Supplementary materials for "Nonresonant nonlinear magnonics in an antiferromagnet"

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Phenomenological fitting of the Kerr rotation dynamics

We use an error function convoluted with a two-exponential decay function to fit the non-oscillatory part of the Kerr rotation dynamics:

$$A_{decay}(t) = a(erf(bt) + 1)(e^{-t/t_s} + ce^{-t/t_f}). \tag{S1}$$

The error function describes the rise dynamics from the convolution of the pump and probe pulses, and t_s, t_f correspond to the slow and fast decay dynamics, respectively. Also, a, b are fitting parameters.

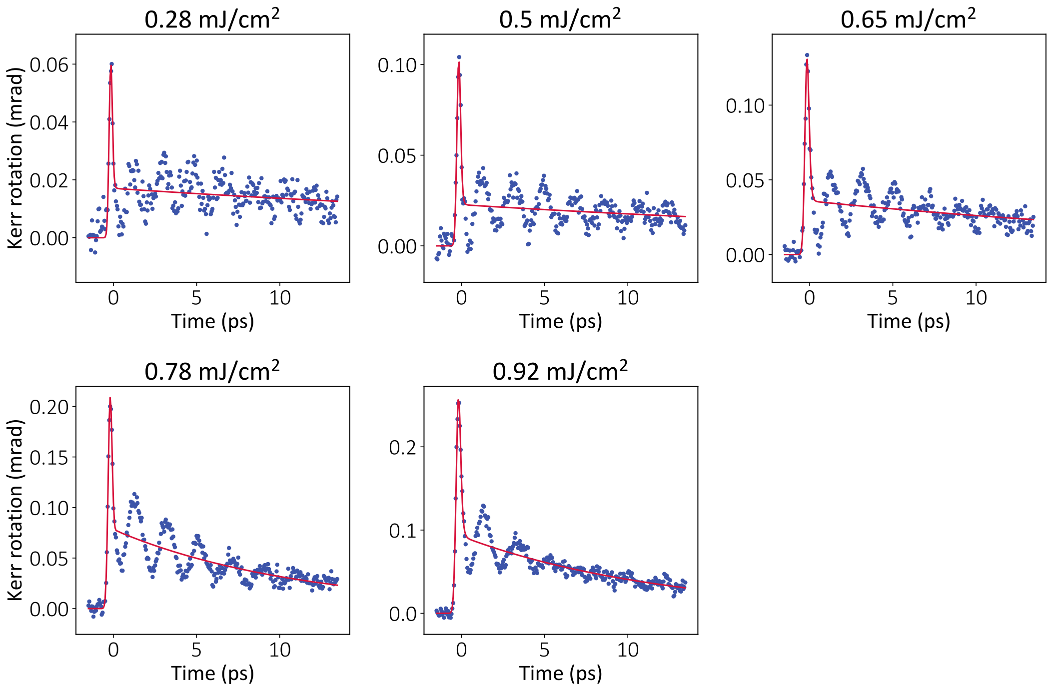
Supplementary Fig. 1 plots the phenomenological fits of below-gap 9 μm pump-induced Kerr rotation dynamics as a function of pump excitation fluence using eq. S1. Fluence dependence of the fitting parameters a, t_s and t_f are also presented in Supplementary Fig. 2. Similarly, the temperature dependence of the dynamics and fitting parameters are depicted in Supplementary Figs. 3 and 4, respectively.

On the other hand, phenomenological fitting of above-gap 1.3 μm pump-induced Kerr rotation dynamics as a function of pump excitation fluence is shown in Supplementary Fig. 5. We note that early time fast Kerr rotation dynamics of 1.3 μm pump is very different from that of 9 μm and it cannot be fitted with two-exponential decay function. Instead of fitting the whole pump probe dynamics, we pick the oscillatory part after the first peak with pump-probe delay time > 1.5 ps for fitting.

