

AN EXPERIMENTAL INVESTIGATION OF THERMALLY
STIMULATED LUMINESCENCE IN ELECTRON-
IRRADIATED BERLINITE (AlPO_4)

By

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CHAPTER I

INTRODUCTION

Berlinite (AlPO_4) is a piezoelectric material that occurs rarely in nature. It has the same crystal structure as quartz, SiO_2 , except that in berlinite, half the Si^{4+} ions are replaced with Al^{3+} and the other half with P^{5+} ions, resulting in a doubling of the unit cell along the c axis [1].

Research on point defects began after the first hydrothermal synthesis of large single crystals by J. M. Stanley in 1953 [2]. Since then efforts to produce large single crystals of sufficient quality to use in commercial applications have proved relatively unsuccessful [3],[4].

Most of the experimental measurements on berlinite which have applicability to the work reported here have been done in areas such as ESR, ENDOR, optical absorption and luminescence. However, there is no evidence of any thermoluminescence (TSL) study on the material.

Emission and excitation spectra for Fe^{3+} in AlPO_4 single crystals with excitation in the near UV are reported and discussed by B. Schmitz and G. Lehmann [5]. Emission bands originating from higher excited states were observed in addition to weak bands between 13300 and 14800 cm^{-1} .

The x-band ESR spectrum of vanadium-doped α -berlinite has been observed at room temperature by R. S. de Biasi [6]. The spectrum

attributed to V^{4+} in a distorted tetrahedral site can be described by a spin Hamiltonian of axial symmetry. Substitutional Cr^{3+} ions were investigated by J. C. M. Henning [7], also using ESR techniques.

A recent study made by L. E. Halliburton, L. A. Kappers, A. F. Armington, and J. Larkin [8] actually triggered the current TSL research on berlinite reported here. They observed the point defects present in $AlPO_4$ after electron irradiation at 77 K and 300 K using Electron Spin Resonance (ESR), Electron-Nuclear Double Resonance (ENDOR) and optical absorption. The presence of a significant concentration of Fe^{3+} ions was verified by the ESR measurements. When the berlinite samples were irradiated at 300 K with 1.7 MeV electrons, six distinct hole-like centers were created. Thermal anneal studies revealed that four of these centers decay near 425 K and the other two decay at about 550 K. When the samples were irradiated at 77 K, two additional defects were observed which were stable only below room temperature. One of these latter centers was identified as atomic hydrogen.

The crystal structure determination using x-ray reflection data was made by Schwarzenbach [9] in 1966. This work was later refined by Thong and Schwarzenbach [10] to include electron density maps and calculation of the gradient of the electric field at the aluminum site. The raman spectra and lattice dynamics of berlinite have been investigated by Scott and coworkers [11,12,13].

Because berlinite has greater piezoelectric coupling coefficients than quartz, it could prove to be an excellent candidate for a surface acoustic wave (SAW) substrate material [14,15,16,17]. However, as mentioned previously, difficulties encountered in synthesizing large, high-quality crystals have seriously hampered efforts at commercial

application.

The purpose of this study has been to investigate the nature of radiation-induced defects in berlinite by using TSL results and attempting to correlate them with the previous ESR measurements of Halliburton et al [8].

CHAPTER II

THEORETICAL DISCUSSION

A. TSL Theory

Thermally stimulated luminescence (TSL) is one of the tools used for trap level spectroscopy. It is often observed that the emission of light by an irradiated specimen can occur at a temperature well below that of incandescence if the specimen is slowly heated. At certain temperatures during the increase, "glow peaks" of light output occur which, if carefully measured and properly interpreted, can provide much useful information about the defect-structure of the material under study. However all the desired information about the microstructure of the crystal imperfections cannot be obtained by studying glow curves alone. This is because all theoretical explanations of glow curves known so far are based on statistical models which describe the re-distribution of electrons within the impurity levels as a result of the gradual warming up of the sample. For this type of calculation a knowledge of trapping and recombination parameters is needed. In fact, no good theory of TSL was developed until late 1930 following the early development of band structure theory.

To find appropriate trapping parameters, other techniques such as space-charge limited current, temperature dependence of photoconductivity, current noise, etc. could be used in principle, but the discussion of these techniques is beyond the scope of this paper.

To illustrate the basic ideas of TSL theory, two simple models will be discussed. The first one is a first order kinetics theory, in which the assumption is made that at a particular temperature there is a certain probability for a recombination of holes and electrons and that the result of this recombination is an observable glow peak. In a second order kinetics theory, however, the assumption is made that the probabilities of recombination or retrapping for the mobile electrons or holes are equal.

To aid in understanding TSL theory it is necessary to mention some ideas from band theory. A pure insulating crystal at zero temperature Kelvin has a filled valence band with no electron in the conduction band. An impurity defect in the crystal introduces a localized energy level between the valence band and the conduction band (Fig. 1) which can — depending on its position in the forbidden band — become a trap level for electrons or for holes following excitation with ionizing radiation at low temperature. When the temperature is slowly elevated so that the electron or hole has enough energy to escape, a recombination might occur resulting in a glow peak. The temperature at which the process happens gives information about the energy of the trap and the spectral output of the glow peak gives information about the nature of the recombination.

The following will serve to illustrate a very simple band structure model and sequence of thermally stimulated luminescence events. (Figure 1). The model implies electron trapping but similar reasoning could be applied in the alternative case of hole trapping and recombination.

1. Excitation of an electron from the valence band due to irradiation at liquid nitrogen temperature.

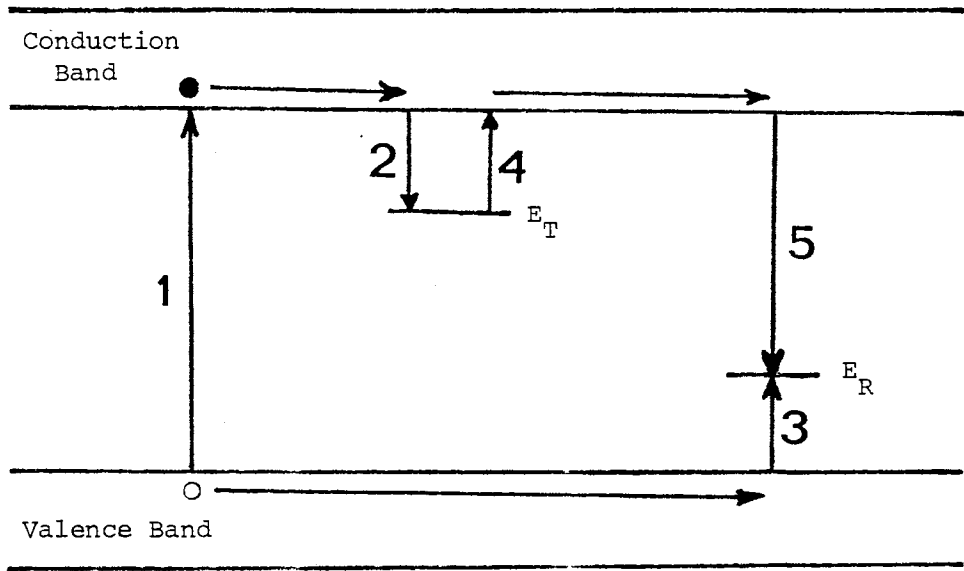


Figure 1. Simple TSL Model

2. Trapping of a conduction band electron.
3. Trapping of a free hole in the recombination center.
4. Thermal excitation of the trapped electron back into the conduction band due to slow elevation of sample temperature.
5. Capture of a conduction band electron at a recombination center accompanied by TSL emission.

B. First-Order Kinetics Model

The first order kinetics model was described by Randall and Wilkins [18] in 1945. The assumption was made that the probability of retrapping is very small in comparison with the probability of recombination and as a result the retrapping factor $R = \frac{\beta}{\gamma}$ is assumed to be zero.

Here:

β = coefficient for the retrapping transition

γ = coefficient for the recombination transition

R = retrapping factor.

Figure 2 indicates the meaning of the model parameters used for both first and second-order theories. Solution of the appropriate equations under the limitation of $R \rightarrow 0$ leads to an expression for the intensity of the glow peak as

$$I = \alpha_0 h_0 \exp\left[\frac{-E}{kT} - \frac{\alpha_0}{q} \int_{T_0}^T \exp\left(\frac{-E}{kT'}\right) dT'\right]$$

where a representative plot of this result is shown in Figure 3 for a chosen set of parameter values.

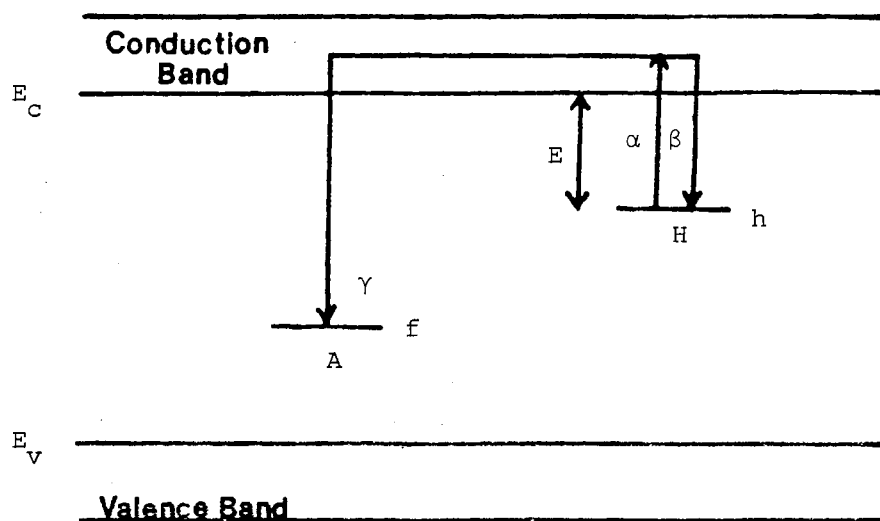


Figure 2. Simple band structure for first order and second order kinetics

E_C = Conduction band energy level

E_V = Valence band energy level

E = Electron trap energy level

α = The probability of excitation into the conduction band

β = The probability of retrapping the electron

γ = The probability of recombination at a recombination center

H = Concentration of trap levels

A = Concentration of recombination centers

f = Concentration of empty recombination centers

h = Concentration of trapped electrons

q = Constant heating rate

α_0 = The value of α at $T = T_0$

h_0 = The value of h at $T = T_0$

k = Boltzman constant.

