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Advances in Magnetic Microscopy

M. R. Freeman and B. C. Choi

A remarkable number of methods for direct, real-space imaging in magnetic microscopy have been demonstrated over the past decade and a half, and the pace of development shows no sign of slowing. Our understanding of magnetism increases as each striking new image of surface and thin-film magnetization is obtained. The continued development of high-performance magnetic information technologies also requires detailed study of the magnetostatics and dynamics of microscopic magnetic structures. Both fundamental curiosity and practical interest now drive us toward innovations in magnetic microscopy for nanometer-length scale and femtosecond temporal resolutions, which are beyond the limits of traditional imaging techniques. This survey is intended to provide an overview of the motivations, accomplishments, and future prospects for this discipline.

Our desire to observe the natural world beyond the limitations of our five senses has driven the development of many new tools. In the case of vision, when objects are too small, distant, or faint, or are moving too quickly or slowly to observe with the unaided eye, we have developed telescopes, microscopes, and cameras to render them visible. The story of imaging of magnetic systems is an interesting case in point, for here we are dealing with a physical phenomenon that is detectable only by our sense of touch with the magnetic force on strongly magnetized objects. The interaction of magnetism with light can be observed by the naked eye only under very special circumstances and only since Faraday's discovery of the magnetic influence on optical polarization. Nonetheless, a very impressive suite of tools has been developed over the intervening 150 years that renders magnetic phenomena and structure as images, thus making them "visible" to the naked eye. Some of these techniques are based on such modern instruments as the scanning tunneling microscope, whereas others harken back to Faraday or even to the direct detection of magnetic forces. Recent advances have given us the ability to directly image magnetic structure on surfaces with atomic resolution and to resolve element-specific contributions to magnetism in complex materials.

The Role of Imaging in Micro- and Nanomagnetism

Magnetism in solids arises on a local scale through quantum mechanical exchange among electrons of neighboring atoms. In ferromagnets, the exchange favors parallel electron spins, and the spatial magnetic structure can range from wonderfully simple—a uniformly magnetized sample—to woefully

complex. Except for special sample shapes, uniform magnetization carries a magnetostatic cost in terms of the energy associated with the long-range interaction between dipoles. The energy can be minimized if the dipoles are not all parallel, hence the formation of magnetic domains. Anisotropy effects that favor the orientation of magnetization along certain crystallographic directions further complicate the situation. The essence of this competition is summarized by so-called "exchange lengths," which dictate the minimum scale on which important variations in the direction of magnetization can occur and are often in the nanometer range. In the non-equilibrium regime, the presence of excess energy leads to additional complication including nucleation and growth of domains, propagation of spin-wave excitations on very short wavelengths, and generation of magnetostatic modes akin to the vibrations of a drumhead. The most successful model of this physics is classical (treating small volumes of material as big magnetic moments) and phenomenological: it is hand-built and constructed to follow reasonable guiding principles such as conserving the magnitude of the big moments, allowing only their directions to change. Only now are the tools becoming available to fully test this description against the complex behavior that can occur even in microscopic specimens and point the way toward improvements. A fully quantum-mechanical treatment of these problems remains intractable, but we can now perform experiments sufficiently detailed and controlled that some might regard them as "analog computations." From an applications perspective, the drive toward goals of magnetic recording at Tbit/in² areal densities and Gbit/s data rates and beyond and of very large-scale integrated magnetic logic, magnetic random access memory, and spintronics calls for very high-performance microscopies applied to materials analysis and device characterization.

