Mass spectrometer controlled electron beam evaporation synthesis of multilayered materials

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A UHV apparatus equipped with two high-power electron guns is extended with a multichannel quadrupole mass spectrometer which accurately controls the deposition rate. A rotating substrate holder is used for deposition of multilayers at room temperature; a fixed substrate holder enables one to prepare layered structures at liquid N_2 temperature. A home-built load lock in combination with an extended travel sample manipulator permits a rapid change of the samples without breaking vacuum. The system has been used to deposit Nb/Cu and Pb/Ge multilayers and preliminary structural measurements indicate the samples are of high quality.

INTRODUCTION

The development of sophisticated evaporation and sputtering systems has given a new impetus in the preparation and study of multilayered materials. These thin-film structures, fabricated by repeated alternate deposition of very thin layers (thickness 0.2–50 nm), have unique physical properties.[1] They are of interest for an increasing number of scientific and technical applications as semiconductor devices, as x-ray mirrors, as samples for the study of new magnetic and superconducting properties, as starting materials for alloying following fast pulsed laser heating or ion-beam mixing, and as samples for the study of diffusion and melting. Different methods have been used to prepare these materials: molecular beam epitaxy,[2] thermal vapor deposition,[3,4] dc and rf sputtering,[5] and ion beam sputtering.[6]

In this paper we report on a versatile ultrahigh-vacuum (UHV) electron beam gun apparatus with a great flexibility (e.g., several materials can be deposited under identical conditions) and an excellent control of the deposition parameters. The main features of the system can be summarized as follows: (i) Two electron beam guns enabling a high-rate, high-purity deposition of metals, semiconductors, and insulators; (ii) The possibility to evaporate five different materials in a single vacuum run; (iii) A rotating substrate holder with the possibility of changing the substrate and metal masks during the same vacuum run and to synthesize up to four identical samples. This turntable driven by a stepping motor and two shutters are computer controlled; (iv) A fixed substrate holder which can be cooled down to liquid N_2 temperature; (v) A load lock and sample manipulator compatible with both substrate holders; (vi) A programmable multichannel mass spectrometer to monitor and accurately control the deposition rate and to analyze the residual gases during deposition; (vii) Two crystal sensors to calibrate the deposition rate and to control the film thickness.

I. DESCRIPTION OF THE APPARATUS

A. Vacuum chamber and system

The water-cooled cylindrical stainless-steel bell jar has a diameter of 0.45 m, a height of 1.5 m, and is sealed with two Wheeler flanges and 38 conflat flanges. Figure 1 shows a schematic view of the evaporation chamber.[7] The bottom of the vertical cylinder is connected to the pumping system and two electron guns are mounted on a baseplate. The top plate contains rotary feedthroughs (not shown) and a removable flange. This flange supports either the rotating or the liquid-N_2 cooled substrate assembly (see further). The cylindrical wall contains the rotary feedthroughs for the computer-controlled shutters and shields, the electrical feedthroughs for the electron guns and crystal sensors, a removable flange for the mass spectrometer, a load-lock assembly, and a large viewing port. The system is successively pumped with a Venturipump, three Vacsorb pumps, seven-110-l/s ion pumps, and a titanium sublimator, surrounded by a liquid-N_2-cooled shield.[7] After baking out the system, a base pressure of 1×10^{-10} Torr is obtained. The background pressure during deposition is in the 10^{-8}–10^{-9}-Torr range and depends on the evaporants, deposition rates, and substrate temperature.

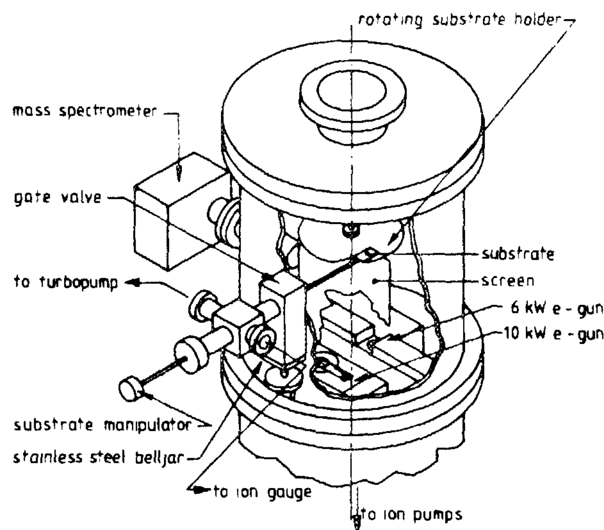

FIG. 1. Schematic view of the UHV chamber.

