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Formulation of "Exciton in a Quantum Box Model" for Multiple Exciton Generation Solar Cell Applications

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***Abstract*—**Exciton in a quantum box model for multiple exciton generation solar cell applications is successfully formulated. We have modeled quantum dot QD as a quantum box and then solved the time independent Schrödinger Equation to determine the Eigen state energies of the confined exciton. In broader perspective the model contributes to the better understanding of the working principles of multiple exciton solar cells and enables the improvement of their performances. The results obtained strongly reveal that multiple exciton generation is more efficient in QDs because of the enhanced Coulomb interaction between the exciton, slow hot exciton cooling and relaxed momentum conservation due to quantum confinement effects. These novel properties allow for proper utilization of the hot exciton created by the absorption of high energetic photons to generate more excitons instead of losing the excess energy to phonon emission. The creation of additional excitons dramatically minimizes the spectral losses due to thermalization which characterized the conventional device thus making it possible to absorb and utilize supra band photons suitable for high efficient multiple exciton generation solar cells. This produces significant increase in solar to electricity conversion efficiencies in the form of increased photo generated current.

***Keywords*—**Theoretical model, Quantum dot, Multiple exciton generation, Excitons, Quantum confinement, Solar cells, Potential well and Schrödinger Equation.

I. INTRODUCTION

Solar cells for conversion of solar energy into electrical energy are of great significance for harvesting, development and efficient usage of renewable (green) energy to control the problem of climate change and pollution that characterized with the use of fossil fuel. The generation of multiple excitons from an absorbed high energy photon provides a mechanism for increasing the efficiencies beyond the Shockley and Queisser established efficiency limit of the conventional solar cell devices. Quantum dots QDs have emerge as important class of innovative materials that can be utilized to realize this novel concept due to the quantum effects that confer on them unique optical and electronic behaviour.

The interest in QDs arises from their specific novel characteristics that substantially deviate from those of bulk solids and molecules. For bulk semiconductors the charge carriers are not confined and are free to move within their respective bands in all the three directions, in this state, the band gap energy maintains its original value owing to the continuous energy state, whereas in QDs, confinement becomes predominant which dramatically modulates the carrier states [2]. QDs are semiconductors in nanometer dimensions providing strong quantum confinements on the carriers in all the three spatial directions [3], in addition to broad absorption characteristics that are tunable due to quantum size effects [4]. Studies have shown that device characteristics based on QDs are strongly altered by quantum mechanical effects which undermine the classical principles upon which most of present day electronic components are based [5]. In this perspective, alternative materials and approaches are underway for novel nano-scale electronic devices, in which the laws of quantum mechanics regulate their behavior in a controllable manner. A fundamental concept in quantum physics is the wave-particle duality, introduced by de Broglie, in which any particle can be associated with a matter wave having wavelength that is inversely dependent on the particle's linear momentum.

Quantum size effects usually manifest whenever the size of a physical system becomes comparable to this characteristic wavelength; a length scale in which the Quantum mechanical model is used to describe motion and energy of the particle instead of the Newtonian (classical) Physics. All the information about the particle is obtained by solving the Schrödinger quantum mechanics, whose solution are mostly standing waves confined in the potential well and the energies associated with two distinct envelope functions are generally, different and discontinuous. This implies that the particle energies cannot assume any arbitrary value and the system exhibits a quantized energy spectrum [6], making QDs typical

example of quantum mechanics in action, which gives rise to zero-dimensional Physics in which the continuous energy bands distribution that characterized the bulk semiconductors broken into discrete atomic-like electronic states [7]. The emission and absorption wavelength of QDs corresponds to their band gap energy [8], this further makes QDs viable and suitable candidate for high efficient solar cells as their band gap can be engineered to desirable energies to better match the solar spectrum, [9].