Mapping Stray Magnetic Fields

The imaging techniques currently in use may be loosely classified into two (slightly overlapping) groups, according to the physical mechanism of interaction between the probe and sample, that is, stray field mapping and magnetization mapping. The earliest magnetic imaging was of fringing fields around permanent magnets, and helped set the stage for the unified understanding of electricity and magnetism. Circular patterns of iron filings around current-carrying wires (Oersted, Davy, Ampère), and the familiar bowing shapes emanating from the poles of bar magnets (Fig. 1) were fundamental observations described as field lines by Faraday (*J*) and later set to mathematics by Maxwell. For a long period, the iron filing method as refined by Bitter offered the greatest spatial resolution. In the Bitter method, the surface of a magnetic material is dusted with magnetic nanoparticles, which are derived either from a colloidal suspension or from an evaporant if the sample is in a vacuum or cryogenic environment. In the settling of this magnetic smoke, the particles agglomerate in the stray micromagnetic fields at domain walls. The final decoration is imaged under an optical or electron microscope, allowing very small (<100 nm) magnetic features to be resolved in multidomain ferromagnets, or in superconductors penetrated by a magnetic field.

Subsequent to Bitter, various magnetic field imaging techniques have been developed. The instrument most widely used now is the magnetic force microscope (MFM). The MFM is a variant of the noncontact atomic force microscope (AFM) first demonstrated in 1987 (2, 3). In MFM, the magnetic contrast is achieved through the magnetostatic interaction between a ferromagnetic tip and the stray micromagnetic fields from the sample, in particular at domain boundaries. During measurements, the probe tip vibrates perpendicular to the sample surface, and the frequency and amplitude of the vibration change in the presence of gradients due to stray magnetic fields. MFM imaging can achieve spatial resolution of less than 10 nm, and the resolution might yet be improved using advanced tip technology, e.g., by modifying probe tips via focused ion beam milling (4). Advantages of MFM include relatively high spatial resolution and simplicity in operation as well as sample preparation. A drawback has been the difficulty of extracting quantitative information directly from MFM images, although the rather complicated interaction between the magnetic tip and sam-

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ple surface is a very active topic of investigation. Hug *et al.* have developed and patented quantitative magnetic force microscopy methods to calibrate an MFM tip, calculate its stray field, and simulate magnetic force microscopy data (5). Perhaps the most important recent extension of MFM is magnetic dissipation microscopy (6). By monitoring the drive power necessary to maintain a constant amplitude of cantilever oscillation, it is possible to extract additional detail, such as the motion or pinning of domain walls, via the energy loss per cycle.

The highest resolution among methods presently available to map the field distribution around and within thin specimens is provided by techniques using electron microscopes. In Lorentz microscopy, for example, a high-energy (100 to 1000 keV) electron beam is incident on a thin (<150 nm) magnetic sample, and the magnetic contrast is derived from the deflection of the electrons due to the Lorentz force upon passing through the magnetic induction in the sample. This can achieve a lateral resolution of better than 10 nm, bearing in mind that the measurement represents an average over sample thickness (7). Representative applications include the study of the detailed magnetic properties of nanoscale magnetic elements and patterned spin tunnel junction material (8).

Another variation based on electron transmission is electron holography, whose application to magnetic domain observation was originally proposed by Cohen in 1967 (9). It is an electron interferometric method, recording an interference pattern from which the amplitude and phase of an object can be reconstructed (10). This technique can measure absolute values of the magnetic flux in and around thin ferromagnetic samples with very high resolution (~2 nm). The scanning of miniature field-sensing electronic devices such as Hall probes (11) or superconducting quantum interference devices (12) are further alternatives in niche areas of application.

Mapping Sample Magnetization

In physics, one most often prefers to know the actual configuration of magnetization within a sample rather than the distribution of stray field around it, although in some contexts (applications such as conventional magnetic recording) it is very important to know the field. Experiments do not provide sufficient information to calculate one from the other. Working from knowledge of the field, even if accurately mapped over a reasonable volume, the inversion to deduce the magnetization profile is not unique. (Most structures are three dimensional, and we normally measure only the magnetization near the surface by microscopy.)

Numerous high-resolution magnetic imaging techniques measure quantities propor-

tional to the local sample magnetization. These consist of interactions of electron, photon, or neutron beams with the sample or of atoms attached to sharp stylus-like probes. The subtle interaction of electromagnetic radiation with magnetized bodies (again, explored in pioneering research by Faraday) has been well exploited in magneto-optic imaging, which became the workhorse method of the 20th century for observing magnetic microstructure.