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B. Electron beam guns

A 6-kW four-hearth UHV and a single-hearth 10-kW electron gun[7] are mounted on the baseplate. The guns are equipped with deflection coils allowing a sweep of the electron beam in both lateral and longitudinal directions. To avoid mutual contamination of the source material and mutual distortion of the fields produced by the permanent magnets, an iron shield is mounted between the water-cooled copper hearth of the guns. Focusing of the electron beams is facilitated by mirrors mounted inside the chamber. Each gun is monitored by a quartz thickness sensor.

C. Substrate holders and load-lock assembly

For the preparation of the multilayers at room temperature, a stainless-steel substrate carrier holder driven by a stepping motor is used [Fig. 2(a)]. Using an extended travel manipulator the substrate carrier can be withdrawn into a load lock which can be isolated from the UHV chamber by a gate valve. The manipulator, the substrate carrier, and the disk are designed in such a way that complete blind operation is possible. A set of metallic masks screwed to the turntable enables the preparation of different material configurations in the same vacuum run. In order to avoid cross contamination of the material fluxes, a vertical screen is mounted between the substrate carrier holder and the electrón beam guns. Two shutters installed on each side of the screen enable the outgassing of the sources and the precontrol of the evaporation rate. The turntable spins with a constant velocity above the screen, carrying the substrate alternatively from one material flux to the other. The velocity of the disk, the number of bilayers, and the opening and the closing of the shutters are computer controlled and displayed during the fabrication of the multilayer.

