Theodore Henry Geballe

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A Biographical Memoir by Malcolm R. Beasley, Aharon Kapitulnik, Richard L. Greene, Frances Hellman, and John M. Rowell

Figure 1 Theodore H. Geballe. *Courtesy Theodore H. Geballe Laboratory for Advanced Materials.*

Theodore "Ted" Geballe believed deeply that new physics would manifest in new materials or those "maverick" materials that the conventional wisdom can't explain. His life bears witness to the aphorism: if you want to find something new, don't look under a streetlight. Ted was particularly interested in new cooperative states of matter and the competition between them, such as, how materials order from interactions at low temperatures, as thermal fluctuations diminish. Ted had a remarkable memory and an equally remarkable associative mind. He understood the vast world of inorganic materials and their chemical compositions, structures, and properties. He had a nose for which materials might exhibit interesting cooperative phenomena or offer fertile ground in which to test some newly proposed ordered state. Ted's greatest love, however, was clearly superconductivity: in what classes of materials does it exist, why does it exist, and what is the mechanism. He was a natural collaborator. At Stanford he was a member of the successful KGB (Kapitulnik-Geballe-Beasley) group that evolved organically as the members successively joined the Stanford faculty.

Ted enjoyed seemingly countless friends. He was kind and caring to his family and to his colleagues, graduate students, postdocs, visitors, and laboratory staff. At his 100th birthday celebration, more than 100 people came from all over the world to honor him, hear scientific talks, and pay tribute to a remarkable scientist and human being.

Childhood and Undergraduate Years (1920–41)

Ted was born and raised in San Francisco, and, with apologies to Tony Bennett, he held this "City by the Bay" deeply in his heart. In his later years at Stanford, he charmed generations of graduate students with stories about his youthful (mis)adventures and his competition with his older brother Ron as they grew up in the city during a kinder, gentler time. Both became distinguished scientists.

Ted's father was a lawyer turned businessman during the Great Depression, and his mother was active in social service organizations for Jewish immigrants. By all accounts, they nurtured a lively intellectual climate in the family. Ted's grandfather Isidor Geballe immigrated to the United States from Posen, then part of Prussia, in the latter half of the nineteenth century because opportunities for Jewish citizens were circumscribed. The family originally settled in New York City but moved to upstate New York, where his father Oscar Geballe was born. They relocated to San Francisco in 1904, seeking opportunity and communities congenial to their Jewish heritage. Ted was born in San Francisco in 1920, three years after Ron.

Ted studied chemistry as an undergraduate at the University of California, Berkeley (UC Berkeley). His propensity and talent for research were evident early on. In his junior year, in a project-oriented lab course, he proposed to measure the work function of metals. By his own account, he failed, but apparently the lab instructor was so impressed with Ted's dedication and ability that he recommended Ted to the famous chemist William Giauque for his senior thesis. Giauque asked Ted to measure the heat capacity of gold at low temperatures with high precision. Ted ultimately published this work after he returned from military service in the army during World War II. During this time, he met his wife-to-be, Frances (Sissy) Koshland, who was an English major. Their union of the humanities and sciences lasted seventy-seven years.

Military Service and Graduate Years (1941–52)

In normal times, most new college graduates seeking a Ph.D. go directly to graduate school. But Ted did not graduate in normal times; World War II intervened. In the spring of 1941, he entered active duty as an officer in the U.S. Army Ordinance Corps, having participated in the ROTC program that UC Berkeley required at that time. For the next four years, he maintained artillery systems in the South Pacific. Toward the end of the war, he witnessed the beginning of the era when the military aimed anti-aircraft artillery electronically rather than mechanically.

After the war, with some soul searching and encouragement from Sissy, he decided to go to graduate school. The admissions process consisted of writing Giauque a letter to ask whether Giauque would accept him as a graduate student. Giauque agreed. That was it.

Giauque was interested in the third law of thermodynamics, which led him naturally to study materials at low temperatures to see whether and how the entropy of a material goes to a constant as the temperature goes to zero. His group worked at the forefront of discovering not only how materials order at low temperatures but also how to develop the complex refrigeration and measurement technologies necessary for such work, a rare combination in those days. Along the way, Giauque won the Nobel Prize in Chemistry for inventing adiabatic demagnetization as a way to cool materials to very low temperatures.

