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Substrate Contribution to Ultrafast Spin Dynamics in 2D van der Waals Magnets

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We propose a model that is able to reproduce the type-II ultrafast demagnetization dynamics observed in 2D magnets. The spin system is coupled to the electronic thermal bath and is treated with atomistic spin dynamics, while the electron and phonon heat baths are described phenomenologically by coupled equations via the two-temperature model. Our proposed two-temperature model takes into account the effect of the heated substrate, which for 2D systems results in a slow demagnetization regime. We applied the framework to a generic 2D system, CrI_3, and we are able to observe a type-II demagnetization process characterized by two steps, the first step being attributed to the free electrons generated when the system behaves as a quasimetal under optical excitation, and the second step, the slower demagnetization region, due to the heated substrate. Finally, after laser excitation, we are able to observe domain formation in CrI_3, similar to recent experimental observations. Our generalized two-temperature model enhances the modeling of ultrafast magnetization dynamics processes by effectively describing the quasimetallic behavior of certain magnetic materials and the influence of a heated adjacent layer.

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Since the discovery of magnetic ordering in two-dimensional systems [1,2], the family of two-dimensional van der Waals magnetic materials (2D vdW) has been considerably enriched, comprising now all types of magnetic ordering including ferromagnets [1,3,4], antiferromagnets [5–7], and even ferrimagnets [8]. Recently, the interaction of 2D materials with ultrafast laser pulses has been investigated both experimentally and theoretically [9–13]. Although many 2D materials have low Curie temperatures which makes the application of intense laser pulses without causing damage difficult, there are several materials such as Fe_3GeTe_2 [4] that present a Curie temperature close to room temperature or in which it is possible to increase the Curie temperature by interface engineering [14] in applications of heterostructures [15]. Hence, devices based on ultrafast excitation of 2D systems can become feasible in the near future.

The control of magnetization via ultrafast laser pulses first shown by Beaurepaire *et al.* [16] has led to crucial discoveries, such as the ability to switch the magnetization in ferrimagnets in the absence of a magnetic field [17] or the possibility to control magnetization via phonons [18]. In terms of theoretical modeling, either a two-temperature (2TM) or a three-temperature (3TM) model is usually employed to approximate the temperature dynamics, where the dynamics of phonons and electrons (and spins in the case of the 3TM) is determined by solving phenomenological coupled equations of the variation of temperature within these subsystems. Generally speaking, in response to a femtosecond laser pulse the 2TM produces a rapid increase of electron temperature to values of around 10^3 K or higher followed by a much slower increase in the phonon temperature. This leads to the continuous quenching and recovery of the magnetization as seen in the pioneering experiment of Beaurepaire *et al.* [16] and corresponds to a type-I demagnetization process [19]. When applying a laser pulse to 2D vdW materials such as CrI_3 [9], Fe_3GeTe_2 [13], and $CrGeTe_3$ [20], these exhibit type-II magnetization dynamics [19]. For a type-II dynamics, a two-step demagnetization process is present: first, the laser pulse provides an initial fast demagnetization and increase in the equilibrium phonon and electronic temperatures followed by a subsequent slower demagnetization at the increased equilibrium temperature.

The difference between type-I and type-II demagnetization processes can also be understood in terms of the timescales and coupling between the three subsystems involved: electrons, spins, and phonons [21]. Type I is observed in materials where the coupling between the electron and spin system is strong, hence the electron-spin equilibration time is shorter than the electron-phonon equilibration time, resulting in the spin temperature being higher than the phonon temperature. For materials exhibiting type-II behavior, the coupling between the electron and spin

