Ponderomotive acceleration of photoelectrons in pump-probe experiments

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The primary objective of our experiment at the Artemis facility was to use femtosecond time-resolved photoemission to study the ultrafast magnetisation dynamics in Gd and Tb as a function of the pump laser wavelength. During these pump-probe measurements we observed strong oscillations in the photoelectron kinetic energy before time zero, i.e. at pump probe delays when the probe pulse arrives before the pump. Under our experimental conditions, one would expect to see no dynamics until the pump pulse has excited the system. The phenomenon is due to acceleration of outgoing photoelectrons by the electric field of the pump laser pulse as it reflects off the sample surface. This is a very general phenomenon in pump-probe time-resolved photoemission experiments, and our results show it will become more significant as researchers use pump laser wavelengths further into the infrared and high-harmonic probe sources. Here we describe a thorough investigation of this effect in photoemission from a clean W(110) surface.

In our pump-probe experiments on the magnetization dynamics of Gd(0001), one obvious feature, shown in Fig. 1, is an oscillation in the kinetic energy before zero time delay (i.e. probe arrives before pump). These oscillations have been observed previously on Gd and explained by Bovensiepen et al. [1] in 2009. The authors invoked ponderomotive acceleration of the outgoing photoelectrons by a transient optical grating formed by interference between the incoming and reflected laser pulse. We observe two significant differences from the published results. Firstly, the amplitude of the oscillations before time zero in our experiment is up to 50 times larger than observed in 2009, and secondly, electrons photoemitted from different initial states can show different oscillation amplitudes in the kinetic energy.

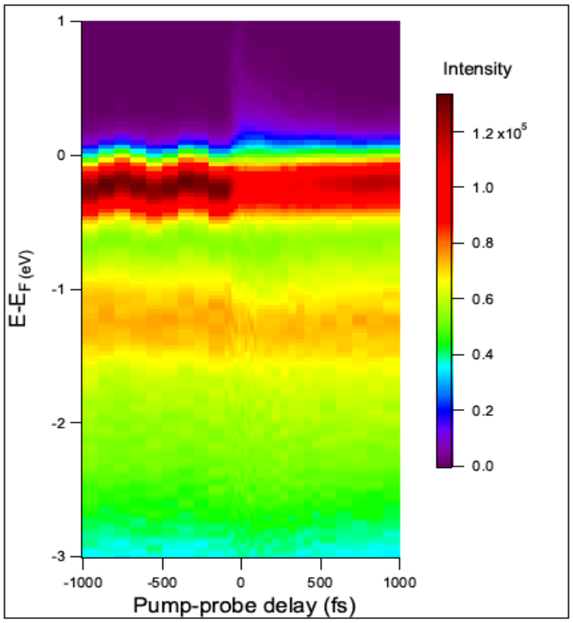

Fig. 1: Time-resolved photoemission spectrum of Gd(0001) following excitation at 1300nm. The magnetization dynamics of Gd occur at pump-probe delays t>0. The oscillations at t<0 are due to ponderomotive acceleration of photoelectrons by the electric field of the pump laser pulse after they have left the sample. This effect can interfere with the dynamics of interest.

As a result of these discrepancies we made a more thorough investigation of the ponderomotive oscillations. We performed time-resolved pump-probe experiments on the W(110) surface that is normally the substrate for our Gd samples. The experiment was repeated with 5 different pump wavelengths: 1610nm, 1450nm, 1384nm, 1300nm, 1148nm. The results are shown in Fig. 2, and reveal a strong dependence on the pump wavelength.

