Low-remanence criterion for helicity-dependent all-optical magnetic switching in ferrimagnets

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We demonstrate that a low-remanent sample magnetization M_R is crucial for all-optical magnetic switching (AOS) in ferrimagnets and ferrimagnet heterostructures. M_R may be devised by the composition of the material. However, it can also be controlled *in situ* by changing the sample temperature because it affects M_R . We show that increasing the lattice temperature by laser pulses or a simple heating resistor enables AOS in a ferrimagnetic Tb-Fe film, which does not exhibit AOS at room temperature. We reconcile earlier contradicting results for AOS in heated and cooled magnetic films by applying the low-remanence criterion. It also applies to other existing rare-earth transition-metal (RE-TM) alloys, such as GdFeCo or Tb-Co, to ferrimagnet heterostructures, and to RE-free synthetic ferrimagnets.

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Reliable, fast, and high-density magnetic data storage technologies are important for efficient future memory devices [1]. For this purpose, materials with a high uniaxial perpendicular magnetic anisotropy and a high coercivity are needed to circumvent the superparamagnetic limit when decreasing the bit size [2]. However, the inalienable high coercivity easily exceeds the achievable magnetic field strength of a read-write head [3]. As a consequence, switching a bit by applying a magnetic field pulse becomes impossible. A well-developed technique to overcome this limitation is the so-called heat-assisted magnetic recording (HAMR). It uses laser heating to lift the sample temperature locally above the Curie temperature, which enables switching at a smaller magnetic field [3]. The major disadvantage of this technique is the issue of correct timing of the heat pulse and the magnetic field pulse [4,5]. One promising route to resolve this problem is to control magnetization exclusively by light without the need for an external magnetic field. After the first demonstration of this so-called all-optical switching (AOS) in amorphous Gd₂₂Fe_{74.6}Co_{3.4} thin films [6], numerous theoretical and experimental investigations have strived to reveal the underlying physical mechanism. A comprehensive review about these efforts can be found in [7]. Soon after the discovery of AOS, it was assumed that higher temperatures are detrimental to AOS because of facilitated switching at low temperatures. Hohlfeld et al. demonstrated that the threshold fluence for AOS in Gd₂₂Fe_{74.6}Co_{3.4} decreases at lower sample temperatures [8]. In contrast, an increase of the AOS threshold fluence was reported for $Tb_{14}Fe_{68}Co_{18}$ when cooling the sample from room temperature down to T = 110 K(Ref. [9]). Recent investigations stress the importance of heating for AOS [9-14] and ascribe it, similar to HAMR, to a thermally assisted [10,14] or even entirely thermally induced process [11,13]. For instance, increasing the repetition rate of laser pulses from single shot up to 500 kHz leads to a decrease of the AOS threshold fluence in $Gd_{24}Fe_{66.5}Co_{9.5}$ (Ref. [14]) due to heat accumulation in the metallic film [15–18], which underlines the role of heating for the AOS process.

We demonstrate that these conflicting observations may be reconciled by taking the sample magnetization instead of the sample temperature into account. We reveal that the occurrence of AOS in $Tb_{34}Fe_{66}$ can be controlled either by laser-induced heating or by substrate heating with a simple heating resistor. In both cases the remanent sample magnetization M_R decreases, which facilitates AOS. We show that the low-remanence criterion applies to amorphous alloys Tb-Fe, GdFeCo, and Tb-Co, artificial heterostructures, and rare-earth-free synthetic ferrimagnets.

We investigate AOS in amorphous Tb₂₉Fe₇₁ and Tb₃₄Fe₆₆ films at laser pulse repetition rates of v = 250 kHz and $v = 10 \,\text{kHz}$ at room temperature. These films with a layer thickness of 16 nm are deposited by dc magnetron sputtering from element targets under ultra-high vacuum conditions on a microscope glass slide. They are sandwiched between a 5-nmthick Pt buffer layer to initiate growth and a 3-nm-thick Pt capping layer to prevent oxidation (Fig. 1). The integral magnetic properties of the magnetic films are measured in a superconducting quantum interference device-vibrating sample magnetometer (SQUID-VSM), revealing a strong uniaxial magnetic anisotropy perpendicular to the sample surface $(M_R/M_S = 1)$. The remanent sample magnetization at room temperature amounts to $M_{\rm R} = (230 \pm 20) \,\text{kA/m} (230 \,\text{emu/cc})$ for $Tb_{34}Fe_{66}$ and to $M_{R} = (112 \pm 10) \text{ kA/m} (112 \text{ emu/cc})$ for Tb₂₉Fe₇₁. Further details concerning sample growth and magnetic properties can be found elsewhere [10].

