ISSN 2229-5518

Ab-initio quantum mechanical model for spin-strain coupling in ferroics

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1 Abstract

The quantum mechanics is employed to analyze and model the novel inter dependence between the electronic properties and how they are coupled with the lattice structure. Various classical as well as numerical models have been compared with the quantum mechanics to strengthen the argument in favor of the latter. Finally, a viable model is presented before the general reader as well as the physicist to assist the fundamental materials science and engineering.

2 Introduction

This section will introduce a quantitative overview of different techniques used for modeling of the magneto electric effects. We observed that the *ab-initio* computations are one of the most suitable approaches in understanding the ferroelectric-ferromagnetic interfaces. The material interactions are mainly based on interface formed by surfaces which comprise of few atomic layers. The earlier models have evolved to accept the importance of these interface effects between the thin films of ferroelectric and ferromagnetic materials. The composite thin films evolved due to the weak magnetoelectric effects found in single phase ferroics like Cr_2O_3. The thin-film composites of ferroelectric and ferromagnetic materials have improved the magnetoelectric effects by 3 orders of magnitude at room temperature. The research into the nano structures of composite films promises to improve the magneto-electric effects further. The nanostructure possess large surface to volume ratios and minimum clamping from the substrate layer. The ferroic interface is the region which approximately consists of limited number of monolayers. These monolayers have different

material properties which are determined by the fabrication methods, dimensions and crystalline properties of the constituent materials.

The *ab-initio* Hartree Fock and Density Functional techniques [1] are proposed to be used to understand the materials interactions at surface interfaces formed by limited number of monolayers on each side of the interface. The *Crystal06 code* [1] may be employed to compute the total energy, electronic and mechanical properties of the bulk $BaTiO_3$ and its material interface with ferromagnetic Fe. The spin device is proposed which is based on the functioning of interfaces. The material interfaces are employed to regulate the exchange interaction of spins by strains. The exchange interaction couples spins with nearest neighbors for split up of spins in spin up and spin down channels. The specific spin is flipped by the application of strain applied by an external terminal attached with the ferroelectric channel. The spin exchange is modulated through the dipoles in the ferroelectric phase inside a ferroelectric channel.

The atomistic nature of ferroelectric and ferromagnetic surface interface interactions can be achieved by *ab-initio* simulations. The crystalline structure of the interface and atomistic interactions determine its electronic properties. The exchange interactions that determine the ferroic properties crystalline field spin orbit coupling and zeeman effects which are arise from electron-ion interactions. In quantum field and perturbation theories the field and material interactions are defined as a correlation function. These correlations resemble traditional Green's function discussed already. The operators representing the material-field interactions resemble boson particles. This formalism is utilized by quantum known as the second quantization.

2.1 Green's function for multi-field materials

In general form, the multi-field materials may have Green's function solution depending upon the boundary conditions and the multi-field material properties. The non-homogeneous differential equation with a Green's function $G(x; y)$ [9] for the boundary conditions of multi-field materials can be expressed as

$$L(x)G(x;\ y) = \delta(x - y) \tag{1}$$

Here L(x) is a linear differential operator and G(x;y) is a Green's function with x and y representing the source and observation points. For an arbitrary function f(x) the linear differential equation

$$L(x)u(x) = f(x, y) \tag{2}$$

With the Green function replacing u(x) with G(x;y) as the response to the arbitrary function f(x,y)

$$G(x; y)f(x) = \delta(x-y) \tag{3}$$

The arbitrary forcing function $f(x, y)$ is replaced with *Dirac Delta* function in the last equation. If the arbitrary forcing function is supposed as $\delta(x - y)$ and the solution of Green's function may become simple. When a differential operator L is applied on Green's function with particular boundary conditions then the same solution could be extended arbitrarily inside the microscopic material. These features of the Green's

ISSN 2229-5518
function coupled with its physical interpretation make it useful function in deriving the multi-field magnetoelectric coefficient. Green's function is also applied on elastic, electro- static, magnetostatic materials prior to its application to multi-field material boundary conditions. The results of Green's functions are well understood for problems for the microscopic boundary conditions. The application of Green's function and other boundary element methods has succeeded to lay down the model. These models predict the macroscopic magneto electric (ME) effect which arise from microscopic interactions between ferromagnetic and ferroelectric material interactions at an interface.

The Green's function is often employed as a solution to the non-homogeneous differential equation excited by an arbitrary point source representing electric potential, magnetic potential and point forces. The multifield material response to an arbitrary excitation is modeled by a differential equation with a Green's function. The Green's function is depending upon the position of the microscopic source and observation points for the microscopic displacement, magnetic and electric potentials. The characteristics of these microscopic point sources are approximated by $\delta(x - y)$. The linear differential operator L and the boundary conditions of the model equation are also defined for solving the Green's function. The Green's function solves the linear differential equation $L(x, y)u(x, y) = f(x, y)$ for permittivity, permeability, piezoelectric coefficient and piezo-magnetic coefficient tensors. The properties of the Delta function are utilized well for the microscopic nature of the magneto-electric interactions across boundary conditions.

