Superconducting films and devices

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Simple superconducting circuits and sensors can be fabricated in the new cuprate high temperature superconductors and conventional low-T_c components can be integrated at a large scale. Reliable multilayer depositions and Josephson junctions need to be developed in high-T_c materials for commercial success. The field has had approximately 12 years to grow since the discovery of ceramic superconductors that enable approximately 10 times higher temperature operation than was previously possible. Compared to more mature technologies, ceramic superconductors should now be about half-way to commercial realization. Niche markets, in which specialized devices strongly out perform the competition, will be the first to develop, however, large-scale superconducting electronics should be no serious threat to semiconducting electronics within the next 10 to 12 years.

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Current Opinion in Solid State & Materials Science 1999, 4:45–52

Electronic identifier: 1359-0286-004-00045

Abbreviations

ALL-MBE	atomic layer-by-layer molecular beam epitaxy
HTS	high temperature superconductor
RSFQ	rapid single flux quantum
SIS	superconductor-insulator-superconductor (tunnel junction)
SNS	superconductor-normal metal-superconductor
SQUID	superconducting quantum interference device
T_c	superconducting transition temperature

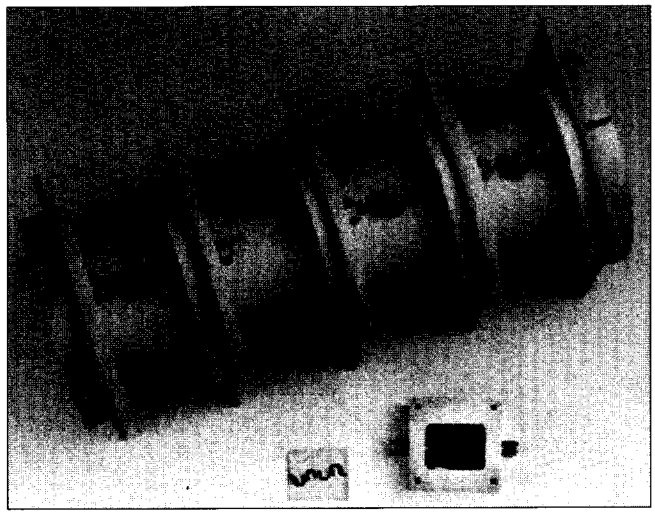
Introduction: superconducting electronic devices

This review will concentrate on superconducting electronic devices, in particular those based on ceramic high temperature superconductors (HTSs). These depend, to a large extent, upon the availability of high quality, thin films and, for active devices, of reliable and reproducible Josephson junctions. We will pick a few examples of devices and emphasize the demands they put on technology. Despite large, challenging materials problems, great efforts have resulted in a considerable understanding of and an ability to optimize the new ceramic superconductors. Technically, the devices often have superb properties, but mostly they are well ahead of market demands, having been developed from a scientific push rather than from a market pull.

Passive microwave components

This is the field upon which most of the American start-up companies concentrate and a commercial breakthrough, or a failure, will affect the future of superconducting electronics to a large extent. Low losses enable high frequency passive microwave structures with high Q-values [1], such as resonators (e.g. $Q = 10^3$ to 10^5 in planar resonators), filters, transmission lines, or phase shifters; an example of a filter is given in Figure 1.

