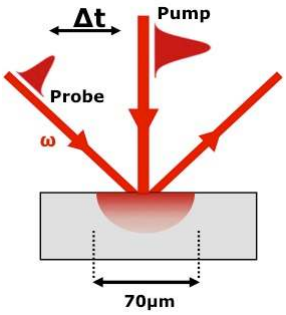
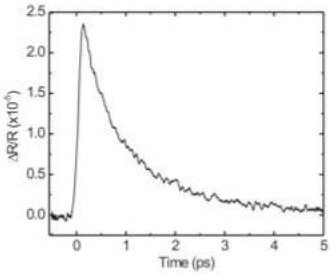
NEWS

- Cliff graduated and joined Paul Scherrer Institute as a postdoctoral scholar. |11/2024
- Prof. Gedik honored as **Committed to Caring.** |09/2024
- Yifan graduated and joined Columbia University as a postdoctoral scholar. |08/2024
- Batyr graduated and joined UC Berkeley as a

Pump Probe Spectroscopy

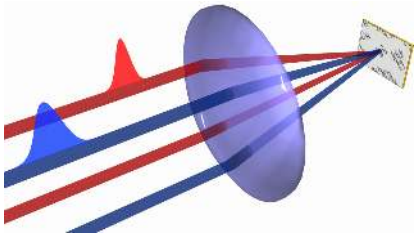
Pump probe spectroscopy is the simplest experimental technique used to study ultrafast electronic dynamics. In this technique, an ultrashort las two portions; a stronger beam (pump) is used to excite the sample, generating a non-equilibrium state, and a weaker beam (probe) is used to m induced changes in the optical constants (such as reflectivity or transmission) of the sample. Measuring the changes in the optical constants as a between the arrival of pump and probe pulses yields information about the relaxation of electronic states in the sample.

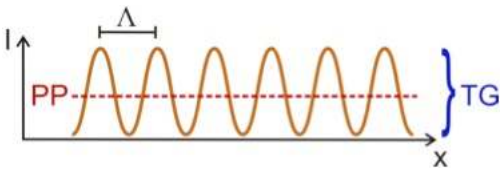




Transient Grating Spectroscopy

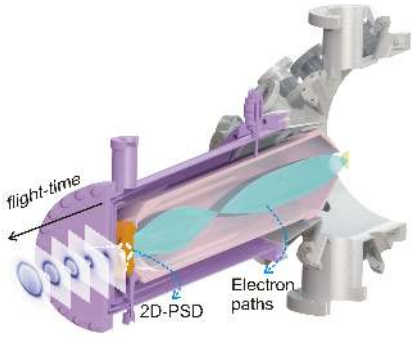
Pump probe spectroscopy described above is well suited for measuring the lifetime of electronic excitations with femtosecond time resolution. In propagation of these excitations in real space, we use transient grating spectroscopy. In this technique, a pair of femtosecond pulses is interfere generate a sinusoidal intensity modulation, that in turn induces a density grating of photoexcitations. Because the index of refraction depends or density, a periodic modulation of the index of refraction is formed. The period of this pattern in real space can be changed either by changing the laser or the angle between the two beams. An incident probe pulse on this pattern is therefore both reflected and diffracted. Measuring the time reflected and diffracted waves enables us to track the propagation of these excitations in real space.

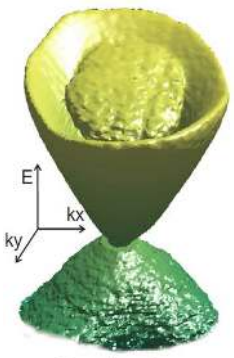




Circular Dichroism and Time-Resolved ARPES

Angle resolved photoemission spectroscopy (ARPES) is a powerful tool for mapping the electronic bandstructure of solids by measuring the energ photoemitted electrons. In our lab we use UV laser pulses and a time-of-flight spectrometer (shown on the left), which simultaneously measures electron energy by its flight time, and momentum Kx and Ky using a 2D position-sensitive-detector. As a result, 3D intensity spectra I(E,Kx,Ky) (the topological insulator Bi2Se3) are obtained.