We can compare the Green's function solution with the ab-initio quantum mechanical computations and we may observe that the Green's function so-

lution for the magneto-electric coupling may not have taken into account the material properties of the interface. After setting up the multifield bound- ary interaction as a linear differential equation having the Green's function solution the next step is to solve the Green's function itself. This is done by various methods such as Radon transform, potential functions and Laplace transform. The main steps of the the solution to the Green's function for a multifield boundary value problem is reported as under:

* The constitutive multifield equations are based on Gauss's law for the electric, magnetic and the microscopic displacement potentials. The constitutive equations for the multifields are formulated from the Gauss's law for the displacement D_{ij} , magnetic flux density B_{ij} and electric displacement u_{ij}

$$D_{ij} = 0, \quad B_{ij} = 0, \quad u_{ij} = 0 \qquad (4)$$

The constitutive multifield equations express the permittivity, magnetoelectric coefficient, stress, piezoelectric coefficient, piezomagnetic coefficient and permeability as c_{ij} , α_{ij} , σ_{ij} , , q and μ_{ij}

$$D_i = c_{i,j} E_j + \alpha_{i,j} H_j + {}_{i,j}\ \sigma, B_i = \alpha_{i,j} E_j + \mu_{i,j} H_j + q_{i,j} H_{i,j} \qquad (5)$$

- The Green's function $G_{MR,in}(x - x^{-})$ is employed to solve the constitutive equation for c_{ij}, α_{ij}, σ_{ij}, , q and μ_{ij}. The E_{iJMn} is a general notation representing E, H and strain. The response to the fields is expressed as $\delta_{JR}\delta(x - x^{-})$.

$$E_{iJMn} G_{MR,in}(x - x^{-}) + \delta_{JR}\delta(x - x^{-}) = 0 \qquad (6)$$

- Solution of the Green's function by radon transform, potentials or

laplace transform methods leads to a matrix solution having Green's function solutions representing permeability, permittivity, piezoelectric coefficient, piezomagnetic coefficient and magneto-electric as coefficients.

- The magneto-electric coefficient is a 3×3 tensor expressed as

$$\alpha_{ij} = k[f(1-f)eq]/[k^m + k^e] \quad (7)$$

The terms f, 1-f represent the piezoelectric and piezomagnetic fractional volume in the composite structure. The term e, q, k^m and k^e represent piezoelectric coefficient, piezomagnetic coefficient, bulk modulus of ferromagnetic, the ferroelectric phases of the composite. The Green's function takes into account the coupling terms and provides the solution to a complex problem consisting of boundary conditions and micro mechanical material parameters. The closed look at this model would clarify that it lacks the crystalline geometry of the interface atoms of the piezoelectric and piezomagnetic materials at material boundaries. The coupling coefficient is not based on the electronic structure and fabrications characteristics of the interface. The magnetoelectric coefficient is based upon elastic constants, bulk moduli, piezoelectric and piezomagnetic coefficients of the constituent materials phases. The magneto- electric coefficient employs experimental values of elastic constants and bulk moduli for the bulk materials. The boundary conditions between piezoelectric and piezomagnetic materials are poorly represented approximations and need to be well defined. The values of the macroscopic material parameters such as piezoelecric coefficients, piezomagnetic coefficients, elastic constants and bulk moduli are used which are approximations.

The microscopic details of the magnetoelecric coefficient lack the surface interface, defects affecting the electronic configuration of the interface. The surface interface effects are the most dominant effects at interfaces between ferromagnetic and ferroelectric materials which are not explicitly treated in microscopic magnetoelectric effects. The experimental values for the piezo-electric, piezomagnetic, elastic constants and bulk moduli of the bulk ferromagentic and ferroelectric materials are employed in computing the ME coefficient. The model must incorporate the surface effects which consist of few atomic layers only. The effects on ME interactions due to defects, impurities, chemical bonding are not taken into account in the existing model. The frequency dependence of the magnetoelectric effect is not clear. The detection of spin polarization in a ferromagnetic part of composite thin film material by the application of electric fields on a ferroelectric thin film part is not experimented as often as the converse of it for ME interactions. The major focus of the fabrication and modeling efforts in ME effect has been the detection of electric polarization in a ferroelectric part of composite material with application of magnetic field in a ferromagnetic section of the thin film. One obvious reason seems to be the fact that the detection ME effect by electric polarization is pronounced as compared with the detection of the spin polarization.

The relevance of quantum mechanics to the magnetoeletric effect comes from the fact it is limited to few atomic layers on each side of interface formed between ferroelectric and ferromagnetic thin film surfaces. The interface magnetoelectric effect for the interface formed between ferromagnetic and ferroelectric materials is limited to two dimensional regions on each side of interface. The quantum ME effect [2, 19] may be termed as quantum form of the macroscopic ME effect reported by

ISSN 2229-5518

Sirinivasan, Petrov and Bichirin [2]. The ferromagnetic and ferroelectric thin films surfaces interactions may be represented by the spin and lattice interactions. These spin and lattice interactions at interface may be represented by quantum particles.

These quantum particles interact through the interaction coefficients

The magntoelectric coefficient is an effective elastic constants or bulk moduli. The application of electric or magnetic fields may deviate the effective elastic properties of the interface from the equilibrium depending upon the magnitude and frequency of the E or H fields. The dependence of interfaces on E and H fields is thrown away for the time being to justify the fact that ME effect is a purely mechanical interaction according to its definition. The external excitations of fields only perturb the mechanical interaction from its equilibrium value. This generalization may help further as the excitation may be expressed as a general term which represents changes in stresses, fields, and temperature.

In quantum field theory the Green's function has infinite dimensions which is different than the basic two-point Green's function. In quantum field theory, it represents a correlation between the quantum field operators. So in quantum field theory the particle interactions essentially become many body in nature. In calculus, it is a non-homogenous differential equation whose solution is a two point Green's function. The relevance of quantum field theory and Green's functions comes from the fact that boundary value problem in macroscopic multifield and the particle interactions in an interface between ferromagnetic and ferroelectric material are focused on interactions that last for microscopic distances only. The main difference lies in the fact that quantum mechanical effects are based on electron and ion interactions around interface and are periodic in nature due to the lattice arrangement of atoms located at the interface.

