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Silicon Nitride Thin Film Production on Si(111)

by

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Submitted To

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Abstract

The production of silicon nitride films by ammonia decomposition on Si(111)-(7x7) has been studied by high-resolution electron energy loss spectroscopy (HREELS), Auger electron spectroscopy (AES) and low-energy electron diffraction (LEED). Silicon nitride films of ≤ 1 monolayer thickness exhibit a characteristic four mode vibrational spectrum after annealing to 1200 K, while multilayer films produce three vibrational modes at 495, 720 and 1020 cm^{-1} . Upon continued heating of the submonolayer and multilayer nitride films, identical vibrational spectra are obtained, suggesting that both layers form Si_3N_4 . These results are in excellent agreement with the vibrational spectra reported in the literature for Si_3N_4 layers grown using N atoms on the heated Si(111)-(7x7) surface, signifying that identical nitride films can be grown using ammonia. Si_3N_4 films grown on Si(111) are able to chemisorb ammonia at 300 K, showing that coordinatively unsaturated silicon sites are probably present on the film surface. This is in agreement with other studies which indicate that the growth mechanism for these silicon nitride films involves silicon enrichment at the film/vacuum interface.

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I. Introduction

Electronic devices which are able to withstand high temperatures are employed in space exploration, nuclear instrumentation, fossil energy plants and aeropropulsion systems [1]. These technologies make use of standard silicon integrated circuit device technology. The thermal limits of operation are necessarily restricted by the thermal stability of the component materials. Due to its greater thermal stability, silicon nitride films have been found to be a good substitute for silicon dioxide films in high temperature capacitors and resistors as dielectric and/or passivating layers [2].

Several methods of preparing silicon nitride films on Si(111)-(7x7) have been employed previously. Some common chemical species used to grow these films are NH_3 [3-6], N atoms and ions [4,7-11] and NO [12]. To prepare the silicon nitride films, these species are exposed to the silicon surface at high temperatures (900 - 1200 K), or at low temperatures (~300 K) followed by heating. Many similarities exist in the high resolution electron energy loss (HREEL) and Auger spectra and the LEED patterns of these silicon nitride films, independent of the reactant gas employed. A brief summary of the surface characterization of these silicon nitride films on Si(111)-(7x7) follows:

1) HREELS - Silicon nitride films grown from N atom adsorption on a Si(111)-(7x7) surface have been studied previously with HREELS [11]. After annealing to ~1240 K these films exhibit four vibrational modes at 495, 735, 970 and 1105 cm^{-1} . Continued heating to ~1350 K produces a vibrational spectrum with one intense loss at 970 cm^{-1} and, depending on the extent of annealing, two

small vibrational features at 485 and 725 cm^{-1} [11]. An identical three-mode spectrum is obtained after an adsorbed layer of NO is subjected to a similar high-temperature annealing procedure [12].

2) AES and Other Studies - Upon nitridation, a new Auger feature develops at ~ 84 eV which has been proposed to result from a silicon nitride species [3,7-10,12]. The characteristic Si(LVV) transition at 91 eV is observed to decrease in energy by 1.5 - 2.0 eV after the growth of the first silicon nitride monolayer (ML) [9,10,12], while the N(KLL) Auger feature at ~ 368 eV remains fixed in energy [3,7-10,12].

Other studies have revealed that the silicon nitride film grows via the production of silicon sites at the silicon nitride/vacuum interface. A recent synchrotron XPS study of silicon nitride films, grown on Si(100)-(2x1) [13], monitored the Si(2p) and N(1s) intensities as a function of the photon energy. By tuning the x-ray energy, the energy of the escaping photoelectron could be varied, thus varying its escape depth and hence the sampling depth. These results indicated that a monolayer of silicon exists on top of a stoichiometric silicon nitride film grown on Si(100)-(2x1). This conclusion is in agreement with a previous nuclear microanalysis study [5]. In this work, $^{14}\text{NH}_3$ and $^{15}\text{NH}_3$ were used to prepare sandwich layers of isotopically dissimilar silicon nitride films. The layers were analyzed before and after reactive chemical etching of the interface. After determining the concentration of isotopically labeled nitrogen at various depths in the film, the authors concluded that the growth mechanism resulted from the appearance of silicon atoms at the film/vacuum interface, and not

from a uni-directional step-by-step motion of nitrogen atoms through the silicon nitride film to the buried silicon nitride/silicon interface [5].