In magneto-optics, small rotations of the polarization plane of light upon reflection (Kerr effect) or transmission (Faraday effect) are used to map the magnetization. The first observation of magnetic domains by the Kerr effect was reported in 1951 (13). Magneto-optical recording is based on the same effects. This method allows an external magnetic field to be applied during measurements without influencing the probe, a clear advantage if magnetization dynamics are to be studied. The spatial resolution of the magneto-optic technique is diffraction limited, but researchers often underestimate the power of optical microscopy: the resolution can be almost an order of magnitude smaller than the wavelength. Argyle *et al.*, for example, achieved a resolution close to 100 nm using a high numerical aperture objective with a laser wavelength of 514 nm (14). It should be remembered when comparing different microscopic techniques that the useful spatial resolution is determined by signal-to-noise ratio as well as spot size or interaction length. A quantitative, "platform independent" means of characterization can be obtained from the signal-to-noise spectrum as function of spatial frequency (measured, for example, on a test specimen with a relatively flat distribution of features as a function of spatial frequency). The resolution may then be simply defined as the frequency (and hence inverse as wavelength or spatial scale) at which signal-to-noise ratio crosses unity (5). At some point, a crossover to near-field scanning techniques is essential, however, if one hopes to extend the lateral resolution of optics to the nanometer scale. This continues to prove quite challenging for magnetic imaging, with no dramatic advances beyond the promising demonstrations of, for example, Betzig (15) and Silva (16), who report near-field aperture and particle scattering approaches, respectively.

Imaging Magnetization in an Element-Specific Way

X-ray microspectroscopy can yield much finer resolution than visible light imaging, even with low numerical aperture optics. A still higher resolution approach is x-ray spectromicroscopy, where photoemitted electrons are imaged using electron optics (photoemission electron microscopy or PEEM). A mag-

netic x-ray spectromicroscope, combining the photoemission and magneto-optic Kerr effect techniques, was demonstrated in 1993 (17, 18). In magnetic PEEM, the contrast arises from the asymmetry in photon-absorption cross sections of atomic core levels that depend on the orientation of local magnetization relative to the optical helicity of incident circularly polarized soft x-rays, known as x-ray magnetic circular dichroism (XMCD). The most powerful feature of this technique is that magnetic domains can be imaged in an element-specific manner. Chemical specificity is obtained from core-level selectivity in tuning the excitation to the absorption edge of the desired element. This capability is useful for interrogating individual magnetic layers within multilayer structures composed of different materials (19) and for the study of alloys.

X-ray microscopy also provides a tool for imaging antiferromagnetism, which breaks the symmetry for core-level absorption of linearly polarized x-rays (causing magnetic linear dichroism) (20). Antiferromagnetic thin films are extensively used in exchange bias applications, where exchange coupling across an interface is used to magnetically pin a ferromagnetic layer to an adjacent antiferromagnet. As the latter is immune to changes in the external applied field, the combination can act as a reference layer in magnetic devices such as nonvolatile random access memory (21). This effect is still poorly understood because of the inability of standard techniques to spatially resolve magnetic domain structure in antiferromagnets. The observation of domains in the exchange-coupled thin-film system Co on antiferromagnetic LaFeO₃ was recently reported (22), preceded by the measurement of antiferromagnetic domains in an epitaxial LaFeO₃ and SrTiO₃(001) thin film (23). In particular, comparison of the in-plane projections of the antiferromagnetic axis and of the ferromagnetic spin directions (Fig. 2) reveals that the ferromagnetic Co spins are aligned parallel or antiparallel to the in-plane projection of the antiferromagnetic axis. This result implies that the alignment of spins in the ferromagnetic layer is directly determined by

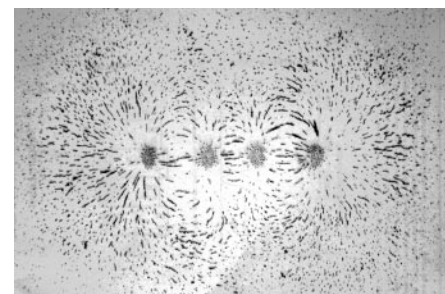


Fig. 1. Now familiar-looking patterns of magnetic field lines around bar magnets, as exposed here by iron filings, were instrumental in early 19th-century science.

the spin directions in the underlying antiferromagnetic layer.

