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Observing Ultrafast Light–Matter Interactions in Real Time

Many ultrafast phenomena have timespans from femtoseconds to picoseconds. Femtophotography—recording 2D spatial information at trillions of frames per second (Tfps)—is indispensable for clearly resolving their spatiotemporal details. Currently, femtophotography is mostly realized by using multi-shot approaches.¹ However, these methods require the dynamic events under observation to be precisely reproducible, which renders them incapable of studying non-repeatable or difficult-to-reproduce ultrafast phenomena, such as femtosecond laser ablation.²

To overcome these limitations for imaging ultrafast events in real time (in the time duration of the event’s occurrence), we recently reported a new ultrafast imaging method—swept coded aperture real-time femtophotography (SCARF)—that can capture non-repetitive 2D ultrafast events at up to 156.3 Tfps in a single-shot measurement.³

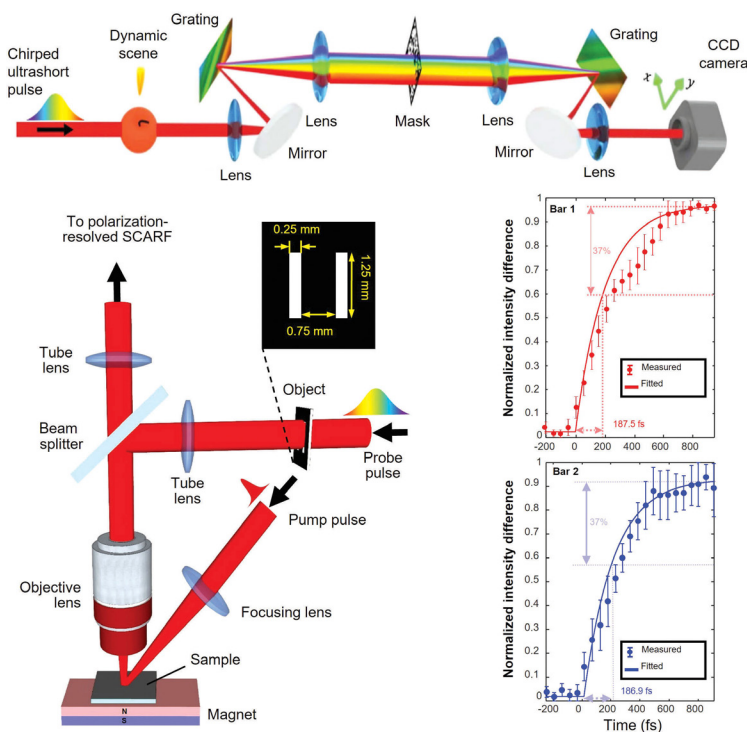
SCARF proceeds in several steps. First, a single linearly chirped laser pulse illuminates a dynamic

scene as a probe pulse. Because of its linear chirp, each wavelength in the pulse’s bandwidth carries a specific timestamp. Then, this probe pulse is imaged by a dispersive 4f imaging system onto a pseudo-random binary transmissive mask. The spectral dispersion shears temporal information contained in wavelengths to different positions for spatial encoding by the mask.

Afterward, the pulse is relayed to a CCD camera by another dispersive 4f imaging system that mirrors the configuration of the first one, which provides the second spectral shearing in the reverse direction. The camera records a compressed snapshot of the dynamic scene’s temporal information that is read out by the single probe pulse. Finally, the dynamic scene is retrieved by employing a compressed sensing-based reconstruction algorithm.

SCARF has been used for observing ultrafast light–matter interactions in real time. In one example, we recorded the transient absorption of a single laser pulse in semiconductors, which will contribute to the study of ultrafast carrier dynamics in graphene and other quantum materials. SCARF has also achieved 2D observation of demagnetization in a metal alloy film. The result reveals a time difference in the onset of this change between the two areas probed due to the oblique incident pump pulse, which cannot be observed by multi-shot approaches and gives new insight into ultrafast magnetic memory.

As an active-illumination single-shot femtophotography method, SCARF not only provides jitter-free operation but has also proven its ability at different spatial scales, with various contrasts, and in both reflection and transmission modes. This generic scheme can be readily integrated into many imaging modalities to bring real-time ultrafast imaging capability to the communities of imaging and sensing, ultrafast science, materials characterization, and next-generation data storage. **OPN**



Top: SCARF system schematic. Bottom: SCARF of ultrafast demagnetization. (Left) Experimental setup. (Right) Time-resolved normalized intensity difference between the *s*- and *p*-polarization light for the two selected areas.