

HIGH QUALITY FLUX CONTROL SYSTEM FOR ELECTRON GUN EVAPORATION.

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Abstract

A high quality flux control system for electron gun evaporation has been developed and tested for the MBE growth of high temperature superconductors. The system can be applied to any electron gun without altering the electron gun itself. Essential elements of the system are a high bandwidth mass spectrometer, control electronics and a high voltage modulator to sweep the electron beam over the melt at high frequencies. The sweep amplitude of the electron beam is used to control the evaporation flux at high frequencies. The feedback loop of the system has a bandwidth of over 100 Hz, which makes it possible to grow superlattices and layered structures in a fast and precisely controlled manner. The drift of the total system is dominated by the temperature drift of the secondary emission multiplier in the mass spectrometer. This drift is typically 1-2% per hour for copper as measured by an independent quartz crystal thickness monitor. The feedback system has successfully been used for the fabrication of $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films.

Introduction

One of the methods to produce high temperature superconductor thin films in situ is by electron beam evaporation techniques, as has been successfully demonstrated by various groups [1,2,3]. One of the requirements for fabricating smooth, in situ films and layered structures of high temperature superconductors in an evaporation type system is the ability to control the relative composition of the film in a precisely controlled manner, preferably well within 1% [4,5].

One of the various types of evaporation sources that can be employed is the electron gun. An electron gun consists of a crucible which contains the evaporant material and a high energy electron beam to heat the material from the top. Advantages of the electron gun are the possibility to evaporate materials which need to be evaporated at very high temperatures due to the large power which can be transferred directly into the evaporation material, and the possibility to cool the crucible, which prevents its coevaporation.

The feedback system for electron gun evaporation we developed is based on work done by Schellingerhout et al., presented in [6], to which we refer for a more detailed analysis. The main conclusion of that work is that the evaporation is a high bandwidth process: it calls for a way to control the flux from the electron gun at high frequencies. This high frequency control is realized in our case by modulation of the amplitude of the electron beam sweep. The electron beam is swept over the melt by adding a high voltage sine to the high voltage supply of the electron gun. If the sweep frequency is much larger than the bandwidth of the evaporation process the electron beam will effectively be smeared out over the melt, and modulation of the sweep amplitude will result in a change of the power distribution which results in a change of the evaporation flux. In this paper we briefly review some of the conclusions drawn by Schellingerhout et al. and show results on the high voltage modulation method which can easily control the evaporation flux from the electron gun up to the cut off frequency of the evaporation process.

Feedback system requirements.

The basic limitation of any feedback system that is used to control a process is that part of the feedback loop which has the smallest bandwidth. If one looks with a high bandwidth (i.e. >1kHz) mass spectrometer at the particles that are evaporated from an electron gun which is equipped with an AC filament one can see strong line frequency components in the mass spectrometer signal as has been observed by Schellingerhout et al [6] (fig.1). The magnetic field of the large AC current flowing through the electron gun filament deflects electrons leaving the surface of the filament. This deflection of the electron beam is seen in the mass spectrometer signal because

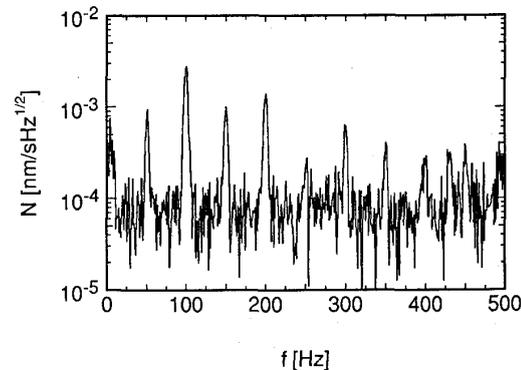


Figure 1. Evaporation rate noise spectral density N (in $\text{nm/sHz}^{1/2}$), as measured by the mass spectrometer, versus frequency f (in Hz). This noise spectrum is recorded for yttrium, at an average rate of 0.05 nm/s , with no feedback applied. The spectrum clearly shows the relatively large harmonics of the line frequency in the evaporation rate signal, which are caused by the AC filament.

the evaporation spot is not sufficiently smeared out by the wiggling of the beam. This implies that the evaporation process in an electron beam source is essentially a high bandwidth process. The high bandwidth of this process opens up the possibility to compose a high bandwidth feedback system. A high bandwidth feedback system can both accurately control the average evaporation flux on the typical time scale of the growth of one unit cell (which is in our case typically 6 seconds for $\text{YBa}_2\text{Cu}_3\text{O}_7$) and has the possibility to modulate the evaporation flux in a controlled manner for e.g. layer-by-layer growth or the fabrication of SIS structures. To obtain such a high bandwidth it is however necessary to use a sensor with a high bandwidth and a way to control the evaporation flux at high frequencies. For optimal performance they should both have a bandwidth larger than that of the evaporation process [6]. It also requires the elimination of the line frequency components by the usage of a DC filament supply.