In this heady and demanding environment, Ted had to choose his thesis topic, always a revealing process. Paraphrasing his own words: stimulated by two student colleagues in Giauque's group who chose to study spin ordering of transition-metal ions in hydrated paramagnetic salts, he went to the library and found a salt, $CuSO_4 \cdot 5H_2O$, that seemed particularly simple because Cu^{++} has only one unpaired electron. Ted's instincts to identify good systems in which to explore physical phenomenon already were evident. His measurements showed only half of the expected Rln2 entropy of the spin ½ ions was removed in an 8000 Oe magnetic field. Because the structure had two different Cu sites, he concluded that only one of the sites had polarized. Later, after he submitted his thesis, he realized that this result was a natural outcome of super-exchange, a concept that was under active theoretical development at the time. All in all, it was an auspicious beginning.

During this time, Ted also took a course from Charles Kittel, who was visiting from Bell Laboratories in New Jersey. The course became the basis for the first edition of Kittel's famous book, *Solid State Physics*. In addition, he attended a seminar by Homer Hagstrum, also from Bell. After the seminar, he asked Hagstrum what research was going on at Bell in solid-state physics. Almost immediately, he received a letter inviting him to visit Bell, after which he received a job offer. Again, hard to imagine from a modern perspective.

The Bell Labs Years (1952–68)

Bell Labs played a seminal role in the emergence of solid-state (now condensed-matter) physics as an important subfield of physics, with symbiotic connections to material science and chemistry and relevance to technology. In the aftermath of World War II, a remarkable number of outstanding young researchers were attracted to Bell. Ted was among them. As he put it in his personal memoir: "New science and technology were pouring out of Bell. When I was offered a staff position there, with the promise of choosing my own research project, it seemed too good to be true. But it wasn't."

After arriving at Bell in 1952, just five years after the invention of the transistor, Ted had to choose a group. He chose the Semiconductor Department, building on the expertise he acquired in Giauque's lab to make electrical and thermal measurements at low temperatures. He made measurements on various semiconductors at low temperatures, for which researchers had not thoroughly explored their properties. Along the way, he found an unexpectedly large thermoelectric power (now known as the Seebeck effect) in high-purity Ge. Discussions with Conyers Herring, a theorist at Bell, revealed that Ted had found a new phenomenon, now known as "phonon drag." These discussions also sparked a lifetime friendship.

The research landscape changed after 1954, when John Hulm and George Hardy discovered superconductivity in the intermetallic compound V_3Si at 17.2K, a T_c roughly twice that of the highest T_c elemental superconductor Nb. Within a few months, Bernd Matthias at Bell discovered superconductivity in Nb_3Sn with T_c = 18.3 K. Ted joined forces with Bernd, forming the now legendary team working on superconducting materials. He also continued work on the low-temperature thermoelectric properties of semiconductors. Ted got off to a fast start in superconductivity, co-authoring the original publication on Nb_3Sn. Work on the entire class of A15-type superconductors naturally followed at Bell and elsewhere.

Ted acquired a special interest in establishing the mechanism of superconductivity across various classes of superconductors. He noted that the isotope effect ($T_c \sim 1/M^{1/2}$, where M is the isotopic mass) expected for materials exhibiting the electron-phonon interaction appeared experimentally in simple *s-p* band superconductors but not in superconductors that contained transition metals. He began with elemental Ru, in which he found no isotope effect, then proceeded to study a wide variety of superconductors containing transition metals in which the isotope effect was absent or notably reduced. Ted thought that a new mechanism of superconductivity might be at play. Ted was on to something, but not exactly what he thought. Rather, in early discussions with Phil Anderson at Bell, it became clear that he had not discovered a new mechanism. Actually, he had identified the need to include the repulsive electron-electron interaction as well as the attractive electron-phonon interaction when calculating the isotope effect for superconductors that contain transition metals (but not s-p band metals). Phil, with Pierre Morel, was just beginning to understand this problem.

