

threshold. A quite general theorem limits the efficiency of an FEL in a storage ring to the fractional energy acceptance of the ring—perhaps 3% in an optimally designed storage ring. The Orsay-Stanford group was gratified that its oscillator efficiency (2.4×10^{-5}) comes close to the limit set by this theorem for the less than optimal ACO ring. The use of optical klystrons also limits the energy acceptance of an FEL and thus its efficiency in a storage ring.

Tapered-wiggler lasers, it is hoped, will eventually operate with efficiencies closer to 20%. But these high-power devices are probably too disruptive for storage ring beams. Restricted to linacs, they may perhaps be limited to infrared applications, though the goal of the rf linac development program by the Math Sciences Northwest-Boeing groups is a tapered wiggler lasing in the visible. Efficiency is of course a much more important issue for the industrial and military uses one has in mind for tapered-wiggler FELs than it is for storage-ring-based visible and ultraviolet FELs. These latter are sought primarily as research tools.

Prospects. In the coming months we can expect a number of other FEL oscillators to become operational. Charles Brau and his colleagues at Los Alamos are in the process of converting its tapered-wiggler amplifier into an oscillator. Claudio Pellegrini's Brookhaven group is building a storage-ring-based uv oscillator. Don Prosnitz and coworkers at Livermore are preparing to apply an alternative linac technology—the ETA and ATA high-current induction linacs—to tapered-wiggler FELs.

Other electron-beam sources under development for infrared FELs are a microtron (a kind of mini-cyclotron) to be used by a group at Bell Labs led by Kumar Patel and Earl Shaw, and an electrostatic accelerator employed by Luis Elias and his colleagues at the University of California, Santa Barbara. The Santa Barbara group hopes to establish a user facility for condensed matter studies with its FEL providing far-infrared laser light tunable from 100 microns to 1 mm, with an average power of several kilowatts. The Orsay-Stanford group is planning to use a positron beam in place of electrons in ACO. Positrons remove one source of storage-ring beam instability—the trapping of positively charged residual gas ions by electrons.

Future plans at LURE call for the building of a "super ACO ring," optimized for free-electron lasers. A 1-GeV storage ring under construction at Stanford will house a 20-meter-long wiggler with which Madey's group hopes to achieve FEL oscillation down to 500 Å in the extreme ultraviolet. At

Frascati's storage ring, Sergio Tazzari and his colleagues are planning to operate an FEL oscillator in the visible. The gain measurements they have already performed in the amplifier mode have yielded results similar to those achieved at LURE. —BMS

References

1. J. M. Billardon, P. Elleaume, J. Ortega, C.

Bazin, M. Velghe, Y. Petroff, D. Deacon, K. Robinson, J. Madey, *Phys. Rev. Lett.* **51**, to be published (1983).

2. G. Neil, J. Edighoffer, C. Hess, T. Smith, F. Fornaca, submitted to *Phys. Rev. Lett.*
3. *Proc. Fourth FEL Workshop*, Eastsound, Wash., 1983, to be published by SPIE: see papers by J. Adamski, W. Gallagher, R. Kennedy, E. Tyson, A. Yeremian, and by D. Quimby, J. Slater.

Second heavy-fermion superconductor

The recent demonstration of superconductivity in uranium beryllide (UBe_{13}) by an ETH-Los Alamos collaboration provides us the second example of an exotic and potentially instructive new class of materials, the "heavy-fermion superconductors." Four years ago, Frank Steglich and his colleagues at Darmstadt and Cologne reported the discovery of the first of these unconventional and somewhat puzzling new superconductors— $CeCu_2Si_2$. The unearthing of UBe_{13} is important not only because it shows that $CeCu_2Si_2$ is not a unique aberration. Until recently, investigations of $CeCu_2Si_2$ have been plagued by the difficulty of fabricating stoichiometrically clean, reproducible samples of this material; different samples were giving distressingly different results. With the advent of UBe_{13} , where one does not have this metallurgical problem, and with new techniques for making crystalline $CeCu_2Si_2$ samples of high quality, one now has two sources of reliable data on the heavy-fermion superconductors.

