



Thoughts on the influence of Alex Mueller on high magnetic field technology

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ABSTRACT

I briefly summarize some of the impacts that Alex Mueller's drive to discover superconductivity in low carrier density oxides has had on high magnetic field technology, especially on the desire to use the cuprate superconductors to generate magnetic fields now almost twice those possible with the Nb-based low temperature superconductors. There were some important lessons for applications of any new exotic temperature superconductor that had to be learned before the Bednorz and Mueller discovery could really impact applications. Above all is the lesson that a high transition temperature is no guarantee of applications unless conductors with high current density can be manufactured. Recent reports and hopes for an ambient pressure room temperature superconductor remind us that the hopes for pervasive superconducting technologies germinated with the discovery of superconductivity in La-Ba-Cu-O and went truly ballistic following the discovery of YBa₂Cu₃O_{7-δ} (YBCO) with a 92 K transition temperature. Now that magnets made out of rare earth alloys of YBCO (REBCO) have entered routine service in NMR labs and prototype magnets made with REBCO appear ready to enable compact fusion reactors, we can finally say that Bednorz and Mueller's magnificent scientific achievement is having real world applications impact too.

1. Introduction

Superconducting wires were seen by that great applied physicist, Kamerlingh Onnes, as being almost immediately useful for the generation of powerful magnetic fields and his 1913 paper to the International Institute of Refrigeration in Chicago lays out the design of a compact 10 T magnet using Pb, Hg or solder-coated nichrome wires at reasonable cost [1]. Sadly his impeccable engineering design, still worth reading today, failed experimentally when he discovered that fields less than 0.1 T quenched the superconductivity of his wires [2] since, though he did not know it at the time, all were type I superconductors with critical fields less than 0.1 T. After fifty years of fascinating and often perplexing science with only minimal applications, interest in superconducting magnets suddenly exploded in early 1961 [3,4] when magnets made out of Nb₃Sn and Nb-Zr wires generated fields of 5–6.9 T [5]. The crucial progenitor of the magnets was the inspired metallurgy of Kunzler and colleagues who placed Sn inside a Nb tube, drew it into wire and heated it to ~ 1000 C to grow a Nb₃Sn layer inside the tube. With wires available, serious magnets came in less than 12 months! The field was fertile and by the end of the 1960s, superconducting magnets were everywhere and the construction of a 6.3 km circumference superconducting particle accelerator, the Tevatron at Fermilab, was well underway.

Alex Mueller's quest for a new superconductor occurred rather quietly, at least as I understand it. 1986 was the 75th anniversary of the discovery of superconductivity and Physics Today had a series of articles reviewing the achievements of the previous 75 years from both scientific and applications perspective [6]. What is really striking is that none considered the possibility of higher T_c materials, apart from brief mentions of stoichiometric Nb₃Ge or Nb₃Si. The Applied Superconductivity Conference in Fall 1986 had many retrospectives on the previous 75 years: From Brian Pippard [7], from Leon Cooper [8], from Gene Kunzler [4], and from Ted Berlincourt [9], who gave a wonderful paper on how Type II superconductivity was really discovered by Lev Shubnikov about 1934 and should not have had to wait until Kunzler's 1960 discovery [3]. My point in recalling these 75th anniversary celebrations is that Alex Mueller and Georg Bednorz were not at the meetings but in their lab looking to make a new class of higher T_c superconductors out of low carrier density oxides like BaPb_{1-x}Bi_xO₃ [10] that would take the field in completely new directions [11]. I do remember Bernd Matthias at a NATO summer School in 1980 being very skeptical about reports of superconductivity in BaPb_{1-x}Bi_xO₃ precisely because its low carrier density made superconductivity implausible based on the general characteristics of metallic superconductivity. His-somewhat derisory comment to the presenter of the work was that it was likely due to Pb-Bi grain boundary phase segregation. Clearly Alex Mueller and

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Georg Bednorz were more open-minded. In any case, their work appears to have been unknown to virtually all in the broader superconductivity community.