3) LEED - Two different LEED patterns have been observed from the nitrated Si(111)-(7x7) surface, depending on the annealing temperature and the surface carbon contamination [3,4,6-12]. An (8x8) pattern has been observed for films after annealing between ~1000 - 1300 K (hereafter referred to as the low-temperature nitride structure) and a "quadruplet" LEED pattern is reported for silicon nitride films which are annealed above ~1250 K. The "quadruplet" pattern occurs on films contaminated with a small amount of carbon impurity (~3 - 5%) [10].

In this study we have used HREELS, AES and LEED to examine thin films of silicon nitride prepared by the thermal decomposition of ammonia on a Si(111)-(7x7) surface. These silicon nitride films exhibit vibrational spectra which are identical to the films prepared by adsorbing nitrogen atoms on a Si(111)-(7x7) surface and annealing to ~1240 K [11]. Continued annealing of our films produced a characteristic three mode vibrational spectrum which has been observed from silicon nitride films prepared at high-temperature (~1350 K) from N atoms and NO adsorbates on Si(111)-(7x7) [11,12]. This three mode spectrum is obtained on silicon nitride films above and below one monolayer coverage, and is believed to be characteristic of bulk Si₃N₄ formation since a film prepared this way is thermally stable and the HREEL spectrum is independent of the reactant gas employed. Using vibrational analysis to monitor ammonia adsorption, we are

able to show that silicon sites capable of bonding ammonia on the surface of the growing nitride film at 300 K are available at various stages of nitridation.

II. Experimental

The experiments reported in this study were performed in a stainless steel ultra-high vacuum chamber described in detail previously [14]. The base pressure of this system was below 5×10^{-10} mbar prior to the preparation of the silicon nitride layers.

The instruments employed in this study are a Leybold-Heraeus double-pass ELS-22 high-resolution electron energy loss spectrometer (HREELS) for vibrational analysis and a Perkin-Elmer single-pass Auger electron spectrometer (AES) for elemental analysis. The HREEL spectra were recorded in the specular direction with a primary beam energy of 6.2 eV and a typical resolution of 90 cm^{-1} . The Auger spectrometer was operated with a 2.5 - 3 keV primary electron beam and a modulation voltage of 3 V.

The silicon nitride films were prepared by dosing a clean Si(111)-(7x7) surface with molecular ammonia. Two thermal procedures were used to prepare the layers. In the first method the Si(111)-(7x7) was dosed with $1 \times 10^{15} \text{ NH}_3/\text{cm}^2$ via a calibrated microcapillary array [15] while the crystal was held at 90 K. This layer was then heated to 900 K to desorb all the chemisorbed hydrogen prior to continued cycles of exposure to ammonia at 90 K. In an alternate method, ammonia was effused continually from the doser [16] onto the Si(111)-(7x7)

surface which was held at 900 K. All of the nitrated layers were subjected to a final heating to 1200 K before recording HREEL or Auger spectra.

III. Results and Discussion

A) The Onset of Silicon Nitride Growth on Si(111)-(7x7).

Figure 1 shows HREEL spectra taken during the growth of silicon nitride films on a Si(111)-(7x7) surface as monitored by HREELS and AES. The early stages of nitridation are observed to produce an intense four peak vibrational spectrum (note an expansion factor of only 75), while Auger spectroscopy reveals only a small N(KLL)/Si(LVV) ratio of ≤ 0.031 [Figs. 1(a-b)]. The four vibrational losses are at 480, 700, 965 and 1090 cm^{-1} . These modes intensify and become better resolved as the nitridation continues. When a N(KLL)/Si(LVV) ratio of 0.085 is reached, a four peak vibrational spectrum is fully developed with losses at 495, 720, 965 and 1125 cm^{-1} [Fig. 1(d)]. As mentioned in the Introduction section, an identical vibrational spectrum has been observed for silicon nitride films grown after the thermal treatment of a layer produced by exposure of a Si(111)-(7x7) surface to atomic nitrogen [11]. This strongly suggests that silicon nitride films grown from either N atoms or NH_3 yield identical surface nitride films on the Si(111)-(7x7) surface. For higher nitrogen coverages, the relative intensity of the 965 cm^{-1} mode decreases compared to the other three modes as seen in Figure 1.