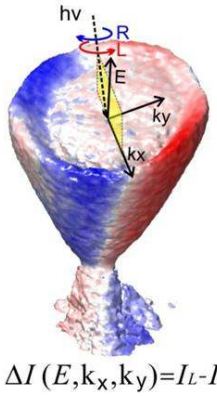




$I(E, k_x, k_y)$

We can also look for circular dichroism, which is the difference in the ARPES intensity spectrum obtained with right- vs. left-circularly polarized li such as the topological insulator Bi_2Se_3, this can be a sensitive measure of the electron spin orientation in momentum space (below left). The ul electrons can also be resolved by performing ARPES in a pump-probe scheme. An 80 fs ultrafast laser pulse pumps the sample to initiate electro excited system is then probed by the delayed UV pulse that photoemits electrons for the ARPES measurement. By sweeping the delay time t bet

while collecting the 3D intensity spectra I(E,Kx,Ky), we obtain a 3D movie of I(E,Kx,Ky,t) that captures electronsï¿½ dynamics in the solids on fe
(below right for Bi_2Se_3).



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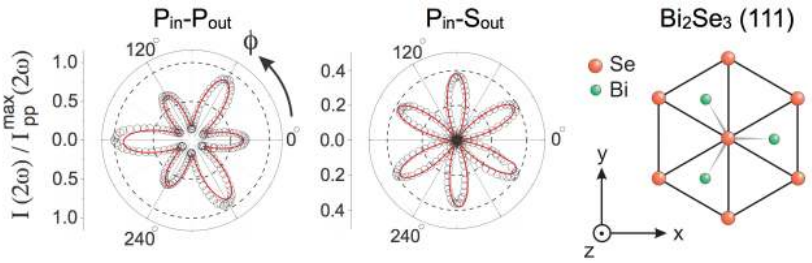
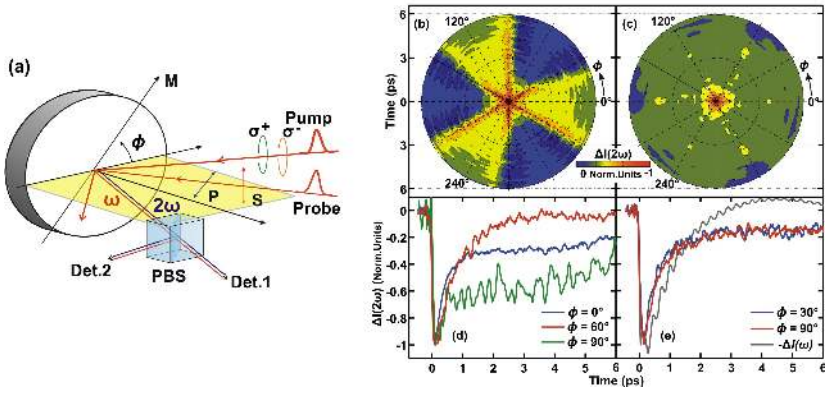
Second Harmonic Generation

A technique our lab has been researching to gain exclusive sensitivity to the surface of a material is optical second harmonic geneartion (SHG). I
electrical polarization of a material $P_i(\omega)$ has a dominant component linear in the driving optical field $E_j(\omega)$ as well as weaker components propor
of $E_j(\omega)$, where ω is the optical frequency and the indices run through three spatial coordinates. Components that contain two powers of $E_j(\omega)$ a
For electric dipole processes, the polarization $P_i = \chi^{(2)}_{ijk}E_jE_k$ is obtained from a third rank susceptibility tensor $\chi^{(2)}_{ijk}$ that vanishes under inversio
dipole induced SHG is forbidden in bulk crystals with inversion symmetry and is only allowed at surfaces or interfaces where inversion symmetry
By measuring the reflected output as the sample is rotated about its surface normal axis, we obtain patterns like those shown below for Bi_2Se_3,
symmetry of the surface electronic polarizability.