As another minor sidebar on their innovation, Don Liebenberg, a program manager at NSF, was concerned at the lack of new materials discovery in superconductivity. He approached Doug Finnemore of Ames Lab, Al Clark of NIST, Mac Beasley of Stanford, Mike Tinkham of Harvard and me about holding a workshop to explore new research opportunities in superconductivity and a workshop with this name was held at Copper Mountain CO in August 1983 and published in early 1984 [12]. When I visited the lab of Chevrel and Sergent, the main lab researching the ultra high field PbMo_6S_8 superconductor at the time ($H_{c2}(0)$ was about 60 T) at the University of Rennes in summer 1985 for a month [13], I was asked to talk about the workshop but other than that I felt that the report disappeared from view rather quickly. A student of mine working on Chevrel phase superconductors, Tomas Willis, kept us up to date on the latest work in superconductivity and often copied the first page of new papers. I remember a very coffee-stained page 1 of the paper, "Possible High Temperature Superconductivity in the Ba—La—Cu—O System" by Bednorz and Muller [14] for which the opening sentence, appropriately in quotations, was "At the extreme forefront of research in superconductivity is the empirical search for new superconductors [1]", where cited reference 1 was our report on the Copper Mountain workshop [15]. For a very long time, this was the only citation of our paper! The quote was indeed direct and at the 2006 M2S meeting in Dresden, I remember a question when I presented this poor citation record as an interesting example of some citations being more important than others, about who had written the memorable lines. I do know that it was not me: I tend to think that it was penned by Doug Finnemore or Mac Beasley. Rereading the workshop report I cannot see that we can claim any credit for Bednorz and Mueller's discovery. Indeed, as described in their Nobel prize lecture [11], the work was stimulated by Mueller's extensive prior work on perovskites and the curious fact that $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ could have a T_c of 13 K with low carrier density and low $N(E_F)$, a sign for him of a new type of interaction potential V much stronger than the electron-phonon interaction responsible for the T_c values of intermetallic A15 compounds like Nb_3Sn .

2. T_c above liquid nitrogen let the world dream of a hugely expanded superconducting future

Even if the applied superconductivity community and the Physics Today readership had not been prepared for Bednorz and Mueller's report, others immediately seized on it, especially in the group of Professor Tanaka at Tokyo University. At the Fall MRS meeting, Koichi Kitazawa of the Tanaka group reported an independent confirmation of the La—Ba—Cu—O result [16] which led to a huge acceleration of interest in the cuprates. Within a week (if I remember correctly) a higher T_c of around 40 K was demonstrated in La—Sr—Cu—O by Bob Cava at Bell Labs [17]. Almost immediately new materials synthesis efforts started up in many labs, including Houston, IBM, Argonne, and similarly in Japan, China, and Europe. At this stage the interest was primarily scientific, but the applications gear engaged immediately that the discovery of Wu and Chu of $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ [18] with T_c of 93 K was reported. A hugely magnified response by thousands worldwide led to immense interest in a superconducting future cooled by liquid nitrogen or even, since for a while T_c was rising very rapidly [19], to room temperature superconductivity. This led to intense debate in the US between members of the condensed matter and materials communities and the high energy physics community about whether investment in small-scale (materials) or large-scale (high energy particle accelerators) should be favored, a question polarized by the recent success of the superconducting Tevatron operating at 1 TeV at Fermilab where the dipole magnets steering the particle beam were made of $\text{Nb47wt}\%\text{Ti}$ wires, a material now in a class dubbed LTS (low temperature superconductors). Success of the Tevatron [20] had generated plans by the high energy

physics community to build a 20 TeV proton accelerator with an appropriately large budget [21]. This Superconducting Super Collider (SSC) was right in the middle of its design, scoping and budget phase as the exciting physics of perovskite cuprate high temperature superconductors (HTS) was developing. So great was the enthusiasm of proponents for HTS that the question of investment in superconductivity often degenerated into an HTS versus LTS competition.

