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SUPERCONDUCTIVITY AND LATTICE INSTABILITIES.(U)
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SUPERCONDUCTIVITY AND LATTICE INSTABILITIES

CUMULATIVE WORK STATEMENT

F44620-72-C-0017

a) Perform electric and magnetic measurements on metals and alloys.

b) Determine the cause of metastabilities in metallic systems.

c) Evaluate the effect of high pressures on the magnetic nature of the metallic state.

d) Measure the influence of strain and its release on the superconducting transition temperature.

e) Verify to the lowest temperature our hypothesis of superconductivity being the normal metallic behavior.

f) Conduct investigations into the synthesis of unstable, metastable and stable metallic phases.

g) Discover systems which will lead to metals having higher superconducting transitions.

h) Exploit and expand recently discovered ternary compounds and phases of superconductors.

i) Determine the cause and prevention of metastabilities in metallic systems.

j) Explore the techniques of cladding, recrystallization by vapor catalysis, and high pressure for stabilizing metallic phases with higher superconducting transition temperatures.

k) Explore the existence of metal-metal solid solution series.

l) Develop the search for new ternary superconducting compounds.

m) Discover new materials systems which will lead to more useful superconductors with enhanced physical, crystallographic and metallurgical properties.

n) Measure and characterize the superconducting properties of these materials.

o) Synthesize new ternary, pseudobinary and quaternary metallic superconducting compounds.

p) Increase the critical transition temperature of these materials.

q) Study non cubic crystal structures with high critical temperatures.

r) Investigate lattice instabilities and metastable phases in these superconducting materials.

s) Measure and characterize the superconducting properties of these materials.

(1) During the past five years we have discovered many new superconductors. Of these, the most important are lithium titanate, the Chevrel phases, new superconductors with the eta-carbide structure and new superconductors with the sodium-chloride structure.

Lithium titanate, when prepared with the spinel crystal structure, is superconducting at temperatures as high as 14 K.[(1)] This is the highest transition temperature known for any spinel structure compound, or, for that matter, any oxygen based compound. Lithium titanate also has the lowest known density of any potentially useful superconductor.

Lithium titanate seems to be one of those rare examples of ideosyncratic superconductivity in the sense that there exist no other superconducting compounds which are related both chemically and crystallographically. For this reason, all attempts to raise its transition temperature by means of chemical perturbation have failed, and for the same reason, we are at a loss to explain why the superconducting transition temperature should be so high in the first place. The superconductivity of lithium titanate thus illustrates what has become an important principle in the search for high temperature superconductors, namely that the existence and transition temperature of ternary compound superconductors cannot be predicted on the basis of the rules which have worked so well in the past for binary compound superconductors.

As it always has been, the search for new superconductors remains an empirical process.

Very important new ternary compound superconductors have been found among the so-called Chevrel phases.[2, 3] These compounds may be thought of as highly distorted cesium chloride type structures, with one type of "atom" consisting of clusters of molybdenum and either sulfur, selenium or tellurium, while the other atom can be almost anything, e.g., copper, silver, tin, lead, zinc, cadmium, or one of the alkaline earths or even rare earths. So far, the highest transition temperatures are found for lead-molybdenum-sulfide at 15 K. While these temperatures, of course, are not the highest known today, the superconducting Chevrel phases do have enormous critical fields - as high as 700 kgauss.[4] If these materials turn out to have favorable critical currents and mechanical properties, they will no doubt have important high field applications.

We have found that the Chevrel phases also have other properties which have never before been observed for any class of superconducting materials. One of these is the extraordinary sensitivity of their transition temperatures to applied hydrostatic pressure, which is the highest yet observed and is often nonlinear.[5] It is believed that the origin of these effects is related to the fact that these compounds seem to be only marginally stable from a crystallographic point of view.

