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Physics of Intense, High Energy Radiation Effects

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Abstract

This document summarizes the work done in our three-year LDRD project titled ”Physics of Intense, High Energy Radiation Effects.”

Acknowledgment

The project team thanks David E. Beutler, Kenneth E. Kambour, Robert Kaplar, Michael L. McLain, Peter A. Schultz, David B. Seidel and C. David Turner for their help with this project. We also than Ann Mattsson, a member of the original project team, for her involvement during the first portion of this project.

Contents

1 Introduction

This LDRD is focused on electrical effects of ionizing radiation at high dose-rates. One major thrust throughout the project has been the radiation-induced conductivity (RIC) produced by the ionizing radiation. Another important consideration has been the electrical effect of dose-enhanced radiation. This transient effect can produce an electromagnetic pulse (EMP).

The unifying theme of the project has been the dielectric function [1]. This quantity contains much of the physics covered in this project. For example, the work on transient electrical effects in radiation-induced conductivity (RIC) has been a key focus for the work on the EMP effects. This physics in contained in the dielectric function, which can also be expressed as a conductivity. The transient defects created during a radiation event are also contained, in principle. The energy loss lead the hot electrons and holes is given by the stopping power of ionizing radiation. This information is given by the inverse dielectric function. Finally, the short time atomistic phenomena caused by ionizing radiation can also be considered to be contained within the dielectric function.

During the LDRD, meetings about the work were held every week. These discussions involved theorists, experimentalists and engineers. These discussions branched out into the work done in other projects. For example, the work on EMP effects had influence on another project focused on such phenomena in gases. Furthermore, the physics of radiation detectors and radiation dosimeters was often discussed, and these discussions had impact on related projects.

Some LDRD-related documents are now stored on a sharepoint site (https://sharepoint.sandia.gov/sites/LDRD-REMS/default.aspx).

In the remainder of this document the work is described in catergories but there is much overlap between the atomistic calculations, the continuum calculations and the experiments.

2 Overall

This LDRD has focused on the RIC and the transient dose-enhanced electrical response of a material during the entire duration of this project. Our first project meetings focused on the electrical effects expressed in terms of radiation-induced conductivity (RIC) for a bulk material. The physics of defects was introduced to understand these phenomena [2, 3]. Calculations focused on insight were performed using the software Radiation Effects in Oxides and Semiconductors (REOS) [2, 3]. Some of these calculations were published in a paper about dose-rate effects in silicon dioxide [4]

Subsequent calculations revealed another role involving defects. Such defects may create trapped charge that could persist for periods of time long compared with the duraction of an experiment. These effects may explain some history effects that are well-known but poorly understood [5, 6]. The focus on the effects of defects also led to calculations to understand their role in radiation effects on capacitors [7, 8].

Continuum REOS calculations were performed to understand RIC data for silicon dioxide samples [9]. These calculations strongly suggested that defects are controlling the RIC in these samples.

One theme of the work in the last year has been the role of transient defects. These are defects that are created during the radiation event but they heal very quickly. Such defects can be expected in wide bandgap insulators. These ideas have been explored with REOS calculations in which hot carriers, excitons and lattice heating are all considered; they were also explored in MD calculations using LAMMPS [10, 11, 12].

As the project evolved, the calculations led us to conclude that there is a need for more physics in calculations that focus on EMP effects caused by dose-enhancement effects. Our work strongly suggests that the present method for empirical inclusion of these effects does not contain enough physics. This method captures the radiation effects in terms of its effect on electrical conductivity. This information is obtained from experiments on various materials. It is included in the EMP calculations as a table of information. At Sandia, this method is used in the Emphasis code that is part of the RAMSES suite of software.

We advocate a new method in which the physics of electrons, holes and traps is included in calculations that are compared with RIC data [13]. This comparison would be used to validate the model. This model would be used in subsequent calculations of the EMP effects.

Calculations that focus on the transient dose-enhancement effects of foam for have been performed. The goal of these calculations has been to explore a very simple material model for the foam material. In this model, the foam consists of a gas, the insulating solid material of the foam and then a gas. These calculations are discussed in this document.