I had a front row seat to this debate since Peter Lee and I were deeply involved in working out the microstructural origins of high critical current density J_c in $\text{Nb47wt}\%\text{Ti}$ in the service of allowing 8 T dipole magnets to be developed for the Superconducting Super Collider. Indeed we were able to double the current density of the Fermilab conductor and enable design of 8 T dipole magnets that shrank the size of the SSC ring by about 5/8 to what Tevatron magnets would have required, thus reducing the SSC ring circumference to "only" 87 km [22,23]. The important lesson of magnet design embedded in this story is that attainable critical current density J_c , not T_c , is the most vital property underlying large scale applications, above all for magnets, just as Onnes had shown in 1913.

So far as I know, Bednorz and Mueller never took a position on the proposition that soon divided the US physics community: if any SSC were to be built it should be built of HTS, not LTS! But many of their most intense admirers were less restrained.

The March 1987 American Physical Society Meeting program had started with a very small smattering of talks about La—Ba/Sr—Cu—O but the 93 K of YBCO led to an avalanche of requests for talks - the "Woodstock of Physics" - that went on for about 12 h, finishing only very early in the morning of the following day. I had a 2 min talk about 4 AM on the electromagnetically granular properties of polycrystalline YBCO powder in tube wire with very low J_c , barely above 100 A/mm², some 30–50 times smaller than in LTS Nb—Ti wires. My colleagues (Eric Hellstrom, Susan Babcock, Tom Kelly and others) and I had also looked to find evidence of oxygen deficiency at grain boundaries, already the likeliest explanation for the granular behavior. Our EELS studies with state-of-the-art HB501 STEM could not find any oxygen loss and were thus inconclusive as to the cause of the granularity. Interestingly, Mueller's studies of superconductivity in granular Al are discussed in Bednorz and Mueller's Nobel lecture: what interested Mueller was that the T_c of granular aluminum was approximately 2.5 times higher than pure Al, even if the T_c only rose to 2.7 K [11]. If only that were true for cuprates too!

A summer of reports and reviews of the potential of cuprates for applications ensued. A National Academy of Sciences Panel chaired by John Hulm, Director of Research at Westinghouse (and also the discoverer of superconductivity in Nb—Ti [24]), was established to sort through the huge excitement generated by Bednorz and Mueller's discovery. One of the key paragraphs of the Research Briefing reads [25]:

"The short-term problems and long-term potential of high-temperature superconductivity may both be easily underestimated. Given this potential and the current limited understanding of the new superconducting materials and their properties, it is essential that government, academic institutions, and industry take a long-term, multidisciplinary view. Since science and technology in this field are strongly intertwined, progress must occur simultaneously in basic science, manufacturing/processing science, and engineering applications. It is also important to maintain an open and cooperative international posture."

This paragraph was in fact remarkably prescient about the complexity and persistence needed to progress towards the grand goal of making HTS cuprates into conductors that could realize the huge application dreams of that moment. Indeed it can be said that major investments were made in superconducting science and technology by both the private and the public sector all over the world and especially for applications in the electric utility sector. Working transmission line, generator, motor, and fault current limiter prototypes were demonstrated in many countries [26]. In the US, starting in about 2010 such programs suffered major cuts as it became apparent that the path to

affordable conductor technologies was not obvious. Thus the successful technical arguments for replacing the copper and iron of electro-technology with superconductors began to fall away because of unsuccessful economic arguments.