The interaction between the ferroelectric and ferromagnetic material at interface is now reduced to an interaction between quantum fields represented as particles. The surface magnetization and surface polarization fields are different than the bulk magnetization and polarization fields. The

ISSN 2229-5518
solution of the macroscopic magneto-electric effect with Green's function techniques make this approach unified as the macroscopic solution carries the interface coupling parameter term which is quantum mechanical in nature. In quantum field theory, every field is represented by operators which inter- act quantum mechanically. The spins are represented by bosons which are named as magnon particles. These magnons are small excitations in the spin arrays which appear as spin waves. The Hamiltonian with Heisenberg's spin exchange interaction [7] between spins of nearest neighbors is expressed as

$$E = -J\, S_{\boldsymbol{j}}\, S_{\boldsymbol{j}+\delta} - 2\mu_O H_O S_{\boldsymbol{j}z} \qquad (9)$$

In equation 9 the terms E, J, $S_{\boldsymbol{j}}$, $S_{\boldsymbol{j}\delta}$, μ_O, H_O and $S_{\boldsymbol{j}z}$ represent the energy, exchange integral, spin angular momentum operator at site $\boldsymbol{j}$ and $\boldsymbol{j}\delta$, Bohr magneton term, applied H field and spin angular momentum operator in positive z direction. The term δ is a vector that connects atom j with the nearest neighbor in bravais lattice. The factor of 2 appears due to the gyro- magnetic term for spin angular momentum. The spin S is further expressed as a complex term having x and y components

$$S_{\boldsymbol{j}}^{+} = S_{\boldsymbol{j}}x + iS_{\boldsymbol{j}} \qquad (10)$$

The $a^{\dagger}$ and $a_{\boldsymbol{j}}$ are atomic variables representing small perturbations

$$S_{\boldsymbol{j}} = S_{\boldsymbol{j}}x - iS_{\boldsymbol{j}} \qquad (11)$$

The spin consists of x, y and z components which are not independent. The Holestein-Primakov transforms the spin operator further into independent creation and annihilation operators which are $b^{\dagger}$, b_k, $a^{\dagger}$ and $a_{\boldsymbol{j}}$

The commutation relations between the operators are

$$[a_{\boldsymbol{j}}, a^{\dagger}] = \delta_{\boldsymbol{j}\boldsymbol{l}} \tag{13}$$

for correct commutation relations for S^{+} and S^{-}. The magnon variables are defined as

$$b_k = N^{-1/2} e^{-ik.x_{\boldsymbol{j}}} a_{\boldsymbol{j}} \tag{14}$$

$$b_k^{+} = N^{-1/2} e^{-ik.x_{\boldsymbol{j}}} a_{\boldsymbol{j}}^{+} \tag{15}$$

And the atomic variables are

$$a_{\boldsymbol{j}} = N^{-1/2} e^{-ik.x_{\boldsymbol{j}}} b_k, \tag{16}$$

$$a_{\boldsymbol{j}}^{+} = N^{-1/2} e^{-ik.} b^{+} \tag{17}$$

The spin may be expressed in atomic or magnon variables as

$$S_{jz} = S - a_j^{+} a_j \tag{18}$$

The characteristics of these variables are expressed by commutation relationships

$$[b_k, b_l^{+}] = \delta_{jl} \tag{19}$$

$$[b_k^{+}, b_k^{+}] = [b_k, b_k] = 0 \tag{20}$$

The interactions [8] which are responsible for the finite lifetime of the magnons are classified according to the number of the magnon operators that are associated with them. We may introduce the impurities and structural defects in the interface regions between ferroelectric and ferromagnetic thin films. These defects act like surface pits which may act as the source of the *two − magnon* processes. In these processes the momentum of the surface ferromagnetic spin system is not conserved. So, the excess of this pesudo-linear momentum is transferred as the linear momentum of the ferroelectric surface lattices. The interaction Hamiltonian for these impurities can be expressed as

$$H_{imp} = F_q b_{k+q}$$

The magnon-phonon interactions at surface interface between ferroelectric and ferromagnetic materials may become has significant contribution due to surface exchange or surface anisotropy. These interactions are modulated by the ionic displacements taking place in surface regions of ferroelectric and across the surface interface. It is generally assumed that ions are rigid and

electrons significantly follow the displacements of the ionic rigid cores. Although ions are supposed to stay in their ground state. On the contrary the ionic cores make virtual transitions to excited states. The electrons from the ferromagnetic side and phonons from the ferroelectric side of interface contribute to the magnon-phonon interactions which are very important for the dielectric-metal and dielectric-dielectric ferroelastic interfaces. The first and higher order magnon-phonon interactions allow relaxation of the magnetization and polarization fields at the interface between the ferroelectric and ferromagnetic materials. The relaxation of magnons may also be expressed as the macroscopic differential equation for magnetization. The differential equation representation is

$$dM(r,t)/dt = \gamma M(r,t) * B_{eff}(r,t) \qquad (22)$$

The damping term γ represents the relaxation mechanisms as suggested by Landau-lifshitz and Felix Bloch and N. Bloembergen for ferromagnetic relaxation. The two-magnon and three-magnon processes are main relaxation mechanisms for ferromagnetic magnetization. The relaxation mechanisms can be thought as the equilibration of the spins to the precessions conserving the z component of spin S_z or S^2. In three-magnon processes the basis mechanism is the dipolar interaction

$$H = [3\gamma^2 \bar{(}2S)]/2 \quad F_{lm}[a^+ a_l a_l - a_l a^+ a^+ a_m] \qquad (23)$$