Figure 1



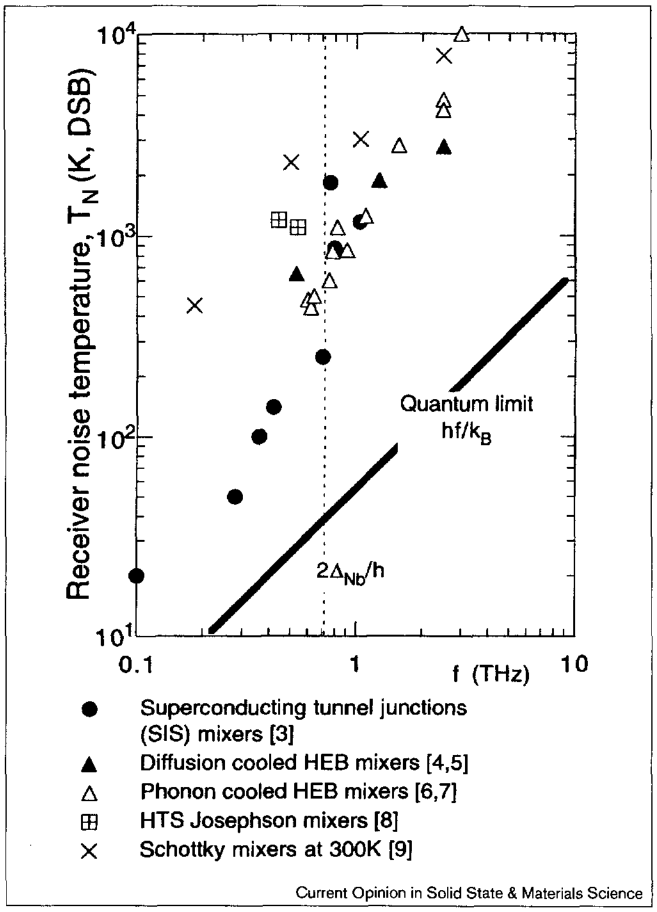
Superconducting filters are based upon the small surface resistance of a superconductor (even at high frequency). Resonators with very high Q-values and filters with sharp 'skirts' can be constructed. An example is given of a 7 pole, planar-type filter (bottom). It is compared with a much bulkier multiresonator filter of conventional technology (top). Similar filters are now being field tested in mobile cellular base stations, where low losses can give better signal-to-noise ratios. Reproduced with permission from [1].

Filters, that may be applied in cellular base stations or for satellite communication, have to be integrated with cooled, low noise amplifiers and cryo-coolers into systems. Superconductors can be combined with ferroelectrics to make tunable filters [2]. The resonator or center filter frequency can easily be varied by applying a voltage across an adjacent dielectric (ferroelectric) layer but the problem is microwave losses in the ferroelectric film or at the interface to the superconductor. Higher power handling capability is desired (e.g. for radar) and, particularly for satellites, a higher temperature of operation (passive cooling).

Low noise, millimeter wave mixers

The SIS mixer, based on superconductor-insulator-superconductor tunnel junctions, is at the heart of low noise receivers that approach a sensitivity limited by quantum fluctuations. They are routinely used in radio observatories at millimeter and submillimeter wavelengths, in the frequency range of 100–1000 GHz [3]. The upper limit in frequency is determined by the superconducting gap energy.

Figure 2



The noise temperatures are given for a number of high frequency mixers that have been developed in different technologies. The full line depicts the one photon limit, $T_N = hf/k_B$. SIS tunnel junction mixers give the best performance (T_N[K] ≈ 0.1f[GHz]) in the range 50–700 GHz. Newly developed hot electron (nonequilibrium) superconducting mixers are expected to give superior noise performance above 1 THz (T_N[K] ≈ f[GHz]). DSB, double side band; HEB, hot electron bolometer; T_N, receiver noise temperature.

Thus, one tries to make HTS tunnel junctions with higher gaps but that are still operable at low temperature, where thermal noise is low. Another way to access higher frequencies is to use a hot electron (non-equilibrium superconducting) mixer or bolometer which can be used at frequencies well above the gap value [4–7]. As the lowest noise is desired, the receivers need to be cooled to low temperature. Thus, low-T_c superconductors (LTS) are used – Nb/AlO_x/Nb tunnel junctions in SIS mixers and Nb or NbN microbridges in hot electron mixers. Noise temperatures versus frequency are compared in Figure 2 [3–9] for a number of mixer types. Such receivers can also be used for environmental studies, such as mapping of the ozone layer, where HTS may be applicable in mechanically refrigerated systems. Integrated receivers with cooled antenna, a local Josephson oscillator, mixer, intermediate frequency matching structures, and a low noise amplifier may be combined (even on a chip) in low power, low weight, multibeam receivers for satellites [10].

Metrology

The Josephson relation, hf = 2eV, gives a precise relation between voltage (V) and frequency (f), where h is Planck's constant and e is the electron charge [11]. Voltage standards at 10 V can be realized by mixing microwaves of extremely well-defined frequency with internally generated Josephson radiation in an array of up to 20,000 series coupled LTS Josephson junctions [12]. Programmable dc and ac LTS and HTS voltage standards are being developed [13,14].

Magnetic (current, voltage) sensors

Superconducting quantum interference devices (SQUIDs) can be fabricated with a magnetic flux sensitivity better than $10^{-5}\Phi_0/\sqrt{\text{Hz}}$ ($\Phi_0 = h/2e \approx 2.10^{-15}$ Vs, is the flux quantum). HTS SQUIDs are approximately as sensitive as LTS ones at f >10 Hz, but 1/f noise sets limits at low frequency. Progress has been made in decreasing the motion of flux quanta in the vicinity of the Josephson junction by incorporating flux locking holes in the transformer structure [15]. HTS rf-SQUIDs also show good performance [16]. The bandwidth of SQUID amplifiers applied, for example, as pico-voltmeters is being expanded [17] with the hope of going beyond 50 MHz.