B) The Development of Silicon Nitride Monolayers on Si(111)-(7x7)

Silicon nitride film growth on a silicon substrate is known to proceed with a rapid growth rate for film thicknesses up to one monolayer, while further nitridation occurs very slowly [9,10,17-21]. This is observed on both the Si(111) [9,10] and Si(100) [17-21] surfaces, regardless of the N-containing reactant gas employed, and is characteristic of the self-limiting growth and passivating properties of silicon nitride films [22].

A qualitative indication of the film thicknesses achieved in this study can be obtained by comparing our exposure conditions to those of Kubler et al. [20]. Their study shows that the fast stage of nitridation occurs below an exposure of ~200 L NH₃ for a Si(100) crystal held at 973 K. Hence at NH₃ exposures of ~200 L at 973 K, approximately one monolayer coverage is expected to be achieved. An Auger analysis of our films produced by this level of exposure provides a reference point for the approximate calibration of monolayer coverage. For 1×10^{17} NH₃/cm² (200 L), the N(KLL)/Si(KLL) Auger ratio of 0.2 is estimated to correspond to 1 ML Si₃N₄.

The changes observed in the Auger spectra upon nitridation are shown in Fig. 2. Two new features develop on the clean silicon spectrum, one at 84 eV and the other at 379 eV. The 84 eV feature initially appears as a small shoulder in the low energy side of the 92 eV Si(LVV) peak at a N(KLL)/Si(LVV) ratio of 0.028, and develops into a resolvable peak at a N(KLL)/Si(LVV) ratio of 0.085 [Figs. 2(a-b)]. This 84 eV peak continues to intensify upon increased nitridation. The second feature at 379 eV is readily identified as the N(KLL) transition [23]. This

N(KLL) peak is observed to increase in intensity, but remains fixed in energy over the range of nitridation studied in this work. The Si(LVV) peak undergoes a shift in energy from 92 to 90.5 eV as the N(KLL)/Si(LVV) ratio increases from 0.19 to 0.32. A similar Si(LVV) shift was previously interpreted as being indicative of multilayer silicon nitride formation [9,10,12].

Therefore, based on the Si(LVV) energy shift [9,10,12] and the NH₃ exposure [20], it is reasonable to propose that the HREEL spectra shown in Figs. 1(f) and 1(g), after exposure to 244 and 874 L NH₃, respectively, represent the vibrational spectra of silicon nitride layers of ~1 and >1 monolayer in thickness, respectively.

The vibrational spectrum undergoes systematic changes upon increased nitridation. The 965 cm⁻¹ loss is not resolved on layers exhibiting N(KLL)/Si(LVV) ratios ≥ 0.19 [Figs. 1(e-g)]. For the three losses remaining, two remain fixed in energy at 495 and 720 cm⁻¹ and the third continually shifts to lower frequency, from 1095 to 1020 cm⁻¹, as the N(KLL)/Si(LVV) Auger ratio increases to 0.35 [Figs. 1(e-g)].

C) Thermal Effects on Silicon Nitride Films of Varying Thickness.

The effects of heating on the vibrational spectra of a silicon nitride layer possessing an initial N(KLL)/Si(LVV) ratio of 0.020 (<1 ML) are compared to the effects of heating a thicker nitride layer (N(KLL)/Si(LVV) ratio = 0.22) in Figure 3. The thicker nitride layer is found to exhibit a vibrational spectrum which is essentially unaffected by annealing at 1200 K for 15 minutes [Fig. 3A(a-b)]. This

behavior is easily contrasted with the vibrational spectrum of the thinner nitride layer ($N(KLL)/Si(LVV) = 0.020$), which is observed to change dramatically after annealing for 15 minutes [Fig. 3B(a-b)]. The four peak vibrational spectrum of this layer is replaced by a characteristic three peak spectrum with losses at ~ 480 , 700 and 970 cm^{-1} [Fig. 3B(b)] [11,12]. An identical three peak vibrational spectrum is observed by annealing the thicker nitride layer, but only after an additional 60 minute annealing period [Fig. 3A(c)]. Continued annealing of the thin nitride layer produces a layer in which only the 970 cm^{-1} loss can be resolved clearly [Fig. 3B(b)], as was observed previously after long annealing periods of the $N/Si(111)-(7\times 7)$ interface [11]. Also shown in Fig. 3 is the $N(KLL)/Si(LVV)$ Auger ratio at the various stages of annealing. For both the thick and the thin nitride layers studied, the $N(KLL)/Si(LVV)$ ratio is observed to decrease slightly upon conversion to the three mode HREEL spectrum.