Another remarkable property of the Chevrel phases is their insensitivity to the incorporation of magnetic rare earth atoms into the crystal lattice. While for almost all other known superconductors the presence of a few atomic percent of, say, gadolinium would have a disastrous effect on the transition temperature, the effect of concentrations as high as seven percent seem to be very small in the Chevrel phases. For example, praseodymium-molybdenum selenide is superconducting well above 9°K and even gadolinium-molybdenum-selenide is superconducting near 6°K.[6] In fact, the onset of magnetic order does not adversely affect the superconducting properties of the rare earth molybdenum selenides, and the magnetically ordered state seems to coexist quite happily with superconductivity at low temperatures.[7] This state of affairs has only recently been discovered, and we do not yet have a full explanation; it is not unreasonable to expect that the phenomenon is somehow connected with the extraordinarily high values of the critical fields of the Chevrel phases.

However, the greatest challenge presented by the discovery of superconductivity in the Chevrel phases is its unpredictability. As with lithium titanate, there are no rules, such as the old electron concentration rules, for predicting the existence or transition temperature of superconductivity in these phases. With all the remarkable properties of the superconducting Chevrel phases, one would like to think that the process of discovery would be greatly accelerated by the application of new rules - which themselves are as yet undiscovered.

The superconductivity of the Chevrel phases has, of course, stimulated our interest in all kinds of chalcogenide materials. Many of the new superconductors discovered during the past five years have the sodium chloride structure. Among these we mention thorium-sulfide, thorium-selenide, scandium-sulfide, scandium-selenide and the related phosphides, zirconium phosphide, hafnium phosphide and thorium phosphide; the highest transition temperature (4.8 K) is observed for zirconium-phosphide.[8] Transition temperatures up to 7 K were also observed in the pseudobinary system silver-tin-selenium with the sodium chloride structure.[9]

New ternary superconductors with the "eta-carbide" structure were found in the systems titanium-platinum-oxygen (2.5 K), zirconium-platinum-oxygen (5.4 K), hafnium-platinum-oxygen (2.2 K), niobium-zinc-carbon (4.6 K) and zirconium-vanadium-oxygen (5.5 K).[10]

The most interesting compound in this system is scandium-chromium-boron. Scandium and chromium by themselves do not even interact but with a few percent of boron as an impurity suddenly they form a superconducting intermetallic compound with transition temperatures in excess of 7°K.[11]

(2) During the past five years we have obtained overwhelming evidence for a connection between moderate to high transition temperature superconductivity and low temperature structural instability.

Five years ago, only the two beta-tungsten structure superconductors niobium-tin and vanadium-silicon were known to undergo low temperature phase transitions and most people thought that these transformations were caused by properties unique to the beta-tungsten crystal structure. Now, on the contrary, we have shown that low temperature structural transformations occur in many superconductors with crystal structures entirely unrelated to the beta-tungsten structure. Table I shows the examples of this behavior which we have discovered here.

Table I

Material	Transition Temperature (°K)	Room Temperature Crystal Structure	Transformation Temperature (°K)	Low Temperature Crystal Structure	References
HfV_2	8.4	Cubic Laves phase	118	Orthorhombic	12, 13
ZrV_2	7.8	Cubic Laves phase	121	Rhombohedral	12, 14
$LaRu_2$	4.5	Cubic Laves phase	30	Tetragonal	15
$CuMo_3S_4$	10.9	Chevrel phase	260	Unknown	16
$CuMo_3Se_4$	5.9	Chevrel phase	123	Unknown	17
$ZnMo_3S_4$	3.0	Chevrel phase	57	Unknown	17

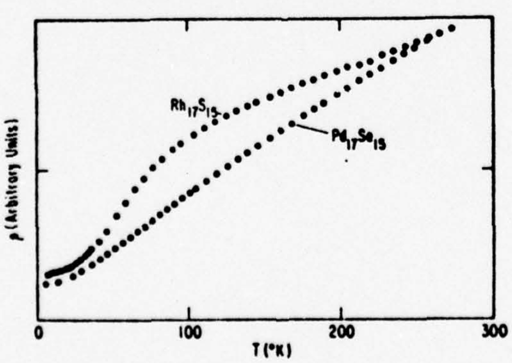
These compounds constitute approximately one third of all structurally unstable superconductors known today, and the discovery of their instability has contributed greatly to the current conventional wisdom that high temperature superconductivity and structural instability are intimately related.(18)