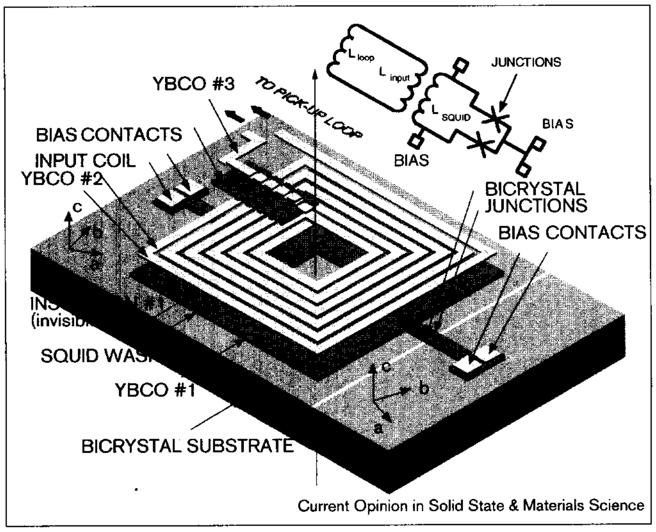
The SQUID, which is outlined in Figure 3, has potential sensitivity for a number of applications, for example, measuring heart beats and currents in the brain of the human body [18], the latter is presently limited to LTS systems, both need clinical acceptance; non-destructive testing of corrosion or fatigue cracks in construction structures, for example, in airplane landing wheels [19]; and geophysical exploration. The lateral resolution (presently ~50 mm) of a scanning SQUID microscope [20] is limited by the size of the hole of the pick-up loop and the distance to the sample that is investigated.

Low noise junctions and a reliable HTS multilayer technology, without pinholes in the insulator, and non-degraded cross-overs and contacts between electrodes in different layers need to be further developed.

Logic circuits

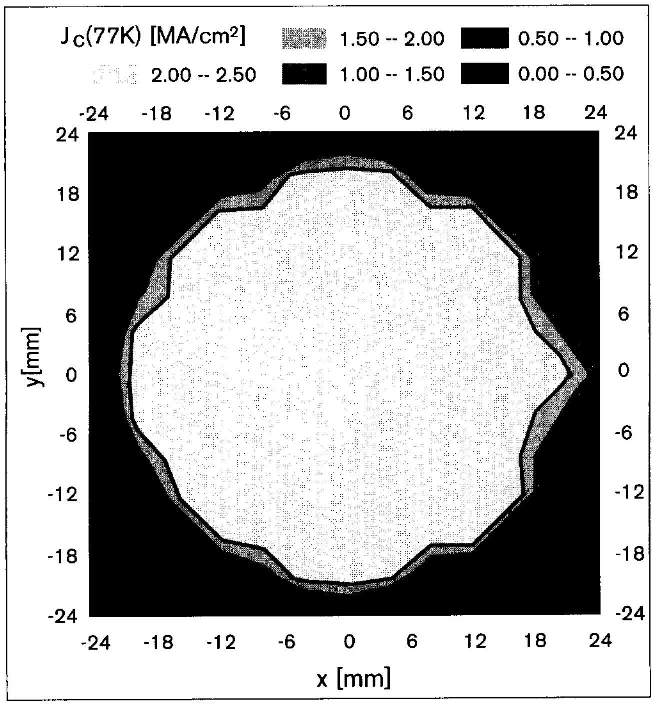
Ultra-fast switching and massive data processing are speculative possibilities for digital circuits (petaflop computing with 10^{15} floating point operations per second has been proposed [21]). Currently, only rapid single flux quantum (RSFQ) logics [22] appear to be truly superior to semiconductor circuits, with switching less than 10 ps and a low power dissipation, of the order of 10 mW per switching gate. Devices such as fast A/D (analogue to digital) converters have been successfully demonstrated using Nb technology and a few thousand junctions [23], whereas HTS circuits have been limited to approximately twenty junctions [24,25]. This is due to a large spread in junction

