A Novel Method for Characterization of Superconductors: Physical Measurements and Modeling of Thin Films

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ABSTRACT

A method for characterization of granular superconducting thin films has been developed which encompasses both the morphological state of the sample and its fabrication process parameters. The broad scope of this technique is due to synergism between experimental measurements and their interpretation using numerical simulations. Two novel technologies form the substance of this system: the magnetically modulated resistance method for characterizing superconductors, and a powerful new computer peripheral, the Parallel Information Processor card, which provides enhanced computing capability for PC computers. This enhancement allows PC computers to operate at speeds approaching that of supercomputers making atomic scale simulations possible on low cost machines. The present development of this system involves the integration of these two technologies using meso-scale simulations of thin film growth. A future stage of development will incorporate atomic scale modeling.

INTRODUCTION

In this paper, we describe a system for characterization of superconductor thin films which encompasses both physical properties of thin films and parameters which control their fabrication. This tool is naturally suited for development of thin film superconductor materials and for quality control. A comprehensive assessment is achieved by physical measurements and computational simulation of the measurement responses. This system is implemented by the integration of two novel technologies: the magnetically modulated resistance (MMR) method for characterization of superconductors,[1-5] and the parallel information processor (PIP)[6-8] for desktop supercomputing with PC computers.

The MMR method measures the magnetic field derivative of resistance as a function of temperature. This measurement has been shown to be sensitive to effects which occur in granular superconductors.[2] Granular superconductors consist of ensembles of small single crystals (grains) which are in contact with one another. Two grains separated by a common boundary constitute a superconductor structure called a weak link which has superconductor properties different in some respects from that of a single grain.[9] Granular superconductors, therefore, exhibit effects that are attributable to the weak links which constitute their morphology. It is important to account for these effects since virtually all high temperature superconductors, for practical purposes, are granular.

Our primary interest here is the characterization of granular thin film superconductors which are of paramount importance in superconductor devices. In the following discussion we will describe briefly how the MMR measurement characterizes superconductors. We then discuss briefly how the MMR measurement can be related to thin film fabrication parameters by computational simulation of the MMR response signal. Numerical simulation is becoming increasingly important in materials research. In this work, it provides an essential link between physical measurements, and sample morphology and process parameters. Calculations of this type require powerful computing machinery. Atomic scale materials simulations often require supercomputers. The PIP technology mentioned above allows the simulations in this project to be done on a PC. We will discuss this new technology, briefly, and show how the physical MMR measurements and computational elements are being integrated into a system for characterizing superconductor thin films. Finally we will indicate how we intend to extend this system in the future.

THE MMR METHOD

The essential features of the MMR technique are shown in the diagram in Figure 1. The superconductor sample is contained in a variable temperature bath. The sample is subjected to a magnetic field consisting of an ac

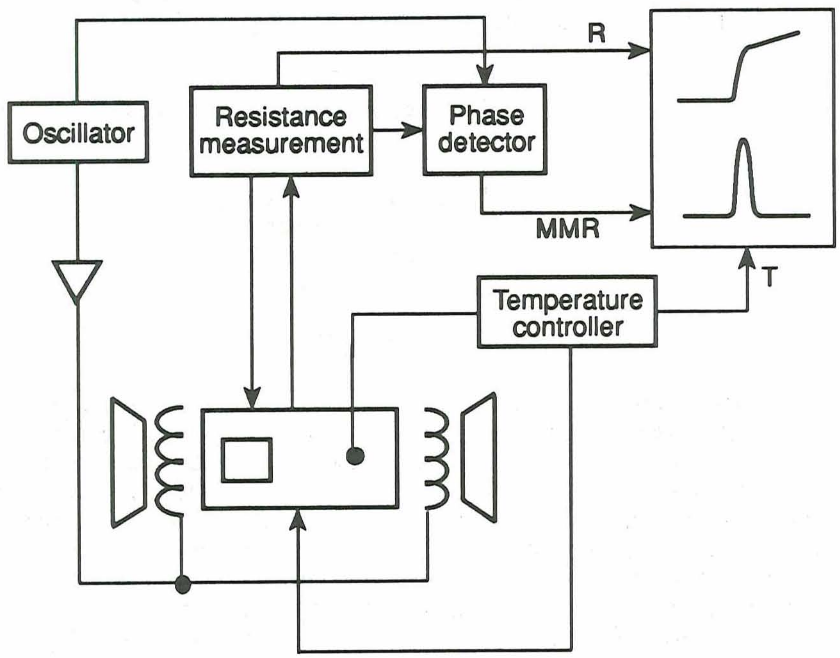

Figure 1. Schematic diagram for an apparatus to measure magnetically modulated resistance.

