Anisotropic laser-pulse-induced magnetization dynamics in van der Waals magnet Fe$_3$GeTe$_2$

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Keywords: van der Waals magnet, laser-induced magnetization dynamics, pump-probe spectroscopy, spin-flip rate

Abstract

Femtosecond laser-pulse excitation provides an energy efficient and fast way to control magnetization at the nanoscale, providing great potential for ultrafast next-generation data manipulation and nonvolatile storage devices. Ferromagnetic van der Waals materials have garnered much attention over the past few years due to their low dimensionality, excellent magnetic properties, and large response to external stimuli. Nonetheless, their behaviour upon fs laser-pulse excitation remains largely unexplored. Here, we investigate the ultrafast magnetization dynamics of a thin flake of Fe$_3$GeTe$_2$ (FGT) and extract its intrinsic magnetic properties using a microscopic framework. We find that our data is well described by our modeling, with FGT undergoing a slow two-step demagnetization, and we experimentally extract the spin-relaxation timescale as a function of temperature, magnetic field and excitation fluence. Our observations indicate a large spin-flip probability in agreement with a theoretically expected large spin–orbit coupling, as well as a weak interlayer exchange coupling. The spin-flip probability is found to increase when the magnetization is pulled away from its quantization axis, opening doors to an external control over the spins in this material. Our results provide a deeper understanding of the dynamics van der Waals materials upon fs laser-pulse excitation, paving the way towards two-dimensional materials-based ultrafast spintronics.

1. Introduction

As conventional data storage and manipulation technologies are reaching their fundamental limits in terms of bit density and processing speed, the need for faster and more efficient solutions has never been more apparent. The possibility of sub-ps control of magnetization with laser-pulses was first discovered in 1996 by Beaurepaire et al, and has been a major subject of research since then [1]. Large strides towards applications that operate on this ultimate timescale have been made with the discovery of fs-laser induced all-optical magnetization switching (AOS) [2], ultrafast spin-current generation [3–5], and spin-wave excitation [6–8]. Simultaneously, several theoretical frameworks have been developed to understand the microscopic mechanisms governing this phenomenon, both in terms of local [9–12] and non-local [13] angular momentum dissipation.

The recent discovery of long-range magnetic order in atomically-thin van der Waals (vdWs) materials [14–18] provides a new and exciting platform for ultrafast spintronics. Their ability to stack without the need for lattice matching makes them ideal candidates for multi-component applications based on for instance spin-current injection, spin-wave excitation and AOS. The first step towards such applications is studying the underlying fundamental phenomena in vdW magnets, namely laser-induced demagnetization and spin precession. Additionally, their ultrafast magnetization dynamics can unveil some of their important magnetic

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and spintronic properties, such as spin-flip rates and electron-phonon coupling. Although magnetic vdW materials have been studied extensively over the past few years, their magnetic response to ultra-short fs laser-pulses remains scarcely explored [19, 20]. Doing so will allow us to unveil unique opportunities for ultrafast and laser-induced spintronics using the intrinsic properties of 2D materials.

One of the most promising vdW ferromagnets is Fe₃GeTe₂ (FGT—figure 1(a), due to its relatively high Curie temperature ($T_C$) of 220–230 K and strong perpendicular magnetic anisotropy [21]. This makes FGT especially suitable for application in spin-valves [22, 23] and allows for efficient spin-orbit torques [24–26]. Furthermore, its properties allow for the stabilization of complex spin structures such as spin spirals and skyrmions [27, 28]. Unraveling the physics governing laser-pulse induced demagnetization dynamics in FGT brings us one step further towards unifying the fields of vdW materials, spintronics and (ultrafast) photonics.

In this work, we study the fs laser-pulse induced demagnetization dynamics of thin-film FGT. Specifically, we measure the timescales involved with the demagnetization process as a function of laser fluence, ambient temperature and applied magnetic field. We compare our experimental results to calculations based on the well-established microscopic 3-temperature model (M3TM) [12], and find very good quantitative agreement. We find that FGT undergoes a two-step demagnetization (type-II), in agreement with an earlier study [20], which we attributed to its low Curie temperature and relatively large atomic moment. Our theoretical analysis allows us to estimate the spin-flip probability, which depend on intrinsic properties of FGT and quantifies how efficiently angular momentum can be transferred within the system upon laser excitation. The analysis indicates a surprisingly large spin-flip probability, contrary to the earlier claims of long spin lifetimes, but in agreement with theoretical expectations of large spin–orbit coupling in FGT. The laser-induced demagnetization is found to be strongly dependent on an applied in-plane magnetic field, consistent with an anisotropy in the spin-flip probability, which has been predicted for other material classes [29, 30]. Moreover, contrary to recent studies in Cr₂Ge₂Te₅ (CGT) [20], we were unable to excite ferromagnetic resonance (FMR) modes. We hypothesize that this is caused by strong dephasing of the FMR mode due to weak interlayer coupling and in-plane anisotropy variations.
2. Experimental details