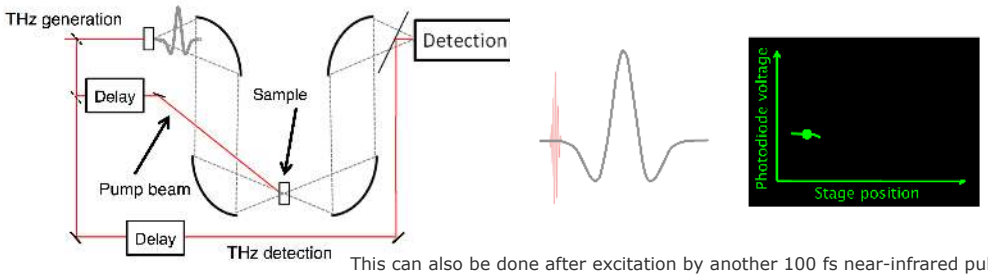


The ultrafast pulsed nature of the laser light used to perform SHG naturally lends itself to the study of ultrafast dynamics on the surface of mate
experiment, a pulse of laser light (pump) is first impinged onto the sample in order to create a non-equilibrium electron distribution. A second ti
(probe) is then used to monitor the temporal evolution of the SHG signal (see below). The relaxation dynamics of the non-equilibrium distributio
understand the microscopic energy loss mechanisms of the surface electrons.



TeraHertz Time-Domain Spectroscopy

Terahertz Time-Domain Spectroscopy (THz-TDS) is an optical technique used to measure equilibrium and non-equilibrium far infrared material pr
of refraction and conductivity. A THz pulse is generated via optical rectification in a ZnTe crystal using a 100 fs near-infrared pulse. The THz pulse
through a sample and subsequently detected in another ZnTe crystal via free space electro-optic detection. The measured signal is proportional t
the pulse, so the magnitude and phase are preserved, and the complex transmission coefficient can be extracted. From this, the full complex val
parameters can be extracted without the use of Kramers-Kronig relations.

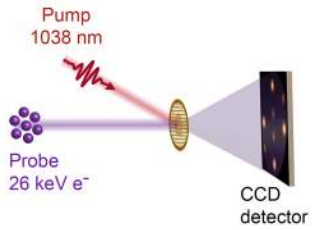


This can also be done after excitation by another 100 fs near-infrared pulse. By varying the time delay b and the excitation pulse, the non-equilibrium complex material parameters can be measured as a function of time, with resolution < 500 fs.

THz-TDS is best used to study systems in which the excitations of interest lie in the meV energy range, e.g., a superconductor with a small energ has been used to study a wide variety of systems, including Cooper pair and vortex dynamics in superconductors, carrier dynamics in semicondu phase transitions, and even metamaterials.

Ultrafast Electron Diffraction

Direct determination of structural dynamics requires the ability of measuring atomic motions with angstrom scale spatial resolution. Conventiona spectroscopy based on measuring transient changes in optical constants is sensitive to dynamics of electronic excitations but can provide only in about structural dynamics. The spatial resolution in these techniques is also limited to micron scales due to diffraction limit.



Ultrafast electron diffraction (UED) can directly couple to structural dynamics and provide sub-angstron together with sub-picosecond temporal resolution. The principle of UED is similar to pump probe spectr laser pulse is split into two; the first part of the laser pulse is directly focused on to the sample to creat state. To probe the induced structural change, the second part is frequency tripled and focused on to a generating an ultrafast electron packet via photoelectric effect. These electrons are then accelerated th (typically through 30 keV, de Broglie wavelength = 0.07 Å) and diffracted from the sample.

The relative arrival time of the probing electron packet and the initiating laser pulse at the sample can changing the relative optical path-lengths of the two laser beams. Recording the diffraction pattern of the electron packet as a function of this tir the equilibrium structure and a movie of the structural evolution with sub-Angstrom spatial resolution (reaching ~0.001 ï¿½ level) and sub-picos resolution.

Pump–probe microscopy

Pump–probe microscopy is a non-linear optical imaging modality used in femtochemistry to study chemical reactions. It generates high-contrast images from endogenous non-fluorescent targets. It has numerous applications, including materials science, medicine, and art restoration.