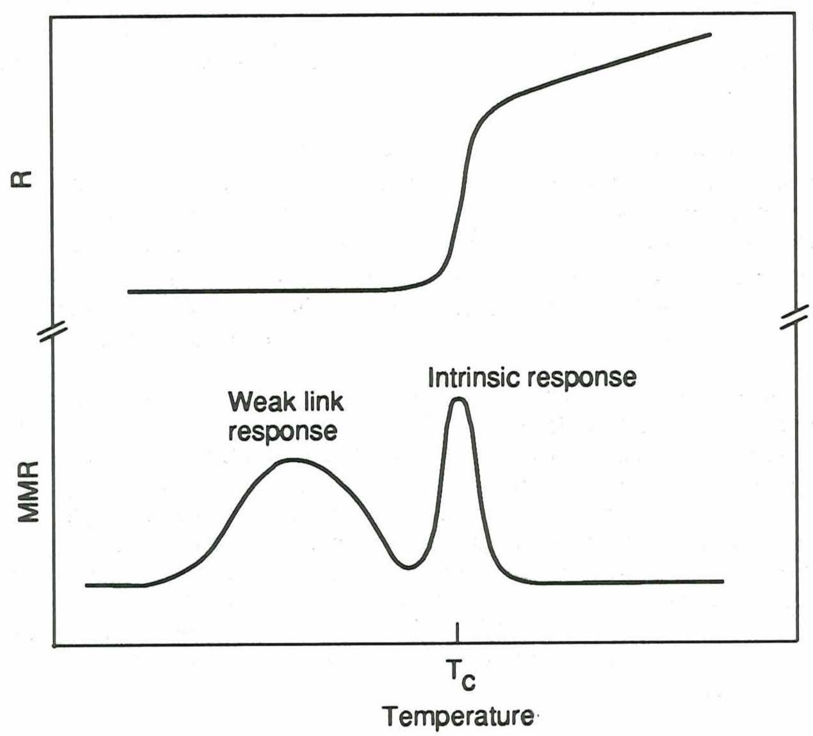

Figure 2. Typical resistance (*R*) and magnetically modulated resistance (MMR) response curves for a granular superconductor.

component, H_{ac}, and a collinear dc component H_{dc} where $H_{ac} < H_{dc}$. The resistance of the sample is measured at the field modulation frequency by phase detection. This signal, which constitutes the MMR response, is measured as a function of temperature. The method for measuring resistance can be any convenient method. We use four point probe dc resistance measurements and X-band microwave surface resistance measurements in our laboratory. (A detailed description of this method is contained in Reference [10]).

A typical MMR response signal for a granular superconductor is shown schematically in Figure 2. Also shown for comparison is the unmodulated resistance which is measured simultaneously. The MMR response contains two features; a peak at T_c which signifies the superconductor phase transition for individual superconductor grains, and a second peak below T_c which is due to superconductor weak link phase transitions. The relative position and shape of the MMR weak link feature depends upon the current density in the sample, the externally applied magnetic field, and the distribution of grain sizes and weak link coupling strengths in the sample. The dependence of the MMR weak link peak on grain size distribution provides the basis for characterization of the morphology of granular superconductors. In the following section, we briefly describe how the MMR signal is related to the sample grain size distribution.