It was expected that the application of hydrostatic pressure would have rather dramatic effects on both the superconductivity and the transformation properties of structurally unstable superconductors. Pressure studies have been made on the systems vanadium-silicon,[19] hafnium-vanadium,[20] zirconium-vanadium,[20] copper-molybdenum-sulfide,[5] copper-molybdenum-selenide,[21] zinc molybdenum sulfide,[5] lanthanium sulfide,[22] lanthanium selenide,[22] vanadium-ruthenium,[23] and gold-zinc.[24] In every case the effects of pressure have been substantial (in some cases quite dramatic), but taken in the aggregate, the behavior of structurally unstable superconductors does not seem to follow any simple rule.

The effect of hydrostatic pressure has also been determined for most of the known Chevrel phases. In general, the effect is larger for this class of compounds than any other. We have concluded from these experiments that the superconducting Chevrel phases which do not show structural transformation have at least a latent instability. This point of view has been confirmed by recent Mössbauer spectroscopy[25] on tin-molybdenum-sulfide and neutron spectroscopy[26] on tin-molybdenum-sulfide and lead-molybdenum-sulfide.

(3) We have developed an empirical correlation between the shape of the electrical resistivity versus temperature curve of a given superconductor and its transition temperature.

During the past five years we have measured the electrical resistivity as a function of temperature for many metallic compounds, both superconducting and non-superconducting. As a result of these measurements we have found that, provided one of the constituents is a transition metal, a strong correlation exists between the shape of the resistivity-temperature curve and the transition temperature of the compound under study. Briefly stated, the correlation is that the resistivity-temperature curves of transition metal non-superconductors are always concave upward (positive curvature), while the resistivity-temperature curves of transition metal superconductors are always concave downward (negative curvature).[27] As an example, the resistivity-temperature curves for isostructural rhodium-sulfide and palladium-selenide are shown below. Rhodium-sulfide is superconducting at 5.8 K and shows strong negative curvature, while palladium-selenide is not superconducting above 1.0 K and shows hardly any curvature at all.