At the same time, Ted and Bernd continued to search systematically for superconductors in various classes of materials of increasing complexity. In the early 1960s, they compiled their (and others') results into a review article, truly a magnum opus, that listed the known superconductors and their transition temperatures in various classes of materials, organized by structural class and elemental constituents. For the first time, they went beyond a simple compilation and gave reasons or conjectures regarding what the results revealed or suggested about the presence of superconductivity in more basic terms. In a later review, Ted discussed cases in which superconductivity was expected, but *not* found, based on their class and constituents. He always treated these maverick materials as potential sources of new physics or materials science.

By the end of the 1960s, Bernd had moved to the University of California, San Diego, and Ted was considering his future. Historically, the strongest players at Bell typically moved either into higher research management or into academic positions. Paraphrasing Ted from his personal memoir: unbeknownst to him, he was one of two people being considered for the position of head of the Physical Research Department at Bell. In the end, the position went to the other candidate, whom Ted felt personally was the better choice. But he did concede that had he been selected, he would have declined the offer for the position at Stanford in applied physics that came a few weeks later. What a consequential moment.

Early Years at Stanford (1968–74)

Ted came to Stanford in 1968 as a professor in the newly formed Department of Applied Physics with a joint appointment in the Department of Materials Science. He hired a postdoc (Rick Greene), a Swiss technician/engineer (Hans Thomas), and three graduate students (Frank DiSalvo, Chris King, and Bob Zubeck). Compared to Bell, the conditions for research were not ideal: an inexperienced postdoc and students and an empty old World War II laboratory with no air conditioning. But Ted was an optimist with a brilliant mind filled with research ideas (even if they were at first incomprehensible to his young research team). Moreover, his relaxed, easy-going personality inspired enthusiasm and joy in his group. For example, I (Rick) never saw Ted lose his temper, despite the many mistakes his group made in those early days.

Ted focused his research on interesting materials physics topics of the day, such as granular superconductors and tungsten bronze superconductors (generically M_xWO_3, where M represents an alkali metal), an early example of a superconducting oxide. Also, in his role as a consultant at Bell, Ted, A. Menth, and Ernest Buehler discovered the first Kondo insulator, samarium hexaboride (SmB_6), which was found—fifty years later—to be a topological Kondo insulator. This work reflected the emerging interest in f-band materials, as the superconducting materials community progressively moved its attention down the periodic table.

The Geballe Group achieved two particularly important results during these early Stanford years. First, they initiated development of the thermal-relaxation method to measure the specific heat of small samples—an activity that continued over several generations of students and has been progressively commercialized by the company Quantum Design. This heat capacity project typified Ted's collaborative style of research; everyone in his group participated and received credit for this significant instrumental advance.

Second, Ted discovered superconductivity in layered transition-metal dichalcogenides (TMDs) intercalated with organic molecules in collaboration with his graduate student Frank DiSalvo, Synvar employee Fred Gamble, and Dick

Klemm, a Stanford undergraduate working with Gamble. Their motivation was to discover an excitonic mechanism for superconductivity. Subsequently, Ted and Frank carried out a comprehensive study of TMDs and their intercalates. They enhanced T_c in TaS_2 intercalated with the organic molecule pyridine $TaS_2(\text{pyridine})_{1/2}$ and apparently observed 2D-like superconducting fluctuations above T_c, suggesting that the superconductivity in these materials was two-dimensional. Mac Beasley, then at Harvard University, privately questioned this interpretation, and Ted and Mac joined forces for the first time to sort out the situation, which involved Ted's student Bob Schwall working at Harvard with Mac's student Dan Prober. During this period, Mac also spent a sabbatical at Stanford and shortly thereafter joined the faculty of the Applied Physics Department.

For the record, Ted had seen a dimensional cross-over effect in the superconducting fluctuations far above T_c. The superconductivity itself was three dimensional.

The Ted-Mac Amateur Hour Era (1974–85)

By the time I (Mac) arrived at Stanford in 1974, Ted had already attracted Bob Hammond to his group, gaining Bob's expertise in advanced thin film co-deposition via multiple feedback-controlled e-beam co-evaporation sources. The goal of their initial project was to investigate the potential of the high-T_c A15 superconductors for application in AC power transmission lines, with the aid of a new group of students (Rich Howard, Bob Norton, and Jesse Salem). The project succeeded. The team found that Nb_3Sn films could match the ac loss performance of Nb—the historical choice for superconducting ac power transmission—with the advantage of higher operating temperatures.

