INVESTIGATION OF TRANSPORT AND MAGNETIC PROPERTIES OF YBCO NANOCOMPOSITES AND DEVICES

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INVESTIGATION OF TRANSPORT AND MAGNETIC PROPERTIES OF YBCO NANOCOMPOSITES AND DEVICES

by

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Department of Physics

Submitted

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to the

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Dedication

I want to dedicate this thesis work

To the Supreme Almighty Shiv Parmatma

And to my Parent

Certificate

This is to certify that the thesis entitled **"Investigation of transport and magnetic properties of YBCO nanocomposites and devices"** being submitted by **Mamta** to the Department of Physics, **Indian Institute of Technology Delhi** is worthy of consideration for the award of the degree of Doctor of Philosophy and is a record of the original bonafide research work carried out by her under my guidance and supervision. The results contained in it have not been submitted in part or full to any other university or institute for the award of any degree or diploma.

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ABSTRACT

$YBa_2Cu_3O_{7-\delta}$ (YBCO) is a most promising and widely investigated type II high T_C superconductor material due to its several interesting properties such as high critical temperature (T_C) of 93 K, low charge carrier concentration, and high upper critical field (H_{C2}) which makes it a suitable candidate for the use in various practical applications. In order to achieve high critical current density (J_C) at higher applied magnetic fields, the immobilization of the vortices by flux pinning is the key requirement. Therefore, there is a need to introduce the additional defects in the YBCO matrix apart from the naturally occurring defects in order to enhance the flux pinning characteristics.

For enhancing the flux pinning characteristics of YBCO, a very preferred strategy is to introduce nano additives in YBCO. In order to carry out work in this direction, YBCO:$NaNbO_3$ nanoparticles (NPs) or nanorods (NRs) (0.25-2 wt%) nanocomposite samples have been synthesized using two-step process i.e. using hydrothermal and solid state reaction method and flux pinning properties are investigated. The orthorhombic crystal structure of YBCO remains preserved in the nanocomposite samples as confirmed by X-ray diffraction pattern. The presence of NPs or NRs in the YBCO matrix is confirmed by morphological analysis. In the temperature dependence of resistance (R-T), above T_C, the thermal fluctuations are responsible for induced conductivity in YBCO and YBCO:$NaNbO_3$ nanocomposite samples which is used to determine various superconducting parameters such as coherence length, lower critical field, upper critical field, and critical current density. The 0.5 wt% NRs or NPs added composite sample has shown significant enhancement in the superconducting parameters as compared to pure sample which is a signature of improved flux pinning characteristics on addition of NRs or NPs in YBCO. The magnetic measurement of YBCO and nanocomposites (YBCO:x $NaNbO_3$) are conducted using PPMS technique. The 0.5 wt% NPs added composite sample has shown significant enhancement of ~2.6 times in the critical current density (J_C) as compared to pure, whereas 0.5 wt% NRs added composite sample has shown enhancement of ~2.5 times which indicates that NPs are acting as more efficient pinning centres at higher temperatures and applied fields. With the increase in concentration of NPs or NRs, the decrement in J_C is attributed to the increased agglomeration of NPs or NRs. Further, the pinning mechanism in all composite samples is explored. With the increase in the concentration of $NaNbO_3$, a switchover from δT_C to δl pinning is observed in maximum temperature range. The weak vortex pinning centres are dominant at a lower temperature while strong pinning centres are

found to dominate at higher temperatures for all samples as observed from the temperature dependence of critical current density.

The superconductor YBCO thin films are used for fabricating Josephson junctions (JJs) which are the basic elements for most superconducting electronic devices used for several applications. $SrTiO_3$ substrates are preferred for fabrication of SQUID devices and it has ~2% lattice mismatch with YBCO thin film. Despite of large lattice mismatch (~9%) with YBCO thin film, MgO single crystal has been used as a substrate for Josephson junctions, microwave devices, Terahertz devices due to its low dielectric loss, stable material properties with very high melting point (~2800 ^{0}C). But, MgO is hygroscopic in nature which leads to the degradation of the substrate surface. Various methods have been reported in the literature which includes solvent cleaning, chemical etching, mechanical polishing, vacuum and oxygen annealing, and Ar ion-beam etching after annealing to obtain the clean MgO substrate surface.