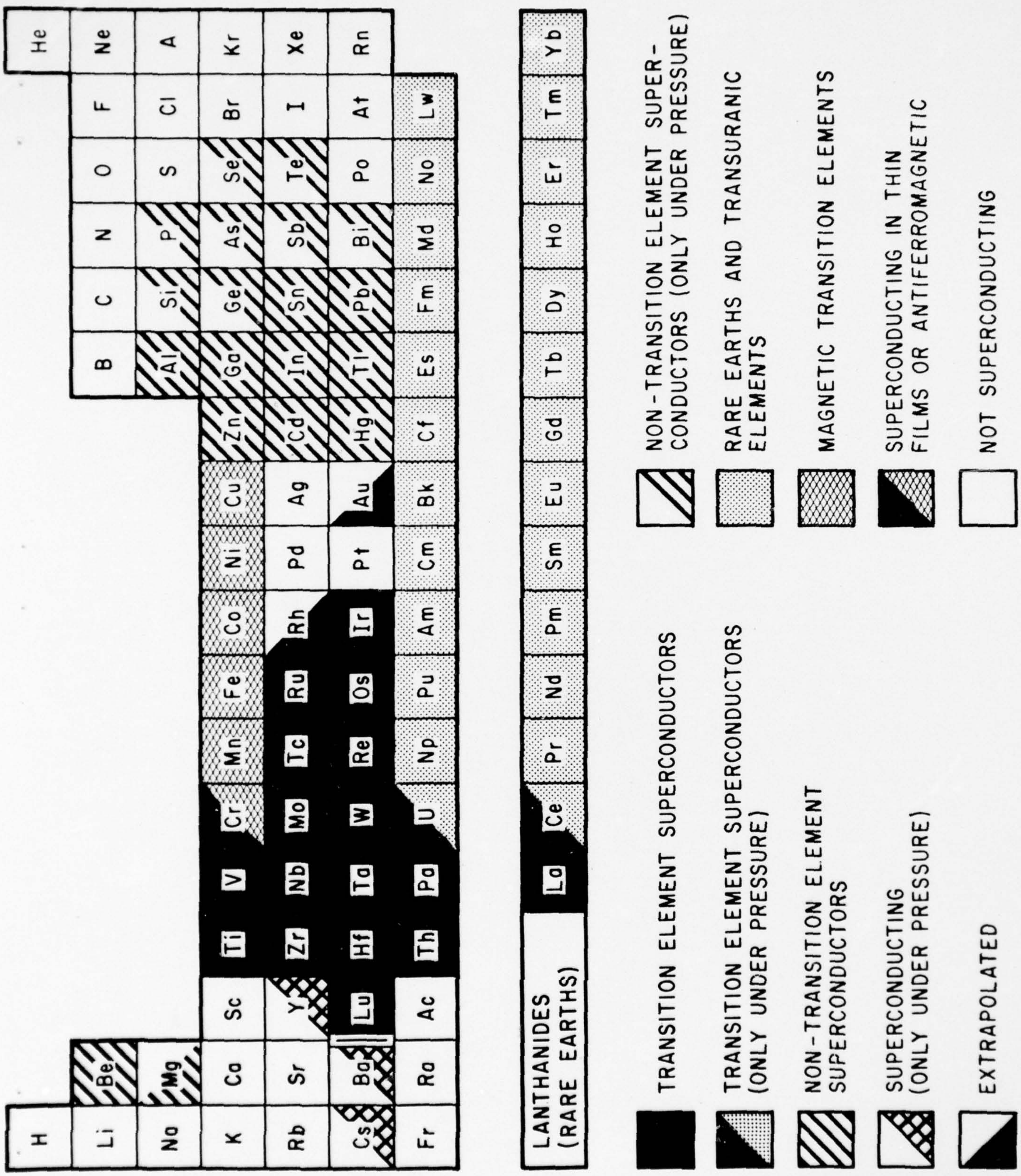


Further examples include the copper dialuminide phases (zirconium-rhodium, zironcium-iridium, zirconium-nickel and zirconium-cobalt) where correlations with specific heat data are obtained,[28] and alloys between the cubic Laves phases lanthanum-ruthenium and cerium-ruthenium for which the superconducting transition temperature and the degree of curvature are strongly correlated.[29]

Our discovery that the negative curvature of the resistivity-temperature curves of transition metal superconductors is an extremely general phenomenon has provoked a great deal of theoretical discussion. The results are not at all explained by established theories for the electrical resistivity (Bloch-Grüneisen theory, as modified by Wilson), and further modifications of the theory to include temperature induced motion of the Fermi level or lattice softening at low temperatures have not led to agreement between experiment and theory. Since the electron-lattice interaction which gives rise to the existence of electrical resistivity is also responsible for superconductivity, we feel that the puzzle of negative curvature in the resistance-temperature curves of superconductors is an important one which should not be allowed to remain unsolved. This is especially true since the measurement of electrical resistivity is a relatively simple one and can be performed on almost all materials -- and what is probably one of the most crucial features -- at temperatures far above the superconducting transition

(4) We have extrapolated to the transition temperatures of magnesium and gold. These experiments consisted of measuring the superconductivity of highly purified alloys of magnesium-cadmium, gold-aluminum, gold-indium and gold-gallium at temperatures as low as 0.007 K and extrapolating to the values of the transition temperature of pure magnesium and pure gold. As a result, we predict that pure magnesium will become superconducting at 0.0004 K,[30] and pure gold will do so at 0.0002 K.[31] Of course, these predictions will be fulfilled only when such low temperatures can be reached, and sufficiently pure samples of these elements are available.