The superconducting transitions of these two materials— $CeCu_2Si_2$ at 0.65 K and UBe_{13} at 0.85 K—come as a great surprise. The Curie-Weiss increase of their magnetic susceptibilities as they are cooled clearly indicates the strong presence of localized magnetic moments at temperatures above about 10 K. Magnetic interactions are anathema to the usual Cooper-pairing mechanism necessary for superconductivity, because the opposite spins of the two pairing electrons would perceive a magnetic perturbation with opposite signs. Furthermore, both materials exhibit enormous electronic specific heats at low temperatures, indicating the presence of delocalized electrons with effective inertial masses in the lattice two to three orders of magnitude greater than that of a free electron. One would expect such extraordinarily "heavy" electrons to condense to a magnetically ordered or charge-ordered state rather than a superconducting one.

The presence of these heavy fermions is further confirmed by the observation of unprecedentedly high values of the rate of increase of the critical magnetic

field with falling temperature at the superconducting transition. It was, in fact, this extraordinarily high value of $-dH_c/dT$ at the critical temperature T_c that led Ernst Bucher and his colleagues at Bell Labs astray in 1974. Having observed a critical-field derivative of about 300 kG/K, they concluded that the superconducting transition they were apparently seeing in UBe_{13} must be due to some filamentary impurity in the sample. Interest in UBe_{13} then subsided until its recent reawakening under the impetus of Hans Ott at the Eidgenössische Technische Hochschule in Zurich.

Cerium and uranium have in common the fact that both possess partially filled inner shells of f (orbital angular momentum of 3) electrons. Cerium is the first of the lanthanides, a sequence of elements characterized by the sequential filling of the inner 4f shell while the further-out 5s, 5p and 6s shells are already occupied with fixed configurations. Uranium is the third of the actinides, the analogous heavier series characterized by sequential filling of the 5f shell. A simpler and more familiar example of this sort of inner shell filling while the outer configuration of valence electrons remains fixed is the series of transition elements such as iron, cobalt and nickel, where the partially filled 3d shell plays a central role in ferromagnetism.

The heavy fermions apparently responsible for superconductivity in $CeCu_2Si_2$ and UBe_{13} are these f electrons. The enormously high electronic specific heats observed in both materials indicate that these f electrons move through the lattice with effective inertial masses as much as a thousand times the mass of a free electron. Unlike ordinary conduction band electrons, which move freely through the lattice with effective masses close to the free electron mass, taking almost no notice of each other, these inner-shell f electrons spend most of their time inside the electron cores of the lattice atoms. Thus they experience strong mutual repulsion and they are highly correlated with one another. It is these strong interactions between f electrons that greatly impede their

mobility and give rise to such extraordinarily high effective masses.

In fact, the *f* electrons would not be delocalized at all—they would be entirely stuck in their atomic cores—were it not for resonant hybridization near the Fermi energy with the conduction electrons of the *s* and *d* conduction bands. Chandra Varma, a theorist at Bell Labs, argues that the delocalization of the *f* electrons in materials such as CeCu_2Si_2 and UBe_{13} depends on the fortuitous fact that in these materials the *f* resonances lie very close to the Fermi level of the *s* and *d* light-electron conduction bands. The strong localization of *f*-band electrons tends to render most lanthanide and actinide materials magnetic. They have thus been regarded as unlikely places to look for superconductivity.

We understand the electronic properties of simple metals quite well. In these materials, where conduction electrons come only from the outermost atomic shells, one can calculate transport properties simply by treating the delocalized electrons as a noninteracting gas. The transition metals present greater theoretical difficulties. Their conduction electrons, coming from an inner *d* shell, are less free of the atomic cores, and hence more correlated and less itinerant. The greater the correlation between conduction electrons, the more difficult is their theoretical description. Much remains to be explained about ferromagnetism.