A solar cell is a device that converts the solar energy into electrical energy through photovoltaic effect, without moving parts or polluting by-products [10]. The semiconductor materials are usually the absorbing material in solar cells. The ultimate goal of solar cells is to harvest and convert the full broad solar spectrum into electrical energy at high power conversion efficiencies to meet the global energy demands. The photon absorption in a semiconductor depends on the material `band gap [11]. Hence semiconductor band gap plays a key role in the range of photons a solar cell can absorb from the solar spectrum [12] and subsequent conversion of solar energy into electrical energy. The limiting performance in conventional solar cells is that upon absorption of photons with energies in excess of the band gap produces hot exciton which often lost as heat via lattice vibration in Pico second regime [13]. The hot exciton cooling is enhanced when the energy level spacing coincide with longitudinal optical phonon energy [14] which is favorable in bulk materials owing to their continuous band distribution.
Thus, thermalization losses are realized in the conventional device because the incident solar spectrum is very broad and poorly matches the fundamental band gap energy of absorber material and as a result the ultimate device performance is drastically reduced, which places an upper limit on the maximum power conversion efficiency to 31% [15] .The search for suitable semiconductor absorbing material on which cooling is slow down to allow for proper exploitation of the hot exciton has led to emergence of semiconductor QDs [16].

The most widely employed scheme to reduce these losses from hot exciton cooling is through carrier multiplication: an optical process in semiconductors whereby more than one exciton can be simultaneously generated upon absorption of a single high energetic photon through impact ionization (inverse Auger process) [17]. For bulk semiconductors both energy and momentum must be conserved during the generation of extra exciton and hence threshold energy needed for carrier multiplication is in excess of five times band gap energy [18]. The combined high threshold photon energies resulting from the restrictions imposed by energy and momentum conservation lies outside solar spectrum and the ultra-fast phonon relaxation render bulk materials inconsequential for carrier multiplication solar cell applications. Therefore for effective absorption and utilization of hot exciton for solar cells application, the threshold energies must be significantly minimized within the limit of solar spectrum.

In QDs the electron-hole pairs exist as exciton and thus the carrier multiplication is referred to as multi exciton generation [19]. Multi exciton generation allows conversion of high energy photon of solar spectrum that would otherwise lost as heat into usable energy. Multi exciton generation in QDs is significantly favored by quantum confinement which relaxes restriction imposed by crystal momentum conservation and thus reduces the threshold energy to the acceptable limit [20]. In addition, because of proximity of the excitons resulting from quantum confinement the Coulomb interaction between electron and hole that drives multi exciton generation is greatly enhanced. The combined enhanced coulomb interaction, slow phonon emission and relaxed momentum conservation make QDs an ideal candidate for multi exciton generation. The creation of additional excitons from hot electrons enhances the quantum yield (i.e., number of excitons per single photon absorption) [21] that will lead to a new solar conversion efficiency limit through increase in the photo generated current.

In general, excitons play major roles in the material interactions with photons. A deeper knowledge of the exciton dynamics, Eigen state energies and spatial extent in QDs is essential for understanding fundamental light harvesting processes in the QD-based solar cells design architecture. Thus simple excitonic models that provide theoretical predictions for fundamental efficiency enhancement is crucial. Presently, the theoretical models of excitons have not been adequately studied or available to a satisfactory extent. In this research we have formulated “exciton in a quantum box model” which has significant implications for improving performances in multiple exciton generation solar cells.

We organized the work in sections as follows. The study background and related literature are discussed extensively in section 1. Section 2 provides detailed theoretical formulation of exciton in a quantum box model which describes the system behavior followed by methodology described in section 3 which deals with the computation details and method of calculation. A detailed analytical results as obtained from the simple model have been provided and been thoroughly discussed as they are relate to multiple exciton generation in section 4. In the concluding section 5 the observed results are summarized.

II.EXCITON IN A QUANTUM BOX MODEL

Quantum dot QDs is a nano size semiconductor particle embedded in a large band gap semiconductor material. The QD then forms a potential well while the larger band gap material creates the potential barrier. In QDs, the allowed region in which this exciton moves is referred here as the potential well while the region where the motion is forbidden is regarded as potential barrier. Thus, a good approximation of an exciton behavior in QD is the particle in an infinite potential well and thus QD is imagined as quantum box. However, in practice QDs confine two particles in contrast to infinite potential well: the electron

and the hole. These two particles interact through the electrostatic Coulomb potential given by Equation. 1 to form an exciton (bound state of electron and a hole).

$$V_{(r)} = \frac{e^2}{\varepsilon r} \quad (1)$$

where

- e is the electronic charge
- r is the electron and hole. Separation
- ε is the dielectric constant of the host semiconductor.