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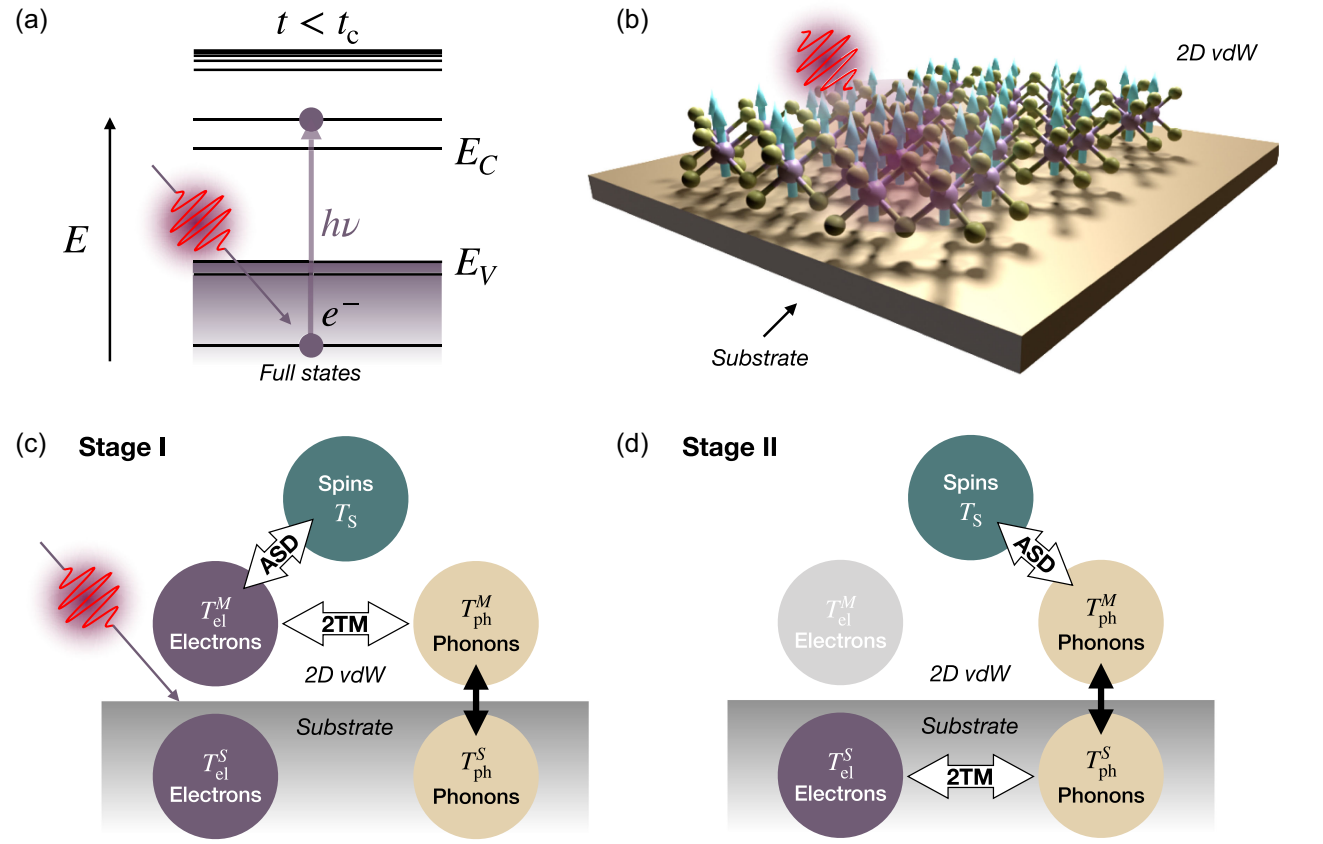

FIG. 1. Illustration of the generalized two-temperature model. Panel (a) shows the quasimetallic nature of CrI_3, with electrons being excited only for a short time of $t < t_c$. In panel (b) is an illustration of a CrI_3 monolayer deposited on a substrate under laser-pulse excitation. Panels (c) and (d) show schematics of the coupled two-temperature model (2TM) applied to both substrate and the magnetic 2D layer and the energy channels involved in the process [electrons represented by T_{el}, phonons T_{ph} and spins T_S of the for the magnetic system (M) and substrate (S)].

system is weak, resulting in faster electron-phonon equilibration and consequently a phonon temperature higher than the spin temperature. In this case, phonons can act as a heat bath for the spin system and contribute to a two-step demagnetization process. It is also possible to see a transition from type-I to type-II ultrafast demagnetization [21–23] by changing the laser fluence or ambient temperature which would affect the coupling and timescales between the three subsystems.

2D vdW materials are in general deposited on a substrate and their low dimensionality can allow the substrate temperature to increase to high values maintained over a long period of time. We propose that this is the source of the second regime of slower demagnetization. The importance of considering the substrate properties has also been suggested in other works, on similar topics that investigate ultrafast magnetization switching. For example, angular momentum transfer from phonons in the substrate can lead to magnetization switching via the Barnett effect [24]. Since 2D magnetic materials have the benefit of use in heterostructures, where each layer has a different functionality, the effect of heated adjacent layers can be considered similarly using the generalized two-temperature model presented here.