In the direction of developing high T_C thin film devices, YBCO Josephson junction (JJ) and SQUID device on the step-edge created $SrTiO_3$ (STO)(00l) substrate has been fabricated using the photolithography and Ar ion beam milling technique. The step height and step angle are measured using atomic force microscopy with corresponding values of 439 nm and 18° respectively. The obtained value of T_C for the YBCO thin films, across JJ and SQUID device on STO substrates are 90.2 K and 88.9 K, and 88.1 K respectively. From the analysis of temperature dependence of critical current of the junction, the presence of SNS type weak link is confirmed. Step edge junction SQUID showed voltage-flux (V-Φ) modulation when biased with current slightly higher to the critical currents. One period of V-Φ modulation corresponds to a magnetic field of 4×10^{-7} T.

Further, a recipe has been developed for reusing the used/degraded MgO substrates in depositing YBCO thin film and Josephson junction devices. Ar ion beam milling (400 V beam voltage, 14 mA beam current and 6×10^{-4} Torr Ar gas pressure) followed by post O_2 atmosphere annealing treatment (750 °C for 2 hours and 900 Torr chamber pressure) has been employed in the recipe. X-ray diffraction, optical microscopy, atomic force microscopy, and Fourier transform electron microscopy techniques are employed after each step of treatment in order to address the changes occurred on the surface of degraded MgO substrate after treatment and compared it with fresh MgO substrate. After treatment, the surface roughness of the degraded MgO substrate (sample 2) reduced to ~ 3.96 nm from their initial value of 129 nm. YBCO thin film and Step edge Josephson junction is fabricated on the untreated, treated and fresh MgO substrates. The obtained value of T_C for the YBCO thin films on treated MgO substrate is

88.3 K which is very much comparable to the YBCO film on fresh MgO substrate. The value of T_C across JJ on the treated substrate is 85.8 K, while on the fresh substrate, the value of T_C is 85.7 K, respectively. The presence of SNS type weak link is confirmed from the analysis of temperature dependence of critical current density.

सारांश

$YBa_2Cu_3O_{7-\delta}$ (YBCO) पदार्थ अपने कई महत्वपूर्ण गुणों, जैसे 93 K के क्रांतिक तापमान (T_C), कम आवेश वाहक सांद्रता, और उच्च क्रांतिक चुंबकीय क्षेत्र (H_{C2}) के कारण एक सबसे उपयुक्त और व्यापक रूप से उपयोग किया जाने वाला टाइप II उच्च T_C का अतिचालक पदार्थ है, जो इसे विभिन्न व्यावहारिक अनुप्रयोगों में उपयोग के लिए उपयुक्त उम्मीदवार बनाता है। उच्च अनुप्रयुक्त चुंबकीय क्षेत्रों में उच्च क्रांतिक धारा घनत्व (J_C) प्राप्त करने के लिए, फ्लक्स पिनिंग द्वारा वोरटिसेस का स्थिरीकरण प्रमुख आवश्यकता है। इसलिए, फ्लक्स पिनिंग विशेषताओं को बढ़ाने के लिए स्वाभाविक रूप से पाए जाने वाले डिफेक्ट्स के अलावा अतिरिक्त डिफेक्ट्स को YBCO मैट्रिक्स में बढ़ाने की आवश्यकता है।