The appearance of the three mode HREEL spectra on both the thin and thick nitride layers implies that a similar nitride structure is obtained upon heating, regardless of the original thickness. We believe this thermally stable layer is stoichiometric Si_3N_4 on the $Si(111)-(7\times 7)$ surface. Analogous results were also obtained upon the oxidation of $Al(111)$ surfaces [14,24]. In these studies, a characteristic aluminum oxide phonon spectra was obtained upon either high exposures of oxygen at 300 K or upon low exposures at 300 K followed by annealing. The formation of Al_2O_3 was confirmed with AES. These results were interpreted as signifying that a critical surface and subsurface oxygen concentration is necessary prior to actual oxide formation [14,24]. After low

exposures of oxygen, the annealing was postulated to cause the oxygen to penetrate to subsurface sites and to form small Al_2O_3 clusters. Thus it is likely that upon heating the thin silicon nitride films, penetration and clustering of the nitrogen occurs which results in the formation of clusters of Si_3N_4 . These yield the characteristic three mode vibrational spectrum and the shift in the Si(LVV) transition energy to slightly lower values. This would explain the decrease in the N(KLL)/Si(LVV) ratio as the thin nitride layer is annealed [Figs. 3B(b-c)].

Low energy electron diffraction studies undertaken during our study revealed only a persistent (1x1) pattern after heating silicon nitride layers from 900 K to the silicon nitride desorption temperature, ~1400 K [9]. Annealing at the silicon nitride desorption temperature for 5 minutes produced a sharp, very well-defined (7x7) LEED structure. Only once was a faint (8x8) pattern detected after forming a silicon nitride film, and the quadruplet LEED pattern was never observed. While the absence of the quadruplet pattern is understandable since our carbon contamination was always small [10], our inability to reproducibly obtain the (8x8) LEED pattern is not understood at present.

D) Adsorption Studies on the Silicon Nitride Films.

Figure 4(a) shows a vibrational spectrum of a thick nitride layer (N(KLL)/Si(LVV) = 0.36) warmed to 300 K prior to adsorbing a saturation dose of ammonia. The loss observed at 3400 cm^{-1} is characteristic of an NH_x species ($x = 1$ or 2) and has been studied in detail by vibrational spectroscopy on the clean Si(111)-(7x7) [25-28] and Si(100)-(2x1) [26,28-30] surfaces. Unfortunately, the

strong Si_3N_4 vibrational modes below $\sim 1400 \text{ cm}^{-1}$ prevent the observation of other $\text{NH}_x(\text{a})$ modes needed to identify either $\text{NH}_2(\text{a})$ or $\text{NH}(\text{a})$ species.

Ammonia is known to dissociatively adsorb at silicon dangling bond sites on both $\text{Si}(111)$ and $\text{Si}(100)$ producing $\text{NH}_2(\text{a})$ and $\text{H}(\text{a})$ [25-31]. Since physisorbed molecular ammonia desorbs from a passivated silicon layer at 115 K [25,26,28], these spectra, measured at 300 K, show clearly that a chemisorbed NH_x species is capable of adsorbing onto the surface before and after extensive annealing. This suggests the presence of silicon adsorption sites on the top surface of the silicon nitride films at various stages of growth. This agrees with the results of Peden et al. [13] as discussed in the Introduction. However, we can not exclude the possibility that Si_3N_4 formation causes clean $\text{Si}(111)$ sites on the substrate to appear, and that NH_x adsorption occurs on these sites also.

The presence of a Si overlayer on films of SiC grown on $\text{Si}(100)$ is well established [32,33], and a similar Si transport process from the substrate Si to the SiC/vacuum interface occurs in SiC film growth as in Si_3N_4 growth.

Summary

The findings of this study are as follows:

- 1) Ammonia adsorption onto $\text{Si}(111)-(7 \times 7)$ followed by heating produces silicon nitride films which are vibrationally identical to nitride films produced using N atoms.
- 2) Submonolayer and multilayer silicon nitride films, upon heating to 1200 K, yield identical vibrational spectra, suggesting that both form stable Si_3N_4

structures upon heating. The decrease in the N(KLL)/Si(LVV) ratio upon annealing the thin nitride layers suggests that nitrogen penetration and clustering is occurring to form patches of Si_3N_4 .

3) Thick silicon nitride films (>1 monolayer) are able to chemisorb ammonia due to the presence of unsaturated silicon sites at the surface. These sites are probably present in an elemental silicon overlayer on the Si_3N_4 film.

