Antiferromagnets see the rainbow

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Since Michael Faraday's discovery in 1845 of the direct interaction of light with magnetic materials our understanding of both has advanced considerably. Yet, the ability to control magnetic order with light has only been demonstrated in the past decade. Recent advances have shown the ability to store information with femtosecond laser pulses in different kinds of magnetic material including ferrimagnets[1], synthetic ferrimagnets[2,3] and most recently ferromagnets[4]. All of these methods of all-optical control of magnetism rely on symmetry breaking either in the dynamic properties of the material[3,5] or through magnetic circular dichroism[6]. Antiferromagnets on the other hand present a particular challenge in that they possess no macroscopic magnetization and are therefore hard to manipulate by direct means.

Writing in *Nature Photonics*, Sebastian Manz and co-workers now report that by exploiting the strong coupling between ferroelectric and magnetic order in $TbMnO_3$, they can achieve deterministic all-optical control of an antiferromagnet[7]. Using different wavelengths of light to control the degree of heating by laser excitation allows the stray electric field to determine the antiferromagnetic ordering in the surface of the material. This opens up the exciting possibility of all-optical control for a previously inaccessible class of magnetic material with potential applications in ultrafast optics and spintronics.

Ferroelectric materials possess a net electric polarization, in the same way that ferromagnetic materials possess a net magnetic moment. It is perhaps surprising then that most ferroelectric materials are in fact non-magnetic. $TbMnO_3$ is an unusual material in that it displays both antiferromagnetism and ferroelectricity at low temperature and because the two properties are strongly coupled[8]. Hence, a strong magnetic field is able to saturate the ferroelectric polarization, and an electric field is able to form a single-domain antiferromagnetic state. In $TbMnO_3$, the ferroelectricity arises due to the incommensurate spiral magnetic structure with a natural periodicity that is different from the underlying crystal lattice. This property breaks inversion symmetry and leads to a net electric polarization in the material. Due to crystal symmetry there are two distinct spin structures possible with an opposite sense of rotation of the spin spiral, with two electric polarization states. At higher temperatures, however, the magnetic structure is different due to thermal spin fluctuations leading to a linear magnetic structure that does possess inversion

symmetry and thus has no ferroelectric polarization. The strong coupling between the ferroelectricity and antiferromagnetism and its temperature dependence therefore provides a way to control the magnetic structure through laser heating.

The experimental set-up of Manz *et al.*[7] exploits the ferroelectric polarization in a novel way to control the formation of ferroelectric domains after laser heating (Fig. 1). A single-crystal $TbMnO_3$ sample is first saturated in a single-domain state with an electric field. The laser heating is applied to the top layer, which causes a change in magnetic order and thus a transient loss of ferroelectric order. Due to the presence of He gas on the surface an inverse temperature gradient develops so that the top of the film cools faster than the middle of the sample. As the surface cools, it feels an electric field from the unheated region causing the opposite polarization state to form. As the cooled region propagates downwards, this domain grows, leaving a surface region with the opposite polarization. Here the inverse temperature gradient is critical to the switching process as it allows the reversed domain to grow sufficiently to be stable while preventing the original domain from propagating up from the unheated region. So far, the laser heating and cooling is only able to reverse the surface region opposite to the rest of the film — a deterministic but not particularly useful process for storing information.

Manz *et al.* have cleverly utilized optical dichroism using different frequencies of light to control the penetration depth of the laser pulse[7]. Using a higher-frequency light ω_2 reduces the absorption and so the laser heats a shallower surface region of the film (Fig. 1b). In this case, the larger reversed region provides a polarizing electric field in the same direction as the lower region of the film, and the surface polarization is reversed along the original direction. Illumination with light of the original frequency (Fig. 1a) restores the two-domain state, representing all-optical control of an antiferromagnetic material by tuning the colour of the light. This extends all-optical switching to the full range of magnetically ordered materials, and opens up exciting fundamental questions and practical possibilities.