The value of these low temperature experiments is that our understanding of superconductivity with respect to the periodic system - our only real guidance in the search for high temperature superconductivity - is thereby extended.


H He
Li Be B C N O F Ne
Na Mg Al Si P S Cl A
K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Br Kr
Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd In Sn Sb Te I Xe
Cs Ba Lu Hf Ta W Re Os Ir Pt Au Hg Tl Pb Bi Po At Rn
Fr Ra Ac Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lw
LANTHANIDES (RARE EARTHS)
La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb
TRANSITION ELEMENT SUPERCONDUCTORS
TRANSITION ELEMENT SUPERCONDUCTORS (ONLY UNDER PRESSURE)
NON-TRANSITION ELEMENT SUPERCONDUCTORS
SUPERCONDUCTING (ONLY UNDER PRESSURE)
EXTRAPOLATED
NON-TRANSITION ELEMENT SUPERCONDUCTORS (ONLY UNDER PRESSURE)
RARE EARTHS AND TRANSURANIC ELEMENTS
MAGNETIC TRANSITION ELEMENTS
SUPERCONDUCTING IN THIN FILMS OR ANTIFERROMAGNETIC
NOT SUPERCONDUCTING

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Ph.D.'s supervised by Professor Bernd T. Matthias--Physics Department--
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Brian C. Sales	Valence Fluxuations on Rare Earth Ions November 23, 1974
David C. Johnston	Superconducting and Normal State Properties of Several Ternary Titanium Oxides January 6, 1975
Arnold R. Moodenbaugh	Superconductivity of Some NaCl Structure Sulfides, Selenides and Phosphides July 1, 1975
Robert N. Shelton	Investigation Into the Effect of High Pressure on Superconducting Ternary Molybdenum Chalcogenides December 6, 1975

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COUPLING

Contract: AFOSR F44620-72-C-0017 Date: 10/1/75 - 9/30/76

1. High Transition Temperature Superconducting Materials

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies.

   c. National Bureau of Standards, Washington, D. C. New superconducting critical temperatures and fields.

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies.

   c. Distinguished Lectures, Joint Center for Materials Science including Los Alamos Scientific Laboratory, Sandia Laboratory, Air Force Weapons Laboratory plus University of New Mexico, New Mexico Institute of Mining and Technology and New Mexico State University. Superconducting materials, their use now and applications for the future.

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies.

   c. U. S. Army Symposium on Ultra High Pressure Phenomena, Rensselaerville, N. Y. Applications of ultra high pressures to superconductors and the applications of the resulting new superconducting materials.

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies, including two branches of DOD (Army and Navy).

   c. Executive Technical Development Program. Office of Special Programs, Polytechnic Institute of New York. General discussion on superconductivity, ferroelectricity and magnetism.

2. Other Topics

a. Bernd T. Matthias, Zachary Fisk and A. C. Lawson, Univ. of Calif., San Diego

b. The International Conference on Low Lying Lattice Vibrational Modes and Their Relationship to Superconductivity and Ferroelectricity, San Juan, Puerto Rico

c. Phase transitions and their role in ferroelectricity, superconductivity and magnetism.

a. Bernd T. Matthias, Zachary Fisk and A. C. Lawson, Univ. of Calif., San Diego

b. 1976 d- and f-band Conference, Rochester, New York

c. New magnetic superconductors.

a. Bernd T. Matthias, University of California, San Diego

b. 1976 Applied Superconductivity Conference, Stanford University, Stanford, California

c. New superconductors, their applications now and in the future.