In this regard, Theodore Geballe (Stanford) points out, the *f*-band lanthanides and actinides should offer many

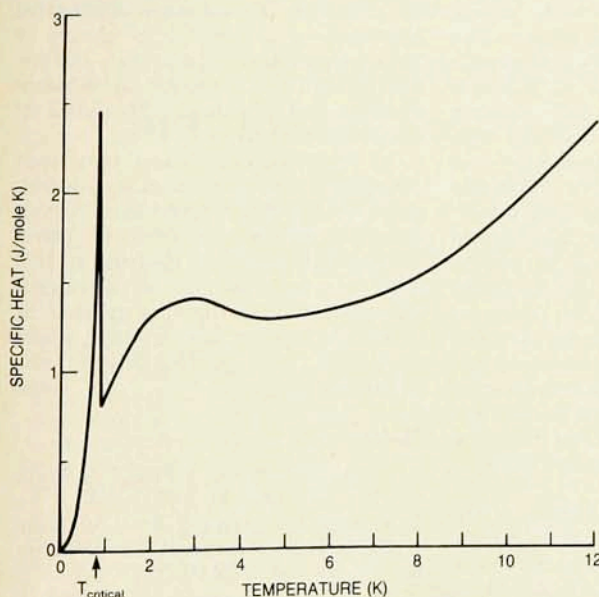
opportunities for finding new effects and thus better understanding. Itinerant *f* electrons are much more strongly correlated than their *d*-shell cousins in the transition metals. "They offer us a rich new playing field," he argues, "because we can control their degree of localization experimentally"—by subjecting them to external variables such as pressure, temperature, magnetic fields and different bonding environments. Uranium, for example, exhibits both charge density waves and superconductivity. Plutonium, to cite another case, goes through six different crystal structures as one cools it, demonstrating a delicate balance between entropy and degree of localization. Having now found two heavy-fermion *f*-band materials that go superconducting, Geballe believes, we are bound to find more, and we should see new, as yet unsuspected ways this delicate balance can manifest itself.

The experimental results on polycrystalline UBe_{13} by Ott and H. Rudigier at ETH and their Los Alamos collaborators Zachary Fisk and James L. Smith closely parallel the earlier CeCu_2Si_2 observations² by Steglich and his colleagues. Above about 100 K, the magnetic susceptibility increases with decreasing temperature in much the same way that a ferromagnetic material behaves above its Curie temperature (Curie-Weiss temperature dependence). Together with the large electronic specific heat, this leads one to anticipate an eventual transition to a magnetically ordered state.

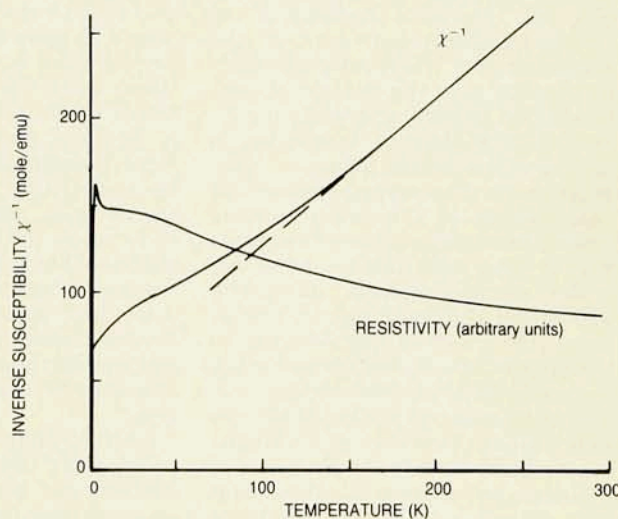
What one finds with further decreas-

ing temperature is something quite different. As the magnetic susceptibility of the UBe_{13} sample begins to fall below the Curie-Weiss curve, the ratio of the specific heat over the temperature grows dramatically, reaching a value of about 1 joule/mole K^2 below 10 K—a hundred times what one sees in the transition metals. One appears to be witnessing a low-temperature transition to a Fermi liquid. The itinerant electrons of an ordinary metal condense to a Fermi liquid at about 10 000 K, a temperature corresponding to the Fermi energy of a typical conduction band. The appearance of a Fermi liquid in UBe_{13} at about 10 K suggests that its electrons come from an extraordinarily narrow conduction band—the 5*f* band—only about 10 K wide. So narrow a band implies an enormous density of electron states in momentum space and thus an effective electron mass perhaps a thousand times that of the free electron. This electron mass is also measured by the ratio of the specific heat over temperature.