Our samples are fabricated through mechanical exfoliation of commercially available FGT crystals (HQ Graphene) in inert nitrogen atmosphere and transferred to an optical cryostat without air exposure, where the sample is then kept in high vacuum (<10⁻⁶ mBar) throughout all experiments. To measure the magneto-optical response we apply an external magnetic field (H_{ext}) at an angle φ_H with respect to the sample plane. Due to the out-of-plane magnetic anisotropy field of FGT (H_{ani}), the magnetization m will be tilted from the out-of-plane direction, according to the effective magnetic field $H_{eff}$. Figure 1(b) shows a diagram indicating all relevant directions and angles.

After selecting a thin FGT flake (figure 1(c)) using an optical microscope, the flake is probed using magneto-optical Kerr effect (MOKE) microscopy at low temperatures (T = 80 K). This way, magnetically homogeneous areas with clear single-domain behavior of several μm² are identified, figures 1(d) and (e). The flake for which the results are shown here was characterized by atomic force microscopy after all other experiments were completed to obtain its thickness, $t = 16.8 ± 0.7$ nm, corresponding to 20 ± 1 monolayers (see section V of the supplementary information).

For our MOKE measurements we use a mode-locked Ti:Sapphire laser centered at 735 nm with a repetition rate of 82 MHz and 85 fs of pulse width at the sample location, with a spot size of 2.9 x 1.7 μm². Using hysteresis curves of MOKE signal versus $H_{ext}$ at $\phi_H = 19^\circ$ (figure 1(f)), we obtain the MOKE amplitude A versus temperature (blue circles in figure 1(g)). We find that the MOKE signal decreases abruptly around 200 K, in agreement with vibrating sample magnetometry measurements done in a bulk FGT flake (figure 1(g), green triangles). The inverse magnetic susceptibility ($\chi^{-1}$) of the bulk crystal is well described by a power law for the paramagnetic phase given by $(T - T_C)^\gamma$. A clear transition point corresponding to the Curie temperature $T_C = 191 ± 2$ K is observed, with $\gamma = 1.3 ± 0.1$, which is close to the typical exponent for a three-dimensional Ising magnet ($\gamma \sim 1.25$) [32]. We note that the Curie temperature is significantly lower than expected for stoichiometric FGT [21, 33–36], indicating that our material is slightly Fe deficient [37, 38]. Following these works, we estimate that the region used for our experiments has an Fe deficiency of less than 7% (i.e. Fe₁₋ₓₐ₉Ge₂ with x < 0.2) assuming no thickness-dependence effects for thicker flakes. This is further confirmed by energy-dispersive x-ray spectroscopy (EDX) measurements, which reveals spatial changes in the Fe content of the bulk crystal (see section IV of the supplementary information).

The laser-induced demagnetization measurements described below are performed by pump-probe spectroscopy using the same setup as above. The higher intensity pump beam, with focused area of 9.6 x 6.6 μm², is obtained from the same laser and overlapped onto the probe spot. For additional experimental details see section I of the supplementary information.

3. Experimental results and discussion

3.1. Demagnetization dynamics of FGT

As can be seen in figure 2(a), a clear two-step laser-induced demagnetization process is observed in our samples, indicative of type-II demagnetization, similar to rare-earth ferromagnets Gd and Tb [39, 40] as well as the vdW magnets CGT [19, 20]. The total magnetization quenching is proportional to the pump laser fluence, which is attributed to the increased heating towards the Curie temperature. Two distinct timescales can be distinguished: $\tau_{m}$ and $\tau_{m'}$. They are commonly referred to as the electron-phonon relaxation and magnetic timescale respectively. How these timescales relate to the physics of laser-induced demagnetization will be discussed later.