The present results suggest that transisient polarization effects may be very important. If this is true, it is hard to capture such physics in a simple conductivity model.

3 Theoretical Foundation

The theoretical foundation for some key issues is discussed in this section.

3.1 Time-Dependent Density-Functional Theory (TDDFT) Calculations

The goal of these calculations is to model the time-evolution and conductivity of a highly excited, non-equilibrium localized electron disturbance in an insulating material such as SiO_2. The hot electrons are expected to scatter inelastically off bound electrons creating a substantial number of electron-hole pairs and eventually relaxing to a thermal distribution of excited carriers before transferring significant amounts of energy to the vibrational modes of the lattice. Our goal is to understand the conductivity of the insulator with highly-excited electrons. When the system is out of equilbrium, an externally applied field can induce a macroscopic current despite the ground-state system being an insulator. Here, we focus on the quantum effects on the electrons and their response in a highly idealized piece of SiO_2 α-quartz and use time-dependent density functional theory (TDDFT) to model the dynamics.

Time-dependent density functional theory provides a convenient framework to describe the many particle interacting electron dynamics [14, 15, 16, 17, 18]. In the KS orbital picture, the KS wave-functions evolve as follows:

$$\frac{d}{dt}\phi_j(t) = -i\hat{H}_{KS}(t)\phi_j(t)$$

with ϕ is the $j-$th KS orbital. $\hat{H}_{KS}$ is the Kohn-Sham (KS) Hamiltonian, $\hat{H}_{KS} = -\nabla^2 + v_{ext}(r) + v_H(r) + v_{XC}(r)$. The first term on the right in the momentum operator, the next is the nuclear coulomb potential, followed by the classical Hartree potential, and finally the exchange correlation potential. The last term is the core of the density functional theory and subsumes the many-body effects allowing the treatment of the system using auxiliary non-interacting KS orbitals. Numerically, the Crank-Nicholson time-evolution method preserves unitary and time-reversal symmetry. At a subsequent time step, the wave-functions are

$$\psi_j(t+\Delta t) = L^{-1}B$$

with

$$L = 1 + i\frac{\Delta t}{2}\hat{H}\left(t + \frac{\Delta t}{2}\right)$$

and

$$B = \left(1 + i\frac{\Delta t}{2}\hat{H}\left(t + \frac{\Delta t}{2}\right)\right)\psi_j(t).$$

This propagation involves two matrix multiplies and an matrix inversion. We can estimate the operator, $\hat{H}\left(t + \frac{\Delta t}{2}\right)$, according to an extrapolation of $\hat{H}_{KS}$ from several previous time steps.

To obtain the dielectric function assuming $q \to 0$, we apply a sudden change in the vector potential. The dielectric function is directly related to the system response:

$$\frac{1}{\varepsilon(\omega)} = \frac{\int_{0^+}^{\infty} dt\ \exp(i\omega t - \eta t)\frac{dA_{tot}(t)}{dt}}{\int_{0^+}^{\infty} dt\ \exp(i\omega t - \eta t)\frac{dA_{ext}(t)}{dt}}.$$

We obtain this from $\vec{D}(\omega) = \varepsilon(\omega)\vec{E}(\omega)$ using $\vec{E} = -\frac{d}{dt}\vec{A}(t)$ and $\vec{D} = -\frac{d}{dt}\vec{A_{ext}}(t)$.