COUPLING

Contract: AFOSR F44620-72-C-0017 Date: 10/1/74 - 9/30/75

1. High Transition Temperature Superconducting Materials

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies

   c. Los Alamos Scientific Laboratories, Los Alamos, New Mexico--The problem of improving the basic properties of present superconducting materials for use in superconducting cables continues being discussed with various groups of researchers at Los Alamos Scientific Laboratories. These discussions have resulted in the development of a new method of synthesis for Nb_3Ge in bulk form with transition temperatures of 22.5°K. A major part of the coupling is, unfortunately, at present available only in classified form.

   a. Bernd T. Matthias, University of California, San Diego

   b. Conference

   c. 1974 Applied Superconductivity Conference, Oakbrook, Illinois. New superconducting critical temperatures and fields.

   a. Bernd T. Matthias, University of California, San Diego

   b. Picatinny Arsenal, John W. Gregorits and Leon W. Saffian; Naval Shipyard, Research and Development, Robert J. Wolfe; CIA, Corroll W. Morrow, and other government agencies, training program.

   c. Executive Technical Development Program. Office of Special Programs, Polytechnic Institute of New York. General discussion on superconductivity, ferroelectricity and magnetism.

   a. Bernd T. Matthias

   b. Conference and interaction with various other agencies.

   c. American Physical Society Meeting, Anaheim, California. Applications of superconductors.

   a. Bernd T. Matthias

   b. Invited lecture

   c. Naval Research Laboratory, Washington, D. C. New superconductors and instabilities.

Coupling

Date: 10/1/74 - 9/30/75

a. Bernd T. Matthias

b. Invited lecture

c. Applied Physics Colloquium, Johns Hopkins University, Silver Spring, Maryland. Properties and applications of superconductors.

a. A. C. Lawson

b. Colloquium

c. Pomona College, October 1974 High Transition Temperatures and Lattice Instabilities

2. Other Topics

a. Bernd T. Matthias, University of California, San Diego

b. Conference

c. Resource Conference on Cu, University of California, San Diego. Copper shortage.

a. Bernd T. Matthias

b. Colloquium

c. Los Alamos Scientific Laboratories, Los Alamos, New Mexico Physics of plutonium.

a. Bernd T. Matthias

b. Conference, Leuven, Belgium (Keynote speaker)

c. This conference was a report to ERDA on the electronic properties of solids under high pressure.

a. Bernd T. Matthias

b. Colloquium

c. Los Alamos Scientific Laboratories, Theoretical Division, classified.

a. Zachary Fisk

b. sample preparation

c. Nb_3Sn samples for specific heat measurements, Dr. Gordon Knapp, Argonne National Laboratory Argonne, Illinois

a. Zachary Fisk and A. C. Lawson

b. collaboration and x-ray measurements

c. Search for Crystallographic Instability in Hexagonal Tungsten Bronzes, H. R. Shanks, Ames Laboratory USAEC, Ames, Iowa

a. D. C. Johnston, A. R. Moodenbaugh and A. C. Lawson

b. sample preparation and collaboration

c. Professor John Cannon, Brigham Young University, Provo, Utah synthesis of new superconductors under high pressures.

a. R. N. Shelton

b. Talk at meeting

c. APS General Meeting, Denver, Colorado March 1975 Pressure Dependence of the Superconducting Transition Temperature for Ternary Molybdenum Sulfides

COUPLING

Contract: AFOSR F44620-72-C-0017 Date: 10/1/73 - 9/30/74

1. High Transition Temperature Superconducting Materials

   a. Bernd T. Matthias, University of California, San Diego

   b. Consultation with other government agencies

   c. Los Alamos Scientific Laboratories, Los Alamos, New Mexico--The problem of improving the basic properties of present superconducting materials for use in superconducting cables have been discussed with various groups of researchers at Los Alamos Scientific Laboratories. These discussions have resulted in the development of a new method of synthesis for Nb_3Ge in bulk form with transition temperatures of 22.5°K.

   a. Bernd T. Matthias, University of California, San Diego

   b. Symposium

   c. Marshall Space Flight Center, Huntsville, Alabama--Discussed the role of superconducting metallurgy in zero gravity fields.

   a. Bernd T. Matthias, University of California, San Diego

   b. Conference

   c. American Academy of Arts and Sciences, San Francisco, Materials Achievements in the Enhancement of Our Health, Safety and Environment: Looking Ahead--Discussed the future difficulties encountered in energy and ecology which can be alleviated to a large extent by a superconducting technology.

   a. Bernd T. Matthias, University of California, San Diego

   b. Conference

   c. Israel Scientific Research Conferences, Haifa, Israel--Discussions including the final "putting to rest" of any high temperature organic superconductivity.