There are of course limitations to the bandwidth of a evaporation flux feedback system. One of these is that the atoms that are evaporated have a finite time of flight in the vacuum system. We estimate this time of flight to be on the order of 1 ms in our system. Another limitation is that at a certain frequency the evaporation spot will not be following changes in the spatial distribution of the electron beam or fluctuations of the emission any more. This frequency is probably material dependent.

Feedback system configuration.

The feedback system here described has been built in a VG MBE system. The MBE system is equipped with two electron guns (Airco Temescal SFIH-270-2) and four Knudsen cells. The Airco gun has a large spot size which has the advantage of a low partial pressure of the evaporant just above the melt which yields an inherently stable evaporation flux. For each electron gun a feedback system has been constructed. The system contains one quartz crystal thickness monitor, which can be moved into the sample position to provide a tooling factor of 100%. The quartz crystal is used to calibrate the mass spectrometers. It has also been used to measure the long term stability of the electron gun feedback system.

The feedback system consists of a evaporation rate sensor, the electron gun and some control electronics. We have chosen a cross beam type quadrupole mass spectrometer as a sensor (VG Masstor 200 DX). This mass spectrometer yields a current of 1 pA (at the input of the SEM) at a rate of 1 nm/s (as measured by the quartz crystal) for copper. The distance between mass spectrometer and source is 65 cm, and between quartz crystal and source 45 cm. Each

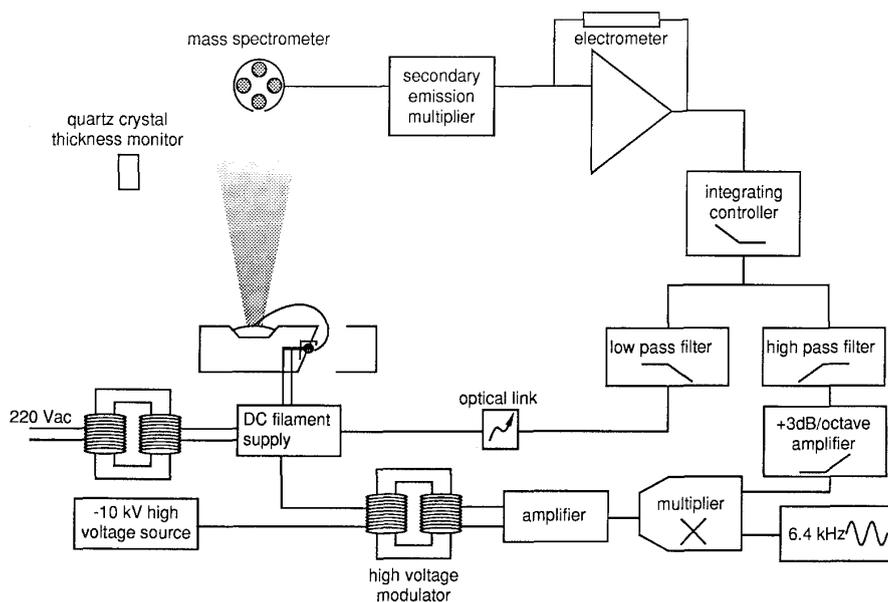


Figure 2. Schematic drawing of the feedback system. The evaporation rate is measured with a one kHz bandwidth mass spectrometer. The evaporation rate of the electron gun is controlled at frequencies below 1 Hz by changing the DC filament current, and at higher frequencies by modulating a 6.4 kHz high voltage sine which is added to the -10 kV high voltage of the power supply.

electron gun has a separate mass spectrometer to obtain an as high as possible bandwidth. The mass spectrometer is equipped with a secondary emission multiplier (SEM) which is essential for a high bandwidth and a high signal to noise ratio. The mass spectrometers are differentially pumped to guarantee its working even at high pressures. The total pressure in the mass spectrometer chamber is a hundred times as low as the pressure in the main chamber when working at high oxygen or ozone pressures. The feedback system has been tested successfully with oxygen and ozone pressures up to 10^{-4} mbar main chamber pressure.

