

Mid-infrared element-specific selective manipulation of the ultrafast magneto-optical Kerr effect dynamics in the ferrimagnet FeCr_2S_4

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FeCr_2S_4 is a ferrimagnet with two oppositely ordered spin sublattices (Fe and Cr), connected via superexchange interaction, giving a nonzero net magnetic moment. We show, using time-resolved measurements of the magneto-optic Kerr effect, how the magnetic dynamics of the sublattices can be selectively manipulated by resonantly perturbing the Fe sublattice with ultrashort laser pulses. The mid-infrared excitation through intra-atomic Fe d - d transitions triggers markedly slower dynamics in comparison to an off-resonant pumping affecting both of the two sublattices simultaneously. By changing probe wavelength to move in and out of resonance with the Fe d - d transitions, we also show the specific contributions of the Fe sublattice to these dynamics.

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Ferrimagnets are an interesting class of magnetic materials that combine some features of antiferromagnets with those of ferromagnets. They are characterized by a finite net magnetic moment given by sublattices with opposite spin orientation but unequal magnetic moments. The relative orientation of the magnetic sublattices is typically determined by the exchange interaction [1]. This leads to intriguing potential applications in spintronics [2], spin textures such as domain walls and skyrmions [3], and in some cases the phenomenon of all-optical switching [4]. For light with photon energies near intra-atomic electronic transitions involving partially filled shells, strong element-specific enhancement of the magneto-optical Kerr effect (MOKE) response can be observed [5–8]. In principle, exciting and monitoring the material resonantly to such transitions via light pulses in a time-resolved approach (TR-MOKE [9,10]) offers a unique pathway for selective manipulation of a single sublattice, provided that other contributions of mixed atomic character are distinctively separated in the energy spectrum. Past studies have shown that resonant pumping can improve the efficiency of processes such as spin-wave excitation and switching, while resonant probing

can help to distinguish the sublattice dynamics [11–14]. To the best of our knowledge, however, no studies so far have reported how the individual sublattice dynamics vary as a result of resonant pumping.

The ferrimagnet FeCr_2S_4 is a semiconductor with a cubic spinel structure (space group $Fd\bar{3}m$) [Fig. 1(a)] [15–18], with some reports suggesting structural distortions below 65 K [19–21]. The iron cations (Fe^{2+}) are found in tetrahedral (T_d) sites, while the chromium ions (Cr^{3+}) are octahedrally (O_h) coordinated. A mixed valence configuration for the Fe and Cr cations was excluded by Chen *et al.* [16]. Recently, the possibility of growing 2D-like crystals with analogous magnetic properties has been demonstrated, which makes the material attractive for applications in ultrathin devices [22]. FeCr_2S_4 becomes ferrimagnetic below the ordering temperature $T_C \approx 170$ K. This alignment is favored by the superexchange interaction between the half-filled Cr t_{2g} orbitals and the partially filled Fe d orbitals, mediated by the sulfide S^{2-} anions [1,23,24]. Polar MOKE studies on bulk samples have revealed a large, temperature-dependent MOKE rotation in the mid-infrared around photon energies of approximately 0.30 eV, enhanced by intra-atomic Fe d - d transitions that become dipole-active due to the absence of local inversion symmetry at the iron site, leading to Davydov splitting [6,7]. Differently, Cr d - d transitions have a much smaller impact, comparable in magnitude to other nonresonant contributions, around 1.60 eV, at the edge of the visible range of the spectrum [7,8]. Around 3.10 eV, the MOKE rotation is about one order of magnitude smaller than in the mid-infrared, and mediated by charge-transfer (CT) transitions between S-3p orbitals and mixed

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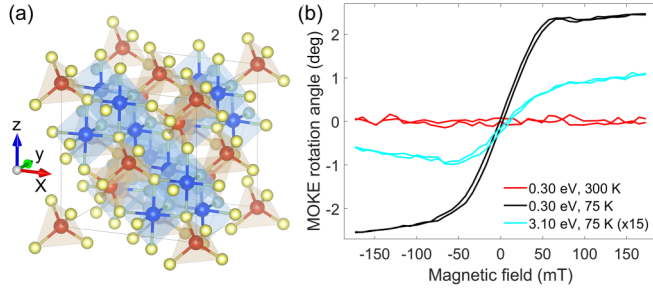


