

The growth of novel silicon materials

Epitaxial growth of germanium-silicon alloys or metal silicides on silicon produces structures that can be used in high-performance optical and electronic devices.

John C. Bean

In the industrialized world, we are seldom more than a few meters from a crystal of silicon. One crystal may be strapped to our wrist, in an electronic watch; others may be buried within a nearby calculator, video recorder or auto ignition system—indeed, one may even be lurking in the quartz movement of a new “antique” clock. These crystals are extraordinarily perfect: No more than one atom in 10^{13} is out of a proper lattice site, and the total impurity concentration may be less than 0.1 parts per billion. Yet a microprocessor or a megabit memory can be fabricated in a crystal of silicon costing only 15–20 cents.

The techniques to purify and grow such silicon crystals were developed in the 1950s and the material was well characterized by the late 1960s, making it accessible for application to technology. The same periods saw the development of designs for the bipolar and metal-oxide-semiconductor devices that still serve as the basis of integrated-circuit technology. The en-

tering vigorous growth of the technology—for all of the resourcefulness involved—has been the result primarily of finding ways to reduce the size of the devices and increase their connectivity. This scaling down has become much more costly as device dimensions drop below the wavelength of visible light. Furthermore, as the characteristic dimensions of the devices reach $\frac{1}{10}$ micron, we may encounter fundamental quantum-mechanical limits on their performance.

Opening new options

Entirely new options would be opened if we could provide the silicon-based technology with a vastly expanded array of materials properties. Ideally, this would be accomplished by combining many materials, in arbitrary geometries, on a silicon-based circuit. It would be necessary, however, in juxtaposing the various materials to preserve the rigorous crystalline structure that gives a semiconductor its outstanding characteristics. To completely avoid defects due to the interfaces, one must limit the components to materials with similar geometric arrangements and nearly identical atomic spacings—that is, one must use

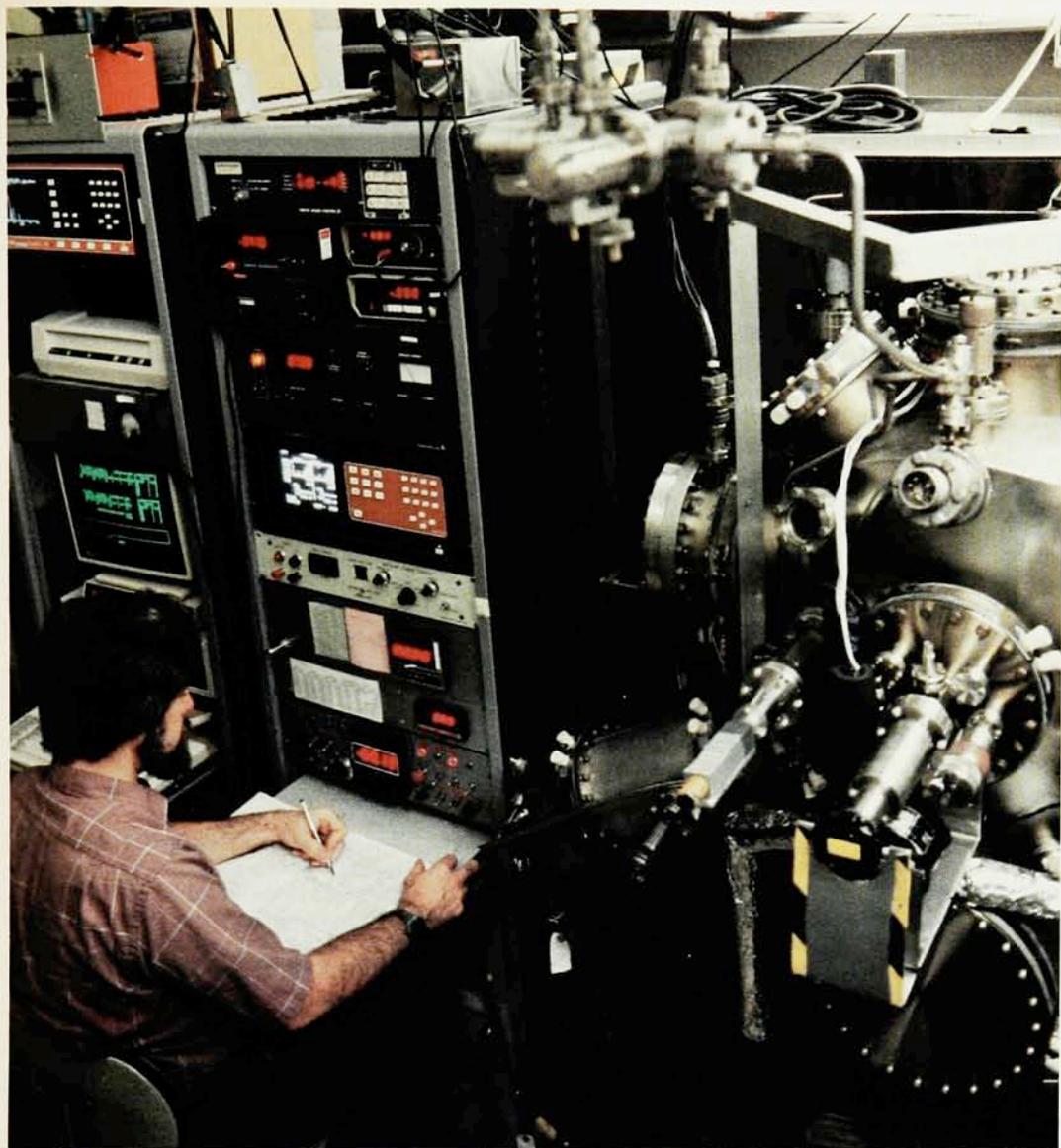
materials with matched lattice structures and lattice constants. Figure 1 shows a molecular-beam epitaxy system used at AT&T Bell Laboratories to grow such structures.