Electron Microscopy

Various imaging techniques based on the interaction of electron beams with a ferromagnetic material have developed into tools for the investigation of surface and thin-film magnetism. The marquee method here is scanning electron microscopy with polarization analysis, or SEMPA (24–27). When a finely focused primary beam of high-energy electrons is scanned over the sample surface, low-energy secondary electrons are emitted from the sample (7, 28). Magnetic contrast can be obtained from the detection of emitted secondary electrons using spin-polarization analyzers such as the Mott detector (29). SEMPA has several unique advantages over most other magnetic imaging techniques: it has the capability of producing a vectorial map of the magnetization; i.e., it measures the magnitude and direction of the magnetization directly. This is because the emitted secondary electrons retain the spin polarization given by the magnetization in the region probed by the incident electron beam. Moreover, SEMPA is sensitive to only the few outermost atomic layers of the sample because of the small inelastic mean free path of the secondary electrons (30). With a short probing depth of the order of 1 nm and focused probe beam size as small as 10 nm, SEMPA is sensitive to minute amounts of magnetic material, as small as 1000 Fe atoms. SEMPA has proven powerful for investigating fundamental problems, such as interlayer exchange coupling of magnetic multilayers, and technological questions, such as the magnetic microstructure of small magnetic device elements. In addition, the SEMPA technique is intrinsically independent of the topography, but the magnetic and topographic images can be acquired simultaneously by measuring the whole intensity and spin polarization of the secondary electrons. Hence, a simultaneous

measurement of the magnetic and topographic images enables in situ comparisons of topological and micromagnetic structures in magnetic thin films.

Like SEMPA, spin-polarized low-energy electron microscopy (SPLEEM) measures quantities directly proportional to the sample magnetization. This technique relies on the spin-dependence of the quasi-elastic scattering of polarized electrons from surfaces. Magnetic contrast is obtained by taking the difference between images measured with each spin direction of the spin-polarized incident beam. SPLEEM is a new approach (31) for resolving surface magnetic microstructure, with lateral resolution presently of the order of 10 nm. This technique has rapidly developed over the last few years for in situ studies of domain structures during thin-film growth, e.g., Co-Au-Co layers grown on W(110) (32).

Glancing at the Interplay Between Magnetism and Atomic Structure

The ultimate resolution goal of most of the microscopic techniques described is to operate below the exchange length, thereby seeing “everything” within the context of classical phenomenology. Beyond this lies another objective: an ability to look directly at the interplay between magnetism and atomic structure with sub-nanometer resolution. In order to achieve this, a new instrument, the spin-polarized scanning tunneling microscope (SP-STM), was proposed and explored starting in the early 1990s (33–35). SP-STM achieves magnetic contrast through the spin-dependent tunneling probability between a magnetic sample and a tip, which behaves as a source of spin-polarized electrons. In principle, tips may be magnetic (or magnetically coated), or optically excited (to create optically induced electronic spin polarization from semiconductor tips). In practice, tip preparation and the separation of artifacts from the measurements have been great challenges.

Another means of exploiting STM tech-

niques for magnetic imaging is ballistic electron magnetic microscopy (BEMM), which has been actively pursued by Buhrman *et al.* (36). BEMM is a variation of ballistic electron emission microscopy (BEEM) (37). Here, an STM tip is used to locally inject current into thin ferromagnetic films separated by thin normal (i.e., non-ferromagnetic) metal layer (upper panel of Fig. 3). Magnetic contrast arises from the spin-dependent scattering of electrons passing through ferromagnetic films. When an unpolarized current from the STM tip is incident to the top ferromagnetic layer, one of the incident spin components will pass through this “spin-filtering” layer with relatively little scattering in comparison to the other spin component. If, at a given lateral position, the magnetization direction of this top layer is strongly aligned (misaligned) relative to that of the bottom spin-analyzer layer, both of the spin components will not (will) be heavily scattered on passage through the structure, yielding a high (low) ballistic current uptake at that position. High-resolution BEMM images are shown in Fig. 3, resolving magnetic domains on the sub-100-nm scale in a Co-Cu-Co trilayer film. The tradeoffs of BEMM compared with SP-STM are straightforward. In the ballistic methods, one loses spatial resolution from

