Magnetic Semiconductors Enable Efficient Electrical Spin Injection

Dreams of exploiting the spin of the electron in solid-state devices underlie the nascent field of "spintronics." Aspirations for semiconductors have been spurred by the development, in metals, of giant magnetoresistance devices—as found in the read heads of current high-density computer hard drives—and nonvolatile magnetic memory (see the PHYSICS TODAY special issue on magnetoelectronics, April 1995).

A central hurdle for spin-polarized transport in semiconductors has been finding an efficient method of spin injection—that is, polarizing the electrons or holes in the semiconductor to begin with. But last December, two groups reported progress in overcoming this obstacle. Using a paramagnetic semiconductor as a spin injector, Laurens Molenkamp and his coworkers at the University of Würzburg in Germany have observed nearly 90% polarization of electrons in gallium arsenide[1] in a field of 3 T. And a collaboration between the research groups of Hideo Ohno at the University of Tohoku in Japan and David Awschalom at the University of California, Santa Barbara, have seen polarized hole injection into a quantum well from a ferromagnetic semiconductor, which does not require an applied magnetic field.[2]

Early efforts

The behavior and manipulation of spins in semiconductors has been extensively studied by optical means for several decades. Circularly polarized light can preferentially promote electrons of one spin orientation into the conduction band of a semiconductor, and the net polarization in the material can be revealed in the polarization of the emitted light or in the induced rotation of an incident probe laser (see the article by Awschalom and James Kikkawa in PHYSICS TODAY, June 1999, page 33). But for many real-world applications, an electrical means of injecting spins is desirable.

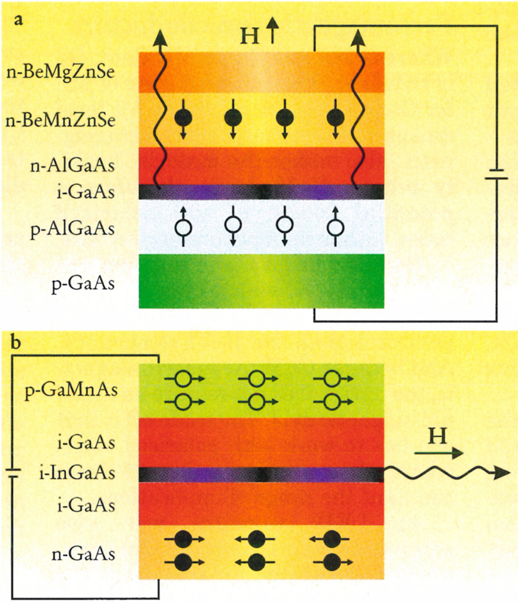
For electrical spin injection, one relies on an applied voltage to pull more electrons of one spin orientation than the other into the device. One therefore needs either a spin-dependent interface or a material that has different densities of states for the two spin orientations—in other words, a magnetic material.

> Semiconductors doped with manganese may provide the answer to a long-standing question in magnetoelectronics.

Attempts at direct electrical injection of polarized electrons and holes have, over the last decade, focused on using a ferromagnetic metal—such as iron, cobalt, or permalloy (80% Ni, 20% Fe)—in contact with a semiconductor. That approach has yielded more controversy than success to date. "The putative spin polarization effects that have been seen have been minuscule—of order 1% or less," comments Michael Roukes (Caltech). "And at that level, there's a host of other parasitic mechanisms that can give rise to the effects seen."

Georg Schmidt, working with Molenkamp and colleagues at the University of Groningen in the Netherlands, has shown theoretically that there are fundamental difficulties to achieving better results with a ferromagnetic metal–semiconductor system.[3] Nearly total spin polarization is needed, but the typical polarization in ferromagnets is only 40–50%. Small devices, in which the transport is ballistic instead of diffusive, might not be subject to the same limitations, however.

The new advances in spin injection come from using all-semiconducting devices incorporating magnetic semiconductors. This approach offers several advantages. Interface properties, such as lattice matching and conduction band offsets, are well understood and controllable. Furthermore, all-semiconducting devices may be integrated better with existing semiconductor and fabrication technology.



ELECTRICAL SPIN INJECTION has been observed using magnetic semiconductors in different layered light-emitting diode structures. **(a)** Manganese-doped BeZnSe is paramagnetic, and has been used to inject spin-polarized electrons into GaAs quantum wells. Unpolarized holes from the other side of the well combine with the polarized electrons to give off circularly polarized light. (Adapted from ref. 1.) **(b)** GaMnAs is a ferromagnetic p-type semiconductor (for a certain range of Mn concentrations) and serves as a source of polarized holes for this InGaAs quantum well. (Adapted from ref. 2.)

The recent electrical spin injection experiments use two different schemes, both involving doping with manganese for polarizing the carriers. The Würzburg group uses a paramagnetic II–VI semiconductor, $Be_xMn_yZn_{1-x-y}Se$, as a spin filter for electrons. The Tohoku–UCSB collaborators use the Mn-doped III–V compound $Ga_xMn_{1-x}As$, which is ferromagnetic, to inject spin-polarized holes. (See the figure at left.)

Paramagnetic semiconductors

Like Zn, Mn is divalent, and thus the partial substitution of Mn for Zn in ZnSe does not add carriers—but it does make the semiconductors paramagnetic. These materials have high g factors (which quantify the coupling of the electron spins to an applied magnetic field)—about 100 times higher than in nonmagnetic semiconductors. Fields of a few tesla create nearly total spin polarization at the Fermi level. The g factors in the Würzburg Mn-doped II–VI compounds are highly temperature-dependent, however, and for the materials to provide efficient spin injection, they must be cooled down to liquid helium temperatures.