YBCO की फ्लक्स पिनिंग विशेषताओं को बढ़ाने के लिए, YBCO में नैनोएडिटिव्स का उपयोग करना एक बहुत ही मुख्य रणनीति है। इस दिशा में काम करने के लिए, YBCO: $NaNbO_3$ नैनोपार्टिकल (NPs) या नैनोरोड्स (NRs) (0.25-2 wt%) के नैनोकंपोजिट सैंपल्स को दो-चरणीय प्रक्रिया, हाइड्रोथर्मल और सॉलिड स्टेट रिएक्शन विधि का उपयोग करके संश्लेषित किया गया है और फ्लक्स पिनिंग गुणों की जांच की गयी है। YBCO की ऑर्थोरोम्बिक क्रिस्टल संरचना नैनोकम्पोजिट सैंपल्स में संरक्षित होने की पुष्टि X-रे विवर्तन पैटर्न द्वारा की गई है। YBCO मैट्रिक्स में NPs या NRs की उपस्थिति की पुष्टि मॉर्फोलॉजी विश्लेषण द्वारा की गयी है। T_C के ऊपर, प्रतिरोध की तापमान निर्भरता (R-T) में तापीय उतार-चढ़ाव, YBCO और YBCO:$NaNbO_3$ नैनोकम्पोजिट सैंपल्स में प्रेरित चालकता के लिए जिम्मेदार होते हैं, जिनका उपयोग विभिन्न अतिचालक मापदंडों जैसे कि कोहेरेन्स लेंथ, निम्न क्रांतिक क्षेत्र, उच्च क्रांतिक क्षेत्र और उच्च क्रांतिक धारा घनत्व को निर्धारित करने के लिए किया गया है। 0.5 wt% NRs या NPs संकलित कम्पोज़िट सैंपल ने शुद्ध सैंपल की तुलना में अतिचालक मापदंडों में महत्वपूर्ण वृद्धि दिखाई है जो YBCO में NRs या NPs को संकलित करने पर बेहतर फ्लक्स पिनिंग विशेषताओं का एक प्रमाण है। YBCO और उसके नैनोकंपोजिट्स (YBCO:x $NaNbO_3$) की चुंबकीय माप, PPMS तकनीक का उपयोग करके की गयी है। 0.5 wt% NPs संकलित कम्पोजिट सैंपल ने शुद्ध सैंपल की तुलना में क्रांतिक धारा घनत्व (J_C) में ~2.6 गुना की महत्वपूर्ण वृद्धि दिखाई है, जबकि 0.5 wt% NRs संकलित कम्पोजिट सैंपल ने ~2.5 गुना की वृद्धि दिखाई है जोकि दर्शाता है कि NPs उच्च तापमान और अनुप्रयुक्त क्षेत्रों में अधिक कुशल पिनिंग केंद्रों के रूप में कार्य कर रहे हैं। NPs या NRs की सांद्रता में वृद्धि के साथ J_C में कमी, NPs या NRs के बढ़े हुए संचय के लिए जिम्मेदार है। इसके अलावा, सभी कम्पोजिट सैंपलों में पिनिंग क्रियाविधि का पता लगाया गया है।

$NaNbO_3$ की सांद्रता में वृद्धि के साथ, अधिकतम तापमान सीमा में δT_C से δl पिनिंग में अंतरण देखा गया है। कमजोर पिनिंग केंद्र कम तापमान पर प्रभावी होते हैं जबकि मजबूत पिनिंग केंद्र सभी सैंपलों के लिए उच्च तापमान पर हावी पाए जाते हैं जैसा कि क्रांतिक धारा घनत्व की तापमान निर्भरता से देखा गया है।

अतिचालक YBCO की पतली फिल्मों का उपयोग जोसेफसन जंक्शनों (JJs) को बनाने के लिए किया जाता है जो कई अनुप्रयोगों के लिए उपयोग किए जाने वाले अधिकांश अतिचालक इलेक्ट्रॉनिक उपकरणों के मूल तत्व हैं। स्ट्रॉन्शियम टाइटेनेट, $SrTiO_3$ (STO) सब्सट्रेट को सुपरकंडक्टिंग क्वांटम इंटरफेरेंस डिवाइस (SQUID) उपकरणों के निर्माण के लिए वरीयता दी जाती है और इसमें YBCO-पतली फिल्म के साथ ~2% लैटिस असमानता है। YBCO पतली फिल्म के साथ बड़े लैटिस असमानता (~9%) के बावजूद, इसकी कम डाइइलेक्ट्रिक हानि, बहुत उच्च गलनांक के साथ पदार्थ के स्थिर गुणों (~2800 °C) के कारण, मैग्नीशियम ऑक्साइड (MgO) के सिंगल क्रिस्टलों का उपयोग जोसेफसन जंक्शनों, माइक्रोवेव उपकरणों और टेराहर्ट्ज़ उपकरणों के लिए एक सब्सट्रेट के रूप में किया गया है। परन्तु, MgO की प्रकृति ह्यग्रोस्कोपिक है जिसके कारण सब्सट्रेट की सतह का क्षरण होता है। स्वच्छ MgO सब्सट्रेट सतह प्राप्त करने के लिए साहित्य में विभिन्न तरीकों, जैसे विलायक सफाई, रासायनिक एचिंग, आयन-बीम एचिंग, यांत्रिक पॉलिशिंग, निर्वात और ऑक्सीजन में एनीलिंग का विवरण दिया गया है।