The great surprise comes as one continues to cool this heavy-fermion quantum liquid below 1 K. Instead of the magnetically ordered state one might have expected, Ott and his colleagues found that the temperature-independent high paramagnetic susceptibility dropped precipitously just below 0.9 K to a negative (diamagnetic) value, signaling a superconducting transition. Simultaneously, the already enormous specific heat suddenly and briefly rose to twice its value (2 joules/mole K), and the resistivity,



Specific heat, resistivity and magnetic susceptibility of UBe_{13} as measured by the ETH-Los Alamos collaboration. Above about 150 K, the susceptibility χ exhibits Curie-Weiss temperature dependence (dashed line), leading one to expect a magnetically ordered state rather than a superconducting transition at lower temperature. The steady rise of resistivity with falling temperature is quite anomalous.



Before the resistivity falls to zero at 0.85 K, we see a peculiar peak at 2.4 K. The electronic specific heat, C , also exhibits a peak just before the superconducting transition. The specific heat at low temperatures is several orders of magnitude higher than that of ordinary metals, indicating a very large effective electron mass. The dramatic rise of the ratio C/T below 10 K suggests a Fermi liquid.

which had been rising with falling temperature, fell abruptly to zero.

This superconducting state of UBe_{13} proves to be extraordinarily immune to destruction by magnetic fields. Brian Maple and his LaJolla colleagues J. W. Chen and Steven Lambert have recently measured the temperature derivative of its upper critical field (UBe_{13} is a type-II superconductor) to be at least 2 megagauss/K at T_c , by far the highest such slope ever seen in a superconductor, and all the more extraordinary at so low a T_c . This result again indicates very high masses for the electrons forming the superconducting pairs.

The superconducting transition, as described by conventional BCS theory, involves the spin-singlet pairing of electrons through lattice phonon coupling to form even-parity bosons. Some sort of Bose condensation is essential if one is to turn a Fermi liquid of electrons into a single macroscopic quantum state in spite of the Pauli exclusion principle. But in these exotic new superconductors, with electrons *almost as heavy as protons*, it is difficult to imagine that the usual BCS mechanism is operative. The Cooper pairing of electrons in BCS theory comes about because the spin-singlet electron-electron attraction via lattice phonons is stronger than the Coulomb repulsion, which is weakened by being spread over the large energy range of the conduction-band width. But in these f-band superconductors, Varma points out, the band width is much smaller than the phonon energy, appearing to render the Coulomb repulsion dominant in the spin-singlet state.

It looks as if the itinerant f electrons that are pairing to produce superconductivity are the same ones that are canceling the dipole moments of the localized f electrons to turn off the magnetization. Whereas for spin-singlet electron pairs the electron-phonon interaction is attractive while the direct electron-electron interaction is repulsive, the reverse is true for spin-triplet pairs. The strong paramagnetic susceptibility of the heavy fermion superconductors just above T_c suggests that in these materials the attractive triplet electron-electron interaction will be the dominant pairing mechanism. It may well be, therefore, that in UBe_{13} and CeCu_2Si_2 we are seeing the first example of superconducting electron pairs in an odd-parity, spin-triplet state instead of the even-parity singlet Cooper pairs of BCS theory.

There is no fundamental impediment to spin-triplet pairing, but in ordinary superconductors nonmagnetic impurity perturbations would tend to destroy the symmetry of a spin-triplet superconducting ground state. In the past, therefore, people have been skeptical about the possibility of getting a mate-

rial clean enough to make triplet superconducting pairing possible. Varma has pointed out, however, that the very large effective electron masses in these exotic materials greatly suppress the destructive influence of nonmagnetic impurities by reducing the superconducting coherence length.

Ott and his ETH colleagues have recently come up with some tentative experimental evidence for triplet pairing in UBe_{13} . They find that its critical temperature is suppressed by the addition of a nonmagnetic impurity (thorium) in a manner one would not expect of a spin-singlet superconductor. Superfluid He^3 is an example of spin-triplet (nuclear) pairing. Fisk suggests that CeCu_2Si_2 and UBe_{13} "may prove to be the electronic analogs of He^3 ." It is certainly clear, he adds, that superconductivity in these materials "is developing out of a very unusual ground state."