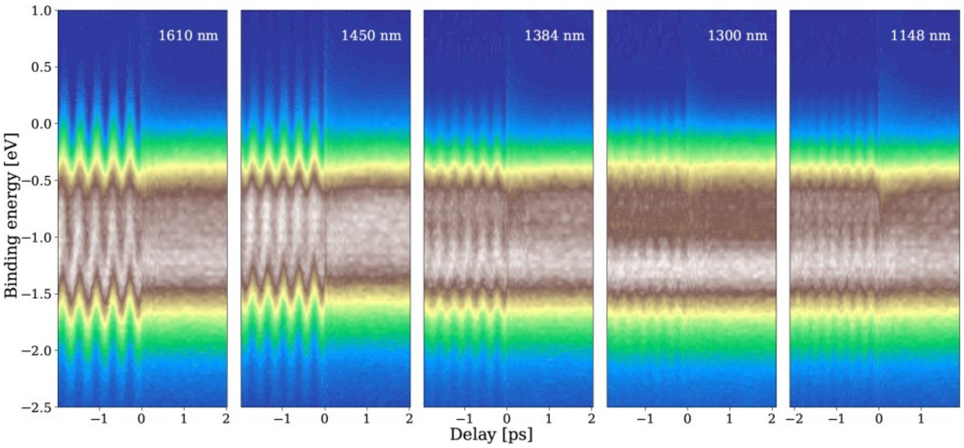

Fig. 2: Oscillatory ponderomotive acceleration by the pump pulse of photoelectrons ejected from W(110) by XUV photons at 36eV. The pump wavelength decreases from left to right: 1610nm, 1450nm, 1384nm, 1300nm, 1148nm.

In [1], the oscillations had an amplitude of about 2meV while we observe amplitudes up to 100meV for the longest pump wavelengths. According to the ponderomotive acceleration model, the higher momentum of the electrons and longer wavelengths of the pump pulse in our measurements should produce larger oscillations compared to the data measured in [1]. Nevertheless, this does not completely account for the magnitude of amplitudes we observe. Uncertainty in the laser fluence may also contribute.

We have improved the model of ponderomotive acceleration by removing some approximations made in [1]. The model now accounts for the acceleration when the pump and probe pulses overlap in time and at positive time delays (probe after pump). This offers the possibility of studying the influence of the oscillations on very fast dynamics. We find that the phase of the oscillations is crucial for the extension of the oscillations into positive time delays. For phases ≤180°, which one expects for light reflected off a non-ideal metal surface, the influence is small, see Fig. 3.

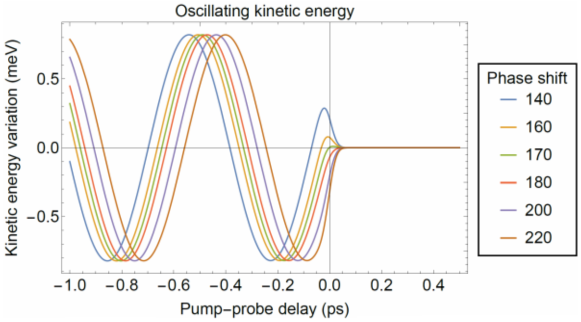

Fig. 3: Kinetic energy variation of photoemitted electrons for different phase shifts of the oscillating light field.

The oscillations in our measurements have phases between 150° and 170°. Thus, we are confident that our observation of a transient increase in the magnetization of the 5d spin system in Gd is not a side effect of the oscillations and will be explored in a fothcoming article.

The photoemission from W(110) comprises two contributions, one from bulk d states, and the other from a mixed bulk and surface resonance. Although these states have nearly the same binding energy, the amplitude of the oscillations is different, see Fig. 4. This is surprising because the electrons should be screened within a few tens of femtoseconds after excitation and not react differently to tha transient potential. This aspect will require further theoretical investigation.

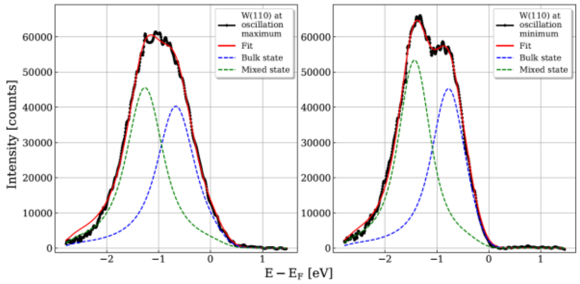

Fig. 4: W(110) spectrum at the maximum (left) and minimum (right) of the oscillations before time zero pumped with 1600nm. The two states shift by a different amplitude, thus overlapping at an oscillation maximum.

References

[1] Bovensiepen et al., Phys. Rev. B 79, 045415 (2009)