To extract the relevant demagnetization timescales, the data are fitted with an analytical solution of the phenomenological 3-temperature model (3TM) [41]. The fitting procedure is discussed in more details in section III of the supplementary information. Although some temperature dependence of $\tau_{m}$ has been reported in literature [42, 43], we assume a temperature (and thus fluence) independence in our analysis to avoid overparameterization of the model. Furthermore, this allows us to study the changes of $\tau_{m}$, which are typically large [12, 44]. A global electron-phonon relaxation timescale of $\tau_{e} = 794 ± 23$ fs is found, which is a typical value for ferromagnetic transition metals and alloys based on them [12, 42, 44–47].

The timescale $\tau_{m}$ is extracted from the fluence-dependent curves (figure 2(a)) and plotted in figure 2(b) as a function of the normalized total demagnetization for each laser fluence. An increase from ~13 to ~20 ps is observed in the studied fluence range. The error bars are relatively large compared to the apparent spread in the data due to a significant cross-correlation between the magnetic timescale and the typical timescale that governs heat dissipation in the flake. Related to its fluence dependence, the magnetic timescale is also dependent on the ambient temperature (figure 2(d)). We observe a critical slowing down of the magnetization dynamics as T approaches $T_C$ which is well reported in literature for standard ferromagnets [44, 48–50].

Detailed information on the dynamic magnetization properties of FGT can be obtained by employing the M3TM [12], which has been tested extensively on a wide variety of materials and can reproduce the right timescales and trends as a function of temperature and fluence using known material parameters.
Figure 2. (a) Laser-induced demagnetization measured at four different laser fluences. The data fitting (black line) was done using the procedure described in section III of the supplementary information. (b) Magnetic timescale $\tau_m$ as a function of the normalized total demagnetization. (c) Laser-induced demagnetization for different ambient temperatures. (d) Magnetic timescale $\tau_m$ as a function of temperature, fitted with equation (3) (black line). The data in a and c are normalized using the methods described in section II of the supplementary information and offset vertically for clarity.

[12, 44, 51–53]. Similar to the phenomenological 3TM for ultrafast demagnetization, the magnetic system is subdivided into three separate systems which can exchange energy as well as angular momentum: the electron (e), the phonon (p) and the spin (s) system [1], each characterized by a temperature, $T_e$, $T_p$ and $T_s$, respectively. A model Hamiltonian is set up to describe the three systems and their interactions. Here, we only focus on the key features of the model; for an extensive treatment, we refer to the original papers [10, 12]. Upon laser excitation, the electron system is heated, bringing the total system in a non-equilibrium state. We assume the subsystems are at internal thermal equilibrium at all times. Furthermore, the heat capacity of the spin system is considered to be negligible. Under these assumptions, energy exchange between the electron and phonon systems is governed by the 2-temperature model [54]. An exemplary solution is presented in figure 3(a), using parameters shown in section VIII of the supplementary information. The laser-pulse significantly heats up the electron system beyond the Curie temperature. Hereafter, the electron and phonon system equilibrate on a timescale given by $\tau_e$.

The angular momentum transfer between the subsystems is represented by $\tau_m$ and taken to follow the Elliot-Yafett mechanism for spin relaxation [55, 56]. Here, an electron-phonon scattering event leads to energy exchange between the electron and phonon system. However, due to spin–orbit coupling, the single-electron state is a mix of spin-up and down states, of the form $\Psi_\vec{k} = a_\uparrow |\uparrow\rangle + b_\downarrow |\downarrow\rangle$ [55, 56]. This leads to spin-mixing of electron states near the Fermi level, quantified by the material dependent spin-mixing parameter $\langle b^2 \rangle$. Consequently, there is a finite probability $a_{sf} \propto \langle b^2 \rangle$ for a spin-flip to occur during an electron-phonon scattering event, thereby transferring angular momentum between the spin system and the lattice. Transforming the model Hamiltonian to rate equations, the normalized magnetization $m$ can be calculated: [12]:

$$\frac{dm}{dt} = R \frac{T_p}{T_C} \left[ 1 - m \coth \left( \frac{T_C}{T_e} \right) \right], \tag{1}$$

$$R = \frac{8 a_{sf} g_{ep} k_B T_C^2 V_{at}}{\left( \mu_{at}/\mu_B \right) E_D^2}. \tag{2}$$

Here, $k_B$, $\mu_B$ and $\mu_{at}$ are the Boltmann constant, Bohr magneton and the atomic moment, respectively. $V_{at}$ and $E_D$ are the atomic volume and the Debye energy. The material parameter $R$ quantifies the demagnetization rate, and is used to determine a material dependent figure of merit $T_C/\mu_{at}$ that provides a simple prediction of the demagnetization rate [12].