As figure 2 shows, silicon has an unusually small lattice constant, which is closely matched by only three other common semiconductors: GaP, AlP and ZnS. Unfortunately, these materials are composed of the elements most effective at generating free carriers in silicon, which has led to an uncontrolled crossdoping effect in all experiments to date.

To many scientists, these limitations have suggested that we should move to the family of semiconductors composed of elements from columns III and V of the periodic table. These materials do not dope one another and, as figure 2 shows, the compounds and their alloys have a wide range of lattice constants, providing many opportunities for single-crystal overgrowth, or “epitaxy.” These semiconductor heterostructures have already been applied in a number of new and powerful device configurations (as Lester Eastman discusses on page 77 of this issue).

I will describe an alternative approach based on the continued use of

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Apparatus for silicon molecular-beam epitaxy built at AT&T Bell Laboratories. This machine can be used to grow materials such as $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ or metal silicides as epilayers on silicon, with lattice structures that are continuous across the interface. The resulting heterostructures often have surprising properties. Figure 1

silicon compounds (or other chemically compatible materials) coupled with means to circumvent or isolate the difficulties presented by imperfectly matched crystalline overgrowth. For the technologist this approach has the powerful attraction of retaining both the economics of silicon and the vast automated-processing knowhow that depends largely on silicon's unique chemical and mechanical properties. One might think that this apparently conservative strategy would offer little meat for the basic scientist. Quite the opposite. Silicon has a highly perfect, covalently bonded structure that is unusually amenable to analysis and modeling on the most basic levels. Indeed, for years silicon has been the prototype system in both surface physics and many areas of theoretical band-structure physics. The inherently tractable nature of silicon-based materials thus provides a unique opportuni-

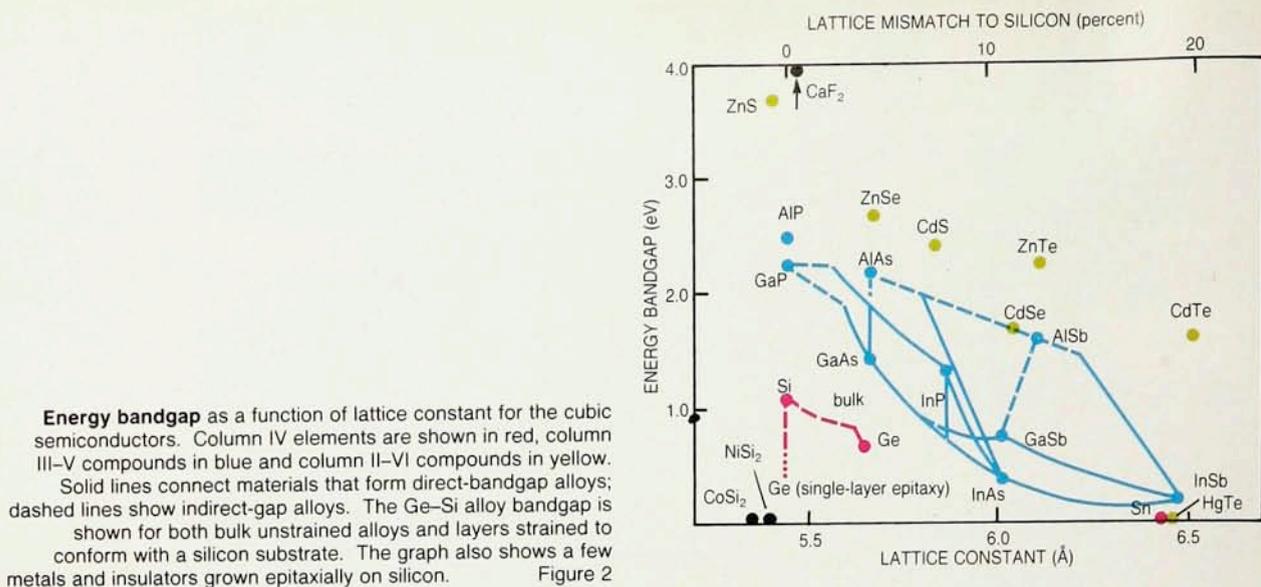
ty for interaction between basic and applied scientists, and it is this interaction that gives the field much of its vitality.