Figure 3



A superconducting quantum interference device, SQUID. Magnetic flux is transformed into a superconducting loop containing two weak links. The voltage across these varies periodically with flux, the period being a quantized unit, $\Phi_0 = h/2e$. Changes in flux as small as $10^{-5}\Phi_0\sqrt{Hz}$ can be detected. The diagram shows the superconducting transformer over the loop (washer) with two bicrystal junctions in HTS material. By introducing holes or moats in the superconducting strips, it is possible to minimize flux jumps and, thus, decrease the low frequency, so-called 1/f, noise. A variant is the rf-SQUID with only one weak link in a loop that is coupled to a circuit that is resonant at radio frequency. YBCO #1–#3 indicate the subsequent epitaxial YBCO thin film layers deposited. #1 And #2 are the required epitaxial insulation layers. The coordinates a, b, c on either side of the bicrystal grain boundary indicate the growth orientation of the YBCO films.

Figure 4



Only a small spread in J_c is registered over a 2" diameter YBaCuO film deposited by co-evaporation. Very thin layers are sequentially deposited on the hot substrate and annealed in a special pocket having a higher oxygen pressure than the high vacuum needed in the evaporation process. This technique was developed at the Technical University of Munich [34]. The measurement of J_c was done using Bean's method of sensing the mutual inductance between double coils scanned across the sample at 77K. Courtesy of H Kinder [39].

parameters for the latter, typically >10% as compared to 2–4% (even <1%) for LTS junctions [26] – which is approximately twice the value needed for acceptable operating margins. The market for HTS logics, integrated with a cryo-cooler at ~20–30K, could be huge if problems of junction quality and integration can be solved and cheap refrigerators be developed. Competing, high speed and densely packed semiconductor logics is also developing rapidly, even if one can see limits in miniaturization, speed, and cost.

Radiation detectors

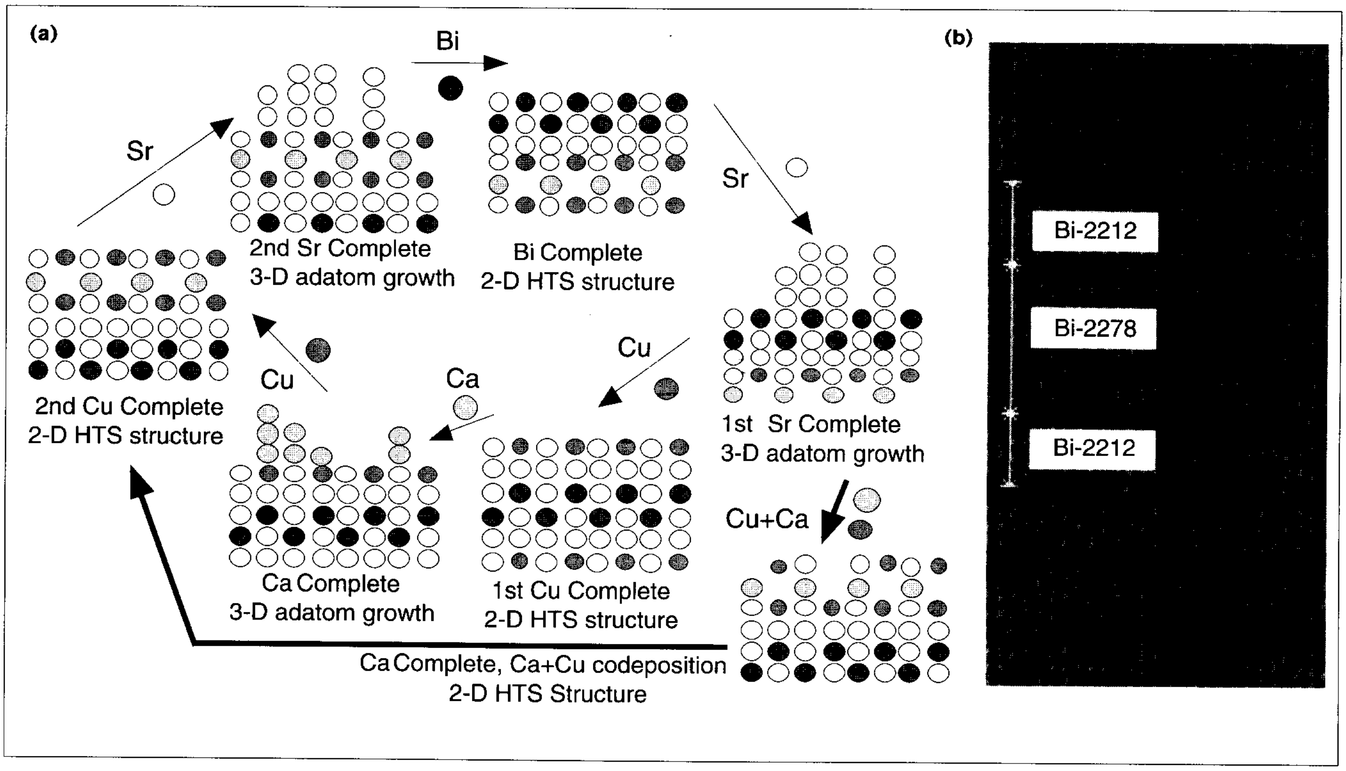
The energy resolution of X-ray photon detectors can be improved by at least an order of magnitude (to <10 eV) using a transition edge bolometer or a tunnel junction for the read-out of changes due to absorbed photons [27]. Elemental analysis in electron microscopes (EDX) would benefit from such detectors and the market could be sizable. Sensitive bolometers, possibly enhanced by Andreev reflections of hot electrons, enable studies of weak sources, such as inhomogeneities in the background radiation in space [28,29]. These sensitive radiation detectors will be limited to low-temperature operation and, presently, to LTS.

