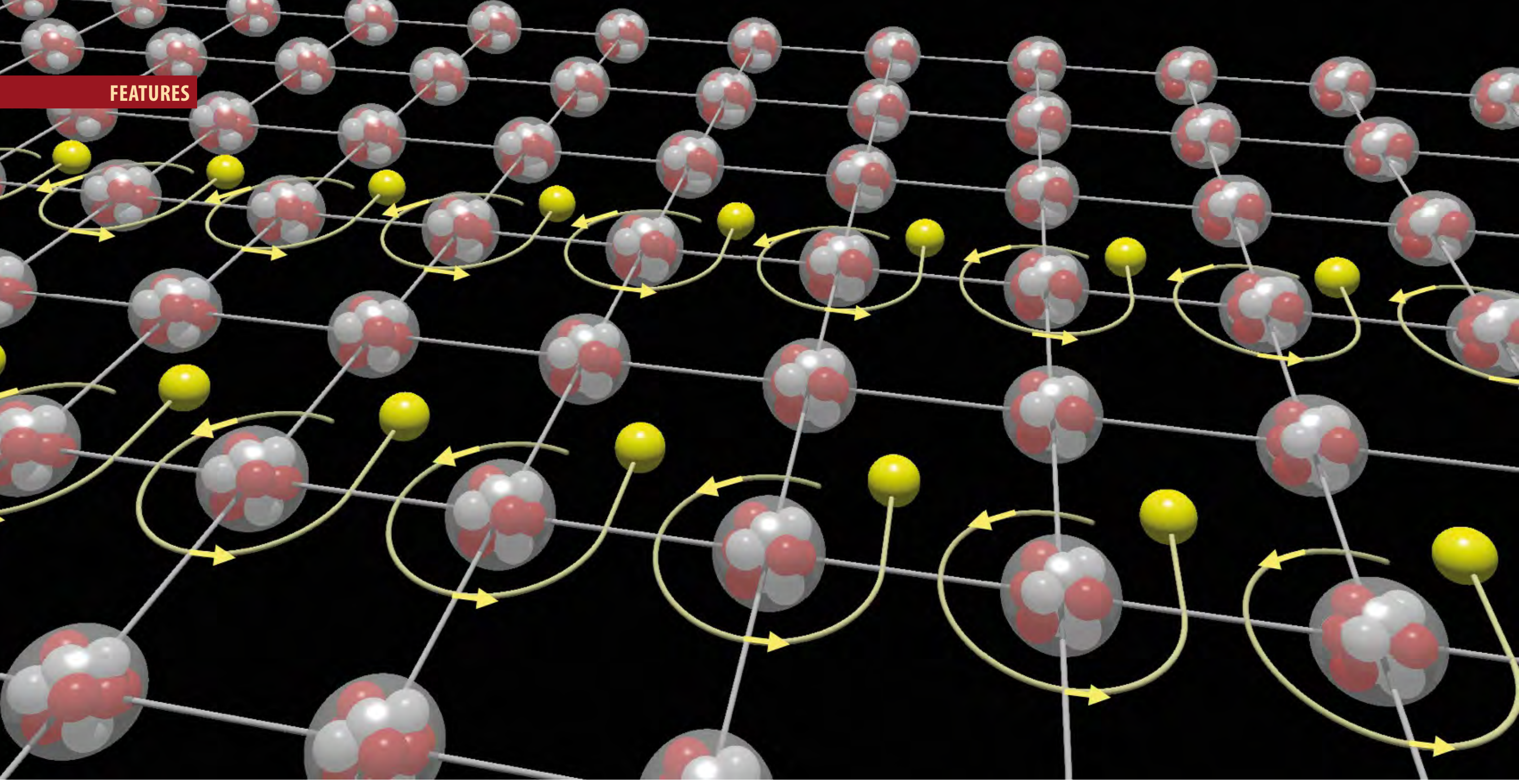

NON-EQUILIBRIUM ORBITAL ANGULAR MOMENTUM FOR ORBITRONICS

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Efficient manipulation of magnetization by electrical current is a key aim in spintronics. The state-of-the-art theories and experiments in spintronics show that harnessing non-equilibrium orbital angular momentum can significantly enhance the efficiency due to novel torques. Devices are based on environment-friendly materials, which has been difficult to achieve by the mechanisms based on spin only, and this has also kickstarted a new emerging field of research: orbitronics.