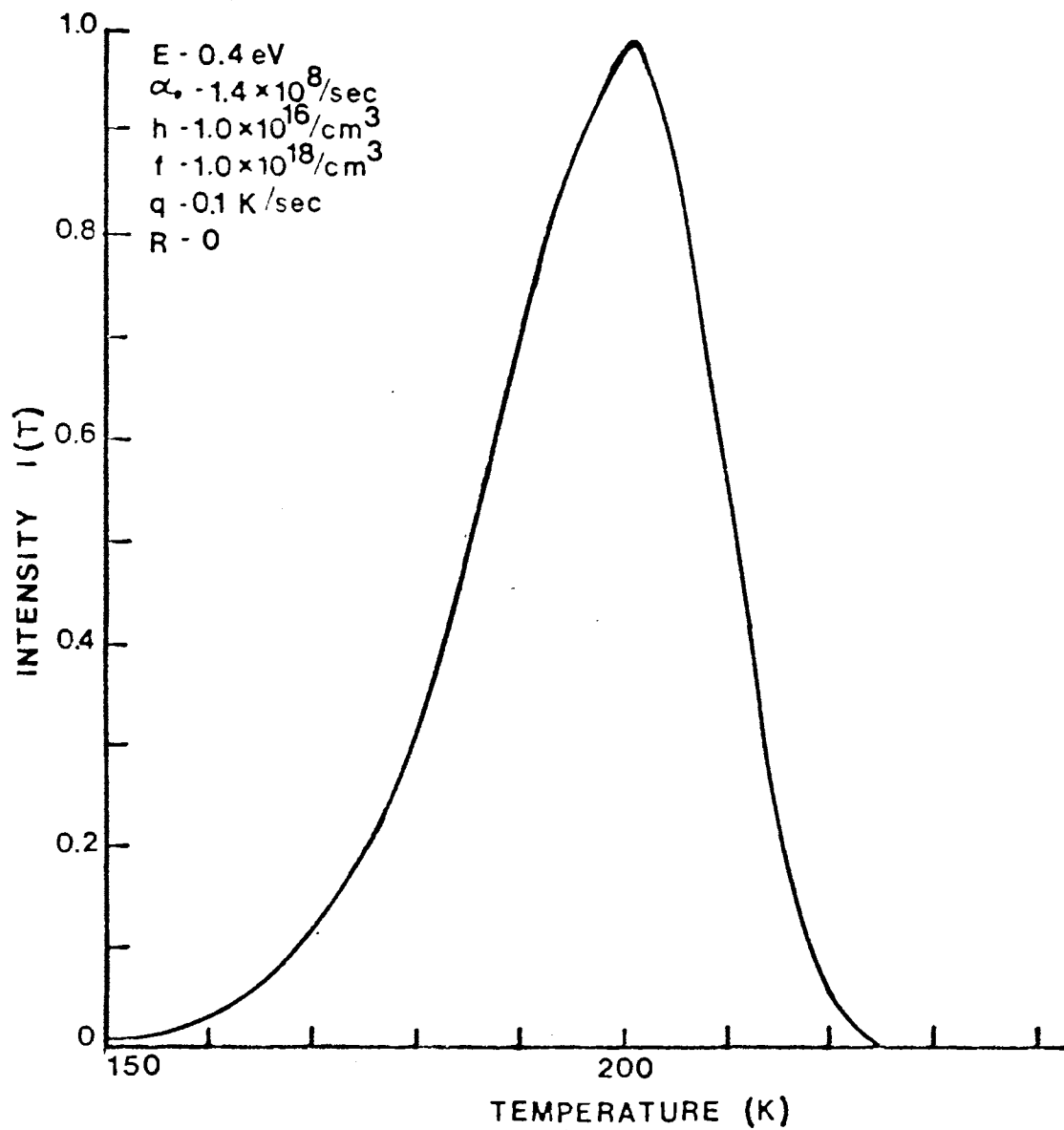


Figure 3. Plot of Intensity Versus Temperature for 1st Order Kinetics

C. Second-Order Kinetics Model

A second order kinetics model has been described by Garlik and Gibson [19]. Here, the assumption was made that the trapping parameters defined previously is equal to one. This leads to an expression for the glow peak intensity as

$$I(T) = \frac{\alpha_o h_o^2 \exp\left(\frac{-E}{kT}\right)}{\left[H + h_o \frac{\alpha_o}{q} \int_{T_o}^T e^{-E/kT'} dT'\right]^2}$$

A representative plot of this intensity vs. temperature is shown in Figure 4. It can be seen that the shape of the curve is quite similar to that predicted by first order kinetics. Figures 3 and 4 are taken from a report by Braunlich [20].

D. Determination of Trap Depth

There are many methods for determination of trap depth E. The initial rise method advocated by Garlik and Gibson [3] is based on the assumption that the traps begin to empty as the temperature is raised and that the integrals in the derivations are essentially independent of temperature. This leads to the expression

$$\ln I = -E/kT + \text{const}$$

It should be noted that if the glow peak is disturbed by overlapping

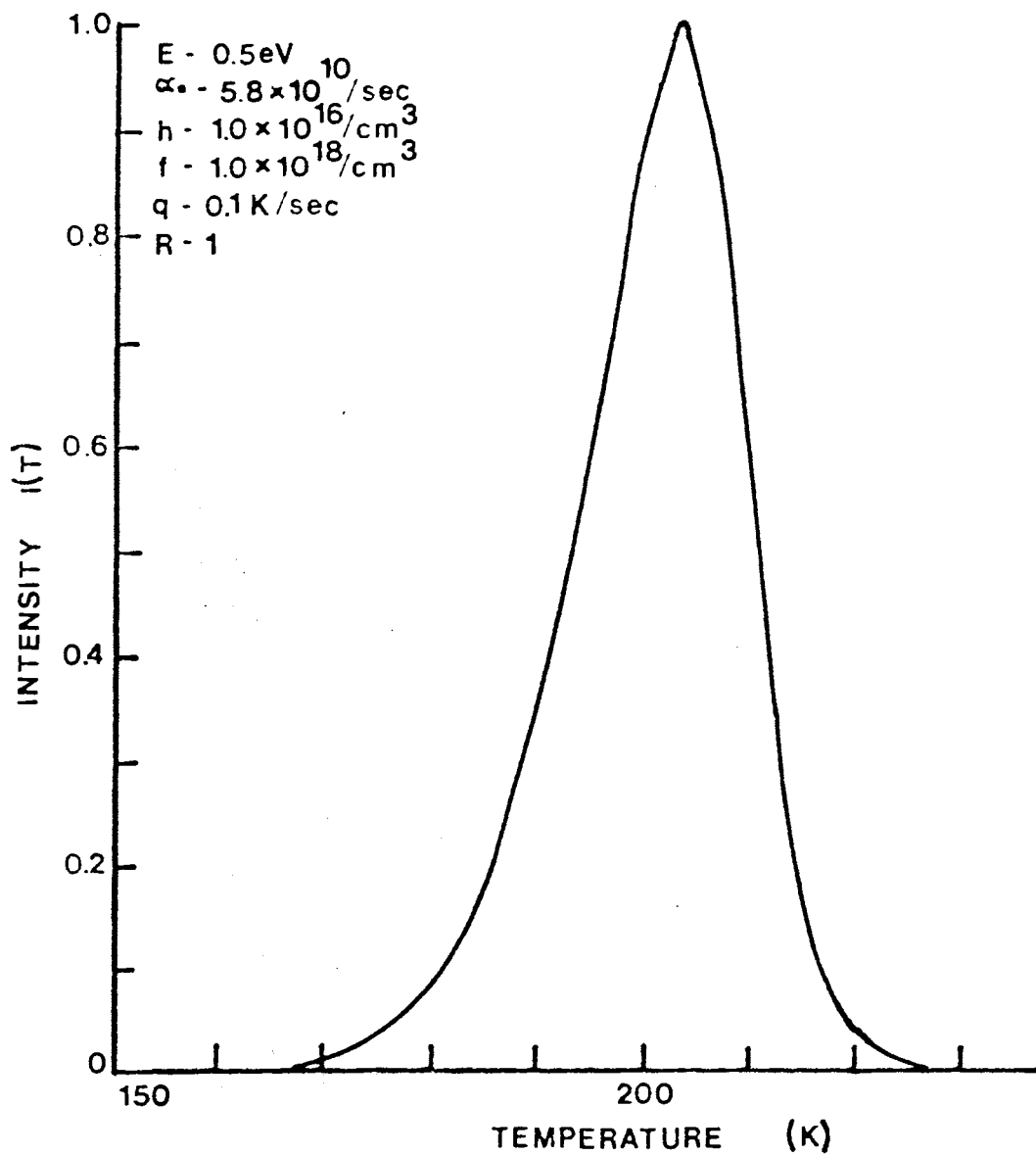


Figure 4. Plot of Intensity Versus Temperature for the 2nd Order Kinetics

neighboring peaks, a cleaning technique by Hoogenstraaten [21] can be applied. Additional methods for calculating trap depths and escape-frequency factors appear in an article by Chen [22]. He also discusses ways of distinguishing between first and second-order kinetics glow peaks.

