**IT4Innovations 5th Open Access Call**

To be submitted before: 2015-04-03 23:59:59

**Name of the project: Optical properties of HfO2 using many body perturbation theory  
Number of core hours requested: 100000**

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**Research area:** Materials Science, Physics

**Popular abstract:**

HfO2 is attracting a lot of attention as a high-k dielectric for electronic applications as well as an optical material for applications such as antireflective coatings, heat-mirrors, or laser mirrors. It has been studied both experimentally and theoretically, however, the calculated and experimental optical data do not agree very well.  
  
We have encountered this problem during our previous projects, in which we established a database of optical properties of TiO2-based binary oxides (Ti1–n*X*nO2) using quantum-mechanical calculations based on the Density Functional Theory employing modified Becke-Johnson potential. While for some materials, e.g. Ti1–nSinO2, the calculated results match well with experiment, for some of the mixtures (for example with Hf) the agreement is far from perfect. It has been argued that is caused by the approximations commonly used to reduce high computational costs.

In this project we will focus more in depth on the binary oxides, namely on HfO2, and will explore more advanced state-of-the-art techniques such as hybrid potentials or many body perturbation theory to describe processes in these materials more accurately, and to help explain discrepancies in our previous works.

Modern computational methods allow for a significant reduction of experimental efforts and costs by providing material trends and selection rules. The computational power of the Salomon supercomputer will enable us to use highly sophisticated approaches, which are computationally too expensive for usage on standard computer resources. All this will assist our experimental efforts focused on application-oriented materials development.

**Scientific readiness:**

**Aims and objectives**

The aim of this project is to calculate properties of selected binary oxides such as monoclinic or cubic HfO2, using state-of-the-art approaches including hybrid potentials and many body perturbation theory.

A thorough study of band structure and optical properties will be conducted, and results will be critically compared with standard DFT potentials, with novel and promising semi-empirical modified Becke-Johnson (mBJ) potential [1], and to experimental results. This will help to better understand limits of the aforementioned methods and to clarify, if the sources of discrepancies with respect to experiment lie in the calculation of the electronic structure (e.g., used functional), or if the problems originate in its post-processing during the calculation of optical properties (e.g. shortcoming of an independent particle approximation often used for calculating the dielectric function).

**Methods and state-of-the-art**

An all-electron fully relativistic DFT package WIEN2k will be used in this project. It is based on the full-potential (linearized) augmented plane-waves + local orbitals method (LAPW+lo) [2], one among the most accurate schemes for band structure calculations. It has an implementation of screened hybrid functional based on the Yukawa potential [3]. Screened hybrid density functionals are the most commonly used extension to the standard LDA and GGA approaches and offers much better precision with regards to lattice constants, electronic structure and band gaps [4,5]. Another suitable approach for studying excited-state properties of extended systems is the Green’s functions based *GW* approximation, or its non-self consistent variant (*G*0*W*0) [6]. This is one of the most precise methods for prediction of band gaps [7]. For the calculation of optical spectra including excitonic effects, a method based on the Bethe-Salpeter Equation will be used [8]. Owing their calculational costs, these methods are not applicable to large systems, and hence only binary dioxide systems will be studied.

**Impact and outlooks**

While there are multiple works reporting optical properties using GW and BSE for the most commonly used dioxides (e.g. TiO2 [9,10], SiO2 [11]), for the less common binary oxides such as HfO2, or ZrO2, the many-body effects on the optical response are not yet so well studied, and hence the proposed work represents a novel and scientifically interesting area.

By comparing state of the art optical calculations on binary oxides with standard LDA, GGA and also mBJ calculations, we will shed light on the role of the excitonic effects in optical spectra. This will help us to better understand our previous works, and to explain discrepancies with respect to experimental measurements. Even though the proposed methods are not suitable for large systems due to their high computational costs, the results from binary system will be still extremely valuable for analysis and interpretation of results also more complex systems.

Calculated optical properties together with the underlying band structure will additionally help to understand nature of electronic transitions, hence facilitating a development of advanced dispersion models [12,13]. Such models are important for routine and accurate evaluation of optical measurements and characterization of experimental samples.

**Computational readiness:**

**Computational approach, parallelization and scalability:**

WIEN2k software used for this project is written in FORTRAN90 and requires a UNIX system (programs are linked together via C-shell scripts). WIEN2k does not require any special libraries, but it uses optimized BLAS and LAPACK libraries if present. It is mostly tested and optimized for the Intel proprietary compiler, but can be built with GNU compiler collection as well.

WIEN2k has a built-in parallelization scheme (so-called k-point parallelization), where subsets of the k-point mesh (sampling the Brillouin zone) are distributed to different processors, and subsequently combined. There are some parts of the main self-consistent file (SCF) cycle which are not parallelized, however their computational costs are negligible compared with the rest of the cycle.

Additional parallelism is also possible at the library level (inside MKL or OpenBLAS).

For large cases or calculation of GW or BSE a MPI fine-grained parallelization is used when necessary, e.g., for memory limited problems or where there are fewer k-points than available processors.

The applicant owns a valid academic license of Wien2k. GW and BSE implementations for Wien2k are, however, not part of the distributed version, but are available on request from authors. If this will constitute any obstacle, we would use an open source alternative for the GW and BSE calculations as implemented in the Exciting code [14], which is available under GNU GPL license. Since Exciting uses the same scheme as Wien2k (all electron full potential LAPW+lo), it is presumed that all following description and estimates apply to Exciting as well.

**Computational resources:**

Resources were estimated on the basis of previous calculations with simple test systems. Depending on the difficulty of the calculations a variable number of calculations is planed. Main focus of this study will be calculations for HfO2 however if CPU time permits (if this systems turns out to be fast to converge with regard to numerical parameters such as k-grid size) more calculations will be performed with ZrO2 or other dioxides. This is to mitigate risks associated with uncertainty in calculations run time.

Planned amount of calculated structures: 2-5  
Core hours needed for hybrid calculations: 10000  
Core hours needed for the G0W0 calculations: 30000  
Core hours needed for BSE calculations: 60000

Total CPU hours needed: 100000

**Economic readiness:**

**Socioeconomic impact:**

In the long run, better understanding of HfO2 optical properties will lead to a development of novel optical components such as multilayer antireflecive coatings, interference filters, laser mirrors, heat mirrors or waveguides with greatly improved optical performance.

Development of dispersion models based on this work will open new possibilities in optical characterization of HfO2 materials. Those models will even enable to obtain material properties that are not usually associated with optical methods, such as mass density or crystallinity. It will reduce need for other experimental methods and lower costs of further experimental research.

**Operational costs:**

This work will be done in parallel to experimental studies performed within the frameworks of funded projects of the CEITEC Plasma Technologies group (group leader Prof. Lenka Zajíčková). The calculations will be done within the PhD project of Pavel Ondračka (principal investigator of this proposal), whose scholarship is paid from the Masaryk University and CEITEC resources. There are no other financial resources available for the PI to cover the operational costs related to the use of the supercomputer.

**Amortization and renewal:**

As described above, there are unfortunately no free financial resources available towards amortization and renewal of the cluster hardware.

**References:**

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1. Comma separated list [↑](#footnote-ref-2)