Real-time simultaneous optical-based flux monitoring of Al, Ga, and In using atomic absorption for molecular beam epitaxy

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We have developed a multichannel atomic absorption measurement system for real-time simultaneous monitoring of Al, Ga, and In molecular beam fluxes. In our configuration, distinct atomic emission lines from three hollow cathode lamps are combined into one beam, thus requiring only one pair of through view ports for the optical probe beam. Based on the dual beam optical configuration, the reference arm compensates for intensity drift of the light sources. In this work, we demonstrate the use of reflection high energy electron diffraction oscillations for calibrating the absorption signal. © *1996 American Vacuum Society.*

I. INTRODUCTION

Molecular beam epitaxy (MBE) growth is currently limited by the inability to continuously monitor the epilayer growth rate and composition. In order to increase growth reliability, a MBE system must monitor growth in real-time, and have the ability to compensate for growth rate drift. It has long been recognized that atomic absorption can be used as a tool for monitoring and control of material deposition processes.[1] Recently, because of a need for better noninvasive real-time feedback during the growth process, increased attention has been turned toward monitoring techniques using atomic absorption of the molecular beam flux.[2–5] In this work, we have developed an integrated optical system which can monitor the atomic absorption of Al, Ga, and In molecular beams (group III elements) simultaneously. With this system, just one pair of through optical ports is necessary. Because our optical-based flux monitor (OFM) independently measures the atomic absorption of each molecular beam, it can simultaneously determine the flux of each of the group III elements. In this article, we compare the atomic absorption intensity of the molecular beam to the growth rate measured by reflection high energy electron diffraction (RHEED) oscillations.

II. ATOMIC ABSORPTION SETUP

The optical setup, as shown in Fig. 1, is a multiwavelength dual beam configuration with two passes of the probe beam through the growth chamber. The atomic emission lines from the three hollow cathode lamps (HCL) are monitored. The lamps are operated under constant current mode. Each atomic line is selected by a narrow band-pass optical filter (10 nm bandwidth) corresponding to emission lines for Al, Ga, and In centered at 395, 417, and 410 nm, respectively. Each emission line is modulated by a mechanical chopper at a different frequency and then combined through a trifurcating optical fiber bundle. The combined beam is then split by a beam splitter into two arms. Each arm is collected by a multimode (1 mm core diameter) optical fiber. One is the reference arm; the other, the probe arm. The signals are detected using two photomultiplier tubes and six lock-in amplifiers.

In our experimental arrangement, we modified one pair of 5° glancing-angle optical ports on our Varian GEN II MBE system for use with the OFM. We do not have to sacrifice the ports used by the RHEED system. The optical ports on the MBE system are 1.5 in. in diameter and approximately 3 ft. apart. Both ports have UHV mechanical shutters. The probe beam passes through the growth chamber in front of the substrate and is then reflected back by a pair of flat mirrors. The returning probe beam is collected by another optical fiber (1.5 mm core diameter) which carries the beam to the signal detector. The advantage of the dual pass configuration is increased absorption. Figure 2 shows the absorption of Al, Ga, and In at the indicated growth rates. Each trace corresponds to the source shutter operations of close/open/close. The absolute growth rates were calibrated by RHEED oscillations.

For data acquisitions, we use a Macintosh Quadra 840AV

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[a] Currently with Pacific Lightwave, 725 Anchor Way, Carlsbad, CA 92008.