THE MMR SIGNAL

In this section, we outline the physical interpretation of the MMR signal without detailed discussion of the underlying physics. The reader who is interested in more detail is referred to Reference [11]. An expression for the MMR signal vs. temperature was inferred under the assumptions that the weak links are Josephson junctions[9] with equal coupling strength per unit area, and that the applied current is unidirectional in the sample. Under these conditions it can be shown that the MMR signal has the form

$$MMR(T) = KWR^2 \frac{e^A}{I} F(T) \frac{d}{dB} \sum_L L \frac{\sin(\pi B/B_o)}{\pi B/B_o} f(L) \qquad (1)$$

where

$$R = \frac{1}{1+e^A}$$

$$A = -\left(1 - \frac{I_c}{I}\right)$$

$$I_c = \sum_L \frac{WK}{B_o} F(T) \frac{\sin \pi B/B_o}{\pi B/B_o} f(L)$$

$$B_o = \frac{K}{L}$$

In the expression above, K is a constant, I_c and I are the critical and applied currents, $F(T)$ is the temperature dependence of I_c, B is the applied magnetic field, B_o the critical field for a junction of length L, W and L are the width and length of a junction, and $f(L)$ is the distribution of junction lengths. The function $F(T)$ is known but is not related to the sample morphology. The junction length is the length along a grain boundary which is perpendicular to the current, and which is bounded by grain boundaries parallel to the current as illustrated by Figure 3. (For our purposes, all grain boundaries are resolved into directions parallel and perpendicular to the applied current.) The junction size distribution $f(L)$ is related to the grain size distribution $g(L_g)$ in a complex way, but this relationship can be calculated by a method of statistical inference. An example of this relationship is illustrated in Figure 4 for a Gaussian grain size distribution. Thus from Equation (1) and the relation between $f(L)$ and $g(L_g)$, we are able to relate the MMR signal to the grain size distribution of the sample.

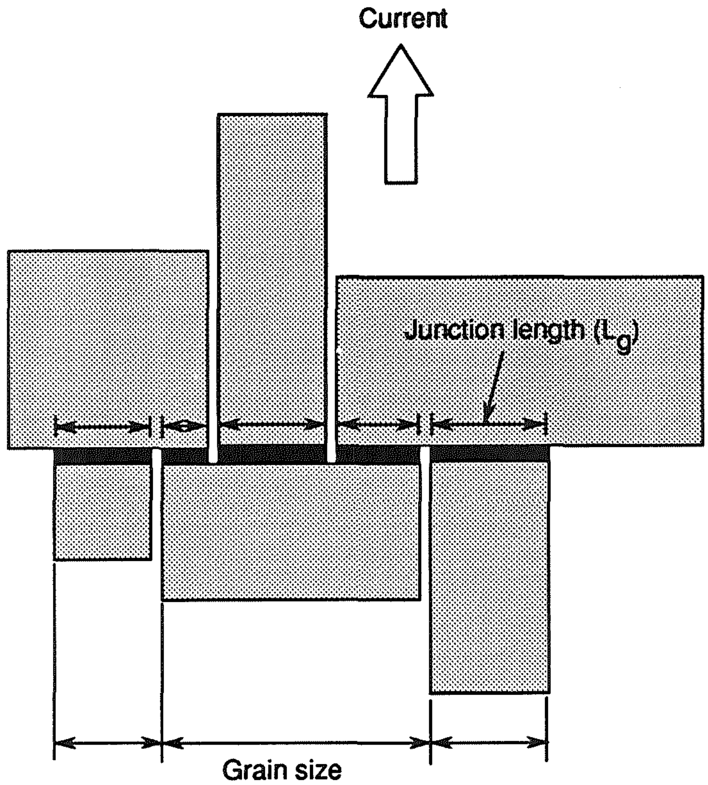

Figure 3. An illustration which shows the relation between grain size (L) and junction size (L_g) in an ensemble of closely packed grains.

