

Quantum analysis of the Cosserat media with application to paramagnetic resonance

M. SIKOŃ*

Faculty of Mechanics, Institute of Machine Design, Cracow University of Technology, 37 Jana Pawła II Ave., 31-864 Kraków, Poland

Abstract. This work is a continuation of the investigation which first results were presented in the Bulletin of the Polish Academy of Sciences: Technical Sciences in 2009. In the work the Cosserat medium is defined there, where inter-atomic actions have non-central nature. Non-central potential energy is introduced. As regards the Schrödinger equation the energetic state of the atom is described in the field of the quantum mechanics. The analysis is carried out on the base of mechanical and magnetic properties of the atom. The obtained solution is applied to write the resonance condition in the EPR spectrometer. The spectra, obtained for a loaded hydrated copper sulfate, were interpreted from the point of view of the presented theory and experimental parameters of the Cosserat medium were written.

Key words: EPR method, Cosserat medium, Schrödinger equation.

1. Introduction

The analysis is related to the earlier research of Voigt [1] and Cosserat brothers [2], as summarized by Nowacki [3], and to the works [4–6]. According to the theory the transmission of mechanical action through the surface dividing two neighboring unit cells of material occurs not only via a force vector, but by a couple vector. Therefore, in addition to force stresses one observes couple stresses. Up to now this has not had a complete experimental verification. However, there are well established techniques of experimental investigations which characterize the Cosserat media at the continuum level [7]. The aim of the work is to describe the experimental parameters of the Cosserat medium in nano-scale and measure these parameters in EPR spectrometer. A system of molecular interactions (forces and couples) is interpreted as the result of non-central interaction and the additional motion of an electron is generated, and it leads to the precession of the electron orbits ω^{couple} [8]. The final result of such interactions is appearance of the additional magnetic field B^{couple} leading to magnetization G^{couple} of the continuum [8].

2. Potential of the non-central action

The system of the two one-electron atoms is assumed. The force action between the atoms $P = -\nabla U^P(r)$ is a result of the potential energy $U^P(r)$; where: r is a distance between the atoms; ∇ is the Hamilton operation. When the action P is conducting non-central by electron e , Fig. 1, the moment action is written:

$$M^{couple} = \rho \times P, \quad (1)$$

where ρ is the position vector of the electron e .

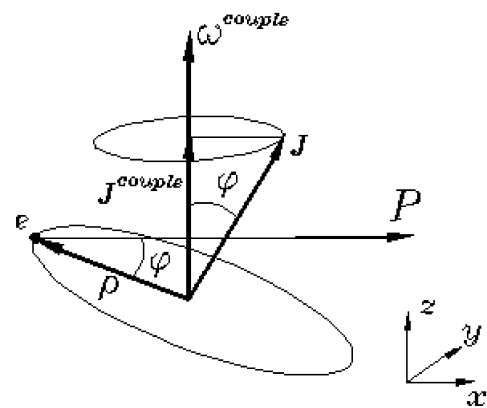


Fig. 1. Electron in the non-central mechanical system of load

The module of the moment (1) equals to: $|M^{couple}| = \rho P \sin \varphi$; where φ is the angle defined as in Fig. 1. The angle φ describes the preferential direction in the space around the atom. This direction is perpendicular to the action P , Fig. 1, and is described by the formula:

$$\varphi = \arccos \frac{|J^{couple}|}{|J|} = \frac{mech_J}{\sqrt{J(J+1)}}, \quad (2)$$

where $|J^{couple}|$ is the projection of the length $|J|$ of the angular momentum J on the preferential direction; $mech_J$ is the mechanical quantum number and J is the resultant quantum number of the moment J .

The change of the angle φ is possible by the rendition of the work: $dA = M^{couple} d\varphi = \rho P \sin \varphi d\varphi$. This work generates the increase of additional potential energy of the atom to the value: $dU^M = dA = \rho P \sin \varphi d\varphi$. By integrating bilaterally the last formula, the potential energy formula is obtained: $U^M = -\rho P \cos \varphi + const$. Finally, $U^M = -\rho P$ is written for $const = 0$.

