

Materials in extreme environments

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The study of materials under harsh conditions is essential to meet a range of energy challenges—from creating better turbines, reactors, and batteries to developing future energy systems in dense plasmas.

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Nature is rich with examples of phenomena and environments we might consider extreme, at least from our familiar experience on Earth's surface: large fluxes of radiation and particles from the Sun, explosive asteroid collisions in space, volcanic eruptions that originate deep underground, extraordinary pressures and temperatures in the interiors of planets and stars, and electromagnetic discharges that occur, say, in sunspots and pulsars. We often intentionally create similar extreme environments—for example, in high-powered lasers, high-temperature turbines, internal-combustion engines, and industrial chemical plants. The response of materials to the broad range of such environments signals the materials' underlying structure and dynamics, provides insight into new phenomena, exposes failure modes that limit technological possibility, and presents novel routes for making new materials.¹

Indeed, exposing materials to those regimes induces new physical phenomena that do not occur under ordinary conditions. Those extreme phenomena are central to many of the most fascinating grand challenges of science, including behavior far from equilibrium (see the article by Graham Fleming and Mark Ratner in *PHYSICS TODAY*, July 2008, page 28), planetary dynamics, the evolution of stars, and the origin of life on Earth.²

Understanding and exploiting extreme environments is critical for facing major societal challenges—particularly in the area of energy—and goes back centuries. For example, Benjamin Franklin wrestled with the hazards of lightning in the mid-1700s, and Benjamin Thompson (later Count Rumford) studied the energetics of gunpowder during the American Revolution.³ Energy technologies often operate at extremes of temperature, pressure, chemical corrosivity, or electric fields. And finding new materials that not only survive but also function under extreme conditions is a major focus in energy research. Researchers must not only observe and understand the behavior of material under such environments but also tailor and control a material's response in order to enhance a device's performance, extend its lifetime, or enable new technologies.

In this article we provide an overview of how materials respond to extremes in energetic fluxes, thermomechanical forces, chemistry, and electromagnetic fields. The subject was the focus of a workshop held 11–13 June 2007 and sponsored by the US Department of Energy's Office of Basic Energy Sciences.¹ Our presentation here contains the highlights; we encourage interested readers to consult the references and the full report from the workshop for more details.

Extreme fluxes

The performance of nuclear fission and future fusion reactors, long-lived radioactive waste forms, nuclear stockpile materials, solar energy conversion devices, and intense laser sources is limited by the damage they can withstand from exposure to energetic particles and intense radiation. Next-generation fission reactors will experience neutron fluxes an order of magnitude higher than do current systems. And the damage from sunlight at Earth's surface, with a peak power of about 1 kW/m², limits the lifetimes of solar collectors (see the article by George Crabtree and Nathan Lewis in *PHYSICS TODAY*, March 2007, page 37). Moreover, lasers used to study fusion will focus light at power densities on the order of terawatts per square meter. The challenge is to synthesize the new materials required to withstand such exceptional particle and photon fluences.

Materials subjected to energetic particles and photons typically fail at fluxes that are a factor of 10 below their intrinsic limits. For example, optically perfect silicon dioxide can withstand energies of 2 MJ/m² without being damaged, but real SiO₂ fails at exposures an order of magnitude smaller, about 200 kJ/m², because of the runaway absorbance of photon energy at nanoscale defects that usually exist on material surfaces.¹ Particle fluxes damage materials through the creation of nanoscale defects in the bulk that subsequently grow by migration and clustering and ultimately create macroscopic defects that lead to failure. For example, neutron irradiation damage begins on the atomic scale with displacements of single atoms to create vacancies and interstitials that diffuse to form larger defects and, ultimately, voids that cause extensive swelling on a macroscopic scale, as shown in figure 1.

Starting small and growing large is a common characteristic of the damage experienced in extreme environments. And the time and length scales involved can be mind-boggling—nine orders of magnitude or more. Capturing the massively multiscale evolution in a single experimental or numerical framework is an important grand challenge still well out of reach. Atomic simulations can follow the early stages of the structural damage evolution in metals, but the sub-picosecond time scale remains beyond observational grasp, and applications to nonmetals remain only poorly understood.