CHAPTER III

EXPERIMENTAL APPARATUS AND PROCEDURES

A. Apparatus

The instrumentation used for the TSL measurements was quite simple. An EMI 9789QB photomultiplier tube was used to detect any luminescence from the sample. It was powered by a Keithley Instrument Model 244 power supply operated at 1000 volts. The photomultiplier tube has a bi-alkaline response which is most sensitive around 450 nm. Since the sample was to also be studied at high temperatures it was necessary to use an infrared filter to filter out any infrared radiation of the sample and cold finger due to this high temperature. The output of the photomultiplier was connected to a Keithley Model 602 electrometer and the TL output of the electrometer connected to the y-axis of a Houston Instruments model 2000 x-y recorder with type 7 modulus. To reduce noise, a low-pass active filter was used between the electrometer and the x-y recorder. The temperature of the sample was measured by a copper-constant thermocouple connected to the cold finger of the cryostat and monitored on the x-axis of the x-y recorder. (Figure 5) To avoid errors in temperature measurement due to a change in ambient temperature, an Omega miniature cold junction was used which provided the electric equivalent of an ice bath reference at 0°C.

The cryostat itself was custom built in the physics shop at

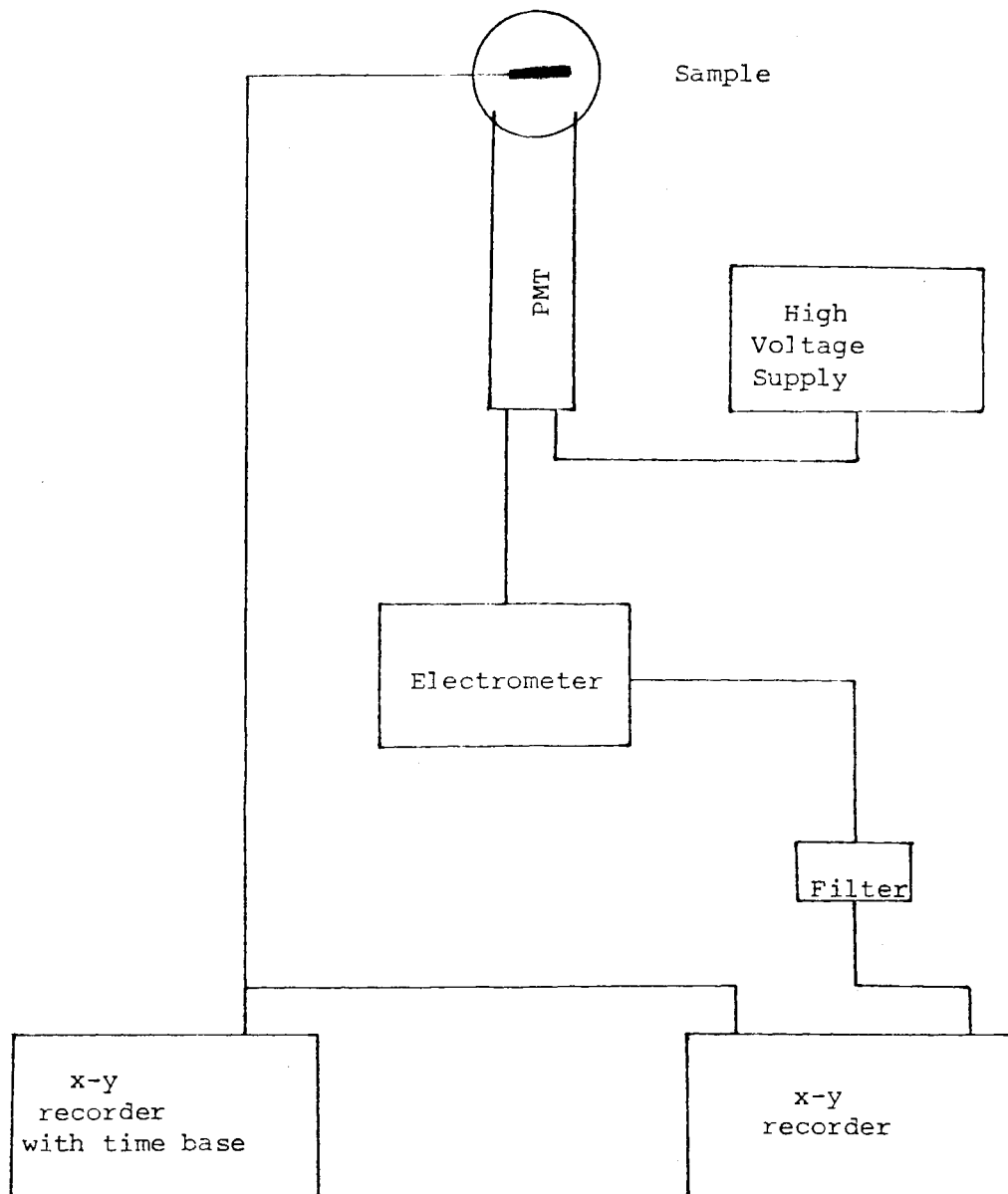


Figure 5. Diagram of TSL Apparatus

Oklahoma State University (Figure 6). The sample was held to the copper cold finger of the cryostat by a copper plate fastened with brass screws to keep the sample secure in position. The sample itself was placed on a piece of soft aluminum foil to make better thermal contact with the copper surface of the cold finger. The base of the cryostat has three quartz windows and one aluminum window which could be rotated so that electron irradiation could be done through the aluminum window and light emission subsequently, observed through a quartz window. The irradiation source used was a High Voltage Engineering Corporation Model AN2000 Van de Graaf accelerator capable of an acceleration potential up to 2 megavolts. For UV light irradiation a 150 watts xenon lamp was used.

B. Sample Preparation

The samples were obtained from Dr. Larry Halliburton who originally obtained them from Airtron. The samples were cut in rectangles of about 5 x 3 mm with a thickness of about 1 mm. They were washed with soap and rinsed with deionized water and then cleaned in an ultrasonic cleaner followed by a final rinse with acetone.

Once cleaned, the sample was mounted on the cold finger of the cryostat and pressure reduced to about 5×10^{-6} torr. The cryostat chamber was filled with liquid nitrogen but because of the thermal resistance of the stainless steel connection to the cold finger, the sample would only cool down to a temperature of about 96 K after a period of approximately 30 minutes.

C. Experimental Procedure

There were two series of measurements done on berlinite samples.

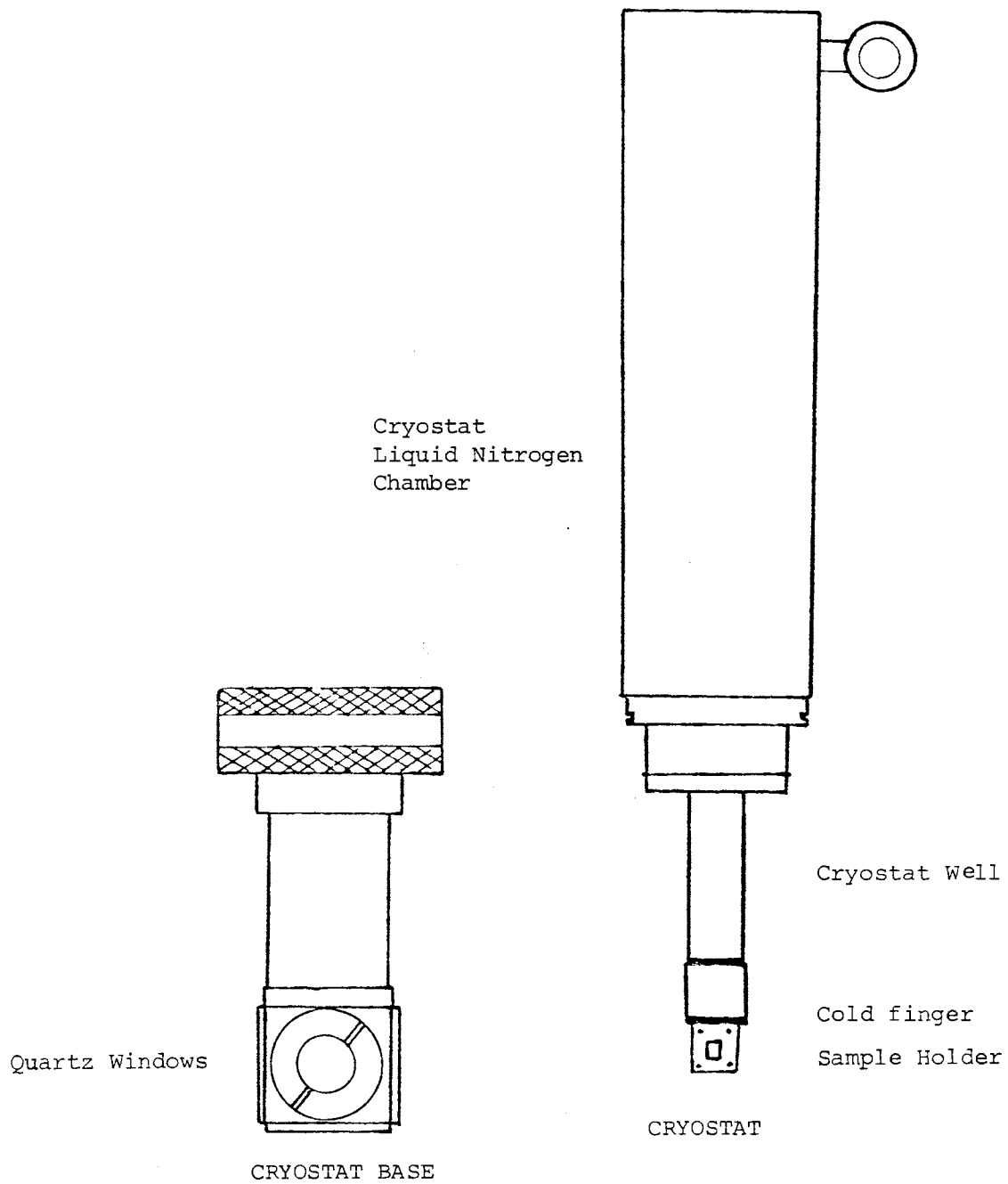


Figure 6. Diagram of TSL Cryostat

The first was to irradiate a virgin sample at liquid nitrogen temperature and to warm it slowly while looking at its luminescence output. Another series of experiments was to electron irradiate the sample at room temperature and then to cool it to liquid nitrogen temperature and irradiate it again. After the second irradiation, it was slowly warmed and the glow peaks recorded. The double irradiation procedure was used to look for results comparable to those observed in earlier work on quartz [23] where irradiation above 200K mobilized alkali ions to move away from aluminum defect centers causing significant changes in TSL results.

