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PULSE LASER ANNEALING OF NIOBIUM FILM ON COPPER FOR SRF CAVITIES

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Abstract

Thin film cavities were proposed as the most promising next generation superconducting cavities. The challenges are improving the surface superconducting performance and reducing defects of the coating film, which can be greatly solved by laser annealing. Laser annealing system has been set up in Peking University, and experiments with niobium thin film sample have been carried out. Superconducting performance and other properties of Nb/Cu samples before and after annealing were compared. Recrystallization happened and surface structure improved a lot according to the results.

INTRODUCTION

The niobium sputter-coated accelerating cavities have advantages of low surface loss, better thermal conductivity, better mechanical stability, and lower cost over bulk Nb cavities [1].

The quality of the niobium thin film is limited by substrate processing [2], impurities, grain structure, surface defects, film-substrate bonding force [3] and etc., which leads to the Q-slope of thin film cavities under high accelerating gradient electric field. Generally, the grain structure of the niobium films deposited on copper prepared by magnetron sputtering is of columnar crystal structure with voids, and the grain size is about tens to hundreds of nanometres (Fig. 1). Excessive grain boundaries and voids per unit area will increase the surface resistance [4]. It is one of the main factors limiting the superconducting radio frequency performance of the film cavities.

During the sputtering process, we can reduce impurities by improving vacuum conditions, using high-purity target material and working gases. In addition, according to the extended SVD (structure zone diagram) model proposed by Andre Anders [5], we can increase the energy of sputtering particles and the substrate temperature to grow larger grains and better compactness Nb films on copper. However, on the one hand, during manufacturing process, setup and operation, the gas impurities adsorbed by the film cavities and the local surface defects generated may also affect its superconducting properties, causing local quenching, etc. on the other hand, due to the melting point of copper (1084℃), we are unable to increase the growth temperature and annealing temperature of copper-based films beyond the melting point of copper.

In order to solve these challenges, we have studied the post-treatment of Nb/Cu samples by laser annealing, aimed at reducing surface defects and gas impurities, improving films morphology and finally improving superconducting RF performance by re-melting niobium films.

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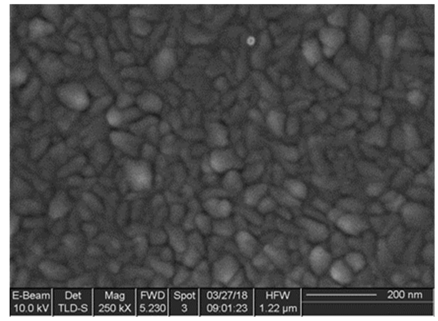
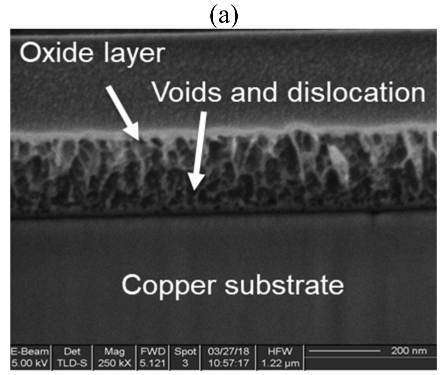


(a)



(b)

Figure 1: The SEM (a) and FIB (b) results of Nb/Cu samples deposited by dcMS [6]. It can be observed that (a) the grain size of niobium film is about several tens of nanometres, and (b) it has a network structure from the cross-section appearance and there are many voids and dislocations inside.

LASER ANNEALING SYSTEM SET UP

Sketch Diagram of Laser Annealing System

Laser annealing uses short pulsed laser to interact with metal, and the heat deposition generated will be concentrated only in the range of film thickness (~μm), forming a local thermal field. When the laser reaches a certain energy density (or the surface temperature rises beyond Nb melting point), the Nb film will melt gradually without the copper substrate affected. To carry out more experiments, we set up the laser annealing system in Peking University. The layout diagram of this system is shown in Fig. 2. The Nd: YAG laser is used and the pulse width is about 6ns. The energy of laser pulse is adjustable in the range of 100~370 mJ. The scanning system consists of a scanner and a PC terminal, with adjustable scanning area, scanning speed and scanning path. Annealing takes place in a vacuum chamber up to 10^{-6} Pa.

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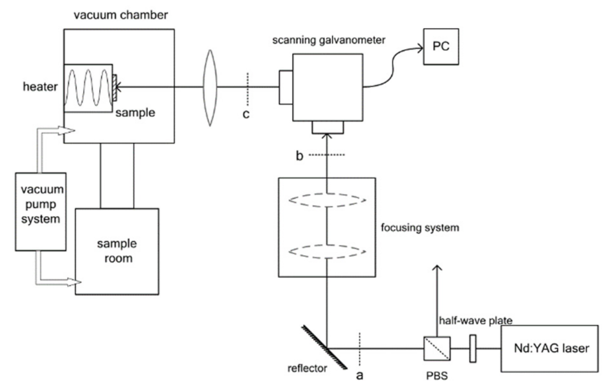


Figure 2: Sketch diagram of laser annealing system. It consists of Nd: YAG laser, scanning system, vacuum chamber and sample room.

