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## C O N T E N T S

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## **Review on the Electromagnetic Mechanism behind the Phenomena of Surface Enhanced Raman Scattering (SERS)**

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**Abstract :** Objective of the review article is to assist the understanding and identification of the electromagnetic mechanism involved in the SERS phenomenon. Electromagnetic (EM) contribution to surface enhancement is though well recognized but charge transfer (CT) contribution to SERS is less clearly understood as yet. It is usually believed that two enhancement mechanisms, one a long-range electromagnetic (EM) effect and the other a short-range chemical (CHEM) effect, are simultaneously operative. The EM mechanism is established on the amplified electromagnetic field produced by optical excitation of surface plasmon resonance of nano-scale surface roughness. When the molecules are adsorbed by the nanocolloidal SERS active metal surface they create hot spots. At the position of the hot spots there will be a huge enhancement of the electromagnetic fields causing amplification of the Raman signal and this enormous application is used in the recent development of science and technology in the fields of physics and chemistry. The various types of EM mechanisms and its recent development are discussed here with detailed theoretical explanations.

**Keywords:** Surface plasmon, Polarizability, Electromagnetic mechanism, Raman bands, Hot spots.

## 1. *Introduction*

The applications of conventional Raman spectroscopy are limited by the extremely small cross section of the Raman scattering process, which are ~ 12-14 orders of magnitude less than the fluorescence cross section. In 1974, a discovery by Fleischmann and coworkers<sup>1</sup>, which showed unexpectedly high Raman signals from pyridine molecules on rough silver electrode, attracted considerable attention<sup>2-4</sup> to the investigators in this field. In this paper<sup>1</sup> Fleischmann et.al. described an unexpected million-fold enhancement of Raman signal from pyridine molecules when they are allowed to adsorb onto electrochemically roughened surface of the silver electrode. In early 1976, R. Van Duyne and D. Jeanmaire observed the result<sup>3, 4</sup> and in 1977, M. G. Albrecht and J. A. Creighton reported similar observation<sup>5</sup>. This surprising discovery touched off a flurry of theoretical and experimental activities. Experiments in different laboratories gave evidence that the enormously strong Raman signals were caused by true enhancement of the Raman scattering efficiency itself and not by more scattering molecules<sup>1-3</sup>. Within the next few years, strongly enhanced Raman signals were reported for various probe molecules<sup>3</sup> when attached to “rough” coinage metal surfaces (mostly on roughened Ag, Cu, Au electrodes and colloids). The effect was named “**Surface-Enhanced Raman Scattering (SERS)**” by Professor Richard P Van Duyne. Later, the advent of scanning probe microscopic (SPM) technique revealed that the order of “surface roughness” is in the nano dimension, which now intimately relates SERS with Nano Sciences<sup>6</sup>. The discovery paved the way to overcome the traditionally low sensitivity problem in Raman Spectroscopy. SERS spectroscopy has now emerged as a powerful technique not only for studying the molecules or ions at trace concentrations down to single molecule detection level<sup>7</sup>, but also helps to understand the surface chemistry



at the nano scale<sup>8</sup>. The discovery of SERS has produced significant impact in fields such as surface chemistry, solid state physics, inorganic chemistry of metals, electrochemistry etc.. Not only so, this has widened many other fields of investigation such as classical electrostatics and electromagnetic theory as applied to small metal particles, problems related to radiating multipoles near metal surfaces, optics of small particles, generation of surface plasmons, surface-photon interaction and second harmonic generation from molecules and surfaces.

## ***2. Understanding of the Electromagnetic mechanism involved in the SERS phenomenon***

Despite intensive theoretical works<sup>9</sup> published in different special issues of scientific journals themed to this phenomenon<sup>10</sup> and publications of excellent reviews<sup>11</sup>, the exact nature of the colossal enhancement in Raman intensity found in SERS is still a matter of controversy. However, it is generally accepted that two enhancement mechanisms, one a long-range electromagnetic (EM) effect and the other a short-range chemical (CHEM) effect, are operative simultaneously. Here only the EM mechanism has been discussed.

### **2.1 Electromagnetic Mechanism**

The electromagnetic mechanism of SERS is now being explained in terms of localized surface plasmon resonance (LSPR) or more specifically, dipolar particle plasmon resonance (DPPR)<sup>12-15</sup> that arises around the plasmonic nanoparticles and are strongly localized at the interstitial gaps within two or more nanoparticle aggregates. The LSPR or DPPR result in huge localizations of electric fields around the probe molecule – plasmonic nanoparticle.

Electric fields of the incident light wave of wavelength  $\lambda$  induce polarizations of the conduction electrons with respect to heavy ionic core of the nanoparticles. Within the quasistatic approximation for  $2R \ll \lambda$ , where  $2R$  is the diameter of the nanoparticle, the electric field polarizes the surface of the nanoparticle uniformly. Under these circumstances, the electric field of light on the spherical nanoparticle can be taken to be constant, and the interaction is governed by rules of electrostatics, rather than electrodynamics. The displacements of the electrons with respect to heavy ionic core due to polarization give rise to Coulomb restoring forces. As a consequence, a dipolar collective oscillation of electrons is created, and is known as surface plasmon oscillation. These collective oscillations of electrons are also referred to as “dipolar particle plasmon oscillation” to differ it from plasmon excitations that occur in bulk metal surfaces. The quantum of energy of such oscillations is called plasmon. If the oscillation frequency of the electrons matches with the frequency of incident laser radiation subject to the fulfillment of dispersion relations, localized surface plasmon resonance (LSPR) or dipolar particle plasmon resonance (DPPR) occurs that result in gigantic enhancement of electric fields around the plasmonic nanoparticles. Various types of electromagnetic mechanism along with its recent developments in the phenomena of SERS have been discussed here.

### **2.1.1 Isolated metal particles**

Isolated metal particle (IMP) model assumes the excitation of a single metal sphere by laser radiation. The diameter ( $2R$ ) of the sphere is small compared to the wavelength of incident light. Raman scattering takes place from the reporter molecules that are adsorbed on the surface of the metal nanocolloid. The metal sphere of dielectric constant  $\epsilon_i$  is surrounded

by the medium of dielectric constant  $\epsilon_0$ .

If the direction of electric field of the incident electromagnetic wave  $\vec{E}_0$  is assumed to be along the z-axis, Maxwell's equations can be approximated by Laplace's equations to determine the electric fields both inside and outside the sphere<sup>16</sup>.

In Raman scattering the applied field induces an oscillating dipole in the adsorbed molecule. This dipole then radiates, and the component of this radiation that has been shifted by the vibrational frequencies of the molecule determines the Raman scattering intensity. Electromagnetic enhancements are generally small for spherical particles because the surface plasmons for most materials occur at frequencies far above the visible region. Larger enhancements arise for prolate spheroidal objects, as the plasmon resonances for such objects occur at frequencies that are red-shifted relative to spherical particles to frequencies in the visible region where the plasmon width is sufficiently small. Therefore in the small particle limit, SERS excitation and extinction plasmon peaks and widths should be the same.

### 2.1.2 Collective Resonances

In most of the real systems, the particles are extremely close so that the coupling of single particle resonances may be considered. The classical surface-plasmon-type enhancement calculations consider the collective interactions explicitly between the resonating structures. Moskovits<sup>17</sup> considered collective resonances of bumps on a metal surface and the dependence of the resonance frequency on the bump density.

The nature of interaction is long range because the excited field at resonance is created at distant far from the particle. For weak coupling system the inter particle distances are very large and frequency shifts occur,

but for the strong coupling case, completely new resonances occur. A huge field is created, in such case, between the two successive particles and the strong local field of one particle polarizes the other<sup>18-20</sup>. A molecule situated at this place is thus envisaged to show large enhancement of spectroscopic signals. In an early literature Arvind et al<sup>18</sup> showed that for a silver grating of period  $d = 1000 \text{ \AA}$  and  $l = 5145 \text{ \AA}$  a resonance of the electric field strength occurs within the channel, strength of which increases with decrease in channel width and the resonance frequency shifts to the blue with decreasing pore height. The substantial property of interaction between a spheres and a sphere-plane structure is illustrated by Arvind and Metiu<sup>21</sup> clearly in their literature. If the distance between the sphere and the plane is large then the resonance will occur by exciting light for  $n = 1$  dipolar resonance of the sphere. According to momentum conservation, light cannot excite the surface plasmon resonance of the flat surface but the dipole induced in the sphere can excite this resonance successfully. When the sphere is brought near to the faces of the surface the interaction between the sphere and its own image takes place and this leads to  $n > 1$  resonance of the sphere. This resonance interacts with the flat surface plasmon and thus excited further. The effect of local field will be strong between the sphere and the plane surface if the distance between the sphere and the plane is very small. For two spheres very close to each other, similar interaction takes place. The field strength at different points in between and outside one-, two- and four-sphere assemblies are discussed in SERS system by Liver at al<sup>22</sup> and Arvind et al<sup>18</sup>. They observed that at a point in between two spheres the field is greater than that of a single sphere or at a point outside the spherical systems by more than an order of magnitude. This suggests that for a molecule located at interstitial points the SERS intensity enhancement is greater by four orders of magnitude. For aggregated nanocolloids they observe a red

shift of frequency at maximum enhancement. For the two- sphere system the field strength is greater than that for the isolated sphere and this explains the large SERS signal from slightly aggregated colloids. Similar types of conclusions done by Inoue and Ohtaka<sup>23</sup> by considering only dipolar coupling between metal particles. For totally symmetric Raman active vibrations, this two-sphere cluster model has also been used to account for the depolarization which is commonly found in SERS<sup>24</sup>. The relationship between the absorption spectrum and the SERS excitation profile for metal colloids or island films may be derived from the boundary condition relationship between the fields inside and outside the metal particles<sup>25</sup>. It is observed that, in the molecules adsorbed on roughened surface<sup>26</sup> and on metal nano colloids<sup>27</sup>, the Raman bands of the absolutely symmetric vibrations are particularly less polarized compared to that for the free molecules. The depolarization ratio for a particular mode of vibration is a function of the relative magnitude of the field components at the surface and the derived polarizability tensor component. Creighton<sup>28</sup> recommended that the depolarization of the Raman scattered light from molecules adsorbed on coagulated metal sols, is due to local electromagnetic anisotropy of the individual spheres within the aggregates. This idea has been supported by the observation of Jiang et al<sup>24</sup>. Theoretical results based on the above-mentioned electromagnetic models are in poor agreement with the experimentally observed depolarization ratios for aggregated colloids and for other SERS active surfaces (the Mie scattering from the aggregated colloids is also found to be depolarized). Ultimately we should get the same results with normal modes of the molecule, for the difference between the two descriptions is a simple change of basis for the representation of the movements. The advantage of the internal coordinates is that it gives a description based on localized movements of groups of atoms, which are

easier to identify spectroscopically in practical situations, either by Raman or IR spectroscopy.