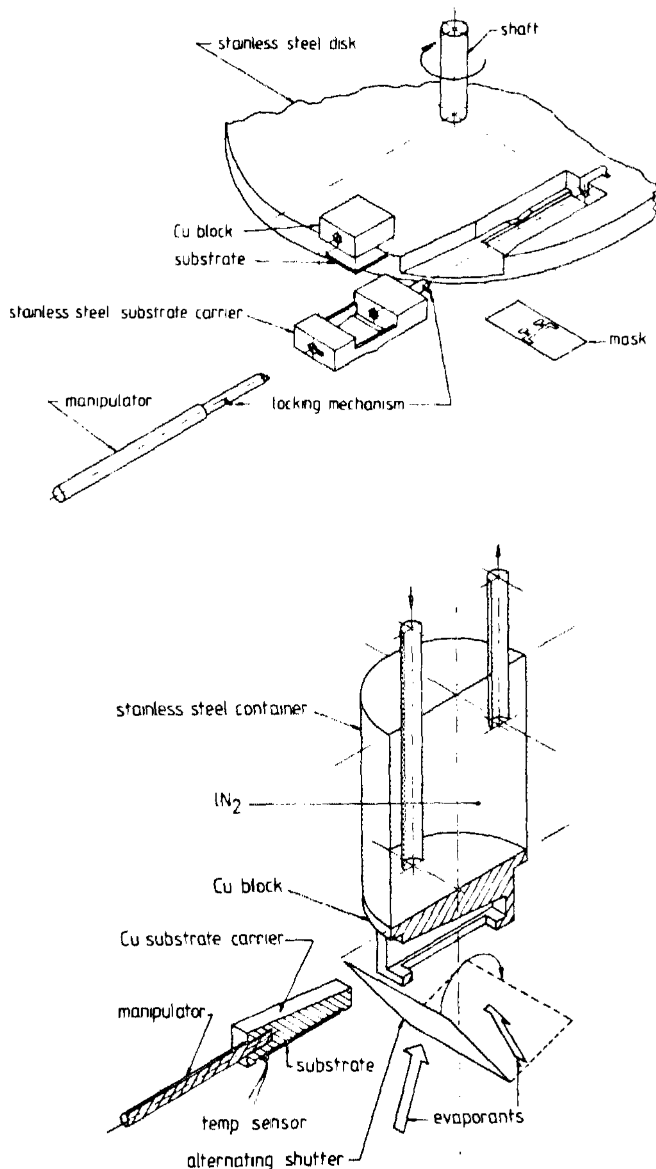

FIG. 2. (a) Detailed drawing of the turntable. (b) Detailed drawing of the cooled substrate holder.

Another substrate holder mounted on the removable flange is used to prepare multilayers on liquid-nitrogen-cooled substrates [Fig. 2(b)]. It consists of a stainless-steel container (liquid N_2) and a copper block in which a conical-shaped copper substrate carrier can be positioned using the manipulator. Test runs showed that the heat flux produced by the electron guns increased the temperature of the sapphire substrate by a negligible amount (<0.5 K). These tests consisted in measuring the resistance variation of a thin calibrated Cu film (covered with Ge) during the growth of the multilayer. The composition-modulated structures are produced by a computer-controlled shutter mechanism (shuttering time 0.2 s) which alternatively shadows off one material flux.

II. DEPOSITION CONTROL

Two deposition techniques are used for the fabrication of multilayers, namely, the constant thickness mode and the constant time mode. We use the second mode in which the deposition time is fixed for each sublayer and, therefore, the accuracy of the sublayer thickness is governed by the stability of the deposition rates. Most workers use crystal sensors to monitor the electron beam power supply for stabilizing the deposition rate. The resolution of a crystal sensor, however, is insufficient for short modulation repeat lengths ($\Lambda < 1$ nm), as mentioned in Ref. 6, because the opening time equals the integration time of the sensor (0.3–1s). For this reason we use a mass spectrometer (integration time 3 ms) to monitor the rate. This method is only useful if one can adjust the evaporation rate in the same time interval. To achieve this fast adjustment we vary the amplitude of the electron beam sweep, thereby changing the power density in the crucible and, therefore, the evaporation rate.

The analyzer of quadrupole mass spectrometer[8] (MSM) is mounted horizontally above the guns (distance 25 cm) as seen in Fig. 1. The analyzer of the MSM is surrounded by a water-cooled shield to prevent contamination from the evaporants causing electrical shorts and malfunctioning. The water cooling maintains a stationary temperature environment and prevents drift of the analyzer electronics due to heating by the high-power electron beam guns. Two small holes (~ 2 mm) in the shield allow the evaporants to enter the center of the ion source of the analyzer.

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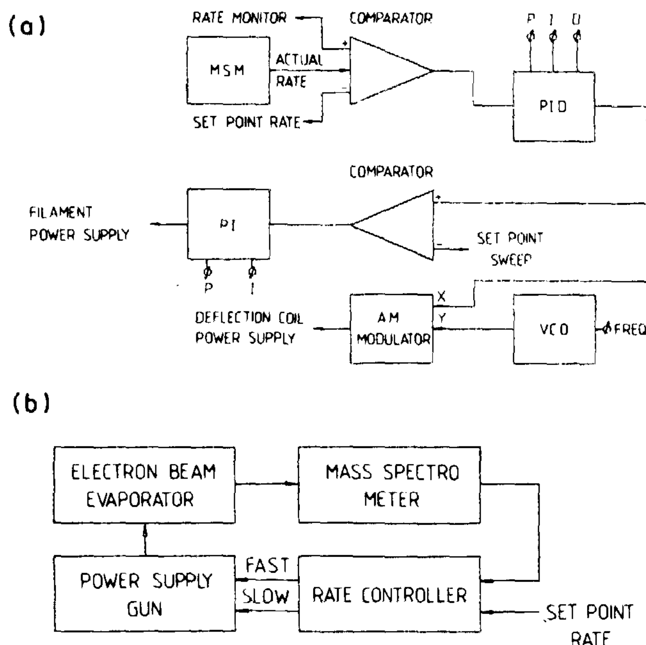

FIG. 3. (a) Block diagram of the deposition rate controller. (b) Schematic of the link between the different devices.