Proper Annealing Parameters

According to the heat conduction equations and our experimental results, the melting phenomenon of niobium film (thickness ~1 μm) can only be observed when a single pulse laser energy is beyond 150 mJ. The surface appearance of annealed samples is strongly dependent on the adhesion force of the thin films. It was found in the experiment results that with the same annealing condition, the samples with poor bonding force delaminated more easily and required higher uniformity of laser scanning. It will be explained in detail in the experiments results below. In order to obtain better copper-based Nb films, it is required that films have better binding force, and the laser scanning path is reasonably set to make the energy deposition more uniform. The intensity distribution of our laser spot is shown in Fig. 3, and the laser repetition frequency is 3 Hz. In order to obtain a more uniform scanning area, we set the scanning speed to 3 mm/s and the path spacing to 0.5 mm, as shown in Fig. 4.

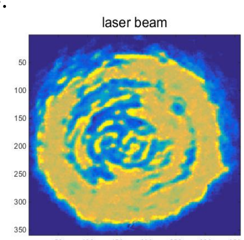
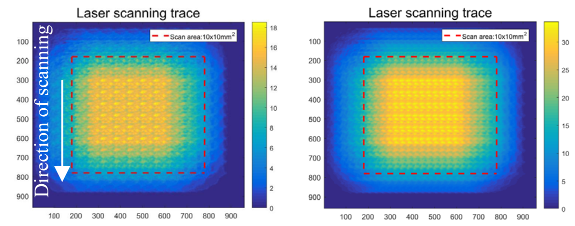


Figure 3: The intensity distribution of single laser pulse. We intend to improve the uniformity of energy deposition by optimizing the scanning path set. The spot diameter is about 6 mm.

When the laser pulse energy distribution is not uniform, reducing the path interval will make the energy deposition more uniform, but too small interval will increase the unit energy deposition. For Nb films with poor bonding force, the thermal resistance at the interface is larger, which may cause Nb/Cu spall by excessive energy deposition.



(a) (b)

Figure 4: Simulation of the effect of scanning path setting on laser energy deposition. The scanning area is 10 × 10 mm^2 (but because of divergence angle the actual annealed area is larger), the laser frequency is 3 Hz, the scanning speed is 3 mm/s, and the path interval is (a) 1 mm and (b) 0.5 mm.

RESULTS AND DISCUSSION

Exfoliation of Nb/Cu Film After Laser Annealing

The Nb/Cu samples of smooth surface we used were all deposited by dcMS. Figure 5 shows the annealing results with the same scanning parameters but different laser pulse energies. For the samples with weak film-substrate binding force, which can be observed the exfoliation of the edge and the sheared edge. After annealing with slightly lower laser energy (such as Fig. 5(b) laser energy is 200 mJ/pulse), the laser would leave clear scanning traces. If the laser energy is increased (Fig. 5(c) laser energy is

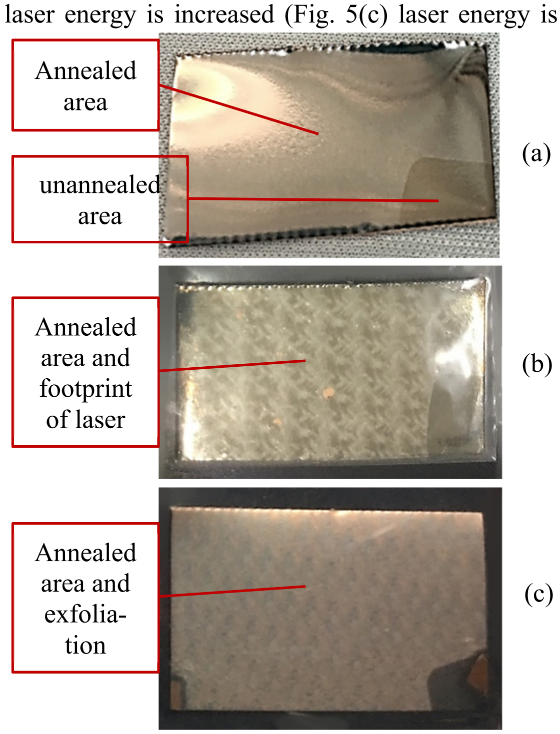


Figure 5: Effect of Nb/Cu binding force on laser annealing. (a) has the best binding force of these three samples. Scanning parameters are the same, and the laser energies are respectively (a) 352 mJ/pulse, (b) 200 mJ/pulse, (c) 300 mJ/pulse. After annealing, (a) a bright surface was obtained, while (b) and (c) both showed severe exfoliation.

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300 mJ/pulse), the surface will be severely etched, even exposing the whole copper substrate.

In addition, we also carried out annealing experiments on Nb/Si samples (Fig. 6). And severe delamination occurred at low energy density (179 mJ/pulse). Compared with Nb/Cu films, the thermal conductivity of silicon substrate is much lower and the adhesion of Nb coating is weaker. Besides, the weak adhesion leads to the high interfacial thermal resistance $R_{Nb/Cu}$. The extra $R_{Nb/Cu}$ is generated because of the interfacial gap, oxide and carbon impurities. It is speculated that the presence of interfacial thermal resistance leads to excessive heat deposition on the interface, and finally local temperature rise leads to the evaporation of Nb. Laser annealing may also be used as a method to measure the adhesion of films.