### 2.1.3 Quadrupole polarizability mechanism

When a SERS active surface is irradiated close to the plasmon resonance frequency, the field is enhanced and provides an optical frequency field gradient. These surface field gradients, for a small sphere, exist adjacent to an imaginary point dipole at the centre of the sphere and are huge for illumination at plasmon resonance frequency. The field is strongly localized between two closed spheres where the surface field gradient becomes larger and contributes significantly to SERS intensity. Sass *et al*<sup>29</sup> recommended that these optical frequency field gradients have substantial involvement to the induced molecular dipoles at the surface via the molecular quadrupole polarizability. In the SERS spectra of benzene<sup>30, 31</sup> and ethylene<sup>32, 33</sup>, selective enhancements of some Raman bands occur which are Raman inactive in Free State of the said molecules. Reduction of symmetry of the molecule in the surface adsorbed state can apparently justify the presence of such bands. However, Moskovits and DiLella<sup>31</sup> interrogated about this idea. Maximum SERS spectra comprehend Raman forbidden bands<sup>31, 34, 35</sup> with minor frequency shift but huge change in SERS intensity. If the theory of symmetry reduction holds, then slight frequency shift indicates small perturbation but the large intensity of the forbidden modes recommends a very large perturbation of the molecule by the surface. In terms of the influence of the surface field gradients in inducing dipole moment through quadrupole polarizability of the molecule, the appearance of the Raman forbidden bands can be elucidated more strongly. It is further supported by the observation that dipole forbidden but quadrupole allowed electronic transitions are present in the fluorescence spectra of some polyenes at SERS active silver surfaces<sup>36</sup>.

#### 2.1.4 Molecule in a trap: Multipolar treatment

In the experiment of Raman scattering, the excitation frequency is generally far less than the surface plasmon resonance of an isolated metallic nano sphere which occurs at  $\frac{\omega_p}{\sqrt{3}}$ <sup>37</sup>. The field becomes largest in the vicinity of the resonance frequency. A small red shift will occur by the coupling of the metal nanocolloid with the active surface of the molecule which produces high fields close to the former and therefore an enhanced Raman signal from the molecule. When two spheres come very close to each other the resonance frequency decreases and produces a high field. If the molecule is trapped in a narrow space between the two curved surfaces of the spherical curvature of the metallic nanocolloid there will be huge enhancement of the field strength which gives large Raman signal<sup>38-42</sup>. So we can say that this type of molecular trapping is one of the main causes for the enhancement of the enhancement of the Raman bands.

When the edge-to-edge separation between two spheres of the same size is smaller than their radius, the dipolar approximation fails and the excitation of higher orders multipoles must be taken into account<sup>56</sup>. Complete multipolar theory for the Raman response of molecules when place in between two spheres was proposed for the first time by Rojas<sup>43</sup>. The red shift of the plasmon resonance by the multipolar terms is enough to place it in the optical region.

#### 2.1.5 SERS on Plane surfaces: Image field theory

Enhancement due to electromagnetic interaction between a molecule and flat surface were explained by the group of researchers as reported elsewhere<sup>44-47</sup>. In a model projected by King et al<sup>46</sup>, the electric field  $\vec{E}$  of the radiation induces an oscillating dipole moment in the adsorbate molecule which induces an image dipole in the metal.

### 2.1.6 Recent Development on the electromagnetic mechanism of SERS

The near-field and far-field spectral response of plasmonic systems are often assumed to be identical, due to the lack of methods that can directly compare and correlate both responses under similar environmental conditions. Recently a widely-tuneable optical technique is developed to probe the near-field resonances within individual plasmonic nanostructures that can be directly compared to the corresponding far-field response. Though the investigations in this area of research are limited, however Van Duyne et. al. worked on SERS of Raman reporter molecules adsorbed on periodically ordered substrates prepared from nanosphere lithographic (NSL) technique<sup>48, 49</sup>. In each of the observations, the best enhancement of Raman signals were accomplished upon laser excitation on the blue edge of the LSPR band in contrast to the incident laser radiation excited right on the LSPR peak. The problem of the choice of excitation wavelength that provides best enhancement of SERS signals was resolved to be on the blue edge of the LSPR band, approximately separated by one half of the vibrational energy mode<sup>50</sup>. However, the recognition of this conjecture as a general rule of thumb for fixing the right laser to attain maximum SERS enhancement was questioned once more while understanding the cases of SM SERS. The relation between the Near-field LSPR maximum and the corresponding Far-field SERS spectra again turns out to be askew with few recent reports which suggest the importance of exciting the probe molecule – nanoparticles system with low energy excitation to achieve maximum enhancement of SERS bands<sup>51-55</sup>. Recent development on the electromagnetic mechanism of SERS incorporates dipole re-radiation (DR) theory and plane wave (PW) approximation methods to estimate the enhancement factor (EF) of SER bands.



Collective resonances at a given wavelength can be arbitrarily classified as having more of a bulk-like or surface-like character, depending on the spatial distribution of the intensity. A bulk-like resonance can have a large contribution to the absorption (proportional to the volume), but may have a relatively weaker field at the surface in a specific point. The opposite can happen for a surface-like resonance. One could argue that there might be a slight compensation effect for large surface-like resonances in the sense that the smaller over all penetration depth contributing to the absorption might be compensated by a much larger field. But in general this compensation is only partial<sup>56, 57</sup>.

Hotspots have been generated in assemblies of single nano-objects, such as nanospheres or nanorods suspended in a homogeneous medium. These hotspots exhibit moderate SERS activity. However, assemblies of rationally designed single nanoparticles with nanoshells, sharp corners, ridges, grooves, tips and/or with intra-particle gaps, such as Au and Ag nanocubes, nanobars, nanostars, triangular nanoprisms, cauliflower-like and other multi-branched nanostructures, and mesocages exhibit much more intense SERS activity<sup>58-60</sup>. SERS hotspots arise also from coupled nanostructures with controllable inter-particle nanogaps. Typical examples are the Au or Ag NP dimers that were used for single-molecule SERS<sup>61, 62</sup> and core@shell nanoparticle dimers<sup>60, 63</sup>, nanoparticle aggregates or oligomers<sup>62</sup>, nanoparticle arrays such as core-satellite structures<sup>64</sup>, nanoparticle assemblies with controllable nanogaps<sup>65</sup> or inter-unit nanogaps in nanopatterned surfaces. These hotspots exhibit excellent SERS activity. Typically, the average SERS intensities from coupled plasmonic nanostructures are four orders of magnitude greater than those from single nanostructures<sup>57</sup>.

The local EM field in the gap between Au or Ag nanoparticle dimers and oligomers with inter-particle nanogaps is very intense due to the strong EM coupling. The extinction spectra of the Au or Ag dimer and the SERS EFs in the nanogap of Ag or Au oligomers depend critically on the gap size. For example, reducing the gap size in an Au nanosphere dimer from 10 to 2 nm, increases the SERS enhancement factor (EF) from  $10^5$  to  $10^9$ . Schatz and co-workers studied the gap-size dependence of the field enhancement factor using both finite element method (FEM) calculations and a semi-analytical model, and showed that for small gaps down to 1 nm, the local field enhancement confined in the gap arises from the waveguide mode, and depends on the local field strength, as the simple antenna theory predicts<sup>66</sup>. A small gap is preferable for single-molecule SERS; however, the local EM field and SERS EF cannot be increased without limit by decreasing the gap size to the sub-nanometre scale. Crozier and co-workers reported that the maximum SERS EFs measured by wavelength-scanned SERS is reached when the gap-width of a nanodisk dimer is decreased to  $0.6 \pm 0.1$  nm. This was attributed to quantum electron tunneling effects between the coupled NPs<sup>[67]</sup>. Aizpurua and co-workers were able to reproduce the experimental observations using a quantum-corrected classic model (QCM) to calculate the local field enhancement<sup>68</sup>. In this model, the junction is modulated by a local dielectric function that includes electron tunnelling and the tunnelling resistivity of the gap region. This is achieved by modifying the Drude dielectric function in the gap region.

For the surface analysis of materials by SERS, it is necessary to take account of the EM coupling effect of the probe materials and the SERS-active nanostructures. These refer to hotspots generated from hybrid structures consisting of plasmonic nanostructures and other materials<sup>48</sup>. These hotspots result from the hybridization of the EM field scattered from

the plasmonic nanoparticles and an EM field reflected from the substrate material surfaces. The resulting SERS EF depends crucially on the dielectric properties of the substrate materials. Several groups have reported SERS results obtained from molecules interacting with a single particle coupled to a metal film<sup>69,70</sup>. Ciraci and coworkers found that for a Au nanosphere on a flat Au film with a nanogap illuminated by a plane wave incident at  $75^\circ$  from the normal, the propagating surface plasmon was excited in addition to an enhanced local field in the particle–substrate nanogap. Furthermore, nonlocal effects affect the results significantly when the particle film gap is reduced to 5 nm, and even become the major contributors when the gap is less than 1 nm<sup>71</sup>. Xia and coworkers experimentally demonstrated that hotspots with SERS enhancements sufficient for single-molecule detection could be produced by depositing Ag nanocubes on a Au or Ag substrate<sup>72</sup>. Specifically, when a Ag nanocube was placed on a Au or Ag substrate, hotspots were created at the corner sites of the nanocube that were in contact with the metal substrate.