The ion current that passes through the quadrupole filter in the mass spectrometer is first amplified by the built-in SEM (fig.2). The output current from the SEM is converted to a voltage by an electrometer. This electrometer is fed by a battery to obtain an as low as possible line frequency pick-up. The electrometer is rigidly connected to the mass spectrometer flange to reduce currents induced by vibrations. The output signal from the electrometer is passed to the control electronics for the electron gun. The control electronics consist of a setpoint comparator which compares the signal measured by the mass spectrometer with a setpoint value, an integral controller which is used to adjust the open loop gain of the feedback system, and electronics to control the evaporation rate from the electron gun.

The output signal from the integral controller is split into a high and a low frequency part. The low frequency signal adjusts the emission of the electron gun by changing the filament current. The standard AC filament supply of the electron gun is replaced by a DC filament supply. This DC filament supply is a voltage controlled power supply which is connected to the high voltage. Line power is supplied to it by a transformer. The control signal for the DC filament supply is transmitted to it with an optical fibre link. The absence of line frequency components in the mass spectrometer signal can be seen in the noise spectra in fig.3. There are two ways of controlling the electron gun at high frequencies: one can modulate the emission of the filament by a Wehnelt electrode or by changing the power density of the electron beam. We have chosen for power density modulation because the Wehnelt method requires the filament to be operated at an higher bias current which reduces the lifetime of the filament. The power density modulation is realized by modulating the amplitude of an high frequency sweep of the electron beam over the melt. The high sweep frequency we use (6.4 kHz, locked to the line frequency) guarantees that the power electron beam is effectively smeared out over the melt [6]. The change in the power density of the electron beam results in a change in the evaporation rate. The sweeping is done by modulating the -10kV high voltage of the electron gun with a

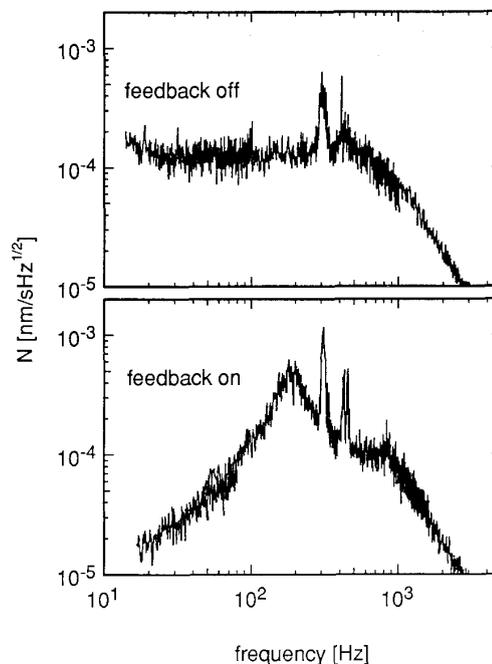


Figure 3. Evaporation rate noise spectral density N (in $\text{nm/sHz}^{1/2}$), as measured by the mass spectrometer, versus frequency f (in Hz). This noise spectrum is recorded for copper, at an average rate of 0.035 nm/s. The electron was equipped with a DC filament supply. The upper curve is recorded with the feedback off, the lower with the feedback on. This spectrum shows no harmonics of the line frequency because a DC filament supply has been used. The suppression of the shot noise in the mass spectrometer signal with feedback on is clearly visible up to 100 Hz, indicating the bandwidth of the feedback loop. The peaks around 300 and 400 Hz are caused by pick-up of acoustic vibrations in the evaporation chamber and can be attributed to the turbo molecular pumps.

high voltage sine (average amplitude is 700V_{RMS}). The sweeping can in principle also be done by coreless magnet coils, or high voltage deflection plates. Magnet coils are however difficult to incorporate in this type of electron gun and would not be effective at high frequencies due to eddy currents induced in the copper cooling block. A disadvantage of deflection plates is that they might be sputtered by ions generated by the electron beam. A secondary advantage of the high frequency is that it makes the evaporation process more stable because of the reduction of the partial pressure of the evaporant just above the melt [6]. The transfer function of the modulation of the sweep for a copper melt is shown in figure 4. The -3 dB/octave fall above 2 Hz is compensated for by a +3 dB/octave amplifier. We associate this cut off frequency with the thermal constant of the melt. The cut off frequency of the evaporation process itself is visible as a kink in the transfer function near 200 Hz.