We therefore treated the exciton as a quasi-particle moving with a center of mass motion. Due to its center of mass motion, we assigned a reduced mass μ to the exciton as shown in equation. 2

$$\mu = \frac{m_e^* + m_h^*}{m_e^* m_h^*} \quad (2)$$

where M^*e and M^*h are effective masses for electron and holes respectively.We introduced the concept of effective mass expressed in units of mass of electron to account for the interaction with the periodic potential. We have also considered this exciton of mass μ be confined in the region bounded by x = 0 and x = L by an infinite two potential barrier i.e., V = ∞ for x ≤ 0 and V = ∞ for x ≥ L so that the exciton moves freely within the quantum box but cannot penetrate the potential wall of infinite height rather bounces back and forth between the dot boundaries as shown in the Figure 1. Owing to this spatial quantum confinement, the electronic states of the exciton in QDs can be found by solving the Schrodinger quantum mechanics.

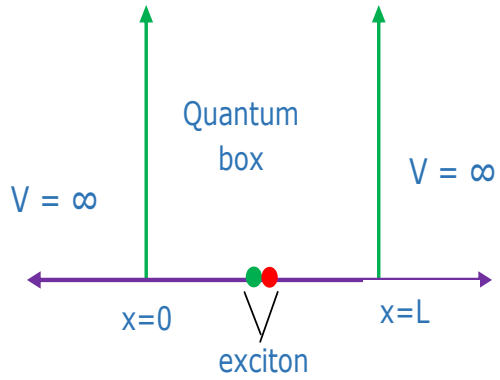

Figure 1: Exciton in the Quantum Box Model

Generally, the dynamics of particles in an infinite confinement potential are governed by the Hamiltonian of the form

$$\frac{h^2}{2m}\frac{d^2}{dx^2} + V_{(x)} = H \quad (3)$$

The energy Eigen state solution of the exciton in the QDs are thus found by solving time independent Schrödinger Equation shown below

$$\frac{h^2}{2\mu}\frac{d^2\varphi_{(x)}}{dx^2} + V_{(x)}\varphi_{(x)} = E\varphi_{(x)} \quad (4)$$

We rearrange the equation to get;

$$\frac{h^2}{2\mu}\frac{d^2\varphi_{(x)}}{dx^2} + [E - V_{(x)}]\varphi_{(x)} = 0 \quad (5)$$

Due to the similarities between the confinement of the exciton and the infinite potential well, boundary conditions of confinement principles were also observed here. Thus $\varphi_{(x)} = 0$ at x= 0 and $\varphi_{(x)} = 0$ at x= L. In analogy with the particle in a potential well problem, the potential function vanishes inside the dot and the resulting Schrodinger equation becomes;

$$\frac{d^2\varphi_{(x)}}{dx^2} + \frac{2\mu}{h^2}E\varphi_{(x)} = 0 \quad (6)$$

The equation 6 can be expressed as;

$$\frac{d^2\varphi_{(x)}}{dx^2} + k^2\varphi_{(x)} = 0 \quad (7)$$

where

$$k = \sqrt{\frac{2\mu E}{h^2}} \quad (8)$$

The differential equation of this kind will have a general solution;

$$\varphi_{(x)} = A\cos kx + B\sin kx \quad (9)$$

where A and B are integration constants that are defined by the boundary conditions. Recall the boundary conditions:

$$\varphi_{(0)} = \varphi_{(L)} = 0 \quad (10)$$

We impose the boundary conditions on the Equation 9, first at x= 0 and we have;

$$\varphi_{(0)} = A\cos 0 + B\sin 0 = A.1 + B.0 = 0 \quad (11)$$

The Equation.11 can only be true if A = 0 so that the solution is now

$$\varphi_{(x)} = B\sin kx \quad (12)$$

We apply the next boundary condition at x = L which gives

$$\varphi_{(L)} = B\sin kL = 0 \quad (13)$$

which means that either B= 0 in which case φ(x) = 0, this implies no exciton in the dot and it is not a useful solution or else sinkL = 0. This can only be true if kL is an integer multiple of π since sine of any integer multiple of π is zero. Therefore,

$$kL = n\pi \quad (14)$$

where n is a quantum number and thus

$$k = \frac{n\pi}{L} \quad (15)$$

Note that n=0 is not an acceptable solution here as it implies k = 0 which again makes the $\varphi_{(x)} = 0$ everywhere. Comparing Equation 8 and 15 we have.

