Ultrafast electron microscopy as a laser-pump/electron-probe technique allows for the investigation of structural and electronic dynamics occurring at sub-picosecond timescales and nanometer length-scales. However, current implementations necessitate compromises in electron source brightness compared to conventional electron microscopy techniques. In-situ transmission electron microscopy with temporally-structured optical sample excitation, i.e. by employing femtosecond laser pulse trains, offers a complementary approach to access ultrafast processes, without the need for customized pulsed electron sources. To this end, we implement free-space-coupled femtosecond sample excitation in a Schottky field-emission electron microscope and investigate the optical response of magnetic domain structures with Lorentz microscopy. Specifically, we study laser-induced domain rearrangements in polycrystalline iron thin films on silicon nitride membranes which are pumped with single sub-50-fs laser pulses. By inverting the observed image contrast at large defocus, we reconstruct the local in-plane sample magnetization based on a transport-of-intensity approach. Prior to laser-excitation, the iron thin films display the well-known magnetic ripple domain structure (cf. Fig. 1A). Upon optical excitation, at laser fluences below a sharp threshold of about 5 mJ/cm², single laser pulses induce local magnetic domain wall. At laser fluences above the threshold, a single laser pulse generates a network of magnetic vortex/anti-vortex (V/AV) structures, as depicted in Fig 1B-D. Subsequent laser pulses lead to nearly complete rearrangement of the V/AV network (left panels in Fig 1C and D). While the network is stable without optical excitation and shows no discernible dynamics on timescales of minutes to hours, V/AV annihilation can be triggered by illuminating the sample with laser pulses below threshold. After several low fluence optical pulses, the equilibrium ripple domain structure is recovered. The generation of a V/AV-network is remarkable as it presumably is the result of a partially melted, non-equilibrium spin system which is quickly quenched to a metastable state. Possible processes leading to a V/AV network are discussed on the basis of micromagnetic simulations and with respect to ultrafast all-optical pump-probe experiments. The nature and dynamics of the laser-driven magnetic reorganization will be further experimentally investigated with temporally-structured illumination utilizing femtosecond pulse pairs separated by variable time delays. In conclusion, we report the optically-induced vortex/anti-vortex generation mapped by in-situ Lorentz microscopy and discuss possible pathways for their generation.

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Fig. 1: (A) Electron micrograph with Lorentz contrast prior to optical excitation. (B-D) After single-fs-pulse laser excitation a network of vortices and anti-vortices appears (C, left panel). The reconstructed in-plane magnetization is displayed in (C, right panel) and (B). Subsequent laser pulses lead to a rearrangement of this network (D).