FIG. 1. Equilibrium properties of FeCr_2S_4 . (a) Crystallographic cubic unit cell, where the Fe atoms are in red, Cr atoms are in blue, and S atoms are in yellow based on the data from Ref. [15] and plotted using VESTA [26] (b) MOKE rotation hysteresis cycles for 0.30 eV (Fe d - d transitions) and 3.10 eV (CT transitions) for $T < T_C \approx 170$ K; for comparison, the hysteresis for $T > T_C$ at 0.30 eV is also shown.

Fe-3 d /Cr-3 d states [6,7,23,25]. The strong enhancement of the magneto-optical coupling at photon energies in resonance with Fe d - d transitions offers an attractive mechanism for both the manipulation and detection of the Fe sublattice. In this Letter, we report on polar TR-MOKE experiments on single-crystal FeCr_2S_4 samples with pump and probe energies both on- and off-resonance with respect to the intra-atomic Fe d - d transitions. We find significant changes in the dynamics as a function of the wavelength of the pump pulse. In particular, we observe a dramatically slower response in the magnetic dynamics when the pump photon energy is tuned to resonance with the Fe d - d transitions. Different probe energies allow us to analyze the roles of the sublattices in the dynamics.

The single-crystal samples were grown using chemical vapor transport as reported by Ohgushi *et al.* [6]. All of our optical measurements were performed on the as-grown (111) surface. Although the easy axis corresponds to the $\langle 100 \rangle$ direction, mechanical polishing has been reported to cause a marked increase in the coercive field [6,7]. This preferential alignment is connected to the temperature-dependent magnetocrystalline anisotropy energy, mainly related to the Fe sites [27]. The experiments were performed using an amplified Ti:sapphire laser system (Coherent Astrella, photon energy 1.55 eV, pulse duration 100 fs, and repetition rate 1 kHz) in combination with an optical parametric amplifier (TOPAS, Light Conversion). Mid-infrared pulses at 0.30 eV were achieved by difference-frequency generation of the signal and idler outputs of the optical parametric amplifier using a gallium selenide crystal, while visible 3.10 eV pulses were obtained by second-harmonic generation through a beta barium borate crystal. More details on the experimental setup, list of the pump and probe spot sizes at the sample position, power reflectivity coefficients, and intensity attenuation lengths are reported in Secs. I and II of the Supplemental Material [28].

In Fig. 1(b), we report the polar MOKE response from FeCr_2S_4 at 0.30 and 3.10 eV through hysteresis cycles. The large few-degree mid-infrared MOKE rotation at 0.30 eV dwarfs the visible response at 3.10 eV at 75 K. The hysteresis vanishes above the critical temperature $T_C \approx 170$ K. These results are consistent with those reported by Ohgushi *et al.* [6], where the enhanced response at 0.30 eV was attributed

TABLE I. Time decay constants for the dynamics near time zero, derived using the fit function reported in the text for the lowest and highest investigated fluences from Figs. 2(a) and 2(b).

Photon energy (eV)	Fluence (mJ/cm ²)	Decay time τ (fs)
3.10	0.5	260 ± 60
3.10	1.9	210 ± 30
1.55	0.4	200 ± 80
1.55	5.0	160 ± 20
0.30	1.7	1600 ± 500
0.30	6.4	890 ± 60

to the strong contributions of intra-atomic Fe d - d transitions, whereas the smaller response at 3.10 eV was ascribed to contributions from interatomic CT transitions. In the following, we focus on the time-resolved MOKE rotation dynamics, in particular on the out-of-equilibrium signal that is odd with respect to the externally applied magnetic field. To obtain it, we acquired pairs of datasets with a magnetic field of ≈ 170 mT, but with opposite directions with respect to the surface normal, namely, θ_+ and θ_- . We express the signal as $\Delta(\theta_+ - \theta_-)/(\theta_+ + \theta_-)$.