An example demagnetization trace, measured at laser fluence $F = 55 \mu J m^{-2}$ and 80 K, is shown in
figure 3(b). Model parameters, as presented in section VIII of the supplementary information, are extracted from literature, and unknown parameters are chosen such that there is a good agreement between experiment and simulations. We observe the best correspondence for spin-flip probability $a_d = 0.14$. This is relatively large compared to elementary transition metals [12, 44, 57]. However, Kuiper et al. have demonstrated that Co/Pt multilayers have significant larger $a_d$ than pure Co due to the proximity of Co to the high SOC Pt [57]. We argue that this is exactly the case in FGT, where the magnetic moment carrying Fe atoms are closely surrounded by the heavy Te atoms.

This corroborated by recent observation of large bulk spin-orbit torques observed in FGT [58, 59]. Although we observe a relatively large $a_d$, the demagnetization process is relatively slow. We attribute this to the sub-room-temperature Curie temperature and large atomic moment, leading to a large figure of merit $T_C/\mu_{at}$ and thus slower demagnetization. We stress that, even though the extracted value can be sample-specific to some degree due to impurity scattering, the intrinsic (e.g. phonon) mediated scattering typically dominates the ultrafast demagnetization [10, 12]. Furthermore, significant deviations can only be expected for layer thicknesses below approximately 5 nm, according to an estimation of the out-of-plane exchange constant [21]. However, spin-flip scattering is non-trivial in the limit of a few monolayers due to the complicated electron structure of FGT, and requires further research to uncover.

The dependence of $\tau_m$ on laser fluence and temperature is well described by the M3TM. The simulation results are plotted as a black line in figures 2(b) and (d). We observe the same upward trend for increasing laser fluence, as well as good quantitative prediction of the demagnetization timescale for realistic model parameters. This indicates FGT behaves very similarly to elementary transition and Rare-earth magnets. The theoretical temperature dependence of $\tau_m$, as derived in section VII of the supplementary information, reads:

$$\tau_m = \left(\frac{1}{2R}\right)\left(\frac{1}{1 + \frac{1}{\tau_C}}\right),$$

which describes the critical slowing down of the dynamics towards the Curie temperature. Here, $\Delta T_h$ is the temperature rise due to the average laser induced heating of the FGT flake at high fluence and temperature. Note that this relation is derived for temperatures close to $T_C$, so deviations are expected for low temperatures. Using the previously determined $T_C$ of 191 K, the data in figure 2(d) is fitted using this relation, yielding $R = 0.15 \pm 0.01$ ps$^{-1}$. Using equation (2), a spin-flip probability of ~0.12 is calculated, which is similar to values found from our time-dependent simulations, and again confirms the large spin-flip probability found for FGT. Furthermore, we extract $\Delta T_h = 21 \pm 4$ K from our fitting procedure, which corresponds to a remaining magnetization quenching just before laser excitation of several percents. Our approach is further validated in section X of the supplementary information, where we estimate the temperature rise using hysteresis loops measured with and without sample pumping at negative time delay supplementary information. The latter yields $\Delta T_h = 16 \pm 7$, in line with the value obtained from fitting equation (3).

3.2. Magnetic-field dependence

Next, in order to study how the symmetry of the FGT crystal structure influences the demagnetization dynamics, we apply an in-plane magnetic field ($\phi_H = 0$) to induce a canting of the magnetization, away from the quantization axis. The measurements are done at an ambient temperature of 160 K and a laser fluence of 140 $\mu$J cm$^{-2}$ to maximize the demagnetization and thus the effect of the applied field.
on the magnetization angle (figure 4(a)). We measure a significant speeding up of the laser-induced demagnetization process for increasing in-plane field, which can be associated with an increase of the spin-flip probability. To visualize the speeding up of the demagnetization dynamics, the delay at the minimum of the demagnetization curve is extracted (figure 4(b)). A strong anisotropy in $\langle b^2 \rangle$ and thus the spin-flip rate is expected in magnetic systems with lowered symmetry and large SOC, of which FGT is a prime example [29]. Since the effect of the magnetization canting is symmetric with the applied field, the first order approximation of the spin-flip probability is given by:

$$a_d \approx a_{d,0} (1 + C \alpha^2)$$

$$= a_{d,0} \left[ 1 + C \left( \frac{H_{\text{ext}}}{H_{\text{ani}}} \right)^2 \right], \quad (4)$$

where $a_{d,0}$ is the field-free spin-flip probability and $\alpha$ the angle between between the magnetization and the anisotropy field (figure 4(c)), given by $H_{\text{ext}}/H_{\text{ani}}$ for small $\alpha$. Here we use $H_{\text{ani}} = 2K_{U}/\mu_0 M_S$ with $K_U$ the anisotropy constant and $M_S$ the saturation magnetization (the values used can be found in section VIII of the supplementary information). The phenomenological parameter $C$ is proportional to the ratio between the two orthogonal components of the spin-flip probability, and therefore a measure for its anisotropy. An increase of $a_d$ leads to lower $\tau_m$, which can be expressed in a shift of the minimum of the demagnetization curve $t_{\text{min}}(m)$. The M3TM is used to find the relation between $a_d$ and $t_{\text{min}}(m)$ (see section IX of the supplementary information). In the lowest order, $t_{\text{min}}(m)$ scales linearly with $a_d$, following:

$$t_{\text{min}}(m) = t_0 - t_1 a_d, \quad (5)$$

where from the M3TM simulations we find $t_0 = 107.8$ ps and $t_1 = 324.7$ ps. Our results shown in figure 4(b) are well fitted using the equation above with $C \approx 13$, indicating a (very) strong anisotropy of $a_d$. We should stress that the observed effect is very large. There is no observable decrease of the MOKE signal when applying an in-plane field. Based on our experimentally determined noise threshold shown in figure 4(e), the magnetization canting is no more than $5^\circ$, which is also in line with calculation based on experimentally determined values for the anisotropy constant [60, 61]. Our results are realistic when compared to calculations for other materials with reduced symmetry [29, 30]. However, in order to gain a deeper microscopic insight on the spin relaxation anisotropy in vdW materials in general and FGT specifically, ab-initio calculations have to be carried out for these materials as well.

Another potential motivation for our field-dependent measurements is to excite FMR modes with the $\Delta K$ mechanism, which is schematically shown in figure 4(d). Here, a demagnetization reduces the anisotropy of the FGT, thereby quickly reorienting the effective field [6]. This leads to the excitation of damped precessional dynamics. Even though FMR modes have been shown for a similar material—CGT, which possesses a much lower perpendicular magnetic anisotropy (PMA) [20]—in a similar experimental configuration, we do not observe any signs of magnetization precession in our measurements. We explain the absence of the FMR mode in our experiments (figure 4(a)) by the weak interlayer exchange coupling $A_{\text{ex}, \perp}$ due to the vdW stacking in FGT [28, 62, 63]. Furthermore, our EDX data (see section IV of the supplementary information) indicate that in-plane anisotropy fluctuations within the probes area ($\sim 3 \, \mu m$) are not unexpected. We hypothesize that these effects lead to a strong dephasing of the measured precessional motion, which could explain the absence of FMR in our measurements.

To test this hypothesis, we use modified LLG equations which take into account the longitudinal magnetization dynamics [64]. Within this framework, the magnetization is given by $\vec{M} = [1 - \Delta M(t)]\vec{M}_0\hat{m}$, where $\vec{m}$ is the unit magnetization vector and $\Delta M(t)$ is the experimental data measured at zero field. Because perturbations are large, an extended LLG equation is used, given by:

$$\dot{\vec{m}} = \frac{\gamma}{1 + \alpha^2} (\vec{m} \times \vec{H}_{\text{eff}} + \alpha (1 - \Delta M(t)) \vec{m} \times \vec{H}_{\text{demag}})$$

$$+ \frac{1}{1 - \Delta M(t)} \frac{d\Delta M(t)}{dt} \vec{m}, \quad (6)$$

where the last term takes into account the magnitude change of $\vec{M}$. The effective field $\vec{H}_{\text{eff}}$ is given by:

$$\vec{H}_{\text{eff}} = \vec{H}_{\text{ext}} + \vec{H}_{\text{demag}} + \vec{H}_{\text{ani}} + \vec{H}_{\text{ex}}$$

$$= \vec{H}_{\text{ext}} - \hat{N} \cdot \vec{M} + \frac{2K_{U}}{\mu_0 M_S} \vec{M}_i$$

$$+ 2A_{\text{ex}, \perp} \sum_i \frac{\vec{M} - \vec{M}_i}{d_i^3}, \quad (7)$$

where $\hat{N}$ is the demagnetizing tensor, with $N_{zz} = 1$ for thin films. We simulate every individual layer as a single macrospin coupled to its neighboring layers $i$ (with magnetization $\vec{M}_i$ and at a distance $d_i$) via an exchange field, of which the strength is given by the interlayer exchange stiffness $A_{\text{ex}, \perp}$. To be able to compare experiments and simulations reliably, laser attenuation is taken into account. This is done by scaling the excitation energy via $\Delta M(t)$ with an exponential decay function characterized by the laser penetration depth $\lambda$. This scaling also applies as weights when averaging the magnetization of the layers to simulate laser probing.

Our simulations indicate similar decrease of the amplitude of the FMR mode for weaker interlayer coupling, caused by a strong dephasing of the
magnetization of the various layers throughout the flake. Assuming a maximal Gaussian in-plane anisotropy variation $\sigma_K$ of 5% and using the parameters presented in section VIII of the supplementary information, we simulate the FMR amplitude as a function of the reduced interlayer exchange stiffness. In figure 4(e), we compare this amplitude to the noise threshold, which we estimate from experimental data at negative time delay. The simulations indicate a significant decrease of the FMR mode amplitude for weaker interlayer coupling, crossing the noise threshold indicated by the light-blue area at around $A_{ex,\perp}/A_{ex,\parallel} = 0.2$, where $A_{ex,\parallel}$ is the in-plane exchange interaction. This gives us an upper limit for the interlayer exchange stiffness, which is in line with first principle calculations [63] as well as other experimental observations [28], where the ratio between the interlayer and intralayer exchange parameter is reported to be in the order of 0.1.

Although FMR oscillations were too small to measure, we note that any FMR signal superimposed onto the demagnetization can in principle lead to a shift of the minimum of the demagnetisation curve. We model this field dependence using equation (6) and plot the results in figure 4(b) with a green line. Although a slight field dependence is calculated, it is an order of magnitude smaller than what we observe in our experiments. Therefore, we believe that the anisotropy of the spin-flip probability is the only viable explanation for the observed decrease of $t_{\text{min}}(m)$ as a function of applied field.

4. Conclusion and outlook

Our results on magnetization dynamics in FGT shine light on the use of ultrafast demagnetization to obtain important magnetic parameters in vdW systems. To some extent, FGT behaves similar upon laser excitation compared to well-studied ferromagnetic systems, such as transition metal and rare-earth ferromagnets. Even though a two-step demagnetization process was obtained, which is usually assigned to a long spin lifetime, a detailed analysis of our results point to the opposite, showing that care must be taken when analyzing such results. Very much unlike the regular transition metal ferromagnets, a strong anisotropy of the spin-flip rate with respect to the magnetization direction was obtained with our M3TM analysis, which provides a powerful method to tune the spin dynamics in two-dimensional magnets.

This property provides a unique advantage of 2D magnets compared over conventional transition metal ferromagnetic systems, and could be further explored to tune the temporal profile of laser-induced spin-currents in vdW magnets [4], or to control the spin and magnon transmission in vdW magnets through a local change of the magnetic anisotropy by electrical gating. Furthermore, the slow dynamics observed in FGT together with its unique magnetic properties make it a prime candidate for all-optical magnetization switching based on vdW materials, when interfaces with a material which exhibits fast dynamics [65]. This would open up new pathways...
towards ultrafast control of magnetization in future data storage devices.

**Data availability statement**

The data that support the findings of this study are available upon reasonable request from the authors.

**Acknowledgments**

We thank Mark C H de Jong his help with A F M measurements, and Jeroen Francke, Bart van Looij and Gerrie Baselmans for technical support. This work is supported by Stichting voor Fundamenteel Onderzoek der Materie (FOM) through Grant No. 10023746 and the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO) through Grant No. 10018479. MHDG acknowledges NWO for financial support through the Grant Veni 15093.

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