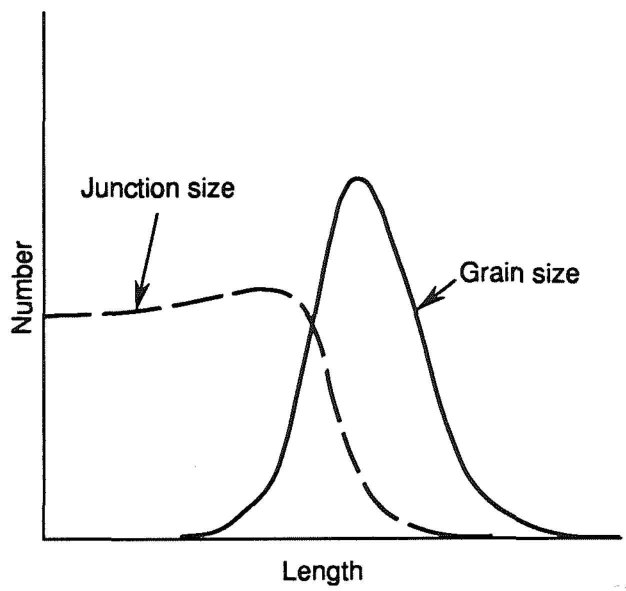

Figure 4. An example of the junction size distribution which results from a Gaussian grain size distribution.

The simulation of the MMR signal described here provides a way, in principle, to assess thin film morphology which is, for our purposes, the grain size distribution of the thin film. In actual practice, one cannot directly solve Equation 1 to obtain $f(L)$. Consequently, one must essentially estimate the function $g(L_g)$ and then compare the calculated MMR response with the experimental response to gauge the accuracy of the estimate. The problem, then, is how to estimate $g(L_g)$. Because the distribution $g(L_g)$ is directly related to the conditions which control the growth of the thin film, our approach is to estimate $g(L_g)$ by modeling the growth of thin films. This provides a way to obtain the grain size distribution from MMR measurements and at the same time extends the application of the MMR method to control of process parameters of thin film growth. In the next section, we discuss our work in modeling thin film growth to obtain estimates of $g(L_g)$.

SIMULATION OF THIN FILM GROWTH

The simulations of vapor phase thin film growth described here, generally apply to state of the art epitaxial thin films such as, for example, $YBa_2Cu_3O_{7-y}$ on $LaAlO_3$ substrates. These types of films consist of grains of various sizes which have a common orientation normal to the substrate. Growth occurs as a result of the vapor phase deposition of the constituent species, which constitute the superconductor, onto the substrate. The vapor phase constituents are commonly created by rf sputtering or laser ablation from a bulk superconductor target. The vapor initially condenses on the substrate and forms a number of seed crystals of the superconductor species. The seed crystals grow until their boundaries encounter neighboring crystals thereby forming common grain boundaries between the neighboring crystals. The size of a particular crystal is then determined by the presence of neighboring crystals which limits their lateral growth to the area encompassed by their grain boundaries.

The grain size distribution is inferred by numerical simulation of this mechanism of thin film growth. The simulation is presently implemented as a meso-scale model with two adjustable parameters. These are the probability P per unit time that a seed crystal will form at any given location on the substrate, and the rate of growth, R, of a crystal on the substrate. The simulation proceeds by determining the location of new seed crystals on the substrate in each unit of time according to the parameter P, and growing each existing crystal on the substrate according to the parameter R. This process continues until there are no voids on the substrate. The resulting grain size distribution is obtained directly by assessing the area of each crystal (grain). Thus, this simulation provides the shape of $g(L_g)$ which is determined by the two parameters P and R.

We plan to extend the film growth simulation using atomic scale simulation of the deposition. The formation of seed crystals and crystal growth will be simulated using interatomic interactions and the physical parameters of the growth process such as substrate temperature, oxygen pressure, and certain parameters of the reactant species. This will permit calculation of the meso-scale parameters P and R. The atomic scale model will be appended to the meso-scale model to provide the grain size distribution which will then be related to physical process variables.

AN MMR CHARACTERIZATION SYSTEM

The concepts discussed in the preceding sections are being integrated into a system for characterization of superconductor thin films. The initial version of this system, shown diagramatically in Figure 5, is based upon a comparison between experimental MMR measurements and numerical simulations of MMR response. The experimental measurements are obtained separately and stored in a computer file for subsequent off-line comparison with simulation. The simulation begins with the meso-scale thin film growth model which calculates the grain size distribution. The grain seed probability and growth rate parameters, P and R, are initially estimates which are entered manually. The junction size distribution, calculated from the grain size distribution, is used to calculate the MMR response. (The MMR response also depends upon some parameters not shown in Figure 5; e.g., magnetic field and current.) Comparison of the simulated and experimental MMR responses yields an MMR error from which new values of P and R are inferred. The process iterates until the MMR error falls within predetermined bounds. In this way the grain size distribution of the thin film is ascertained.