Several series of measurements using UV light — either as a primary irradiation source or as a bleaching source following a electron irradiation — were also carried out.

D. Sample Irradiation

The cryostat was placed a distance of about 12 cm from the Van de Graaf window to be electron irradiated through its own aluminum window. The irradiation was done with the Van de Graaf set at 1.7 MV accelerating potential and adjusted for a return current of about 10 μA which resulted in a beam of approximately $0.2 \mu\text{A}/\text{cm}^2$ on the sample. In order to avoid raising the temperature of the sample, the irradiation was done in four one-minute doses separated by two-minute intervals.

UV irradiations were done for 10 minutes using a source distance of 1.5 meters.

E. Observation of Luminescence and Heating Method

The cryostat's base was rotated to observe the glow peaks through the fused-quartz windows. To warm the sample from liquid nitrogen temperature, a rubber air hose was lowered into the crystal's liquid nitrogen chamber and the extra liquid nitrogen was evaporated by blowing air into the chamber. The rate of heating was closely watched by means of a Houston Instrument model 2000 x-y recorder with a type 5 and a type 7 module. The type 5 module was used in order to utilize the x-axis as a time base and the temperature was monitored on the y-axis. A calculated plot of temperature vs. time was drawn with a slope of 10 K/min and the rate of air flow was manually adjusted so that the pen would follow the pre-plotted line. Using this method the heating rate was kept fairly accurately at 10 K/min except at temperatures close to room temperature.

To raise the temperature of the sample above the room temperature a heating cartridge was lowered into the cryostat's well and the heating rate was adjusted using a Variac to periodically increase the potential difference across the heater. The heating rate in this range was approximately 15 K/min.

CHAPTER IV

EXPERIMENTAL RESULTS

When a virgin sample of berlinite was irradiated at liquid nitrogen temperature with 1.7 MeV electrons there were five glow peaks observed below room temperature with the last peak a broad one which upon subsequent runs split into two separate peaks. (Fig. 7). The measurements were repeated with the same sample cooled down to liquid nitrogen temperature and again irradiated with the same dose of electrons. The glow peaks were observed at the same temperature as before (Fig. 8) but the relative amplitudes of the last four peaks were changed. The TSL results in Figure 7 and Figure 8 are characteristic of these observations. It should be noted that for the experiments described thus far, the sample was never warmed above room temperature nor was it irradiated at room temperature.

In the next set of measurements the sample was electron irradiated at ~ 300 K (room temperature) for 5 minutes and cooled to liquid nitrogen temperature and then given a second dose of electron irradiation (4 one-minute doses with 2 minute intervals in between). The TSL glow peaks observed below room temperature (Figure 9) were basically at the same temperatures as previously seen. However, it was noted that the relative amplitudes of the last four peaks remained fairly stable from one run to another although overall amplitudes were reduced. The ultraviolet bleaching results also shown in Figure 9

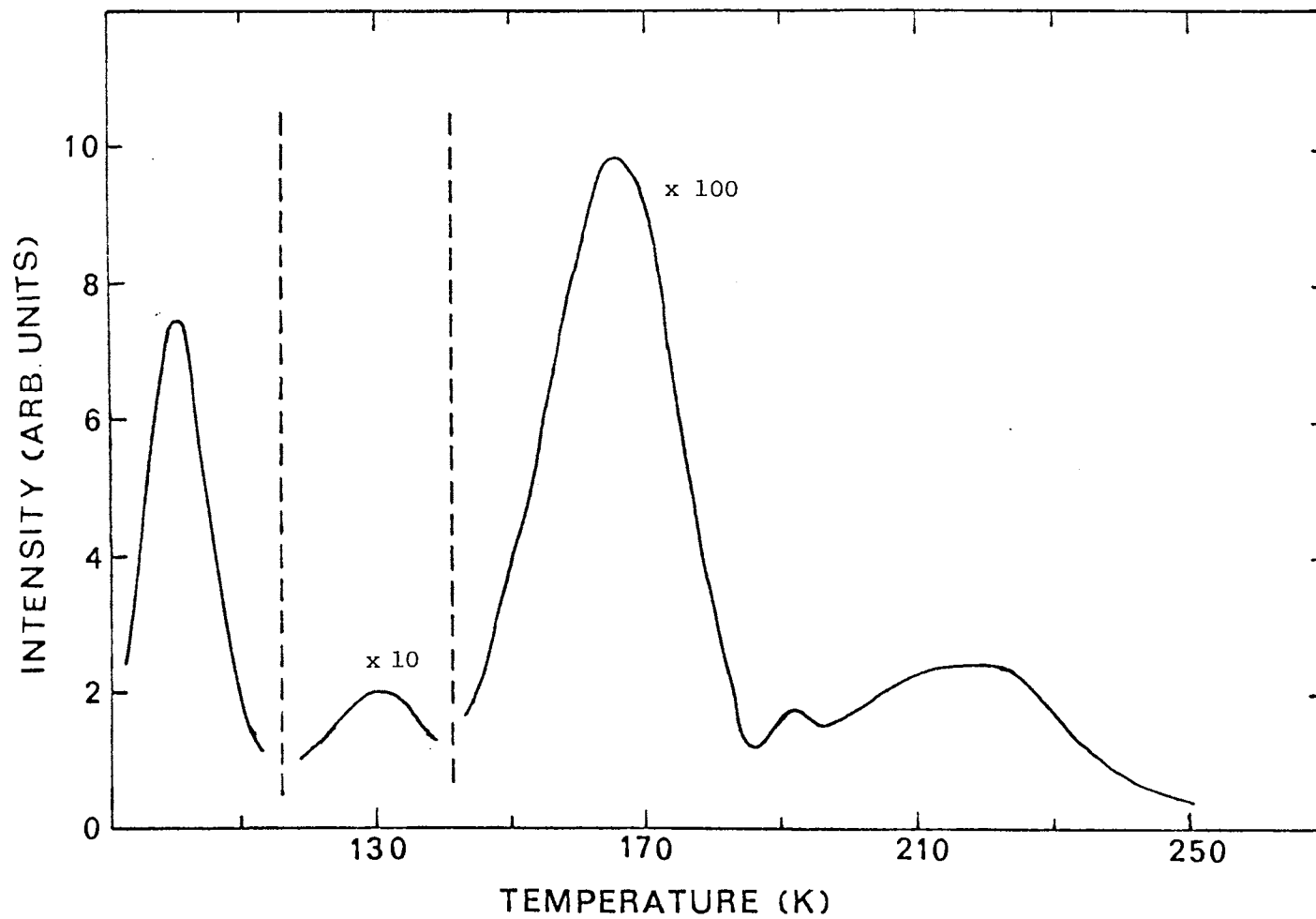


Figure 7. Low temperature TSL of AlPO_4 Electron Irradiated at 96 K (1st measurement)