Compared with the all-optical switching of ferrimagnets and ferromagnets, the reversal mechanism found by Manz *et al.* is very different and relies on the strong coupling of ferroelectric and antiferromagnetic order[7]. However, it raises important questions about the optical control of antiferromagnetic materials. Dichroism is known to be important for all-optical switching in ferrimagnets and ferromagnets in controlling the amount of heat transferred to the sample. The same is apparently true in the present work, but is this generally true? In principle, circularly polarized light could also be used to drive antiferromagnetic switching through magnetic circular dichroism, where different helicities of the light have different absorption coefficients. This raises the further question of whether direct angular momentum transfer to the antiferromagnet through the inverse Faraday effect is also able to

directly reverse the antiferromagnetic order. Our present understanding is that in general this is not possible for ferrimagnets and ferromagnets, but this may not be the case for antiferromagnets due to the spiral ordering of the antiferromagnetic spins. Little is known about the atomic-scale spin and ferroelectric dynamics and their role in the formation of the reversed domains and their exact interaction with light. Addressing these questions could raise the possibility of direct switching of an antiferromagnetic material on the picosecond timescale as seen with ferrimagnets.

Antiferromagnets have so far had limited practical applications due to their lack of a macroscopic magnetization. However, the field of spintronics and magnonics have opened up a range of exciting possible applications in the manipulation of pure spin currents where antiferromagnets are of great interest due to their intrinsically faster spin dynamics. The all-optical control of antiferromagnets adds a further dimension, opening up new opportunities in data-storage and information-processing applications and offering a direct way to control their magnetic order. While an exciting possibility, practical applications will require advanced materials to make such an effect practically useful. The first challenge will be developing materials that exhibit antiferromagnetism and ferroelectricity at room temperature. Hexaferrites are promising candidates, but their complexity makes them challenging for applications. Another enticing possibility is a synthetic multiferroic heterostructure, which combines different types of ferrioc behaviour. For example, one could combine ferroelectrics and ferromagnets to develop a material that exhibits the desired physical behaviour for all-optical switching while being able to separately optimize the different materials, for example using high-temperature antiferromagnets such as IrMn. The final challenge for applications is enabling these effects at the nanoscale, which requires advances in nanophotonics in both light delivery and sensing.

The work of Manz *et al.* has added antiferromagnets to the growing number of magnetic materials that can be manipulated by light, raising questions about the microscopic mechanisms and whether this possibility is general or specific to ferroelectric materials. If the fundamental and practical challenges of all-optical magnetic switching can be overcome then this could be the genesis of magnetophotonics, where the original discovery by Faraday becomes a cornerstone of exciting new technologies.

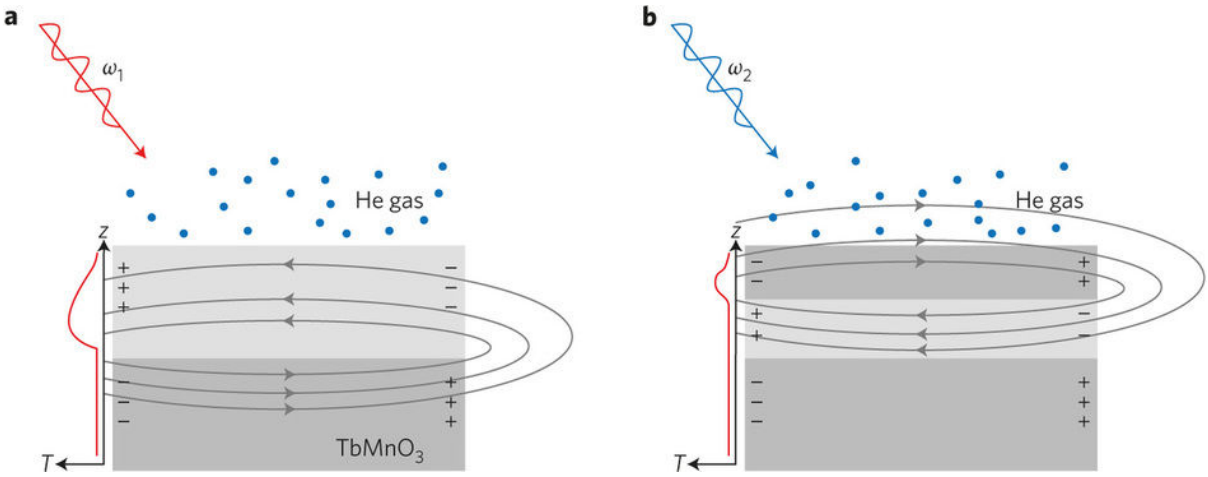

Figure 1 | Schematic diagram showing switching process for $TbMnO_3$. **(a)** Illumination with the initial pulse causes the stray electric field to reverse the top layer of the film due to the inverse temperature gradient, shown schematically with a temperature profile $T(z)$. **(b)** A second pulse with higher frequency, ω_2, limits the light absorbed in the film and causes a thinner top region to reverse in the original direction, giving a complete switching process.

References

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