The parameters P and R are also determined in this system, although this knowledge is primarily of academic interest. In order to relate this to more fundamental physical parameters we intend to extend this system using atomic scale modeling of the deposition process. Figure 6 shows this extended system. The P and R parameters are calculated in the atomic scale model, as discussed previously, using process variables such as substrate temperature, oxygen pressure, and certain parameters of the reactants (e.g., momentum, excitation, etc.). Thus, in this system, the MMR response will be related to the thin film process parameters.

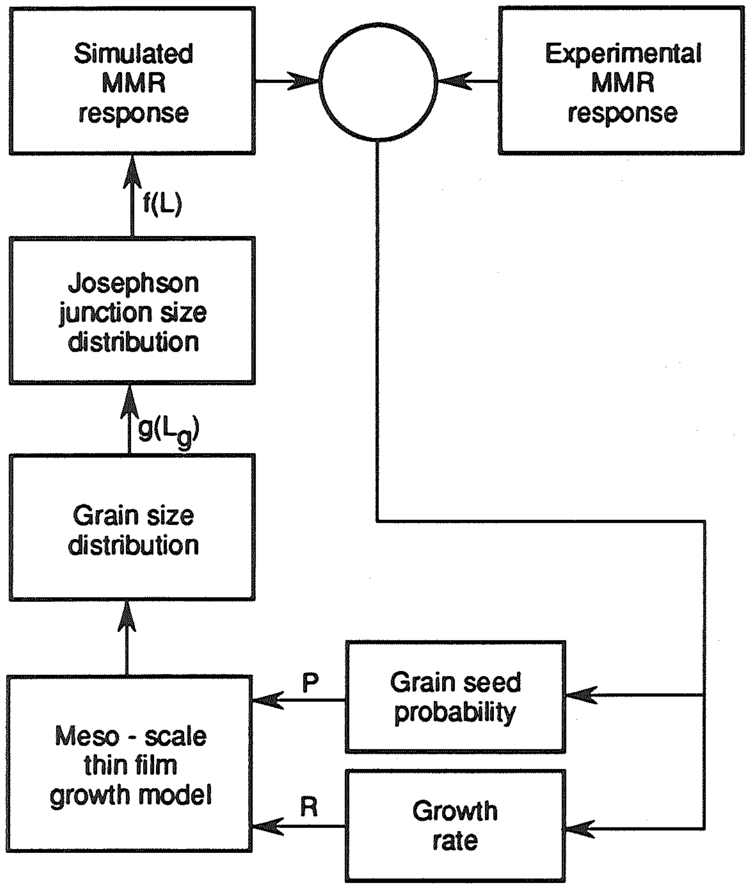

Figure 5. Schematic diagram of an MMR characterization system which uses meso-scale modeling.

The systems described above, particularly the extended system which uses atomic scale modeling, involve extensive computations which requires supercomputer capability to achieve results in reasonable times. A high speed processor card has been developed which provides supercomputer capability on a PC platform and allows the MMR characterization systems to be implemented in a small package at low cost. The heart of the new computer technology is a parallel information processor board which can reside in any 80x86 PC, and is capable of 60 million floating point operations per second (MFLOPS). The PIP board also has 512 kilowords (32 bits/word) of high speed random access static memory on board and is expandable to 20 megawords. Up to eight PIP boards can be installed in a single PC to achieve 480 MFLOPS computing power, approximately one half the peak computation rate of a CRAY I. A user-friendly interface for this computing system is being developed which allows programming in Fortran, Basic, C, and a high level tool for integrating various software modules with flow charts. This PIP technology has been licensed to Scientific Data Systems Inc., which expects to begin commercial distribution in late 1992.