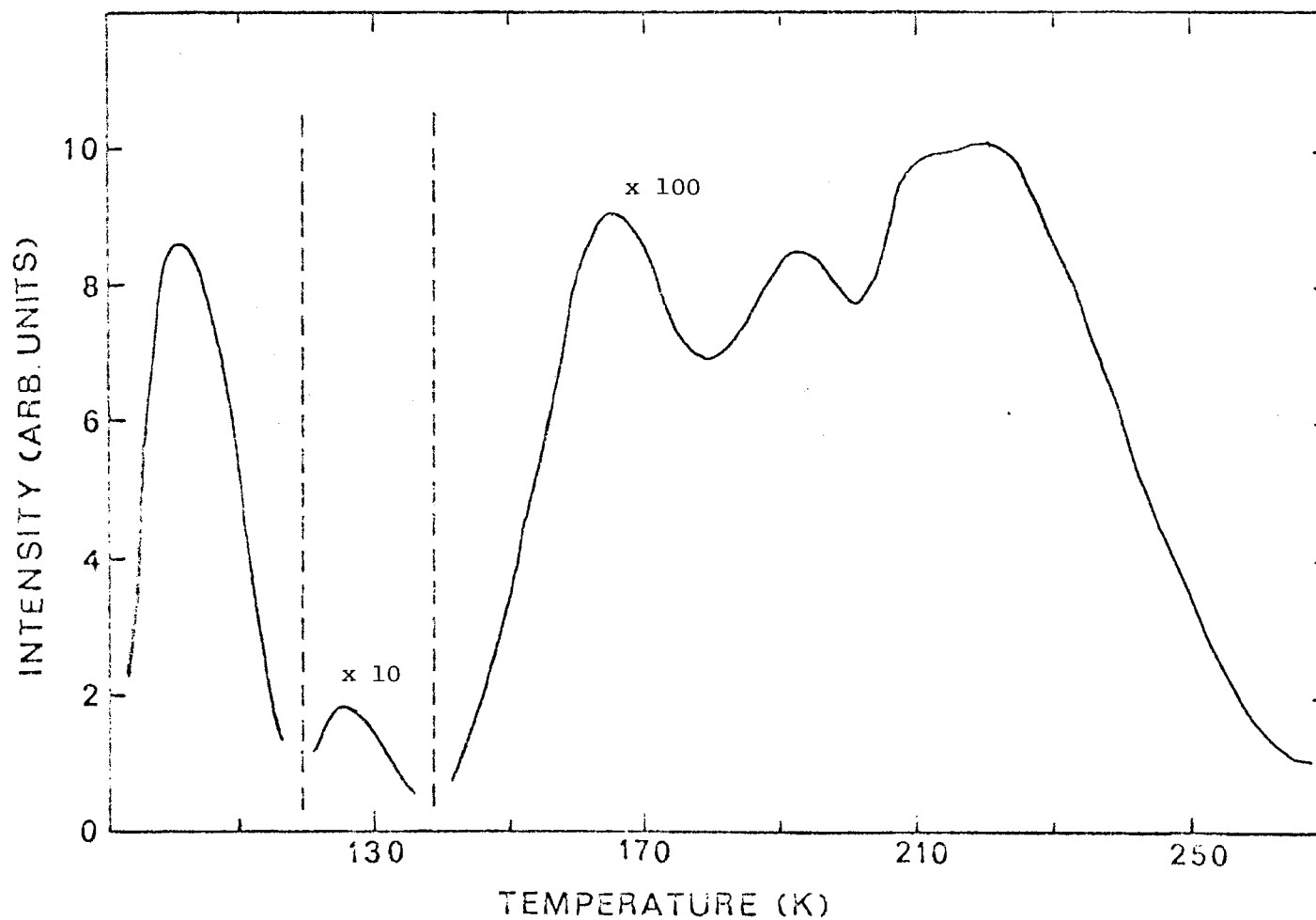


Figure 8. Low temperature TSL of AlPO_4 Electron Irradiated at 96 K
(A later measurement)

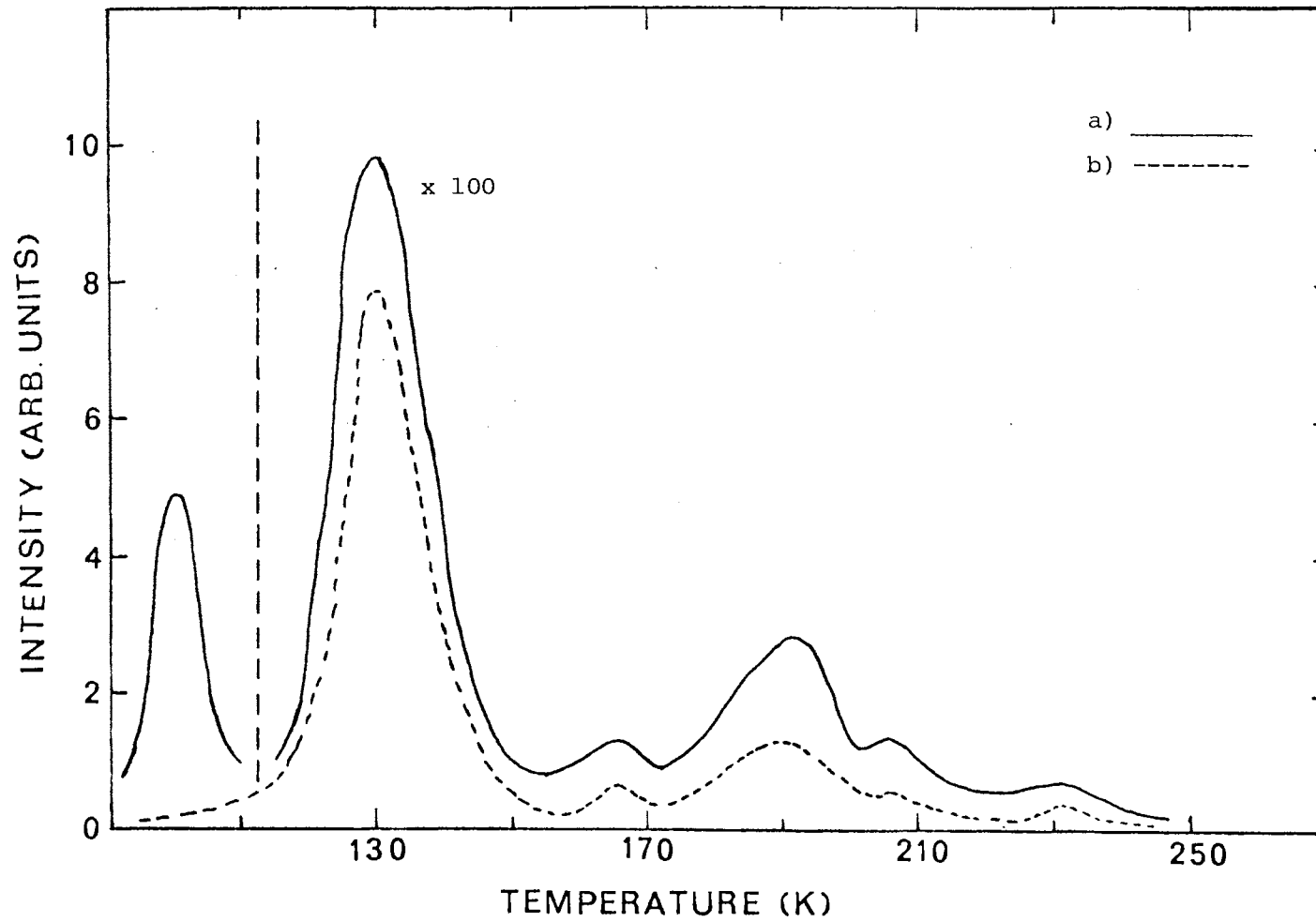


Figure 9. Low temperature TSL of AlPO_4 : a) Electron Irradiated at 300 K and 96 K. b) Same double irradiation followed by uv bleach at 96 K

(dashed line) will be described later. Figure 10 shows a continuation of Figure 9 as the heating and glow peak observations were extended above room temperature to 520 K. Two additional peaks of roughly comparable amplitude appeared in this temperature range the first time the experiment was done.

Next, following the previous work in which the specimen had been heated to 520 K, it was again electron-irradiated at liquid nitrogen temperature with no room temperature irradiation (Figure 11). The observed glow peaks were identical in position to those observed in previous results (Figure 9), except for the lowest temperature peak, the amplitudes were slightly higher. This figure also includes an ultraviolet irradiation curve to be discussed later.

Figure 12 is a continuation of Figure 11 showing results above room temperature. Once again the peak temperatures agree with those in Figure 9 but it was noted that the amplitude of the first peak was now about three times that of the second. This figure, too, contains ultraviolet bleaching data for later discussion and comparison.

When a second virgin sample was subjected to 4 minutes of electron irradiation at liquid nitrogen temperature it exhibited four distinguishable peaks with only a hint of two more over lapping peaks in the liquid nitrogen to room temperature range (Figure 13). When the same sample was room temperature irradiated and then electron irradiated again at liquid nitrogen, however, it exhibited six distinguishable peaks matching the previous results in temperature positions as shown in Figure 14. The comparable data for the first sample are given in Figures 7 and 9.