Physical principles of spintronic memory

In spintronics memory devices, a bit of information is encoded in the configuration of magnetization, *e.g.* "0" and "1" for the states with magnetization up and down, respectively. Reading the magnetic bit has become possible thanks to the discovery of giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR), in which the electrical resistance of a ferromagnet/nonmagnet/ferromagnet (FM/NM/FM) heterostructure varies significantly depending on whether the two FMs have parallel or antiparallel magnetization configurations [1]. As shown in Figs. 1a and 1b, the physical principle of for instance the GMR effect is analogous to an optical polarizer. When electrical current flows across the FM/NM/FM junction, the first FM acts as a spin polarizer which filters only the electrons whose spin direction is the same as that of the FM. If the magnetization direction in the second FM is parallel to that of the first FM, the electrons can easily pass through (Fig. 1a). In the anti-parallel configuration, on the other hand, the spin-polarized electrons by the first FM are blocked by the second FM (Fig. 1b).

An obvious way to "write" the magnetic bit of information is to use a magnetic field, but this method exhibits poor scaling. Also, generating a magnetic field requires an additional conductor in which electrical current flows. The idea of manipulating the magnetization by electrical current was proposed in 2000's, by injecting spin-polarized electrons into a FM (Fig. 1c) [2]. The injected spins interact with the magnetization, and eventually, the outgoing electrons will acquire the spin polarization of the FM. Since the total angular momentum is conserved, the difference of the spins of the in and out states implies that the magnetization must experience a torque as a back action. The corresponding process is called the spin torque, and it enables efficient manipulation of magnetization by electrical current. The GMR and spin torque are the key mechanisms behind spintronic memory devices such as magnetic random-access memory (MRAM), which can serve as a robust hardware for in-memory and neuromorphic computing devices as well as for storage-class memories.

Generating non-equilibrium spin

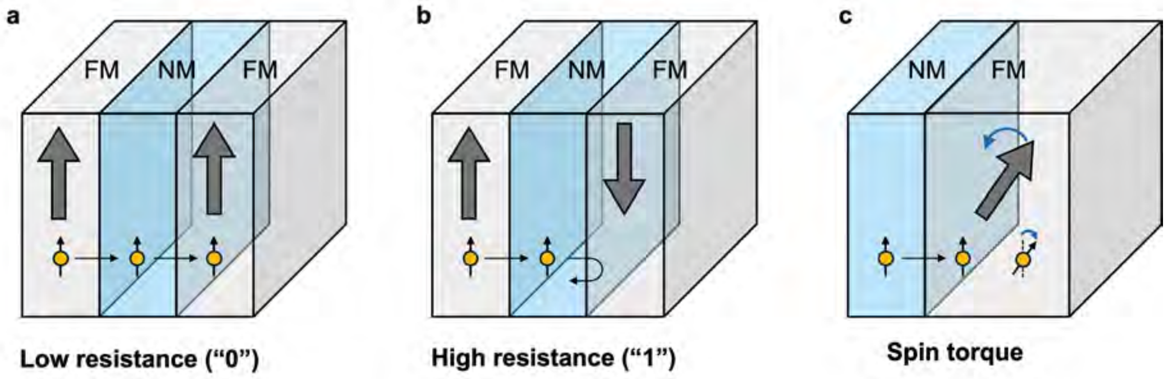
Although a FM may be used as a spin polarizer, other mechanisms exist for generating non-equilibrium spin even in NMs by utilizing relativistic spin-orbit coupling (SOC). Two well-known mechanisms are the spin Hall effect (SHE) and spin Edelstein effect (SEE), in which an external electric field induces non-equilibrium spin current and density, respectively (Figs. 2a and 2b) [3]. These have become of major interest in spintronics research for more than ten years, as magnetization switching by the electrically generated non-equilibrium spin was experimentally demonstrated [4]. This was surprising because the SOC effects have been so far regarded weak, giving mostly perturbative corrections to other strong effects that do not require the SOC. In fact, it has been found that the efficiencies of the SHE and SEE can be large in materials with heavy elements whose SOC is sizable. Since then, researchers have proposed various novel material systems that may enhance the relativistic effects by, *e.g.*, Rashba-type interfacial states, Dirac surface states of topological insulators, oxide heterostructures, metal alloys, *etc.* Despite noticeable progress within more than a decade, the efficiency of SHE and SEE could not be improved significantly beyond a certain limit. Also, many studies focused on materials including heavy elements (Ta, Pt, Au, Bi) and even rare-earth elements (Gd, Tb, Dy, Ho) owing to their large SOC. However, dependence on heavy and/or rare-earth elements has raised a concern in spintronic device applications because they are not only scarce and costly but also environmentally harmful. In fact, many of them are included in the EU's List of Critical Raw Materials [5].