$$k = \sqrt{\frac{2\mu E}{h^2}} = \frac{n\pi}{L} \quad (16)$$

Therefore,

$$En = \frac{n^2h^2}{8\mu L^2} \quad (17)$$

Conventionally, there is a spherical confinement of exciton in QDs rather than a square, we therefore interchange the length L with radius R. The energy Eigen state solution for exciton (exciton kinetic energy) becomes

$$E = \frac{n^2h^2}{8\mu R^2} \quad (18)$$

The Equation 18 is a simple model for exciton kinetic energy in the QDs.

Here E is the kinetic energy of exciton in the QDs, n is the quantum number, R is the dot radius, μ is the reduced mass of the exciton and h is Plank's constant. The model strongly indicates that energy levels of the exciton are quantized; limited to discrete values and this arises out of a need to fulfill the imposed boundary conditions on the system. The fact that n cannot be zero means that the lowest possible kinetic energy of the exciton is never zero but:

$$E = \frac{h^2}{8\mu R^2} \quad (19)$$

The Equation 19 is ground state kinetic energy model of the exciton in the QDs

To model two particle exciton such as electron and hole, we introduced an electronic interaction energy to account for the Columbic attraction between the negatively charge electron and positively charge hole each with charge of magnitude e. According to Coulomb's law, the electrostatic interaction energy is on the order of

$$E = \frac{-\beta e^2}{4\pi\varepsilon_0\varepsilon_r R} \quad (20)$$

We have multiply the Coulomb interaction effect by a coefficient β which arises as a result of wave function overlap between the electron and hole in QDs and it is approximately 1.8 (Brus 1984) in the first excited state. The minus sign indicates that two particles (electron and hole) are of opposite charges. Finally, exciton is not confined in an empty space but rather inside a host semiconductor crystal, thus semiconductor fundamental band gap energy becomes baseline energy of the system. Thus minimum energy needed to excite the quantum dot is thus made up of three main energy contributions. The first energy contribution is the infinite crystal band gap of the host semiconductor which is the characteristics of the material, another crucial contribution is the exciton kinetic energy which represents the infinite potential well contribution due to quantum confinement of exciton and the third term represents the electrostatic energy contribution due to coulomb's attraction between the electron and the hole as shown below

$$E_{g(QD_s)} = E_{g_{(bulk)}} + \frac{nh^2}{8\mu R^2} - \frac{1.8\,e^2}{4\pi\varepsilon_0\varepsilon_r R} \quad (21)$$

The Equation 21 is a simple model for exciton excitation energy of the QDs.

Where

- $E_{g(QD_s)}$ is the bandgap of QD_s
- $E_{g_{(bulk)}}$ is the bandgap of the bulk semiconductor
- n is the quantum number that labels the different confined energy levels of exciton within QD structure.

The ground state excitation energy which is the commonly observed state can easily be found, taking n=1 and is given as

$$E_{g(QD_s)} = E_{g_{(bulk)}} + \frac{h^2}{8\mu R^2} - \frac{1.8\,e^2}{4\pi\varepsilon_0\varepsilon_r R} \quad (22)$$

The inverse quadratic dependence on R in the exciton kinetic energy compared to the inverse linear dependence on R in the electrostatic interaction energy implies that in the strong confinement regime (small R limit) exciton kinetic energy dominates and thus electrostatic energy contribution could be ignored.

III. METHODOLOGY

The variation in band gap energy as a function of size were computed for CdSe QD using Equation 22 in order to ascertain the impact of quantum confinement on multiple exciton generation in QDs. The input parameters used in the theoretical calculation have been listed in the Table 1and Table 2 below.

Table 1: Experimental Parameters used for the Computation [24].