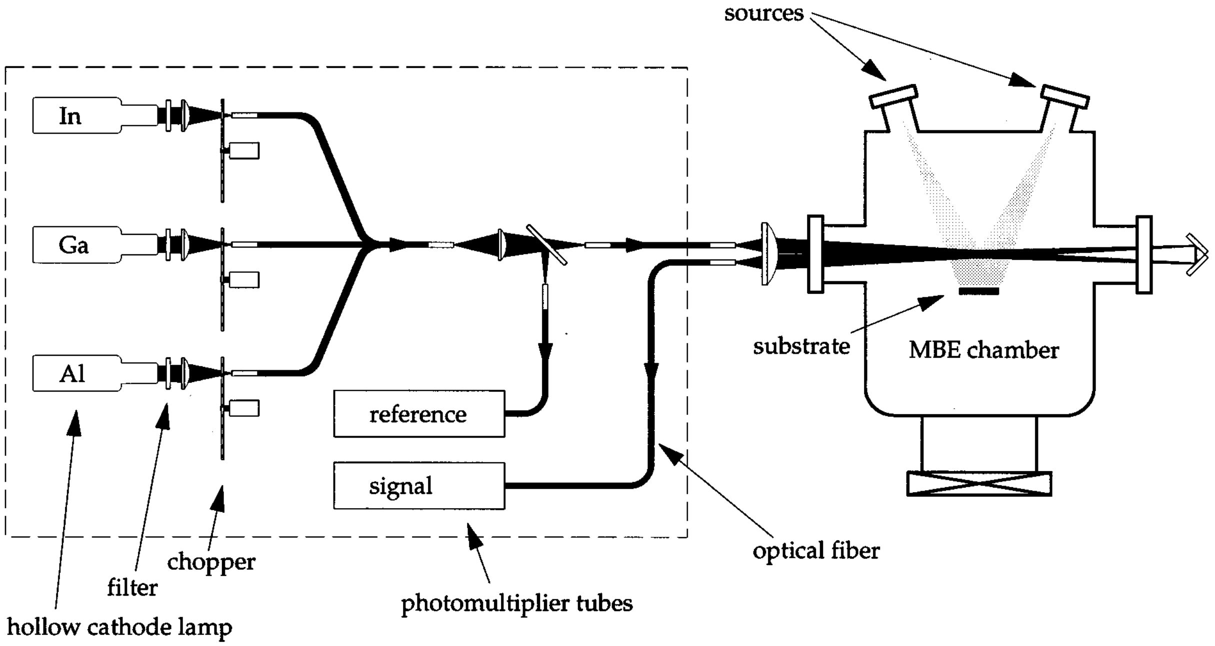

FIG. 1. Schematic diagram of the atomic absorption flux monitor. The dashed line encloses the components of the system that are mounted on a 2 ft.×3 ft. optical table.

computer equipped with a multichannel 16 bit analog to digital converter board. The signal for each atomic line is defined as

$$signal \equiv \frac{I_{\text{probe}}}{I_{\text{ref}}}, \tag{1}$$

where I_{probe} and I_{ref} are the measured intensity signals from the respective probe and reference lock-in amplifiers. This referencing technique compensates for the drifting output of the HCL.

Without using heated view ports, As coating causes the *signal* to decrease at a rate of about 0.1%/min. With the view ports heated to 250–300 °C range, the drift rate during growth is reduced to about 7×10^{-3}%/min. We believe that at this temperature range, there is still some coating of the view ports during growth, because during our long term base line test (over 10 h) with all the sources off, the drift rate is less than 4×10^{-4}%/min. This systematic drift can also be removed from the data by linearly fitting to the drifting base line.

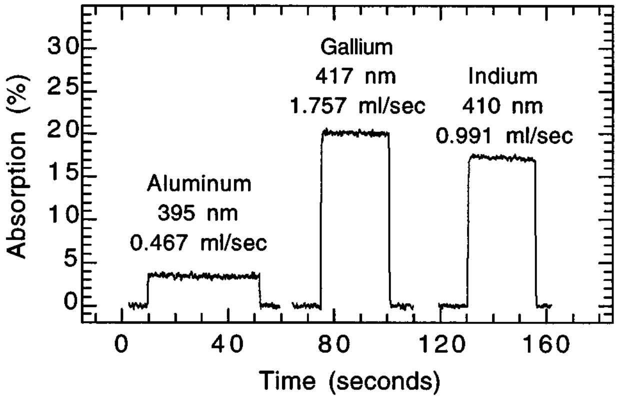

FIG. 2. Absorption signals for three different sources. The growth rates were determined by observing RHEED intensity oscillations. The monitored wavelengths are as indicated. The data were taken at 200 samples/s. The noise is at about 0.1% level.

Each I_{probe} is composed of three components,

$$I_{\text{probe}} = R + T_0 - A, \tag{2}$$

where R is the unintentional reflected radiation from the view port on the fiber side of the growth chamber, T_0 is the transmitted radiation with no beam flux, and A is the radiation absorbed by the beam flux. When one of the view port shutters is closed, only the stray reflection from the view port is observed, $I_{\text{probe}} = R$. When the probe beam passes through the growth chamber while a particular material species is not being grown, $I_{\text{probe}} = R + T_0$. The useful parameter is the normalized absorption which is defined as

$$\gamma \equiv \frac{A}{T_0}. \tag{3}$$

Its relationship to the growth rate will be discussed in Sec. III. Under the growth condition that re-evaporation of the monitored atomic species is negligible, the growth rate is a one-to-one monotonically increasing function of absorption. Fortunately, re-evaporation is not a problem for our growth condition—sufficiently low substrate temperature[6] with As overpressure and growth rate limited by the incident flux of the group III elements.