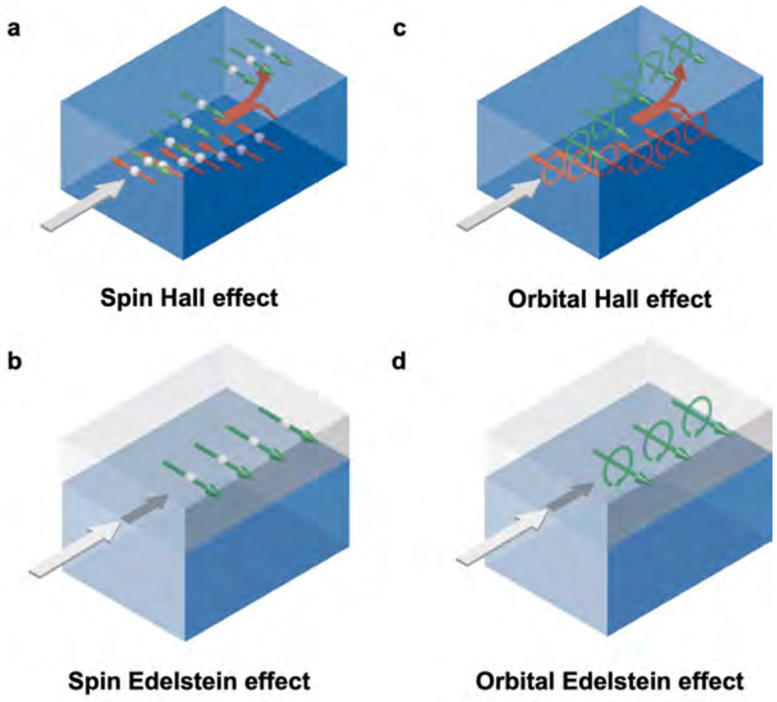
Orbital degree of freedom: Quenched in solids?

As the name of the field suggests, spintronics research has focused primarily on the spin degree of freedom, and the role of other degrees of freedom has been relatively overlooked. However, quantum mechanically, spin is only one part of the electron's angular momentum, with its orbital degree of freedom being a second source. Although this is a well-known statement, the orbital angular momentum (OAM) has been considered unimportant for spintronics because the orbital motion of electrons is suppressed in a crystalline environment. This is known as "orbital quenching" which is discussed in textbooks on solid-state physics.

Nonetheless, the idea of generating current of OAM-polarized electrons was theoretically proposed in 2005 in hole-doped Si [6] and in 2009 in transition metals [7]. These works predicted an orbital analog of the SHE, namely the orbital Hall effect (OHE). Similarly, the orbital Edelstein effect (OEE), which is an orbital analog of the SEE, was also theoretically proposed in 2018 [8]. The OHE and OEE are schematically illustrated in Figs. 2c and 2d, respectively. However, these works did not trigger much interest at the time, probably due to the focus of the community on spin-based effects, such as *e.g.* the discovery of the spin Hall effect in 2004, and their interpretation in terms of spin only. The rich physics of the orbital degree of freedom in transport phenomena started gaining significant attention only since very recently.

▾ **FIG. 1: Physical principles of spintronic memory. (a,b)** A magnetic bit of information can be read for instance by the giant magnetoresistance in the FM/NM/FM structure, where the first and second FMs act as a spin polarizer and analyzer, respectively. **(c)** When the injected spin is not parallel to the magnetization in the FM, magnetization dynamics is induced as a result of the back action of the change of the spins of conduction electrons.





▲ **FIG. 2:** Schematic illustrations of **(a)** spin Hall effect, **(b)** spin Edelstein effect, **(c)** orbital Hall effect, and **(d)** orbital Edelstein effect. When an electric field is applied, the spin(orbital) Hall effect generates a transverse spin(orbital) current. In the spin(orbital) Edelstein effect, spin(orbital) is accumulated by an external electric field.