*e-mail: sikon@mech.pk.edu.pl

The minimum energy U^M corresponds to the place of the stable equilibrium of the atom when the vectors ρ and \mathbf{P} have a parallel orientation. The analysis of the mechanical state of the atom under the action of the non-central loading, Fig. 1, allows to write the equivalent formula which describes the potential of the non-central action:

$$U^M = -\omega^{\text{couple}} \mathbf{J}. \quad (3)$$

For description of the total potential energy of the atom it is required to include not only the energy which is the result of the change of the distance between the atoms r but also the effect of the rotation of the atom φ :

$$U = U^P(r) + U^M(\varphi). \quad (4)$$

3. Introduction to the molecular dynamic of the Cosserat medium

Considering the system $i = 1, 2, 3, \dots, N$ of the interaction atoms of the Cosserat medium, the moment action of the i -atom is defined at the first step. For the atom with Z electrons this moment is written as a sum:

$$\mathbf{M}_i^{\text{couple}} = \left[\sum_{k=1}^Z (\rho_k \times \mathbf{P}_k) \right]_i, \quad i = 1, 2, 3, \dots, N \quad (5)$$

where ρ_k , \mathbf{P}_k are the position vector and the force which belongs to electron k in the i -atom, respectively. The sum (5) can be described according to the phenomena of the precession of the electron orbits in the field of the non-central mechanical actions:

$$\left[\sum_{k=1}^Z (\rho_k \times \mathbf{P}_k) \right]_i = \omega_i^{\text{couple}} \times \mathbf{J}_i, \quad i = 1, 2, 3, \dots, N, \quad (6)$$

where \mathbf{J}_i is the resultant angular momentum of the atom number i which equals to the sum of the orbital L_i and spin \mathbf{S}_i angular momentum; ω_i^{couple} is the precession of the vector \mathbf{J}_i in the plane perpendicular to the action $\mathbf{P}_i = \sum_{k=1}^Z \mathbf{P}_k$.

For the system of the $i = 1, 2, 3, \dots, N$ atoms of the Cosserat medium, the equations of the molecular dynamics is written in the form of the Newton equation for straight line motion $m_i \frac{d^2 r_i}{dt^2} = \mathbf{P}_i$, where: m_i , r_i , \mathbf{P}_i stand for mass, position and force action of the atom number i . Additionally, the equation of the motion of the angular momentum \mathbf{J}_i is including every i -atom under the action of the moment $\mathbf{M}_i^{\text{couple}}$:

$$\frac{d\mathbf{J}_i}{dt} = \mathbf{M}_i^{\text{couple}}, \quad i = 1, 2, 3, \dots, N. \quad (7)$$

When the force $\mathbf{P}_i = -\nabla U_i^P$ is related to the potential of the central actions $U_i^P(r)$, the moment $\mathbf{M}_i^{\text{couple}}$ is described by the potential of the non-central action $U_i^M(\varphi)$

$$\mathbf{M}_i^{\text{couple}} = -\frac{dU_i^M}{d\varphi_i} = \omega_i^{\text{couple}} \times \mathbf{J}_i, \quad i = 1, 2, 3, \dots, N, \quad (8)$$

where φ_i is the angle which describes the preferential direction in the space around the i -atom.

The aim of the analysis of the molecular dynamics of the Cosserat medium is the solution of the additional system of Eqs. (7) independent for the Newton equation for straight line motion.

However, the aim of the presented work is to determine the precession ω_i^{couple} experimentally in the spectrometer EPR.

4. Analogy of the description in macroscale

The description of the motion of the angular momentum \mathbf{J}_i of the i -atoms under the action of the moment \mathbf{M}_i , presented by the system of equations (7), concerns the nanoscale. In the macroscale description, the macroscopic vector of the angular momentum in the elementary volume V is defined in this form:

$\mathfrak{S} = \frac{1}{V} \sum_1^N \mathbf{J}_i$. The state of the mechanical polarization is written by the formula:

$$\frac{d\mathfrak{S}}{dt} = \frac{\mathfrak{S}^{\text{couple}} - \mathfrak{S}(t)}{T}, \quad (9)$$

where $\mathfrak{S}^{\text{couple}}$ is the stationary value of the vector \mathfrak{S} ; T is the time constant of the mechanical polarization. The rotation of the vector \mathfrak{S} to the stationary state $\mathfrak{S}^{\text{couple}}$ is connected with the rotation of the elementary volume V and belongs to the continual Cosserat medium.

The mechanical polarization of the vector \mathfrak{S} leads to the couple stresses.