Acknowledgments

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Figure Captions

Figure 1. HREEL spectra showing the development of the vibrational structure of silicon nitride thin films on a Si(111)-(7x7) surface. The films in (a)-(e) were prepared by ammonia doses of 1×10^{15} NH_3/cm^2 onto Si(111)-(7x7) at 90 K followed by heating to 900 K prior to cooling to 90 K for any subsequent doses. Number of ammonia doses: a) 1; b) 2; c) 3; d) 4; e) 8. Spectra f) and g) were prepared by dosing Si(111)-(7x7) at 900 K with 1.2×10^{17} NH_3/cm^2 and 4.3×10^{17} NH_3/cm^2 , respectively. All films were heated to 1200 K in a final annealing step.

Figure 2. Auger spectra showing the changes observed upon the nitridation of Si(111)-(7x7). The principal changes in the spectra is the development of an 84 eV Si(LVV) Auger feature attributed to silicon nitride.

Figure 3. A comparison of the thickness dependent thermal conversion of a nitride layer to a more stable three peak vibrational structure characteristic of Si_3N_4 , and the N(KLL)/Si(LVV) ratio measured for each layer.

Figure 4. Vibrational spectra showing ammonia adsorption to a thick silicon nitride layer (a) before extensive annealing, and (b) after extensive annealing. Ammonia is observed to chemisorb on both surfaces at 300 K.

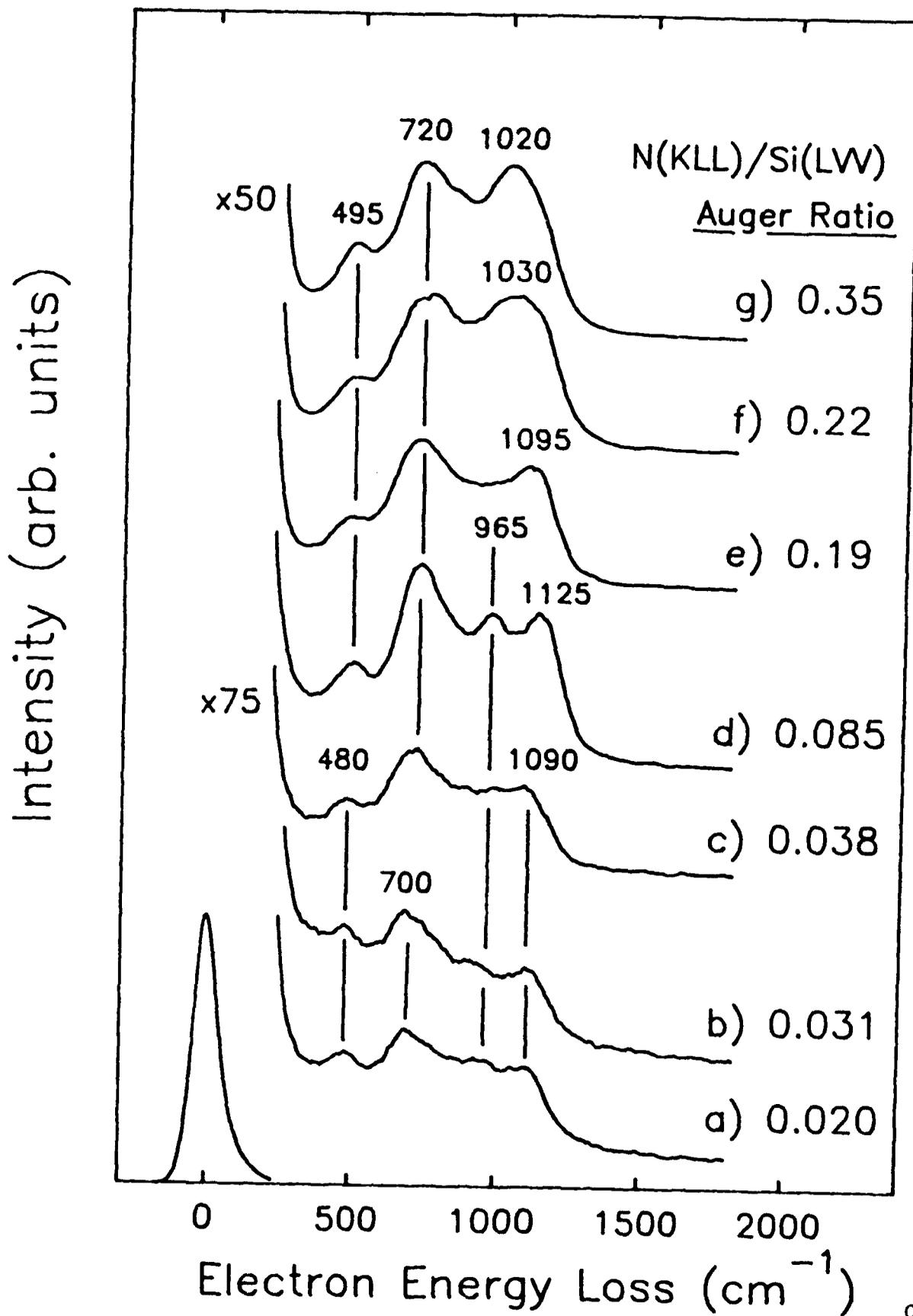
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16. It was found that a procedure in which a silicon crystal held at 900 K and positioned only 3 mm from the glass microcapillary array in the doser resulted