To investigate the AOS ability, we use a magneto-optical Faraday imaging setup with a continuous-wave He-Ne laser $(\lambda = 632.8 \text{ nm})$ for illumination [10] [Fig. 2(a)]. The orientation of the sample magnetization appears as black (M^-) and gray (M^+) contrast (Fig. 3). A regenerative laser amplifier system (Coherent RegA 9040) with a variable pulse repetition rate ν between 10 and 310 kHz is used to provide the switching

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FIG. 1. (Color online) Heatable sample holder and layer stack. The sample is sandwiched between the heated aluminum top plate and a copper fastener. The heater is a 5-W, $330-m\Omega$ heating resistor connected to an adjustable high-current power supply. To ensure proper heat conduction between the sample holder and the sample, it is embedded into a conductive paste. The temperature close to the sample position is measured by two thermocouple probes located on opposite sides of the sample.

pulses. The center wavelength is 800 nm and the pulse duration at sample position amounts to 100 fs. The polarization of the pulses is set by an achromatic quarter-wave plate. The laser pulses are attenuated by a combination of a zero-order half-wave plate and a polarizer. The switching pulses are focused onto the sample by an aspherical lens (f = 20 mm) to a diameter of 77 µm FWHM, as measured by the Liu method [19].

The all-optical switching experiment is performed as follows: First, the sample is homogeneously magnetized in one direction by applying an external magnetic field, e.g., M^- [Fig. 3(a)]. Subsequently, the sample is linearly translated $10\,\mu\text{m/s}$ below the focus of circularly polarized light [6,10,14,20] (e.g., right handed, σ^+) for a given laser fluence. This procedure is repeated by stepwise increasing the laser fluence until a domain with opposite contrast as the initial state is observed (see gray rectangular domain structure with magnetization M^+ in Fig. 3(b)). Afterwards, the written structure may be erased at the same fluence but opposite helicity (e.g., left handed, σ^{-}). If this erasing attempt is successful, AOS is witnessed [Fig. 3(c)]. If the written area cannot be erased, a purely thermally induced demagnetization process occurs [PTD, cf. Fig. 3(f)]. In this case, a second stripe at the same fluence but opposite helicity as in the previous image is written to the right to illustrate this helicity-independent effect.

At 250 kHz we observe AOS at a threshold fluence of $F = (1.0 \pm 0.3) \text{ mJ/cm}^2$, in accordance with Ref. [10] [Figs. 3(a)-3(c)]. The switching experiments are performed



FIG. 2. (Color online) Beam geometry for (a) Faraday imaging and (b) Kerr magnetometry.

for both orientations of the sample magnetization to verify the results. For both directions we obtain similar threshold fluences. The switching causes a homogeneous domain pattern with opposite orientation as the initial state when using circularly polarized laser pulses.

Lowering the repetition rate from $\nu = 250$ kHz to 10 kHz should still result in AOS. Only the threshold fluence should increase [14] due to less heat accumulation in the magnetic film. However, when repeating the AOS experiment for $\nu = 10$ kHz at room temperature, AOS is not observed. Instead, the switching attempt results in a pure thermally induced demagnetized state (PTD) at a threshold fluence of $F = (1.4 \pm 0.2)$ mJ/cm² [Figs. 3(d)-3(f)]. The formation of this multidomain structure arises from stray field contributions of the nonirradiated surroundings [10]. It is facilitated in samples with a high remanent magnetization M_R and counteracts AOS, because the magnetization of the switched region is always opposite to the direction of the rest of the film, and erasing with the opposite helicity is not possible anymore.