5. Quantum nature of the non-central potential energy

The assumption to eliminate the orbital angular momentum by the action of the crystal field, $\mathbf{L} = 0$, $\mathbf{J} = \mathbf{S}$ takes place. The energy contribution due to the electron spin \mathbf{S} under a non-central mechanical loading \mathbf{P} , Fig. 1, can be written in this form:

$$U^{\text{couple}} = -\omega^{\text{couple}} \mathbf{S}. \quad (10)$$

There ω^{couple} is the precession vector of spin due to loading \mathbf{P} . Equation (10) can be re-written in the operator form as follows:

$$\hat{S}\Phi = \Omega^{\text{couple}} \Phi, \quad (11)$$

there Φ is the wave function, Ω^{couple} represents the eigenvalues of the operator \hat{S}

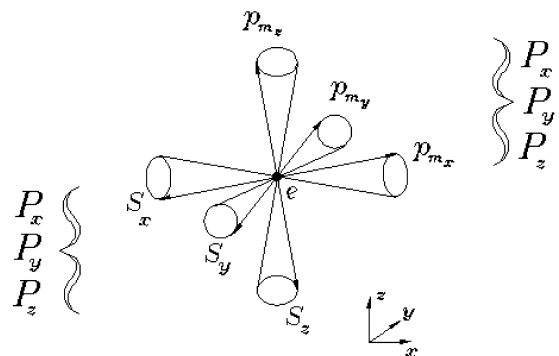


Fig. 2. Precession of the spin components (S_x, S_y, S_z) and magnetic moments ($p_{m_x}, p_{m_y}, p_{m_z}$) in nonsymmetrical field (P_x, P_y, P_z)

Equation (11) can now be written in terms of the components P_x, P_y, P_z Fig. 2, which correspond to the Schrödinger equation written in the Cartesian coordinate system:

$$\hat{S}_i \Phi_i = \Omega_i^{couple} \Phi_i, \quad i = x, y, z. \quad (12)$$

By application the Pauli operators:

$$\hat{S}_x = \frac{\hbar}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad (13)$$

$$\hat{S}_y = \frac{\hbar}{2} \begin{pmatrix} 0 & -\sqrt{-1} \\ \sqrt{-1} & 0 \end{pmatrix}, \quad (14)$$

$$\hat{S}_z = \frac{\hbar}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (15)$$

and the spin wave functions:

$$\Phi_x = \begin{cases} \Phi_x^+ = \begin{pmatrix} 0 \\ 1 \end{pmatrix} & \text{for } mech_{S_x} = \frac{1}{2} \\ \Phi_x^- = \begin{pmatrix} -1 \\ 0 \end{pmatrix} & \text{for } mech_{S_x} = -\frac{1}{2} \end{cases} \quad (16)$$

$$\Phi_y = \begin{cases} \Phi_y^+ = \begin{pmatrix} 0 \\ \sqrt{-1} \end{pmatrix} & \text{for } mech_{S_y} = \frac{1}{2} \\ \Phi_y^- = \begin{pmatrix} \sqrt{-1} \\ 0 \end{pmatrix} & \text{for } mech_{S_y} = -\frac{1}{2} \end{cases} \quad (17)$$

$$\Phi_z = \begin{cases} \Phi_z^+ = \begin{pmatrix} 1 \\ 0 \end{pmatrix} & \text{for } mech_{S_z} = \frac{1}{2} \\ \Phi_z^- = \begin{pmatrix} 0 \\ 1 \end{pmatrix} & \text{for } mech_{S_z} = -\frac{1}{2} \end{cases} \quad (18)$$

where $mech_{S_i}$, $i = x, y, z$ denotes the spin quantum number for the axes x, y and z , Eq. (12) has the following form:

$$\hat{S}_i \Phi_i = \hbar mech_{S_i} \Phi_i, \quad i = x, y, z. \quad (19)$$

They are identically satisfied. Using (10) and an analogy between equations (12) and (19) the eigenfunctions for both sets of equations can be cast in the form:

$$U_i^{couple} = \omega_i^{couple} \hbar mech_{S_i}, \quad i = x, y, z. \quad (20)$$