Again, Figure 15 is a continuation of the Figure 14 data in the

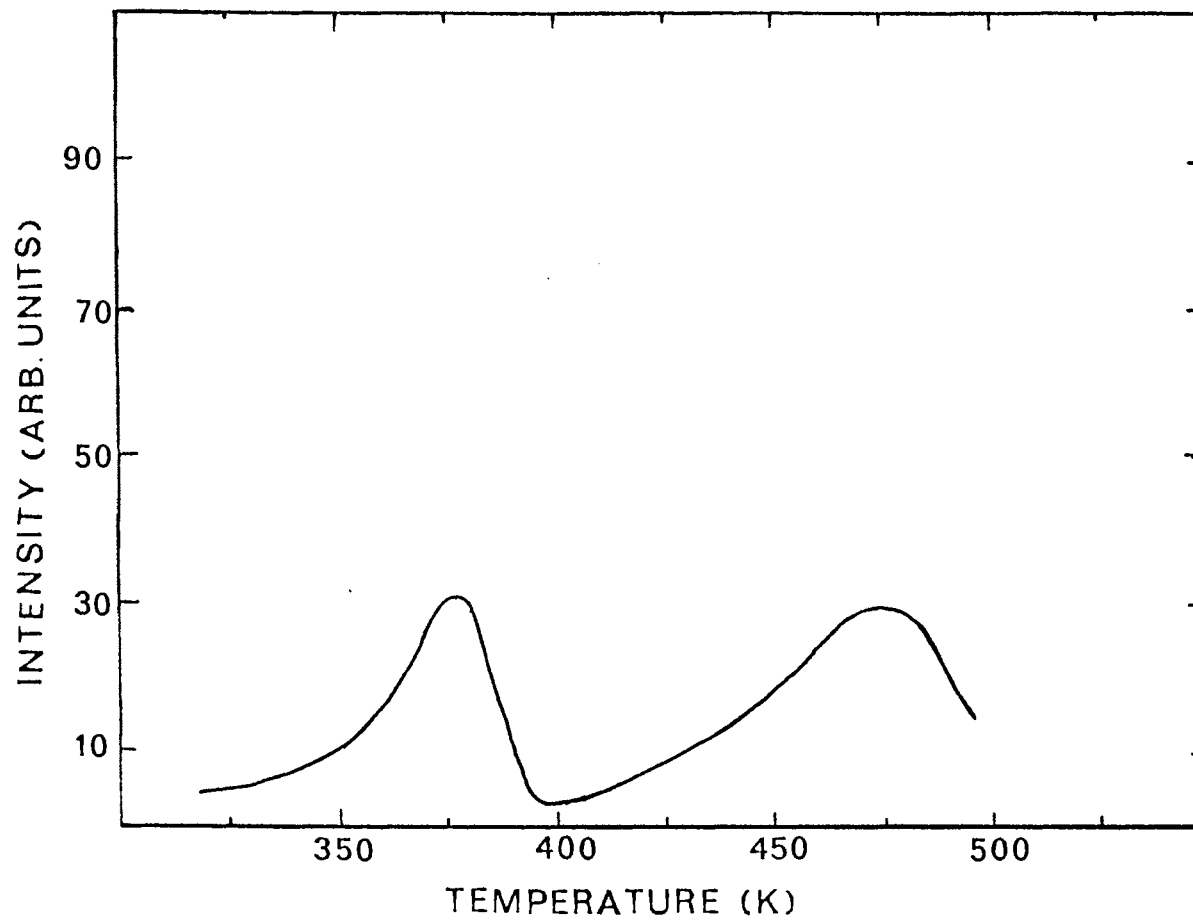


Figure 10. High temperature TSL of AlPO_4

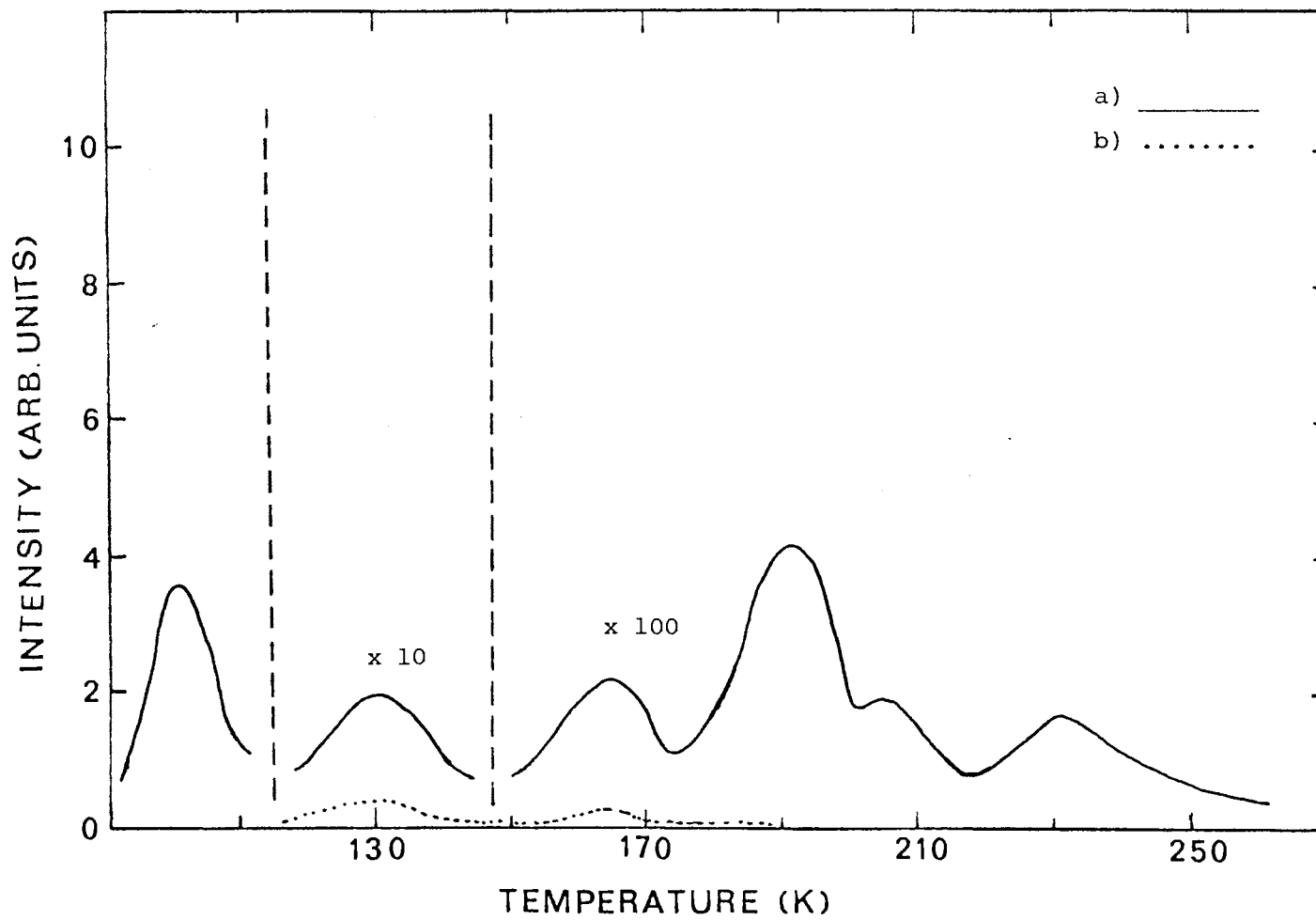


Figure 11. Later low temperature TSL of AlPO₄: a) Electron Irradiated at 96 K
 b) UV Irradiated at 96 K

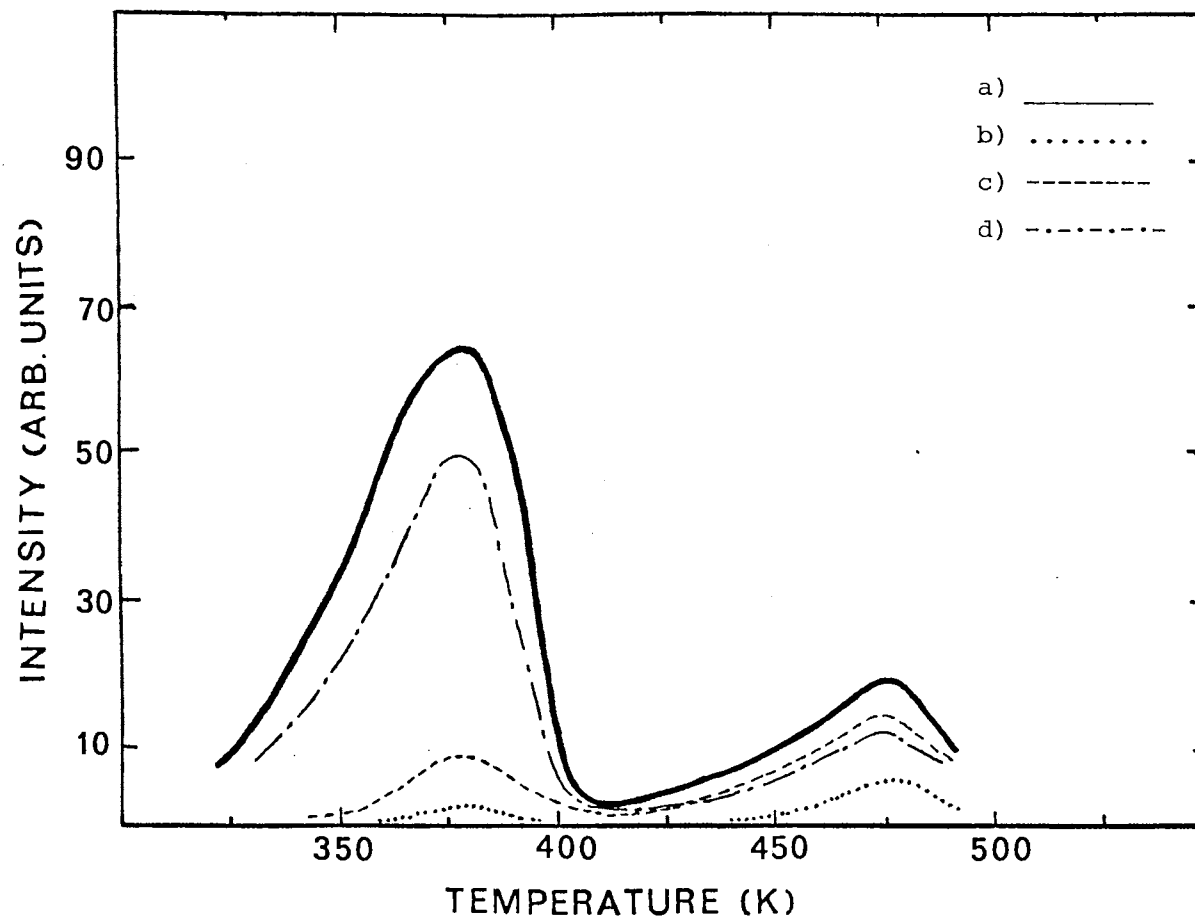


Figure 12. Higher Temperature TSL of AlPO_4 : a) Electron Irradiated only at 96 K. b) UV Irradiated only at 96 K. c) UV Bleach at 96 K after 96 K Electron Irradiation. d) UV Bleach at 300 K after 96 K Electron Irradiation

