

Shedding light on condensed matter, literally.

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We all know that plants use the energy from sunlight to transform water and carbon dioxide into oxygen and sugars, while converting the sunlight's energy into chemical energy. This can be regarded as one example of photoinduced phenomena, since shining light induces the synthesis of sugars. A more physics-related and well-known example is the solar cell, which is the application of the photoinduced voltage in p-n junctions (i.e. the interface of semiconductors with different doping).

The topic I will introduce in this essay is the ultrafast photoinduced phenomena in condensed matter, which is something probably less-known compared to the two examples I showed earlier. The main idea is to instantly induce some photoinduced phenomenon by shining an extremely short pulse of light onto a sample, and then observe how the photoinduced phenomenon evolves as time passes. Some of the already known ultrafast photoinduced phenomena include ultrafast photocurrents, currents that are induced by photoexcitation, and ultrafast photoinduced phase transitions. How the time evolution of such a phenomenon should behave is completely nontrivial, making it a “dark problem”. The dynamics of such phenomena contain rich information of the material's character that cannot be accessed without photoexcitation, and thus examining ultrafast photoinduced phenomena is an important approach for the fundamental understanding of condensed matter.

How do we measure the time evolution of ultrafast photoinduced phenomena? The typical method to determine the properties of solids is to measure the

resistivity. This will involve measuring some voltage or current by using electronic devices, which is the method usually referred to as dc transport measurements. While this method is valid in determining the static properties of solids, it cannot be used to measure the time evolution of photoinduced phenomena. This is because the time resolution of electronic devices is on the order of nanoseconds (10^{-9} seconds), while the typical time scales for dynamics in solids (e.g. relaxation of excited carriers, energy transfer between electrons and phonons, etc.) are in the sub-picosecond (10^{-12} second) range. This specific time scale in solids will also affect the choice of the light source. If we control the light source's on/off by electronic devices, the light source will have a pulse duration on the nanosecond scale. This does seem ultrafast in the human perspective, but can hardly be regarded as an instant excitation in the solid's perspective. We will need light sources that emit pulses with the duration on the sub-picosecond scale or even shorter.

Reading up to this point, the reader may feel that the experimental approach to ultrafast photoinduced phenomena is hopeless. However, this is not the case. The recent advancement in femtosecond laser technology now enables us to use strong laser pulses with pulse durations of 100 femtoseconds (10^{-15} seconds) or shorter. This light source provides us with the instant excitation we need to investigate the ultrafast photoinduced phenomenon in solids. There is another advantage of this technology, as its application leads to time-resolved measurements with time resolution as short as the pulse width (~ tens of femtoseconds).

The history of ultrafast photoinduced phenomena is rather short, as it was only realized after the development of femtosecond lasers. The first reports of femtosecond laser pulses came out in the 1970s, and its application to investigate condensed matter emerged from the end of the 1980s to the beginning of the 1990s. The

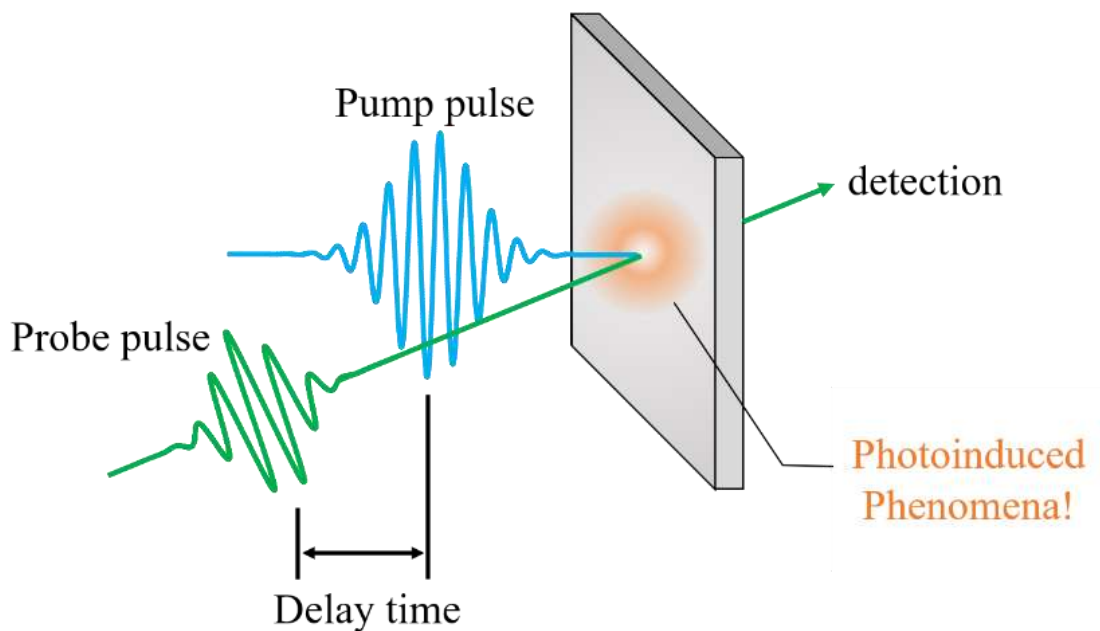
main technique used in these measurements is called pump-probe spectroscopy. It remains the central method to directly measure the time evolution of photoinduced phenomena.

In regular spectroscopy measurements, we shine light onto a solid and measure the intensity of the transmitted or reflected light. The transmittance or reflectance of the sample contains information on the solid's properties. One can say this is analogous to taking a picture of a ball. From the picture, you can recognize the color, size, and texture of the ball, but you cannot understand the dynamic properties of it (i.e. how much velocity it would gain by dropping it from a certain height, or to what height would the ball bounce after that). If you wanted to understand these characters, you would need to first lift the ball to a certain height and drop it, wait for a certain amount of time, and then take a picture of the ball. By repeating this procedure while varying the waiting time (0.1 seconds, 0.2 seconds, 0.3 seconds,) and by combining them, you obtain an animation that resembles the motion of the ball. This is exactly what we do in pump-probe measurements, except the waiting time interval is much shorter. We first excite the sample with a strong laser pulse (pump pulse), and then shine a weaker pulse (probe pulse) after a certain time interval (also called "delay time"). The transmittance or reflectivity of the probe pulse is measured while varying the delay time, and by doing so, one can measure the time evolution of the solid's character after strong and instantaneous photoexcitation.

Using this method, a variety of ultrafast phenomena have been found in solids. One of the most famous examples is photoinduced superconductivity [1, 2]. By exciting a normal conductor with the pump pulse, it turns into a superconductor for about a picosecond (though I note that there are arguments against this interpretation as well). I already discussed that ultrafast photoinduced phenomena contain essential information

for the frontier areas of science. However, these examples of photoinduced superconductivity show that investigation of ultrafast photoinduced phenomena can lead to applications as well. The fact that functional properties emerge by photoexcitation, and that we can manipulate them on an ultrafast time scale opens up the possibility of various applications beneficial for the society.

To summarize, ultrafast photoinduced phenomena are an important topic both for fundamental science and applications. It is surprising that we have a method to experimentally access such extreme phenomena, but over the last few decades pump-probe spectroscopy has turned out to be a powerful tool.



Reference

- [1] Mitrano, M., Cantaluppi, A., Nicoletti, D. *et al.* Possible light-induced superconductivity in K_3C_{60} at high temperature. *Nature* **530**, 461–464 (2016).
- [2] Suzuki, T., Someya, T., Hashimoto, T. *et al.* Photoinduced possible superconducting state with long-lived disproportionate band filling in FeSe. *Commun Phys* **2**, 115 (2019).