In a magnon-phonon interaction at interface, the effective exchange integral and the dipolar interaction term depends upon the position

of ions in a lattice and external perturbations. The distance between ions change in a predictable manner. The distance between the nearest neighbors may be expressed as

$$R_{ab} = R_b - R_a = R_{ab} - \delta R_b - \delta R_a \qquad (24)$$

The term $\delta R_{a,b}$ expresses the instantaneous displacements of the respective ions in a lattice. These ionic displacements may be expressed as linear combinations of the phonon creation and annihilation operators. If we consider a single ion the α component of the displacement of l ion is expressed as

$$\delta R_{\alpha,l} = [(\hbar/NM_{at})\ 1/\ (2\omega_s(k)[e^{(s)}(k)e^{ik.R_l}\ a_s(k) + e^{(s)}(k)e^{-ik.R_l}a^{+}(k)\ \mathbf{(25)}$$

Where $a^{+}(k_s)$ and $a_s(k)$ creates and annihilates a phonon wave vector k inside the Brillion Zone (BZ). The s and $\omega_s(k)$ expresses the polarization and the corresponding dispersion relation. Then replacing the $e_s(k)$ and simplifying further leads to

$$\delta R_l = 1/\sqrt{(N)}\ g_s(k)e^{-ik_s.R_l}\ [a^{+}(k) - a_s(-k)] \qquad (26)$$

In a Heisenberg Hamiltonian effective exchange integral $J(R_{lm})$ can be expanded in a Taylor series around the equilibrium ion distances as

$$J(Rm) = J(Rm) + \delta J(R_l m)\ (\delta R_l - \delta R_m) + \ldots\ldots\ldots \ (27)$$

After the substitution of the effective exchange term and the ionic displacement term from last two equations, the magnon-phonon interaction

Hamiltonian appears as

$$H_{mp} = \sum_{k,q} \varphi^{\delta}_{k,q} b^{+}_{k-q} b_k (a^{+}_{qs} - a_{qs} \qquad (28)$$

wherea s the term $\varphi^{\delta}_{k,q}$ is defined as

$$\varphi^{\delta}_{k,q} = 4S \div (\bar{N})_{\delta} g_s(k)_{.\delta}(e^{-ik} - 1)_{\delta}(1 - e^{-iq}) \qquad (29)$$

The equations express the magnons and phonons coupling across the interface in a region that is only couple of atomic lengths apart. The atomic displacements δ in the atomic layers around the interface region are significantly important for the interface spin exchange and interface lattice displacements. The limited number of atomic layers comprising the interface region points at the importance of the ab-initio Hartree Fock and Density Functional Theory being the appropriate method to investigate the interface properties.

The interaction between phonons may be utilized to compute the life times and energy renormalization of phonons and magnons at the interface.

Due to the magnetic, electric, stress and thermal perturbation in a ferro-magnetic or ferroelectric phase of the composite the magnon population in ferromagentic phase deviates from their equilibrium and decay via phonons. The deviation of the phonons from their equilibrium population may decay through scattering and end in magnons excitations. This is particularly relevant here with the experimental evidence of magnetoelectric effect by Sirinivasan and other groups. The impedance of the composite thin film structure is supposed to be high such that no charge transport takes place during these interacting processes. Any leakage currents may alter the magnon-phon re- laxation mechanisms in a sufficient way.

The relaxation of the excess energy possessed by phonons in a ferroelec- tric phase to ferromagnetic phase and magnons from ferromagnetic phase to ferroelectric phase may be controlled by the spin-lattice and magnon-phonon τ_{mp} relaxation time. The precondition for utilizing this excess energy is that magnon relaxation times τ_{mm} and phonon relaxation time τ_{pp} is longer than the magnon-phonon interaction relaxation time τ_{mp}. It can be assumed that the scattering times are very short for the poor crystalline

quality and the amorphous or polycrystalline structures. **It** is supposed that the unperturbed and perturbed energy spectra of magnons and phonons are different from each other [8] in FM both at low temperatures and near near critical temperatures (Tc). However there is an interval for values of temperature T and k such that the dispersion relations of magnons and phonons inter- sect. This intersection implies the mixing of magnon-phonon modes. The mixed magnon-phonon modes are magneto-elastic modes which appear only at the long wave vector k limit. The bilinear magnon-phonon interaction appears only at non equilibrium. If we consider the equation expressing dipolar Hamiltonian

$$(-3\gamma^2 S\ (2S))/4\,[F(R_{lm})a_l] \qquad (30)$$

If we expand the $F(R)$ in Taylor series around the equilibrium position

$$F(R^0_{lm}) = F(R^0_{lm}) + \delta F(R_{lm})^0(\delta R_l - \delta R_m) + \ldots\ldots\ldots \qquad (31)$$

expanding $\delta(R)$ in phonon operators we obtain a hamiltonian that is bi-linear in phonon operators.

$$H_{mp} = \sum_q b^+(G_q a^+_{-q} + G^* a_q) \qquad (32)$$

ISSN 2229-5518

The hamiltonian may express the mixing of the magnon and phonon excitations on the ferromagnetic and ferroelectric interface surface. The region where the magnons and phonons seem to overlap in ω, q plane is called the nominal crossing as the dispersions never cross each other in real sense. Generally, magnon has finite frequency at k equal to zero. The acoustic phonons have dispersion relation linear in k. At small k both excitations are well defined and magnons are always in the top branch in dispersion. There are regions where magnons and phonons turn into mixed magnetoelastic waves. At higher values of k the magnons and phonons always interchange and the lower branch becomes a magnon and the upper one switches to a phonon mode. This can be expressed by three part Hamiltonian containing Hamiltonian for magnon, phonon and inetraction hamiltonian separately as i perturbation theory

$$H_o + H_{mp} = \omega_k b^+ b_k + \sigma_k a^+ a_k + a^+$$

The parts first and second part are unperturbed Hamiltonians representing spin and phonon energies in the ferromagnetic and ferroelectric sections of composite structures. The third part represents the interaction between the magnons and phonons at the interfaces forming a type of new material by spin and lattice interactions.