When a bare Au nanosphere dimer is placed on a Si or Pt surface, hotspots can be simultaneously produced at the particle–substrate and inter-particle nanogaps at certain incident wavelengths. For dielectric materials, the average SERS EF at the surface in the presence of an Au, as was found for the dimers of nanocubes or nanocylinders, the local field in the gap region can be significantly enhanced by plasmon coupling waveguide cavity modes to dipolar antenna modes. Aizpurua et al demonstrated that the antenna mode could hybridize with symmetry-allowed waveguide modes<sup>[73]</sup>. Baumberg and coworkers systematically compared the two fundamentally different resonant gap modes: transverse waveguide (s) and antenna modes (l), which, although both are able to strongly confine light in

gaps, have very different near-field and far-field radiation distributions. They showed experimentally and theoretically, that changing the nanoparticle shape from a sphere to cube and varying the gap size alters the coupling of s and l modes, resulting in strongly hybridized (j) modes<sup>74</sup>.

Schatz et al investigated the correlation between dipole re-radiation and  $|E|^4$  for a single nano sphere and its dimer<sup>75</sup>. They found that for small nanospheres, when the detection direction is perpendicular to the incident direction and polarization, the dipole re-radiation will be approximately equal to what is obtained using  $|E|^4$ . For large nanospheres, however, differences between the two approaches arise primarily due to multimode excitation. Moreover, for molecules positioned at some locations and for some detection directions, the two approaches can produce significantly different results. For the backscattered signal from nanosphere arrays, they found that the dipole re-radiation and  $|E|^4$  results were very similar in the long wavelength limit; however, some deviation occurred at short wavelengths and for observer positions other than backscattered<sup>76</sup>.

In shell-isolated nanoparticle-enhanced Raman spectroscopy (SHINERS), hotspots are produced by placing core-shell NPs composed of plasmonic Au or Ag cores with ultrathin (1–5 nm) chemically and electrically inert shells composed of, for example, SiO<sub>2</sub>, or Al<sub>2</sub>O<sub>3</sub> as probes on a surface. The advantages of SHINERS are two-fold: (1) the ultrathin but pinhole free shells separate the cores from the material surface (and environment), thus ensuring that there is almost no chemical interference from the Au and Ag cores; (2) the chemically inert shell effectively avoids inter-particle and particle-metal substrate fusion, which significantly improves the stability of the NPs and the probe structures. SHINERS have been used for the in situ characterization of chemisorption and electro-

catalytic reactions on various transition-metal single crystal surfaces that are difficult to probe using Raman spectroscopy<sup>77, 78</sup>.

Recently, many researchers have reported that plasmonic nanostructures with Fano resonances arise from spectral interferences between broad super radiant modes, such as a dipolar antenna mode, and sharp sub-radiant modes, such as the high order modes and photonic crystal modes. The interferences can produce narrow and asymmetric extinction and/or scattering features and a dramatic local field enhancement. Such features have made many groups use or propose to use plasmonic Fano resonance in applications such as sensing, lasing, switching and surface-enhanced spectroscopies, such as SERS and surface enhanced infrared spectroscopy. Comprehensive descriptions of the fundamental Fano resonance theory and its progress in plasmonic nanostructures can be found in recent reviews<sup>[79]</sup>. Recently, SERS of probe molecules adsorbed on nonplasmonic nanostructured substrates have been reported. Across the interface, the tangential component of the electric field is continuous, which gives the existence of surface waves. As no energy flows across the interface of the dielectric, surface waves can be exploited for investigating the surfaces with minimal perturbation of the probe molecule. John R. Lombardi et al<sup>80</sup>, in an extensive review recently reported that the dielectric nanoparticle can be used as a new frontier for SERS. It can serve as either a complement or an alternative to conventional metal-based SERS active substrate. J. Cambiasso et al<sup>81</sup> suggested that the strongest enhancement factors will be achieved for dielectric non-plasmonic SERS with the materials of high-refractive-index. Dielectrics must exhibit a higher refractive index than the surrounding media (including the analyte), to meet conditions for total internal reflection above the critical angle and propagate

the resulting progressive wave through the adsorbed analytes. For large difference in refractive-index between dielectric medium and analyte, the critical angle becomes small, and the penetration depth can be strongly reduced. At the same time, the materials should be optically transparent under the conditions of measurements, exhibit a featureless or clearly identifiable Raman spectrum to avoid overlapping with the spectrum of the analyte, and not be fluorescent.

### ***3. Conclusions***

Different electromagnetism mechanisms involved in the phenomena of SERS have been explained here. Huge enhancement of the Raman signals are very much responsible for the photo induced optical excitation imposed on the adsorptive sites of the metal-molecule interface. Isolated metal particle model accepts the excitation of a single metal sphere by laser radiation. For collective resonance, when the molecule is considered as a whole, we should get the same results with normal modes, for the difference between the two descriptions is a simple change of basis for the representation of the movements. In Quadrupole polarizability mechanism the optical frequency field gradients have significant involvement to the induced molecular dipoles at the surface of the metal-molecule interface. When the edge-to-edge separation between two spheres is smaller than their individual radius, the dipolar approximation fails and the higher order multipolar treatment must be taken into account. Image field theory is applicable on the flat surface where the electromagnetic field induces the oscillating dipole moment of the adsorbate. The recent developments of electromagnetic enhancement in the phenomena of SERS have been discussed here elaborately.

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## **Quantum Optical Frequency Encoded Oscillator Using Pauli Y gate and EDFA**

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[**Abstract:** Oscillators are essential and effective components of any digital processing system. Photons have already proven their ability in fastest computing. Even in quantum computing it can take important role to ensure fastest as well as secured computation. The author here proposes a new photonic scheme for developing a quantum optical oscillator using Pauli Y gate utilizing frequency encoding technique.]

**Keywords:** Quantum logic gates, Frequency encoding, Oscillator, Reversibility

### ***1. Introduction***

Quantum computing or quantum information processing has become very meaningful term nowadays. Several quantum logic gates had already been proposed and implemented by different researchers using quantum properties of light<sup>1-6</sup>. Exploiting the nonlinear properties of Pockels materials, Semiconductor optical amplifiers, photonic crystals etc. several optical devices and photonic logic gates had been developed<sup>7-13</sup>.

Light has become an important candidate in Quantum information processing. Unlike bits a quantum computer uses qubits or quantum bits as their basic building blocks. Qubits are superpositions of pure states  $|0\rangle$  and  $|1\rangle$  with certain probability. A general qubits state can be represented (Dirac notation) as  $|\Psi\rangle = C_0 |0\rangle + C_1 |1\rangle$ ; where the orthogonal pure states are superposed with probabilities  $|C_0|^2$  and  $|C_1|^2$  such that  $|C_0|^2 + |C_1|^2 = 1$ . In matrix notation orthogonal basis states are represented as  $|0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$  and  $|1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ ; consequently the general quantum state of qubit takes the form  $|\Psi\rangle = \begin{pmatrix} C_0 \\ C_1 \end{pmatrix}$ . Processing of quantum information is handled by different quantum logic gates. Among various single qubits gates Pauli gates are the most fundamental one. When a quantum gate operates on such a qubit state  $|\Psi\rangle$ , a new quantum state  $|\Psi'\rangle$  will be formed. A quantum gate is represented by a unitary  $2^n \times 2^n$  matrix depending on the number of qubits (n) required for its logical operation. Reversibility is the inherent prime feature of quantum gates which promises unique correspondence between inputs and outputs. Pauli Y gate has immense usefulness in the computation domain. Single qubit quantum Pauli Y gate is represented by a  $2 \times 2$  matrix of the form  $\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$ . The Pauli Y gate changes the single qubit state  $\begin{pmatrix} C_0 \\ C_1 \end{pmatrix}$  to  $\begin{pmatrix} -iC_1 \\ iC_0 \end{pmatrix}$  following the matrix equation:

$$Yq = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} C_0 \\ C_1 \end{pmatrix} = \begin{pmatrix} -iC_1 \\ iC_0 \end{pmatrix}.$$

Pauli Y gate had been implemented by these authors using polarization encoding and frequency encoding technique<sup>14,15</sup>. The authors in this paper propose a new scheme for development of a quantum

oscillator based on frequency encoded quantum Pauli Y gate and EDFA (Erbium Doped Fibre Amplifier). EDFA is a very successful optical amplifier having wide practical applications in several domain of optical processing and communication systems, testing devices etc. <sup>[16-18]</sup>. For implementation of this scheme two frequencies  $\nu_1$  and  $\nu_2$  have been considered to form a quantum state as  $|\Psi\rangle = \begin{pmatrix} \nu_1 \\ \nu_2 \end{pmatrix}$ . This quantum oscillator will be very effective in quantum information processing owing to superfast operational speed over terahertz limit.

## ***2. Frequency encoding***

Encoding/decoding is an important part for implementation of every all-optical device. Conventional encoding processes are frequency encoding, phase encoding, spatial encoding, intensity encoding, polarization encoding etc. Frequency encoding is the most rudimentary one, used massively in all-optical computing as it is associated with least bit-error rate and equipped with all inherent beauties of photons such as fastest speed, least noise, highest parallelism etc. Authors have exploited/utilised this encoding/decoding technique in quantum computing as the frequency is a quantum phenomenon of light. Here two frequencies  $\nu_1$  and  $\nu_2$  for two different light rays serve as the initial qubit state as  $\begin{pmatrix} \nu_1 \\ \nu_2 \end{pmatrix}$ .