The group added n-type dopants to their magnetic semiconductor, making it a source for spin-polarized electrons. Electrons are the preferred carriers in these materials because they have longer spin coherence times than holes, which suffer from strong dephasing due to spin-orbit coupling.

The Würzburg experimenters used a light-emitting diode (LED) structure to monitor the efficiency of spin injection. (A US patent for a spin-LED was issued last year to Berry Jonker of the Naval Research Laboratory.) At the heart of the Würzburg diode is a GaAs quantum well. Because it has a lower conduction band energy and a higher valence band energy than the adjacent layers, the well traps electrons and holes entering it. When the electrons and holes recombine, they emit light.

The paramagnetic semiconductor was put on one side of a GaAs quantum well and p-type nonmagnetic semiconducting layers on the other. When a bias voltage is applied across the device, electrons enter the Mn-doped layer and scatter into the lower Zeeman level; for a sufficiently thick layer, the electrons are spin-polarized by the time they enter the quantum well. The paramagnetic semiconductor thus acts as a "spin aligner." Unpolarized holes enter the well from the other side.

The polarization of the emitted light from the quantum well—about 45% in the Würzburg experiments—is a measure of the polarization of the recombining carriers. The quantitative relationship between the polarizations has one complicating factor, however. In GaAs and other semiconductors with the same zincblende structure, there are two valence bands that are degenerate in the bulk, but become split in energy in narrow quantum wells.

Molenkamp tells us that in the device discussed in their paper, he and his coworkers did not see any splitting between the valence bands in the excitation spectra, and so they took them to be degenerate. Due to an additional transition allowed by the degeneracy, they concluded that the electron spin polarization in the well is twice the optical polarization, or roughly 90%. In subsequent experiments with narrower quantum wells, adds Molenkamp, they do have band splitting, but observe an optical polarization of up to about 90%, indicating a comparable spin polarization.

At a spintronics workshop in Santa Barbara in January, and at the annual meeting of the American Association for the Advancement of Science in February, Jonker reported that he and his coworkers at NRL and the State University of New York at Buffalo have performed similar experiments using paramagnetic ZnMnSe as the spin aligner.[4] They, too, observe about 50% optical polarization. Accounting for the valence band splitting in their quantum well, the researchers quote a lower bound of 50% on their electron polarization. Jonker notes that the paramagnetic material they used, $Zn_{0.94}Mn_{0.06}Se$, has a 0.5% lattice mismatch with the underlying GaAs layers, which is relatively large by semiconductor standards. "The spin transport across the interface is a fairly robust process," concludes Jonker. "It happens in spite of this rather large lattice mismatch."

Fully lifting the degeneracy between the spin-up and spin-down states in the Mn-doped II-VI compounds requires external magnetic fields on the order of 3 T. One possibility for providing this polarizing field may be to use a ferromagnetic metal contact—not to provide the polarized spins, but the fringe field required for full spin alignment in the Mn-doped paramagnetic semiconductor.

Ferromagnetic semiconductors

Whereas Mn and Zn are divalent, Ga is trivalent. Thus, substitution of Mn for Ga in GaAs, which at the 5% level makes the semiconductor ferromagnetic, also makes the material heavily p-type. GaMnAs is therefore a potential source of spin-polarized holes, rather than polarized electrons.

GaMnAs has a Curie temperature of up to 110 K, which may allow for spin injection at much warmer temperatures than the II–VI compounds. And because GaMnAs is ferromagnetic, the device does not require an external magnetic field. The trade-off is that one has to work with polarized holes, whose shorter polarization lifetimes can limit the range of applications.

An LED geometry was also employed by the UCSB–Tohoku team, which sandwiched an InGaAs quantum well between layers of GaMnAs and n-doped GaAs that provided unpolarized electrons. The circular polarization of the emitted light was proportional to the temperature-dependent magnetic moment of the GaMnAs and showed telltale hysteresis in an applied external magnetic field. "It's surprising that it works at all," says Awschalom. "After travelling hundreds of nanometers, the holes should have no polarization left."

The orientation of the ferromagnetic LED was different from that used in the II–VI experiments: The magnetic field was parallel to the layers instead of perpendicular (see the accompanying figure), and the experimenters measured the light emission parallel, not perpendicular, to the surface. This geometry was necessary because the magnetic GaMnAs layer would have altered the polarization of any light passing through it, masking the polarization of the carriers producing the light.

Although the horizontal geometry avoids many possible ambiguities, it is not without its own difficulties. The selection rules relating the polarization of the light to the polarization of the carriers are straightforward with vertical emission, but are more complicated for edge emission. Consequently, it is difficult to infer an actual spin polarization—and hence the injection efficiency—from the measured 2% change in optical polarization.

Tomasz Dietl of the Institute of Physics at the Polish Academy of Sciences, working with Ohno and collaborators at the University of Grenoble 1, has recently developed a model for the ferromagnetism of GaMnAs.[5] They have used their model to predict the Curie temperatures for other Mn-doped semiconductors. For two of them—GaN and ZnO—the calculated Curie temperatures are above room temperature, a definite advantage for device applications.

Although these efforts report achieving spin injection electrically, they are still relying on optical detection. "This work hints at the possibility for real electrical spin injection devices," notes Roukes, "but they're still very far away." Research toward them continues, though, including efforts to better understand spin transport in the solid state, develop higher-temperature magnetic materials, and inject spins into two-dimensional electron gases and quantum dots.

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