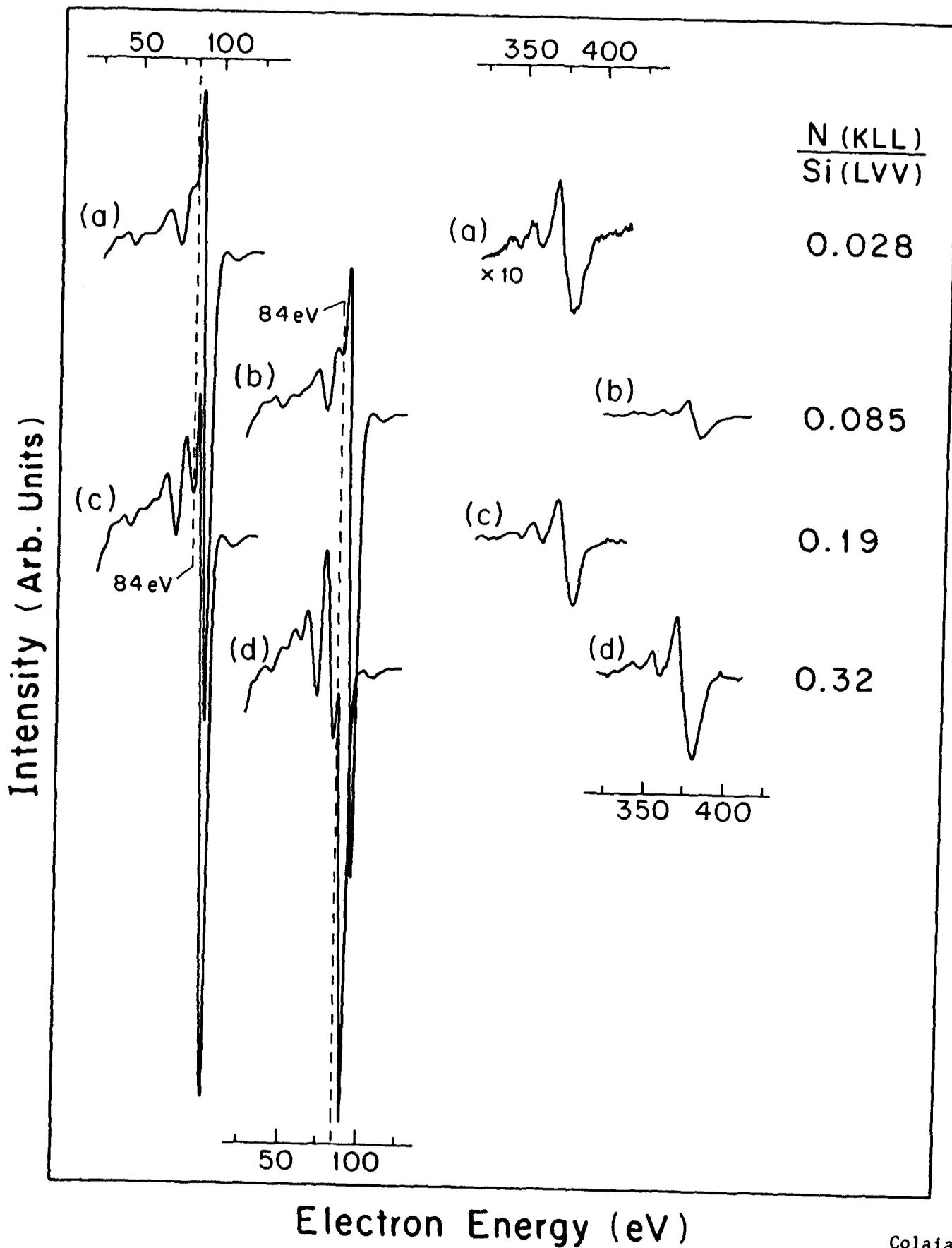
in a carbon contaminated silicon nitride layer. This problem was overcome by removing the glass microcapillary array from the doser head.