But most important, Bob created a powerful new facility to synthesize thin films of advanced complex, multi-component materials and multilayers with atomic-layer control. In addition, Ted, then director of the Center for Materials Research at Stanford (funded by the National Science Foundation), fostered the development of an advanced co-sputtering facility by Troy Barbee and Doug Keith with similar capabilities. It would be hard to overstate the impact of these two facilities in opening new research possibilities at Stanford and elsewhere.

The first major development occurred when John Rowell, who was visiting at the time, observed that the Nb_3Sn films in the AC power line project exhibited far better surfaces than the bulk Nb_3Sn crystals used previously for electron tunneling studies. With high-quality tunnel junctions, one can extract the spectral weight of the superconducting pairing interaction $\alpha^2 F(\omega)$, which provides details of the pairing mechanism. Such data on the s-p band elemental superconductors show that their superconductivity resulted from the electron-phonon interaction.

Almost immediately, Mac's new student David Moore made Pb/Nb_3Sn tunnel junctions, extracted $\alpha^2 F(\omega)$, and confirmed definitively electrons pair in Nb_3Sn via the electron-phonon interaction. Eventually, Ted's students Ken Khilstrom, Raynien Kwo, and Bob Feldman extended these studies to all the high-T_c A15 superconductor, including the challenging metastable Nb_3Ge. It is fair to say that at the conclusion of this work, the group had definitively established the mechanism for superconductivity in the A15 class of superconductors, two and a half decades after their discovery.

Also, during this time, Ted's students Greg Stewart, Steve Early, and Frances Hellman greatly extended the earlier calorimetry work. They developed a "calorimeter on a chip" that increased the sensitivity of the relaxation method and enabled measurements on thin-film samples, including the A15 superconductors. After graduating with her PhD, Frances improved the relaxation method even further for which she won the Joseph F. Keithley Award for Advances in Measurement Science from the American Physical Society.

All this work was done with Ted's and Mac's students in constant interaction. The students christened themselves the "Ted-Mac Amateur Hour," in a play on words for a popular TV program of the time.

Never one to rest on his laurels, Ted, with Artie Bienenstock, joined the emerging community of researchers studying the properties of quenched-condensed physical-vapor-deposited amorphous materials, as opposed to the more traditional splat cooling. They wanted to understand the effects of homogeneous disorder on cooperative phenomena, in particular superconductivity and magnetism in amorphous transition metal/metalloid alloys (specifically Mo-Ge and Fe-Ge). They co-sputtered films on a rapidly spinning substrate holder, requiring multiple passes to deposit an atomic layer. Detailed analysis showed that the superconducting films were amorphous (that is, homogeneously disordered) with nearly constant resistivities down to film thicknesses of order 20 angstroms. In all cases, the superconducting properties approximated the predictions of BCS theory, although their transition temperatures were modest at best.

Impressed by the quality of these amorphous superconducting films produced by Ted and his students, Mac and his student John Graybeal realized that that these beautiful amorphous films could potentially serve as a model system to test the linear reduction of T_c as a function of R_{sq} predicted by the Berezinskii–Kosterlitz–Thouless (BKT) theory of 2D topological phase transitions in superconductors. They found a linear reduction alright, but with a slope nearly ten times higher than predicted by BKT theory. Ted was right—there was something to learn about homogeneously disordered superconductors: Why were their transition temperatures so low?

Theorists already suspected that Coulomb interactions were important in homogeneously disordered metals. Fukuyama, Ebisawa, and Maekawa had shown via BCS theory that when they included localization and Coulomb interactions, T_c decreases linearly (to first order) with increasing resistivity ρ in 3D and resistance per square R_{sq} in 2D. Moreover, their theory gave a good quantitative account of our data.

But there was more to the story. Bob Dynes, Bill McMillan, and their co-workers at Bell Labs were finding that the metal/insulator transition observed in amorphous Nb-Si alloys—a sibling to our amorphous Mo-Ge alloys—obeyed a scaling theory, including localization and Coulomb interactions. The scaling theory predicted a square-root cusp in the electron density of states $N(E) = N(0)\ [1 + (E/\Delta)^{1/2}]$ where N(0) goes to zero at the metal/insulator (M/I) transition. In the fancy language of renormalization group theory, as the system approaches the MI transition, it flows to the Mott insulator fixed point away from the Anderson localization fixed point. Clearly a tendency toward insulating behavior through Coulomb interactions was pulling T_c down.