## ***3. EDFA***

Erbium doped optical fibre amplifier is an established faithful amplifier in long haul, multichannel digital and analogue applications and its amplification can lead to develop all-optical switching for optical

information and data processing systems. Since the information carrying signal is directly amplified in the optical domain without conversion to the electrical domain, so the system is bit rate transparent, have large gain band width ( $\sim 40$  nm), low insertion loss, low noise figure etc. The amplification is dominated by stimulated emission of radiation of erbium ions ( $\text{Er}^{3+}$ ) in host silica fibre. The signal interacts with the doping ions in presence of a pump laser beam. The signal beam which is to be amplified and a pump beam are coupled into the erbium doped fibre and after an optimum length of the fibre; the pump power is abruptly attenuated while the signal power is enhanced strongly. Usually a pump beam (obtained from semiconductor lasers) at a wavelength of 980nm or 1480nm along with a signal beam in the wavelength region 1530-1570 nm are coupled into the erbium doped silica fibre. The amplified signal passes through an optical isolator and a wavelength filter, the filtrate being the signal beam alone.

#### ***4. Electro-optic modulator for phase balancing***

Potassium di-hydrogen phosphate (KDP), lithium niobate ( $\text{LiNbO}_3$ ), ADP etc. are some most widely used electro-optic Pockels materials. These crystals when excited by an electric field can modulate the phase or intensity of an incident laser light. Under the influence of external electric field applied to these crystals, the crystals become electrically birefringent that is refractive index ( $n_i$ ) of the crystals changes with the applied electric field. This electro-optic effect has wide range of applications in optical switching, amplitude, and phase or in frequency modulation of light, etc. In a typical electro-optic amplitude modulator the electro-optic crystal is placed in between two crossed polarizers. The

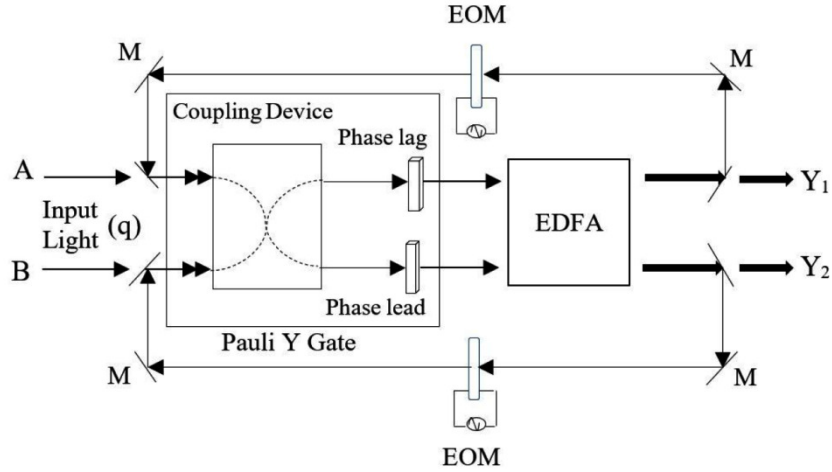


two orthogonal components of the incident radiation suffer different refractive indices along two orthogonal directions due to application of external potential which in turn creates an electrical field induced birefringence proportional to the applied electric field.

### ***5. The proposed scheme***

Proposed oscillator utilizes Pauli Y gate, optical amplifier and electro-optic phase controller for its construction. The output qubit of Pauli Y gate is initially passed through an optical amplifier which amplifies the optical signals coming from Pauli Y gate to desired level. Either semiconductor optical amplifier or fibre optic amplifier like EDFA will serve as amplifying tool for the optical signals. In the proposed scheme EDFAs have been used in this role. As discussed in the earlier section the optical signal while propagating through this fibre amplifier gets amplified and the amplified signal then passed through an electro-optic phase modulator. Finally, these modulator outputs serve as the inputs of the Pauli Y gate with a feedback mechanism in such a way that  $Y_1$  output merges with the input terminal A and  $Y_2$  with other input terminal B. During feedback, the optical signals suffer from different unavoidable foreseeable hazards like signal attenuation, change in phase of the optical signal etc. To compensate for the signal attenuation loss, EDFA is used in the pathway of both signals. On the other hand, during feedback through air both signals may suffer change in phase. To minimise this effect electro-optic modulators are used in the respective paths which balance the change in phase suffered by both signals during journey through proper adjustment of biasing voltage. The optical

scheme for implementation of the quantum oscillator is shown in figure 1.



**Fig.1**

A schematic diagram of developing optical quantum oscillator. (Here A and B are input terminals, EOM's are the electro-optic modulators, M's are the mirrors,  $Y_1$  and  $Y_2$  are the output terminals respectively.)

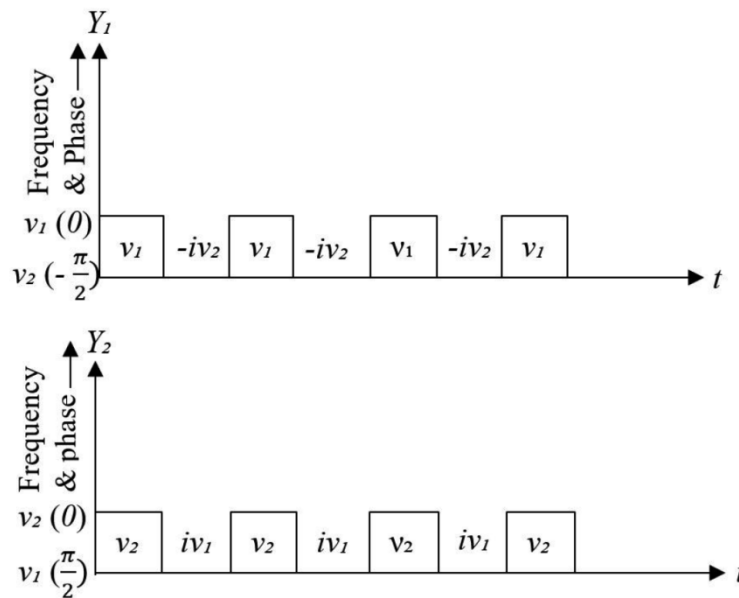
## 6. Operation

Frequency encoded Pauli Y gate, as proposed by these authors, consists of a fibre optic cross coupling system, a  $\frac{\pi}{2}$  phase delay and a  $\frac{\pi}{2}$  phase leading mechanism<sup>15</sup>. Pauli Y gate converts the input qubit  $\begin{pmatrix} v_1 \\ v_2 \end{pmatrix}$  to output qubit  $\begin{pmatrix} -iv_2 \\ iv_1 \end{pmatrix}$  governed by the matrix equation  $Y \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} = \begin{pmatrix} -iv_2 \\ iv_1 \end{pmatrix}$ . “+i” and “-i” designate  $\frac{\pi}{2}$  phase lead and  $\frac{\pi}{2}$  phase delay of the two optical signals. When two monochromatic optical beams of frequencies  $v_1$  and  $v_2$  propagate as two inputs A and B through this Pauli Y gate, at the output end, the optical outputs take the form  $Y_1 = -iv_2$  and  $Y_2 = iv_1$ . This Pauli Y gate fulfils the basic

requirements of a quantum gate that is reversibility. These outputs are sent back to inputs A, B and initial inputs are withdrawn. A second trip through Pauli Y gate system gives the initial state back (reversibility), i.e,  $Y'_1 = \nu_1$  and  $Y'_2 = \nu_2$ . A further round cycle gives again the state  $\begin{pmatrix} -i\nu_2 \\ i\nu_1 \end{pmatrix}$  and it will continue to oscillate between these two states as the feedback system runs the oscillator.

### 7. Analytical discussion

Analysis of this photonic scheme reveals that the input A of frequency  $\nu_1$  after single passing through Pauli Y system becomes  $-i\nu_2$  and after one more passing becomes  $\nu_1$ . An additional passing gives  $-i\nu_2$  again which becomes  $\nu_1$  for another extra passing. This shows that the input frequency  $\nu_1$  oscillates between two states  $\nu_1$  and  $-i\nu_2$ .



**Fig.2**

Outputs of quantum oscillator.

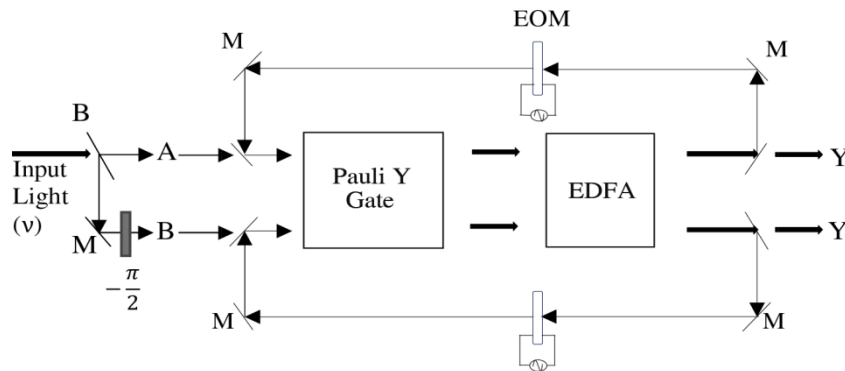
Similarly the second input with frequency  $\nu_2$  also toggle between the states  $\nu_2$  and  $i\nu_1$  as shown in figure 2. Therefore, in this photonics scheme, the two signals of different frequencies will oscillate with either  $\frac{\pi}{2}$  phase lead or  $\frac{\pi}{2}$  phase delay or in phase. The operation can be clearly depicted by the truth table given in table 1.

**Table 1**

Input (A)	Output ( $Y_1$ )	Input (B)	Output ( $Y_2$ )
$\nu_1$	$-i\nu_2$	$\nu_2$	$i\nu_1$
$-i\nu_2$	$\nu_1$	$i\nu_1$	$\nu_2$
$\nu_1$	$-i\nu_2$	$\nu_2$	$i\nu_1$

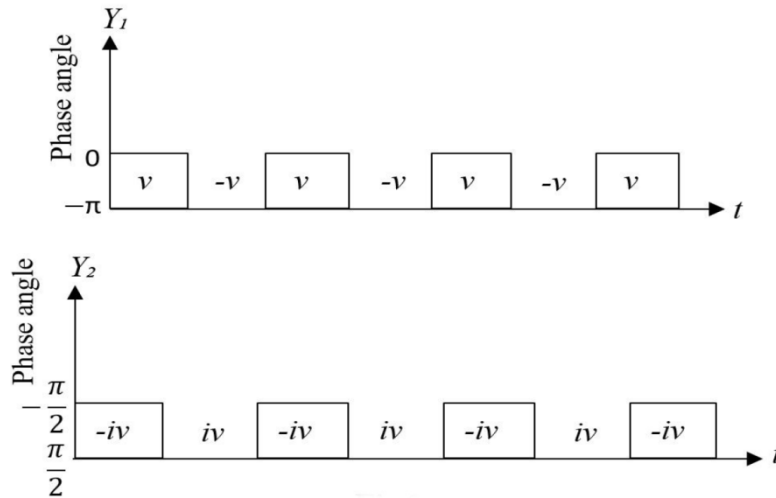
Truth table for frequency encoded quantum oscillator.