Advantages

The classic method of nonlinear absorption used by microscopists is conventional two-photon fluorescence, in which two photons from a single source interact to excite a photoelectron. The electron then emits a photon as it transitions back to its ground state. This microscopy method has been revolutionary in biological sciences because of its inherent three-dimensional optical sectioning capabilities.

Two-photon absorption is inherently a nonlinear process: fluorescent output intensity is proportional to the square of the excitation light intensity. This ensures that fluorescence is only generated within the focus of a laser beam, as the intensity outside of this plane is insufficient to excite a photoelectron.[1]

However, this microscope modality is inherently limited by the number of biological molecules that can undergo both two-photon excitation and fluorescence.[2]

Pump–probe microscopy circumvents this limitation by directly measuring excitation light. This expands the number of potential targets to any molecule capable of two-photon absorption, even if it does not fluoresce upon relaxation.[3] The method modulates the amplitude of a pulsed laser beam, referred to as the *pump*, to bring the target molecule to an excited state. This will then affect the properties of a second coherent beam, referred to as the *probe*, based on the interaction of the two beams with the molecule. These properties are then measured by a detector to form an image.

Physics of pump–probe microscopy

Because pump–probe microscopy does not rely on fluorescent targets, the modality takes advantage of multiple different types of multiphoton absorption.

Two-photon absorption

Two-photon absorption (TPA) is a third-order process in which two photons are nearly simultaneously absorbed by the same molecule. If a second photon is absorbed by the same electron within the same quantum event, the electron enters an excited state.[4]

This is the same phenomenon used in two-photon microscopy (TPM), but there are two key features that distinguish pump–probe microscopy from TPM. First, since the molecule is not necessarily fluorescent, a photodetector measures the probe intensity. Therefore, the signal decreases as two-photon absorption occurs, the reverse of TPM.[3]

Second, pump–probe microscopy uses spectrally separated sources for each photon, whereas conventional TPM uses one source of a single wavelength. This is referred to as degenerate two-photon excitation.[3]

Excited-state absorption

Excited-state absorption (ESA) occurs when the pump beam sends an electron into an excited state, then the probe beam sends the electron into a higher excited state. This differs from TPA primarily in the timescale over which it occurs. Since an electron can remain in an excited state for a period of nanoseconds, thus requiring longer pulse durations than TPA.[5]

Stimulated emission

Pump–probe microscopy can also measure stimulated emission. In this case, the pump beam drives the electron to an excited state. Then the electron emits a photon when exposed to the probe beam. This interaction increases the probe signal at the detector site.

Ground-state depletion

Ground-state depletion occurs when the pump beam sends the electron into an excited state. However, unlike in ESA, the probe beam cannot send an electron into a secondary excited state. Instead, it sends remaining electrons from the ground state to the first excited state. However, since the pump beam has decreased the number of electrons in the ground state, fewer probe photons are absorbed, and the probe signal increases at the detector site.[3]

Cross-phase modulation

Cross-phase modulation is caused by the Kerr effect, in which the refractive index of the specimen changes in the presence of a large electric field.[6] In this case, the pump beam modulates the phase of the probe, which can then be measured through interferometric techniques. In certain cases, referred to as cross-phase modulation spectral shifting, this phase change induces a change to the pump spectrum that can be detected with a spectral filter.[3]

Optical design

Excitation

Measuring nonlinear optical interactions requires a high level of instantaneous power and very precise timing. In order to achieve the high number of photons needed to generate these interactions while avoiding damage of delicate specimens, these microscopes require a modelocked laser. These lasers can achieve very high photon counts on the femtosecond timescale and maintain a low average power. Most systems use a Ti:Sapph gain medium due to the wide range of wavelengths that it can access.[3][7]

Typically, the same source is used to generate the pump and the probe. An optical parametric oscillator (OPO) is used to convert the probe beam to the desired wavelength. The probe wavelength can be tuned over a large range for spectroscopic applications.[7]