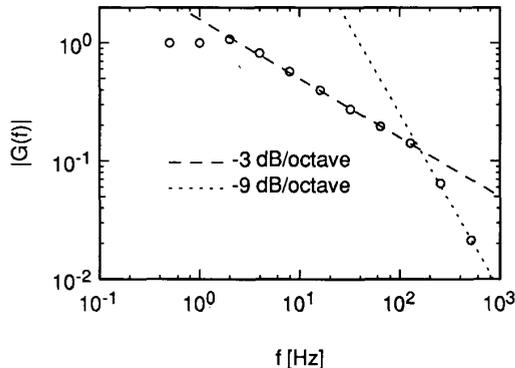


Figure 4. Amplitude of the complex transfer function $G(f)$ of the 6.4 kHz high voltage modulator versus frequency (in Hz). The evaporation material is copper. We associate the kink at 2 Hz with the thermal time constant of the melt and the one at 200 Hz with the cut-off frequency of the evaporation process.

Feedback system performance

The system as described above has very stable low frequency properties and a high bandwidth. Stable operation has been tested in the range from 0.001 to 1 nm/s for different materials (Y, Cu, BaF₂). A typical evaporation rate we use is 0.05 nm/s. The long term drift as measured with an independent quartz crystal monitor is typically 1-2% per hour for copper (fig.5) and can mainly be attributed to the thermal drift of the gain of the mass spectrometer SEM (fig 6). Drift due to crater formation in the melt could be ruled out by recording the evaporation rate measured by the quartz crystal as function of time and remelting the melt during that experiment several times. No deviation from a curve similar to that of fig.5 was seen. The large

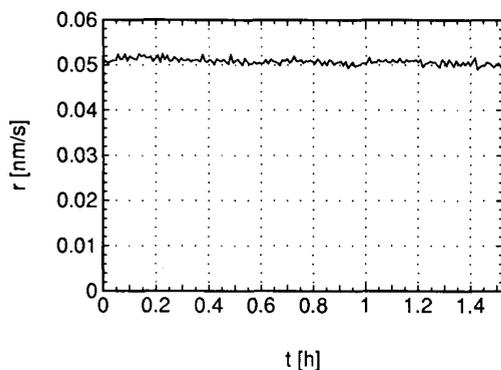


Figure 5. Copper evaporation rate (in nm/s) vs. time t (in hours). The evaporation rate was measured with an independent quartz crystal thickness monitor, by recording the thickness of the accumulated layer every 100 seconds. An average drift of the evaporation rate of 1.4% caused by temperature drift of the mass spectrometer is visible.

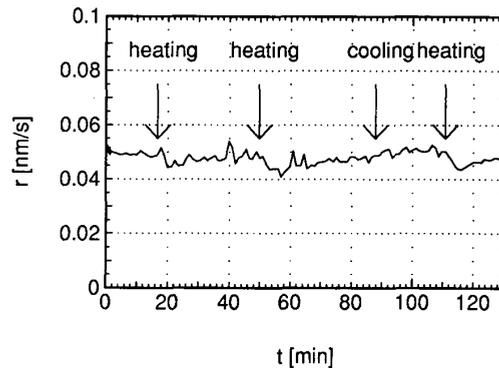


Figure 6. Copper evaporation rate (in nm/s) vs. time t (in hours). The evaporation rate was measured with an independent quartz crystal thickness monitor, by recording the thickness of the accumulated layer every 100 seconds. The arrows indicate where the enclosure of the mass spectrometer was deliberately heated or cooled for a couple of minutes.

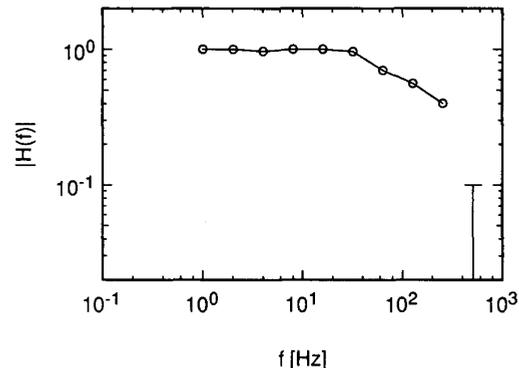


Figure 7. Amplitude of the complex transfer function $H(f)$ of the feedback system vs. frequency f (in Hz), indicating a bandwidth larger than 100 Hz.

temperature drift of the room (2°C per hour) which renders the drift caused by the power dissipation of the electron gun supplies. At the moment a water cooling is added to the mass spectrometer enclosure to stabilize its temperature.