It is possible to use this photonic system as a phase oscillator of a monochromatic signal of frequency  $\nu$ . For this purpose a monochromatic signal of frequency  $\nu$  serves as 1<sup>st</sup> input component of the single qubit state and the same signal split by a beam splitter having its phase delayed by  $\frac{\pi}{2}$  as the other qubit component. The output  $Y_1$  of this oscillator will oscillate between two states one having same phase as initial one and the other by  $\pi$  phase lag. Similarly, the output  $Y_2$  will oscillate between the states with phases  $\frac{\pi}{2}$  and  $-\frac{\pi}{2}$  shown in fig. 3b. This photonic system will act as a phase oscillator of an arbitrary monochromatic input signal (Fig.3a).



**Fig.3a**

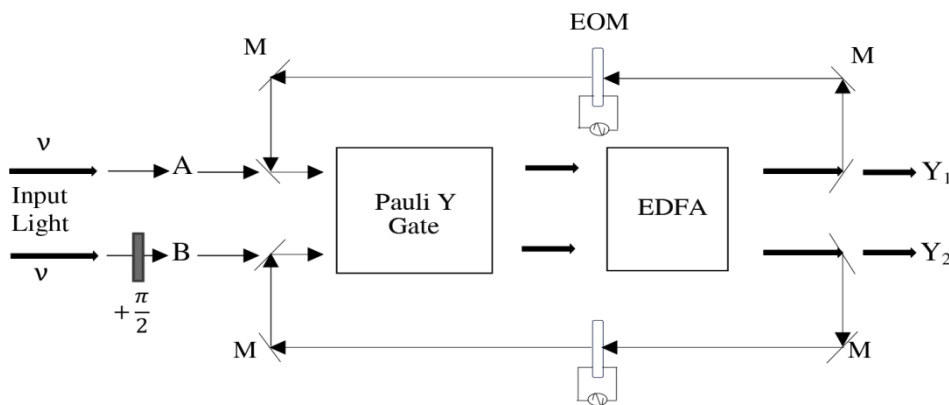
Optical implementation of quantum phase oscillator.



**Fig.3b**

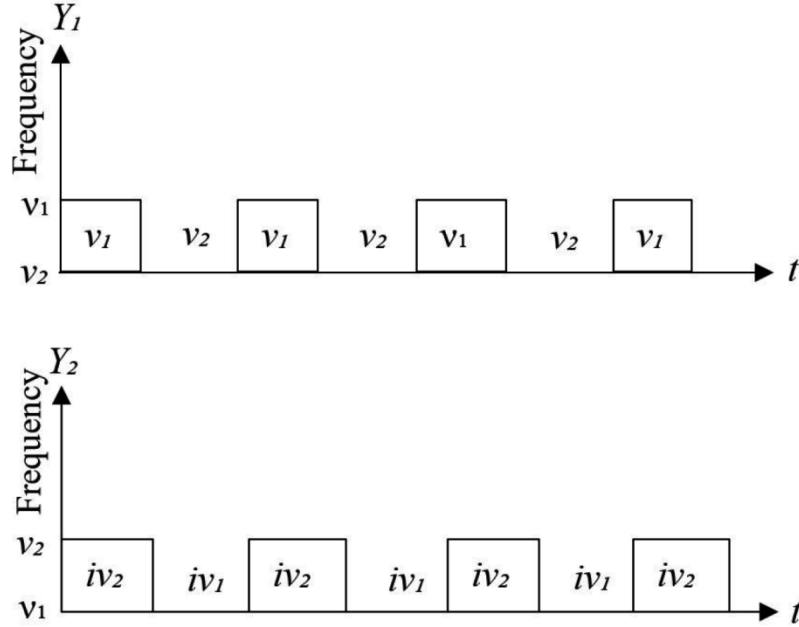
Operational output of quantum phase oscillator.

Again, if two signals with different frequencies are applied at the input face of the system and the 2<sup>nd</sup> signal of frequency  $\nu_2$  is employed with a phase lead by  $\frac{\pi}{2}$ , then the system will act as a frequency oscillator (Fig. 4a). Its outputs will toggle between two applied frequencies as shown in 4b.



**Fig.4a**

Optical implementation of quantum frequency oscillator.



**Fig.4b**

Operational output of quantum frequency oscillator.

### 8. Conclusion

Proposed frequency encoded quantum oscillator efficiently oscillate between two arbitrary frequencies with either  $\frac{\pi}{2}$  phase lead or delay in a channel and the other channel continues in phase. One can select the frequency which will be in phase or which will suffer a  $\frac{\pi}{2}$  phase change according to the requirement. In addition, one can determine whether a particular frequency of light suffer a change in phase by  $+\frac{\pi}{2}$  or  $-\frac{\pi}{2}$ . None the less, this all-optical scheme exhibits the role of a phase oscillator as well as frequency oscillator by proper selection of input qubit i.e., if the initial qubit is frequency as well as phase encoded. As

the whole system is all-optical, the operational speed approaches THz limit with assurance of high degree of data security.

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## **PROGRAMME**

### *Inaugural Session*

Introductory Lead by **Dr. Susil Kumar Sarkar**, Secretary, CITP

Welcome address by **Prof. Jayanta Kumar Bhattacharjee**, Director CITP

Inaugural address by **Prof. Debasish Mondal**, TIC, SPCMC

### *Technical Session*

#### **Prof. K. C. Kar Memorial Lecture**

Introduction to Prof. K. C. Kar by **Dr. Mrinal Kanti Chakrabarti**, CITP

Topic : **Climate Change - a Physicist's perspective**

Time: 4 pm

#### **Prof. Sibaji Raha**

Former Director, Bose Institute, Raja Ramanna Fellow.

#### **Prof. P.P. Chattarji Memorial Lecture**

Introduction to **Prof. P.P. Chattarji** by **Prof. Dulal Chandra Sanyal**, CITP

Topic: **Glimpses of Mathematics in Ancient India**

Time: 5 pm

#### **Dr. Partha Sarathi Mukhopadhyay**

Associate Professor in Mathematics  
R K M Residential College, Narendrapur

*Vote of Thanks* by **Dr. Amitava Sil**, HOD Physics, SPCMC

### **Introductory Note**

The association of St. Paul's Cathedral Mission College (SPCMC) with Calcutta Institute of Theoretical Physics (CITP) was not new. In fact, the department of physics of the college had opportunity to organise seminars in an occasion or two in collaboration with CITP before the pandemic broke. In the later half of the year 2021 there already had begun a norm worldwide to replace Seminars and Workshops by Webinars – a newly coined term that means 'Web-based-Seminar'. Like all other online social gathering, it had the challenge of handling technical issues but at the same time easier to find a common date and time for all the resource persons. During the pandemic period by organising many such events of our college online we gathered experience. So, when the proposal came from Dr. Susil Kumar Sarkar, the Secretary of CITP for organising jointly the event of 69<sup>th</sup> Anniversary of CITP, we promptly accepted it. In an online meeting with our departmental teachers and few key members of CITP we finalised the program schedule. It was decided that there will be two lectures: Prof. Sibaji Raha, the former director of the Bose Institute will deliver the Prof. K. C. Kar Memorial Lecture followed by Dr. Parthasarathi Mukhopadhyay of RKM Residential College, Narendrapur who will deliver the Prof. P. P. Chattarji Memorial Lecture. Dr. Pintu Mandal of the Department of Physics, SPCMC was given the responsibility of supervising the technical part of conducting the webinar on Google Meet platform. Dr. Aditi Sarkar of the Department of Physics, SPCMC agreed to design and distribute certificates to the participants. The other teachers of the department shared many such related responsibilities willingly. It was wonderful to see the whole team working together harmoniously.

On 25<sup>th</sup> September, 2021 the Webinar began on time.

In the beginning Dr. Susil Kumar Sarkar, the Secretary of CITP on the occasion of 69<sup>th</sup> Anniversary of the institute remembered its founder Prof.

K. C. Kar and narrated the life and work of the eminent theoretical physicist in a very impressive way. He described how Prof. Kar founded the institute at his own residence using his personal resources. Encouraged by his teacher Sir C. V. Raman, how he began his research while in M.Sc. class on Indian Musical Instruments. Later Prof. Kar turned his attention to many other areas of physics. He published altogether 123 research papers, out of which 44 had been cited 86 times. We were amazed to know that his most cited paper which was published in 1923 was first cited in 1925 and cited for last time in 2012. Dr. Sarkar told us how Prof. Kar started the publication of the Indian Journal of Theoretical Physics once again using his personal resources. He also said that CITP organises seminars every year to commemorate his legacy but last year it failed due to the break of pandemic. At the end of his speech Dr. Sarkar requested Prof. Jayanta Kumar Bhattacharjee, the Director of CITP and distinguished Emeritus Professor of Physics, IACS to deliver his introductory note. Prof. Bhattacharjee, on behalf of the Calcutta Institute of Theoretical Physics welcomed all on this 69<sup>th</sup> anniversary celebration of the institute. He reminded that the institute has survived almost seven decades and thanked all the people who helped keep it going. He mentioned about the two lectures to be given, welcomed the speakers and finally passed the stage to Prof. Debasish Mondal, the Teacher-in-Charge of St. Paul's Cathedral Mission College. Prof. Mondal, on behalf of his college, welcomed all the dignitaries. He reminded that St. Paul's C. M. College is a very old educational institute which was established by the Church Missionary Society in the year 1865. He then, in the line of the topic of the two lectures, said few sentences on climate change and the importance of statistical analysis for better understanding of global weather pattern. He also recalled the contribution of Indians in mathematics like negative numbers and zero and reminded us that even before the birth of Christ, Indian astronomy, which is a branch of mathematics as well as a branch of physics, was very much developed. That

