IN THE UNITED STATES PATENT AND TRADEMARK OFFICE BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

WU et al.

Interference No. 102,447

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\$ Examiner-in-Chief
\$ Ronald H. Smith

DECLARATION OF RULING MENG

Box Interference Commissioner of Patents and Trademarks Washington, D.C. 20231

Attention:

Ronald H. Smith Examiner-in-Chief

Dear Sir:

- I, RULING MENG, do hereby declare and state that:
- I received a B.A. degree in Material Science from the Mining and Metallurgy College in Hunan, China in 1958. receiving my degree, I have worked as an instructor at the Mining and Metallurgy College, Hunan, China (1958-59); a Research Assistant, Metallurgy and Material Science Division, Institute of Mining and Metallurgy, Chinese Academy of Science (1959-73); and a Research Associate, Institute of Physics, Chinese Academy of Science (1973-84) during which time I also held visiting positions as a Research Associate, Department of Physics at University of Houston, Texas (1979-81), University of Konstanz, West Germany (1981-82), and University of Houston, Texas (1984-86). currently a Senior Research Scientist, and Lab Manager of the Experimental Solid State Physics Lab of the Department of Physics, University of Houston, Texas and of the Texas Center for Superconductivity; a position I have held since 1987. 79252/1/DECLARATION-MENG

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that, during the period 1984 to 1986, I held the position of Research Associate, Department of Physics at the University of Houston, Texas, working at the direction and under the supervision of Dr. C.W. (Paul) Chu.

- 2. I first began to make and examine compositions composed of rare earth metal-alkaline earth metal-copper-oxygen for superconducting properties in November 1986. Dr. C.W. (Paul) Chu under whose directions and supervision I worked directed me to make compositions of La-Ba-Cu-O to a nominal formula as described in Bednorz & Muller, Z. Phys. B.-Condensed Matter, 64, pp 189 193 (1986) and then test such compositions for the existence of the properties of a superconducting material -- namely, the exhibition at or below a certain critical temperature (T_{c1}) of zero electrical resistance and a diamagnetic property.
- 3. During November 1986 I prepared several samples of La-Ba-Cu-O compositions to a nominal formula of La_{4.25}Ba_{0.75}Cu₅O_{5(3-y)}, as disclosed by the Bednorz & Muller article. Each sample thereof was prepared under various firing conditions, as recorded in my handwriting in Exhibit A at or about the time each such sample was prepared, and thereafter tested by me, or tested by others under my direction and supervision, to measure the sample's electrical resistance at various temperatures from that of about ambient and progressing to the boiling point of liquid helium which is about 4°K. Further, samples which exhibited the attainment of zero electrical resistance at or below a certain critical temperature were measured for the exhibition of a diamagnetic effect by me, or

by others working under the direction and supervision of Dr. Chu and myself.

- 4. My work with the La-Ba-Cu-O compositions, as described in paragraphs 2-3 above confirmed that a composition described by Bednorz & Muller as one of nominal formula La_{4,24}Ba_{0,75}Cu₅O_{5(3-y)} did, in fact, possessed superconducting properties even though I produced my replicas, as directed by Dr. Chu, by a solid state reaction technique rather than by a precipitation technique as described by Bednorz & Muller.
- During the remainder of November of 1986, and bridging into December of 1986, as directed by Dr. Chu, I, or others working under the direction and supervision of Dr. Chu and myself, began at least two activities; one being to subject the La-Ba-Cu-O compositions produced to a Bednorz & Muller nominal composition of La4.25Ba0.75Cu5O5(3-y) which we had confirmed to be superconductors to application of extreme pressure while testing how such pressure affected the Tc1 property of such composition; the second activity being to produce La-Ba-Cu-O composition to nominal formulas different than that described for a La-Ba-Cu-O composition by the Bednorz & Muller article and to test such different nominal formulations of La-Ba-Cu-O for exhibition of superconducting properties. Among the first samples of La-Ba-Cu-O of a differing nominal composition which I prepared in accordance with Dr. Chu's instructions were La-Ba-Cu-O samples of the following nominal formulas, designed by my identification designation ("J"):