Of the materials systems currently under study, I will highlight two that give some flavor of the research: the GeSi/Si system, in which lattice mismatch is overcome by the tendency of thin layers to deform mechanically into precise atomic registration, and the metal silicides (actually, near-noble metal disilicides), in which defects can be largely confined to interfaces. These two kinds of materials, in which alternating semiconductor and metal layers are added to the silicon system in a single-crystal form, may permit both arbitrary stacking of materials and three-dimensional integration. More importantly, these systems provide two examples of the successful integration of materials with lattice mismatches of several percent. Sever-

al hundred electronic materials have lattice structures with a comparable registry to silicon.

Semiconductor heterostructures

There are several reasons for wanting to combine silicon with another semiconductor. For one, on its electronic merits alone, silicon is in fact a mediocre material. It has only average high-field breakdown characteristics, free-carrier mobilities and carrier saturation velocities. Its high minority-carrier lifetimes are offset by its indirect bandgap, which precludes its use in light-emitting diodes or laser structures and handicaps it as a photodetector. It is used in integrated circuits more because of its superb oxide (see the article by Frank J. Feigl on page 47), its unique etching properties and its phenomenal mechanical strength. One can therefore hope to compensate for silicon's electronic deficiencies by



combining it with another, more distinguished, semiconductor.

Even without this bootstrapping effect, the epitaxial combination of any two semiconductors makes possible devices based on the abrupt change of properties at the interface. For instance, a sudden change in bandgap produces discontinuities in the valence- and conduction-band edges that can be used to trap free carriers and segregate them from the parent ionized donor or acceptor impurity atoms, as illustrated in figure 3; the result is to diminish the scattering of carriers by electric fields and to increase the carrier velocities. This effect is exploited in the modulation-doped field-effect transistor. If two semiconductors are layered on a fine enough scale, the artificial periodicity (or superlattice) produces fundamental and often desirable changes in the band structure of the composite material.

Of the semiconductors shown in figure 2, all but germanium are composed of materials that will produce doping in silicon. And while germanium shares silicon's crystalline structure, its atomic spacing is about 4% larger. This suggests that at an interface between Ge and Si, every 25th row of Si atoms will have only three bonding neighbors, as shown in the crystal structure at the upper right of figure 4. The remaining "dangling" bond can serve as a site for carrier generation or recombination, seriously degrading the electronic properties of the interface.

This difficulty has recently been overcome by the use^{1,2} of molecular-beam epitaxy, a technique for growing overlayers on crystals by the condensation of low-vapor-pressure molecules or atoms from a molecular beam in a high vacuum. The low vapor pressures of the materials allow one to block the beam with a room-temperature shutter

in the vacuum chamber, providing nearly instantaneous control of the deposition fluxes. By using simple molecules or atoms one can largely avoid the need for decomposing the molecules at the surface, allowing one to lower the temperature of the substrate to the point where atomic arrangements are frozen in place in all but the surface planes of the growing crystal.