We suppress vector notation. A δ function spike is a typical model of the applied field E. The electric spike is given by the vector potential, $A_{ext}(t) = A_0\theta(t)$ and can be used to extract the dielectric response. In this case,

$$\frac{1}{\varepsilon(\omega)} - 1 = \frac{1}{A_0}\int_{0^+}^{\infty} dt\ \exp(i\omega t - \eta t)\frac{dA_{ind}(t)}{dt}.$$

Thus, to find the dieletric function, we need to find $A_{ind}(t)$, the induced polarization vector potential, given an impulse A_0. Note, that the intial state of the system can be a quasi-equilibrium state. Thus, we have a way to calculate non-thermal dielectric functions. This differs from finite temperature response functions arrived at via the Kubo-Greenwood method in two important ways. First, we are not restricted to equilibrium situations. Second, we are including TDDFT effects beyond the non-interacting orbital or RPA picture. In energy space parlance, this amounts to including the effects of a exchange-correlaiton kernel. By including $v_{XC}(r,t)$ we have a framework in which to extent to nonadiabatic effects. Note that ε is in general a dyadic tensor. We restrict ourselves to a perturbing field oriented in the z axis and only collinear response. Then we find A_{ind}.

The introduction of the vector potential, $\vec{A}(t) = \hat{z}(A_{ext}(t) + A_{ind}(t))$, will modify the KS equation we use to propagate the orbitals. To find the new equation, apply gauge transformation on the wave-function:

$$\phi_i(r,t) = \exp(iA\cdot r)\psi_i(r,t)$$

and find

$$\hat{H}'_{KS} = -\nabla^2 + v_{ext}(r) + v_H(r) + v_{XC}(r) + i\vec{A}(t)\cdot\nabla + \frac{1}{2}\vec{A}^2(t)\hat{S}.$$

This Hamiltonian is used to time-propagate the KS equation.

We now need a separate differential equation for the time-evolution of the vector potential, A. The average polarization per unit cell and volume averaged current.

$$\frac{d}{dt}P(t) = \frac{1}{4\pi}\frac{d^2}{dt^2}A(t) = \frac{1}{\Omega}\int_{\Omega} d^3r\, j(r,t)$$

using $\vec{P} = -\frac{1}{4\pi}\frac{d}{dt}\vec{A_{ind}}(t)$.

$$\frac{\Omega}{4\pi}\frac{d^2}{dt^2}A_z(t) + i\sum_{occ.}\langle\phi_i|\nabla_z|\phi_i\rangle + A_z N_e = 0$$

.

Our plan is to implement TDDFT time evolution in the electronic structure code, SeqQuest. SeqQuest uses pseudpotentials and a high quality local orbital basis of contracted Gaussian functions in a linear combination of atomic orbitals (LCAO) approach to solve the Kohn-Sham equations fully self-consistently. Most of the necessary matrix elements are already available in Sequest; however, it is possible that elements of the form

$$\langle \phi_i | \nabla_z | \phi_j \rangle$$

must be coded. This method will involve a further modification to quest to allow vector potentials.

There are two major theoretical challenges in simulating this system within time-dependent density functional theory (TDDFT). First, a computational engine must be constructed to time-evolve the Kohn-Sham (KS) equations. Second, a physically reasonable initial state must be chosen. Since the KS equations do not in general represent physical quantities, the initial state must be presented as a physical initial charge distribution and mapped back unto the KS orbitals.

First, we find the initial state. For each k point in the Brillioun zone, we solve a Schroedinger-like KS equation.

$$\hat{H}_{KS}(k)\phi_i(k) = \varepsilon_i(k)\phi_i(k).$$

Since the basis set is non-orthogonal, we would be solving $\mathbf{H}(0)\mathbf{C}(0) = E_0\mathbf{S}\mathbf{C}(0)$ with $\mathbf{S}$ an overlap matrix, $\mathbf{C}$ is the coefficient and weight vector, and $\mathbf{H}$ is the Hamiltonian matrix. However, once the initial eigenvector matrix, $C(0)$, is found then we define $V(t) = \sqrt{n_i(0)}C_i(t)$ the occupation weighted eigenvector and time evolve according to the matrix multiply $H_{ij}(t)V_j(t)$.

Additional assumptions in this early implementation are fixed nuclear position. We will start by using the adiabatic local density approximation for the time-dependent potential. This amounts to assuming that the exchange-correlation potential at any given time snapshot can be calculated from the instantaneous density by appealing to a uniform reference system. The limitations of this approximation are that the many-body potential will not retain memory of earlier system states.