is why, he reasoned from the story of Bible, the wise men who came from the east (India), just looking at the movement of the stars found the exact place where Christ was born. At the end of his speech, he thanked all the teachers of the department of physics of his college for the preparations they took in organising the webinar. Finally, it was time to begin the technical sessions. Dr. Mrinal Kanti Chakrabarti, distinguished member of the CITP, who also happened to be the former Teacher-in-charge of St. Paul's Cathedral Mission College, chaired the first session. The first lecture was Prof. K. C. Kar Memorial Lecture. Dr. Chakrabarti introduced the speaker, Prof. Sibaji Raha as an eminent physicist who is currently holding the chair of prestigious Raja Ramanna Fellow of the Department of Atomic Energy, Government of India and is associated with Bose Institute. Prof. Raha was born in Kolkata and after graduation from Presidency College went to USA for higher education including masters and Ph.D. He did his Ph.D. from Texas University USA and after conducting researches in various laboratories of Germany and France he came back to India and joined Saha Institute of Nuclear Physics, Kolkata as a Faculty member of High Energy Physics. He shifted to Bose Institute in 1991 and continued there for a long period. He held the post of Director of Bose Institute for a long 10 years from 2006 to 2016 and from 2019 he is Raja Ramanna Fellow. Besides he created National Centre for Astrophysics and Space Science at Darjeeling and Kolkata. Prof. Raha is the chairman of Joint Scientific Council of International Facility for Anti-proton and Ion Research. Dr. Chakrabarti described Professor Raha as a physicist of varied interest who has worked on High energy particle physics, nuclear physics, astrophysics, astro-particle physics, cosmology. Prof. Raha also contributed immensely on other rather non-technical topics like Global Change Scenario. He has authored about 400 scientific papers in international journals and also written extensively in non-technical publications on various issues. Professor Raha is associated with CITP as one of the eminent members of the international editorial

board of the institute journal as an adviser. Dr. Chakrabarti gratefully remembered his kind cooperation in organising seminar of this institute and invited Prof. Raha to deliver the Prof. K. C. Kar Memorial Lecture.

Professor Raha began his talk 'Climate Change: A physicist perspective' by giving a striking analogy between the evolution of the Earth system with the life cycle of a human being. The climate change crisis that we are facing today is similar to the lifestyle diseases that a person faces at midlife. To overcome such diseases, it is not only important to control our life-style but at the same time, a thorough understanding of the basic sciences related to physiological and biochemical changes due to aging is necessary. Unfortunately, the science that drives the Earth system is not very clear to us. Environmental Science, a subject in which the entire world is now investing maximum resources, is primarily studied from empirical view point. The basic fundamental issues at core often remain unaddressed and to some extent also ignored. The speaker in his talk promised to bring out some of those issues to the audience.

Climate change is mostly natural and has variety of cosmological and geological origin. In today's context, however, it refers to an additional and relatively rapid change induced by human actions. Our usage of fossil fuels like oil, natural gas, coal causes anthropogenic emissions which are primarily the main cause of global warming. But the basic science of climate change is still very poorly understood. Till date only one Nobel Prize has been given in this field in Chemistry in 1995 to Paul J. Crutzen, Mario J. Molina and F. Sherwood Rowland for ozone formation and decay in the atmosphere. The success of this research motivates one to emphasize basic scientific research in this area. Prof. Raha pointed out another aspect of the motivation behind such research. He said that in order to derive a full knowledge of the effect of human behaviour on various aspects of the climate, we also need to discriminate between what features of the climatic system we can control and what features we cannot. In the rest of his talk

Prof. Raha mainly focuses on the effect of variation of cosmic ray flux and solar radiation on climate change.

Historically the relationship between climate and the sun was first suggested in 1801 by William Herschel who proposed that the British wheat prices varied with the sunspot numbers. The idea stemmed from a valid observation that the variation in solar radiance could be correlated with the climate change on earth and different climates will affect the production of wheat and which will be reflected in the market price of wheat. There are of course very well-known correlations between the solar cycle variations and the tropical sea surface temperature. All of these are direct links. The indirect links are also noted. For example, solar radiation input and the cloudiness are seemed to be related. The solar radiation coming onto the earth can vary at the top of the atmosphere. The question that has puzzled scientists all along is how does this variability come down to the lower atmosphere? One school of thought to which Prof. Raha and his associates subscribe, claims that the most effective carrier of this variability down to the lower atmosphere is the cosmic ray particles. One of the most striking examples of the variation of cosmic ray on solar phenomena was observed during the total solar eclipse of October 1995 which was visible from large parts of India. During that period, a direct correlation between the amount of soft cosmic ray flux measured on the Earth's surface and the duration of the total solar eclipse was observed. One other feature, which has been seen for quite some time, is the amount of global cloud cover observed by satellites and the relative percentage variation of the cosmic ray flux. What one sees is that the lower the cosmic ray flux, the lower the cloud fraction. As if the more incoming cosmic rays facilitates more production of clouds. Prof. Raha explained the possible scientific reason behind it. Cloud is essentially a conglomeration of billions of little droplets of water and it forms when humid air is cooled and condenses on tiny particles suspended in the atmosphere which we largely call aerosols. Aerosols act as seeds of

condensation and the more the number of aerosols the better is the chance of finding cloud condensing nuclei (CCN). If we start with a collection of uncharged aerosols the CCN activation will be enhanced due to charge attachment and the activation of the CCN means a rapid growth of an aerosol into a larger droplet and this is where the cosmic ray comes in. Since cosmic rays are charged particles entering the Earth's atmosphere from outer space, collision of these particles with the aerosols will transfer charge and so the aerosol-charging will be facilitated by the cosmic ionization. Thus, one of the very basic expectations of the cloud microphysics is that if the cosmic ray flux is high the aerosol-charging will become more facilitated and that will lead to higher cloud cover. These processes were suggested almost 20 years ago but unfortunately there is no direct observation quantifying the modulation of the charge density near the clouds with the change in cosmic rays.

Prof. Raha told us about a satellite called the MODIS (Moderate Resolution Imaging Spectrometer) which investigates the cosmic ray cloud connection to a large extent and these data are available worldwide. This has led to a very detailed investigation of the response of clouds to sudden decrease in the flux of cosmic ray. As a result, this has given rise to a field of study called Cosmo-climatology. The main findings of the study is that the variation in the cloud properties from MODIS over the southern hemisphere subtropical oceans do not show any statistically significant correlation with variations in the global cosmic ray flux associated with the For bush decrease events. On the other hand, in the domains studied namely the 'Mid-Atlantic Region' showed correlations and values of cloud parameters are consistent with the cosmic ray induced CCN formation. At this point Prof. Raha explained that Forbush decrease is the decrease in the cosmic rays preceding solar flares. The general inference is that the stronger Forbush decrease events do show high correlations but the small number of events does not allow us to draw any firm conclusions.



Now as long as we're talking about satellite observations, they usually lead to average measurements. So, there is some relevance of designed experiments, if we can frame them up. The CLOUD project in CERN which ran about 12 years had a proton synchrotron as a source of cosmic rays and since one can change the flux and the energy of the proton this would be adjustable cosmic ray flux. It also had a large aerosol chamber where supersaturated water vapour could be contained and the conditions anywhere in the atmosphere could be recreated. This chamber then could be exposed to the artificial source of cosmic ray, adjusted to mimic the natural cosmic rays. Giving such an example of experimental setup, Prof. Raha reminded that the cosmic ray cloud connection leads to an understanding of the microphysics that requires observational inputs like the atmospheric conditions and the local cosmic ray environment. The clouds do not travel intercontinental distances. They form, develop, move a few kilometres, may be tens of kilometres and dissipate. Clouds are therefore a local phenomenon. So, it is important to know the cosmic ray environment of the region where the cloud is getting formed.

Prof. Raha reported that they started a project about 15 years ago under the aegis of the Department of Science and Technology, Government of India called the establishment of the National Centre for Astro-particle Physics and Space-science at Darjeeling, a place which Prof. Raha described as the most suitable for such study. Darjeeling also happens to be one place where Bose Institute had a campus established by the founder himself many years ago and therefore when Prof. Raha and his team were looking for a suitable site to locate a laboratory where all these various measurements could be carried out, the Bose Institute site came into their consideration. For the purposes of this kind of study they had a small cosmic ray detector array. For this atmospheric aspect they also clubbed all of those in the space science objective. Primarily the goal was to monitor all the physical, chemical and the radiometric environment of the region. The data that has

been collected there at Darjeeling provides all the things that are needed for an understanding of the cloud microphysics. This project was initiated sometime in 2007 and within 14 years they could generate substantially important data both for national as well as international databases. The aspect of understanding of microphysics of the cloud requires much more work and there it is continuing. Prof. Raha was hopeful that in next 5 years or so he would be able to report on the findings that will come out of this study.

Before concluding Prof. Raha made a very important remark: What is pure science today leads to technology tomorrow. If technology comes from a thorough understanding of basic science, then that would be sustainable. Otherwise, technology based on empirical findings will need frequent revision. To highlight importance of basic science research he cited the example of the Cosmic Ray Laboratory of TIFR which was established 60 years ago at Ooty in Tamilnadu for researches in basic science. In 2016 following a large magnetic storm due to Solar activity it reported registering a high cosmic ray burst 30 to 35 minutes before the actual magnetic shock came and hit the atmosphere. The report created a huge amount of interest in the international community due to its potential merit of finding advance warning of magnetic storm of large scale and avoid disaster to civilization on the Earth. The Laboratory in Ooty was established for understanding basic facts of nature but what we are now realising is that an understanding of the basic facts of nature ultimately will point to our sustenance of life as we know it.