We start by considering the case of a nonresonant probe (3.10 eV), where the magneto-optic effect is mediated by CT transitions. We explored three distinct time ranges with different time steps for selected incident fluence values [Figs. 2 and 4(a)]. Three pump photon energies were employed: 3.10, 1.55, and 0.30 eV. In all these cases, the probe attenuation length is smaller than or equal to that of the pump. The left side of the panels in Fig. 2 shows the initial dynamics over a 6 ps window around the temporal overlap between pump and probe pulses ($t = 0$). In the nonresonant cases [3.10 and 1.55 eV, panel (a)], a sudden drop occurs at time zero, followed by slower dynamics, which becomes more pronounced as the excitation energy increases. A major difference occurs once the pump photon energy is resonant with the Fe d - d transitions [0.30 eV, panel (b)]. Here, the dynamics do not exhibit a sharp drop at time zero, but rather a more gradual decrease. After this, the response then follows a trend similar to that observed for nonresonant pumping after about 2 ps. To describe the response after time zero, we use the empirical fit function $f(t) = G(t) \times \{\Theta(t)[A(1 - e^{-t/\tau}) + Bt + C]\}$, where $\Theta(t)$ is the Heaviside function, $G(t)$ is a Gaussian with unit area whose full width at half maximum (FWHM) is determined by the pump and probe pulse durations, A , B , and C are fit parameters representing the relative amplitudes of the components of the response, and τ is a time constant. The time constants for the lowest and highest fluence values from each set of data among panels (a)–(c) are reported in Table I. For all the wavelength combinations, the FWHM of $G(t)$ was estimated to be shorter than 200 fs. More details on the fit results and pulse lengths can be found in Sec. I of the Supplemental Material [28]. On the right side of the panels in Fig. 2, we show the TR-MOKE rotation dynamics over a 150 ps range. Here, together with a gradual recovery MOKE signal after the excitation, we observe the emergence of a 24 GHz oscillation, which disappears at high fluences for all the three pump photon energies. In the longest time-delay window [1600 ps,

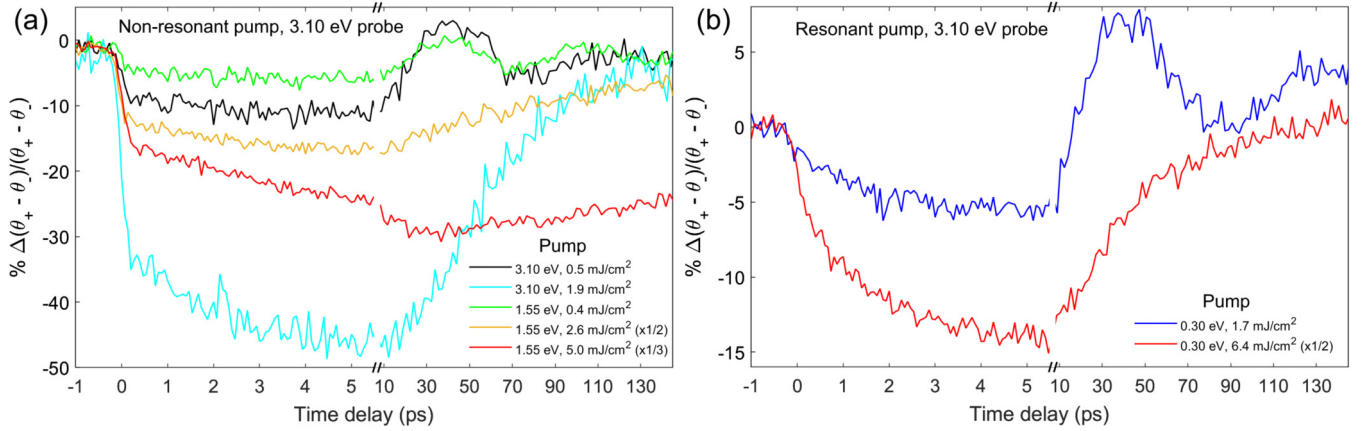


FIG. 2. TR-MOKE signal for FeCr₂S₄ using (a) nonresonant 3.10 or 1.55 eV pump photon energy and (b) resonant 0.30 eV pump photon energy, together with a nonresonant 3.10 eV probe photon energy. All the measurements were acquired at $T = 75$ K.