Thin superconducting films

The main attribute of the new ceramic superconductors is the layered, anisotropic structure, with conducting CuO_2 planes separated by spacers of alkaline earth or rare earth metals and other metal oxides that function as charge reservoirs [30]. Superconductors with only one CuO_2 plane exhibit critical temperatures, T_cs, up to ~93K (with YBaCuO being the most commonly used compound). Layered cuprates with two to three CuO_2 planes in the unit cell (as in BiSrCaCuO, TlCaBaCuO, or HgBaCaCuO) can have T_cs of up to 110 to 135K (even up to 160K with an applied pressure). The complex chemistry, with three to four metals and oxygen, makes epitaxial growth of HTS films a challenging task.

One approach is to deposit a cuprate thin film on a heated substrate by a stoichiometric transfer of molecules from a bulk target in a reduced oxygen plasma. The flux can be obtained by sputtering [31], pulsed laser deposition [32], or ion beam deposition. Another method is to use molecular beam epitaxy when, for example, beams of thermally evaporated metals [33,34] (or metal-organic gases [35]) and activated oxygen are directed towards the substrate, where

Figure 5



(a) A schematic presentation of the atomic layer-by-layer beam epitaxial (ALL-MBE) growth of a $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) monolayer. The growth cycle is marked by the thin arrows. Note that the Sr and Sr/Ca 3D domains disappear after the deposition of Cu and Bi layers. The formation of 3D Ca domains can be avoided in a dual Cu+Ca flux, the path of which is marked by bold arrows. The scheme is a real space interpretation of RHEED (reflection high-energy electron diffraction) patterns taken during the growth of the Bi-2212 film. **(b)** A transmission electron micrograph of a metastable single $Bi_2Sr_2Ca_7Cu_8O_x$ (Bi-2278) layer inserted into Bi-2212. Reproduced with permission from [33].

they react and grow into a film that is lattice matched to the substrate. The first approach is more simple and allows flexible growth of different high quality films, whereas the second approach demands individual monitoring and control of each flux in order to hit the relatively narrow range of stoichiometry. However, the second method tends to give smoother films and is much faster than the first it also seems to be the industrially preferred one, at present, and its use is growing.

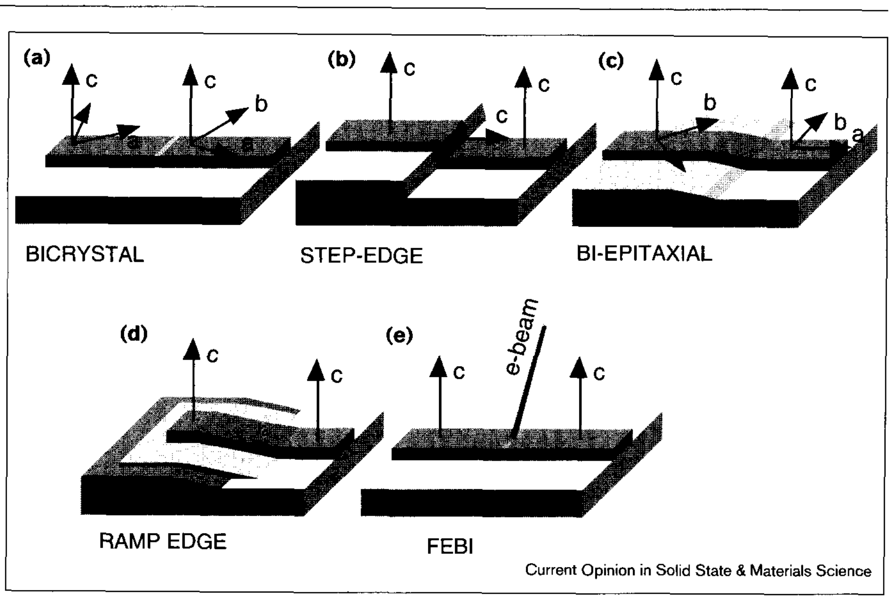
A more complex method is layer-by-layer epitaxial growth, where the individual atomic layers are deposited one by one, for example, by opening shutters in front of the thermal sources sequentially [33] or by multi-target laser ablation [32,36].