Irradiation can also be used as a tool to create new materials and structures and it can drive materials into novel thermodynamic states. For example, shaped laser pulses can

be used to anneal a material and produce specific microstructures or repair materials that have been damaged by prior irradiation. Specific electronic excited states of an atom can be controlled with laser pulses on the time scale of attoseconds, the time required for light to cross the diameter of an atom. And with femtosecond pulses, the time scale of lattice vibrations, the positions and orientations of atoms and molecules can be controlled. Indeed, it is becoming possible to design light pulses that direct specific chemical reactions or phase transitions by first positioning the atoms in favorable configurations with a femtosecond pulse and then triggering the transfer of an electron from one atom to another with an attosecond pulse.⁴ Oxides can be irradiated with swift heavy ions to create tailored arrangements of imbedded nanoparticles and other unique structures.⁵ Researchers are just beginning to exploit the potential of light-matter interactions to control complex systems (see the article by Ian Walmsley and Herschel Rabitz in *PHYSICS TODAY*, August 2003, page 43).

Pressure, temperature, and strain rate

The behavior of materials subjected to thermomechanical extremes of high pressure and stress, high strain and strain rate, and high and low temperatures is at the heart of many energy problems. New materials that can withstand high temperatures and stresses are required for efficient next-generation turbines and heat exchangers in engines and electrical power plants, where temperatures and pressures reach 1030 K and roughly 40 MPa (400 bar).⁶ Next-generation nuclear reactors will operate around 1300 K, significantly higher than the 625 K used now. Deep inside Earth, the ultimate source of geothermal energy, one finds a domain of megabar pressures and many thousands of degrees, with a central pressure of 360 GPa (3.6 Mbar) and temperatures of 4000–6000 K. But the operating conditions we can expect of any future fusion-power technology are even more severe:⁷ pressures in the gigabar regime, temperatures in the keV range (over 10^7 K), and strain rates above 10^7 s⁻¹.

One principal materials challenge is understanding the origin of enhanced mechanical properties, such as strength, under extreme conditions. Recent deformation studies have demonstrated that pronounced improvements in strength may be achievable with nanocrystalline materials and multilayered, ultrathin films, compared with conventional materials with the same compositions. For example, novel nanoscale architectures such as micropillars may lead to materials systems that are stronger and stiffer than the constituent materials.⁸

Another overarching challenge is finding a way to circumvent nature's traditional linkage of high strength with poor ductility and low toughness. Carbon-based materials