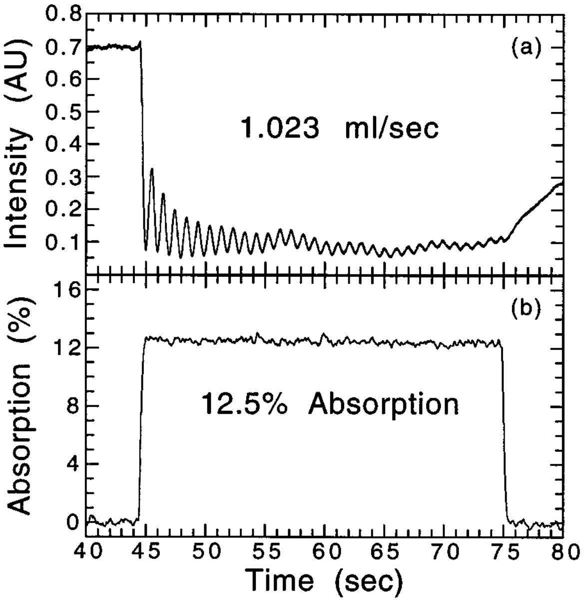

FIG. 3. (a) RHEED intensity oscillations and (b) the Ga atomic absorption signal for GaAs grown at 1.023 ML/s. The absorption and RHEED data were taken simultaneously.

III. CALIBRATION BY RHEED

Since OFM measures the atomic absorption of the molecular beam fluxes and not the growth rates directly, the OFM signals must be calibrated using other techniques. For MBE, RHEED is the most practical choice. During calibration, the sample is not rotated. We position the calibrating RHEED electron beam at the center of the sample where the monitored growth rate should be independent of sample rotation. Typically, sample rotation is preferred during growth because it improves epitaxial uniformity across the wafer. The disadvantage of rotation is that many monitoring techniques which directly probe the substrate such as RHEED, pyrometric interferometry, reflection and transmission spectroscopy, and ellipsometry become more difficult to implement. To partially circumvent this problem, many investigators have synchronized their data acquisition to sample rotation[7] or averaged over several rotation periods.[8] Therefore, the measurement speed for these other techniques is restricted by the period of sample rotation, while an OFM is free from this limitation.

To calibrate the OFM, we measure the beam flux absorption and RHEED oscillation signals simultaneously. Figure 3 shows the RHEED and the Ga absorption data for GaAs calibration. The growth rate in monolayers per second is determined from the period of RHEED oscillations. Unlike the OFM signal, RHEED oscillations decay away over time; therefore, they become ineffective for monitoring growth of thicker layers. In Fig. 4, atomic absorption signals as a function of growth rate for Ga and In are shown. RHEED calibrations were measured on GaAs and InAs substrates, respectively. The lines through the data are the modified Beer's law fits of the form

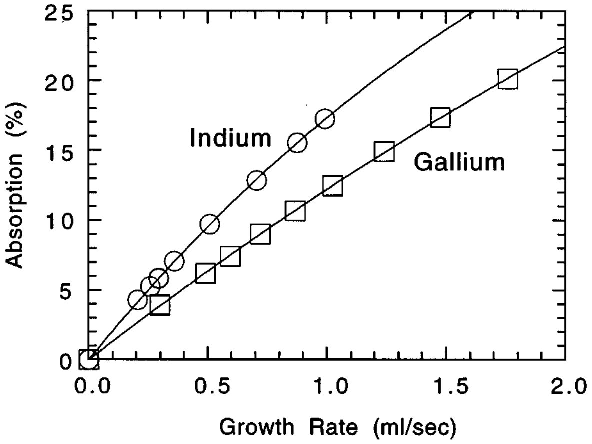

FIG. 4. Absorption vs growth rate for InAs and GaAs. The curve fits shown through the data follow the form of the modified Beer's law given by Eq. (4). This curve deviates from the data by no more than 0.5% over the range of interest. The substrate temperatures were 620 and 520 °C for the GaAs and InAs, respectively.

$$\gamma = 1 - \exp[-(\alpha + \beta r) r], \tag{4}$$

where r is the growth rate, and α and β are the two fit parameters for the growth rate dependent absorption coefficient, $(\alpha+\beta r)$. The growth rate dependence of the absorption coefficient is likely due to (a) the differences between the linewidth of atomic emission for a HCL and the absorption linewidth of the beam flux,[5] and (b) α not being a constant because the beam flux velocity changes with changing cell temperature.[2] The discrepancy between the fit and the data is about 0.5%. However, by assuming a constant absorption coefficient ($\beta=0$) for the fit, the discrepancy between the fit and the data increases significantly to about 2.5%.

IV. CONCLUSION

We have developed a compact multichannel optical-based flux monitoring system for MBE based on atomic absorption. RHEED oscillations were used to calibrate the OFM so our system can accurately monitor in real-time the individual growth rates of Al, Ga, and In.

Several factors limit the accuracy of an OFM. Because atomic absorption is not a direct measurement of growth rate, this type of system is ultimately limited by the accuracy of the growth rate calibration measurement technique; in our case, the limit is the accuracy of the RHEED oscillation calibration. Another source of error for the OFM is the signal drift due to the As coating of the view ports during growth. This problem can be reduced substantially by using heated view ports. Future work on our OFM will include further study of long term stability.

Based on our experience, OFM has a promising future in the area of real-time monitoring and control of MBE growth since the signal is well understood and easy to interpret. OFM is also well suited to control of production MBE systems since the absorption signal increases with higher fluxes and increased molecular beam cross sections.

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