Single crystal substrates of oxides like $SrTiO_3$, $LaAlO_3$, $NdGaO_3$, and MgO are compatible with HTS films. Other substrates may be more desirable, for example, due to better microwave performance, availability, or price, but the diffusion of atoms from the substrate to the superconductor may be devastating to the latter. Buffer layers of $SrTiO_3$, CeO_2, YSZ, and others have been used to prevent such interdiffusion [37].

$(RE)Ba_2Cu_3O_{7\delta}$ films, with RE = Y, Nd, Dy, and Sm, have been grown on a variety of substrates up to 8–9 inches in diameter and, in some cases, coated on both sides. The best films are of high single crystalline quality with X-ray rocking curve widths of ~0.04–0.07°, complete in plane alignment and have a surface roughness in the range 1–4 nm. Films with $T_c \geq 90K$, a critical current density of $j_c \approx (1–5) \times 10^6$ A/cm^2 and a microwave surface resistance of 10^{-4} Ω at 10 GHz and 77K have been deposited by laser ablation [38], sputtering [31], and co-evaporation [39] in several laboratories. The spread in superconducting parameters is as low as 1–3% over a 2″ substrate, see Figure 4.

The values quoted are valid for single layer films. Many applications, however, need multilayers of superconductors and insulators, for example, SQUID transformers, transmission lines over ground planes in high speed logics, or tunable filters. Even small deviations in stoichiometry during deposition cause defects that are amplified with

Figure 6



Examples of different Josephson junctions in HTS. **(a)** Bicrystal grain boundary junction in which a misorientation is transferred from the two crystal parts of the substrate to the epitaxially grown HTS. The Josephson current is exponentially dependent on the misorientation angle. **(b)** Step-edge grain boundary junction. If the microbridge is all HTS, the step is usually less steep than drawn in the figure and two, or more, grain boundaries exist, one of which dominates the behavior. The region of a steep step can also be filled by a nonsuperconducting metal. **(c)** A bi-epitaxial junction using a seed layer to give another orientation. The misorientation is usually 45°, which gives a rather small critical current. **(d)** A ramp-edge junction in which a thin insulator is formed between the insulator-covered bottom HTS electrode, which has been cut under an edge, and the upper irradiated HTS electrode. **(e)** Focused electron beam (or ion-implanted or ion-damaged) junction.

film thickness and cause both electrical shorts and undesired grain boundaries. Multilayers of insulators can disrupt columnar growth and fill up holes between superconducting layers [40].

Excellent control of composition and sharp interfaces between layers, even enabling tunnel junction formation, have been obtained by atomic layer-by-layer molecular beam epitaxy (ALL-MBE [33]), see Figure 5. This method may be used to fabricate metastable crystal structures with, for example, a larger number of stacked CuO_2 planes than is possible in thermodynamic equilibrium. Thus, it might be possible to form novel superconductors with a higher T_c.

Josephson weak links and tunnel junctions

Josephson effects are central in all active superconducting devices. A well-established, standard technology exists for producing reliable Josephson tunnel junctions in low-T_c materials [41]. An insulating barrier of well-defined thickness is formed by oxidizing a thin Al layer in a multilayer of Nb/Al/Al-oxide/Nb. A reproducibility and a uniformity of tunneling parameters, like the Josephson critical current, I_c, and the normal state resistance, R_n, of the order of one percent have been obtained. NbN/MgO/NbN junctions for digital operation at 10K, which is much more economical than operating at 4K, have been developed but the control of the barrier thickness is more difficult than for the forgiving Al. It is much more difficult to fabricate high-T_c junctions as their coherence length is much shorter, the interfaces are distorted and there is no self-limiting formation of the barrier thickness as for Al. Best results give a reproducibility of the order of 10%. One desires a high I_cR_n value but it is usually limited to 0.1–1 mV, that is, no higher than that for low-T_c superconductors. Several approaches have been tried to fabricate HTS Josephson junctions or weak links, and some of these are illustrated in Figure 6.