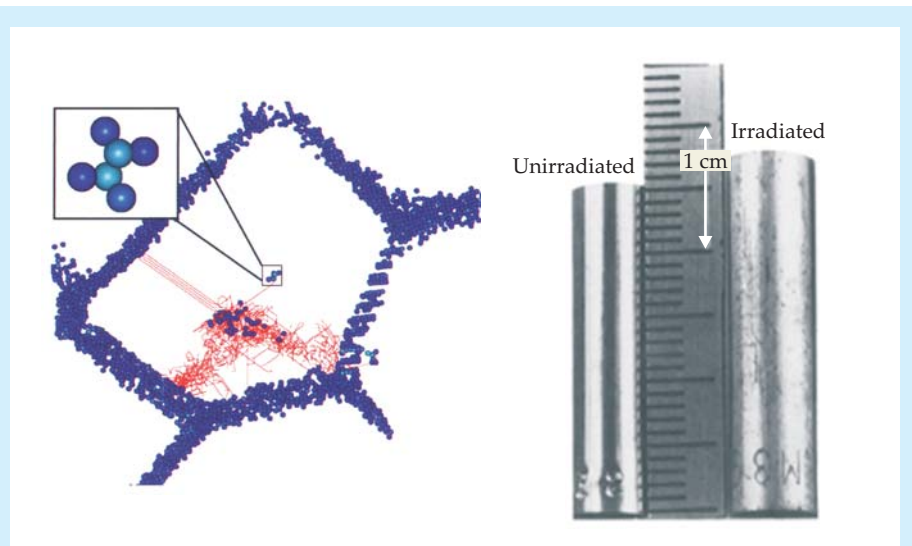


Figure 1. A molecular-dynamics simulation (left) of neutrons (red) irradiating a crystalline alloy of steel shows the grain boundary structure and damage—displaced iron and chromium atoms inside the grain—produced by a 20-kV cascade of neutrons. The inset shows bonded iron and chromium atoms and the white space inside the grain represents perfectly ordered crystal. (Adapted from T. Allen et al., *MRS Bull.* **34**, 20, 2009.) The photograph (right) of two otherwise similar “cold-worked” stainless steel rods reveals the dramatic swelling that occurs when one of them is exposed to a flux of 1.5×10^{23} neutrons per square meter in an experimental breeder reactor. (Adapted from L. K. Mansur, *J. Nucl. Mater.* **216**, 97, 1994.)

may provide unique opportunities in that respect (see box 1). Imaging techniques that can “watch” atoms move and document the clustering of atomic and nanoscale defects on the much larger length scales that control mechanical properties are needed to understand and further optimize enhancements in strength, ductility, and hardness.

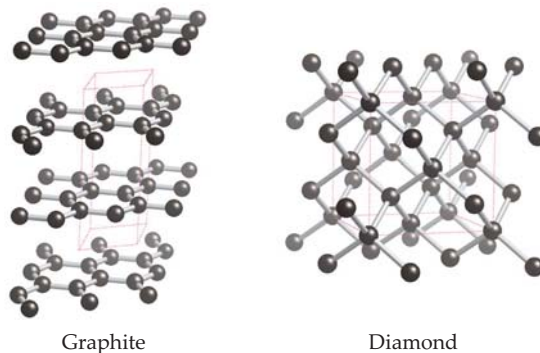
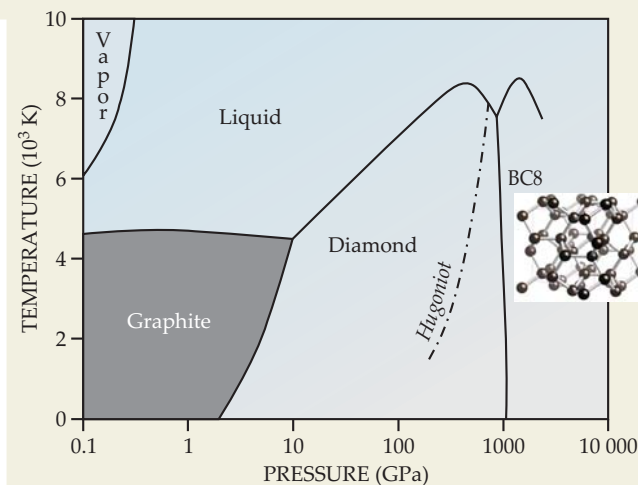
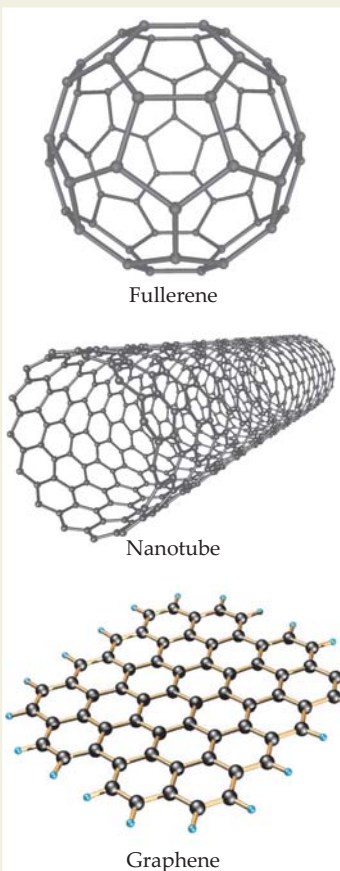
Pressure, temperature, and strain rate are variables that can be independently controlled to explore and manipulate matter. Pressures in the multi-megabar range and temperatures in the thousands of kelvin are now accessible in the laboratory using both static and dynamic compression methods. Thanks to changes that occur in the distribution and dispersion of electronic energy levels at high pressures, otherwise incompatible elements combine to form new alloys and compounds (see the article by Russell Hemley and Neil Ashcroft in *PHYSICS TODAY*, August 1998, page 26). Furthermore, compression is often associated with unexpected properties; the highest-temperature superconductivity among compounds ($T_c = 166$ K for $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$) and among elements ($T_c = 25$ K for calcium) emerges under pressure, as does an enhancement in the critical temperature of the recently discovered iron-based superconductors.⁹