To understand the absence of AOS, we have to investigate the influence of laser irradiation on the stationary magnetic properties of the sample at the two different pulse repetition rates. Therefore, stationary *in situ* hysteresis loops are measured by magneto-optical Kerr-effect (MOKE) magnetometry in polar geometry with and without irradiation with laser pulses. For this purpose the imaging laser reflected from the sample surface is steered into a polarization-sensitive two-channel photodetector [21]. The Faraday imaging unit behind the sample is replaced by two permanent magnets [Fig. 2(b)]. The magnets generate a homogeneous magnetic field within the probed area with a field strength up to 5 kOe, which is sufficient to fully saturate the magnetization of the Tb₃₄Fe₆₆ film. For measuring hysteresis loops, the



FIG. 3. (Color online) All-optical helicity-dependent magnetic switching (AOS) in Tb₃₄Fe₆₆ at a pulse repetition rate of 250 or 10 kHz. The left column shows the initial homogeneously magnetized state, the middle column after exposure with right-handed circularly polarized (σ^+) laser pulses, and the right column images the state after exposure with left-handed circularly polarized (σ^-) laser pulses at the same fluence as used in the corresponding previous image. (a–c) 250 kHz pulse repetition rate and substrate temperature of T = 300 K. The threshold fluence for all-optical switching (AOS) is $F = (1.0 \pm 0.3)$ mJ/cm². (d–l) 10 kHz repetition rate and different substrate temperatures of T = 300 K and $F = (1.4 \pm 0.2)$ mJ/cm². (g–i) AOS onset at a substrate temperature of T = 345 K and $F = (0.9 \pm 0.1)$ mJ/cm². (j–l) AOS at a substrate temperature of T = 370 K and $F = (0.5 \pm 0.1)$ mJ/cm².



FIG. 4. (Color online) Hysteresis loops of Tb₃₄Fe₆₆ measured by magneto-optical Kerr magnetometry (MOKE): (a) With laser irradiation at a pulse repetition rate of $\nu = 250$ kHz at the AOS threshold fluence of $F = (1.0 \pm 0.3)$ mJ/cm². The substantial decrease of the coercivity and remanence due to irradiation with the switching pulses is visible. Solid lines represent averaged values. (b) With laser irradiation of $\nu = 10$ kHz pulse repetition rate at the PTD threshold fluence of $F = (1.4 \pm 0.2)$ mJ/cm². Here, the magnetic properties are unchanged with and without laser irradiation.

magnitude of the magnetic field is adjusted by changing the distance between the sample and the permanent magnet and is simultaneously monitored by a Hall probe located near the sample in the homogeneous field region. For the measurements with simultaneous laser irradiation the fluence is adjusted either to the PTD threshold at v = 10 kHz or AOS threshold at v = 250 kHz and the polarization is adjusted to linear (π), left (σ^{-}) and right (σ^{+}) handed circular, respectively. For averaging, at least five hysteresis loops are recorded in each configuration, i.e., with and without laser pulses and for each polarization. As summarized in Fig. 4(a), a strong effect of the laser irradiation at a repetition rate of v = 250 kHz on the magnetic properties of the sample is apparent, which is independent from the polarization of the switching pulses. The coercivity of the sample decreases from 2.3 to 1.2 kOe, and also the remanence decreases. The drastic changes of the magnetic properties can be explained by an increase of the sample temperature within the probed area due to laser irradiation and hence heat accumulation [15-18], which facilitates AOS. An influence of the laser irradiation on the stationary magnetic sample properties is not found at a pulse repetition rate of $\nu = 10 \,\text{kHz}$ [Fig. 4(b)]. Therefore, laser-induced heating is negligible at v = 10 kHz, similar to single-shot experiments. This experimental result indicates that the sample temperature (i.e., room temperature) is too low for AOS to occur at $\nu = 10$ kHz. In contrast, the elevated sample temperature and hence reduced remanent magnetization $M_{\rm R}$ and coercivity $H_{\rm C}$ facilitates AOS in Tb₃₄Fe₆₆ at a pulse repetition rate of 250 kHz [10].