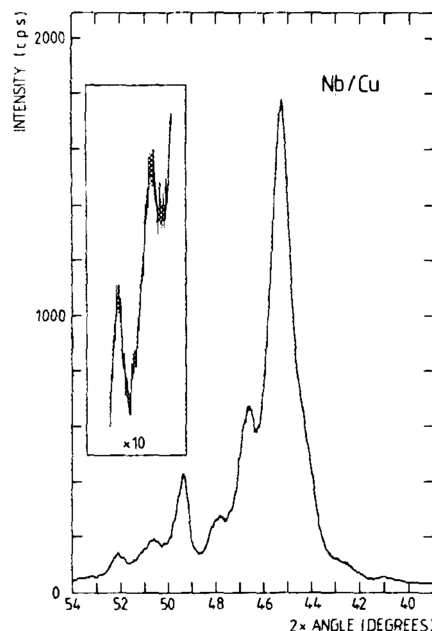

FIG. 5. X-ray θ–2θ scan of a Nb/Cu multilayer.

After two years, the contamination of the ion source is still negligible.

The rate controller is diagrammed in Fig. 3(a). The MSM output signal is compared with the set-point rate and further amplified to give an error signal. This error signal is then fed to the PID (proportional, integrating, and differentiating) regulator where it is transformed into the control signal. The set-point rate and set-point sweep bias voltages are supplied by precision voltage references.

The error signal is used in two ways. First, to vary the input power of the electron beam by changing the current through the filament. This is satisfactory for error signals whose main frequency components are less than 1 Hz. The filament emission response is no longer in phase with the change in heating current at a frequency of about 1 Hz. This results in the feedback changing from negative to positive and wild oscillations of the filament current occur. The filament controller which provides both linear proportional and integral control (PI) is used to eliminate these oscillations. The integration is adjustable over a wide range to accommodate a variety of control situations.

Second, the amplitude of the beam sweep is modulated, thereby changing the power density in the crucible and, therefore, the rate by up to 50%. This second feedback loop[9] (the sweep controller) consists of a PID controller, a voltage controlled oscillator (VCO), an analog multiplier, and the

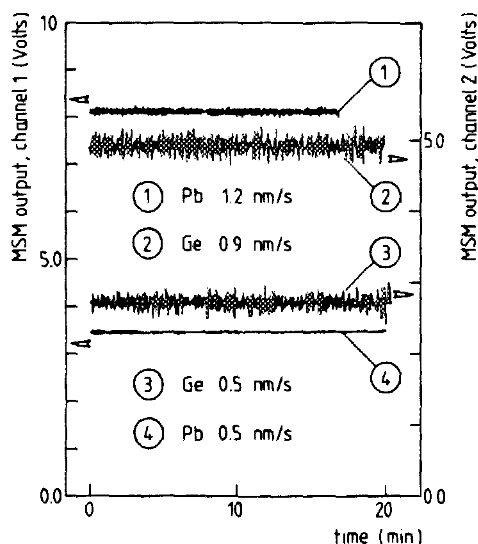

FIG. 4. Curves of the instantaneous rate stability for Pb and Ge.

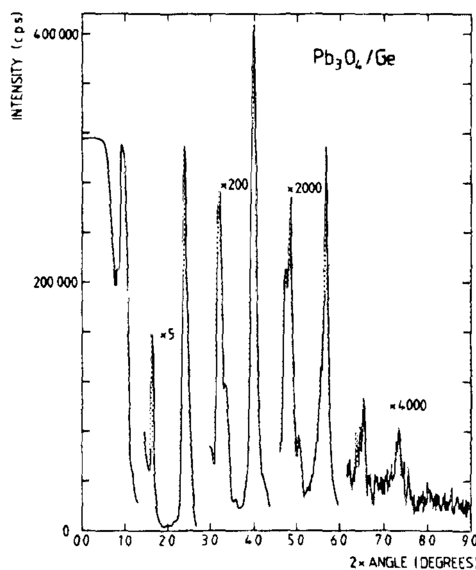

FIG. 6. Glancing angle x-ray θ–2θ scan of a Pb_3O_4/Ge multilayer.

