Nuclear quantum effects in electron microscopy

CIM project proposal

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1 Background

Transmission electron microscopy is a powerful instrument providing insight into materials at spatial resolutions well below the resolution of optical microscopes. Individual atoms can be resolved in crystalline specimens. Moreover, by observing inelastic processes, i.e., processes where the electrons from the beam lose some energy while passing the sample, we can perform atom-by-atom elemental mapping. Figure 1 shows a high-resolution image of a crystalline specimen, spectra and elemental maps. In the left panel we can see electron energy loss spectra measured at two different pixels (position on a sample). They reveal peaks at specific energy levels (denoted Ti-$L_{23}$, Mn-$L_{23}$ and La-$M_{45}$) which are signatures of presence of specific elements. Right panel a color map for all the three element types (A-La, B-Ti, C-Mn) and their overlay (panel D).

Atoms in such images are represented as “clouds”, despite that the electron scattering occurs mostly on atomic nuclei, which are minuscule. The reason is that atoms in a sample are never steady. Not even at theoretical temperature of zero Kelvin. Explanations lie not in a classical physics but in the world of quantum mechanics. Quantum mechanics teaches us that even at the very lowest temperature, the atoms are not standing still but perform so called zero point vibrations, also called “nuclear quantum effects” (NQE). Contrary to Nature, in theoretical simulations we have the possibility to turn on or off the quantum mechanical effects and observe, how they change the simulated images of electron scattering. Fig. 2 shows such a comparison for a hypothetical Gd crystal scattering an electron beam at a very low temperature of 5 K. The NQE are either ignored (left) or fully taken into account (right) and the differences are obvious. Clearly, if we want to explain (or predict!) the experimental observations, we have to carefully consider the quantum mechanical effects.

2 What to do?

The project topic proposed here deals with careful consideration of the nuclear quantum effects and their implications on observations in transmission electron microscopes.

One of the most accurate methods to deal with NQE is the path-integral molecular dynamics (PIMD), which originates from works of Richard Feynman, who has shown that an evolution of a quantum
mechanical system of vibrating atoms can be mapped (bijection), using the path integral formulation, on a set of \( n \) copies of classical systems of vibrating atoms. The catch is that these \( n \) copies are not independent, but they are mutually connected by harmonic forces. A sketch of this idea is in Fig. 2.

Recently, in works of Ceriotti et al.\cite{Ceriotti} it was shown that NQE can be included into simulations in a simpler way. Atoms in crystals at a specific temperature vibrate randomly about their equilibrium positions. This is simulated by random stochastic forces, which deliver the atoms “kicks”, as the system evolves in time. These forces, in a classical simulation, do not have any frequency dependence and have no memory effects. Such dynamics is represented as a Markovian process. Ceriotti and coworkers have shown that by including memory effects into the noise it is possible to simulate NQE without performing expensive PIMD simulations.

This idea was developed further, because soon it was realized that such so called “colored noise” can be used for a number of specific purposes. For example to only excite atomic vibrations of specific frequencies. Also, it is possible to mix ideas of PIMD and colored noise to perform very accurate and yet fast simulations of NQE.

We intend to apply and develop further the methods of non-Markovian dynamics and study their implications for the simulated images of experiments in transmission electron microscopy. Such simulations use intensively the methods of mathematical statistics to evaluate mean values of operators. Simulations of electron microscopy images (scattering of electrons) involve integration of differential equations, similar to well-known Schrödinger equation in quantum mechanics.

### 3 What for, with whom and where?

Using precise simulations of atomic vibrations, correctly treating the quantum mechanical effects, we aim to propose new experimental methods utilizing transmission electron microscopy 1) to measure temperature of the samples with nanometer precision, 2) to measure the spectrum of atomic vibrations of materials. As the third point, we intend to characterize the influence of atomic vibrations on inelastic electron scattering experiments (such as those observed in Fig. 1) - a domain, where atomic vibrations are typically ignored due to complexity of the resulting simulations.

In cooperation with experimentalists from cooperating laboratories (Nagoya University, Japan; Oak Ridge National Laboratory, USA; IFW Dresden, Germany, among others) the developed methods will be tested “in the field”.

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### References