We therefore investigate whether an increase of the sample temperature by a physical process other than heating by laser pulses is able to reestablish AOS at a repetition rate of $\nu = 10$ kHz. From previous studies in GdFeCo it is known that switching occurs for laser pulse durations of 100 fs and 10 ps (Ref. [22]). Therefore, we exclude an ultrafast rise of the electronic temperature to be responsible for AOS. Instead, the comparatively slow increase of the lattice temperature should be important. We externally control the sample temperature by a heatable sample holder via Joule heating (Fig. 1). For this purpose a heating resistor $(330 \text{ m}\Omega, 5 \text{ W})$ is placed on an aluminum sheet of 1 mm thickness by a thermal adhesive and connected to a high-current power supply. The temperature of the sample is measured by two thermocouple probes (K-type) located at opposite ends of the sample to assure a thermal equilibrium within the probed sample region. Both the thermocouples and the sample are embedded in thermally conductive paste between the top aluminum holder and the bottom copper fastener to ensure good heat transfer from the heating stage to the sample and to the thermocouple probes. This setup allows us to control the substrate temperature from 300 up to 380 K by simply applying different currents at the heating resistor.

At an elevated substrate temperature of T = 330 K, PTD still occurs [$F = (1.0 \pm 0.1)$ mJ/cm²]. But at T = 345 K AOS becomes partially possible at a threshold fluence of $F = (0.9 \pm 0.1)$ mJ/cm², as shown in Figs. 3(h)–3(i). At 370 K, AOS reappears at $F = (0.5 \pm 0.1)$ mJ/cm² [Figs. 3(k)–3(l)]. Obviously, AOS may be thermally controlled, either by tuning the repetition rate of the laser pulses or by substrate heating (Joule heating).

To corroborate the influence of heating for AOS, the magnetic properties at different sample temperatures are studied. We measure stationary *in situ* MOKE hysteresis loops at T = 300 K, 345 K, and 370 K for a qualitative analysis. As shown in Fig. 5(a), the coercivity decreases



FIG. 5. (Color online) Hysteresis loops of Tb₃₄Fe₆₆ at various sample temperatures of T = 300, 345, and 370 K, measured by a MOKE and by SQUID-VSM. For elevated sample temperatures the coercivity as well as the remanence decreases, which facilitates AOS at a pulse repetition rate of $\nu = 10$ kHz at T = 370 K.

for increasing sample temperatures. Also, the remanence decreases at T = 370 K. However, MOKE is not a suitable technique to obtain quantitative values of the sample magnetization, because it is mostly sensitive to the Fe sublattice magnetization at $\lambda = 632.8$ nm [23]. Hence, we measure the stationary magnetic properties at elevated sample temperatures also by SQUID-VSM magnetometry. Out-of-plane hysteresis loops are recorded for the Tb₃₄Fe₆₆ film [Fig. 5(b)] which reveal a decrease of the remanent magnetization from $M_{\rm R} = (230 \pm 20)$ kA/m (230 emu/cc) at 300 K to (152 ± 20) kA/m (152 emu/cc) at 345 K, to finally (106 ± 20) kA/m (106 emu/cc) at 370 K.

We find that the increase of the sample temperature either by laser-induced heating at v = 250 kHz or by substrate heating at v = 10 kHz leads to a decrease of the remanent sample magnetization and the coercivity, which promotes AOS in Tb₃₄Fe₆₆. At v = 10 kHz the laser-induced increase of the sample temperature is negligible [Fig. 4(b)]. Thus, we can exclude any additional laser-related decrease of the stationary remanent sample magnetization $M_{\rm R}$ or coercivity $H_{\rm C}$, necessary for AOS to occur. Moreover, the decrease of the coercivity by laser irradiation at v = 250 kHz as well as at the AOS onset temperature of T = 345 K at v = 10 kHz is the same: $H_{\rm C} = 1.2 \text{ kOe}$ [Figs. 3(a) and 4]. Hence, the laserinduced increase of the sample temperature at v = 250 kHz is at least 45 K, which causes a reduction of $M_{\rm R}$ of about 34%, as measured by SQUID-VSM.