The initial state is chosen as a linear combination of virtual KS orbitals reproducing the density of the desired hot localized electron. There is no rigorous relationship between the eigenvalues of the KS orbitals and the local temperature. An approximate value can be obtained through a single evaluation of the DFT energy on the modified density.

There is no unique way to set up this loosely defined initial state. The most naïve approach would be to simply charge fill one virtual KS orbital. This however would result in a delocalized excitation, but is likely to be the first approach since it provides a test of the time-evoluation algorithm. A pure state trivially time-evolves using the exact KS Hamiltonian; however, the practical approximate KS potential will likely cause this pure state to time-evolve and artificially thermalize. A measurement of the time-scale of this thermalization will provide a time bound beyond which we can not longer trust a practical approximate application of TDDFT.

A more realistic initial state is an ensemble of KS orbitals. We should create an initial state as Wannier distribution of conduction states localized at a particular atomic site in a supercell and its images in other supercells. This gives an effective hot electron density.

In order to asses the time-dependent properties of the system, we can compare the initial and time-dependent densities of a super cell. The difference should be minimal. A more resolving comparison would be to re-express the time-dependent density matrix in the initial basis and to project the time-evolved basis onto the initial. The difference gives a measure of thermalization.

$$\delta(t) = \text{Trace}(\rho_{KS}(t)\rho_{KS}(0)).$$

Note $\delta(0) = N$ but $\delta(t) \neq N$ for a mixed ensemble.

3.2 Molecular Dynamics Calculations

The key advance in our use of molecular dynamics calculations focused on the two-temperature method (TTM) for inclusion of ionization effects into such calculations. The basis for our work has been described in papers focused on ionization effects in metals [19, 20]. In this approach, the ionization portion of the energy loss of a particle heats the electronic subsystem. These hot electrons subsequently heat the lattice subsystem. This heat in the lattice can contribute to defect creation. This effect was considered in a series of calculations [11, 12].

During this LDRD, a new approach was developed that should allow this method to be used for insulators. In this approach, a continuum calculation is used to define the density and temperature of the electrons and holes created by the ionizing radiation. This plasma is allowed to cool as these electrons and holes recombine. This recombination, described by a carrier lifetime, generally involves defects. It can in fact involve defects created during the cooling process.

3.3 Defect Creation Mechanisms

The ionizing radiation creates electrons and holes that cause electrical effects. Such effects tend to involve carrier recombination at defects. In addition, the ionizing radiation can also create defects. Such defects may be heal rapidly. For example, atoms displaced from a lattice site may return to that lattice site very rapidly.

The primary mechanisms described in the literature are the thermal spike, excitons and the Coulomb explosions [21]. These mechanisms may all contribute to defect creation in insulators.

3.4 Transient Dose-Enhancement Effects

Ionizing radiation that strikes an interface between dissimilar materials can generate an electromagnetic pulse (EMP) [22, 23]. The basic effect involves an electrical current consisting of secondary electrons from one material that are stopped in the other material [24, 25, 26]. This current of secondary electrons is also called dose-enhancement. This current creates the EMP that can produce disruptive effects elsewhere. In addition, in a device connected to a power supply, the transient

current drawn in response to this charge flow can have harmful effects. These EMP effects will be called system-generated EMP (SGEMP) throughout this document.

These phenomena have been known for several decades of time. The usual analysis involves a calculation of the transient response of a material to an electrical charge. In such calculations, the response of the insulating material is computed in terms of the transient radiation-induced conductivity (RIC) [24, 25, 26]. Furthermore, in applications, the conductivity is assumed to reach the steady-state value very quickly. This assumption enables the use of empirical RIC data for the computation of the EMP effects.

4 Continuum Calculations

The REOS continuum calculations are involved in all aspects of this LDRD. In a sense, these calculations are the bridge between the atomistic calculations and the experiments.

These calculations include electron and hole transport, exciton effects and chemical reaction reactions between defect species [27, 28]. In addition, the energy of hot electrons and holes is tracked following exposure to ionizing radiation.