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power supply of the lateral deflection coil. This feedback loop has been designed to deal with deposition rate fluctuations up to 10 Hz. A triangular current waveform, supplied to the deflection coils, is used to sweep the electron beam on the molten metal being evaporated. The frequency of the triangular waveform is adjustable from 10 to 100 Hz by means of a VCO.

The interaction [Fig. 3(b)] between the filament and sweep controller works as follows. When the deposition rate is too small, the sweep controller decreases the sweep voltage in order to increase the rate. This results in a rise of the MSM output which is fed back to the rate controller. The filament controller will increase the filament current enabling the sweep voltage to shift back gradually to the middle of its range.

III. SYSTEM PERFORMANCE

The performance of this rate controller depends on the evaporation rate, the kind of material, and the level of the melt in the crucible. For a crucible–substrate distance of 35 cm the fluctuations on the instantaneous rate for Pb is 1–2% for low deposition rates (0.1–1 nm/s) and 2–5% for high deposition rates (1–4 nm/s). For Ge we achieve, respectively, 3–5% for low- and 5–8% for high-deposition rates (see Fig. 4). The rate fluctuations read simultaneously from a quartz sensor are typically 3–5 times smaller.

The long-time stability is very important and depends essentially on (i) the resistance variation of the sweep coils due to the temperature increase (this effect is minimized by cooling the coils with air), (ii) the long-time stability of the MSM electronics (the manufacturer guarantees 1×10^{-2} m u/°C drift for the analyzer and 10 ppm/°C for the electrometer amplifier), (iii) the temperature drift of the set-point rate reference: 3 ppm/°C.

Different materials have been evaporated in the vacuum system such as Nb, Cu, Si, Ge, Ti, Al, Sn, Pb, and SiO_2. The superconducting transition temperature T_c of Nb films condensed at a rate of 5 nm/s on a substrate at room temperature was 9.2 K, indicating the very clean conditions during evaporation.

We also prepared a series of metal–metal superlattices of Nb/Cu on substrates at room temperature.[10] The θ–2θ scan in Fig. 5 shows high-angle superlattice peaks, resulting from the coherent scattering between the layers ($\Lambda = 8.2$ nm).

Another series of Pb_3O_4/Ge and Pb/Ge multilayers were condensed on sapphire substrates cooled to liquid nitrogen. A θ–2θ scan of a Pb_3O_4/Ge multilayer showing 9–11 peaks at glancing angle is shown in Fig. 6 ($\Lambda = 10.8$ nm). This high number of small-angle peaks (usually one observes only 2–4 peaks) is due to the accurate control of the modulation repeat length and the large difference in mean atomic scattering factor of Ge(32) and Pb(82). A profile of the same multilayer taken in a TEM[11] is shown in Fig. 7. We clearly observe layered growth and the typical micrograph image of amorphous Ge and monocrystalline Pb_3O_4 (Moiré pattern). A detailed study of the structural analysis and the superconducting measurements of the Pb/Ge system will be published elsewhere.

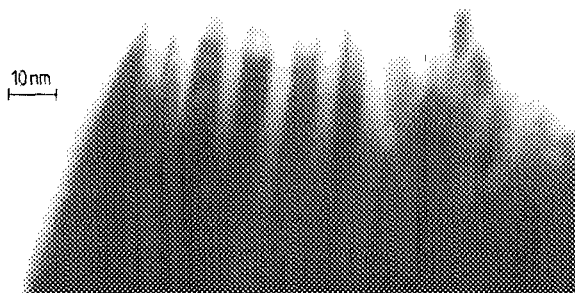

FIG. 7. Electron micrograph profile of a Pb_3O_4/Ge multilayer.

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