijk}^{12} = h_{ij} = (1 + z_{ijk}^{11})^{-\frac{1}{2z_{ijk}^{5}}} \right\}; \]
\[ z_{13} = \left\{ z_{ijk}^{13} = \left( x_{i1} - x_{j1} \right)^2 + \left( x_{i2} - x_{j2} \right)^2 + \left( x_{i3} - x_{j3} \right)^2 \right\}; \]
\[ z_{14} = \left\{ z_{ijk}^{14} = u_{14} - u_{4}^{-1} \exp(-u_{3}\sqrt{2u_{4}(z_{ijk}^{13} - u_{2})}) \right\}; \]
\[ z_{15} = \left\{ z_{ijk}^{15} = u_{14} - u_{4}^{-1} \exp(-u_{3}\sqrt{2u_{4}(z_{ijk}^{13} - u_{2})}) \right\}; \]
\[ z_{16} = \left\{ z_{ijk}^{16} = f_z(z_{1i}) \right\}; \]
\[ z_{17} = \left\{ z_{ijk}^{17} = V_{ij} = z_{ijk}^{9} \exp(z_{ij}^{9} - z_{ij}^{12}z_{ij}^{15}) \right\}. \]

(i = \{1,T \}, \quad j = \{T \}, \quad i \neq j, \quad k = \{T \}, \quad k \neq i, j). \]

The total energy of the atoms' system with the help of new variables may be rewritten as follows:
\[ E(z(u)) = \sum_{i=1}^{I} \sum_{j=1}^{I} z_{ij}^{17}, \]

Variables \( z_1, z_2, \ldots, z_{17} \) (the phase variables) are determined by the specified above multistep algorithm \( z_i = F(l, Z_i, U_i) \), \( (I = \{1,T \}) \), where \( Z_i \) is the set of elements \( z_{ni} \) in the right part of the equation \( z_i = F(l, Z_i, U_i) \), and \( U_i \) is the set of elements \( u_{ni} \) that appear in the right side of this equation. Note that each component \( Z_i \) depends on a number of other components \( z_{ij} \) or \( z_{ij}^{17} \).

Below are represented the general formulas of the Fast Automatic Differentiation (see [3]). They will be used later to calculate the gradient of the total energy \( E \) of atoms' system with respect to parameters of the Tersoff Potential.

Let vectors \( z \in \mathbb{R}^n \) and \( u \in \mathbb{R}^m \) satisfy the following system of nonlinear scalar equations (multistep process):
\[ z_i = F(i, Z_i, U_i), \quad 1 \leq i \leq n, \quad (2) \]
where $Z_i$ is the set of vectors $z_j$, that appear at the right part of equality (2), and $U_j$ is the set of vectors $u_j$, that appear at the right part of the same equality (2). Usually vectors $z \in \mathbb{R}^n$ and vectors $u \in \mathbb{R}^m$ are called dependent (phase) and independent (control) variables respectively. Let differentiable function $W(z,u)$ define mapping $W: \mathbb{R}^n \times \mathbb{R}^m \rightarrow \mathbb{R}^1$. Then the composite function $\Omega(u) = W(z(u),u)$ is differentiable, and its gradient with respect to the independent variables $U_i$ is given by the formula

$$
\frac{d\Omega}{du_i} = W_{zi}(z,u) + \sum_{q \in \mathcal{Q}_i} F^T_{zi}(q,Z_q,U_q)p_q.
$$

(3)

The multipliers $p_i \in \mathbb{R}^n$ are the conjugate variables that are defined by the following system of linear algebraic equations:

$$
p_i = W_{zi}(z,u) + \sum_{q \in \mathcal{Q}_i} F^T_{zi}(q,Z_q,U_q)p_q,
$$

(4)

where $\mathcal{Q}_i$ and $\mathcal{K}_i$ are the index sets:

$$
\mathcal{Q}_i = \{ j : 1 \leq j \leq n, \quad z_j \in Z_i \}, \quad \mathcal{K}_i = \{ j : 1 \leq j \leq n, \quad u_j \in U_i \}.
$$

The partial derivatives of function $\Omega(u) = E(z(u)) = \sum_{i=1}^{I} \sum_{j=1}^{J} z^i_j(u)$ with respect to independent variables $u_m$, $(m = \cap \mathcal{Q}_i)$ (components of gradient), according to equations (3), (4) are determined by the relations:

$$
\frac{\partial \Omega}{\partial u_m} = \sum_{q \in \mathcal{K}_m} F^T_{zi}(q,Z_q,U_q)p_q, \quad \mathcal{K}_m = \{ j : \quad u_m \in U_i \}, \quad m = \cap \mathcal{Q}_i,
$$

where the conjugate variables $p_i$ are defined by the following system of linear algebraic equations:

$$
p_i = E_{zi}(z) + \sum_{q \in \mathcal{Q}_i} F^T_{zi}(q,Z_q,U_q)p_q,
$$

(5)

$$
\mathcal{Q}_i = \{ j : \quad z_j \in Z_i \}, \quad (i = \cap \mathcal{Q}_i).
$$

Note that $E_{u_m}(z(u)) = 0$ because function $E(z(u))$ clearly does not depend on the vector components $\tilde{u}$.