Our experiments revealed the decisive role of the stationary magnetic properties of the Tb-Fe film on AOS. However, it is still unclear if either the decrease of the remanent sample magnetization $M_{\rm R}$ or the decrease of the coercivity $H_{\rm C}$, or maybe both, are important for the AOS process. To answer this question we investigate a sample with a different Tb concentration. Tb₂₉Fe₇₁ has a low-remanent magnetization of $M_{\rm R} = (112 \pm 10) \,\text{kA/m} (112 \,\text{emu/cc})$ at $T = 300 \,\text{K}$ (Ref. [10]), which is almost equal to that of $Tb_{34}Fe_{66}$ at 370 K [$M_{\rm R} = (106 \pm 20) \,\text{kA/m} (106 \,\text{emu/cc})$]. In contrast, the coercivity of $Tb_{29}Fe_{71}$ amounts to $H_C = 5.7$ kOe, which is more than 4 times higher compared to the previous sample of $Tb_{34}Fe_{66}$ with $H_C = 1.2$ kOe at 370 K. To exclude any laser-induced heating effects, the experiment is carried out at $v = 10 \,\text{kHz}$ repetition rate at room temperature. Thus, only if AOS relies on a low-remanent magnetization $M_{\rm R}$, should all-optical switching be witnessed. Indeed, we find AOS with a threshold fluence of $F = (1.7 \pm 0.2) \,\mathrm{mJ/cm^2}$ at room temperature (Fig. 6). The used fluence is in the same range as the threshold for PTD in Tb₃₄Fe₆₆ at 10 kHz. Hence, this observation verifies the decisive role of a low-remanent sample magnetization for the AOS process, while at the same time the coercivity seems to be less important because it strongly differs for the two samples.

According to the low-remanence criterion, we are now able to reconcile previous conflicting measurements of AOS with cooled [8] and heated [9–11,13,14,24,25] magnetic films. In general, increasing the sample temperature decreases the remanent magnetization $M_{\rm R}$ and facilitates AOS. However, in Gd₂₂Fe_{68.2}Co_{9.8} decreasing the sample temperature towards the magnetic compensation point $T_{\rm M}$ around 100 K (Ref. [26]) also leads to a reduction of $M_{\rm R}$ and hence to the reappearance of AOS, if the sample is cooled instead of heated [27–29]. This



FIG. 6. (Color online) All-optical helicity-dependent magnetic switching (AOS) in Tb₂₉Fe₇₁ at $\nu = 10$ kHz. Substrate temperature is T = 300 K. (a) Initial homogeneously magnetized state M^- . (b) AOS at a threshold fluence of $F = (1.7 \pm 0.2)$ mJ/cm², after irradiation with right-handed circularly polarized laser pulses (σ^+). (c) Erasing of the written domain structure at the same fluence as in (b) by using left-handed circularly polarized laser (σ^-) pulses.

observation also applies to Tb-Co, where AOS is witnessed when crossing the magnetic compensation point, i.e., a low M_R is established [24]. We therefore find that the low-remanence criterion for AOS is not only limited to Tb-Fe but also applies to GdFeCo and Tb-Co (Fig. 7), which we will discuss in more detail in the next section.

AOS occurs in Tb-Fe for a maximum value of the remanent sample magnetization of $M_{\rm R} = 220 \,\text{kA/m} (220 \,\text{emu/cc})$ at $\nu = 250 \,\text{kHz}$ (Ref. [10]). Above this magnetization threshold, only PTD is found (Fig. 7). To verify that such a lowremanence criterion also applies to other RE-TM alloys, we first investigate the reported AOS ability in GdFeCo (e.g., Refs. [6–8,11,22,26,30]) concerning remanent sample magnetization $M_{\rm R}$. For Gd_x(Fe₉₀Co₁₀)_{100-x} AOS is observed in the range between x = 22 and 28 atom % where $M_{\rm R}$ is below 125 kA/m (125 emu/cc) [31] (Fig. 7).

For Tb-Co similar low values for $M_{\rm R}$ are found [24,32], where $\text{Tb}_{23}\text{Co}_{77}$ [$M_{\text{R}} = 100 \text{ kA/m} (100 \text{ emu/cc})$] and Tb_{26} $Co_{74} [M_R = 150 \text{ kA/m} (150 \text{ emu/cc})]$ exhibit AOS using laser pulses of 400-fs duration at a repetition rate of v =6.3 kHz (Fig. 7). For 10-ps laser pulses at v = 5 kHz the remanence threshold in Tb-Co increased to about 375 kA/m (375 emu/cc). The higher value for M_{R} with picosecond switching pulses is due to the strong influence of the pulse duration on the demagnetization ability. For longer pulses a higher fluence is needed to obtain a similar reduction of the sample magnetization compared to short pulse excitation [33]. This effect is caused by the different temporal and spatial heating characteristics for femtosecond and picosecond durations. In both cases electrons are directly excited by the laser pulses and the lattice is only indirectly affected via the electron-phonon interaction [34]. However, with longer laser pulses the fraction of energy which is transferred during the pulse from the excited electrons to the phonons increases [35]. In that way, the initial difference of the electronic and the phononic temperature becomes smaller. For 10-ps-long laser pulses the electron and lattice temperatures