QDs	m_e^*	m_h^*
CdSe	$0.13m_o$	$0.45m_o$

Table 2: Approximate values of some physical constant

	Value
Electronic Charge (e)	1.6×10^{-19}C
Speed of light (c)	3×10^{8}m/s
Planck's constant (h)	6.63×10^{-34} J.S
Electron rest mass ($m_{0)}$	9.11×10^{-34} kg
Electron volt (eV)	1.6×10^{-19} J
Permittivity of free space	8.85×10^{-12} fm^{-1}
Pi (π)	3.142

IV. RESULTS AND DISCUSSION

The simple obtained models show that spatial confinement of excitons within the quantum box has two fundamental consequences that significantly modulate the carrier states. Firstly, the transformation of the continous energy level into quantized discrete electronic states which shifts the effective band edge to higher energies. The increased spacing between the discrete electronic states is largely dependent on the confinement strength. Accordingly, a large optical blue shift is observed in the excitation spectrum as compared with the parental bulk whose electronic energy levels are distributed in continuous bands as shown in the Figure 2 below.

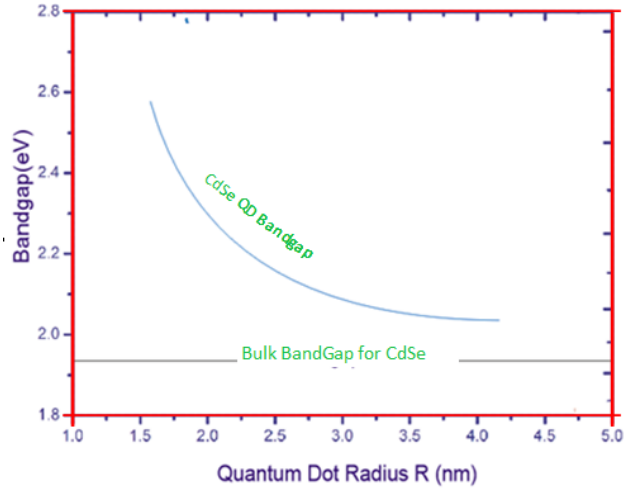

Figure 2: Band gap verses QD Radius

Secondly, confinement brings electrons and hole closer and therefore enhances the radiative recombination probability. The computed band gap for CdSe QD using our model is compared to the experimental values as observed by Harbold and Monica (2008) in the Figure 3.There is a good accord between our theoretical values and the experimental results which justifies the success of our models.

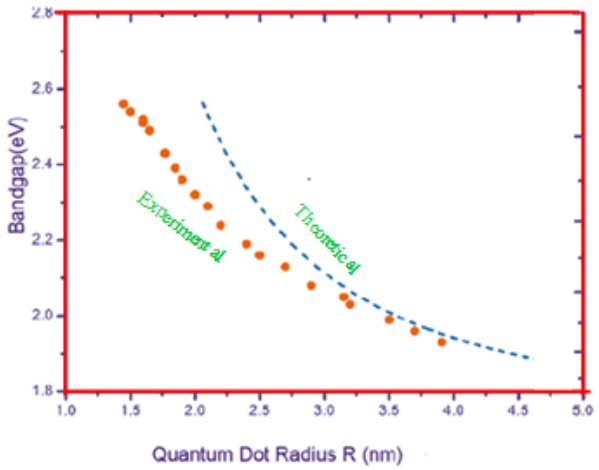

Figure 3: Computed/Experimental Band gap verses QD radius

Due to the proximity of the exciton resulting from quantum confinement the Coulomb interaction between electron and hole that drives multi exciton generation will be greatly enhanced in QDs. Also, due to quantization effects in QDs crystal momentum conservation is expected to relax. The relaxation of crystal momentum conservation reduces the threshold energy required for multiple exciton generation process in QDs within the acceptable limit. In addition, the wide separation between the quantized energy states in QDs can greatly exceed the longitudinal optical phonon energy and therefore gives rise to significant reduction in the carrier-longitudinal optical scattering rates that dominate energy relaxation process of the bulk and as a result a substantial increase in the optically excited hot exciton cooling time is expected in QDs in contrast to sub-picoseconds carrier relaxation time reported for the parental bulk.

The slow hot exciton cooling dynamics called phonon bottleneck is attributed to the potential mismatch between the wide electronic energy states and phonon energy in the QDs, making it possible for the rate of impact ionization (inverse Auger effect) to compete favorably with the rate of hot exciton cooling. Thus the competition between the hot exciton cooling and carrier multiplication significantly favors multiplication process in QDs. The hot exciton thus has life time that is strongly dependent on the confinement strength, the stronger the confinement the longer the hot exciton life time. This makes it possible to exploit hot exciton created by the absorption of high energy photons to produce more excitons instead of losing its energy to phonon emission.