4 The ferroic Spin transistor model

The bulk inversion asymmetry (BIA), Rashba inversion asymmetry (RIA) effects has failed to fulfill the promise of a room-temperature spin FET. The major technology road blocks to implementation of room-temperature spin FET is the relativistic nature of BIA, RIA effects which is weakened at room temperature. This is due to the due to the sensitivity of these effects to high temperatures. The difficulties to inject the efficient spin polarized electrons in a 2-DEG pheterostructures channel have increased the challenges for implementation of the Rashba effect.

The solution to the spin injection and spin transport challenge lies in the control of spin polarization in a channel by spin-strain coupling which is stronger than RIA at room temperature being non-relativistic in nature. The magnetoeelctric effect ensures the control of spin exchange inetraction in a ferroic phase of the channel with the electric field activated polarization of the ferroelectric phase. The interface coupling of lattice and spin ensures the effective spin polarization by lattice-spin coupling. In RIA effect, the spins of the spin up and spin down electrons are split with the application of electric field as these fields are perceived as magnetic field in the electron's frame of reference. However, the variation in the crystalline field by lattice strains affects the spin exchange interaction between the nearest neighbors. The effect on the spin exchange in spin-strain coupling results in

the modulation of the exchange interaction by the variation in electronic orbital overlap due to the lattice displacements. There can be a perfect control on the lattice displacements by the applied fields from the gate terminal on the ferroelectric phase of the channel. **It** must be emphasized here that the spin exchange interaction can be regulated by the lattice displacements if the material is ferroelastic in nature.

5 Rashba effect, Zeeman Effect, spin exchange interaction and magnetoelectric effect

The effect of spin-orbit coupling is weaker than crystal filed splitting in the transition 3-d metals Fe [8] surrounded by oxygen atoms or other transition metals like Ti and alkaline Ba. However the the coulombic, spin-orbit cou- pling and zeeman effects [8] should always be considered as a part of the Hamiltonian representing the ferroic channel.The Hamiltonian representing the lattice displacements [8] must also be the term in the combined Hamilto- nian. The interaction between the ferroelectric and ferromagnetic materials will deviate the the crystalline field perturbed by the displacements of ions which may vary the crystalline field splitting between e_g and t_{2g}. The oxida- tion state of metal, the type of ligands and the type of coordination complex determines the position of 3-d orbitals which are 05 in number. During the displacements of ions of octahedral complex the spins in 3-d sub orbitals are changed because of the change in the position of the ligands. In this way the change in the spin occupation of 05 sub orbitals of 3-d Fe and their respective spins may be changed. According to crystalline field theory the field formed by the atoms around every Fe atom determines the number of spins in e_g and t_{2g} energy levels. The gap between the e_g and t_{2g} energy levels and the position of Fermi energy vs

the e_g or t_{2g} pumps the particular spin to the injector. The degeneracy of the spins is lifted by applied H field in the octahedralcomplex of ferrite when the spin is pumped from the spin spitted injector. The double exchange interaction couples the spin in the ferroic channel in a spin device. The considerable magnetoelectric effects have been seen in $FeBaTiO3$ and $CoFe2O4 - BaTiO3$, $NiFe2O4 - PZT$ bilayers and multilayers [2] which suggests that the coulombic, crystalline field splitting and spin-orbital effects in ferroics takes place along with the intra-atomic coulomb effects to result in magnetoelectric effects. In ME effect, the output electric field has been detected in the ferroeelctric part of the bilayer which suggests that the impacts of crystalline field variations in ferromagnetic side of the bilayer perturbs the ions from their equilibrium position in ferroelectric part.

In Rashba effect the spin orientation in a spin FET may be controlled by the application of gate voltage at low temperatures only as it suffers from the scattering of spins moving in the transistor channel. The magnetoelectric effect is proposed to control the spin orientation by application of the gate voltage. The hysteretic resistance varied by the applied voltage, *electro-resistance [*19], may be coupled with the spin exchange interaction effect in a ferroic. So, the spin exchange interaction may be coupled with the polarization of the ferroelectric phase. Whereas the spin-orbit coupling and crystal field splitting effects are also combined in strengthening this interaction. In a ferroic channel the gate effect is expected to be stronger due to the fact that the magnetoelectric effect (ME) is added with the spin orbit, crystalline field and zeeman effects. It is important to note that none of these effects are expected to contrast the magnetoelectric effect. The electric field in the gate terminal is efficiently coupled with the channel which

may flip the spin orientation as a function of the gate voltage. The ferromagnetic, ferroelctric and magnetoelectric components of the energy terms are solutions in the form of magnon, phonon and magnon-phonon interactions.

The variation in the crystalline field interaction by the displacement of ions may be expressed as the perturbation of the basic Hamiltonian where the perturbation potential is expanded in Taylor series with respect to the strains. These strains are initiated by the gate voltage which appears as ionic displacements from their mean equilibrium positions. The impact of the ionic displacements on the spin exchange is proved indirectly by the magnetoelectric effects in bilayers and multilayers of multi ferroic magnetoelectrics.

6. Working mechanism of three-terminal ferroic spin device

The application of the magnetic field lifts the degeneracy of electron energy may split the energy for spin up and down in the source, channel and detector regions. The s-p-d exchange interaction takes place in the interfaces and bulk regions of the $BaTiO_3 - BaFeO_3$ device. The crystalline field split-up in the transition metal oxides may be controlled by the application of gate voltage.

The electric field from the gate terminal displaces the ions from their mean positions in the channel. The displacement of ions varies the octahedral crystalline field and spin-orbit coupling effects. The crystalline field effect is more pronounced than spin-orbit coupling in transition metal oxides. The t_{2g} and e_g bands are shifted in such a way that the fermi energy inside the d orbital is shifted across the spins in e_g sub-orbitals $d^2, d^2 - y^2$ or t_{2g} sub zorbitals d_{xy}, d_{xz}, d_{yz}.