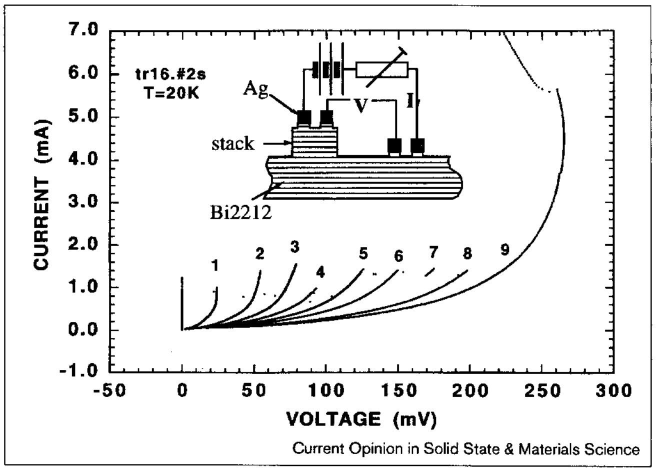
The first development was to use natural and man-made grain boundaries, across which Josephson weak links are formed. Microbridges in polycrystalline films belong to this category, as well as bi-crystal [42], bi-epitaxial [43], and step-edge [44] junctions. The junction properties can be controlled, for example, by the misorientation angle in the bicrystal junction or by the step-edge height and angle.

Another approach is to use man-made artificial barriers, such as a normal metal (Al with its natural oxide layer) across a step edge [45], forming an SNS junction, or ramp-edge junctions [46,47] with a variety of different barriers. Co-doped YBaCuO [48] and Ga-doped PrBaCuO [46,47] have been used but much attention is presently given to ion-damaged and subsequently annealed barriers [49].

A third category is based upon the local damage caused by irradiating thin strips with high energy electrons or ions to form SNS or SS'S structures. For example, narrow beams of electrons in transmission electron microscopes with 200–350 keV energy have been used to write lines across microbridges [50,51]. Initial problems with stability in time were overcome by annealing. Beams in the range 20–40 nm, of 150–200 keV ions (Ga, Be, Ne, O) have been implanted (dose of 10^{13}–10^{16} ions per cm^2) to cause local disorder either in the substrate (before deposition) or in the HTS film itself [52,53]. This gives rise to a local region of empirical superconductivity.

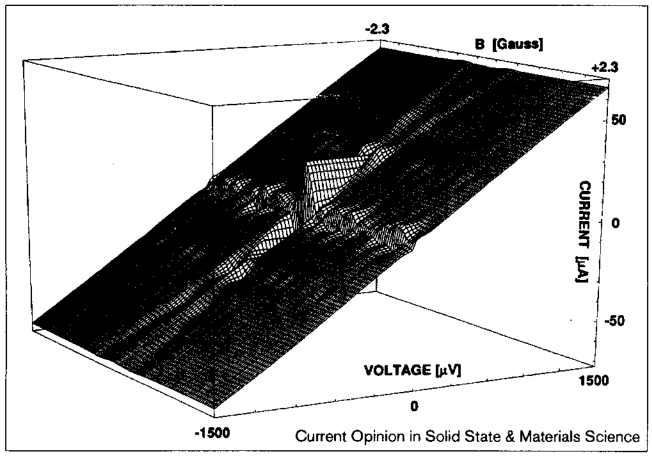
A fourth method uses tunneling between the CuO_2 layers in the c-axis direction of a single crystal or an epitaxial film. One can either etch columns into an HTS superconductor, like Bi-2212, to form so called intrinsic junctions [54] or one can deposit layers by ALL-MBE as described above [33]. If the structures are relatively small, one can avoid

Figure 7



Intrinsic tunnel junctions can be formed between superconducting CuO_2 planes for current transport in the c-direction of a single crystalline HTS. The insert shows how a stack of junctions can be etched into a Bi-2212 crystal while the sample is cooled to 77K and I–V curves are monitored. The current-voltage curves show that as the Josephson current of one of the nine series-coupled junctions is exceeded, there is a jump in voltage to next branch. The different curves are traced out as the current is decreased/increased on each branch. The back bending at high voltage may be due to a non-equilibrium state in which a large number of quasi-particles are injected at energies above the energy gaps of the junctions (it could also be due to internal heating).