2. High Critical Fields in Superconductors

   a. Bernd T. Matthias, University of California, San Diego

   b. Conference

   c. Grenoble, France--The high critical fields of the new sulfides are now being verified from Europe to the United States. As a consequence, there may be rather decisive changes in the future applications of superconducting materials.

COUPLING

Contract: AFOSR-F-44620-72-C-0017 Date: 10/1/72 - 9/30/73

1. Superconductivity
   a. Lt. Col. Edward Daggit, U. S. Army Armament Command, Rock Island, Ill.; Allen M. Moss, Theodore W. Stevens, Robert Leonardi, U. S. Army, Picatinny Arsenal, N. J.; Raymond F. Siewert and Samuel L. Taffel, Naval Air Systems Command.
   b. Discussion
   c. Executive Technical Development Program, Office of Special Programs, Polytechnic Institute of New York. General discussion on the applications of superconductivity

2. Superconducting Accelerators
   a. Los Alamos Scientific Laboratories, Inauguration of the Linear Accelerator
   b. Presentation and discussion
   c. The problems involved in the application of superconductors for magnets and cavities.

3. Applications for High Transition Temperature Superconducting Materials
   a. ARPA Workshop for Superconducting Technology
   b. Presentation and discussion
   c. A major part of the meeting was taken up by arguments on "organic superconductors." In the meantime, the then prevailing skepticism has been more than justified.

COUPLING

Grant No: AFOSR-F44620-72-C-0017 Date: 10/1/71 - 9/30/72

1. Crystallographic Instabilities and High Temperature Superconductivity

   a. Professor Bernd T. Matthias; University of California, San Diego

   b. Testimony before Subcommittee on Science, Research and Development for hearings on Energy Research and Development, Rayburn House Office Bldg., Washington, D. C. May 23, 1972 "Electric Power Transmission: The Use of Superconductors"

   a. Professor Bernd T. Matthias; University of California, San Diego

   b. Conference: National Commission on Materials Policy, Lake Arrowhead, California June 12 - 16, 1972 "Materials for Electricity and Magnetism, Advanced Material Usage in the Southwest"

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During the past five years ~~we have made~~ have been made four distinct advances in the science of superconductivity: (1) ~~We have discovered~~ many new superconductors were discovered, including lithium-titanate (T_c = 13.7K) and numerous Chevrel phases (T_c = 15.2K lead-molybenum-sulfide). The latter are remarkable not only for their extraordinarily high critical fields, which are close to 700 kg in some cases, but also for the fact that really high critical temperatures have for the first time been discovered in non-cubic materials. ~~At the same time, the old electron-per-~~

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atom rules do not seem to work for the Chevrel phases. We have also discovered superconductivity in many related oxides, sulfides and selenides. (2) We have obtained overwhelming evidence for a connection between moderate-to-high-temperature superconductivity and low temperature structural instability. Our work on both the occurrence and the pressure dependence of superconductivity and lattice instability in zirconium-vanadium, hafnium-vanadium, lanthanum-ruthenium, ruthenium-vanadium, copper-molybenum-sulfide, copper-molybenum-selenide, zinc-molybenum-sulfide, gold-zinc, lanthanum-sulfide, and lanthanum-selenide has shown that the connection between the two phenomena is very general. (3) We have developed an empirical correlation between the shape of electrical resistivity versus temperature curve of a given superconductor and its transition temperature. Despite our incomplete understanding of its nature, we have found this correlation useful in predicting superconductivity in some cases. (4) We have extrapolated to the transition temperatures of magnesium and gold (5 x 10-4 and 2 x 10-4K, respectively). This information is fundamental for our understanding of the periodic system vis-a-vis superconductivity.