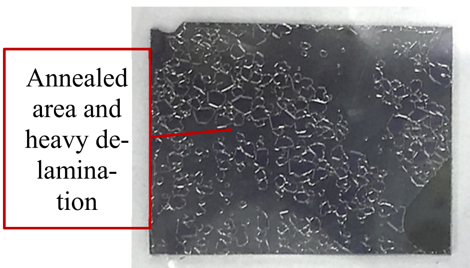

Figure 6: A result of Nb/Si annealing. The bonding force is weaker than Nb/Cu samples. Laser energy used was 179 mJ/pulse, and severe delamination occurred on the surface.

Reduction of Surface Defects

After annealing, the surface often becomes brighter (Fig. 5(a)) and it can be observed that a significant reduction in holes and pits by metallomicroscope (Fig. 7). Increasing laser energy and annealing times can make the defects disappear gradually.

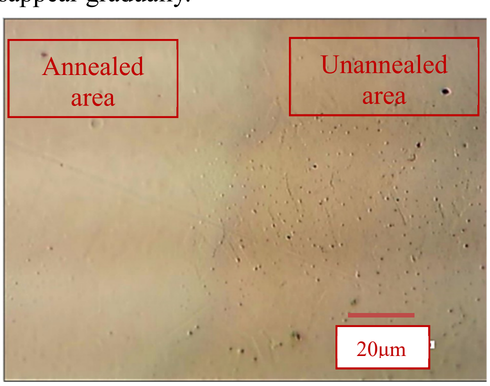

Figure 7: Metallomicroscope image of annealed Nb/Cu film. Laser energy used is 370 mJ/pulse. The defects are obviously reduced after annealing.

Grains Re-Melting

The melting process was observed by increasing the annealing times (Fig. 8). Since the laser pulse width is only 6ns and the repetition frequency is low (3 Hz), better annealing effect can be obtained by increasing the annealing times.

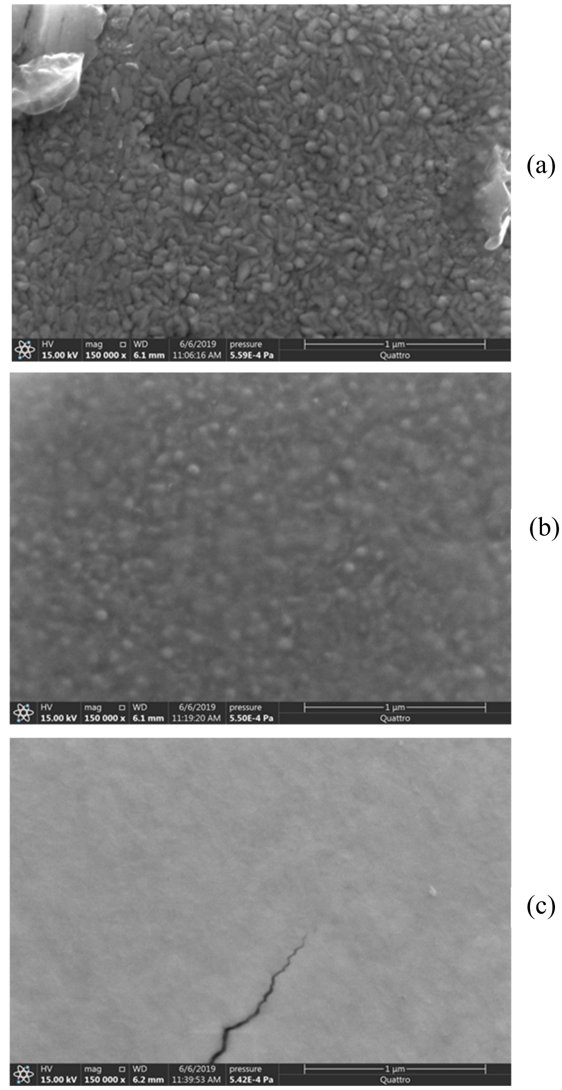

Figure 8: (a) The grain boundary is very clear without annealing in the SEM images. Annealing energy is 270 mJ/pulse, annealing times respectively are (b) 10 times and (c) 20 times. The grain boundary gradually disappeared and re-melting occurred.

CONCLUSION

The laser annealing experiments of Nb/Cu film in Peking University are reported. The laser pulse energy range for annealing is 100~370 mJ, and the scanning path is adjustable. Due to the nonuniformity of our laser spot, it is necessary to reduce the path interval to obtain uniform energy deposition for the surface. The adhesion of Nb coating is an important factor affecting the quality of laser annealing. Samples with poor film adhesion may be exfoliated before the uniform melting of niobium film. For samples with better adhesion, the annealing quality can be improved by increasing unit energy deposition (such as increasing laser energy density, annealing times, etc.). Further experiments including superconducting performance tests are carrying out too.

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