The application of pressure in simple oxides gives rise to entirely new classes of ferroelectrics with colossal dielectric properties that may allow the materials to capture, store, and transmit strain energy.¹⁰ New superhard materials have been made, including osmium boride, doped diamond, and cubic carbon- and boron-nitrides.¹¹ Amorphous metals, including bulk metallic glasses, offer the high hardness and superelastic response that are important for next-generation protective coatings. Materials for vibrational damping, strength reinforcement, and hydrogen storage are being produced under extreme thermomechanical conditions, including high strain

Box 1. Carbon polymorphism

Thanks to their unique structures and varied dimensionalities, the polymorphs of carbon are ideal materials for many extreme environments. Fullerene (carbon-60), graphene, and carbon nanotubes are examples of low-pressure—indeed, thermodynamically metastable—structures whose unique electronic and mechanical properties arise from the strong carbon–carbon bonds, low dimensionality, and electronic structure of single layers of graphite. Diamond exhibits extreme properties, including ultrahigh strength, unmatched thermal conductivity, ultrahigh melting temperature, radiation hardness, high magnetic-field compatibility, low friction and adhesion, biocompatibility, and chemical inertness. It is also unique electronically: Diamond is a wide-gap insulator, but once electrons are excited above the gap, they exhibit the highest mobility of any known material. Diamond can also be turned into a superconductor by heavily doping it with boron—up to several percent by weight¹⁶—and can be synthesized with enhanced toughness by introducing a dislocation microstructure not found in natural diamond. Diamond is being used in combined static and dynamic compression experiments to explore unusual states of matter in new pressure–temperature (P – T) ranges.¹⁵

Various degrees of extreme conditions are needed for the synthesis of all the phases of carbon beyond graphite. The phase diagram of carbon at high pressures and temperatures has been documented using laser- and magnetic-compression techniques. The P – T path explored by those shock compression measurements is known as the Hugoniot. Recent measurements include confirmation of the predicted negative P – T melting slope and a metallic liquid phase as well as support for the solid metallic structure known as BC8, found for the heavier group IV elements and predicted theoretically for carbon.¹⁷



rates where the evolution of point and extended defects can be tailored to produce desired properties.

Extreme chemistry

Extremely reactive environments are found in many advanced power systems such as fuel cells, nuclear reactors, and batteries. Those systems are often limited by the inability of materials to perform reliably when exposed to reactive liquids or gases. But reactivity is often enhanced not just by the nature of the chemical species but by elevated temperature, pressure, an applied or inherent electrical potential, and even spatial confinement (see box 2). For example, electrochemical processes affect high-field conduction of ions across an oxide film at the interface of a metal with an aqueous solution.

In electrical energy storage devices, interfaces are even more complex, with charge and mass transport between electrode materials resulting in structural changes and phase transitions that ultimately limit the energy density, charge-discharge rate, lifetime, and safety of the devices. A basic

knowledge of the energetics and reaction dynamics of interfaces under extreme chemical conditions is key to developing stable and even protective interfaces between a material and its environment. The nature of that reactivity is also affected by the morphology and structure of the interface and any defects present there. The roles of these features are poorly understood.

The development of robust interfaces is essential not only for the enhanced performance of energy technologies but also for the discovery of new forms of chemical behavior, new synthesis approaches, and materials with superior properties such as ultrahardness, corrosion resistance, and high strength. A principal challenge is to understand the chemical and physical processes at interfaces in order to create stable structures, protective surface layers, and functionality such as selective chemical and catalytic reactivity and the controlled deposition of new phases.