J-1 (La_{0.4}Ba_{0.6})₂CuO₄

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J-2 (La<sub>0.6</sub>Ba<sub>0.4</sub>)<sub>2</sub>CuO<sub>4</sub>
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J-3 (La_{0.7}Ba_{0.3})₂CuO₄

J-4 (La_{0.8}Ba_{0.2})₂CuO₄

J-5 (La_{0.85}Ba_{0.15})₂CuO₄

J-6 (La_{0.9}Ba_{0.1})₂CuO₄

These samples are recorded in my handwriting in my notebook, together with my calculations of the amounts of reagents needed for their production as shown in Exhibit B.

- 6. From the results of the work which I performed according to Dr. Chu's instructions on La-Ba-Cu-O compositions as described in paragraphs 2-5 above, it was determined that (1) certain La-Ba-Cu-O compositions were superconductors, (2) that compositions of La-Ba-Cu-O of nominal formulations different than that described by Bednorz & Muller for a superconducting La-Ba-Cu-O composition were superconductors; (3) that under extreme pressure application such superconducting compositions of La-Ba-Cu-O exhibited zero electrical resistance at a critical temperature, T_{cl}, which was unexpectedly higher than that at which the sample composition exhibited zero electrical resistance when subjected only to ambient (atmospheric) pressure. These results were reported to Dr. Chu at or about the time the results were obtained.
- 7. Following the reports to Dr. Chu of the observation of the results measured in La-Ba-Cu-O systems, as described in paragraphs 3-6 above, while Dr. Chu was out of town in about mid-December 1986 tending to other duties, Dr. Chu called me to describe his ideas for making various elemental substitutions in a

La-Ba-Cu-O superconductor system to increase the $T_{\rm cl}$ temperature at which a so substituted composition would exhibit zero electrical resistance without the need for application of great pressure to such substituted compositions.

- 8. Of the substituted compositions described to me by Dr. Chu during that December 1986 telephone conversation, Dr. Chu described his idea of substitution for Ba as the alkaline earth metal element in a La-Ba-Cu-O system by Sr and Ca.
- 9. Further, during the mid-December 1986 telephone conversation with Dr. Chu, as described in paragraph 8 above, Dr. Chu also informed me of his idea of superconducting rare earth metal-alkaline earth metal-copper-oxygen compositions which in comparison to one of La-Ba-Cu-O would exhibit zero electrical resistance at a greater temperature (T_{c1}) without application of pressure. Dr. Chu described his idea for compositions wherein the rare earth element of La was substituted for by non-magnetic rare earth and similar elements, including Y or Lu. During this telephone call Dr. Chu described to me his belief that substitution for La in a La-Ba-Cu-O composition would produce a superconductor which superconducts at a T_{c1} temperature greater than that of a La-Ba-Cu-O composition.
- 10. As related in paragraph 9 above, among other things, Dr. Chu instructed me by that telephone call to next prepare, among other substituted compositions, Y-Ba-Cu-O compositions in accordance with the same program by which I had earlier prepared and tested different nominal formulations of La-Ba-Cu-O for

superconductive properties.

- 11. During the latter part of December, after Christmas, W.K. Wu and his research graduate student, James Ashburn, arrived at the laboratories of the University of Houston campus at which I worked to have samples, which I understand Dr. Chu had suggested to them (Wu/Ashburn) to prepare, tested for exhibition of superconductive properties. Based on information and belief which I derived from Dr. Chu and independently from M.K. Wu and Ashburn during their attendance at the University of Houston campus, I understood that the sample brought by Wu/Ashburn to be tested were compositions of I further understood that Wu/Ashburn had already La-Sr-Cu-O. tested these compositions for electrical resistance versus temperature and determined that, absent the application of great pressure thereto, such La-Sr-Cu-O samples exhibited a T_{el} greater than that of a La-Ba-Cu-O of similar nominal composition without the application of pressure.
- 12. As described in paragraph 11 above, at our labs at University of Houston, while Wu/Ashburn were in attendance, in the supervision of our group at Dr. Chu's directions I, with the assistance of others in our group, tested the Wu/Ashburn made samples of La-Sr-Cu-O for (1) zero electrical resistance at specific temperatures, (2) for diamagnetism, and (3) for T_{cl} under application to such materials of great pressures. These results were reported to Dr. Chu. The results of this testing were encouraging of a belief that a La-Sr-Cu-O composition possessed a higher T_{cl} than do compositions of La-Ba-Cu-O. However, because the