Fig. 4(a)], the response is similar for all pump photon energies, consisting of a slow monotonic recovery after the initial excitation. As the pump fluence is increased, such as in the 1.55 eV case, the recovery in the first hundreds of picoseconds becomes at first slower, but then follows dynamics similar to that observed for the lower fluence.

We now turn our attention to the case of probe photon energy resonant to the Fe $d-d$ transitions [0.30 eV, Figs. 3 and 4(b)]. Again, three time ranges were collected, albeit only for two pump photon energies (1.55 and 0.30 eV). We chose not to acquire data for a pump photon energy of 3.10 eV since its attenuation length is approximately a factor of 10 smaller than at 0.30 eV. Starting from the 6 ps window shown on the left side of the panels in Fig. 3, the response is predominantly flat after excitation. For the 1.55 eV case, a small prepulse, arriving 19 ps before and estimated to be eight times smaller than the main pulse, was present in the pump. For degenerate pump and probe photon energies at 0.30 eV, it was not possible to fully remove the contribution of interference effects, which created coherent artifacts, localized mainly around the first few hundreds of femtoseconds after time zero [Figs. 3(b) and 4(b)] [29,30]. Their identification is described in Sec. III of the Supplemental Material [28]. We believe that this effect

is not strongly present in the degenerate case at 3.10 eV [Fig. 2(a)], since the pump and probe beams were generated by two distinct nonlinear crystals leading to different time profiles [29]. Using the mid-infrared pump, although limited by the coherent artifacts at time zero, we do not observe a slow decrease of the out-of-equilibrium signal as in the case of the visible probe [Fig. 2(b)]: After 1 ps the response is already flat. Over the 150 ps range (right side of the panels in Fig. 3), we observe a response similar to that of the visible probe: A 24 GHz oscillation is present at the lower fluences, which gradually disappears, together with a strong signal decrease at the highest excitations. A comparison of the data shown in Figs. 3 and 4(b) shows that the 24 GHz oscillation exhibits a gradual, strong, redshift to around 2 GHz at the highest fluences.

One major contribution to the differences among the time dynamics at similar excitation fluences likely stems from the strong differences in attenuation length. This may explain, for example, why, for a 1.55 eV excitation with a given fluence, the 0.30 eV probe reveals dynamics, which resembles those measured at lower fluences for the 3.10 eV probe [Figs. 2(a) and 3(a)]. A similar argument applies for the differences among distinct pump photon energies, but same probe,