The prototypical 2D magnetic material is CrI_3. It exhibits interesting magnetic effects, containing a ferromagnetic order at the monolayer limit, however, for even numbers of atomic layers, an antiferromagnetic order is stabilized [3]. In the CrX_3 family, $CrBr_3$ and $CrCl_3$ are considered to be ferromagnetic insulators while studies show CrI_3 to be either insulator [25] or a ferromagnetic semiconductor [26], and during the laser excitation is considered a quasimetal [27]. As an insulator or semiconductor the system couples weakly to incoming laser light, but intense pulses can transiently promote electrons from lower energies to the conduction band, as illustrated in panel (a), Fig. 1, leading to a sudden change in the interaction between the spin and electron system and an elevated electron temperature. This can lead to an ultrafast demagnetization process, followed by slow cooling after the laser pulse as the electrons suddenly equilibrate back to their ground state and the dynamics become dominated by the slow spin-phonon coupling and naturally leading to type-II behavior. In the following we outline the modified 2TM as illustrated

in Fig. 1 and carry out atomistic calculations which reproduce the type-II behavior in CrI_3 and also produce the domain structures observed experimentally by [10]. For completeness, we also apply the generalized 2TM on another 2D system, $CrGeTe_2$ proving the capabilities of the framework to model 2D systems in general, results being provided in Supplemental Material [28].

In order to model the ultrafast demagnetization in a 2D vdW system we employ a generalized two-temperature model (2TM) based on the work of Chen *et al.* [29]. During the laser excitation, the photon energy is transferred from the laser pulse to the electrons, and then equilibrates with the phonons and the spin system. The spin system is coupled to the electronic thermal bath and is treated within the atomistic spin dynamics via the software package VAMPIRE [30,31], while the electron and phonon heat baths are described phenomenologically by coupled equations as described in [29].

In the case of CrI_3, the system behaves as a quasimetal [27] hence we consider that electrons are excited only for a few hundred femtoseconds, which in our model is introduced as a cutoff time t_c, which practically implies connecting the spin system to the electronic thermostat only for the respective cutoff time, but heating the substrate for a longer time. An illustration of this process is shown in Fig. 1, stage I, panels (a) and (c). CrI_3 behaves as a quasimetal only for a finite amount of time, afterward the laser pulse cannot couple anymore to the electrons in the system and the material is heated up only by the substrate. We propose that the type-II magnetization dynamics appears in these materials as an effect of the heated phonons, since the substrate can couple over a longer timescale to the laser pulse. As the substrate has a different specific heat capacity compared to the 2D magnetic system, it will heat up more slowly and will lead to a slow demagnetization of the magnetic system, corresponding to stage II of the process illustrated in Fig. 1(d). Hence, the magnetization dynamics in 2D vdW materials can be described via a two-temperature model provided the quasimetallic nature of the CrI_3 system and heating from the substrate are included. These assumptions have been incorporated in the following coupled equations for the magnetic system (M) and for the substrate (S). During the times $t < t_c$ where the system behaves as a quasimetal we solve the following coupled equations and couple the magnetic system to the electronic temperature:

$$C^M_{\mathrm{el0}} T^M_{\mathrm{el}} \frac{dT^M_{\mathrm{el}}}{dt} = -G^M_{\text{el-ph}}\left(T^M_{\mathrm{el}} - T^M_{\mathrm{ph}}\right) + P(t), \qquad t < t_c \quad (1)$$

$$C^M_{\mathrm{ph}} \frac{dT^M_{\mathrm{ph}}}{dt} = -G^M_{\text{el-ph}}\left(T^M_{\mathrm{ph}} - T^M_{\mathrm{el}}\right), \qquad t < t_c \quad (2)$$

where C^M_{el0}, C^M_{ph} are the electron and phonon heat capacity of the magnetic system, $G^M_{\text{el-ph}}$ represents the electron-phonon coupling factor, T^M_{ph}, T^M_{el} the phonon and electron temperatures of the magnetic system and $P(t)$ is the time dependent laser pulse power. After the electronic transitions have subsided, the magnetic system will be coupled to the temperature of the heated substrate T^S_{ph}. At this point no more energy will be deposited in the system. The substrate has different heat capacities and electron-phonon coupling factors. Similarly we solve the two-temperature model for the substrate:

$$C^S_{\mathrm{el0}} T^S_{\mathrm{el}} \frac{dT^S_{\mathrm{el}}}{dt} = -G^S_{\text{el-ph}}\left(T^S_{\mathrm{el}} - T^S_{\mathrm{ph}}\right) + P(t), \qquad t > t_c \quad (3)$$

$$C^S_{\mathrm{ph}} \frac{dT^S_{\mathrm{ph}}}{dt} = -G^S_{\text{el-ph}}\left(T^S_{\mathrm{ph}} - T^S_{\mathrm{el}}\right). \quad (4)$$