test results were stated by Dr. Chu to be too poor to support a publication, he directed me to prepare higher quality La-Sr-Cu-O samples for evaluation.

- 13. Dr. Chu's instruction to me to prepare higher quality samples of La-Sr-Cu-O consequently delayed the time at which I could begin preparation of samples of Y-Ba-Cu-O which had been assigned to me. During a discussion which occurred after Christmas in late December 1986, which was attended by myself, M.K. Wu, and others, the concept of substitution of Y for La in a composition of La-Ba-Cu-O to produce a composition of Y-Ba-Cu-O which superconducts at a T_c temperature greater than that of a La-Ba-Cu-O composition was described to M.K. Wu.
- 14. During the period of December 1986 after Christmas and continuing through January 28, 1987 while I continued to make and study approximately 47 different samples of La-Ba-Cu-O, I also prepared and evaluated thirty-eight different samples of La-Sr-Cu-O compositions. My sample code for a La-Sr-Cu-O sample was "SL" followed by a number or numbers by which I could identify from my records the nominal formulation and sintering conditions of the sample. In the period December 23, 1986 through January 1, 1987 I prepared and evaluated six samples, SL1-SL6 (Exhibit C). Between January 2-18, 1987 I prepared and evaluated twelve additional samples, SL7-SL17 (Exhibits C and D) and between January 18-28, 1987 I prepared twenty additional samples, SL18-SL31 (Exhibit E).
- 15. As my work with the La-Sr-Cu-0 system was progressing, I began to turn my attention to the making of Y-Ba-Cu-0 compositions

which I would begin upon conclusion of my work with La-Sr-Cu-O compositions.

- 16. The formulas of Y-Ba-Cu-O which I first listed for production, as shown by Exhibit F, are "214" nominal formulations wherein the rare earth to alkaline earth metal ratio, Y to Ba, which together comprise the "2" element begins with a slightly greater amount of rare earth metal --y-- (6:4, like the La to Ba ratio in the "J-2" sample of paragraph 5).
- 17. On January 15, 1987, I identified in my notebook several substituted compositions for production and examination. Among the compositions so identified is one of the formula $(Y_{0.6}Ba_{0.4})_2CuO_4$ (which may also be written as $Y_{1.2}Ba_{0.6}CuO_4$) as shown in Exhibit F attached hereto which is in my handwriting and made by me on January 15, 1987.
- 18. By January 28, 1987 I had substantially completed my assignment to prepare and investigate higher quality samples of La-Sr-Cu-O and on January 29, 1987 I again returned by attention to the project which Dr. Chu had previously assigned to me, namely the making of rare earth metal-alkaline earth metal-copper oxides in which, compared to a La-Ba-Cu-O system, La is replaced with Y or Lu. To this purpose, with respect to a Y-Ba-Cu-O system, on January 29, 1987 I proceeded to list samples of Y-Ba-Cu-O for production in accordance with a convention I followed when I first made samples of La-Ba-cu-O which differed from the nominal formula of Bednorz & Muller, as discussed in paragraph 5. I listed in my notebook (Exhibit G) the following samples for production, which I

later identified by a "YB" designation, as follows:

 $YB-101 (Y_{0.8}Ba_{0.2})_2CuO_4$

YB-102 (Yo Bao) CuO,

YB-103 (Y_{0.4}Ba_{0.6})₂CuO₄

 $YB-105 (Y_{0.3}Ba_{0.7})_2CuO_4$

YB-104 (Y_{0.2}Ba_{0.8})₂CuO₄

In addition to the above Y-Ba-Cu-O composition I further identified twenty other compositions involving various types of substitutions, including four compositions of Lu-Ba-Cu-O.