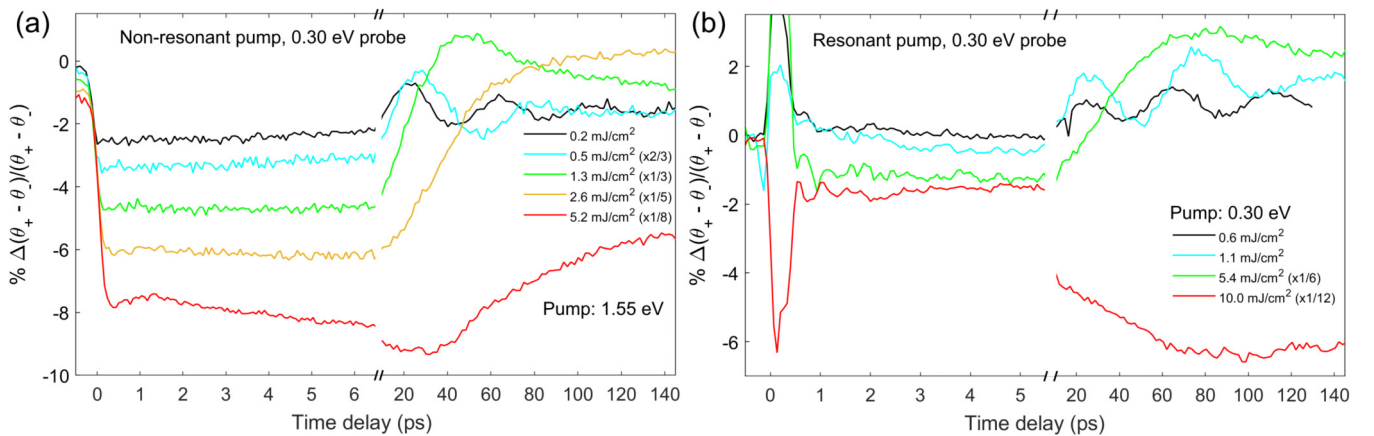


FIG. 3. TR-MOKE signal for FeCr₂S₄ using (a) nonresonant 1.55 eV pump photon energy and (b) resonant 0.30 eV pump photon energy, together with a resonant 0.30 eV probe photon energy. All the measurements were acquired at $T = 75$ K.

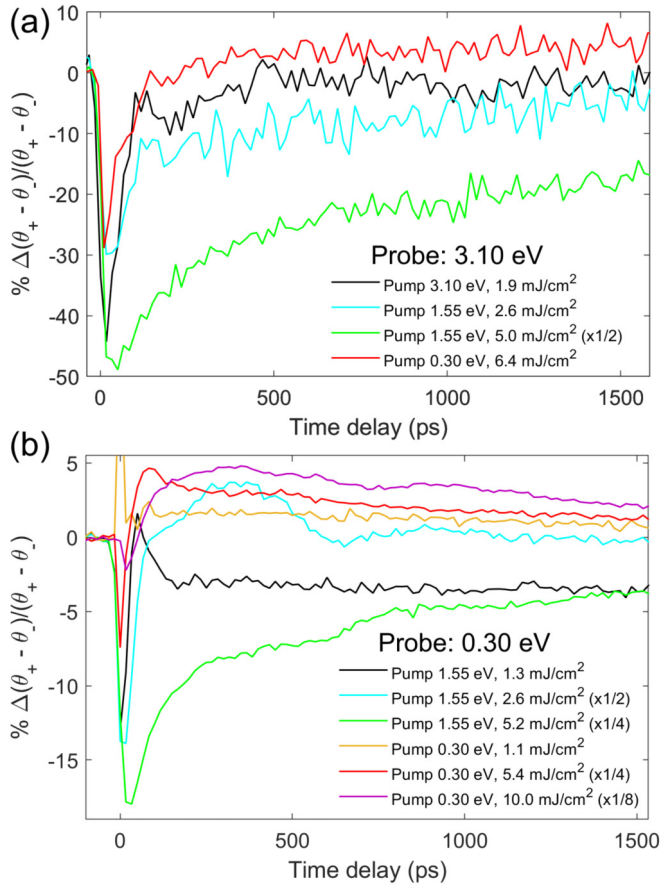


FIG. 4. Long-delay TR-MOKE signal for FeCr_2S_4 using (a) 3.10 eV or (b) 0.30 eV probe photon energy. All the measurements were acquired at $T = 75$ K.

even after taking into account their reflectivities. However, the dynamics of a material is determined not simply by the absorbed energy, but also by the orbitals and quasiparticles involved. An example is the TR-MOKE dynamics in Fig. 2 in the first picoseconds. Considering the average absorbed excitation energy density, the highest fluence for the 3.10 eV pump in panel (a) corresponds to $9.8 \mu\text{J}/\text{m}^3$, whereas it is $4.3 \mu\text{J}/\text{m}^3$ for 1.55 eV excitation in panel (b) and $2.2 \mu\text{J}/\text{m}^3$ for 0.30 eV excitation in panel (c). Despite this, even decreasing the pulse energy by a factor of 10 for the 1.55 eV pump does not reproduce the slower dynamics seen for the 0.30 eV pump. As shown for antiferromagnetic iron oxides [13], resonant pumping of Fe $d-d$ transitions can weaken the magnetic interactions by altering the electronic configuration of the sublattices. This can transiently make a different alignment more favorable. The data of Fig. 2 show that, using a 3.10 eV probe to monitor the response sensitive to modifications that occur in both sublattices [6,7,23,25], a 0.30 eV excitation leads to a slower TR-MOKE dynamics in comparison to other excitation photon energies that are not resonant with the intra-atomic Fe $d-d$ transitions. This dependence of the speed of the initial response on the pump photon energy does not occur when probing resonantly to the Fe sublattice (Fig. 3), where a fast drop was observed regardless of the selected pump. This suggests that the slower response in the