On the other hand, Steglich and his coworkers cite³ experimental evidence against triplet pairing in CeCu_2Si_2 . They find that the low-temperature ratio of the paramagnetic susceptibility over the specific heat coefficient γ is about one half, whereas one would expect this ratio to be greater than one for a triplet superconductor. They also argue that the T_c dependence on nonmagnetic impurities is more likely to be due to lattice disruption than triplet pairing.

Another uranium compound, U_6Fe , appears to be something of a bridge between the heavy-fermion superconductors, characterized by strong magnetic renormalization, and the conventional superconductors with strong electron-phonon renormalization. The superconductivity of U_6Fe was discovered by John Hulm and B. S. Chandrasekhar 25 years ago, but only recently has the work of Lance DeLong, John Huber (University of Kentucky) and their La Jolla collaborators, Maple and K. N. Yang, shown⁴ that this low- T_c superconductor (3.7 K) is characterized by anomalously high values of the specific heat, magnetic susceptibility and critical field. "It's not quite as exotic as UBe_{13} and CeCu_2Si_2 ," DeLong concedes, "but it's an interesting and potentially fruitful demonstration of the enhancement of these parameters and the decrease of critical temperature with increasing magnetic interaction."

Another interesting example of the antagonistic interaction of superconductivity and magnetism is the "reentrant superconductor" ErRh_4B_4 , whose peculiar vanishing superconductivity was discovered by Bernd Matthias, Maple and their LaJolla coworkers (see PHYSICS TODAY, April 1977, page 17). In such materials, superconductivity gives way to long-range ferromagnetic

ordering at temperatures far below T_c . The essential difference between these materials and the new heavy-fermion superconductors appears to be that in the former, one is seeing a competition between two distinct electronic systems—a magnetic system and a superconducting system. In the case of UBe_{13} and CeCu_2Si_2 , by contrast, it is apparently the same electrons—the partially filled f band—that are vacillating between a magnetic and a superconducting state.

"No theorist has done anything yet about heavy-fermion superconductivity that makes any sense to me," says Philip Anderson (Princeton). No one has as yet put forward a quantitative theory of the superconducting transition in these materials, but Varma believes that the concept of delocalization by resonant hybridization near the Fermi level between the f-band and the overlapping light-electron conduction bands at least explains the high effective masses, if not the superconductivity. When two different electronic states are nearly degenerate in energy, he argues, "the material exploits this degeneracy to fluctuate quantum mechanically between these states." This "fluctuating-valence" picture, however, has been challenged by John Wilkins (Cornell) and Duncan Haldane (University of Southern California). They argue that fluctuating-valence behavior in f-band materials—first studied experimentally by Geballe and coworkers in the 1960s—is better described by generalizations of a model proposed by Anderson in 1961 to explain the magnetic behavior of impurity atoms in metals. An alternative approach—a Kondo lattice theory of fluctuating valence behavior—has recently been put forward⁵ by Masashi Tachiki and Sadamichi Maekawa of Tohoku University.

At least we may know very soon whether the superconducting electron pairs in the heavy-fermion materials are spin singlets or triplets. David Bishop and Bertram Batlogg at Bell Labs are carrying out an ultrasonic attenuation experiment suggested by Varma to answer this question, whose resolution is a prerequisite to any sensible theory. —BMS

References

1. H. Ott, H. Rudigier, Z. Fisk, J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983).
2. F. Steglich, J. Aarts, C. Bredl, W. Lieke, D. Meshede, W. Franz, H. Schäfer, Phys. Rev. Lett. **43**, 1892 (1979).
3. W. Lieke, U. Rauchschwalbe, C. Bredl, F. Steglich, J. Aarts, F. DeBoer, J. Appl. Phys. **53**, 2111 (1982).
4. L. DeLong, J. Huber, K. N. Yang, M. B. Maple, Phys. Rev. Lett. **51**, 312 (1983).
5. M. Tachiki, S. Maekawa, Tohoku University preprint (1983). □