B05 Towards Femtosecond Spectroscopy of Spontaneous Phase Transitions

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Almost all phenomena we encounter in daily life can ultimately be traced back to processes at the atomic level, which occur within a few femtoseconds or picoseconds. Since conventional electronic measurement methods cannot resolve such time scales, optical methods based on femtosecond laser technology are often used to investigate these dynamics. These technologies are usually based on triggering the investigated process with a strong pump-pulse and then probing with a second, time-delayed probe pulse. However, most dynamics in nature does not involve femtosecond triggering. Therefore, pump-probe laser spectroscopy is not really suitable for understanding spontaneous reaction dynamics under natural environmental conditions, since the pump pulse always places the substance in a rather unnatural starting condition. Here, we investigate how to potentially solve this fundamental problem of femtosecond spectroscopy with the help of a double-probe approach. In contrast to the classical pump-probe technique, we completely avoid any excitation by means of a pump pulse, but instead allow the process to be triggered by spontaneous fluctuations. In this way, the process takes place spontaneously in its natural, undisturbed form. In order to nevertheless obtain a high time resolution, the investigated material is measured repeatedly by pairs of femtosecond probe pulses, and the results of the two measurements are compared. By changing the time interval between the two pulses, information about the dynamics can be obtained without the need for a dedicated excitation (compare Fig. 1).

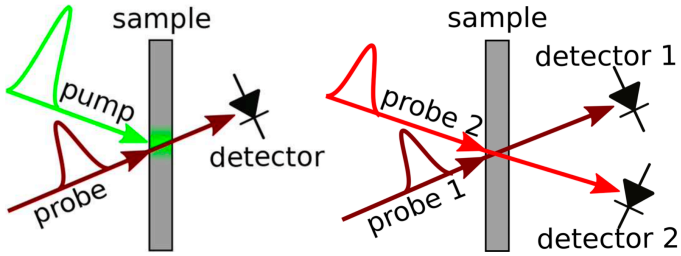

Fig. 1: *Exemplary figure showing classical pump-probe approach (left) and double pulse method (right).*

Two promising processes for the application of this new measurement approach are investigated in the scope of this project: First, the magnetic Barkhausen effect describes spontaneous changes in the magnetization of ferromagnetic materials under the influence of time-varying magnetic fields. Since magnetization can be measured very efficiently by means of the Faraday effect, this phenomenon is ideal for a first application of our double-probe method. However, the dynamics may occur only on nanosecond timescales, and therefore a femtosecond time resolution may not be required. Second, vanadium dioxide has a metal-insulator transition near room temperature, which is well studied after ultrafast heating, but whose dynamics upon cooling could not be explored so far. The application of our double-probe method to the transition will allow us to study this technologically and fundamentally relevant phase transition as it spontaneously occurs. We are already able to produce VO_2 crystals [1] and can also simulate the heating and cooling processes [2] that will be applied to trigger the transition.
In short, the new double-probe spectroscopy to be developed [3] will enable us to measure spontaneous reaction dynamics with the ultimate time resolution that is offered by femtosecond laser technology, without putting the material into unnatural physical states by any pump pulses.

References:

[1] S. Sprinkart, D. Kazenwadel, R. Hartmann, P. Baum, *Phys. Rev. Res.,* in print (2023).
[2] D. Kazenwadel, N. Neathery, P. Baum, manuscript in preparation (2023).
[3] D. Kazenwadel and P. Baum, manuscript in preparation (2023).