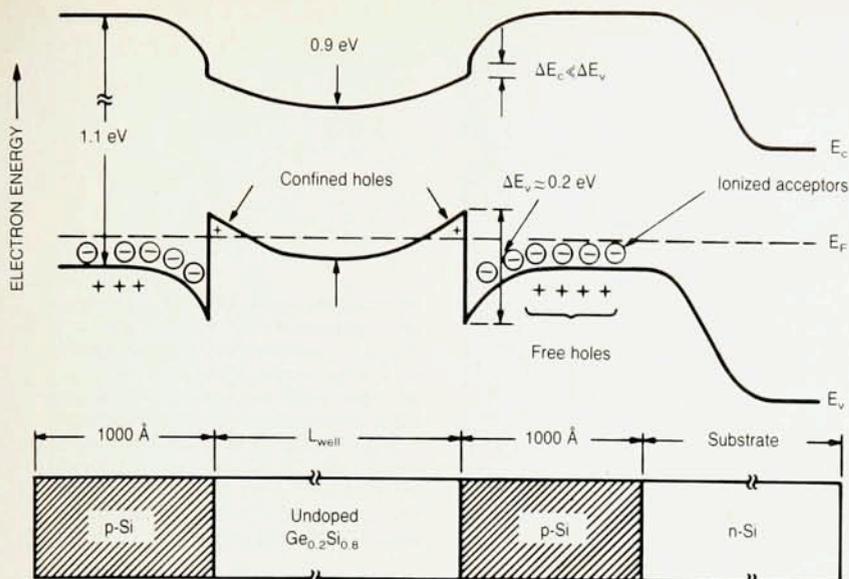
One can in this way grow defect-free $\text{Ge}_x\text{Si}_{1-x}$ alloys on Si. As long as the layer of alloy is thin enough, the first few atomic planes of $\text{Ge}_x\text{Si}_{1-x}$ will readily compress in the growth plane to match the smaller lattice spacing of the Si substrate crystal, as in the crystal structure shown at bottom left in figure 4. For thin layers, the strain energy is more than offset by the absence of dangling-bond energies, making this the lowest-energy configuration. As the thickness of the alloy layer increases, of course, the saving in dangling-bond energy is overwhelmed by the strain energy in the accumulating alloy layers; as a result, the alloy eventually reorders to its undistorted arrangement, forming arrays of interfacial dangling bonds, as in the structure shown at upper right in figure 4. At equilibrium, this critical reordering thickness decreases from about 25 to 3 atomic layers as the Ge fraction in the alloy increases from 0.2 to 1.0. Fortunately, the reordering process involves gross atomic rearrangement—which, in effect, provides an energy barrier that cannot be surmounted at the temperatures involved in molecular-beam epitaxy. As a result, $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ "strained-layer epitaxy" can be maintained to the thicknesses graphed in figure 4, which are much larger than those for other epitaxial growth techniques—large enough to be of use in electronic devices.

The volume of strained $\text{Ge}_x\text{Si}_{1-x}$ can be further extended by inserting layers of Si between layers of alloy to form a superlattice structure such as that shown in the cross-sectional transmission electron micrographs of figure 5. This particular superlattice contains layers mismatched by almost 2%. Defects are avoided by a strain in the $\text{Ge}_x\text{Si}_{1-x}$ layers that can actually be discerned in the detailed micrograph as a slight bend in the atomic rows at the Si-alloy interfaces. In this superlattice, the total thickness (1500 Å) of strained $\text{Ge}_{0.40}\text{Si}_{0.60}$ is approximately 50 times the equilibrium value for a single layer of this composition on Si.

From the material to devices

One might have expected the strained-layer lattices to move from the materials-science lab, where they were invented, to the physical scientists for characterization, and then on in a straight line to the device engineers for use in electronic components. They did, several times: In applications of truly novel materials, we find that the device engineer, by invalidating inappropriate models, often motivates basic research rather than just consuming its product. Finely dimensioned structures such as strained-layer superlattices contain configurations of forces and possess properties that readily defy both intuition and conventional physical measurement. For this reason, the most sensitive basic characterization of the material is frequently provided not by standard physical techniques, but by measuring the performance of devices whose designs are based on the band structures and interfacial properties assumed for the material.

This iterative process produced³ several surprises in the $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ system. First, $\text{Ge}_x\text{Si}_{1-x}$ optical detectors absorbed light at a longer wavelength



Modulation-doped $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ heterostructure. This schematic cross section shows the sequence of layers structure (below) and the corresponding band structure (above). A large discontinuity in the edge of the valence band at the alloy-silicon boundary can confine holes generated by dopant atoms in the undoped Si layers, reducing the scattering of the holes by the ionized acceptor atoms and increasing the hole mobility. Figure 3

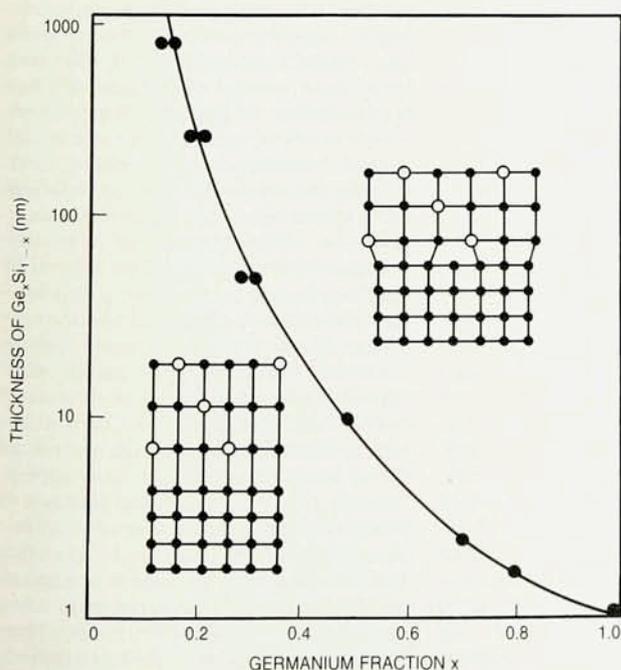
than was expected from earlier work on bulk Ge-Si alloys; next, the modulation-doping results were not only unpredictable but contradictory.