The multiple exciton generation solar cell is a photovoltaic device designed to enhance solar to electricity conversion efficiencies by utilizing the excess energy in the absorbed energetic photons that would otherwise lost as heat. It is essentially a process in which hot exciton created by absorption of solar photon higher than the excitation energy (fundamental bandgap energy) is utilized to promote two or more excitons before they are thermalised through phonon emission. Thus, an electron from valence band is promoted by absorption of photon makes a transition to a higher level in the conduction band and generates one exciton. If the energy of the solar photon is at least twice band gap of the given QDs the exciton will posses high kinetic energy that can be released to promote a second exciton jumping from the valence band to the conduction band through impact ionization. In this way one photon generates two excitons as shown in the Figure 4

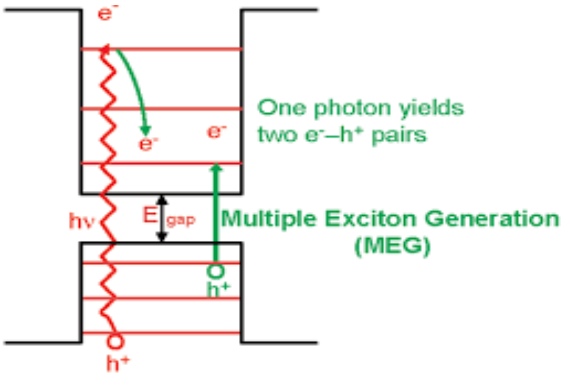

Figure 4: Multi Exciton Generation Mechanism

The multiple exciton generation is a novel concept to minimize loss processes associated with the incomplete absorption in the conventional cells and signifies a promising route to boost light absorption. In this way the excess kinetic energy thus contribute to conversion efficiencies through generation of more exciton rather than heating up the cells.

V. CONCLUSION

Exciton in a quantum box model for multiple exciton generation solar cells and their performance improvements was formulated within the frame work of effective mass approximation. The simple obtained model shows that spatial confinement of exciton within the quantum box enhances their coulomb interaction, slows hot exciton cooling (phonon emission) and relaxes momentum conservation which collectively favor inverse Auger process in QDs. These novel optical and electronic behavior greatly indicate potential route for multiple exciton being extracted from single light absorption in QDs contrast to the bulk which generally emit one exciton per photon absorption. The creation of additional exciton significantly minimizes the thermerlization losses that characterized the conventional devices and provides significant increase in power conversion efficiencies beyond Shockley and Queisser established limit via the increased in the photo generated current.

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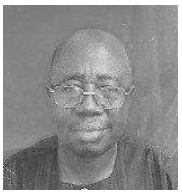
Ikeri Henry is presently pursuing Ph.D degree in Solid State Physics from Enugu State University of Science and Technology, Enugu, Nigeria. His research work focuses on theoretical characterization and the models for high performance Quantum Dot based on third Generation Solar Cells. He obtained his M.Sc. Solid State Physics with distinction in 2017 from Enugu State University of Science and Technology, Enugu, Nigeria. His areas of research interest are Nanotechnology, Material Science, Nano Electronics, and Solar cell Technology. He has published a number of research papers in a reputed peer review academic national and international Journals/Conferences. He is working as a Lecturer and currently Head of Department of Electrical/Electronics Engineering, Eastern Polytechnic Port Harcourt, Rivers State, Nigeria.



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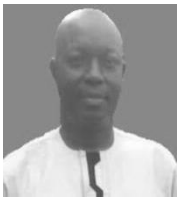

Prof. Paul Ukoro Asogwa had his secondary education at St. Teresa's college, Nsukka and proceeded to the University of Nigeria, Nsukka where he obtained Bsc, Msc degree and a PhD in Physics and Astronomy department specializing in solar energy Physics. He joined the services of UNN in 1994 and is currently a professor of solar Energy physics/ materials science at the Department of physics and Astronomy University of Nigeria, Nsukka. He is an avid researcher who has participated in many National and international Conferences/Workshops and a member of many scientific organization.