The spins split -up in the injector are are coupled with spins in the channel by double exchange interactions. The singlets or triplets may be injected to detector depending upon their proximity to Fermi energy due to the coupling between the spin exchange and electric field controlled crystalline field splitting. The zeeman, Rashba, spin-orbital coupling and the crystalline field effects are not as strong as the magentoelectric (ME) coupling. But the coupling voltage in 1mV - 1 V range is generated by 10-30 KV electric fields.

6.1 Exchange interaction

The *exchange interaction* is a quantum mechanical effect which increases or decreases the expectation values of the particular energy and distance between two distinguishable or indistinguishable particles when their wave functions overlap. So the exchange interaction for distinguishable identical particles push them farther apart as compared to indistinguisable identical particles as the latter has exchange symmetry. This being the case the spin exchange asymmetry holds two electrons with spin 1/2 farther apart as com- pared to two photons or phonons with their wave functions overlapping. The spin statistics theorem suggests that wave functions of spin 1 particles remain same after swapping and the wave function of spin 1/2 particles change sign after swapping.

Suppose $\psi_1(r_1)$and $\psi_2(r_2)$ represent the wave functions of two identical particles. If these particles interact with each other there can be total spin of 1or zero as two possible cases. The interaction for spin be- ing 1 will be $\psi_A(r_1, r_2) = \psi_1(r_1)\psi_2(r_2) - \psi_2(r_1)\psi_1(r_2)/(2)^{1/2}$and with to- tal spin being zero the interaction will be $\psi_S(r_1, r_2) = \psi_1(r_1)\psi + 2(r_2) - \psi_2(r_1)\psi_1(r_2)/(2)^{1/2}$. If the interaction energy between these expressed as $V_I(r_1, r_2)$ then the difference between the energies shown in

expression will be $J = 2\,\psi^*(r_1)\psi^*(r_2)V_{\boldsymbol{i}}(r_1, r_2)\psi_2(r_1)\psi_1(r_2)dr1dr2$. By taking into account the different spins of the interactions with spins being singlet or doublet we may add a new term in Hamiltonian which takes into account the spin-spin interaction $-J\,S_1S_2$. The term J being positive suggests the spins being alignedand J being negative stands for spins anti-parallel. In both cases the consequences of spin-spin interaction are magnetic due to electric repulsion and Pauli's exclusion principle. So the electric repulsion will be higher for two spin half particles with spins parallel depending upon the precondition that Pauli's exclusion principles are not violated. The *Double exchange interaction mechanism* in $Fe - BaTiO3$ is an important exchange interaction mechanism as the e_g orbitals of transition metal Fe exchange spins with the second nearest neighbor atoms of Fe, Ba and Ti via oxygen 2-p orbitals.

These p-d exchange interactions determine the ferromagnetic and antiferromagnetic properties depending upon the overlap between p-d orbital via O atoms. The double exchange is different from super exchange interaction which is between same atomic species having same valence electrons. In addition the super exchange is between two positive ions separated by a negative ion. The super exchange results in antiferromagnetic interactions unless the positive ions are located at 90 degrees with one another. While in su- per exchange interaction electrons actually hop between the positive ions via intermediate ligand oxygen. The references suggest that the $BaFeO_3$ and $Fe - BaTiO3$ exchange interactions result in ferromagnetic properties so these interactions are double exchange. But this can be further investigated

by detecting the orientation of spins in detector terminals. The Zeeman

ISSN 2229-5518
effect is defined as the splitting of electron energies due to the application of mag- netic field which were degenerate due to electronic configurations with elec- trons having same energies. The zeeman and spin-orbit coupling effects are related as shows the expression $H = H0 + Vm$. The $V_m = -\mu B$, $\mu = \mu_b gJ/\hbar$ and $gJ = (l + gS)$.

6.2 Rashba effect and exchange interaction

The selection rules precondition the transition of electrons between different orbitals. The spin-orbit interaction is the interaction between the spin and motion of the orbiting electron. $\delta H = -\mu B$, In electron's rest frame of reference the electric field appears as magnetic field, $B = v * E/c^2$, supposing $E = \boldsymbol{E/r}$ and $p = m_e * V$, So $B = m * vE/mc^2$, substituting $E = -gradV$, we get $E = \delta V/\boldsymbol{r}$ which is same as $1/e(\delta U/\boldsymbol{r})$. So the final expression is $B = (1/mc^2)(1/e)\delta U/\boldsymbol{r}(L.S)$. It must be clarified that the impact of the Rashba

effect to be present in the channel. The magnetoelectric effect control of the spin may be stronger effect than the Rashba or spin-orbit effect. Suppose the electric is applied in z is named as E_z. In the carrier frame of reference the electric field appears as magnetic field. The component of velocity parallel to interface is $v(x, y)$. The appearance of magnetic field $B = -[v(x, y)/c^2] * E$ tends to flip the spin of carrier. In terms of the Hamiltonian $H_R = \alpha k_x \sigma_x +$

$k_y \sigma_y$. The σ_x and σ_y are the spin matrices and α is a function of the potential barrier profile.

The applied field H can be related with ω the cyclotron resonance fre- quency as $\omega = eB/m$, where e is electron charge and m is mass of an electron, the mass m must be replaced by the effective mass m^* for the material and γ is a relativistic term defined as $\gamma = 1/[1 - (v/c)^2]^{1/2}$. The range of values

of frequency is in GHz and the B field is in 100 Gauss or .01 Tesla. The harmonic oscillators are defined by $^{2.}\ JS_{\boldsymbol{i}}S_{\boldsymbol{j}} + {}^{2.}\ hw(n + 1/2) + \delta$.

At interface the Electric field from gate terminal connected to ferroic material oscillates these harmonic oscillators.