In our early Bell Labs experiments we grew $\text{Ge}_x\text{Si}_{1-x}$ on Si, doped one of the two layers to make it n- or p-type, and looked for the transfer of free electrons or holes between adjacent layers. Measurements of the low-temperature Hall effect and of the Shubnikov-de Haas magnetoresistance indicated that the only strong transfer was for holes from p-silicon to undoped $\text{Ge}_x\text{Si}_{1-x}$. This result suggests a band alignment like that shown at the top of figure 3, where the alloy bandgap falls within the silicon bandgap and most of the difference in bandgap appears as a discontinuity of the valence-band edge.

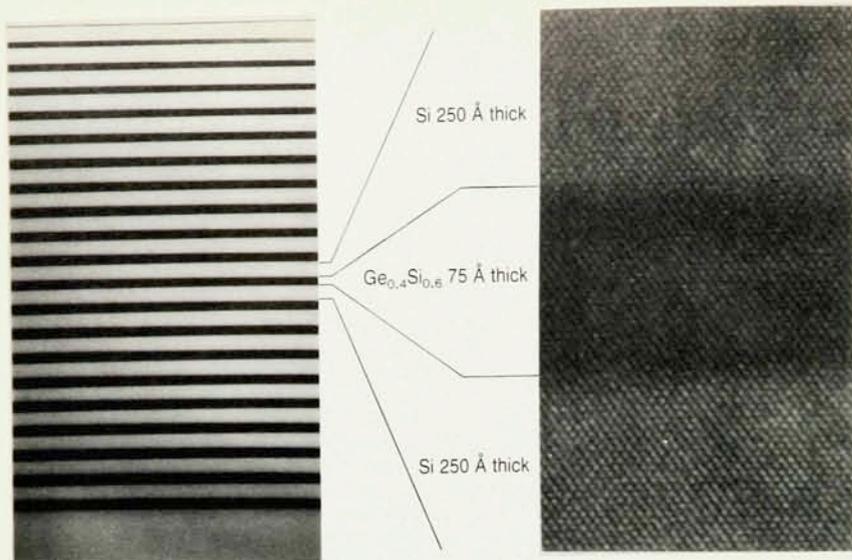
Subsequent experiments at AEG in Ulm, West Germany, and the Munich Technical University were equally unequivocal in showing a strong transfer of electrons from n-doped $\text{Ge}_x\text{Si}_{1-x}$ to undoped silicon. This suggested a staggered band alignment, with the edge of the alloy conduction band above that of silicon.

A reexamination of the band-structure physics revealed that both results were correct and also explained the anomalous optical absorption. The key finding was that the crystals in these layers are so strained that their band structure is not just perturbed, it is grossly altered. Working from earlier derivations of strain Hamiltonians and deformation potentials, one can show that the bandgaps of $\text{Ge}_x\text{Si}_{1-x}$ and Si

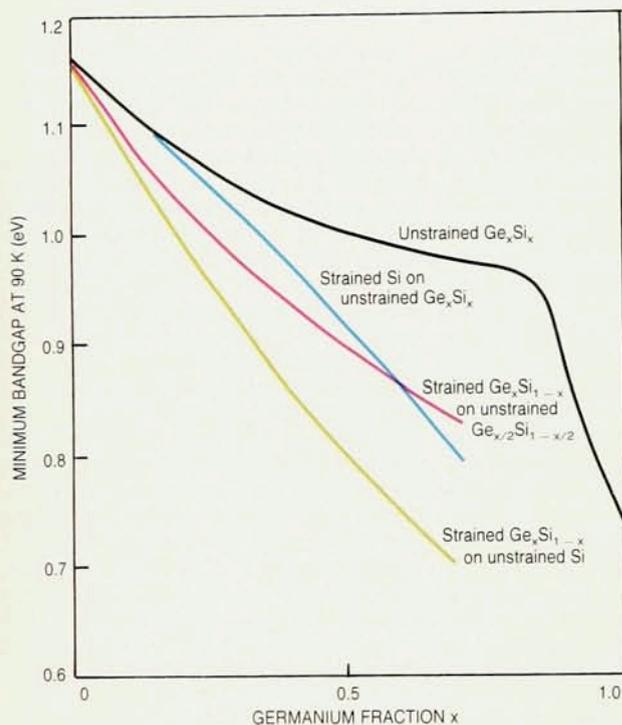
depend strongly on the relative allocation of strain; figure 6 shows the calculated results for the minimum bandgaps of several materials as a function of the atomic fraction of germanium. In fact, as a result of the strong strain dependence, the bandgap of $\text{Ge}_x\text{Si}_{1-x}$ strained to conform with a silicon substrate falls below that of pure bulk Ge for x above 0.6, which explains the anomalous photodetector results. When one combines these results with an interpolation of pseudopotential calculations (performed by Richard Wright and Chris van de Walle at the Xerox Palo Alto Research Center) for the Ge/Si valence-band discontinuity, one also finds that strain affects the shift of the valence-band edge at the Si- $\text{Ge}_x\text{Si}_{1-x}$ interface, changing