Phase of the linearly polarized 9 μm response

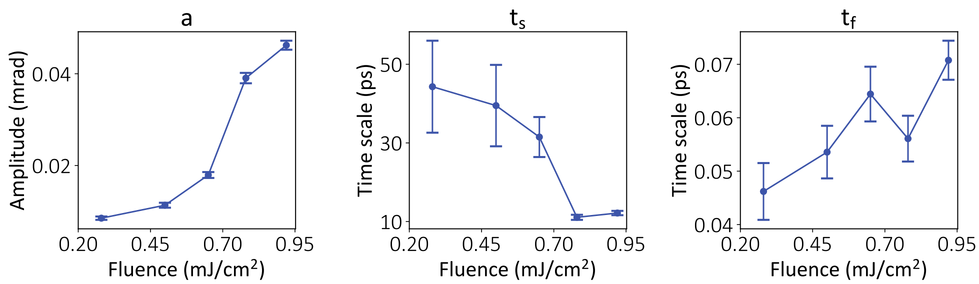
The linearly polarized 9 μm data are shown in Extended Data Fig. 5c,d of the main manuscript. The Fourier-amplitude spectrum contains weak spectral weight near 0.5 THz; however, the Fourier magnitude does not determine the oscillation phase, and the small oscillatory signal and nonoscillatory background do not permit a reliable phase extraction. We therefore do not assign a phase to this response.

Fitting of fluence-dependent 9 μm pump - Kerr rotation probe dynamics



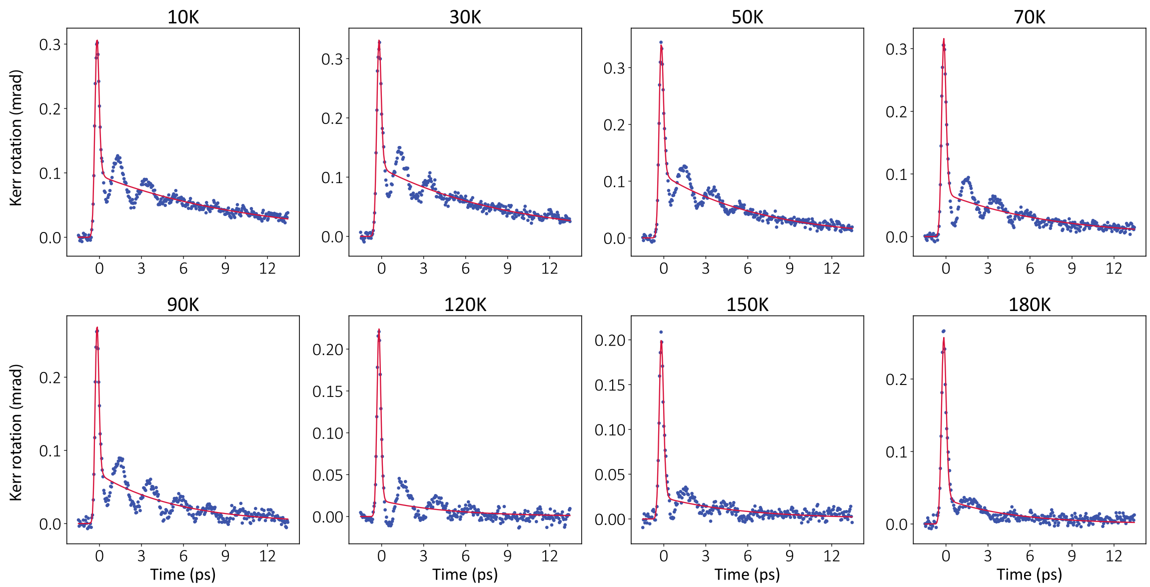
Supplementary Fig. 1. The fitting of the non-oscillatory part of the fluence-dependent 9 μm pump - Kerr rotation probe dynamics. Blue dots are the experimental data points, red lines are the fitting results.