Each of the procedures which comprise the system shown in Figure 5 has been completed. The procedures will be integrated on a 386 PC with a single PIP card. We expect to incorporate an atomic scale model with the MMR characterization system within a year. The extended system using atomic scale modeling will require additional PIP cards.

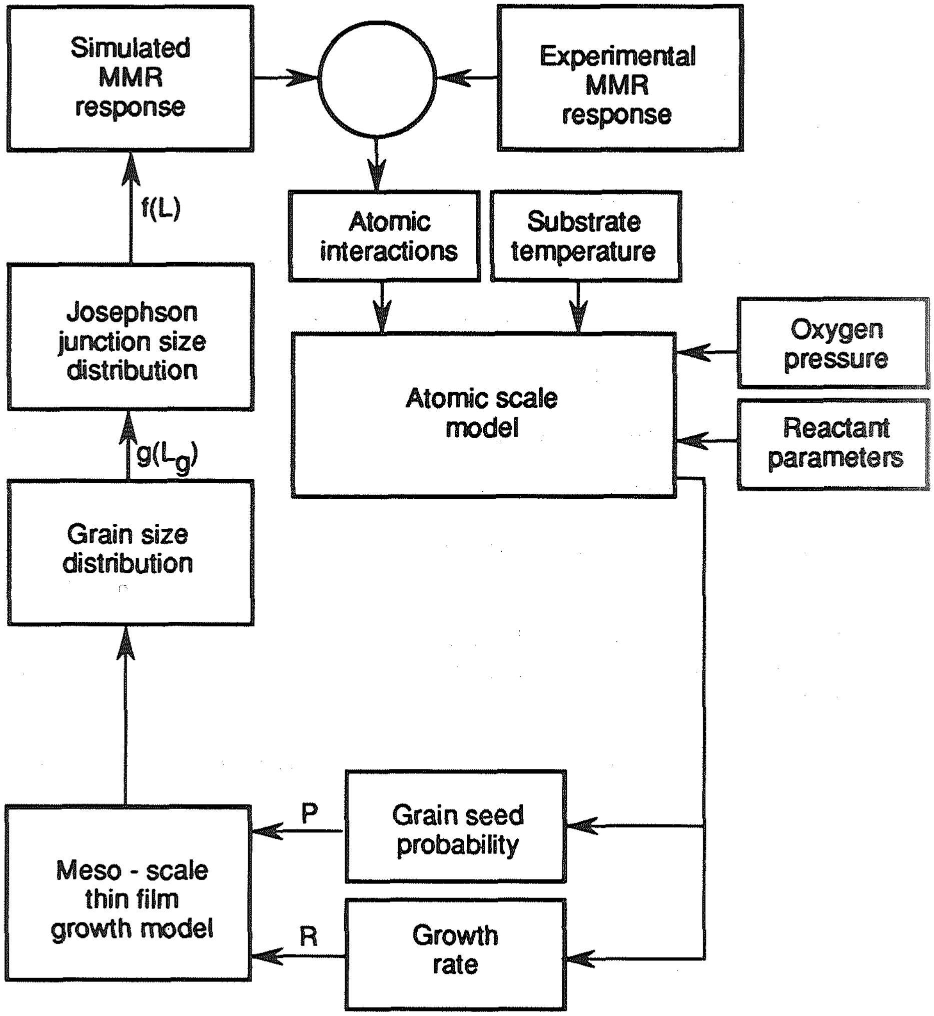

Figure 6. Schematic diagram of an MMR characterization system which uses atomic scale modeling.

CONCLUSION

The superconductor characterization system described here is noteworthy because of the extent of the information inferred from a relatively simple physical measurement. In its early usage, the MMR method provided evidence for the presence of weak links in superconductor samples in both granular samples and single crystals. At this stage, it was and remains a unique capability. The present stage of development in which the grain size distribution is inferred and is related to process parameters is due to the use of numerical simulations of the measured response function. This type of enhancement of physical measurements is being rendered practical by the emergence of powerful, low cost computer technology such as that embodied by the PIP card. There is little doubt that the combined use of measurements and large scale simulation in materials characterization and process control will become common as use of the new computer technology becomes widespread.

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