Self-healing protective surfaces have been demonstrated to extend the lifetime of materials. However, the protective

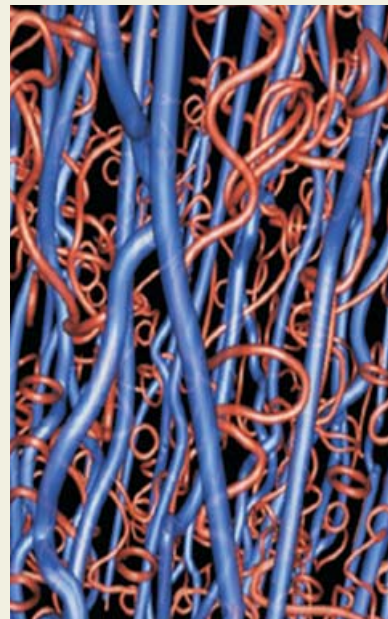
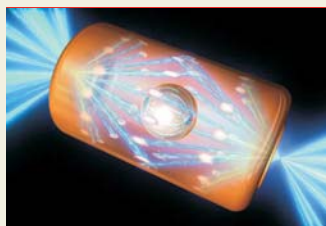
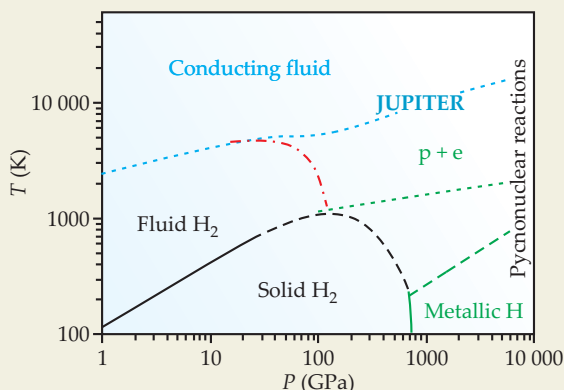
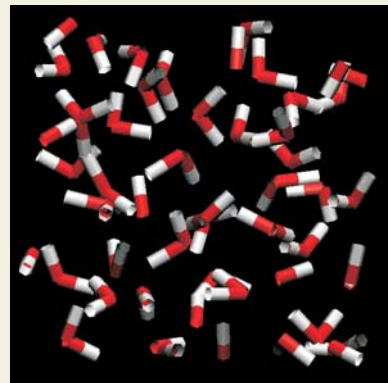
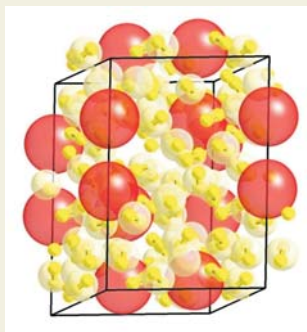
Box 2. Element one at the extremes

Hydrogen and hydrogen-bearing materials are on the frontier of extreme-conditions research for their extreme chemical reactivity, high chemical energy density, and novel electronic and magnetic properties. The high reactivity in hydrogen-rich materials is leading both to the creation of entirely new classes of hydrogen-storage materials—for example, the novel Xe-H₂ compound pictured at top left (adapted from M. Somayazulu et al., *Nature Chemistry*, in press)—as well as to unwanted corrosion and hydriding of materials due to the high proton mobility as in hot dense water, illustrated by the molecular-dynamics snapshot at top right (adapted from N. Goldman et al., *Physical Review Letters* **94**, 217801, 2005).

The phase diagram indicates the fields of stability of known and predicted forms of pure hydrogen below and above 100 GPa. The transition to the metallic state is predicted to occur in dense solid H₂ except at high temperatures, at which a dense plasma of protons (p) and electrons (e) forms. Jupiter's deep interior is close to this regime. At high pressures, pure hydrogen is predicted to transform into a quantum metal that would be a unique form of matter—at low temperature a state that is a superfluid and a superconductor. The image at bottom right shows a snapshot of the magnetic state, represented by electronic (blue) and protonic (red) vortices, in the interior of that predicted superconducting superfluid (see E. Babaev, A. Sudbø, and N. W. Ashcroft, *Physical Review Letters* **95**, 105301, 2005).