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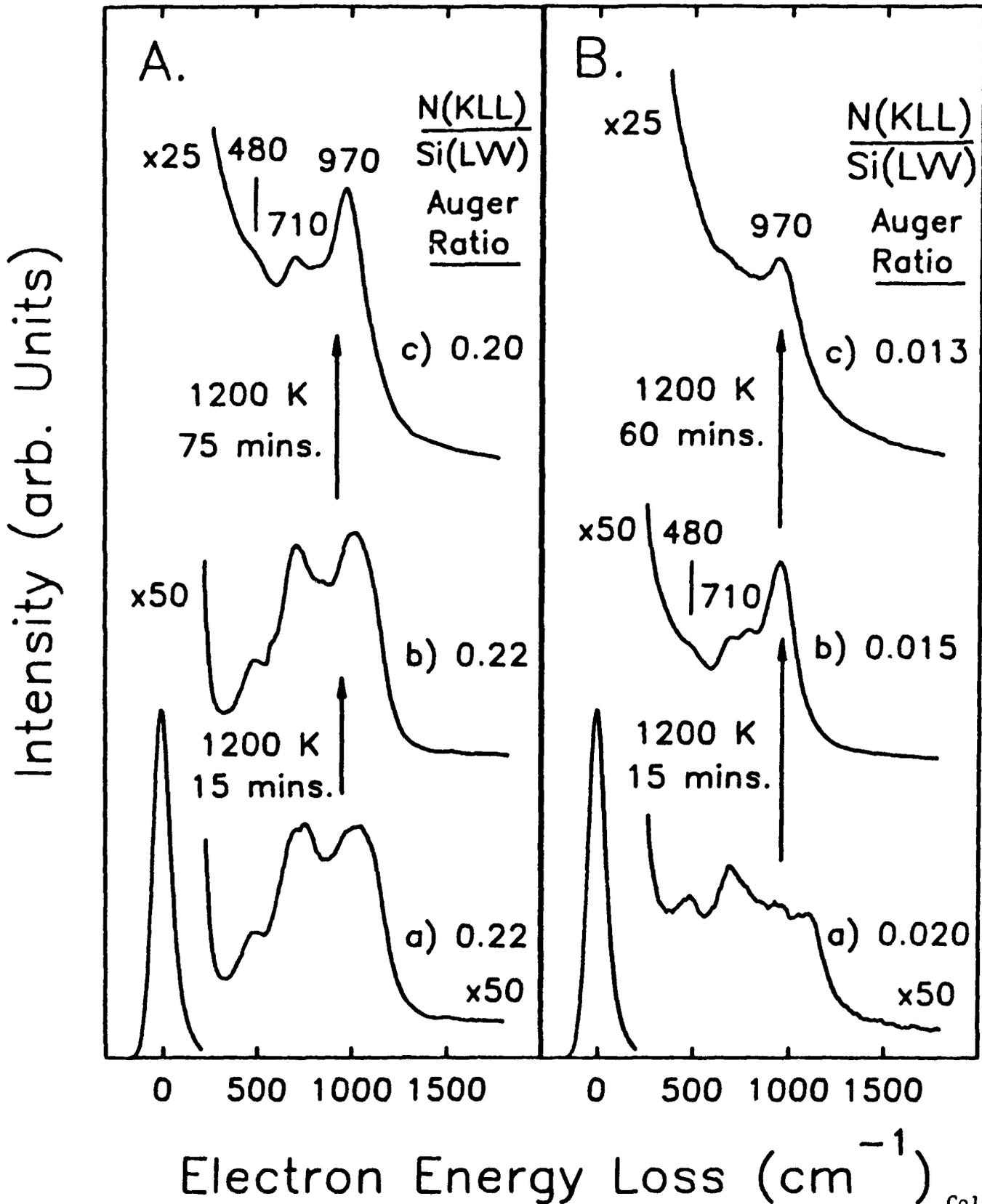
HREEL Spectra Showing the Development of Silicon Nitride Films