Prof. Dulal Sanyal, a distinguished member of the CITP, chaired the second technical session in which the Prof. P. P. Chattarji Memorial Lecture was scheduled. In his introductory address, Prof. Sanyal told us that the full name Prof. P. P. Chattarji was Prafulla Prasun Chattopadhyay. Prof. Chattarji was born on 1<sup>st</sup> October 1925 in a small village of district Birbhum. He was a brilliant student who stood first in the district of

Murshidabad in Matriculation Examination in the year 1941. He graduated from Krishnath College, Baharampore and did his Post-Graduation from Applied Mathematics Department of Calcutta University. He obtained D. Phil. degree in Applied Mathematics under the supervision of late Prof. B.V. Sen. During the early days of his working life, he first served as a teacher of Mathematics at Krishnath College, Baharampore, then at Calcutta Technical School and at IIT Kharagpur. He joined the Department of Applied Mathematics of Calcutta University in the year 1962 and served there till his retirement. Many students carried research under his supervision and obtained Ph. D. or D. Phil. degree. A good number of his research students are established in various colleges and universities in India and abroad. He was a very popular, soft spoken and kind hearted teacher. He had co-authored some books on Astronomy and Technical Mathematics. Prof. Chattarji had long association with CITP, Calcutta Mathematical Society and some other academic organisations. The eminent mathematician, scholar and teacher, passed away on 4<sup>th</sup> January 2020. What we call Memorial Lecture today, used to be known as endowment lecture only few years back and Prof. Sanyal became nostalgic to remember that he was lucky enough to present one such endowment lecture when Prof. Chattarji was alive.

Prof. Sanyal then introduced the speaker of Prof. P. P. Chattarji Memorial Lecture, Dr. Parthasarati Mukhopadhyay as a young Mathematician. Prof. Mukhopadhyay is now an Associate Professor of Mathematics at Ramkrishna Mission Residential College at Narendrapur. He stood first in First Class in Pure Mathematics from Calcutta university and obtained his PhD degree in Pure Mathematics from CU in 2000. His field of research is Algebra. He is member of various national committees of mathematics in India. Dr. Mukherjee is author of many books on mathematics like Abstract Algebra, Rudiments of Mathematics, History of Mathematics and so on. Dr. Mukherjee also translated the famous book named *The Man who knew*

*infinity*, the biography of Ramanujan. The translated Bengali book was formally released by Prof. Amartya Sen, the Nobel Laureate.

After this short introduction, Prof. Dulal Sanyal invited the speaker to present the Prof. P. P. Chattarji Memorial Lecture on the topic *Glimpses of mathematics in Ancient India*.

Prof. Mukhopadhyay began his talk with a statutory warning that the subject of Ancient Indian Science, Mathematics in particular, are frequently being addressed nowadays by politicians in India which has put the scholars, who wants to pursue the subject for its own sake, in a very difficult situation. These people who are not academicians are making tall claims without knowing the subject. On the other hand, there are also some scholars who are keen to establish that all the claims about ancient Indian mathematics are absolutely false and all the knowledge came from Greece or Eurocentric doctrine. So, the speaker advised the audience to try themselves to find out the truth and make their own opinion about it instead of listening to others. Since the subject of ancient Indian mathematics is vast with huge repository the speaker suggested that he would pick up one or two topics, discuss those in real depth and then leave it to the audience for pursuing it more in future. Dr. Mukhopadhyay assured that these days there is no dearth of good reference books and anyone who wishes can study the whole aspect in a much deeper and better way.

In an interview to Times of India, Professor Manjul Bhargava, the first ever Fields Medallist of Indian origin, once said that “*Students in India should be taught about great ancient Indian mathematician like Panini, Pingala, Hemachandra, Aryabhata, Bhaskara ... many of these works were written in Indian languages in beautiful poetry and contain important breakthroughs in the history of mathematics*”. Dr. Mukhopadhyay emphasised the words ‘important breakthroughs’ and repeated that those were not just ‘ordinary’ results. In another interview when Manjul was asked to name his most

favourite five books that had most lasting impact on him, his first choice was 'Shulba-Sutras' by Baudhayana. He said that the book goes back to 800 BC and was perhaps the first place where Pythagoras theorem came on. The speaker pointed out that this refers to an era about 300 years before that of Pythagoras! Manjul's second choice was Chandrasutra by Pingala, which is a book written around second or third century BC, where in the last chapter one may find topics which are related to decimal to binary conversion, Pascal triangle, Fibonacci sequence and many more.

Dr. Mukhopadhyay gave one customary warning that whenever 'India' is referred with respect to its wisdom and knowledge achieved in the past, it is not the geographical location of India that we know today. It is rather the whole subcontinent including some parts of Afghanistan, some of Myanmar and so on. He showed the photographs of ruins of few famous Indian learning sites and mentioned the names of Takshila, Nalanda, Vikramshila, Udantapura, Shramapura. He made an attempt to answer a frequently asked question: Had there been so much of knowledge in ancient India where had it vanished? Why there was no continuation of that knowledge? One possible answer emerges when we look at the geographical locations of these centre of learnings and notice a pattern in the shift of its locations with time. It originated with Takshila in the Gandhara region at northwest corner of the then India, which is now the land of Afghanistan. The institute bloomed there but after few foreign invasions it was completely demolished in 5<sup>th</sup> century. The centre for learning then shifted to Nalanda at central east of India but invaders kept on coming. After a point of time in history, the learning Centres came to the other side of the Bindhya mountains, went to Kerala coast with a hope that possibly northern invasions never could cross Bindhya. But such is the unfortunate history of India that the next phase of invasion started along Kerala coast with the Europeans. All the manuscripts and materials full of knowledge were either destroyed or taken away. Many

of those are now found in repositories of various different libraries of Europe.

Dr. Mukhopadhyay then mentioned some big names of ancient Indian mathematical luminaries, which he said to be a very incomplete list. He mentioned the names of Baudhayana, Panini, Pingala, Aryabhata, Bhaskara and Brahmagupta. He did not forget the Kerala school but remembered the name of Guru Madhava. He mentioned about the Bakshali manuscript, the first ever known written mathematical document of India. Any discussion on ancient Indian mathematics cannot be made without a reference to the great invention of zero, the decimal place value system in tandem with the numeral zero, which is believed to be happened in India. The story and history of zero is so rich and so deep that other mathematical achievements of ancient India appear insignificant compared to it. He gave references for further learning on this topic and invited to visit the 'Zero Gallery' at his college where the story of 'zero' across different civilization and time is told. He also gave us a very important information which goes against a very common belief that Aryabhata invented zero. The fact stands that Aryabhata never invented zero. He used zero in his calculations for extracting square roots, cube roots of numbers and in many other occasions but he did not invent zero. In fact, at least 700 years earlier to Aryabhata, in Pingala's book Chandra Sutra, we find a concrete reference to zero. It is a sorry state of affair that there exist plenty of such examples depicting lack of correct information about ancient Indian mathematics.

Dr. Mukhopadhyay then explained a specific work of Aryabhata where the scholar had attempted to give the value of Pi. In those days there happened to be different kind of fire altars of different geometrical shapes like semicircle, circle, rectangle, square and many more. The civilization used to believe that those fire altars were very sacred and areas of those fire altars of different shapes had to be equal for ritualistic reasons. This belief of those ancient civilization was very important for the development of Indian

mathematics. Dr. Mukhopadhyay then explained a two-line sutra written in Sanskrit taken from Aryabhatia- the work of Aryabhata. It gives the approximate size of circumference of a circle of diameter 20000 unit. When translated in modern language of mathematics, it gives the approximate value of Pi correct up to 3 decimal places. The most important aspect about this finding is that this gentleman Aryabhata knew that the value is approximate and not exact.

Next the speaker told us about Sulba-Sutras. 'Sulba' was the name of a kind of knotted strings to measure the fire alters of different shapes. Again, he repeated, that people living in those civilization believed that exact shape and size of fire altars are absolutely important to enjoy the benefits of those rituals. Because they were so particular of this exactness, the geometry in India thrived. The rules of different kinds of geometrical constructions are found in 'Sulba-Sutras'. Among 'SulbaSutrakars' – the writers of 'Sulba-Sutras', Dr.Mukhopadyay mentioned the name of four luminaries: Boudhayana, Manava, Apasthambha and Katyana. The translation of their work reached west much earlier in 1875 through the translation of the sutras by G. Thibaut. Later in 1932 Bibhutibhushan Dutta and A. Seidenberg in 1962 did similar work among many others. A recent example is the book written by Jean-Michel Delire from France on BoudhayanSulvasutra in 2016. Michel Danino, a Padmashree and faculty of IIT Gandhinagar, wrote a review of this book in 2018 where he says "*In a long discussion on so called Pythagoras theorem, which Boudhayana expressed differently (as the property of the diagonal of a rectangle rather than the hypotenuse of a right-angled triangle)...Delire makes a praiseworthy attempt to grasp the Sulvasutras on their own terms rather than through the prism of either Greek or modern approaches*". The speaker then analysed the 'Boudhayana Sulvasutra' word by word while translating it from Sanskrit for the audience. It says that '*what length and breadth of a rectangle do together, the diagonal does the same thing alone*'. The historians of mathematics see

Pythagoras theorem in the above statement but it is quite natural to raise objection to such conclusion. The speaker then explained why such objections are not pertinent. He said that Boudhayana was not writing a geometry book like Euclid did for us. Sulvasutras are collection of rules for society which was on oral tradition. Sutras were meant to be cryptic, as short as possible, unambiguous, like a gist, of universal applicability, without any extra cosmetic word and unique. The explanation was there but it was meant to be transmitted verbally from Guru to Sishya and as a part of Indian culture those were never written explicitly. The speaker explained that to understand fully the 48<sup>th</sup> Sulvasutra discussed above, one has to read 50<sup>th</sup> sutra where it becomes apparent what is meant by 'doing' in 48<sup>th</sup> rule. He then translated the sutra word by word which addresses the problem of amalgamating two squares of different sizes to construct one bigger square having area equal to the sum of areas of the given two. The 50<sup>th</sup> Sulbasutra is the rule of construction of the bigger square. The inherent logic magically explains why the historians of mathematics see Pythagoras theorem in the 48<sup>th</sup> rule.