At very high pressures, pycnonuclear, or density-induced, reactions occur. In pursuit of inertial confinement fusion, researchers have turned to large laser platforms to explore the behavior of hydrogen over a broad range of extreme conditions. The lower left image shows a roughly 2-mm-diameter ICF target pellet inside a gold and uranium hohlraum

capsule designed for the National Ignition Facility; NIF's laser beams penetrate the capsule through openings on either end and impinge on the hohlraum material to create a plasma that compresses and heats the target sufficiently to trigger nuclear fusion. The success of both ICF and magnetic confinement fusion requires detailed understanding and control of the component materials in the devices.⁷



films can break down due to chemical, mechanical, and other factors. Understanding the interfacial processes behind the formation and degradation of those films will lead to the development of new materials that provide long-lived stability under extreme conditions.

Familiar concepts of chemical bonding and chemical affinities become blurred in extreme environments. Emerging experimental and theoretical methods are providing some insight into behavior in extreme conditions and allowing researchers to explore, and ultimately define, fresh approaches to the creation of materials with novel physical and chemical properties.¹² Conventional chemical rules fail at extreme pressure, where basic concepts like *s*, *p*, *d*, and *f* electrons are no longer useful. New concepts of bonding and reactivity need to be formulated, informed by experimental and computational

input on atomic and electronic structures under extreme conditions. The challenge prompts several questions. Can synthesis techniques be extended to make materials with extraordinary properties? Are there new routes by which materials can be produced and stabilized without resorting to extreme conditions? What is the nature of materials at temperatures above 10⁴ K (the eV range) and pressures in the TPa range before the identities of the component atoms disappear into a plasma? How do defects form and move under complex stress states?

Electromagnetic fields

Our ability to meet burgeoning electricity demands is ultimately tied to our ability to harness high electric and magnetic fields. Electricity accounts for about 40% of the primary energy consumed in the US, and the demand is projected to

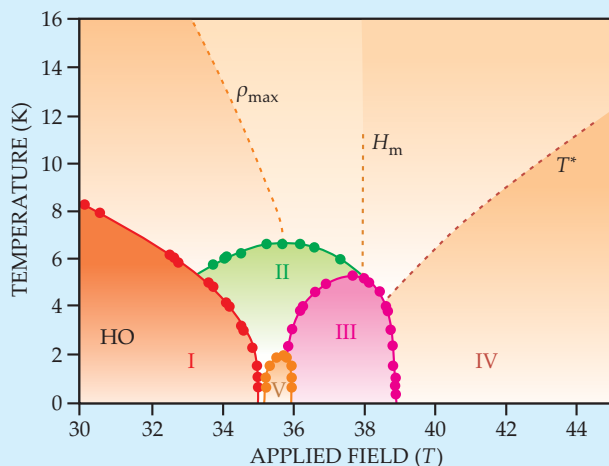


Figure 2. Magnetic phase diagram of the heavy-electron compound URu_2Si_2 . The dramatic effect of high magnetic fields on electronic structure is reminiscent of the wealth of new crystalline structures induced by high pressure. The single phase at room temperature and zero magnetic field gives way to a proliferation of exotic electronic states at high fields and low temperature. Despite a large entropy change on entering the “hidden order” (HO) phase, the order parameter of the phase remains elusive. With increasing magnetic field, the HO phase gives way to a rich variety of strongly correlated electronic behavior. For example, the material exhibits a maximum in resistivity (ρ_{max}), the onset of magnetic order (H_m) and electron–electron scattering (T^*), electronic restructuring of the Fermi surface (phases II and III), and an inferred quantum critical point in phase V. Conventional normal metallic behavior is evident in phase IV. The subtle energy differences among the strongly correlated electronic states are rearranged by the magnetic field to produce the complex phase diagram. (Image adapted from K. H. Kim et al., *Phys. Rev. Lett.* **93**, 206402, 2004; see also J. Janik et al., *J. Phys.: Condens. Matter* **21**, 192202, 2009.)

increase 50% by 2030; meeting this demand will require new approaches to energy production, transmission, and use. The efficiency of long-distance transmission of electric power is increased by transforming power to higher voltages and lower currents. The highest voltage now used in the US is 765 kV. Even higher efficiency could be achieved with higher voltage. Efficient long distance electricity transmission will be required to increase renewable generation of electricity with the sun and wind, sources of which are often located far from high population demand centers.