Auger Spectra Showing the Development of Silicon Nitride on a Si(111) Substrate



Comparing the Effects of Heating Thin and Thick Silicon Nitride Films



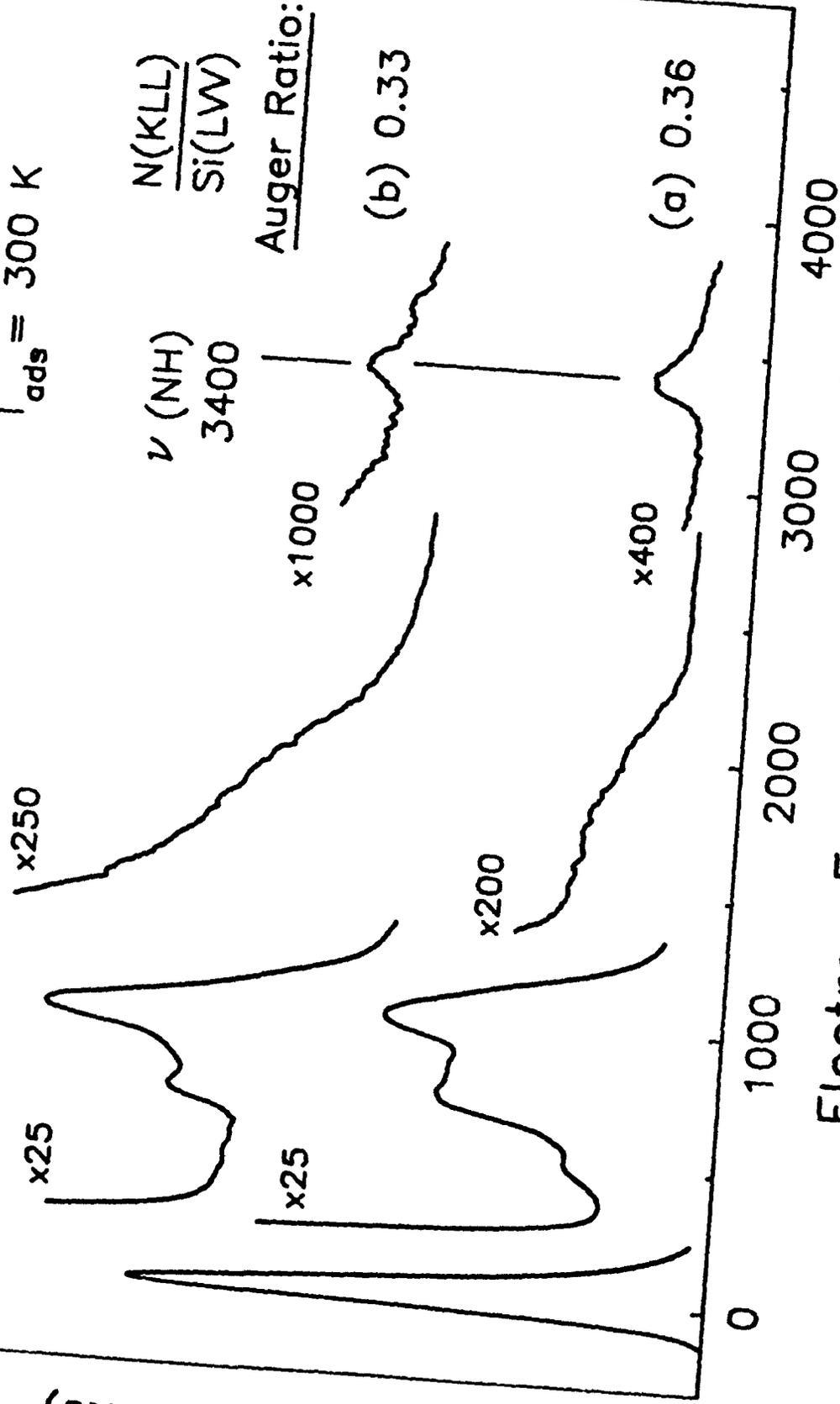
Adsorption of Ammonia on Silicon Nitride Films



$\text{NH}_3 = 1 \times 10^{15} / \text{cm}^2$

$T_{\text{ads}} = 300 \text{ K}$

Intensity (arb. units)



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