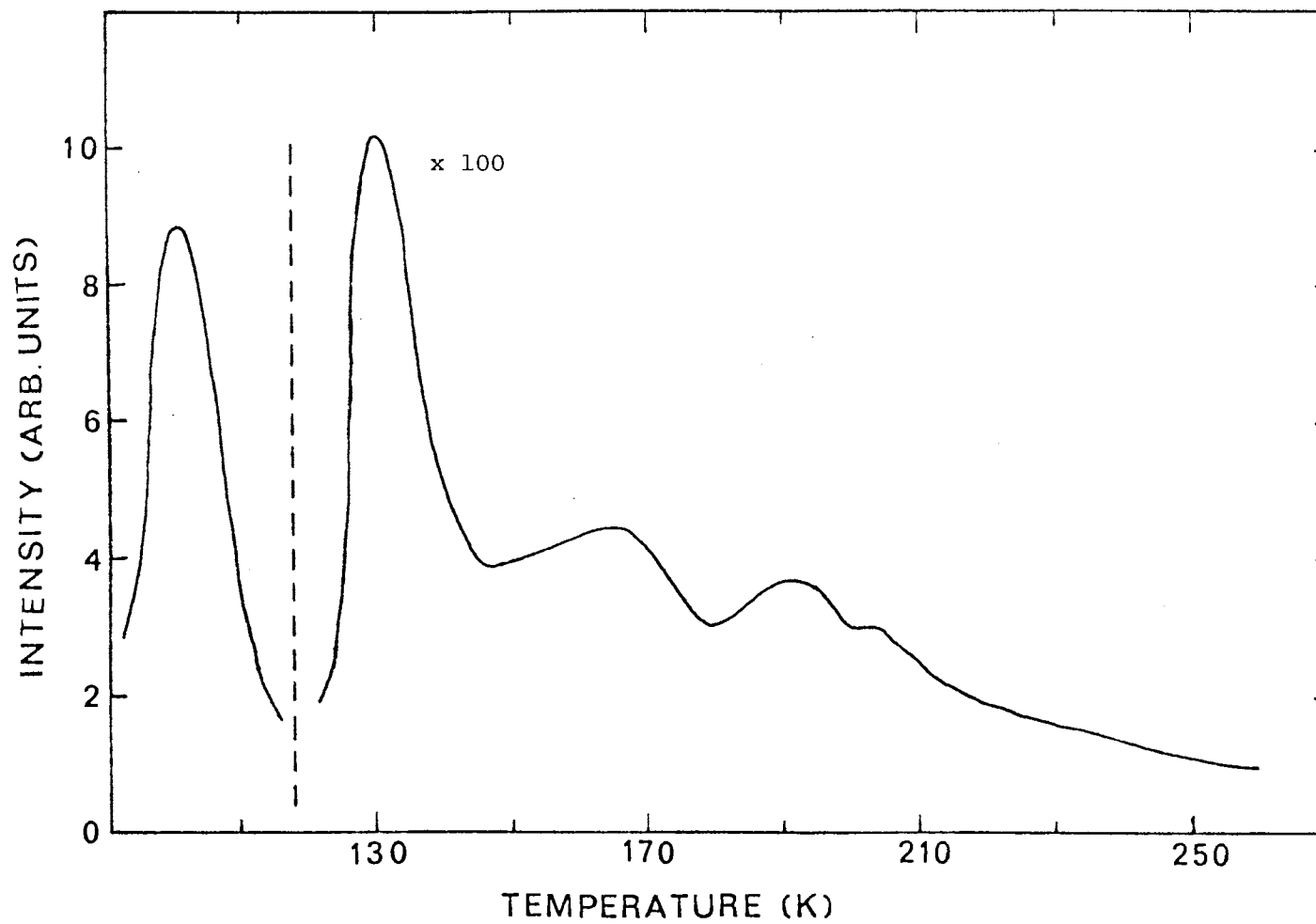


Figure 13. Low Temperature TSL of AlPO₄ (Second Sample) Irradiated at 96 K

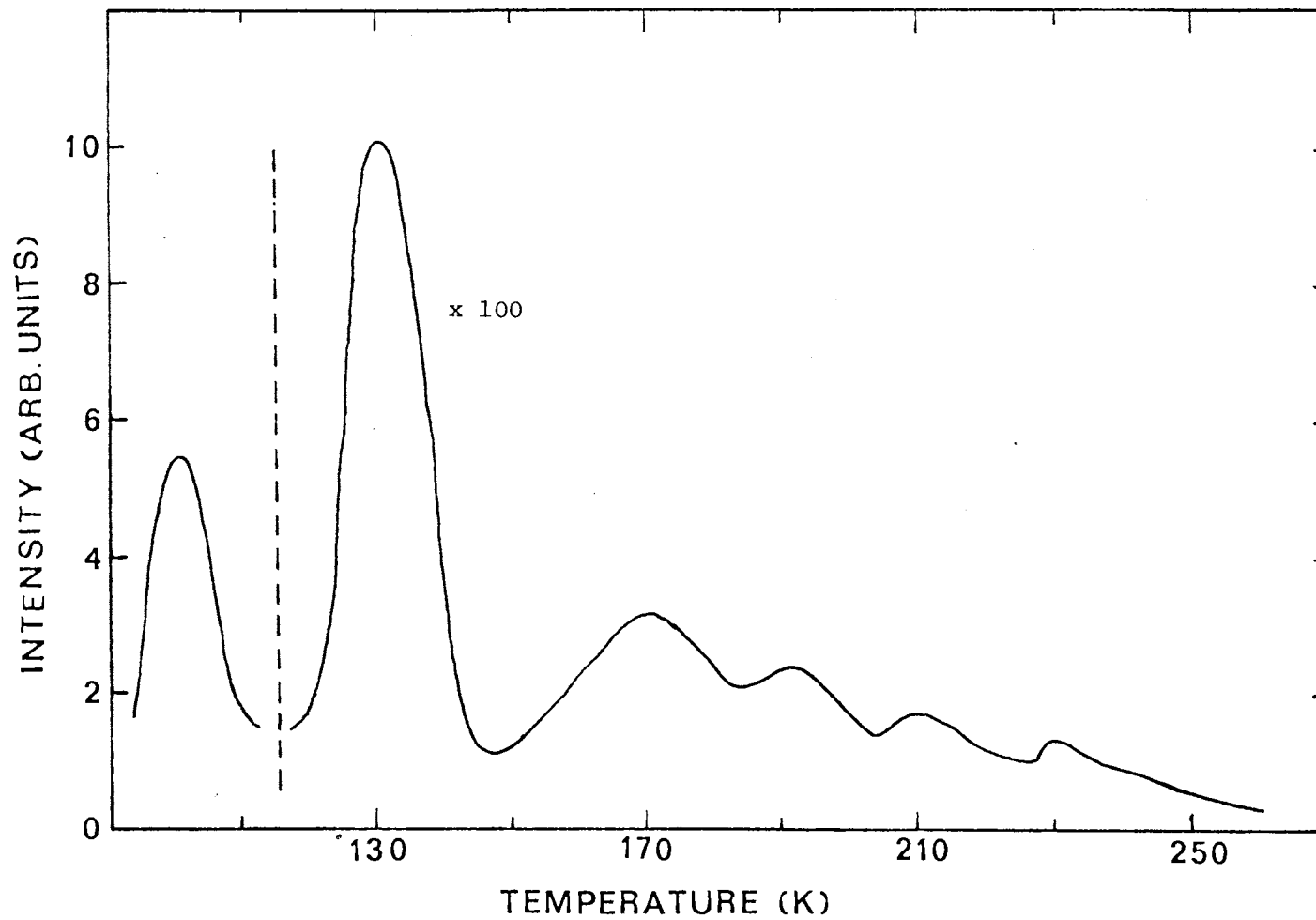


Figure 14. Low Temperature TSL of AlPO_4 (Second Sample) Irradiated at 300 K and 96 K

