Controlling Free Induction Decay using Strong Fields and XUV Pulses

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Abstract

We present an experimental study of controlled Free Induction Decay in the extreme ultraviolet (XUV) regime. An attosecond pulse train is used to coherently promote argon to a superposition of the ground state ([Ne]$3s^2 3p^6$) and a series of excited states ([Ne]$3s^1 3p^6 np^1$), which are embedded in the [Ne]$3s^2 3p^5$ continuum. This superposition coherently emits light with the same directionality and divergence as the incoming XUV light. Applying a strong infrared probe pulse either break the coherence, or control the direction and phase of the emitted light.

When an ensemble of atoms is exposed to a short, coherent light pulse it will respond collectively and the excited atoms will start to act as oscillating dipoles. These dipoles may continue to oscillate coherently for a long time after the excitation pulse has passed resulting in forward scattered light known as Free Induction Decay (FID) [1,2]. This forward scattered light has the same spatial properties as the excitation pulse, but the phase is shifted by π. The overlap between the two fields will therefore yield the normal absorption spectrum observed in optical spectroscopy. FID has been observed from THz radiation to the optical regime, but it is more challenging when shorter wavelengths are used.

We propose a method to observe FID at even shorter wavelengths in the extreme ultraviolet (XUV) regime. The coherent XUV light is generated using high-order harmonic generation from a carrier-wavelength tunable Ti:Sapphire laser. The laser system is a conventional chirped pulse amplification Ti:Sapphire based laser with a programmable acousto-optic filter in the amplification chain to minimize gain-narrowing. By restricting the bandwidth of the seed for the amplification chain the carrier wavelength of the output can be tuned from 770-820 nm, while maintaining sub-40 fs pulses.

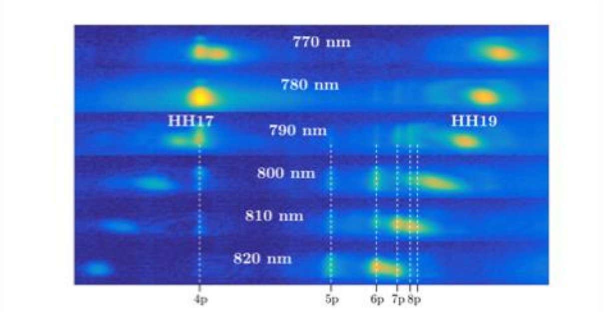

Fig. 1. Harmonics 17 and 19 transmitted through argon as a function of carrier-wavelength of the infrared laser. A number of window resonances in the absorption cross-section of the [Ne]$3s^2 3s^5$ continuum are observed [3].

Figure 1 demonstrates the tunability of the XUV light. Harmonics generated in an argon gas jet are refocused into a second pulsed gas jet of argon; the light transmitted through the second jet is then recorded by a flat-field spectrometer. Whenever parts of the harmonic light spectrally overlap with inner-shell transitions in argon, a coherent wave packet is excited, which emits light with similar directionality and divergence as the incoming XUV light.

In order to overcome the challenges of observing FID in the XUV region we use a short IR pulse to change the direction of the light emitted by the ensemble of oscillating dipoles. We demonstrate that the spatial phase of the FID can be controlled if an IR probe pulse is focused at the ensemble at a small non-collinear angle, illustrated in Fig. 2. In addition to these snapshots we will present time-evolution of these dipoles as a function of the pump-probe delay.

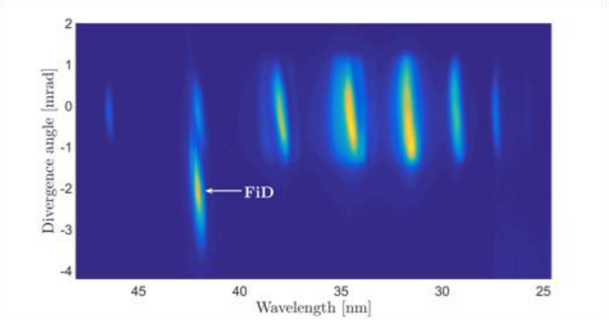

Fig. 2. The spectrum transmitted through argon under the influence of a strong non-collinear infrared probe pulse. The probe pulse modulates the spatial phase of the excited dipoles, thus modifying the directionality and divergence of the FiD.

References

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