However, for certain types of two-photon interactions, it is possible to use separate pulsed sources.[3] This is only possible with interactions such as excited-state absorption, in which the electrons remain in the excited state for several picoseconds. However, it is more common to use a single femtosecond source with two separate beam paths of different lengths to modulate timing between the pump and probe beams.[3][7]

The pump beam amplitude is modulated using an acousto-optic or electro-optic modulator on the order of 10^7 Hz. The pump and probe beams are then recombined using a dichroic beamsplitter and scanned using galvanometric mirrors for point-by-point image generation before being focused onto the sample.[3]

Detection

The signal generated by probe modulation is much smaller than the original pump beam, so the two are spectrally separated within the detection path using a dichroic mirror. The probe signal can be collected with many different types of photodetectors, typically a photodiode. Then, the modulated signal is amplified using a lock-in amplifier tuned to the pump modulation frequency.[3]

Data analysis

Similar to hyperspectral data analysis, the pump–probe imaging data, known as a delay stack, has to be processed to obtain an image with molecular contrast of the underlying molecular species.[3] Processing pump–probe data is challenging for several reasons – for example, the signals are bipolar (positive and negative), multi-exponential, and can be significantly altered by subtle changes in the chemical environment.[8][9] The main methods for analysis of pump–probe data are multi-exponential fitting, principal component analysis, and phasor analysis.[3][7]

Multi-exponential fitting

In multi-exponential fitting, the time-resolved curves are fitted with an exponential decay model to determine the decay constants. While this method is straightforward, it has low accuracy.[7]

Principal component analysis

Principal component analysis (PCA) was one of the earliest methods used for pump–probe data analysis, as it is commonly used for hyperspectral data analysis. PCA decomposes the data into orthogonal components. In melanoma studies, the principal components have shown good agreement with the signals obtained from the different forms of melanin.[10] An advantage of PCA is that noise can be reduced by keeping only the principal components that account for majority of the variance in the data. However, the principal components do not necessarily reflect actual properties of the underlying chemical species, which are typically non-orthogonal.[3] Therefore, a limitation is that the number of unique chemical species cannot be inferred using PCA.[3]

Phasor analysis

Phasor analysis is commonly used for fluorescence-lifetime imaging microscopy (FLIM) data analysis[11] and has been adapted for pump–probe imaging data analysis.[8] Signals are decomposed into their real and imaginary parts of the Fourier transform at a given frequency. By plotting the real and imaginary parts against one another, the distribution of different chromophores with distinct lifetimes can be mapped.[3][7] In melanoma studies, this approach has again shown to be able to distinguish between the different forms of melanin.[8] One of the main advantages of phasor analysis is that it provides an intuitive qualitative, graphical view of the content[7] It has also been combined with PCA for quantitative analysis.[12]

Applications

The development of high-speed and high-sensitivity pump–probe imaging techniques has enabled applications in several fields, such as materials science, biology, and art.[3][7]

Materials science

Pump–probe imaging is ideal for the study and characterization of nanomaterials, such as graphene,[13] nanocubes,[14] nanowires,[15] and a variety of semiconductors,[16][17] due to their large susceptibilities but weak fluorescence. In particular, single-walled carbon nanotubes have been extensively studied and imaged with submicrometer resolution,[18] providing details about carrier dynamics, photophysical, and photochemical properties.[19][20][21]

Biology

The first application of the pump–probe technique in biology was in vitro imaging of stimulated emission of a dye-labelled cell.[22] Pump–probe imaging is now widely used for melanin imaging to differentiate between the two main forms of melanin – eumelanin (brown/black) and pheomelanin (red/yellow).[23] In melanoma, eumelanin is substantially increased. Therefore, imaging the distribution of eumelanin and pheomelanin can help to distinguish benign lesions and melanoma with high sensitivity[24]

Art

Artwork consists of many pigments with a wide range of spectral absorption properties, which determine their color. Due to the broad spectral features of these pigments, the identification of a specific pigment in a mixture is difficult. Pump–probe imaging can provide accurate, high-resolution, molecular information[25] and distinguish between pigments that may even have the same visual color.[26]

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