first picoseconds of Fig. 2(b) is connected to a change in the superexchange interaction between the excited Fe states and the Cr sublattice, triggered by the selective modification of the Fe electronic configuration. In fact, excitations at either 1.55 or 3.10 eV photon energies perturb directly both the two sublattices, not relying mainly on an indirect Cr spin reorganization via superexchange [7,23]. In the simplified semiclassical picture of the Heisenberg model, these pump-induced changes can be viewed as variations of the effective magnetic fields generated by each sublattice, which depend on the exchange interaction and spin orientation [1,25]. Interaction with the phonon bath is expected to occur on a longer time scale [31].

The 24 GHz oscillation was previously reported by Ogasawara *et al.*, and assigned to the precession of Cr spins [25]. This was motivated by the lack of oscillations at 0.5 eV probe photon energy (pump at 1.55 eV), ruling out a significant role of the Fe spins, limited by the relatively high magnetocrystalline anisotropy of this material [27]. Despite this, our data in Fig. 3 clearly show oscillations at 24 GHz when probing resonantly to the Fe sublattice (0.30 eV). These experimental results can be reconciled by noting that a photon energy of 0.5 eV is at the very edge of the MOKE resonance associated with the Fe $d-d$ transitions [6,7,25]. Thus, MOKE data at this photon energy may have other contributions that are not specific to the Fe sublattice. Moreover, in Ogasawara *et al.* [25], a slight oscillation appears to be in fact present in the datasets at 70 and 90 K. The crystalline anisotropy, although predicted to be on the order of a few tesla at 4 K [25], is heavily influenced by temperature [27]. This is also evident from the coercive field growth while cooling [22]. Therefore, we conclude that under our conditions ($T = 75$ K), the magnetocrystalline anisotropy, while present, is considerably reduced. Since the oscillation frequency is close to that observed when probing at low fluences for 3.10 eV (Fig. 2) and 1.9 eV [25], we conclude that the two sublattices are coupled in their precession motion by the superexchange interaction. The precession oscillations occur at different fluence conditions depending on the photon energy combination due to the different attenuation lengths, as discussed in the previous paragraph. No further oscillatory contributions were observed (see Sec. IV in the Supplemental Material [28]). For the nonoscillatory component of the long-time dynamics shown in Fig. 4, there are marked differences in the dynamics observed with 0.30 eV probe for the two different pump photon energies. On these time scales, we expect that the pump excitation has locally thermalized, but with a strong depth gradient that slowly relaxes via diffusion. This depth gradient may influence strongly the magneto-optic response, causing the strong dependence on pump wavelength that we observe.

Additional data on the FeCr_2S_4 dynamics can be found in the Supplemental Material [28]. Section V of the Supplemental Material [28] presents out-of-equilibrium hysteresis curves. In Sec. VI of the Supplemental Material [28], we report the TR-MOKE ellipticity. Here, we also observe slower dynamics for the resonant 0.30 eV pumping compared to the nonresonant pumps at 1.55 and 3.10 eV. This confirms that a nonmagnetic response is not responsible for the differences in the dynamics between rotation and ellipticity signals [10,32]. Finally, in Sec. VII of the Supplemental Material [28],

time-resolved reflectivity data show a non-negligible impact of the external magnetic field for temperatures below T_C .

In summary, we have shown that, by selectively pumping the Fe sublattice in the mid-infrared, we are able to modify dramatically the timescale of the TR-MOKE response connected to both the Fe and Cr sublattices, compared to nonresonant case. We propose that the possible mechanism consists of a distinct modification of the superexchange interaction, based on which electronic levels are perturbed by the pump. Moreover, our Letter highlights the importance of resonant probing to distinguish the dynamics of the sublattices and to understand whether their spins are involved in specific collective modes. This approach can be extended to other magnetic materials provided that a clear spectral

separation among the localized energy levels of the atoms is present.

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Data availability. The data that support the findings of this article are openly available [33].

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