The displaced atoms oscillate at $\hbar\omega(n + 1/2)$. The strains in the ferroic material couple the ferroelectric and ferromagentic phases represented by oscillators by the interaction part of the Hamiltonian. The columbic forces are coupled through these strains as the ions in ferroelectric part displace from their mean positions. The spin orientation is changed by the harmonic oscillators coupling as the d orbitals of the ferroic materials are positioned around the Fermi level and are controlled due to the displacement of transition metal atoms. The triplet and singlets spins which are split apart start transporting due to the degeneracy being lifted up with the application of the magnetic field present at the injector terminal.

7 surface interfaces modeling

The interfaces between two different ferroic materials are vital for the spin injection, spin transport control and detection. The surfaces of the hetero structures at the interfaces get characteristics from the surface energy states [19], chemical bonding and electrostatic dipoles which are significantly different from the bulk. The interface electronic structure will dominate the tunneling across the interface formed by the surfaces of the materials at in- terface. The interface states get their characteristics from the solutions of Schrodinger equation as the localized state. In a perfectly periodic bulk solid the Block theorem enforces the periodic part to the wave function solution being a plane wave e^{ikr} having complex wave vector with a real $k(r)$ and a function μ_k representing the periodicity of lattice. The solution of the Schrödinger equation for the bulk is $\varphi(r) = e^{ik(r)}\mu_k(r)$. At surface interfaces the symmetry is broken on both sides of the interface.

The solutions of the complex wave vector components perpendicular to the interfaces k_{perp} are possible.

The crystalline symmetry and electronic configuration of the surface of a bulk material is different. The lower symmetry and modified electronic configuration determines the properties of surface layers to be different form the bulk. The surface layers are normally undergoing a relaxation normal to the surface or lateral to the surface or both. In all cases the distances between the atoms are changed as compared with the bulk. The lattice constants between the surface layers and the bulk are shorter depending upon the type of the relaxation the surface layer has undergone. In case of the surface reconstruction, the surface layers are formed with different lattice geometry than the bulk. The adsorption of the atoms from another material at surface sufficiently alters the formation of the surfaces and it is different from the surfaces under vacuum. These relaxation, reconstruction and adsorption at surfaces alter the electronic properties of surfaces and interfaces depending upon the composition, crystalline symmetry, and ambient conditions during the formation of the particular material interface.

These evanescent waves are propagating on the surfaces of the constituent materials. The metal-insulator surface states are formed by the propagating metal waves from the metal side matching the evanescent states on the insulator side thereby penetrating the insulator and forming metal insulator gap states (MIGS) which is true for $Fe - BaTiO3$ surface interface. For two oxides surface interfaces formed by $NiFe2O4 - BaTiO_3$ or other materials, the evanescent states on both sides of surface interface decay away from the interface. Matching of the decaying states on both sides of interface can ensure an intrinsic surface state which decay uniformly on both sides of

interface. The hybridization of the surface states with bulk states may allow them to propagate far inside bulk. The surface states are special type of states where the evanescent states in the material matches the decaying states in the vac- uum. The interface states have large density of states which has important consequences for the band alignment and the position of Fermi energy in- side the materials at the interface. The surface dipoles are formed at the $Fe - BaTiO_3$ metal-semiconductor interface which influence the formation of schottky barriers.

In case of Fe doped $BaTiO3$ the space charge regions of equivalent widths are supposed to be formed and the junction is ohmic type. In both cases the spins may be injected but the matching of resistances of the two materials on each side of interface enhances the percentage of spin injection. In ferroics the issue of spin injection is avoided by spin exchange interaction on both sides of interface coupling spins in source with channel in Fe doped BaTiO3 materials. The chemical bonds formed between Fe and BaTiO3 may result in the formation of ionic or covalent bonds between itinerant Fe electrons with valence electrons in the dielectric BaTiO3 with high affinity. The charge transport may result in the dielectric $BaTiO_3$ from Fe itinerant electrons carrying spins with them along with the charge. The spin dependent electron tunneling has been demonstrated earlier and magnetic tunnel junctions (MTJ) incorporate the spin dependent tunneling of electrons in an oxide separated by ferromagnetic materials. The Tedrow, Meservy and Juliere later have presented models that explain the spin exchange interaction and intermediate layer properties. The thickness of the layers perpendicular to interfaces are a couple of nm for these interactions. The replacement of ordinary oxides with ferroelectric and ferroic materials like Fe doped BaTiO3 ensures that the spin exchange interacts from the ferromagnetic

source to ferroelectric or ferroic material. The hysteretic resistance of the ferroelectric material con- trolled by gate electric field ensures the electric and magnetic field control of the spins and dipoles in a ferroic channel. The spins injected in the chan- nel may be detected in the drain terminal without electron transport in the channel as the dipoles transfer the spin through exchange interaction to the drain terminal.

8 Ballistic regime and magnetoelectric effect

When the sizes of regions is equal to or less than the electron mean free path the material properties are changed from classical to ballistic regime. The recent advances in the fabrication have been successful in making thin films of few nano meters thickness perpendicular to the interface between different materials. In the ballistic transport regime the scattering is very low and the polarization of charges is a function of chemical potentials in left and right electrodes μ_l and μ_r . In Landaur-Buttiker formalism the current is a function of transmission probability, the Fermi-Dirac distribution functions summed over all energies with the condition that there are occupied and unoccupied states available at the source and drain terminals.

$$d/dt[q\delta] = 2e/\hbar \quad T(E)(f_L(E) - f_R(E))dE \qquad (34)$$

At the bias from a terminal

$$G = d/dt[q\delta]/V = (q\uparrow\delta)^2/\hbar T(E_f) - (q\downarrow\delta)^2/\hbar T(E_f) \quad (35)$$

Here $V_L = \mu_L - \mu_R$. The conductance in the channel is a function of trans- mission probability $T(E)$ and the conductance quantum $\hbar/(q\delta)^2\sigma$. The spin degeneracy has been replaced by the spin channels. In a layered hetero structure the thin layers of nano meters dimensions perpendicular to the interface plane undergo the conductance quantization. In layered regions formed be- tween the source and channel the periodicity is broken perpendicular to the interface. The periodicity in the plane parallel to the interface is not sufficiently broken. The transmission coefficient T is represented as a function of parallel and perpendicular wave vectors k_par and k_perp. It must be noted that the transversal wave vector across the interface is not conserved on both sides of interface for charge less or charge transport cases. So fabricating a defect free interface and model it with a realistic model that takes all defects into account is the best approach which is possible if a large super cell is considered or mean field techniques are applied.