4.1 Dose-Rate Effects

Early in this project, calculations were performed that focus on the physics at various dose rates to obtain insight about the role of defects. These calculations revealed that the presence of electron and hole traps could lead to very non-linear dependence on radiation dose-rates. In particular, these calculations showed that the progression of behavior to be expected began with non-linear, then linear and then finally non-linear again at the highest dose rates [4, 29]. These calculations led to a unified point of view concerning very low dose-rate phenomena, to be discussed, and the very high dose-rate phenomena that occur in a RIC experiment at high dose-rates.

4.2 Enhanced Low Dose-Rate Sensitivity (ELDRS)

The physics developed to handle dose-rate effects was applied to the problem of enhanced low dose-rate sensitivity (ELRDS) that leads to reduced radiation effects at very low dose rates in bipolar junction transistors [30]. Recent work has led to the hypothesis that elevated concentrations of molecular hydrogen can be associated with this effect. Accordingly, a mechanism involving chemical reactions of molecular hydrogen was explored using REOS calculations. The results show a dose-rate dependence that compares well with the data [4, 30].

The dose-rate dependence arises because not all the holes created by ionizing radiation are able to react with molecular hydrogen at high dose. In this case, all the available hydrogen at these sites is consumed. At low dose-rates, in contrast, the hydrogen is supplied rapidly enough that each hole is able to crack molecular hydrogen to release a proton.

This hydrogen cracking mechanism has been supported by recent calculations that suggest that an oxygen vacancy defect, when it has trapped a hole, is responsible for the release of atomic hydrogen.

In future work, it is expected that these calculations will be pursued in two distinct directions. In one direction, the physics and reactions will be made as simple as possible to enable rapid calculations to compare with data. In another direction, more physics will be included in the calculations. For example, other hydrogen release mechanisms, which were discussed but not implemented in the earlier paper, will be used in a new set of calculations. One goal of this effort

is to examine a discrepancy between the calculations and data that are independent of dose rate.

4.3 RIC Experiments

Calculations to interpret radiation-induced conductivity (RIC) experiments were done [9]. These calcultions focused on the control of RIC by hole traps in SiO_2.

4.4 Transient Dose-Enhancement Effects

A series of calculations focused on transient dose-enhancement effects. These calculations compared REOS results with a simple analytic model. These results show that a simple model can be used for insight for case in which the geometry is very simple [31].

An extensive series of calculations was done that included the effects of defects in controlling the RIC response of a material. These calculations revealed that the temporal behavior of this data can be very complicated. This suggests that a stead-state experiment is not likely to be adequate in capturing the time-dependent conductivity that is necessary for an EMP calculation. In this LDRD, our calculations in which the consequences of defects were taken into account revealed that these assumptions are not true in general. These calculations showed that RIC data very often had much temporal variation before steady-state was reached.

Our results suggest that an improved approach would use a physics-based model for the RIC in which much of the physics depends on the defects in the material. The physical parameters of this model would be obtained by comparison with transient and steady-state RIC data. Then this model would be used to described the transient conductivity in an EMP calculation.

4.5 Foam

The calculation of transient dose-enhancement effects in foam is very challenging because the structure is not known. In this LDRD, a very simple structure was hypothesized. The intention of this prototypical structure is to capture the main effects.

The prototypical structure captures the essence of a foam in its radiation effects. A real foam has large variations in material density. For our purposes, it can be regarded as consisting of cells whose walls are composed of a solid material and whose interior is composed of gas. To represent the essence of this complicated structure, the simple structure consists of two cells separated by a wall.

The electrical effects for this structure involve the capture of the secondary electrons in the wall. These electrons are assumed to create electron-ion pairs in the gas regions during their slowing down process.

The calculations for this structure explored the effects of variation of the conductivity for the gas. The low conductivity gas produced a transient current that became smaller during the electrical pulse. This reduction was caused by the electron-ion pairs in the gas. The gas with the higher conductivity produced a similar transient reponse but in this case the current reversed sign before the end of the radiation pulse.