19. Further, on January 29, 1987, I identified further compositions for production and testing in accordance with Dr. Chu's direction to make compositions involving substitution variously for the La or Ba of a La-Ba-cu-O composition by other smaller radii atomic elements as shown in Exhibit G attached hereto which was made by me and is in my handwriting. The amounts of reagents required for the production of the Y-Ba-Cu-O compositions as shown in Exhibit G are shown in Exhibit H attached hereto, which is in my handwriting and made by me during January 29-30 1987. Among the Y-Ba-Cu-O compositions considered for production, one is specifically of the formula Y_{1.2}Ba_{0.8}Cu₁O₄, which is YB-102 (page H65 of Exhibit H).

20. In accordance with my prior calculations as recorded in Exhibit H, I began to prepare compositions of Y-Ba-Cu-O, one of which, designated as YB-102, was of the formula $Y_{1.2}Ba_{0.8}Cu_1O_4$ as described in Exhibit H. I prepared the Y-Ba-Cu-O compositions by weighing out the required amounts of Y_2O_3 , BaCO₃ and CuO powder

reagents, mixing the powdered reagents by grinding them in a mortar with a pestle until the powder mass was homogeneously mixed. Thereafter the homogeneous powdered mass was subjected to pressure in a die to convert it to pellet form, multiple pellets were made, and each pellet was sintered at 1000°C, the various pellets being sintered for different lengths of time.

- 21. Y-Ba-Cu-O compositions of nominal formula (Y_{0.6}Ba_{0.4})₂CuO₄, which may also be written as Y_{1.2}Ba_{0.8}CuO₄, were prepared under various firing conditions and were tested for electrical resistance versus temperature (R/T) on February 1, 1987 (Exhibit I; which is a measurement of electrical resistance as a function of temperature). The results of this R/T testing are listed in my Stenographic Notebook (Exhibit J). One of two samples sintered for twenty minutes exhibited zero electrical resistance at 89°K (T_{c1}) with a beginning onset temperature (T_{co}) at 94°K. The second sample exhibited zero electrical resistance of 90.1°K with an onset temperature of 94°K.
 - 22. Beginning February 2 and continuing through February 3, 1987 Samples of Y_{1.2}Ba_{0.8}CuO, were subjected to various pressures (3 klb, 6 klb, 9 klb, 12 klb, 15 klbs and 2 klb) and tested for electrical resistance versus temperature (Exhibit K).
 - 23. The Y_{1.2}Ba_{0.8}CuO₄ samples described in paragraph 21 were subjected to powder x-ray diffraction analysis on February 2 and 4, 1987. From the x-ray diffraction analysis it was determined that neither of the multiple phases comprising the sample had a crystalline structure like that of K₂NiF₄. The x-ray spectrum is

Exhibit L.

- 24. On February 5, 1987 the Y_{1,2}Ba_{0,8}CuO, samples described in paragraph 21 were tested for diamagnetism at temperatures from 140K to 4.2K and were found at 4.2K to exhibit at least about 24% of the diamagnetic signal of a lead sample of similar dimensions as shown by Exhibit M.
- 25. The Y_{1.2}Ba_{0.8}CuO₄ samples described in paragraph 21 were tested on February 6, 1987 for resistivity over a temperature range of about 100K to 30K while subject to various magnetic field strengths and the zero resistivity state at a magnetic field strength of 5.7T was found to remain as high as 40°K, as shown in Exhibit N.
- 26. From the data acquired on the $Y_{1.2}Ba_{0.8}CuO_y$ samples as described in paragraphs 21-26 above, the upper critical field $H_{c2}(T)$ was determined on February 6, 1987.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the Chu Application or any patent issued thereon.

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