In response to all this, Belitz and coworkers then generalized the famous McMillan equation for the T_c of strong coupling superconductors in terms of an experimentally accessible parameter $Y' = N_o/N(0) - 1$, which is a measure of the strength of the Coulomb interactions. Later, Mac's student Katherine Luna showed how, with experimental knowledge of Y' and the measured value of T_c, one could, with reasonable assumptions, back out the disorder-free T_{co} of the superconductor in question. More on all this below.

The KGB Era (1985–2020)

While all the work above was going on, Aharon Kapitulnik joined the faculty in applied physics. Early on, Mac and Aharon realized that the potential of Ted's amorphous Mo-Ge films for new physics had hardly been exhausted. Over the next several years with yet another group of students that included David Ephron, Mike Hahn, Monica Helerqvist, Whitney White, and Ali Yazdani, they studied the properties of these superconducting amorphous thin films in the region of the quantum phase transition at large R_{sq} where $T_c \rightarrow 0$. They were also able to study comprehensively the dislocation/anti-dislocation 2D vortex lattice melting transition predicted by the BKT theory for 2D superconductors in a magnetic field. Along the way, the Ted-Mac Amateur Hour evolved into the equally whimsically named KGB Group.

As is now legendary, in 1986 Bednorz and Mueller discovered superconductivity in the La-Ba-Cu-O system with transition temperatures up to 35K. Others rapidly confirmed their discovery, ushering in the era of high-T_c cuprates. Very soon after, by substituting Y for La, T_c was raised to above 90K. The early days were understandably chaotic; researchers discovered many new higher-T_c cuprates and carried out a wide range of initial measurements on poorly characterized materials. KGB was in the mix. To be fair, the stakes were high and the materials complex.

Matters progressed, however, and better characterized materials and increasingly reproducible experimental results eventually came to the fore. The KGB group made two particularly important contributions to this evolution, based on Ted's insight to use SrTiO3 as substrate for epitaxial growth of the cuprates. In the first contribution, the group demonstrated that largely grain-boundary-free films of YBCO could be deposited on SrTiO3 substrates using electron beam co-deposition in the presence of oxygen. Moreover, these films exhibited very high critical current densities, confirming by implication that grain-boundary weak links were limiting the critical current densities in bulk samples. This result strongly influenced the subsequent development of so-called deposited "coated conductors" for high-field, high-current density applications of the cuprate superconductors. In the second contribution, this time using co-sputtering, the group discovered the underdoped cuprate Y2Ba4Cu8O16 (sometimes called 248 or 124 YBCO), which in equilibrium is only stable at high pressure, a classic case of epitaxial strain mimicking high pressure. These two contributions clearly demonstrated the importance of epitaxial co-deposition in the study and applications of the cuprate superconductors.

In time, a phenomenological phase diagram for the hole-doped cuprates emerged. It displays a range of interesting regimes, going from the parent anti-ferromagnetic insulating phase, onto the high-T_c superconducting dome, and finally to a Fermi liquid regime at high doping. And not to forget a "strange metal" regime at higher temperatures above the superconducting dome.

Only a year before the discovery of high Tc cuprate superconductivity, at age sixty-five, Ted had yielded to the mandatory retirement requirement of Stanford at the time. Of course, he did not stop working; he only stopped taking on graduate students. Perhaps ironically, he made some of his most important contributions to this emerging phenomenological understanding during his emeritus period. He immersed himself in the literature on the cuprates, old and new, always with an eye to whether they confirmed or challenged the prevailing few. He was a relentless gadfly—in the honorific Socratic meaning—right up to his passing. In his last publication, which appeared in the year he passed, he and his fellow travelers laid out a passionate case that a class of extremely over-doped superconducting cuprates synthesized using high pressure oxygenation did not fit into the prevailing phenomenology—specifically that superconductivity persisted to doping levels beyond the usual superconducting dome of the standard phenomenological phase diagram.