Fig. 2. Domain structures in the antiferromagnetic LaFeO_3 (A) and ferromagnetic Co (B) layer at the same region of the sample. The spectra shown underneath illustrate the origin of the intensity contrast in the images. Comparison of the images shows that the Co domains align with the antiferromagnetic domains (light and dark regions inside outlined areas). [Adapted with permission from (22)]

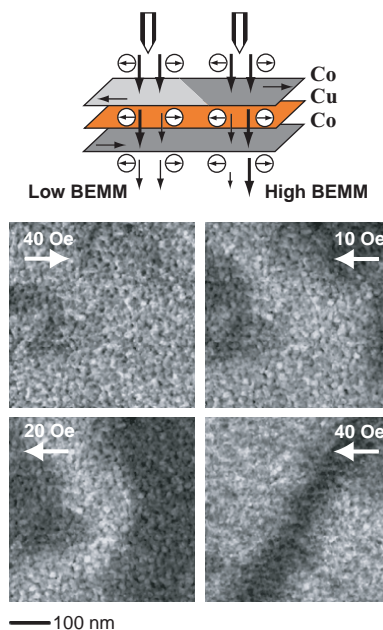
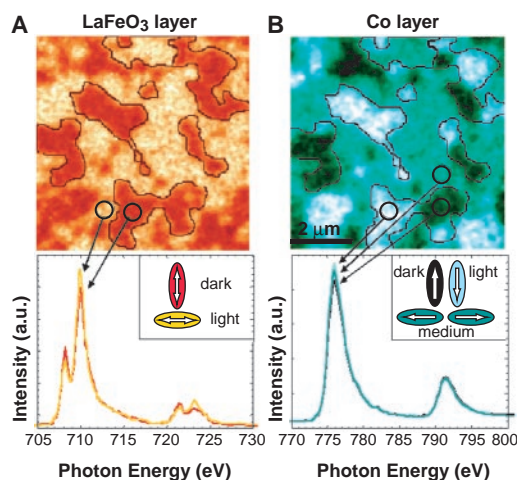


Fig. 3. Working principle of BEMM analogous to “polarizer-analyzer” experiments in optics. A high ballistic electron current flows through the magnetically aligned ferromagnetic layers, and a low ballistic current flows through layers which are in antiferromagnetic alignment. BEMM images in four different magnetic fields of a Co-Cu-Co trilayer film grown on a Au/Si(111) substrate. The darkest regions are regions of antiferromagnetic alignment between the two Co layers. [Adapted with permission from (36)]

angular spread of the emitted electrons, and the sample must have an appropriate layered structure, but it is not necessary to prepare a spin-polarizing tip. The ability to look at buried layers is extremely useful for device structures. Similarly, spin-polarized tunneling-induced luminescence microscopy has produced exciting results correlating spin-injection efficiency with nanoscale surface structure on GaAs (38). Another approach with great potential is the method of local sensing of magnetostrictive effects using force microscopies (39).

The power of SP-STM has been demonstrated in a series of recent papers by the Wiesendanger group, who unraveled the predicted two-dimensional antiferromagnetic state of a pseudomorphic Mn monolayer grown on W(110) surface, where adjacent atoms at nearest-neighbor sites are chemically equivalent but have magnetic moments with opposite directions (40). As an example of SP-STM measurements (Fig. 4), an atomically resolved magnetic image is shown taken with a W-tip coated by 5 to 10 monolayers of Fe (41). In agreement with first-principles calculations (42), the magnetic ground state is a $c(2 \times 2)$ antiferromagnetic configuration. SP-STM opens the door to the investigation of atomic-scale magnetism with complex magnetic structures (43), as demonstrated by its capability to resolve antiferromagnetism at the ultimate limit.