उच्च T_C वाली पतली फिल्म के उपकरणों को विकसित करने की दिशा में, YBCO जोसेफसन जंक्शन (JJ) और SQUID डिवाइस को स्टेप-एज (STO) (00l) सब्सट्रेट पर बनाया गया है, जिसे फोटोलिथोग्राफी और Ar आयन बीम मिलिंग तकनीक का उपयोग करके तैयार किया गया है। स्टेप- हाइट और स्टेप-एंगल के मानो को, परमाणु बल माइक्रोस्कोपी का उपयोग करके क्रमशः 439 nm और 18° के संगत मापा गया है। STO सबस्ट्रेट्स पर JJ और SQUID डिवाइस में YBCO की पतली फिल्मों के लिए T_C के प्राप्त मान क्रमशः 90.2 K और 88.9 K, और 88.1 K हैं। जंक्शन की क्रांतिक धारा की तापमान पर निर्भरता के विश्लेषण से, S-N-S टाइप कमजोर लिंक की उपस्थिति की पुष्टि हुई है। स्टेप-एज-जंक्शन-SQUID में धारा का मान जब क्रांतिक धारा के मान से ज्यादा होता है, तो वोल्टेज-फ्लक्स (V-Φ) मॉडुलेशन मिलता है। V-Φ मॉडुलेशन की एक अवधि 4×10^{-7} T के चुंबकीय क्षेत्र के बराबर होती है।

इसके अलावा, YBCO पतली फिल्म और जोसेफसन जंक्शन उपकरणों को बनाने में प्रयुक्त/अपघटित MgO सबस्ट्रेट्स का पुन: उपयोग करने की एक विधि विकसित की गयी है। इस विधि मे, Ar आयन-बीम मिलिंग (400 V बीम वोल्टेज, 14 mA बीम करंट और

6×10^{-4} Torr Ar गैस दबाव) के बाद O_2 वायुमंडल-एनीलिंग ट्रीटमेंट (2 घंटे के लिए 750 °C और 900 Torr चैम्बर दबाव) किया गया है। एक्स-रे विवर्तन, ऑप्टिकल माइक्रोस्कोपी, परमाणु बल माइक्रोस्कोपी, और फूरियर ट्रांसफॉर्म इन्फ्रारेड माइक्रोस्कोपी तकनीकों को ट्रीटेड सब्सट्रेट की सतह में आये परिवर्तनों को मापने के लिए किया गया है ताकि उसकी तुलना शुद्ध MgO के सब्सट्रेट से की जा सके। ट्रीटमेंट के बाद, प्रयुक्त/अपघटित MgO के सब्सट्रेट (सैंपल 2) की सतह का खुरदरापन प्रारम्भिक मान, ~129 nm से घटकर 3.96 nm हो गया है। प्रयुक्त/अपघटित, ट्रीटेड, शुद्ध MgO के सब्सट्रेट की सतह पर बनी YBCO की पतली फिल्म और स्टेप-एज जोसेफसन जंक्शन के लिए प्राप्त Tc का मान 88.3 K है, जोकि शुद्ध MgO सब्सट्रेट पर बनी फिल्म के Tc के तुलनात्मक है। ट्रीटेड सब्सट्रेट पर बनाये गए JJ के लिए Tc का मान 85.8 K है, जबकि शुद्ध MgO के सब्सट्रेट पर, T_C का मान 85.7 K है। जंक्शन की क्रांतिक धारा की तापमान पर निर्भरता के विश्लेषण से, S-N-S टाइप कमजोर लिंक की उपस्थिति की पुष्टि हुई है।

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