When the mechanical loads are applied to a Cosserat medium it is expected that the energy levels will split according to the value of the quantum numbers $mech_{S_i} = \pm 1/2$, $i = x, y, z$, resulting in the following energy levels:

$$mech_{S_i} = \frac{1}{2}, \quad U_i^{+couple} = U_o + \frac{\hbar}{2} \omega_i^{couple}, \quad i = x, y, z, \quad (21)$$

$$mech_{S_i} = -\frac{1}{2}, \quad U_i^{-couple} = U_o - \frac{\hbar}{2} \omega_i^{couple}, \quad i = x, y, z, \quad (22)$$

here U_o denotes the energy level of the undisturbed state. The difference between two adjacent energy levels is:

$$\Delta U_i^{couple} = \hbar \omega_i^{couple}, \quad i = x, y, z. \quad (23)$$

For splitting energy (21) and (22) we obtain the couple line frequency:

$$\omega_i^{+couple} = \omega_o + \frac{\omega_{S_i}^{couple}}{2}, \quad i = x, y, z, \quad (24)$$

$$\omega_i^{-couple} = \omega_o - \frac{\omega_{S_i}^{couple}}{2}, \quad i = x, y, z, \quad (25)$$

here ω_o is the frequency of the precession of an undisturbed electron.

Precession of an electron generates the additional magnetic field B^{couple} , Fig. 3.

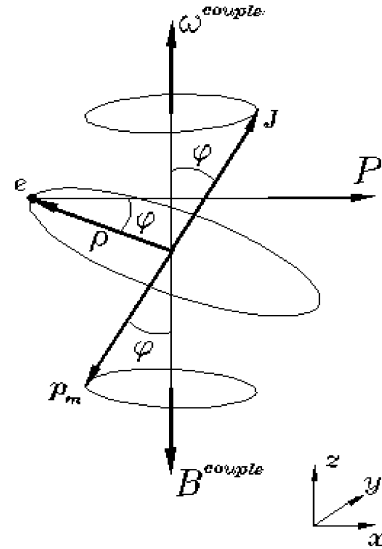


Fig. 3. Magnetic field generation in noncentral interatomic action

Spin energy under the influence of the mechanical load can now be written as follows:

$$U^{couple} = -p_{m_S} B^{couple} = -p_{m_{S_i}} B_i^{couple}, \quad i = x, y, z. \quad (26)$$

Since:

$$p_{m_i} = -(g_s)_{ij}^{couple} \vartheta mech_{S_i}, \quad i = x, y, z, \quad (27)$$

where $(g_s)_{ij}^{couple}$ is the tensor spin coefficient of spectroscopic energy levels splitting accounting for noncentral interaction, ϑ is the Bohr's magneton. The energy levels can now be represented as:

$$mech_{S_i} = \frac{1}{2}, \quad U_i^{+couple} = U_o + \frac{1}{2} (g_s)_{ij}^{couple} \vartheta B_i^{couple}, \quad (28)$$

$$i = x, y, z,$$

$$mech_{S_i} = -\frac{1}{2}, \quad U_i^{-couple} = U_o - \frac{1}{2} (g_s)_{ij}^{couple} \vartheta B_i^{couple}, \quad (29)$$

$$i = x, y, z,$$

and then an expression for the difference between two adjacent levels is written:

$$\Delta U_i^{couple} = (g_s)_{ij}^{couple} \vartheta B_i^{couple}, \quad i, j = x, y, z. \quad (30)$$

Comparing (23) and (30) we obtain the condition:

$$\hbar\omega_i^{couple} = (g_s)_{ij}^{couple} \vartheta B_i^{couple}, \quad i = x, y, z. \quad (31)$$

Due to the interaction spin-orbit, the precession of spin and the resulting magnetic field B^{couple} can generate orbital motion of the electron. Then condition (31) we write in the form:

$$\hbar\omega_i^{couple} = g_{ij}^{couple} \vartheta B_i^{couple}, \quad i, j = x, y, z, \quad (32)$$

here ω_i^{couple} , g_{ij}^{couple} , B_i^{couple} represents both spin and orbital effect of the analyzed phenomena. In the case of hydrostatic loading we have:

$$\omega_i^{couple} = \omega^{couple}, \quad i = x, y, z, \quad (33)$$

$$B_i^{couple} = B^{couple}, \quad i = x, y, z, \quad (34)$$

$$g_i^{couple} = g^{couple}, \quad i = x, y, z. \quad (35)$$

There g^{couple} denotes the hydrostatic effect factor of the stressed lattice. For hydrostatic state the condition (32) can be cast into the form:

$$\hbar\omega^{couple} = g^{couple} \vartheta B^{couple}. \quad (36)$$

6. Magnetic polarization of the Cosserat medium

Taking into consideration the magnetic property of the atom, the moment action is written in the form:

$$\mathbf{M}^{couple} = \mathbf{p}_m \times B^{couple}. \quad (37)$$

For the system of the $i = 1, 2, 3, \dots, N$ interaction atoms of the Cosserat medium, the additional motion equation of the molecular dynamics can be described in the form:

$$\frac{d(\mathbf{p}_m)_i}{dt} = \gamma (\mathbf{p}_m)_i \times B_i^{couple}, \quad i = 1, 2, 3, \dots, N, \quad (38)$$

where γ is the magnetomechanical ratio; $(\mathbf{p}_m)_i$ is the magnetic moment of the i -atom; B_i^{couple} is the magnetic induction associated with the non-central interatomic action of the i -atom.

The aim of the presented work is to determine the magnetic induction B_i^{couple} by the experimental procedure in the EPR spectrometer.

By the analogy with the mechanical polarization, the process of the magnetic polarization of the macroscopic magnetization vector $\mathbf{G} = \frac{1}{V} \sum_1^N (\mathbf{p}_m)_i$ can be described by the formula:

$$\frac{d\mathbf{G}}{dt} = \frac{\mathbf{G}^{couple} - \mathbf{G}(t)}{T}, \quad (39)$$

where \mathbf{G}^{couple} is the magnetization vector in the stationary state and T is now the notation time constant of the magnetic polarization.

The phenomenon of the magnetic polarization leads to the magnetization of the volume V of the Cosserat medium in the field of the mechanical action.

7. Cosserat medium in cavity of the EPR spectrometer

The condition of the resonance for the Cosserat medium in the EPR spectrometer can be written in the following form:

$$\hbar(\omega + \omega^{couple}) = (g + g^{couple}) \vartheta (B_o + B^{couple}), \quad (40)$$

there ω is an angular velocity of the transverse field in the spectrometer, g is the spectroscopic factor for an unloaded crystal, g^{couple} is the spectroscopic factor describing the interaction resulting from the stressed crystallographic lattice, ϑ is the Bohr's magneton, B_o is the induction of the constant magnetic field of the spectrometer, while $\hbar = h/2\pi$, in which h is the Planck constant.

Energy absorption within an atom of the medium placed in the cavity of the spectrometer can be described by, cf. [8]

$$N = \frac{\omega B_1 T_2 [G_o (\omega_1 + \omega^{couple}) - G^{couple} (\omega_o - \omega_1)]}{1 + [(\omega_o + \omega^{couple}) - \omega]^2 T_2^2}. \quad (41)$$

Here B_1 is the amplitude of the alternating magnetic field of the spectrometer, G_o denotes magnetization caused by the constant field within the spectrometer, T_2 is the lateral relaxation time, $\omega_1 = \gamma B_1$ and $\omega_o = \gamma B_o$, where γ is the magneto-mechanical coefficient.

Equations (40), (41) differ in the case when there are no couple stresses, $\omega^{couple} = 0$, $g^{couple} = 0$, $B^{couple} = 0$ and $G^{couple} = 0$, [9].

It is expected that the presence of the couple stresses will be revealed by changes in the resonance frequency, resonance magnetic induction, the factor of spectroscopic fission, and resonance energy absorption (which is proportional to the intensity of the EPR signal).

8. Measurements

All measurements were done on the spectrometer ELEXYS 500 manufactured by Bruker of Karlsruhe within the frequency band X at 9.5 GHz. The spectra were recorded at modulation amplitude of 5 mT and power of the microwave 10 mW. The specimen tested were the crystals of hydrated copper sulfate $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ while the tests were performed at room temperature. External hydrostatic loading was executed by means of chemical and thermal contraction occurring within an epoxy resin in which the crystals of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ were embedded. The epoxy resin E51 was hardened with triethylenetetramine (TETA) diluted in toluene. The hardener was applied in the proportion of 10 weight units per 100 weight units of the resin. EPR spectra of unloaded ("free") crystals were then compared with those obtained for crystals embedded in the resin and subjected to contraction. A background spectrum of the resin itself was also measured and its intensity was found to be several orders of magnitude lower than the intensity of the spectrum obtained for the $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ spectrum. Thus, it was concluded that the "noise effect" was negligible.