3. The path to high current density cuprate conductors

Bednorz and Mueller's interest was, I believe, always primarily scientific rather than technological, though Alex Mueller did join the Technical Advisory Board of American Superconductor (AMSC) Inc at the invitation of Alex Malozemoff (where I was also a member). In this role Alex Mueller offered genial encouragement to the AMSC team in their quest to change the way that the world would use electricity. But the path to this goal was to make superconductors with high J_c and the practicalities of this task soon devolved principally to avoiding the superconducting granularity that impeded the high J_c circulating within each grain [27,28]. Starting with $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Bi-2223) with a T_c of 110 K, they developed a deep understanding of the complex mechanical and growth alignment processes needed to develop texture in rolled powder-in-tube tapes of the compound and so enabled many of the early prototypes [29]. But after about 15 years of hard work by AMSC and many others around the world, the inability to develop more than a uniaxial texture in Bi-2223 turned attention to tapes of biaxially aligned $\text{YBa}_2\text{Cu}_3\text{O}_{7-s}$ (or more generally REBCO, where the RE stands for rare earth). REBCO thin films not only had much higher intragrain critical current densities but with biaxially-aligned grains long length polycrystalline conductors without extensive grain boundary interruption became possible. These so-called coated conductors were first manufactured using the IBAD (Ion Beam Assisted Deposition) process by Iijima and colleagues in Japan [30]. This also stimulated much interest in mechanical deformation routes such as RABiTS to achieve texture [31] in the substrate. These developments were principally aimed at enabling electric utility applications. The fabrication difficulties of making the complex, multi-layer tapes required several years to debug but early in the new millennium very high current density coated conductors competitive with Nb—Ti and Nb_3Sn conductors in their low temperature domain started to appear [32]. As the next section shows this is what finally made possible the generation of magnetic fields above anything possible with the LTS conductors.

4. The emergence of HTS magnets in field domains impossible for LTS conductors

Although a variety of test coils using Bi-2223 and Bi-2212 were developed in Japan, largely in the service of advancing towards the a 1 GHz proton resonance NMR spectrometer which would require 23.5 T, the magnet success that really got my attention was the use of Bi-2212 tapes made by Oxford Superconducting Technology (OST) to generate a 5.11 T field when tested inside the 20 T large bore resistive magnet at the National High Magnetic Field Laboratory (NHMFL) [33]. For the first time HTS conductor had been used to generate a field, 25 T, quite impossible with any LTS conductor. The LTS magnet record then was about 22 T, a value only slightly exceeded when Bruker BioSpin achieved 1 GHz LTS NMR magnets operating at 23.5 T in about 2010, the still valid summit of LTS magnet records [34]. However, the fact that the whole conductor current density J_E was less than 100 A/mm² made these Bi-2212 conductors unattractive as the basis for a real magnet technology.

I arrived at the NHMFL in Fall 2006 with a mandate to get HTS materials into high field magnets as soon as possible. SuperPower had made a great success of the pilot production of REBCO coated conductor in 2007 and demonstrated a 1.5 T coil operating at 77 K at the summer DOE Peer Review. They gladly accepted our offer to test their magnet at 4.2 K in the 19 T large bore resistive magnet. To our great trepidation and happy surprise, the magnet generated a total field of 26.7 T at a tape current density J_E of 350 A/mm², almost 4 times that of the earlier

NHMFL/OST magnet made with Bi-2212 tapes [35].

With this demonstrated achievement of high current density and high fields, I was able to get the NHMFL to commit to an HTS magnet program too. Denis Markiewicz, an old Nb_3Sn tape hand from his former career at Intermagnetics General, enthusiastically embraced the idea of pancake winding REBCO tapes. We were able to make a series of small test coils with fields that ranged from 33.8 T in pancake form [36] to 35.4 T in layer wound form [37], all operating in the 38 mm cold bore cryostats of the 50 mm bore 31T Bitter magnets of the NHMFL. Progress was extremely rapid at this time as is summarized in a paper by Huub Weijers [38] to the 2009 Magnet Technology Conference from which I excerpt the following paragraph:

“This (earlier) rather evolutionary and steady advance was decisively broken by the very first REBCO coils which generated larger magnetic field increments with far less conductor [39,40], than earlier BSCCO coils. These coils feature winding current densities at least double the highest values observed in BSCCO coils, despite a nearly non-existent development activity specific to REBCO coils. At the time of the first test of the SuperPower/NHMFL coil, even the most basic high field data with which to reasonably predict coil performance was lacking, as were reliable coil construction procedures in this new high stress and high energy density domain. In fact this first SuperPower coil [40] had degraded conductor resulting from the overbanding applied during coil construction to protect the outer pancake joints. This damage did not however prevent the achievement of very high winding current densities of 275 A/mm² at almost 27 T. A subsequent GdBCO insert built by ISTECH [41] and operated in a narrow bore 28.3 T Hybrid magnet brought the maximum central field inside an HTS insert to 29.3 T; this too despite reported damage to the conductor during winding. A comparably sized YBCO insert was then constructed at the NHMFL and operated in a 31 T resistive magnet generating an additional 2.8 T at a remarkable winding current density of 440 A/mm² and stress levels of up to 380 MPa [36]. A group at Tohoku University, Sendai, operated spirals of YBCO coated conductor at 4.2 K under hoop stress levels of about 1 GPa [41]. In summary, YBCO coated conductors grown on strong substrates have clearly shown that superconducting magnets in the domain above 30 T are possible.”