OAM in non-equilibrium

In *equilibrium*, the OAM is strongly suppressed when compared to the spin, with difference that at times reach orders of magnitude. This is because the OAM is induced by SOC as a secondary effect in the presence of the spontaneous ordering of the spin which is more robust. Analogously, it has been commonly assumed that the *non-equilibrium* OAM and its related effects mentioned above must be suppressed in crystalline solids unless the SOC is strong. However, some of us have explicitly demonstrated that the hierarchy between the OAM and spin in non-equilibrium is exactly opposite to that in equilibrium: upon applying an external electric field, non-equilibrium OAM or its current can be generated robustly despite the orbital quenching and even in the absence of the SOC [9]. When sizeable SOC is present, the spin "follows" the OAM. This is because the electric field couples directly to the orbital part of the wave function, *i.e.* a charge distribution, via a dipolar coupling. The work has uncovered an intrinsic magneto-electric mechanism by which the OAM is induced by an external electric field, even if the OAM is completely quenched in equilibrium, demonstrating that the equilibrium and non-equilibrium properties of the OAM are completely different. Consequently, it has been theoretically shown that the OHE can be huge in 3d metals such as V, Cr, and Mn, in which the SOC is weak and thus the SHE is negligibly small [10]. Interestingly, these materials have been traditionally considered not very useful for spintronics. The theoretical prediction of large OHE was experimentally confirmed recently by optically measuring the orbital accumulation driven by the OHE in Ti and Cr thin films [11,12].

Orbital torque for spintronics

Fundamentally, the physical principle of magnetization dynamics is the conservation of total angular momentum, including both the spin and orbital contributions. The well-known mechanism of the spin torque, however, considers only the conservation of the total spin, ignoring the orbital contribution. The possibility of inducing magnetization dynamics by non-equilibrium OAM, which is nowadays called the "orbital torque", has been theoretically proposed in 2020 (Fig. 3a) [13]. In the mechanism of the orbital torque, the SOC is still necessary for the OAM to interact with the magnetization. This might be considered as a bottleneck of the efficiency, but the orbital torque can be comparable or even larger than the spin torque because the efficiency of the electrical generation of non-equilibrium OAM, *e.g.* by the OHE or OEE, is often much higher than that the equivalent effects for the spin.

Numerous experiments have demonstrated high efficiency of current-induced magnetization dynamics even in systems without any heavy element by utilizing the OAM instead of, or, together with the spin. This has attracted significant attention and has become one of the most important topics in spintronics research nowadays because the orbital torque provides a way to avoid using heavy and/or rare-earth elements for device application, thus avoiding the problem of critical raw materials. Spintronics can go green by harnessing non-equilibrium OAM.

A novel strategy to significantly enhance the efficiency of current-induced magnetization dynamics is to convert the OAM into the spin in another layer, which is inserted between the OAM-generation layer and the FM (Fig. 3b) [14]. The insertion layer acts as a highly efficient "orbital-to-spin" converter if the SOC is sufficiently strong such as Pt. For example, this idea has been first demonstrated in FM/Pt/surface-oxidized Cu, where the surface-oxidized Cu exhibits strong OEE and Pt converts the OAM into spin [14]. While even an ultrathin (~ 1 nm) Pt insertion layer is already sufficient to convert the OAM to the spin, it has been shown that also the ferromagnets themselves can convert the OAM into spin (Fig. 3a). Experimental demonstrations include some early work using permalloy [15] and more recently Ni was shown to be very efficient for this conversion [16, 17].

Orbitronics: Beyond spintronics

The traditional solid-state physics has taught us that the OAM is suppressed in equilibrium, but recent works have shown that the OAM manifests crucially in non-equilibrium, which can also interact with other degrees of freedom such as local moments and be converted into the spin. Because of the possibility of enhancing the efficiency of current-induced magnetization dynamics with abundant and environment-friendly materials, utilizing non-equilibrium OAM has become one of the most important topics in spintronics research. Meanwhile, there are yet numerous unknowns on the fundamental properties of the orbital dynamics and transport, such as orbital relaxation, dephasing, and its lifetime. As a result, an interdisciplinary field of research called "orbitronics" working with generation, detection, transport, and manipulation of the OAM, has emerged [18]. All the time we see more and more exciting results being reported, and new materials and systems are being explored. This strongly suggests that orbitronics has potential to become one of the major topics in condensed matter physics, which can be useful for devices but may also solve grand puzzles in other fields such as superconductivity, multiferroics, and ultrafast phenomena. ■

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Dongwook Go obtained his PhD from POSTECH in South Korea in 2019, where he worked as a postdoc for one year. Since 2020, he joined the group of Yuriy Mokrousov at JGU Mainz as a postdoctoral researcher.

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▼ **FIG. 3:** (a) Schematic illustration of the orbital torque, in which magnetization dynamics is induced by the injection of OAM-polarized electrons. (b) The OAM can be converted to the spin in the insertion layer by the SOC, by which the OAM can be harnessed for enhancing the efficiency of the spin torque. For the demonstration of the effect, an additional Pt layer needs to be placed such that the SHEs from the two Pt layers cancel out and only the orbital-to-spin conversion contributes to the torque on the FM.