Figure 8



Fiske steps. Magnetic field dependence of the I–V curves for a 0–32° bicrystal junction at T = 4K. At zero voltage we see a Fraunhofer type modulation and, at higher voltages, the modulation is due to Fiske modes, that is, standing waves of electromagnetic radiation in the junction. (Notice that the I–V curves were traced only from zero voltage to maximum/minimum bias voltage to avoid the hysteresis in the I–V curves.)

misfits in the layering and obtain 'traditional' tunneling curves with hysteresis. An example of tunneling in an intrinsic junction is shown in Figure 7.

An ideal Fraunhofer type variation of the Josephson current with magnetic flux is seldom seen in an HTS junction. This may be due to metallic shorts in the barrier but more probably this is due to tunneling *via* localized states [55]. The latter would explain the observed proportional dependence of I_cR_n to $\sqrt{I_c}$. The localized states may be due to stresses, defects or inclusions in the interface regions (which are seen in electron micrographs). A band bending model, with midgap states, was recently proposed by Mannhart *et al.* [56]. It is related to well known semiconductor junction behavior – HTS can be seen as strongly doped semiconductors with larger screening lengths than normal metals.

A question is whether the grain boundary should be considered as a normal metal or as an insulator (SNS or SIS junctions). The presence of well-developed geometrical resonances at certain frequencies of the Josephson oscillation, see Figure 8, indicates a relatively high Q-value, as in an insulator or semiconductor [57].

A non-zero current within the gap voltage of an HTS junction without shorts, even at low temperature, is a token of a varying energy gap or states in the 'gap'. Low-T_c superelectrons are paired with opposite spins and velocities, a so-called s-wave symmetry (S = L = 0). The layered HTSs are considered to have not only a very anisotropic gap parameter but even a d-wave symmetry (which means that the Cooper pairs have opposite spins but a more complicated orbital motion, S = 0, L = 2, and that the gap, or order, parameter changes sign, i.e. phase, in diagonal directions of the ab-plane). The existence of fluxons with half the flux quantum value, $\Phi_o/2$, at tri-crystal boundaries (where three differently oriented crystals meet) is considered as proof of d-wave symmetry [58]. However, other tunneling experiments may be explained by a mixture of s- and d-wave symmetry (or, possibly, by an extended s-wave symmetry) [59,60].

As superconducting films usually do not grow perfectly, grain boundaries are also not perfect and often display kinks at the 10–15 nm scale [61]. A varying tunneling direction would mean that the gap would vary in space in a d-wave situation and even π-junctions with opposite current directions may appear in parts of the interface [62], thus, resulting in an unsymmetric field behavior and a lower I_cR_n product.

The smaller gap energy for tunneling in the c-direction (of intrinsic junctions) than in the a-, b-directions of, for example, Bi-based cuprates indicates a nonequilibrium state (heating) or tunneling *via* metallic, or semiconducting, layers in between the superconducting CuO_2 layers [63]. A resonance type structure in the conductance at certain bias voltages may be due to a coupling to lattice vibrations at corresponding frequencies.

Conclusions

There has been a rapid development in films and devices since the discovery of high temperature superconductivity in oxides – despite the fact that the materials are extremely complicated and that basic concepts are not yet understood. The sensitivity of SQUID sensors, for example, has improved by more than eight orders of magnitude since the first HTS versions were made. The low-T_c SQUIDs are still the choice for most sensitive brain magnetometry and Nb-based digital processors are much closer to large scale realization than HTS RSFQ circuits. In any case extremely low noise detectors have to be cooled to low temperature to avoid thermal noise; however, HTS devices may have advantages in niche applications. Passive microwave devices with low losses and SQUIDs for non-destructive testing, geophysical exploration and heart screening seem to be the closest to a breakthrough.

Multilayers of films, for multi-level integrated circuits and sensors, and predictable, fully reproducible and reliable Josephson junctions need to be further developed to allow large scale application. The development of economic refrigerators for operation at 10–30K is also important. Presently, a large portion of HTS electronics are envisaged to operate at temperatures intermediate between those of liquid helium and liquid nitrogen.

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