In our project, the localized states on the both sides of the interface may contribute to the coupling by the electron-electron, electron-phonon and electron-magnon scattering. In a ferromagnetic-dielectric interface the spin degeneracy is lifted and the conductance becomes spin-dependent controlled by the material and fabrication characteristics of the interface. It must emphasized here that the Tedervow, Meservey, Stears and Juliere models of spin polarization are based on the spin exchange of the charge across a ferro- magnetic metal-insualtor interfaces and ferromagnetic metal-paramagnetic metal interfaces in ballistic regimes. In a metal-ferroelectric

interface and metal-ferroic interface the N^b, P^b, P^b, J^b *allistic*, J^b ***iff*** refers to the density of the bound charges per unit volume. These charges form the dipoles in a ferromagnetic or ferroelectric insulator and are sufficiently different from the metallic charges. Another important difference with traditional spin polarization model and a ferroic channel model is the transition metal doping of the ferroelectric $BaTiO_3$. The Fe doped BaTiO3 is a Dilute Magnetic Semiconductor (DMS) with an n type transition metal impurity. The spin polarization across the interfaces can be expressed by spin-dependent density of states (DOS). The DOS are split by exchange spin in the Fe-BaTiO3 interfaces and expressed as $N_{\uparrow}(E)$ and $N_{\downarrow}(E)$. The spin-dependent DOS is expressed by

$$Nb\sigma = 1/(2pi)^3 \quad dS^{\alpha\sigma}/v_{k\alpha\sigma} \qquad (36)$$

Where σ represents all the spin index being up or down and the integral is taken over all Fermi surface interface, $v_{\sigma\alpha}$ is the Fermi velocity at interface and α represents all bands over which sum is taken. The spin polarization of the DOS at Fermi energy is expressed as

$$P_{Nb} = N_{b\uparrow} - N_{b\downarrow} \div N_{b\uparrow} + N_{b\downarrow} \qquad (37)$$

This equation can expresses the spin split of the bound charges adequately in Fe-BaTiO3 heterostructures as the bound charges contain the magnetic properties. For doped Fe-BaTiO3 dielectric hetero structures the charge transport phenomenon is negligibly small for low Fe concentrations in $BaTiO_3$. The equation that expresses the spin polarized transport of charges

$$P_{Jb} = N_{Jb\uparrow} - N_{Jb\downarrow} \div N_{Jb\uparrow} + N_{Jb\downarrow} \qquad (38)$$

The dipole charge densities of spin up and spin down electrons act like two separate channels.

$\alpha\sigma$

b

In diffusive transport regime the Boltzmann equation with relaxation time approximation is

$$J^{d}iff_{\sigma} = \tau k\alpha\sigma\, v k\alpha\sigma\, dS^{\alpha\sigma} \quad (40)$$

The scattering time depends upon the type of scattering center, electronic configuration and the spin polarization dependency. All above equations provide different interpretations of spin polarization and there is no single definition of the spin polarization. The qualitative understanding of the spin dependent scattering, tunneling and the spin polarization can be expressed with the s-d hybridization states at interface, defects types, material, the thickness and the height of the potential barrier. The spin polarization across $FeBaTiO_3$ and Fe alloy $BaTiO_3$ interfaces will be due to the majority spin. According to Stearns the spin polarization is DOS based and depends upon the difference of the Fermi wave vectors of the majority and minority spin itinerant electrons.

$$Pk = kspinup - kspindown/kspinup + kspindown \quad (41)$$

According to Slonczewski, the spin polarization depends upon the potential barrier as well as ferromagnetic source and detector materials

$$Pb,tunn = \kappa^2 - kspinup kspindown/\kappa^2 + kspinup kspindown \quad (42)$$

When the barrier height is large and condition $\kappa >> k_spinup, k_spindown$, we get the original expression for the $P_{b,k}$. And the spin polarization can also be interpreted as

$$Pb,tunn = Tb,spinup - Tb,spindown/Tb,spinup + Tb,spindown \quad (43)$$

The definition of T is $4\kappa k_sigma/\kappa^2{}_s + k^2 igma$ and it is the spin dependent transmission probability from source to the channel in a ferroic

FET. If we can superimpose the transmission coefficients of the spins of the bound charges on two sides of the interface as

$$T(k\|)\sigma = T_L(k\|)\sigma e^{\kappa d} T_R(k\|)\sigma$$

Here d expresses the barrier thickness. This equation ignores the scattering and is valid for the large barrier height. The interface conditions are not explicitly included in this expression. For the channel being a ferroelectric material and the source being the ferromagnetic material the interface properties can be modulated by the applied electric field at the gate terminal. The resistance of the channel region is varied with the gate field thereby producing the giant Electro resistance effect (GER), which is the difference in the conductance of the channel for the polarization pointing in the left and right direction. The interface transmission probability for the spin can be controlled with the variation of interface electrostatic potential, variation in bonding strength and strain associated with the piezoelectric response.

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