The noise level visible in the curve without feedback at low frequencies (fig 3) is the shot noise in the ion current that impinges the SEM. The noise falls off at 1 kHz because the bandwidth of the electrometer is limited to 1 kHz. The reduction of the noise by the feedback system is clearly visible up to 100 Hz. At low frequencies the shot noise present in the ion current is imposed on the evaporation flux with reversed sign. The transfer function of the feedback system is shown in figure 7. Figure 8 shows the step response of the feedback system, indicating the very fast response time below 10 milliseconds. From these measurements we estimate the bandwidth of the feedback system to be at least 100 Hz. The peaks around 300 Hz in the noise spectrum can be related to microphonic pick up of vibrations in the MBE system which are caused by the turbo molecular pumps. The pick-up of these signals limits at the moment the bandwidth of the feedback system together with the time constant of the evaporation process. Attempts are being made at the moment to increase the signal to noise ratio and to diminish the relative size of the acoustic peaks by adding permanent magnets to the ionization chamber of the mass spectrometer. The feedback system has been successfully used for the fabrication of superconducting YBa₂Cu₃O₇ films with T_c 's up to 88 K by the BaF₂ method and up to 80 K by the ozone method.

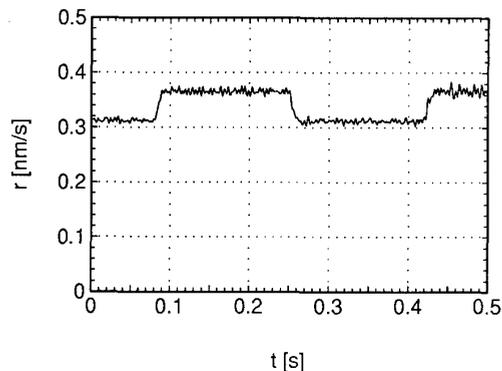


Figure 8. Step response of the evaporation rate r (in nm/s) vs. time t (in seconds) with feedback applied, as measured by the mass spectrometer. The step response time of the feedback system is shorter than 10 ms, indicating a bandwidth larger than 100 Hz.

Summary

A high bandwidth feedback system for electron gun evaporation has been developed and tested. This feedback system has the capability of controlling the evaporation flux from the electron gun up to the cut-off frequency of the evaporation process. It can control the average flux on the time scale of the growth of one unit cell well within 1% and it has the possibility to modulate the evaporation flux accurately on very short time scales. The system has been successfully tested with evaporation rates of 0.001 to 1 nm/s, for various materials and under ozone and oxygen pressures up to 10^{-4} mbar. Long term drift of the feedback system is typically 1-2 % per hour for copper and is dominated by thermal drift of the SEM of the mass spectrometer. We estimate the bandwidth of the feedback loop to be larger than 100 Hz from noise, transfer function and step response measurements.

Acknowledgements

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References

1. R.M.Silver, A.B.Berezin, M.Wendman, and A.L.de Lozanne, "As-deposited superconducting Y-Ba-Cu-O thin films on Si, Al_2O_3 , and $SrTiO_3$ substrates", *Appl.Phys.Lett.*, Vol.52, 2174 (1988).
2. N.Missert, R.Hammond, J.E.Mooij, V.Matijasevic, P.Rosenthal, T.H.Geballe, A.Kapitulnik, M.R.Beasley, S.S.Laderman, C.Lu, E.Garwin, and R.Barton, "In situ growth of superconducting YBaCuO using reactive electron-beam evaporation", *IEEE Trans.Magn.*, Vol.25, 2418 (1989).
3. D.D.Berkley, B.R.Johnson, N.Anand, K.M.Beauchamp, L.E.Conroy, A.M.Goldman, J.Maps, K.Mauersberger, M.L.Mecartney, J.Morton, M.Tuominen, and Y-J.Zhang, "In situ formation of superconducting $YBa_2Cu_3O_{7-x}$ thin films using pure ozone vapor oxidation", *Appl. Phys. Lett.*, Vol.53, 1973 (1988).
4. N.G.Chew, S.W.Goodyear, J.A.Edwards, J.S.Satchell, S.E.Blenkinsop and R.G.Humphreys, "The effect of small changes in composition on the electrical and structural properties of $YBa_2Cu_3O_7$ thin films", submitted to *Appl. Phys. Lett.*.
5. V.Matijasevic, P.Rosenthal, K.Shinohara, A.F.Marshall, R.H.Hammond, and M.R.Beasley, "Reactive coevaporation of YBaCuO superconducting films", submitted to *J.of Mat.Res.*
6. A.J.G.Schellingerhout, M.A.Janocko, T.M.Klapwijk, and J.E.Mooij, "Rate control for electron gun evaporation", *Rev.Sci.Instrum.*, Vol.60, 1177 (1989).