Accommodation of strain in epitaxial growth. The curve shows the experimentally determined limits for defect-free strained-layer epitaxy of $\text{Ge}_x\text{Si}_{1-x}$ on Si. The insets show the atomic arrangements for the strained but defect-free material found in sufficiently thin layers and for the bulk material in which misfit dislocations accommodate the lattice mismatch. The diagrams greatly exaggerate the degree of lattice mismatch—which is about 4.2% for pure Ge on Si. Figure 4



Strained-layer superlattice consisting of alternating layers of $\text{Ge}_x\text{Si}_{1-x}$ and Si. These transmission electron micrographs of a cross section show both the arrangement of the superlattice (left) and the atomic arrangement of the layers (right). The silicon layers (light bands) are 250 Å thick; the alloy layers (dark bands) are 75 Å thick. The detailed micrograph shows clearly the absence of crystal dislocations at the interfaces. Figure 5



Minimum bandgap of strained $\text{Ge}_x\text{Si}_{1-x}$ and Si illustrates the pronounced effects of strain. The curves show the bandgap as a function of the fraction of Ge in the alloy for unstrained alloy, alloy strained to conform with silicon, and silicon strained to conform with alloy. Figure 6

it from -20 to $+150$ meV; this shift can account for the discrepancy in the modulation-doping data.

Strained $\text{Ge}_x\text{Si}_{1-x}$ thus provides semiconductor technology with several new degrees of freedom: a variable bandgap adjusted by either Ge content or strain partition, and an adjustable alignment of band discontinuities between layers. Consider the possible implications of varying just one of these parameters in a single application, the fiberoptic detector. Glass fibers have two low-attenuation win-

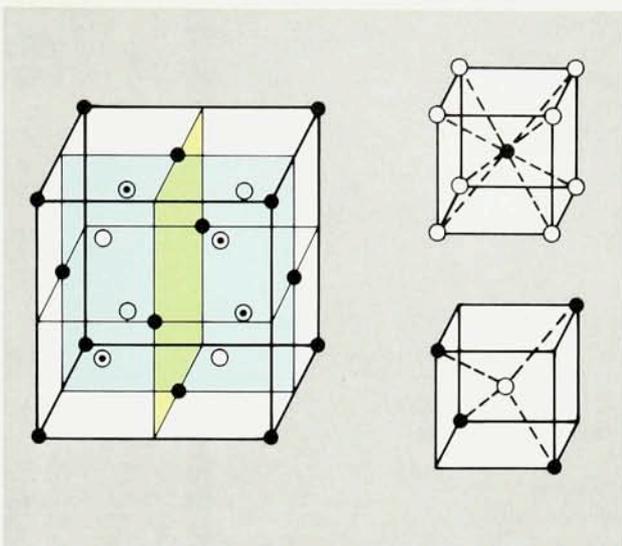
dows at wavelengths of about 1.3 and 1.5 microns (corresponding to bandgaps of 0.95 and 0.8 eV). The bandgaps of strained layers of $\text{Ge}_x\text{Si}_{1-x}$ fall within this range, making it feasible to fabricate fiberoptic detectors with silicon technology. The range of bandgaps, of course, corresponds to a range of indices of refraction for the layers. This opens the possibility of using the variation in refractive index to produce heterostructures that have the long optical path necessary to achieve strong absorption in the indirect band-

gap material (Si), while retaining a short photocarrier collection length in the neighboring layers of the direct-gap alloy. The result is a fast and efficient detector. It is generally combined with an avalanche multiplication layer of silicon, which provides internal gain and exploits one area in which silicon surpasses other semiconductors: the selective multiplication of only one carrier species.

Such devices have been fabricated and have achieved⁴ a gain-bandwidth product of over 48 GHz and detected signals of 800 megabits/sec over a 45-km fiberoptic link with negligible error rates. These figures of merit make the strained-layer devices possible contenders for shorter telecommunications links, where millions of detectors at minimal prices will be required. But the possibilities don't stop there. As silicon technologists contemplate integrated circuits covering an entire wafer and containing millions of elements, they face serious problems of data transmission within the chip. An obvious solution would be to use multiplexed high-bandwidth optical links. The availability of a fast and efficient silicon-based detector takes this scenario an important step closer to reality.