For simplicity, we consider in the first instance a nonmagnetic substrate that is described in terms of its thermal properties: electronic specific heat C^S_{el0}, phononic specific heat C^S_{ph}, and electron-phonon coupling $G^S_{\text{el-ph}}$ [Eqs. (3) and (4)—superscript "S" referring to substrate] that enters into the generalized two-temperature model described above. In Ref. [10], the substrate is considered a hexagonal boron nitride, which was placed on a SiO_2/Si substrate. A similar substrate was also considered in [20]. As details of these properties for realistic substrates is limited, we have used generic parameters to show a proof of concept, such as decreased electron-phonon coupling. It has been recently proposed that certain resonant optical transitions can influence the spin dynamics via spin-flip excitons [32]. The spin-flip excitons could enhance the coupling between electronic and spin degrees of freedom, potentially increasing the efficiency of demagnetization within the two-temperature framework. Similarly, the substrate could potentially affect these resonances, modulating the effective electron-phonon coupling used in the model and therefore influence the magnitude of demagnetization.

The electronic or phononic temperature enters into the thermal field used in the Landau-Lifshitz-Gilbert equation. The laser power density takes a Gaussian form $P(t) = (2F_0/\delta t_p\sqrt{\pi/\ln 2})\exp[(-4\ln 2)(t/t_p)^2]$ where F_0 is the laser fluence (in units of energy density), t_p the pulse temporal width and δ the optical penetration depth, assumed to be $\delta \sim 10$ nm. The 2TM can be extended to include a heat-sink coupling term, $-\kappa(T_p - T_0)$, which is a thermal diffusion to the environment. In the above equation, $\kappa = 2 \times 10^9\ \mathrm{s}^{-1}$ represents the thermal relaxation rate, and quantifies the rate at which the heat is transferred to the environment at temperature T_0, with T_p representing the phonon temperature. This allows for the magnetic system to relax at hundreds of picosecond-nanosecond timescales to the equilibrium environment temperature. For modeling 2D magnetic systems CrI_3 we use the atomistic spin dynamics (ASD) software package VAMPIRE [30,31]. ASD simulations assume a fixed lattice of atoms to which

is associated a magnetic moment or spin $\mathbf{S}_i = \boldsymbol{\mu}_i/\boldsymbol{\mu}_s$ that can precess in an effective field $\mathbf{H}_i$ according to the Landau-Lifshitz Gilbert equation:

$$\frac{\partial \mathbf{S}_i}{\partial t} = -\frac{\gamma}{(1+\lambda^2)}\mathbf{S}_i \times (\mathbf{B}_i + \lambda \mathbf{S}_i \times \mathbf{B}_i). \tag{5}$$

In our model, the Hamiltonian of the system contains a Heisenberg exchange term, biquadratic exchange, uniaxial anisotropy, and a Zeeman term as shown in Eq. (6):

$$\begin{aligned}\mathscr{H} = &-\frac{1}{2}\sum_{i,j}\mathbf{S}_i^{\alpha}\mathscr{J}_{ij}^{\alpha\beta}\mathbf{S}_j^{\beta} - \frac{1}{2}\sum_{i,j}K_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)^2 \\ &- \sum_i D_i(\mathbf{S}_i\cdot\mathbf{e})^2 - \sum_i \mu_i \mathbf{S}_i\cdot\mathbf{B},\end{aligned} \tag{6}$$

where i, j represent the atoms index, $\alpha, \beta = x, y, z$, $\mathscr{J}_{ij}^{\alpha\beta}$ represents the exchange tensor, K_{ij} the biquadratic exchange interaction, D_i the uniaxial anisotropy, which is orientated out of plane [$\mathbf{e} = (0, 0, 1)$] and $\mathbf{B}$ the external magnetic field applied during the field cooling.

The effective field $\mathbf{B}_i$ is calculated from the Hamiltonian of the model $\mathbf{B}_i = -(1/\mu_i)[\partial\mathscr{H}/\partial\mathbf{S}_i] + \xi_i$, augmented by a thermal noise ξ_i that acts as a Langevin thermostat and it is assumed to be a white noise.