Imaging Spin Dynamics

An important subject to be addressed by magnetic microscopy is magnetization dynamics on all time scales, including the ultrashort regime. High frequency (microwave) and fast time-domain phenomena are fundamental properties that are also closely related to applications such as magnetic data storage. Considering the Gbit/s magnetic switching speeds of present technology, the temporal

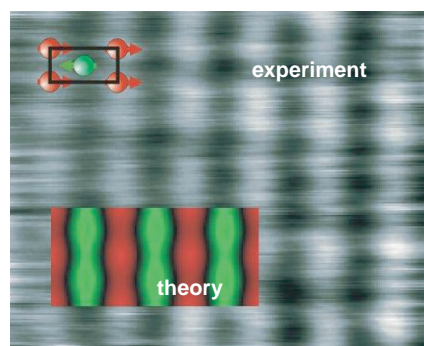


Fig. 4. STM image of a Mn monolayer on W(110) measured with a magnetic Fe tip. Theoretical image and unit cell of the calculated magnetic ground-state configuration are directly compared. Periodic parallel stripes along the [001] direction implies that the magnetic ground state of Mn atoms is the $c(2 \times 2)$ antiferromagnetic configuration. [Adapted with permission from (22)]

resolution necessary for characterizing dynamics in real devices is deep into the subnanosecond regime.

An experimental tool for imaging fast magnetization dynamics has been demonstrated in time-resolved ferromagnetic resonance imaging experiments (44), an extension of nonimaging pump-probe magneto-optical measurements on dilute magnetic systems (45). We stroboscopically imaged micromagnetic samples to access spatiotemporal information about the magnetization state. This approach has been further extended to true “experimental micromagnetics” by including all vector components, as in the imaging of precessional motion of the magnetization reported by Acremann *et al.* (46). Recently, spatially detailed movies of magnetization reversal were reported (47), documenting the change in magnetization reversal mechanism caused by the presence of a transverse field (Fig. 5). Time-resolved imaging with the second-harmonic magneto-optic Kerr effect (48), a good complement to linear magneto-optics, offers extreme sensitivity to the magnetization at surfaces and interfaces (49).

One of the most exciting prospects in the field of magnetization dynamics is time-resolved magnetic PEEM (TR-XMCD and TR-XMLD). Recent efforts to use the pulsed x-ray beam from synchrotron radiation sources for the study of time-resolved and element-selective magnetization dynamics have been fruitful (50, 51).

Prospects

All of the techniques described above continue to evolve in their capabilities, and new methods continue to appear. Another generation of time-resolved PEEM will provide a lateral resolution of a few nanometers combined with a temporal resolution of 50 ps, along with chemical specificity. Another developing technique of great interest in this area is magnetic imaging using resonant scattering of coherent x-rays to allow domain reconstruction from experimental speckle

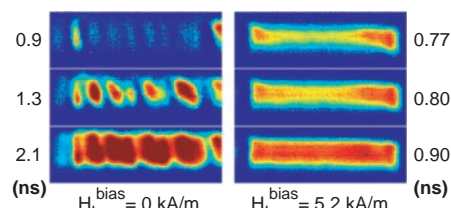


Fig. 5. Spatial profiles of the easy axis magnetization component measured at different time delays after a magnetic pulse was applied. The magnetization reversal mode drastically changes from domain nucleation process (left column) to quasi-coherent domain wall motion (right column) as the transverse bias field H_t^{bias} varies from 0 to 5.2 kA/m. The field of view of each frame is $12 \mu\text{m}$ by $4 \mu\text{m}$ and contains the entire $10 \mu\text{m}$ by $2 \mu\text{m}$ sample. [Adapted with permission from (47)]

patterns. The implementation of these kinds of state-of-the-art techniques at future facilities such as the Linac Coherent Light Source may revolutionize investigations of magnetic dynamics.