Other degrees of freedom offered by strained layers of $\text{Ge}_x\text{Si}_{1-x}$ can be—and have been—exploited in other devices. Many device concepts can be borrowed wholesale from earlier work on III-V semiconductor heterostructures. But in addition, we find that the harder we look at these highly strained structures the more exotic they appear. One of the most exciting findings is that strain induces a regular atomic ordering of the Ge and Si atoms within the alloy layers. This was first suggested by structural measurements using electron and x-ray diffraction. More recently, an analysis of Shubnikov-de Haas data indicated the existence of a

Crystal structure of NiSi_2 and CoSi_2 . The black spots indicate atoms of Ni or Co; Si atoms are shown in white. As shown at right, each Si atom is bonded tetrahedrally to four metal atoms; each metal atom is bonded to the eight silicon atoms that surround it, forming the corners of a cube. The lattice has the same structure as CaF_2 ; removing from the lattice the four sites marked with dots makes the lattice identical with that of silicon. Figure 7



piezoelectric power-loss mechanism. Such a loss mechanism requires a preferred orientation of polar bonds, which in turn suggests that there is an ordered structure. An atomic reordering could change the basic symmetry of the lattice, allowing a variety of additional optical and electronic properties not allowed in the bulk crystal because of its symmetry.

Metallic heterostructures

Epitaxial metal-silicon heterostructures also offer challenges and opportunities for the basic scientist; on the technological side they are of interest as replacements for the polycrystalline metallic interconnect "wiring" now used in integrated circuits. Considerations of power loss and transmission speed are driving integrated-circuit designs to lower operating voltages, where interconnect resistivity and uniformity become increasingly important. If the metal line has a width comparable to its typical grain size, neither the averaging effect of an ensemble of grains nor the perfection of a single crystal is available to ensure uniformity. The boundaries between grains also offer rapid and unpredictable paths for impurity diffusion. Furthermore, the processing of the wafers is severely constrained if the metallization cannot withstand the same temperature as silicon without undergoing degradation or interaction. The temperature instability effectively rules out pure metals in favor of materials that have already reacted with silicon to form a stable compound, that is, the metal silicides.

With these technological rationales securely supporting him, the basic scientist can use metal silicides to investigate the mysteries of the semiconductor-metal interface. Paramount among these mysteries is the nature of a metal-semiconductor Schottky bar-

rier. These barriers, which have been studied for decades, play an important role in silicon technology and a crucial role in III-V semiconductor devices, where the absence of a high-quality oxide rules out metal-oxide-semiconductor structures. But in spite of their importance, we still cannot provide a first-principles derivation of the barrier heights. Carrier injection and hot-carrier transport ("hot" in this context referring to a carrier with energies well above equilibrium values) are also important topics, research into which would be furthered by a well-behaved metal-semiconductor interface.

In the last five years molecular-beam epitaxy has produced⁵ high-quality single-crystal overgrowth of the metal silicides NiSi_2 and CoSi_2 on silicon. It would be stretching matters, however, to say that the resultant interfaces are well behaved. As in the $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ case, the closer one looks, the more peculiar (and interesting) these interfaces appear.

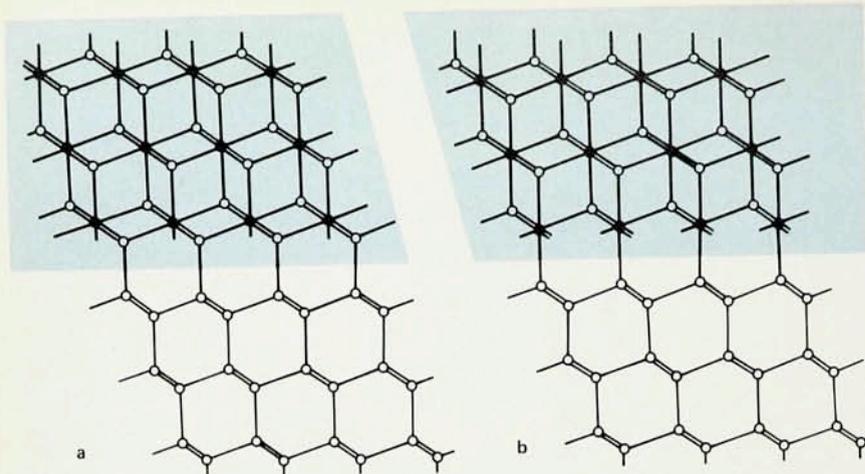
These metal silicides share the so-called calcium fluoride structure illustrated in figure 7. With the subtraction of four lattice sites (for example, the four interior sites, arranged in a tetrahedron, marked with black dots in the figure) this unit cell would be identical to that of silicon, and indeed the silicon atoms in the silicides retain the same tetrahedral bond geometry found in pure silicon. At room temperature, the cell dimensions for the nickel and cobalt silicides are, respectively, only 0.4% and 1.2% smaller than in silicon. Grown on silicon, these materials do not form strained layers but retain their bulk spacings; the resulting lattice mismatch is accommodated by rows of misbonded atoms at the interface, as in the structure shown at upper right in figure 4. These rows are widely spaced (80–250 atoms apart) and have a minimal effect on the properties of the

adjacent metal and semiconductor layers.