We next apply the model for our choice of a 2D vdW material, CrI_3. During the time the system behaves as a quasimetal, the electrons are responsible for the thermalization of the system, afterward this role being attributed to the heated substrate as illustrated in Fig. 1. The temperature profile of the spin thermostat is given by numerically solving Eqs. (1)–(6) and then using the resulting electronic temperature for the spin dynamics. The obtained temperature profile leads to similar demagnetization profile results as observed experimentally, with a two-step demagnetization process, the first step being attributed to the free electrons generated when the system behaves as a quasimetal, and the second step, the slower demagnetization region, to the heated substrate. We next apply the generalized two temperature model to our spin dynamics calculations.

Figure 2 shows the temporal behavior of the phonon temperature [dashed lines—panels (a), (b)], spin temperature [continuous lines, panels (a), (b)], and magnetization [panels (c), (d)] after the application of a laser pulse of various fluences and 85 fs width. Other parameters used in the simulation are presented in Table 1, Supplemental Material [28]. The exchange interactions have been previously parametrized in [33,34] and have also been used in Ref. [10] for atomistic spin dynamics simulations. However, in comparison to Ref. [10] only a generic two-temperature model was been used, the resulting demagnetization being of type I only. For our improved simulations, the magnetization shows a very rapid decrease in the first picoseconds [Fig. 2, panel (c)], followed by a slower demagnetization up to 400 ps [specific to type-II demagnetization, Fig. 2, panel (d)] and then a slow recovery on the nanosecond timescale. This behavior is similar to what has been observed experimentally in [9] and specific to other materials in the class of 2D systems. Moreover, similarly to what has been observed in [10], after the laser excitation, the resulting magnetic configuration consists of domains [Fig. 2, panels (e) and (f)] in the case of increased laser pulse fluence. The fact that our results show both a type-II demagnetization and domain formation confirms the success of our generalized two-temperature model. To show the generality of our model, in Supplemental Material we also present the results of ultrafast magnetization dynamics in a different 2D magnetic material, $CrGeTe_2$—Fig. S1 [28]. Similarly a type-II demagnetization profile is observed.

The existence of domains in our system is also suggested in Fig. 2(b), in which we analyze the spin temperature T_S in relation to the phonon temperature T_{ph}. We observe that for laser pulse fluences where there is no creation of domains (such as $F = 0.1$ mJ/cm^2, blue lines), the spin and phonon temperature equilibrate very quickly, while in the case of domain formation (fluence $F = 0.22$ mJ/cm^2), even after 1 ns the spin system has not yet reached equilibrium. Moreover, for large fluences, we also observe a plateau in the spin temperature after 0.5 ns as illustrated in Supplemental Material, Fig. S2 [28]. Analyzing the temperature at which the plateau appears (by calculating the maximum in the difference between the spin and phonon temperature after 0.5 ns) we observe that the plateau appears at about 45 K which corresponds to the Curie temperature of the system. Hence, the plateau feature we observe in the spin temperature can be attributed to the critical slowing down at the phase transition.

In summary, we have developed a generalized two-temperature model including novel aspects of 2D materials and applied it in atomistic spin dynamics simulations of the 2D van der Waals magnet, CrI_3. The proposed model takes into account a heated substrate, and with this addition, the resulting spin dynamics gives a type-II demagnetization with further domain formation. With the recent development of 2D materials, our proposed model will be of interest to study the interaction of 2D system with the laser pulses, while obtaining the magnetic features observed experimentally. The benefits of the model proposed here is that it can be applied to other quasimetallic systems, by the introduction of the cutoff time. Although this model has been develop to illustrate the effect of a heated substrate to the ultrafast magnetization dynamics of 2D systems, it can be applied to heterostructures, where regions or layers with different thermal properties can act as a source of prolonged heat to magnetic materials. In particular, it should be