4.6 Negative-Bias Temperature Instability (NBTI)

Other calculations were performed to explore charge collection by the defects in the silicon dioxide. These calculations were linked to experiments done at the Air Force Weapons Laboratory (AFRL) by R. A. B. Devine and others [32, 33, 34].

5 Atomistic Calculations

5.1 Time-Dependent Density-Functional Theory (TDDFT) Calculations

In this section, we report progress in the development and utilization of time-dependent density functional (TDDFT) calculations at Sandia. TDDFT is a powerful tool used to model the effects of energetic electrons on the atomic lattice. It treats the electrons according to the quantum mechanical Schrödinger equation with heavy nuclei, and thus, no empirical fits to data are required. The electrons are time evolved allowing the dynamic modeling of the electron-nuclear interactions. TDDFT's advantage over a full solution of the quantum mechanical problem is its great reduction in computational cost while preserving much of the quantum mechanical accuracy and generality.

A mature and reliable capability to perform TDDFT calculations on solids is not yet available at Sandia. In order to establish this capability, we have implemented several new features into SeqQuest, a highly scalable, finite basis set code that is currently maintained at Sandia. One of the major advantages of using a finite basis set code is that the basis functions are highly optimized and the pseudopotential is local. Several computational challenges that arise when combining TDDFT with non-local pseudo-potentials can be avoided.

The TDDFT calculations simulate the time evolution of both electron and nuclear systems with the electrons moving according to a Schrödinger-like equation and the nuclei obeying a classical equation of motion. Bonding and inter-atomic interactions are naturally handled in this formulation through the Schrödinger-like equation obviating the need for effective potentials that would be required in a classical molecular dynamics simulation. TDDFT has been successfully carried out on many finite systems (atoms, molecules, clusters) with coupled ion-electron motion for many years. However, applying TDDFT to solids introduces several challenges. Primary amongst these is the issue of long-range polarization effects in otherwise periodic repeat cells. This is best handled by inclusion of the classic electro-magnetic vector potential, A, as an auxiliary quantity in the calculation. In this way, a separate equation of motion must be evolved for the vector potential, and the electronic Hamiltonian now depends on the vector potential in a complicated fashion.

Implementation of this real-time version of TDDFT requires the introduction of the vector potential and momentum overlaps to the SeqQuest code. Over the course of this LDRD, we have implemented the momentum overlaps and the vector potential. The vector potential has been added to the code, and it's time evolution is allowed for and is calculated through integration of its equation of motion. The momentum matrix elements that are essentially overlaps between differing basis functions with a gradient operator between has also been implemented.

The TDDFT algorithm to be used in SeqQuest is as follows. 1. Calculate initial KS ground state wave-function 2. Repopulate the density matrix according to non-equilibrium distribution assuming A=0 everywhere. 3. Propagate the new density matrix over time according to TDDFT. This requires calculation of the density and Hamiltonian at each time step but not repopulating the density matrix. 4. Analyze the new density matrix at each time step. 5. Perturb the system with a laser pulse (vector potential step function in time). 6. Calculate the dielectric function of the

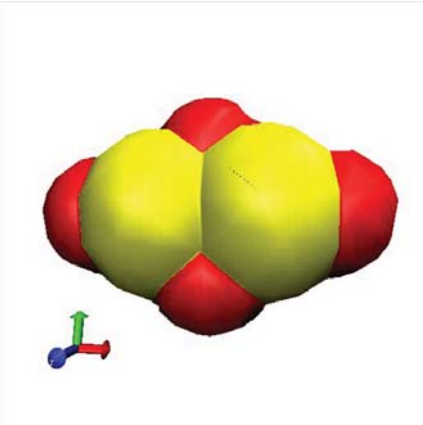

Figure 1. Si_2O_4 relaxed nuclear positions prior to electron heating. TDDFT calculations using OCTOPUS with a PBE-GGA functional.

system at quasi-equilibrium.