FIG. 7. (Color online) Overview of all-optical switching (AOS) in $Gd_xFe_{100-x-y}Co_y$ [6–8,11,22,26,27], Tb_xFe_{100-x} [10], Tb_xCo_{100-x} [24], and [Co/Ir/Co/Ni/Pt/Co/Ir]×5 [25]. Red dots and light-red area indicate AOS. Black squares and light-gray area denote pure thermal demagnetization (PTD). Red (black) arrows represent the decrease (increase) of sample magnetization due to heating; the solid red arrow shows our result obtained in $Tb_{34}Fe_{66}$. The dashed red and black arrows visualize the trend in Tb_xCo_{100-x} . (See main text for details.)

are almost equal [22], and the electron-phonon cooling and heating times are slowed [35] by about a factor of 7 compared to excitation with 100-fs pulses [22]. In addition, the laterallaser-heated area enlarges for increasing pulse durations [36]. These effects result in a higher AOS threshold fluence for increased pulse durations [22,30]. They also lead to an elevated lattice temperature in the center of the laser-heated area, which causes a reduction of remanence (indicated by red dashed arrows in Fig. 7) and promotes AOS, similar to our observation for Tb-Fe.

However, this effect only occurs for samples where the magnetization of the rare-earth sublattice prevails. For samples where the magnetization is dominated by the transition metal, an increase in sample temperature results in an increased magnetization. Therefore, $Tb_{16}Co_{84}$ with $M_R = 250 \text{ kA/m} (250 \text{ emu/cc})$ does not exhibit AOS [24] (Fig. 7, black dashed arrow), because here a slight increase of the



FIG. 8. (Color online) Remanent sample magnetization $M_{\rm R}$ versus temperature for a Tb₁₉Co₈₁ amorphous alloy film of 20 nm thickness, measured by SQUID-VSM magnetometry in the temperature range T = 4-370 K. $M_{\rm R}$ amounts to 300 kA/m (300 emu/cc) at room temperature and increases for rising temperatures.

sample temperature does not result in a reduction but rather in a rise of M_R for a Co-dominated Tb-Co sample (Fig. 8).

The low-remanence criterion is not only limited to ferrimagnetic alloys. AOS was recently discovered in an artificial zero-moment magnet [37]. This heterostructure consists of two layers of Tb-Fe, where each has a remanent magnetization above 220 kA/m (220 emu/cc) and could not be individually switched by AOS. In contrast, the bilayer exhibits zero remanence at room temperature due to an antiferromagnetic coupling of the two sublayers and consequently shows

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AOS [37]. AOS was also witnessed for a synthetic REfree ferrimagnet consisting of antiferromagnetically coupled [Co/Ir/Co/Ni/Pt/Co/Ir] \times 5 multilayers (Fig. 7) with a room temperature remanence of about 150 kA/m (150 emu/cc, Ref. [25]). Therefore, we find that the low-remanence criterion for AOS is general because it also applies to other RE-TM alloys, heterostructures, and even RE-free synthetic ferrimagnets.

In conclusion, we have demonstrated that the occurrence of AOS in $Tb_{34}Fe_{66}$ can be controlled by laser-induced heating or by substrate heating with a simple heating resistor. Both lead to a decrease of the remanent sample magnetization M_R , which facilitates AOS. Furthermore, we showed that the low-remanence criterion is not limited to Tb-Fe. It also applies to GdFeCo and Tb-Co amorphous alloys and even to artificial heterostructures and RE-free synthetic ferrimagnets. Our results are important for a technological implementation of AOS, because the AOS ability can now be controlled *in situ* by tailoring the sample remanence. Furthermore, our work may be extended to switching on the nanoscale. For example, local Joule heating via a current flow through a nanowire [38] may control AOS on small spatial scales.

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