Spatiotemporal imaging studies of combined electronic and spin transport have been successfully demonstrated in semiconductor systems (52) and are important to the development of spin electronics. We can anticipate the extension of such studies to metallic systems using some of the techniques described above for direct views of processes including spin-transfer torque (53).

Arguably the greatest promise the scanning probe methods hold is the possibility to build and study magnetic systems from the ground up, combining the techniques of atom manipulation with SP-STM and other pioneering probes of atomic-scale magnetism.

In the words of E. O. Wilson (54), “the search for the ultimate. . . through direct visual observation by steady advances in the resolving power of microscopes. . . satisfies [an] elemental craving: to see all the world with our own eyes.” By some combined spatiotemporal metric, we are still far from the physical limits, although motivated experimentalists are turning dream into reality at a rapid clip. Further developments and accomplishments to anticipate include exchange force microscopy (55), few- or single-spin detection and volume imaging by magnetic resonance force microscopy (56), single-shot and real-time ultrafast microscopy to capture stochastic and chaotic processes, and, lastly, correspondence with numerical modeling (“computational microscopy”). These are exciting times for participants and spectators alike as an era of creativity, ultimately unleashed by the earlier mastery of electromagnetism, continues to unfold.

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REVIEW

Spintronics: A Spin-Based Electronics Vision for the Future

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This review describes a new paradigm of electronics based on the spin degree of freedom of the electron. Either adding the spin degree of freedom to conventional charge-based electronic devices or using the spin alone has the potential advantages of nonvolatility, increased data processing speed, decreased electric power consumption, and increased integration densities compared with conventional semiconductor devices. To successfully incorporate spins into existing semiconductor technology, one has to resolve technical issues such as efficient injection, transport, control and manipulation, and detection of spin polarization as well as spin-polarized currents. Recent advances in new materials engineering hold the promise of realizing spintronic devices in the near future. We review the current state of the spin-based devices, efforts in new materials fabrication, issues in spin transport, and optical spin manipulation.

Until recently, the spin of the electron was ignored in mainstream charge-based electronics. A technology has emerged called spintronics (spin transport electronics or spin-based electronics), where it is not the electron charge but the electron spin that carries information, and this offers opportunities for a

new generation of devices combining standard microelectronics with spin-dependent effects that arise from the interaction between spin of the carrier and the magnetic properties of the material.

Traditional approaches to using spin are based on the alignment of a spin (either “up” or “down”) relative to a reference (an applied magnetic field or magnetization orientation of the ferromagnetic film). Device operations then proceed with some quantity (electrical current) that depends in a predictable way on the degree of alignment. Adding the spin degree of freedom to conventional semiconductor charge-based electronics or using the spin degree of freedom alone will add substantially more capability and performance to electronic products. The advantages of these new devices would be nonvolatility, increased data processing speed, decreased electric power consumption, and increased integration densities

compared with conventional semiconductor devices.

Major challenges in this field of spintronics that are addressed by experiment and theory include the optimization of electron spin lifetimes, the detection of spin coherence in nanoscale structures, transport of spin-polarized carriers across relevant length scales and heterointerfaces, and the manipulation of both electron and nuclear spins on sufficiently fast time scales. In response, recent experiments suggest that the storage time of quantum information encoded in electron spins may be extended through their strong interplay with nuclear spins in the solid state. Moreover, optical methods for spin injection, detection, and manipulation have been developed that exploit the ability to precisely engineer the coupling between electron spin and optical photons. It is envisioned that the merging of electronics, photonics, and magnetism will ultimately lead to new spin-based multifunctional devices such as spin-FET (field effect transistor), spin-LED (light-emitting diode), spin RTD (resonant tunneling device), optical switches operating at terahertz frequency, modulators, encoders, decoders, and quantum bits for quantum computation and communication. The success of these ventures depends on a deeper understanding of fundamental spin interactions in solid state materials as well as the roles of dimensionality, defects, and semiconductor band structure in modifying these dynamics. If we can understand and control the spin

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