Imaging of dynamics and electronic structure of materials

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Current state of the art

When solids are exposed to intense laser fields, nonlinear optics of solids advances to its extreme regime. A hallmark effect of extreme nonlinear optics is the emission of high harmonics of the laser from the bulk of materials[1–5]. High harmonic generation (see Fig.1) enables radically new approaches for spectroscopy and structural probing of the condensed matter. These approaches could be proven useful under conditions for which conventional spectroscopies become impractical. Moreover, it has been demonstrated recently, that high-harmonic emission can be used to image electric signals in electronic circuits[6]. This promises new diagnostic imaging techniques for the semiconductor industry. It has also been shown that that HHG is sensitive to topological properties of solids [7]. This opens an avenue towards time resolved HHG imaging of topological phase transitions. Furthermore, experimental and theoretical studies have shown the capacity to retrieve the valence electron potential of solids from harmonic spectra [8]. This is an exciting new aspect as common x-ray scattering studies cannot differentiate the valence electron density from the the total electron density in crystals.

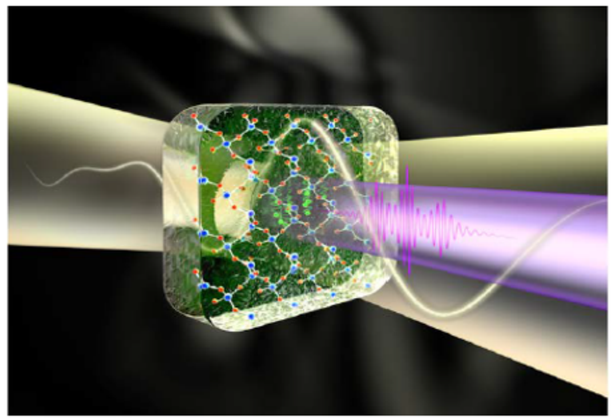

Figure 1: Artist's impression of high harmonic generation in bulk solids by an intense laser field.

This project focuses on yet another exciting aspect of solid-state physics in which we believe that high harmonic generation and precise measurement could enable the direct imaging of the electronic wavefunction of impurities in solids. Impurities and defects in the crystalline structure of solids have been long considered a nuisance in electronic and material science as they locally distort the properties of a crystal giving rise to undesirable effects. Yet modern developments in the field of quantum computing and communications has dramatically changed this point of view. Indeed, impurities and defects are now considered important candidates for the realization of qubits in solid state quantum computers. Whereas electron microscopes can measure the electron density of impurities localized close to surfaces of materials, HHG microscopy could allow the probing of defects in the bulk of materials. If ultrashort pulses are used to probe the quantum properties of defects it should be also possible to probe dynamics in these systems at ultrashort time scales. In a theoretical study we have shown [**?**] that HHG can be used to measure the wavefunction of impurities. More recently it has also been shown theoretically that high harmonic emission is not only sensitive to the presence of defects but can provide insight into the spin polarization state of the defects. The above works suggest a broad range of new possibilities for detail experimental explorations of these systems that would allow harnessing their unique properties in future electronic technologies.

Research goals and working program

At the focus of this project is the use of high harmonic generation in bulk solids for the study of impurities and defects. Important goals include: (a) Experimental demonstration of the capability of high harmonics in solids to detect impurities. (b) Excitation and real-time tracking of electronic phenomena in impurities (c) Direct reconstruction of the quantum wavefunction of impurities in space and time.

The basic pillars of this project include a variety of experimental and theoretical tools: (a) intense, light pulses confined to and controlled within sub-cycle time intervals which can induce the generation of high harmonics in solids, (b) analytical tools that can probe the induced currents/radiation and concomitant structural modifications (c) advanced theoretical tools that provide high fidelity modelling of the dynamics and can make the necessary predictions for the optimal interaction regimes and materials to be employed.

In the planed experiments impurities will be introduced to a variety of insulating samples using ion beam implantation. To ensure a significant signal level, impurities will be implanted at various densities. The high harmonics will be studied in a setup as shown in Fig.2. To perform tomographic experiments, we will follow the methodology presented in our recent theoretical work namely will study the characteristics of the emitted harmonics to reconstruct the spatial properties of the impurity. The study of dynamics associated with the implanted impurities/defects will be possible by the adequate excitation of electronic coherences using ultrabroadband laser light pulses which linearly or nonlinearly promote population to excited states of the system. High harmonics generation will in turn probe the dynamics of the excited system in a pump probe setting. Theory will also play an essential role in this endeavor. Expanding further the ideas of high harmonic tomography to three dimensions and for model systems that describe the experiments with high accuracy it will become possible to visualize the world of material defects in space and time.

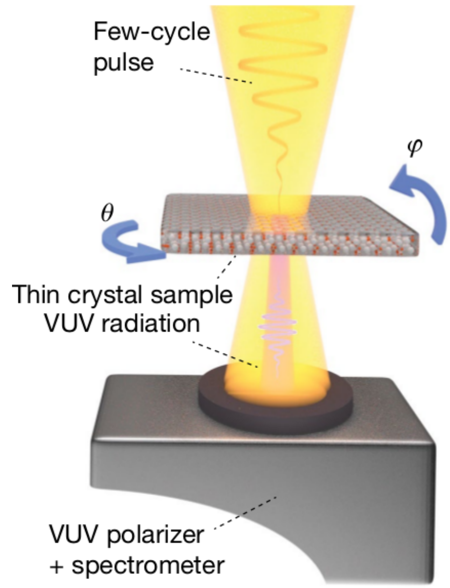

Figure 2: Schematic of a high harmonic generation and measurement experimental setup. A few-cycle laser pulse impinges on a crystaline medium in which impurities have been introduced by ion-beam implantation. The emerging high harmonics from the sample are recorded by a VUV spectrometer. Rotation of the crystaline sample allows for three dimensional probing of the crystal structure.

References

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