In accordance to (5), for all $i = \cap \mathcal{Q}_i$, $j = \cap \mathcal{Q}_i$, $j \neq i$, $k = \cap \mathcal{Q}_i$, $k \neq i,j$ conjugate variables corresponding to the phase variables $z_1 , z_2, ..., z_{17}$ are defined by the equations:

$$
p_{i j = k}^j = \sum_{l=1}^{I} \sum_{i = 1}^{J} \left( \frac{z^j_i}{u^i} \cdot \frac{p_{i j = k}^j}{u^i} + \frac{z_{15}^j}{u^j} \cdot \frac{p_{i j = k}^j}{u^j} \right),
$$

$$
\frac{\partial \Omega}{\partial u_m} = \sum_{i=1}^{I} \sum_{j=1}^{J} \left( \frac{z^j_i}{u^i} \cdot \frac{p_{i j = k}^j}{u^i} + \frac{z_{15}^j}{u^j} \cdot \frac{p_{i j = k}^j}{u^j} \right).
$$

III. DETERMINING THE COMPONENTS OF THE GRADIENT

Partial derivatives of the function $\Omega(u) = E(z(u))$ with respect to independent variables $u_m$, $(m = \cap \mathcal{Q}_i)$ are determined by these relations:
The received formulas for calculation of the gradient of the total energy of atoms’ system with respect to Tersoff parameters are quite complex and bulky. Therefore, there is a natural question whether it would be better to use simpler approaches to calculate the gradient of the function \( \Omega(u) = E(z(u)) \), for example, finite difference method.

The problem of selection of Tersoff parameters for one-component silicon crystal was considered in [2]. To determine the initial approximations in this work an admissible set \( X = [\xi, \bar{\xi}] = \{u \in R^{10} : \xi_i \leq u_i \leq \bar{\xi}_i \} \) was chosen in such a way that it clearly contains all the possible values of parameters, namely:

\[
\xi = (0.5; 0.5; 0.5; 0.5; 0.1; 5 \cdot 10^{-8}; 0.5; 10000; 1; -2), \\
\bar{\xi} = (10; 5; 5; 5; 2; 3 \cdot 10^{-6}; 3; 200000; 30; -0.1).
\]

In [4] the comparison of function gradients, calculated by the finite differences and by using Fast Automatic Differentiation formulas (6), was presented. In TABLES I and II are given the values of the gradient for two specific parameters of the simulated substance: \( u_3 = \beta, \ u_6 = \gamma \).

Input parameters \( u_3 = 1.0 \) and \( u_6 = 10^{-7} \) were selected from the range of acceptable values. The value \( \Delta \) in TABLES I and II indicates the increment of parameters \( u_i \) that were used during calculation of the gradient using the method of finite differences.

As can be seen from TABLE I, during the calculation of the third component of the gradient value \( \Delta \approx 10^{-7} \) is the most suitable choice. It is about \( 10^{-5}\% \) from initial approximation \( u_3 = 1.0 \). TABLE II shows that during the calculation of the sixth component of the gradient \( \Delta \approx 10^{-11} \) is the most appropriate choice. This is about \( 10^{-2}\% \) from the initial approximation \( u_6 = 10^{-7} \). The results of the comparison are the following:

1) when computing the gradient of a 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 any parameter, if its value changed, the research must be carried out again;
4) to calculate the gradient of a complicated function using finite differences one must \( (m = 10) \) times calculate the value of the function itself.

\[ \text{TABLE I} \]
In contrary to it, the Fast Automatic Differentiation enables us to calculate gradients of any complicated function with the machine accuracy for arbitrary number of parameters. The machine time that is needed to calculate the gradient does not exceed three times that of the calculation of the function itself (see [3]).

IV. CONCLUSION

The efficient algorithm to calculate gradients of the total energy of atoms’ system with respect to Tersoff parameters is presented. The algorithm is based on the Fast Automatic Differentiation technique. The formulas to compute the gradients are derived. These formulas allow to compute the gradients with the machine accuracy and computation time that is needed to calculate the gradient does not exceed three times that of the calculation of the function itself. The following conclusion is made: the calculation of above-mentioned gradient using finite difference method is linked to substantial difficulties.

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REFERENCES


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