This extremely rapid progress incited Denis Markiewicz and me to write a proposal to the National Science Foundation to build a 32 T magnet for NHMFL users. This was funded in 2010 [42] and the magnet was completed in 2017 [31]. It consists of a compact 15 T 25 cm bore LTS magnet made by the Oxford Instrument Company Ltd into which two REBCO magnets designed by Markiewicz and Weijers were placed. The magnet was successfully tested in 2017. It has now served many users. The NHMFL was of course not the only lab pursuing such high fields. The Sendai lab of Tohoku University has operated a 25 T hybrid magnet (Bi-2223 insert magnet inside a Nb_3Sn magnet) for many years that is now widely used for testing REBCO fusion coated conductors [43]. In parallel with our 32 T magnet was a major effort by Bruker BioSpin to use REBCO inserts to extend their champion 1 GHz LTS NMR magnet designs into first the 1.1 and then 1.2 GHz (28 T) regime [34]. Starting in 2021 they have delivered now almost ten of these NMR spectrometers that utilize HTS inserts generating about one quarter of the total field, making this the first genuinely commercial magnet development for HTS materials. Substantial R&D efforts to apply REBCO exist in Europe, Japan, China and the USA. Particularly to be noted is the large commitment made by Commonwealth Fusion Systems to making REBCO magnets a central technology for their SPARC prototype tokamak fusion reactor. In Fall 2021 they tested a prototype tokamak toroidal field test coil yielding a peak field of 20 T operating at 20 K [44]. This magnet used almost 400 km of 4 mm wide REBCO coated conductor, about 20 times that of all conductor purchased by the NHMFL for our HTS magnet program.

Finally I should note major efforts to develop Bi-2212 into a high current density, high field superconductor. Both Bi-2223 and REBCO are aspected tape conductors exhibiting significant mechanical and electromagnetic anisotropy and in this respect are not the first choice of LTS

magnet designers used to round, multifilament, twisted conductors. Bi-2212 was recognized early on as the HTS compound that allowed superconducting continuity in multifilament round wire geometries but as noted in the 25 T NHMFL/OST test coil [33], the actual wire current densities were high enough to generate a new record field but not high enough to sustain real applications. On arriving at the NHMFL, my colleagues and I decided to follow up and understand why round wire, multifilament Bi-2212 wires could develop significant grain-to-grain connectivity despite their apparent lack of texture. We teamed up with the magnet group at LBL who had always expressed interest in Bi-2212 because of its round wire form which allowed the Rutherford cable almost universally used for accelerator dipole and quadrupole magnets. A still -running joint effort between the LBNL magnet group and our efforts in the Applied Superconductivity Center at the NHMFL has been able to develop Bi-2212 to the state that whole wire J_c values of more than 800 A/mm² at 30 T and 4,2 K have been demonstrated [45,46]. Indeed we regard this conductor as now being ready for commercial introduction and ongoing efforts to make general purpose 25 T magnets with Oxford Instruments and Cryomagnetics have this as a goal. A key aspect of Bi-2212 wires as opposed to REBCO coated conductors is that they are significantly more stable than Nb₃Sn wires and thus do not exhibit the unfortunate “training” behavior of many Nb₃Sn coils, especially those of dipole or quadrupole geometry [47]. But they are also not so stable that local loss of superconductivity, for whatever reason (fluctuations in longitudinal properties of the single filament, sudden damage, or other local heating) can cause intense local heating or even burn out [48]. Indeed one can say that the need to effectively current share in magnets so as to effectively protect against quench burn out remains one of the major unresolved issues in making high field magnets from REBCO coated conductors. In this respect the inherent current sharing of round, multifilament, macroscopically isotropic Bi-2212 offers a distinctive array of advantages provided that the complexity of its wind-and-react processing can be tolerated [45,46].