Knowledge of the time evolved vector potential in response to certain pulse stimuli, allows the calculation of the dielectric response of the material. This post-processing step has also been implemented.

Another major challenge to practical implementations of TDDFT is the choice of electronic time integrator. A necessary requirement for any time integrator is that it be unitary, stable, and allow for longer time steps. A great deal of work has established that Crank-Nicholson integrators are among one of the most robust methods used for finite systems. It is unclear what the best choice for solids is. We have implemented and tested these CN integrators for several model systems using the Mathematica software determining that CN is a reliable integrator when dealing with a Gaussian basis as used in SeqQuest.

As part of this LDRD, a publically available TDDFT code, OCTOPUS, for finite systems has been compiled and run to examine the effects of hot electrons in clusters and molecules of SiO2. Several illustrative simulations demonstrate that energetic electrons can stretch and alter bond lengths and with sufficient energy can explode the clusters. Figures 1 and 2 illustrate the initial and final states of a small silica cluster wit hot electrons.

DFT and TDDFT can be used in addition to giving fundamental insight into small-scale phenomena to provide parameters for larger scale and more course grained simulations. Along these lines, ground-state DFT calculations were used to model a fictitious system of hot electrons in a frozen SiO2 lattice. Information from these calculations provided inputs for TTM of radiation damage in insulators resulting in a JCP paper.

Full implementation of TDDFT in SeqQuest requires continued support. In particular, the nuclear equation of motion must be implemented. For the most part, this can be included through

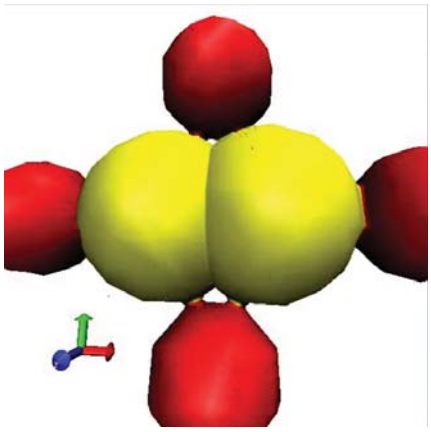

Figure 2. Si_2O_4 relaxed nuclear positions subsequent to electron heating.

modification of the pre-existing geometry optimization routines. However, before such implementation should be considered, the electronic contributions must be thoroughly tested. This is possible by verifying that the TDDFT response to optical stimuli agrees with other published data of the dielectric function of various materials. The most likely failure point is the implementation of the momentum overlaps. SeqQuest is already highly optimized code and direct implementation of the algorithm into the existing code structure allows the possibility for memory management and coding issues. The cleanest test of the momentum overlaps would require comparison to similar elements calculated in pre-existing and well tested codes. However, few codes are capable of calculating these elements, and at this time, no Gaussian-based code for solids is capable of this. A second test involves the creation of a fairly complicated Mathematica or post processing script to convert output data from SeqQuest into these elements.

Additional work in support of understanding electronic radiation effects in insulators involved travel to national conferences to discuss results and obtain feedback from experts in the field regarding progress of this effort. In particular, two trips are worth detailing here. Several members of the team participated in a workshop on elementary excitations in solids last fall. The focus was on the development of computational methods like TDDFT to provide reliable predictions of the electronic response of solids. In particular, details of former pre TDDFT calculations of similar hot electron effects were discussed. Another important visit was to UC Irvine where much of the detail of this LDRD was discussed. Suggestions about how to implement this method to solids and possible issues with the vector potential formalism were discussed.

The key advances in TDDFT are the following: 1. Momentum operator / Berry's phase matrix elements have been coded into Sequest. 2. A time integrator for the vector potential has been coded and tested. 3. A Mathematica script has been developed to model a Kohn-mode motion of an electronic density as a testing case for integrators and local basis set methods in TDDFT. 4. Some proof of principle calculations using the octopus code illustrate how heated electrons will effect the geometry of atoms in cluster models. Some density functional advances have been

described in a forthcoming publication [35] and were presented at the Kavli Institute of Theoretical Physics workshop.