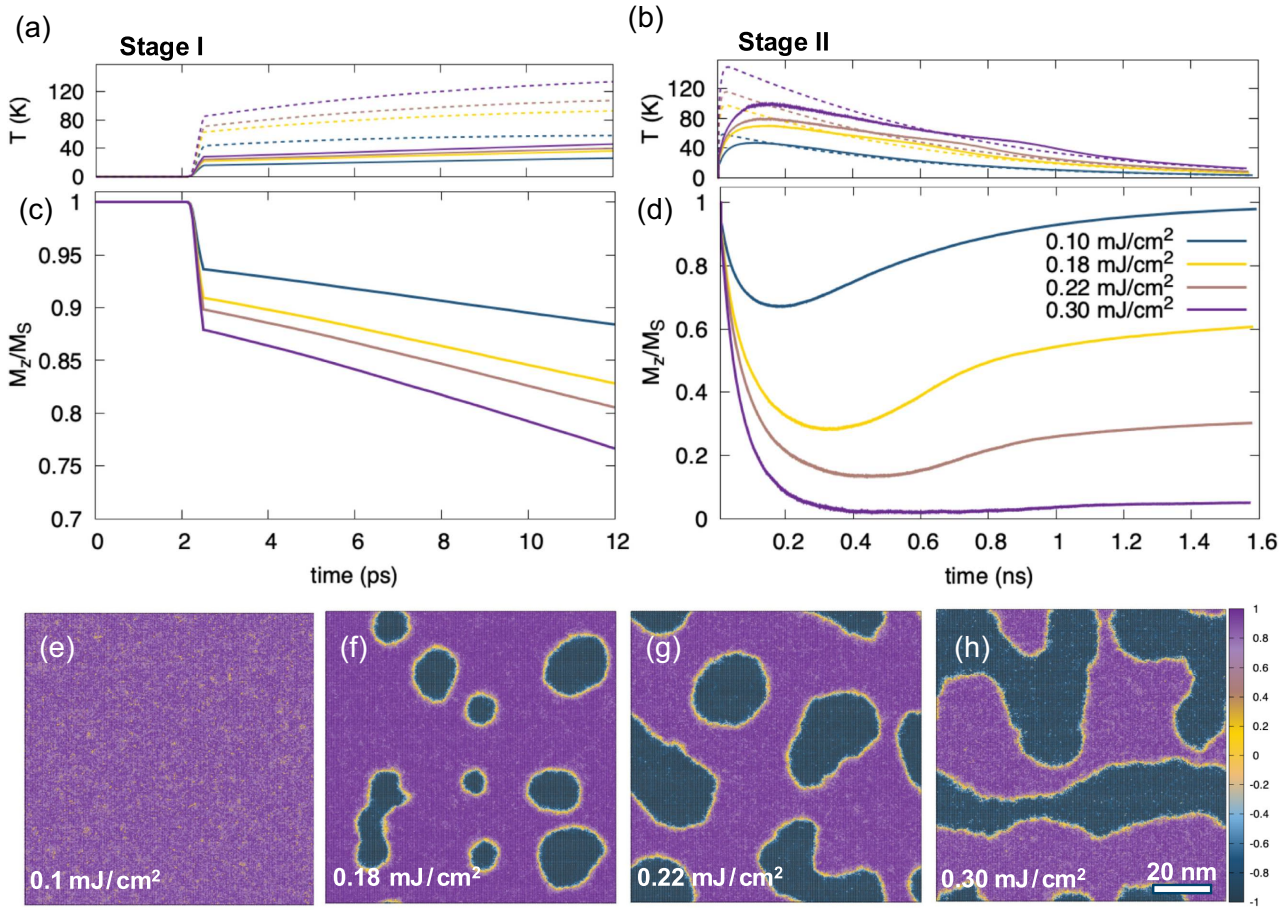

FIG. 2. Induced temporal electron, phonon, and lattice temperatures and magnetization dynamics in CrI_3. Panels (a), (b) show the evolution of spin (continuous line) and phonon (dashed lines) temperatures and panels (c), (d) the z component of the magnetization for varying laser pulse fluences. The magnetization shows a very rapid decrease within the first picosecond after the laser pulse application (stage I), followed by a slower demagnetization up to hundreds of picoseconds (stage II—specific to type-II demagnetization) and then a slow recovery in the nanosecond timescale. Panels (e), (f): induced spatial magnetization profiles after the laser pulse. Spin configurations are plotted at $t = 1.5$ ns for varying fluences. The color bar shows the direction of the z component of the magnetization. We observe that during ultrafast excitation, domains are formed for increased laser pulse power.

possible to engineer the temporal dynamics of the magnetization through a mixture of different magnetic and nonmagnetic layers providing an additional degree of freedom in comparison with more commonly studied metallic systems. Based on our findings the substrate behavior provides an interesting extra approach to the manipulation and control of magnetization dynamics specific to 2D materials.

Although not explored here, substrate engineering could potentially serve as a new avenue for controlling topological spin states such as merons and skyrmions (Refs. [11,12]) in 2D magnetic systems, as these states are often induced by heating protocols.

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M. S. implemented the generalized 2TM, performed the atomistic simulations, analyzed the data, produced the figures, and drafted the manuscript. M. S. and R. W. C. conceived and designed the study, with input from RFLE. All authors contributed to the final version of the manuscript.

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