As a final note, I happily acknowledge the collaborations started with Professor Seungyong Hahn, originally Professor of Mechanical Engineering in the FAMU/FSU College of Engineering and now Professor at Seoul National University. His advocacy of the “No Insulation” coil winding technique for REBCO magnets as an important enabler of current sharing and thus more quench tolerant behavior enabled very compact, high current density test magnets to be built, culminating in a series of three so-called Little Big Coils, the third of which reached 45.5 T when tested inside the NHMFL 31 T magnet [49]. Operating at over 1400 A/mm² J_E at 45.5 T, these coils self-damaged due to their extremely high hoop stresses, amplified by large screening current stresses generated by the tape geometry [50]. However, a recent coil generated 44.5 T with much less damage and thus hopes for generation of 50 T or even higher magnets may not be unreasonable. Indeed a project to design a 40 T user magnet is under way at the NHMFL [51], and similar efforts are now underway in Europe and Asia too. In short, even if the effort needed to make Bednorz and Mueller’s cuprates suitable for magnets has suffered delays and setbacks, the wonderful properties offered by the cuprates have sustained the large efforts needed to make them capable of generating magnetic fields now almost twice as high (45.5 T versus 23.2 T) as is possible with LTS conductors.

5. Closing words

As I write this in summer 2023, it is 37 years since submission of Bednorz and Mueller’s epochal article on a “possible” high temperature superconductor. In retrospect we can see that their restrained claims were in many ways a very appropriate counterpoint to the over-reaching claims of instant applications made by so many others in the first 3 or 4 years after the discovery of YBCO, the first liquid nitrogen cuprate. But the last 5 years have seen that real, significant applications are finally flowing, including BioSpin’s 1.2 GHz NMR systems, the 32 T NHMFL and the 25 T Tohoku user research magnets. And if, as Commonwealth

Fusion Systems, Tokamak Energy and many others hope, the cuprates enable pervasive and affordable fusion power, Alex Mueller’s drive to discover new low carrier density oxide superconductors will exert an even more beneficial impact on the world’s future. Indeed as we move towards ever greater electrification of the world, it seems possible that driving production costs of cuprates down as happened 40 years ago for Nb—Ti may make REBCO coated conductors pervasive and essential to the electric power industry, the original driver for applications flowing from Bednorz and Mueller’s great discovery.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

This long historical reminiscence has reminded me of many colleagues and collaborators who participated in the developments briefly sketched here. I have noted several by name in the article, especially my senior colleagues in the Applied Superconductivity Center in Madison and later in Tallahassee who were partners in our programs to develop and then apply conductors to magnets. Ulf Trociewitz added hugely to this effort, especially in Bi-2212 magnets, when I came to the NHMFL. Lance Cooley now nurtures these programs as present ASC director. Underpinning all of this work was continuing and strong support from our program monitors at the Department of Energy, especially Dave Sutter, Jim Daley, Bruce Strauss and Ken Marken. In particular I cherish the interactions arising from two vital collaborations nurtured by these program managers, the now 40 year-running Low Temperature Superconductor Workshop (originally the Nb-Ti Workshop) and the Wire Development Group collaboration for which Alex Malozemoff of American Superconductor was the lead. They all took a long view of the potential of the cuprates and thus played an essential role in sustaining work on both LTS and HTS materials through many setbacks and they enabled support for more than 60 PhDs, as well as many MS and BS students. Finally I express my gratitude for present support to the Department of Energy Office of High Energy Physics (DE-SC0010421 and SC0012083) and Office of Fusion Energy Sciences (DE-SC0022011), to Florida State University for a discretionary support for high field HTS technology development and to the NHMFL, which is supported by NSF under Award Number DMR-2128556, and by the State of Florida.

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