Further testing is required before these TDDFT methods are fully available at SNL.

Some density functional advances have been described in a forthcoming publication [35].

5.2 Molecular Dynamics (MD) Calculations

Several types of molecular dynamics (MD) calculations were performed to ascertain the effects of hot electrons and holes. As discussed previously, these calculations used a two-temperature MD method in which carrier temperatures were allowed to be governed by the energy dissipated by the ionizing particle [11].

One consideration for the transient electrical effects was the notion that the ionization process could produce transient defects that would heal soon after the ionization event. These ideas were explored in both continuum and atomistic calculations. In atomistic calculations, these ideas were first explored for a Lennard-Jones crystal.

The TTM was also applied to an exploration of transient defects created in a quartz crystal [12].

Finally, several calculations were done to examine the Coulomb explosion mechanism [36]. These calculations included the motion of electrons together with the motion of the atoms in a crystal with a Lennard-Jones force field. These calculations illustrated the conditions that would lead to large effects caused by this mechanism.

6 Experiments

The success in comparing our calculations with RIC data from for silicon dioxide led to a new experiment. As stated above, the comparison suggested that defects control the RIC data [9]. In these new experiments, the RIC for small silicon dioxide capacitors was measured [9, 7]. However, these devices were too small to produce useful data.

Another set of new experiments focused on RIC from sapphire. These experiments have been completed. The data is being analyzed.

The work in this LDRD stimulated Mike McLain, a regular attendee of the meetings, to submit an LDRD focused on RIC experiments on Kapton, an important insulating material.

7 Other

Some simulations were focused on radiation detection. This issue arose because there is an ongoing concern about performance of ionizing radiation detectors at high doses.

8 Impact

This LDRD suggests that additional physics should be included in calculations of SGEMP.

This LDRD led to the inclusion of the two-temperature model (TTM) in LAMMPS MD calculations [11]. This feature has been extensively used by the LAMMPS user community.

The work on this LDRD helped guide our thinking about 3D radiation detectors. As a consequence, a new LDRD project (10-0702) was funded to develop such gamma-ray detectors. The present design relies on detectors for which Xenon is the active material.

The research in this LDRD is expected to be helpful in understanding why there is a discrepancy among various classes of radiation detectors used at Sandia. The REOS capability for handling hot electrons, hot holes and excitons is expected to be helpful for radiation detector calculations.

The REOS capability for carrier heating effects has led to its consideration for use in an ongoing LDRD focused on laser damage of semiconductor devices.

9 Future Work

We suggest that some of the ideas developed in this project may lead to improved calculations of SGEMP effects in solid insulators. Our findings suggest that a physics based model should be developed. In such a model, the properties of defects would be obtained by comparing calculated RIC with data. Then this physics-based model should be compared with experiments on simple structures whose properties can be readily calculated.

Our work on foam suggested that an experiment could be performed on the idealized model of the foam to test the calculations for this model. Such experiments would be a good step prior to more refined calculations to be compared with experiments on real foam structures with a simple geometry.

Some portions of this project may lead to a collaboration with Michael King, a summer intern from Vanderbilt University. He expressed interest in exploring the Coulomb explosion mechansim in his upcoming thesis work.

Our work may provide a theoretical foundation for an improved two-temperature model that is applicable to insulating materials.

10 Manuscripts, Publications and Presentations

10.1 Manuscripts

Several unpublished manuscripts to guide our work were created [2, 3, 8, 27, 28, 9, 5, 6, 13, 31, 29].

10.2 Publications

Several publications were created during the course of this LDRD [4, 30, 32, 33, 1, 34, 11, 12].

10.3 Presentations

Several presentations were made as a consequence of this LDRD project [37, 38, 39, 40, 41, 7, 42].

11 Summary

Most of the LDRD effort has been focused on transient electrical effects such as RIC. The remaining work involves the assimilation of the various portions of this project to produce a more coherent description of the phenomena.

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