Dr. Mukhopadhyay then took another Sulbasutra, this time of Katyayana, where essentially the rule of geometrical construction of square root of a number is stated. The problem is to find the length of a square whose area is the sum of  $n$  squares of equal size. The intension and approach were purely geometric although essentially it is algebraic from present-day point of view. The method of construction when translated and explained word by word by the speaker, the realisation was awesome.

Finally it was the rule number 54 of Baudhayana Sulba that was addressed by the speaker. It was about the construction of a square whose area is equal to the area of a given rectangle. In his book 'The origin of Mathematics' Seidenberg says "*This is entirely in the spirit of The Elements of Euclid Book II indeed. I would say it's more in the spirit of Book II than Book II itself.*" The exact construction was explained by Dr. Mukhopadhyay while



translating the original sulba-sutra word by word from Sanskrit. The effect was again awe-struck feeling that enlightens that how rich was mathematics in ancient India in its own terms.

In the conclusion Dr. Mukhopadhyay asked the audience to judge themselves whether they would now keep the tall claims about ancient India like existence of internet, jet plane, test-tube babies which are mostly beliefs of a section of scholars who does not care to give any proof, at the same footing with the existence of Pythagoras theorem in ancient literature of India much earlier than Pythagoras time.

### **Interactive Session**

Participants in the webinar included college teachers and students and scientists from different institutes of Kolkata. At the end of each talk, ten minutes were allotted for comments and questions. Both the speakers were enthusiastic and interested in hearing from the audience. Teachers and students asked questions to clarify some important points pertaining to the topics presented by the speakers and the discussion became very lively and interesting. It is noteworthy that the speakers appreciated the students for raising relevant questions. Both the lectures evoked strong interactive sessions between students and scientists and the purpose of organising this webinar was fulfilled.

### **Vote of Thanks**

Dr. Amitava Sil, the Convener of the webinar and Head of the Department of Physics, SPCMC expressed his sincere thanks to the distinguished speakers for their excellent presentation and making the webinar meaningful and interesting. Dr. Sil also expressed deep gratitude to Dr. Debasish Mondal, the Teacher-in-charge of SPCMC and Prof. Jayanta Kumar Bhattacharjee, the Director of CITP for their interesting and thought-provoking addresses. He thanked all the audience for their significant

presence and taking part in interactive discussion of the webinar. Special thanks were given to Dr. Pintu Mandal of the Department of Physics, SPCMC for taking the responsibility of supervising the technical part of the webinar on Google Meet Platform and Dr. Aditi Sarkar for her sincere effort in designing and distributing the certificates among participants. Finally Dr. Sil thanked all the members of the organising committee and other people who were directly or indirectly helped us in making the webinar successful.

Dr. Mrinal Kanti Chakrabarti  
*Sr. Member, CITP*  
*Joint Convener*

Dr. Amitava Sil  
*HOD, Physics, SPCMC*  
*Joint Convener*

### Dignitaries present in the Webinar



Prof. D. Mondal

Prof. J. K. Bhattacharjee

Dr. M. K. Chakrabarti

Dr. A. Sil



Prof. D. C. Sanyal

Dr. S. K. Sarkar

Prof. S. Raha

Dr. P. S. Mukhopadhyay

### Climate Change: A physicist perspective

**Prof. Sibaji Raha**

Former Director, Bose Institute

Raja Ramanna Fellow

### Abstract

Climate change is mostly natural and has variety of cosmological and geological origin. In today's context, however, it refers to an additional and

relatively rapid change induced by human actions. In order to derive a full knowledge of the effect of human behaviour on various aspects of the climate, we need to discriminate between what features of the climatic system we can control and what features we cannot. This requires a clear understanding of the basic science of climate change. In today's world every nation is investing its resources to combat global warming but ironically in this context Environmental Science is primarily studied from empirical view point. The basic fundamental issues at core often remain unaddressed and to some extent also ignored. In order to understand the basic science of climate change we have addressed here one important aspect of the issue: the effect of variation of cosmic ray flux and solar radiation on climate change. The link between the two is sometimes direct, like the well-known correlations between the solar cycle variations and the tropical sea surface temperature. One example of indirect links is the observed relation between solar radiation input and the cloudiness. The variation of solar radiation at the top of the atmosphere may affect the lower atmosphere via cosmic ray particles. One of the most striking examples of the variation of cosmic ray on solar phenomena was observed during the total solar eclipse of October 1995 when a direct correlation between the amount of soft cosmic ray flux measured on the Earth's surface and the duration of the total solar eclipse was observed.

One other feature which has been studied for quite some time, is the amount of global cloud cover observed by satellites and the relative percentage variation of the cosmic ray flux. It has been found that more incoming cosmic rays facilitates more production of clouds. A possible scientific reason behind it may be as follows. The water droplets in cloud forms when humid air is cooled and condenses on aerosols, the tiny particles suspended in the atmosphere, which act as seeds of condensation. The occurrence of rapid growth of an aerosol into a larger droplet is known as activation of cloud condensing nuclei (CCN). The activation process can be enhanced by

charge attachment to aerosols. Since cosmic rays are charged particles, aerosol-charging will be facilitated by collision of these particles with the aerosols. Thus, one of the very basic expectations of the cloud microphysics is that if the cosmic ray flux is high the aerosol-charging will become more facilitated and that will lead to higher cloud cover. These processes were suggested almost 20 years ago but unfortunately there is no direct observation quantifying the modulation of the charge density near the clouds with the change in cosmic rays.

Response of clouds to sudden decrease in the flux of cosmic ray was investigated using the data obtained from MODIS (Moderate Resolution Imaging Spectroradiometer) satellite. The events of decrease in the cosmic rays usually precede solar flares and are known as Forbush decrease events. The general inference is that the stronger Forbush decrease events do show high correlations but the small number of events does not allow us to draw any firm conclusions.

Understanding of the microphysics behind cosmic ray-cloud connection require observational inputs like the atmospheric conditions and the local cosmic ray environment. This is because clouds are local phenomenon and hence it is important to know the cosmic ray environment of the region where the cloud is getting formed. Since satellite observations usually lead to average measurements, such data are not suitable for this purpose. To meet the requirement, we started a project about 15 years ago under the aegis of the Department of Science and Technology, Government of India called the establishment of the *National Centre for Astro-particle Physics and Space-science* inside Bose Institute campus at Darjeeling, a place which is most suitable for such study. For this purpose, we had a small cosmic ray detector array. We also clubbed all of those in the space science objective to study the atmospheric aspect. Primarily the goal was to monitor all the physical, chemical and the radiometric environment of the region. The data that has been collected there at Darjeeling provides all the things that are

needed for an understanding of the cloud microphysics. This project was initiated sometime in 2007 and within 14 years we could generate substantially important data both for national as well as international databases. The aspect of understanding of microphysics of the cloud requires much more work and there it is continuing. We are hopeful that in next 5 years or so we would be able to report on the findings that will come out of this study.

What is pure science today leads to technology tomorrow. If technology comes from a thorough understanding of basic science, then that would be sustainable. Otherwise, technology based on empirical findings will need frequent revision. An understanding of the basic facts of nature ultimately will point to our sustenance of life as we know it.

## GLIMPSES OF ANCIENT INDIAN MATHEMATICS

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### **Abstract**

Though India in Antiquity is generally well-known among common educated folks for its lofty philosophical ideas and thoughts, its unique mathematical achievements have not come to the limelight with equal importance and glory, possibly but for one instance: transformation of the philosophical idea of *SUNYA* into the invention of mathematical ZERO as a number in its own right in tandem with the decimal place-value notation, which is recognized nowadays worldwide as a cornerstone of human

civilization, peerless among the advancement of knowledge as a whole. However, this unique accolade has somewhat eclipsed many a praiseworthy and deep mathematical idea that evolved and flourished in India during ancient time, most of which now go by the names of one European mathematician or the other from decidedly later period of time. This happened mostly due to our general ignorance that crept in through either colonial bias of some early British historians of this period, or political/ideological compulsion of upholding the “Greek supremacy” by some relatively modern “Eurocentric” historians/scholars, which can be traced in the historiography of ancient India. On the other hand, recent times have witnessed a few tall claims about ancient Indian scientific achievements, mostly made from non-academic quarters, without much of proper scientific, analytic or historical back-up, which tends to put any sceptic mind in the denial mode about any and every achievement as a whole. However, not an iota of braggadocio is needed to appreciate the mathematical achievements of ancient India, if we objectively look at the long list of unparalleled feats attained. To mention only a few, these include results of plane Geometric exact constructions to be found in the *Sulba Sutras*, based on the enunciation of the so called “Pythagorean theorem”, much ahead of the Greek “origin” of plane geometry; a lineage of combinatorial marvels starting from *Pingalachandasutra* of third century B.C., leading to results, now commonly known as “Pascal’s triangle”, “binary numbers and conversion of binary to decimal and vice versa”, “binomial coefficients” etc. about two thousand years ahead of their rediscovery in Europe by Pascal and Leibniz respectively, to the *prakrit* works of Virahankain 600 C.E. which is now referred to as Fibonacci numbers; Arithmetical gems in the *Bakshali* manuscript, e.g., sum of arithmetical progression, calculation of weighted arithmetic mean, rule for finding approximate square root of a non-square integer, which is akin to Newton’s calculus-based later work known as Newton-Raphson method;

solutions of indeterminate equations--- *Kuttaka* algorithm of Aryabhata, his calculation of approximate value of  $\pi$ ; the algebraic principle of *Bhavanaby* Brahmagupta (one of whose brilliant work is now erroneously referred to as Pell's equation) towards solving some specific quadratic "Diophantine equations", which was later furthered by Bhaskara II and Jayadeva through *Chakravala* algorithm; the series expansions of trigonometric functions achieved by the Kerala school of Madhava of Sangamagram and his disciples, e.g., Madhava-Gregory series of arctangent function, some two hundred years before inception of calculus in the West, etc.

In this lecture, we shall try to throw some light on a few of these achievements in a nutshell manner.

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