**IT4Innovations 3rd Open Access Call**

To be submitted before: 4.4.2014

**Name of the project: Optical properties of TiO2 based alloys from first principle calculations  
Number of core hours requested: 104000**

**Name and surname of primary investigator: Pavel Ondračka**

**Affiliation of primary investigator:** Department of Physical Electronics, Faculty of Science, Masaryk University (PhD student, Advanced Materials and Nanosciences program)  
Central European Institute of Technology, Masaryk University

**e-mail:**[pavel.ondracka@gmail.com](mailto:pavel.ondracka@gmail.com)

**Names and surnames of other investigators**[[1]](#footnote-1)**:** David Holec a, Daniel Franta b,c, Lenka Zajíčková b,c

**Affiliations of other investigator**1**:**a Department of Physical Metallurgy and Materials Testing, Montanuniversität Leoben  
b Department of Physical Electronics, Faculty of Science, Masaryk University  
c Central European Institute of Technology (CEITEC)

**e-mail**1**:**[david.holec@unileoben.ac.at](mailto:david.holec@unileoben.ac.at), [franta@physics.muni.cz](mailto:franta@physics.muni.cz), [lenkaz@physics.muni.cz](mailto:lenkaz@physics.muni.cz)

**Research area:** Materials Science, Physics

**Popular abstract:**

Titanium dioxide (TiO2) thin films are extensively studied because of their interesting optical, electrical and chemical properties. TiO2 is a wide band gap semiconductor, hence it is transparent over a wide range of wavelengths. TiO2 has a high refractive index and high static dielectric constant. The optical properties of TiO2 motivate research on applications such as multilayer antireflective coatings, interference filters, or waveguides.

Regardless its exciting properties, the current material quality of TiO2 thin films still limits their performance. Columnar morphology generally observed in TiO2 coatings leads to increased optical losses and degradation of its insulating behavior. It has already been shown that mixing Si-O and Ti-O bonds leads to more homogeneous material (i.e. suppression of the columnar structure of pure TiO2 films) and an increased fraction of amorphous phase. Consequently, controlled implantation of Si into TiO2-based materials is a perspective way how to alter the dielectric constant and refractive index, and simultaneously to improve insulating properties. Additionally, it is anticipated that the material will become transparent for a broader range of wavelengths. Other alloying elements, e.g. Nb or Ta, has been proposed to act similar to Si.

Modern computational methods allow for a significant reduction of experimental efforts and costs by providing materials trends and selection rules. In particular, quantum-mechanical calculations based on Density Functional Theory provide highly accurate and affordable predictions. Harnessing available computational power provided by Anselm supercomputer enables high throughput calculation approach, which will be used to create a “materials library” of calculated properties of various TiO2-based alloyed materials. This will serve as a starting point for subsequent experimental work focused on further application-orientated optimization.

**Scientific readiness:**

**Aims and objectives**

The aim of this project is to calculate properties of various Ti1–*nXn*O2 solid solutions, based on anatase and rutile structures for low concentrations *n*, and on corresponding *X*O*m* structures for high *n*. A particular attention will be paid to optical-related properties such as band gaps or dielectric function. Supercells representing concentrations *n* ranging from 0 to 1 will be constructed, structurally optimized, and their properties will be calculated and evaluated. Broad scope of this study, allowed by available computational power, will enable systematic identification of new alloying trends (structure and composition based) in studied materials. The main considered alloying element will be Si, but other promising candidates, e.g. Zr, Hf, Sc, Nb, Ta, Al or Ge, will be also explored.

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

Ti1–*n* *Xn*O2 solid solutions will be modeled using Special Quasi-random Structures (SQSs) [1] with optimized short-range order parameters describing the structural disorder (SQS generation and supercell structural optimizations will be done using co-investigater’s computational resources). The supercell sizes will range between 50 and 100 atoms to make the study computationally feasible. The quantum mechanical calculations will be performed within the framework of Density Functional Theory (DFT) [2,3], which replaces a solution of the Schrödinger equation for many-body wave-function with a search for charge density.WIEN2k DFT package will be used for this project. It is based on the full-potential (linearized) augmented plane-waves + local orbitals method [4], one among the most accurate schemes for band structure calculations. WIEN2k is an all-electron fully relativistic code. As a part of it comes also the recently developed modified Becke-Johnson (mBJ) exchange-correlation potential which yields highly accurate energy band gaps for most semiconductors and insulators, with experimental agreement of the same order as much more computationally expansive hybrid functional or GW methods [5]. Using full-potential all-electron scheme in combination with the mBJ potential is critical for calculations of correct optical properties.

**Impact and outlooks**

Multiple works concerning experimental [e.g., 6, 7, 8] and calculated [e.g., 9, 10, 11] properties of doped TiO2 exist, however not at the scale proposed by this project. Wide scope of this study will allow for systematic and “coherent” identification of alloying trends (in optical, electronic and other material properties) in those systems. These will be subsequently used to predict and select the best candidates for future experimental investigations.

Calculated optical properties together with the underlying band structure will also help to better understand the nature of electronic transitions in these materials, hence facilitating a development of advanced dispersion models. 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) and applicant possesses valid academic license. 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.

Additionally, MPI fine-grained parallelization can be used when necessary, e.g., for memory limited problems or where there are fewer *k*-points than available processors.

One typical SCF cycle on a test Ti1–*n*Si*n*O2 48-atom supercell takes approximately 8 CPU-hours, out of which only about 6 minutes are pure serial code, and the rest makes use of the built-in *k*-point parallelization scheme.

**Computational resources:**

As no Sandy Bridge processors were available to benchmark, few typical calculations of Ti1-*n*Si*n*O2 were done on a quad-core Intel Xeon CPU W3550 @ 3.07GHz processor to help estimating the needed CPU hours. One Intel Xeon core was assumed to have approximately the same power as one Intel Sandy Bridge core at Anselm. From these calculations, an estimation of the total CPU time needed was done as follows:

Planned amount of calculated structures: 150 (30 structures or elements x 5 concentrations each)  
Usual number of SCF cycles needed to converge a calculation using the mBJ potential: 60-80  
Core hours needed for one typical cycle with 48 atoms cell size: 8  
Additional core hours needed for larger test cases (to test cell-size effects) and amorphous structures: 20000  
  
Total CPU hours needed: 150 \* 70 \* 8 + 20000 = 104000

**Economic readiness:**

**Socioeconomic impact:**

In a long run, this research could lead to development of components such as multilayer antireflecive coatings, interference filters, or waveguides with greatly improved optical performance.

**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-1)