A key problem to address is electrical breakdown. Although breakdown itself is a dramatically sudden event, its precursors are remarkably slow. The seeds of breakdown are tiny structural defects in the dielectric that slowly collect charge under the influence of the electric field. After an incubation time that is often decades long, the charged defects become spaced closely enough to trigger spontaneous discharge, in which trapped electrons find a continuous path from defect to defect through the entire dielectric in a single discharge that lasts a few picoseconds. Tantalizingly, the ultrafast breakdown event remains just beyond current limits of experimental observation, though new fourth-generation

light sources will have the resolution time to capture it.

The magnetic field at Earth’s surface, roughly 30 to 60 μT (0.3 to 0.6 gauss), is tiny compared with steady fields of up to 35 T achievable in the laboratory with resistive solenoids and 45 T with resistive-superconducting hybrid magnets. With such large laboratory fields, researchers are on the verge of being able to reveal and control the electronic energy levels of correlated electron systems by raising and lowering orbital and spin energies, by exerting torque on magnetic moments, and by producing novel magnetically ordered states. The richness of correlated-electron behavior in magnetic fields rivals that of chemical bonds under pressure: Both exhibit a host of phase transitions to exotic states. An example is URu_2Si_2 , a heavy-electron metal that exhibits a wealth of correlated-electron phase transitions clustered around a quantum critical point at about 35 T, as shown in figure 2.

The most powerful permanent magnet is $\text{Nd}_2\text{Fe}_{14}\text{B}$, which produces at its surface a field of 1.6 T, small compared with the field of solenoids. There is no fundamental reason, however, why permanent magnets with magnetic fields at least an order of magnitude larger cannot be made. Their discovery and design are likely to involve complex structures like that of $\text{Nd}_2\text{Fe}_{14}\text{B}$, which integrates the two essential features of high-field permanent magnets: a high density of atoms with strong magnetic dipoles, such as iron or nickel, in a host structure with sufficient orbital anisotropy to force the dipoles to point in a common direction.

The production of high magnetic fields in electromagnets is limited by the strength of materials; they must withstand the magnetic pressure required to create a high energy density in magnetic fields. Fields up to 20 T can be produced in superconducting magnets, and the prospect of 30 T is within reach with the use of high-temperature superconductors. Further advances may soon produce fields that approach the limits imposed by the strength of known materials—about 50 T for the materials with the highest strength (roughly 1 GPa) now in use.

Nanoscience has given us far stronger materials, such as carbon nanotubes (see box 1), whose strength-to-mass ratio is 360 times that of steel. The existence of such strong materials suggests that much higher magnetic fields could be produced if only we could figure out how to translate the strength achievable on the nanoscale to the macroscale. Indeed, apart from the practical restriction of materials strength, there is no known theoretical limit to magnetic fields; neutron stars produce fields of 10^6 to 10^8 T.

High electric and magnetic fields offer opportunities for researchers to control the synthesis and properties of new materials. For example, electric fields of a few volts per micrometer switch electro-optic materials between transparency and opacity by controlling the orientation of liquid crystal molecules. And large magnetic fields of 30 T can dramatically alter the microstructure, phase composition, and effective solubility of C and Ni in steels.¹³ Processing in high electromagnetic fields represents an unexplored methodology for synthesis of new materials with tailored functionality.