Fluence dependence of 9 μm pump fitting parameters



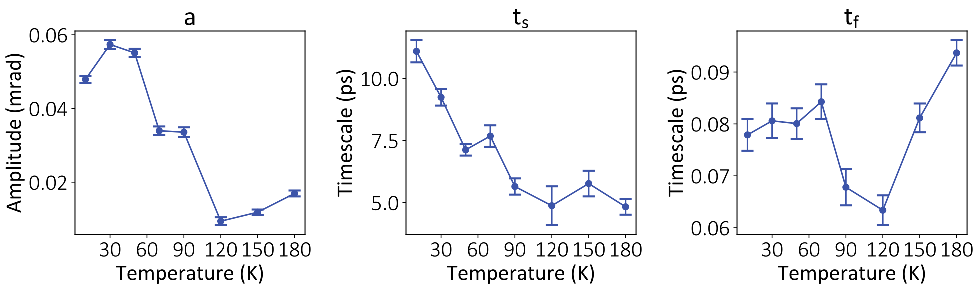
Supplementary Fig. 2. The fluence-dependent fitting parameters: amplitude a, slow decay t_s, fast decay t_f.

Fitting of temperature-dependent 9 μm pump - Kerr rotation probe dynamics



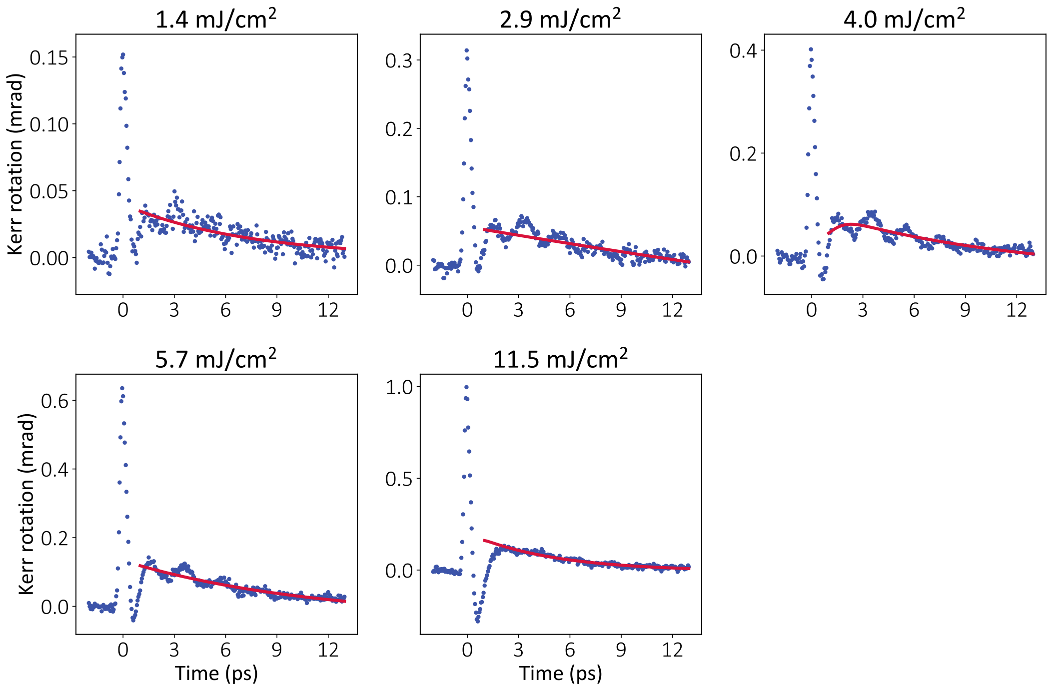
Supplementary Fig. 3. The fitting of the non-oscillatory part of the temperature-dependent 9 μm pump - Kerr rotation probe dynamics. Blue dots are the experimental data points; red lines are the fitting results.

Temperature dependence of 9 μm pump fitting parameters



Supplementary Fig. 4. The temperature-dependent fitting parameters: amplitude a, slow decay t_s, fast decay t_f.

Fitting of fluence-dependent 1.3 μm pump - Kerr rotation probe dynamics



Supplementary Fig. 5. The fitting of the non-oscillatory part of the fluence-dependent 1.3 μm pump - Kerr rotation probe dynamics. Blue dots are the experimental data points, red lines are the fitting results.