Given the similarity in crystal structures, one would expect these metal silicides to grow on silicon with a strict continuation of the lattice symmetry. However, on the (111) crystal face of silicon the metal silicide prefers to grow with a stacking sequence that produces a 180° lattice rotation. I use the word "prefer" because with a certain amount of coercion, the unrotated configuration also can be grown for NiSi_2 on Si (but not for CoSi_2 on Si). Ball-and-stick models demonstrate the similarity of the two interface configurations, but calculations have not yet shown why the rotated structure should be strongly favored.

Further complicating the structure of the interface, the plane of metal atoms just at the interface can have either five or seven bonds with neighboring silicon atoms, depending on the precise registration of layers at the interface.⁶ Figure 8 shows a two-dimensional model. Comparison of transmission electron micrographs with computer simulations suggests that for NiSi_2 , both rotated and unrotated interfaces produce sevenfold coordination. For CoSi_2 , however, the metal atoms at the (rotated) interface appear to be fivefold coordinated.

Common polycrystalline Schottky diodes display barrier heights remarkably independent of the detailed metal structure or orientation. It has been proposed that this uniformity is due to surface states and the averaging of fine-scale metal properties over the diode area, but this has not been confirmed. Because the epitaxial metal silicide-silicon interfaces are at least uniform and well defined—confusing as they are in other respects—they may help explicate the intrinsic mechanisms of barrier formation. Measurements⁷ by Ray Tung at AT&T Bell



Interface between a metal disilicide (blue) and silicon. This projection shows the alternative arrangements of atomic bonds at the boundary plane. Note that in one case the metal atom at the interface has a fivefold coordination, whereas in the other the coordination is sevenfold. Figure 8

Labs, confirmed⁸ by Robert Hauenstein and his colleagues at Caltech and General Electric Research, do indeed show systematic variations of barrier height with interfacial structure. For NiSi₂, the rotated and unrotated interfaces with sevenfold-coordinated metal atoms have barriers of 0.79 and 0.65 eV, respectively, on n-type silicon. The fivefold-coordinated rotated CoSi₂ interface has a barrier of 0.64 eV. Although models do not as yet account for these data, they finally give us a clear case where barrier properties depend on fundamental materials parameters. This at last provides a way of discriminating between proposed theories.

The current absence of models has not prevented workers from exploiting yet another quirk of the CoSi₂-Si system. For some reason, films of highly perfect single-crystal CoSi₂ contain occasional pinholes. These openings can be as small as 10-100 nm across. Impurities do not appear to be responsible, and a simple strain-relief mechanism seems an unlikely explanation, given that pinhole spacings can be as large as a hundred times the layer thickness. When these CoSi₂ layers are overgrown by a second layer of silicon, the holes are filled with pillars of silicon that provide natural electrical channels through the silicide between the cladding silicon layers. The conductance of these channels is controlled by the depletion of carriers from the pillar boundaries, which are in contact with the metal. One can modulate the thickness of the depletion layer by applying a bias voltage to the metal layer, thus producing transistor action.^{9,10}

There is nothing new in this type of transistor action, as it forms the basis

for the so-called permeable-base transistor, which has been under study for some years. The significant point is that a conventional permeable-base transistor depends on a *tour de force* in photolithographic processing to produce microscopic openings in metal-on-semiconductor films. This potential application transforms the naturally occurring channels in CoSi₂ on silicon from an irregularity into a potentially powerful attribute. Early transistor results are encouraging, and models suggest the possibility of efficient operation at frequencies well over 1 GHz. If these predictions are borne out, this would be yet another example of a completely unanticipated property producing important technological benefits.

Opportunities

I have discussed only three of the hundreds of materials that are candidates for epitaxial overgrowth on silicon. Several patterns are nevertheless evident. The crystalline combination of two materials invariably produces physical properties not found in the component materials; these can be the result of stresses, fine dimensions or artificial periodicities. The uniformity and regularity of materials based on crystalline silicon make it unusually easy to define—and eventually model—these properties. In turn, the models available for these materials provide a rigorous framework often missing in conventional materials science and frequently lead to the discovery of further novel characteristics. The field is thus a natural ground for the strong interaction of the materials scientist and physicist.

The fact that silicon processing capa-

bilities are incredibly well developed and used in a wide variety of contexts means there are many opportunities to apply a novel property or material to devices. At its heart this technology is built on a single semiconductor with a limited set of rather unexciting electronic characteristics, constraining the technology severely. But by adding a single additional material compatible with silicon and silicon processing, one significantly extends the range of physical properties—often vastly beyond the initial expectations.

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