Extremes in combination

The behavior of materials in multiple extreme environments presents additional complexity but also leads to new physics. Intriguing and diverse examples are found in the case of Si (see figure 3). The exposure of Si to a plasma generated by a femtosecond laser in a sulfur hexafluoride environment produces conical structures with a highly nonequilibrium phase near the surface. The surface is rich in sulfur (at more than 1 atomic percent), and the chemical change there creates mate-

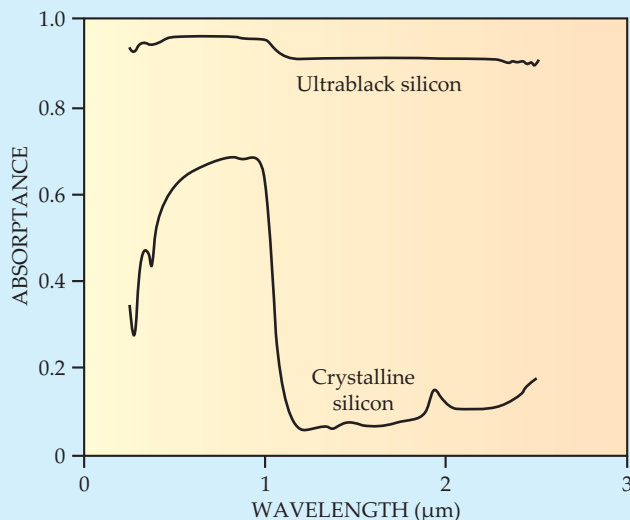
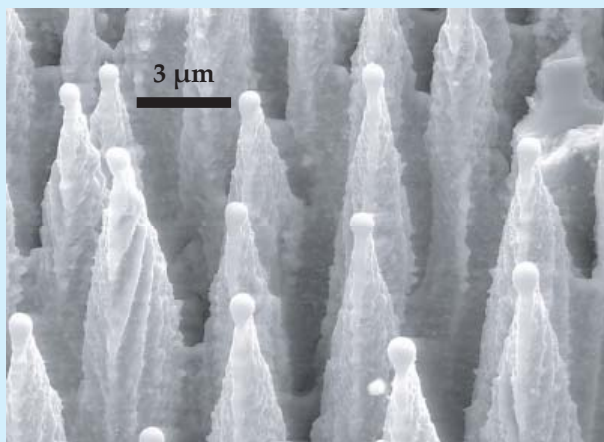


Figure 3. Silicon exposed to a plasma generated by a femtosecond laser in a chalcogen-rich environment produces ultrablack silicon,¹³ a material whose surface becomes restructured into conical spikes (left), each topped with a spherical ball. The optoelectronic properties of ultrablack silicon (right), in particular its response to IR wavelengths, make it an exceptional light-harvesting material. (Adapted from M. A. Sheehy et al., *Mater. Sci. Eng., B* **137**, 289, 2007.)

rial called ultrablack silicon that is remarkably absorbing over nearly the full optical spectrum—even at IR wavelengths where pure silicon is transparent. Such surfaces have promise for application in photovoltaic devices.

A completely different example of the effect of multiple extreme environments is the coupling of chemical reactivity and mechanical properties. In Si, for example, simulations reveal that stresses can be blunted at propagating crack tips.¹⁴ And the combination of x-ray flux and high pressure can be used to form novel metastable molecules and high-energy materials (see the article by Wendy Mao, Carolyn Koh, and Dendy Sloan in *PHYSICS TODAY*, October 2007, page 42). High chemical reactivity induced by electric fields or laser excitation has the potential to become an entirely new method of synthesis by driving materials into new states of matter.¹⁵

Studies of materials in extreme environments are essential for meeting a range of energy challenges—from creating better turbines and reactors, to improving energy storage in batteries, to developing future energy systems in dense plasmas. The problems are particularly difficult because of gaps in our understanding of multiscale phenomena—how the creation of atomic defects gives rise to nanoscale and ultimately macroscale effects. The design and synthesis of more robust materials require characterization on the scale of fundamental interactions—for example, in situ measurements of the motion of atoms and defects in materials exposed to extreme environments.

Accurate predictions and modeling of materials' performance require advanced computational techniques. Because extreme environments induce energy changes that approach or exceed that of the chemical bonds and electronic states of the component materials, development of new experimental, theoretical, and computational tools will allow us to understand a rich area of fundamental science that challenges conventional concepts of condensed matter. Indeed, new physics and chemistry are already emerging from combined experimental and theoretical investigations of materials under extreme conditions.

We are grateful to the panel chairs and participants of the workshop for their input.

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