9. Results

The results of the spectral measurements are shown in form of graphs illustrating the dependence of the EPR signal I on the constant magnetic field B_o as shown in Fig. 4. For comparison the spectra for unloaded and loaded specimens are shown side by side.

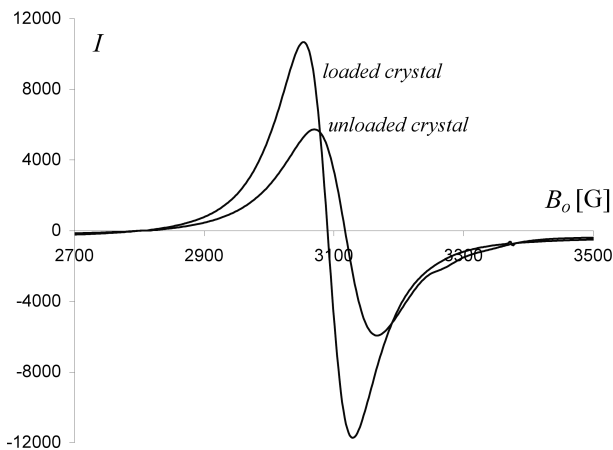


Fig. 4. EPR spectrum of the loaded and unloaded crystal $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$

Available parameters:

Sampling Time [s]	0.16384
Field Mod. Amplitude	0.0005
Field Mod. Frequency [Hz]	100000
Microwave Frequency [Hz]	9.75098×10^9 – unloaded crystal 9.46777×10^9 – loaded crystal
Microwave Power [W]	0.0100825
Receiver – Gain	20
Receiver – Time Constant [s]	0.04096
g – factor	2.233 – unloaded crystal 2.187 – loaded crystal
Resonance induction [G]	3117 – unloaded crystal 3090 – loaded crystal
Intensity of EPR signal [units]	51714 – unloaded crystal 102571 – loaded crystal

10. Discussion

A detailed comparison of the EPR spectra obtained for loaded and unloaded crystals $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ reveals substantial differences in the resonance frequencies, resonance magnetic induction, the g-coefficient and the intensity of the signal. The observed differences are in agreement with the theoretical predictions obtained from formulae (40) and (41).

These differences are interpreted as manifestation of the non-central interactions between atoms. At the level of an unit cell volume the effect should be understood as a proof of

existence of couple stresses predicted by the Cosserat Theory for the continuum. These stresses result as the averages of the atomic interactions. The end result of the additional motion of the electrons in form of precession is the *couple stresses*. The angular velocity of the precession is $\omega^{couple} = 0.28321 \times 10^9$ Hz. The sense of the vector of precession is opposite to “z” axis, see Fig. 1. The magnetic field generated by the couple stresses is of the magnitude $B^{couple} = 27$ G. The Larmor precession for $B^{couple} = 27$ G is calculated: $\omega_L = \frac{e}{2m} B^{couple} = 0.23744 \times 10^9$ Hz, where e is an elementary charge and m is an electron mass. Difference between ω^{couple} and ω_L is explained by an influence of the crystal lattice of the $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$.

The value of the g-coefficient changes from 2.233 to 2.187 for an unloaded and loaded crystal. An intensity of the EPR signal due to the couple stresses is $I^{couple} = 50\,857$ units.

Results described here provide a new light into molecular motion and deformation at nano-scale level. The present model provides details of the atomic interactions not available in the currently applied models of atoms of a system of material points, cf. [10]. This is a valuable insight into the reality of nanomechanics applied at an atomistic level.

Investigation of the relations between a system of atoms interacting via nano-couples acting at the atomic level and the couple stresses observed in the continuum is the essence of the Nanomechanics pertaining to the Cosserat media. As the next step we consider incorporation of the nanoeffects in a form of interatomic couples into the theory of accumulation and propagation of defects in materials, cf. [11]; and one obtains the macro-description of fracture occurring in the Cosserat media at the continuum level. An analysis of systems of atoms subjected to an external stress is helpful in many areas of Nano-engineering and it may be applicable in design of molecular devices, in which the behavior of atoms is controlled by the external loads.

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