But that's not all. Given any chance, Ted would remind us of his favorite maverick superconductors. Most prominently, these included candidates for negative-U pairing and a handful of so-called high temperature USOs (unidentified superconducting objects). Negative-U pairing is most associated with materials that exhibit charge disproportion (such as, ($2Bi^{4+} \rightarrow Bi^{3+} + Bi^{5+}$) that leads to an effective attractive electronic pairing interaction. Via an Air Force Office of Scientific Research (AFOSR) program courageously funded by Harold Weinstock through the Multidisciplinary Research Program of the University Research Initiative (MURI), a group of us joined forces to look at a few of Ted's mavericks. The team included Bob Cava, Gabi Kotliar, Ian Fisher, Emilia Morosan, and me (Mac), with Ted as a "kibitzing" collaborator. Ian also had a regular AFOSR grant for some of his work. To close this memoir, we describe two highlights from this work that speak to the issue of negative-U superconductivity.

The Bismuthate Superconductors

In 1975, Art Sleight discovered the first bismuthate superconductor (with a maximum T_c = 12K) by hole doping the parent compound $BaBiO_3$, a charge-disproportionated charge-density-wave (CD-CDW) insulator. The parent compound itself is interesting in that local density approximation (LDA) for electronic structure calculations predicts $BaBiO_3$ to be a conductor. Shortly thereafter, Cava and co-workers showed that T_c could be raised further to 37K using electron doping. These results have defied definitive understanding for three decades, in particular whether it is a negative-U superconductor or not.

The AFOSR MURI program identified two new critical pieces of the puzzle. Cesare Franchini had shown that an advanced, more complex density functional could account quantitatively for the insulating charge density state of $BaBiO_3$. The Kotliar group then applied this approach to the superconductivity of the bismuthates and found that it accounted quantitatively for the maximum T_c of 37K in electron-doped $BaBiO_3$. The physical interpretation of these results is that superconductivity observed here stems from a dynamical-correlation-enhanced electron-phonon interaction (that is, a new class of highly correlated superconductors). For hole-doped $BaBiO_3$, Katherine Luna and Paula Giraldo-Gallo of the Beasley-Fisher team found that as the doping decreased from high levels (such as in the metallic region), the material underwent a metal-insulator transition identical in character to that seen in amorphous Nb-Si alloys. Moreover, using the Luna procedure to extract the disorder-free T_{co}, we found T_{co} = 17K for hole-doped $BaBiO_3$, as compared to the observed maximum of 12K. In addition, no reduction of T_c was found for the electron-doped materials. Clearly, greater disorder exists on the hole-doped side of the phase diagram, which explains, at least in part, the observed electron-hole asymmetry.

Superconducting Tl-doped PbTe

PbTe is a narrow band-gap semiconductor that can be doped (electrons and holes) to reach carrier densities of order 10^{20} cm^{-3}. Remarkably, when doped with Tl (and so far, only Tl), it becomes superconducting with a T_c = 1.5K, stunningly high for such a low carrier-density material. Tl is known to exhibit charge disproportion ($2Tl^{2+} \rightarrow Tl^{1+} + Tl^{3+}$), which suggests that Tl-doped PbTe may be a negative-U superconductor. In support of this interpretation, Yana Matsushita, a student working with Ian, found that the temperature dependence of the resistivity of this material shows a logarithmic rise at low temperatures reminiscent of the Kondo effect, but in the absence of magnetic impurities. Working with theorist Jörg Schmalian, they interpreted this as evidence for a charge Kondo effect. This is a striking discovery. Moreover, the case for negative-U superconductivity is getting stronger, with evidence accumulating for the presence of the mixed Tl valence. Time will tell.

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Thus ends the remarkable career of a remarkable human being. His fingerprints are on many topics that have been and, in many cases continue to be, at the heart of condensed-matter physics. Perhaps even more important, he was among the pioneers of the field that, in the face of harsh criticism from some quarters that solid-state physics was "squalid-state physics," successfully brought us to the point where to seek new physics in so-called quantum materials is a rallying cry of condensed-matter and materials physics.

For his significant research and his leadership roles throughout his career, Ted won the Oliver E. Buckley Prize (jointly with Bernd Matthias) of the American Physical Society, the Von Hippel Award from the Materials Research Society, the Bernd T. Matthias Prize for new materials, and election to the National Academy of Sciences. In addition, at Stanford, the new advanced materials lab was named the Geballe Laboratory for Advanced Materials in his honor.

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