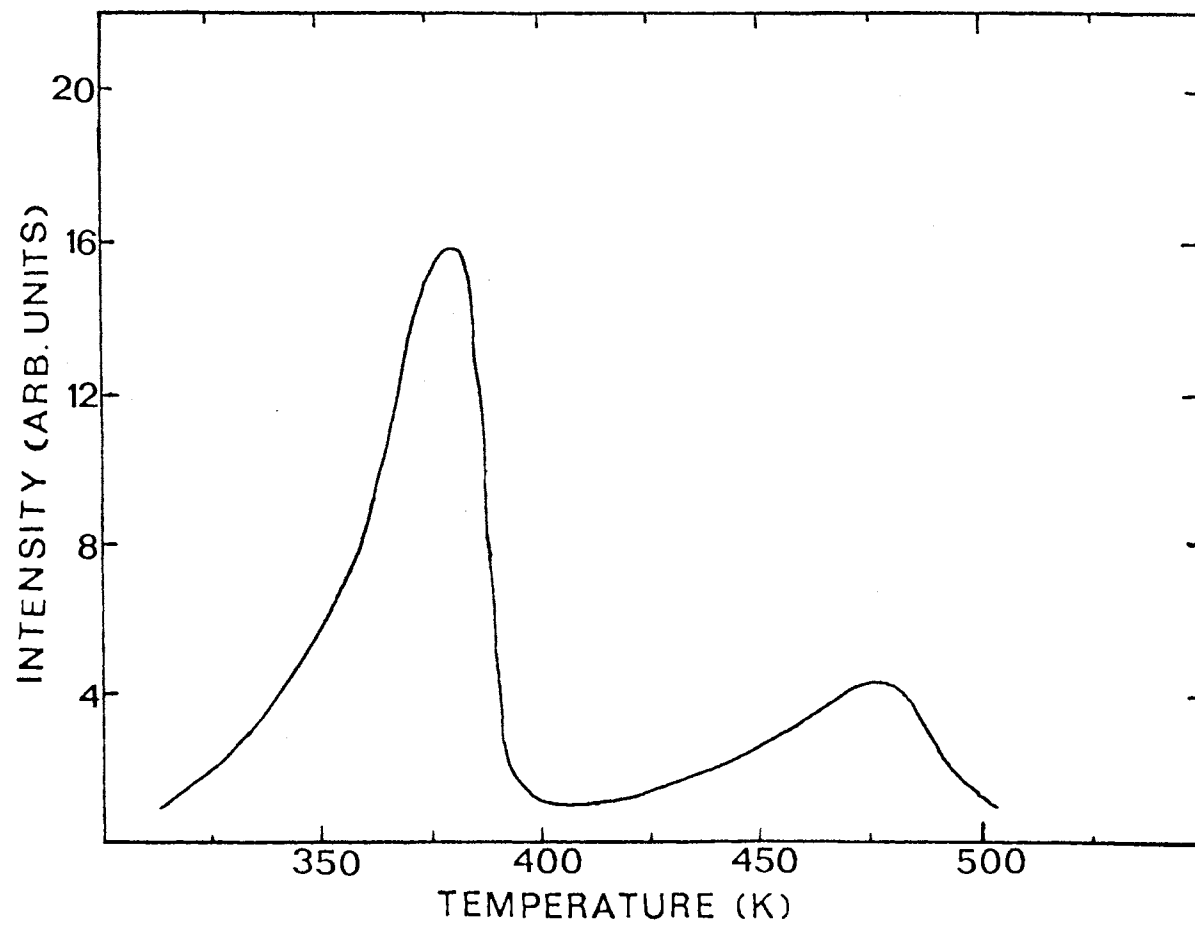


Figure 15. High Temperature TSL of AlPO_4 (Second Sample) Irradiated at 300 K and 96 K

above room temperature range and shows that as before there were two peaks at 380 K and 480 K.

To investigate the effects of another type of irradiation on berlinite, the sample was illuminated with ultraviolet (UV) light at liquid nitrogen temperature for 10 minutes using a source distance of about 1.5 meters. The resultant glow peaks were very small in amplitude and were observed at 130 K and 165 K as shown in Figure 11 with dotted lines, and at 380 K, 480 K as shown in Figure 12 also with dotted lines.

To observe some effects of ultraviolet bleaching, the sample was illuminated with 10 minutes of uv light at liquid nitrogen subsequent to the normal double irradiation procedure (electron irradiation at room temperature followed by a second electron irradiation at liquid nitrogen) used prior to the TSL run previously described in Figure 9. After the bleaching, a new TSL run showed the resultant peaks to be reduced in amplitude (dashed lines Figure 9) and the first peak to be absent — presumably because it was thermally removed by a slight increase of sample temperature associated with the UV irradiation. The above room temperature peaks were markedly reduced as shown in Figure 12 by dashed lines.

It was also observed that when the UV light was shined on a liquid nitrogen irradiated sample at room temperature, it appeared to be less effective in reducing the amplitude of the above room temperature peaks than when the UV illumination was carried out at liquid nitrogen temperature. This is shown in Figure 12 by the alternately dashed-dotted curve.

CHAPTER V

DISCUSSION

In general there were eight peaks observed in the temperature range of 96 K to 520 K.

The first glow peak appeared at ~ 100 K and its amplitude was on the order of one hundred times that of the other peaks. When the sample was irradiated and left at 96 K for a period of more than 60 minutes this peak essentially disappeared, indicating that the 96 K temperature provides enough thermal energy to at least partially depopulate the trap associated with it.

It was also desired to look at the spectral response of this peak but because of its sharpness in time duration, it was not practical. However, if the sample was irradiated at room temperature, then cooled to liquid nitrogen temperature in styrofoam cup, and again electron-irradiated there, it was observed that when it was subsequently quickly warmed up in air, the first glow peak observed by eye showed a violet color. This was distinctly different than the yellow-green color associated with higher temperature peaks. This observation indicates that the recombination mechanism of the first glow peak must be different from the others.

The second peak was fairly stable in temperature position and amplitude from run to run and appeared at about 130 K. This glow peak was always observed to have an amplitude comparable to or somewhat

higher than those observed above it in the 96 K to 300 K region. Nevertheless, the other four low peaks at 165 K, 190 K, 206 K and 220 K were individually distinguishable but they overlapped to an extent that suggested that before one recombination process was completed, the next had started.

The other two peaks above room temperature at 380 K and 480 K may not each be the result of just one recombination process. That they could indicate an overlap of recombination processes at small intervals of temperature is suggested by their shape, since they both slowly rise on the low side and then at 380 K and 480 K start to drop off more sharply.

The analysis of the results of UV illumination at liquid nitrogen temperatures indicated that it was relatively ineffective in populating defects although the most effect was noted on the peaks at 130 K, 165 K, 380 K and 480 K. However, when UV light was used to bleach the specimen after it had been electron-irradiated at low temperature, it was found that the amplitude of all peaks was somewhat reduced as might be expected. But when the bleach was done at room temperature, it was considerably less effective in reducing the amplitude of the 380 K and 480 K peaks than the liquid nitrogen temperature bleach. This suggests the lack of available low temperature traps after warming the sample to room temperature made it difficult to depopulate the higher temperature traps.

A. Correlation of TSL and ESR Results

The ESR work done by Halliburton et al [8] on berlinite shows that the decay of the observed defect centers due to thermal annealing

can be separated into three temperature regions of interest as shown in figure 16. In the 77-300 K temperature range two defects resulting from 77 K electron irradiation were observed to decay as the temperature was increased. The first one attributed to atomic hydrogen (H°) decayed near 125 K. It is tempting to correlate this with the first glow peak at 100 K temperature observed in the TSL. A similar type of emission was observed in quartz TSL results near 120 K and assumed to be associated with the presence of atomic hydrogen [23]. The second decaying center in this range is a B-7 center designated by Halliburton et. al. They observe it to slowly thermally anneal over the 100 K to 250 K temperature range. This appears to correlate with any of the several glow peaks found in the 120 K to 300 K temperature range of the TSL results. However, it is very difficult to associate any single peak with B-7 center decay observed in the ESR.

In the second region of 300 to 500 K, four defect centers are reported and designated as B-1, B-4, B-2 and B-3 centers. They all appear to decay near 425 K. However, the current TSL study shows two broad peaks at 380 K and 480 K which may be more indicative of actual decay combinations and decay overlaps.

In the third region, the 500 K to 600 K temperature range, two defect centers decays are reported and designated B-5 and B-6 centers. They anneal at about 550 K, above the temperature limit of the TSL study. Higher temperatures were beyond the capability of the cryostat apparatus used.

It has been suggested by Halliburton et al [8] that all the B centers may be variations of a basic defect consisting of a hole trapped in a nonbonding oxygen p-orbital. This hole-bearing oxygen would be

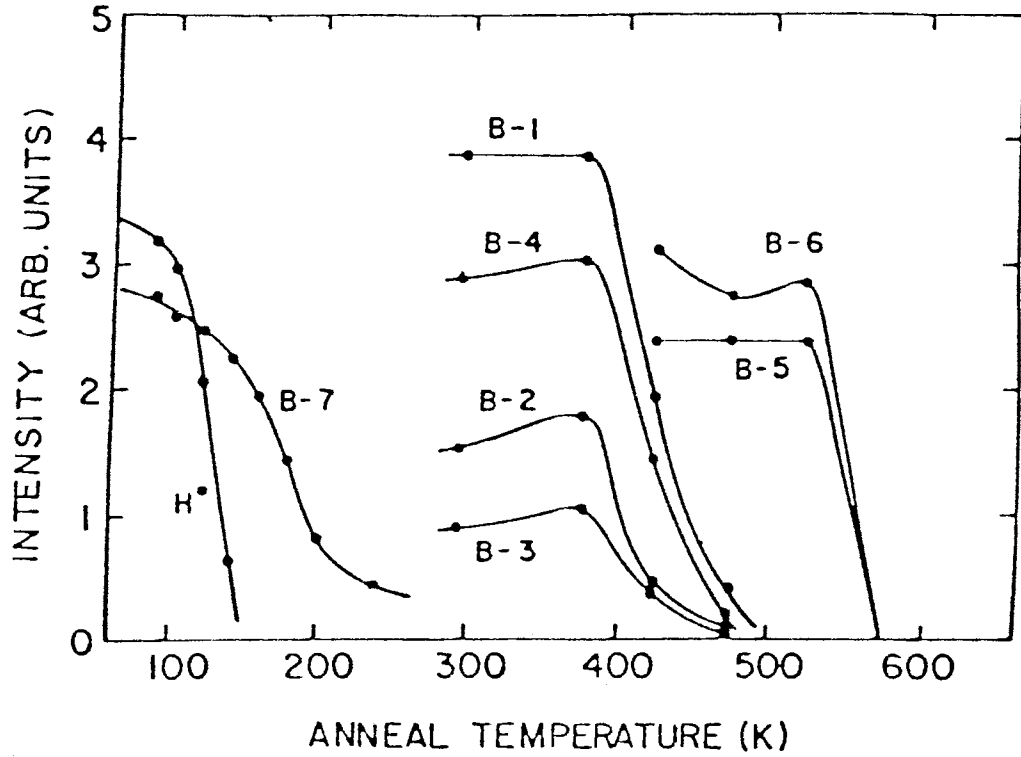


Figure 16. Thermal Stability of the Radiation Induced Defects in Berlinite Determined by 3-min Pulse at Progressively Higher Temperatures The Monitoring Temperature was 90 K. Taken from Halliburton et al [8].

stabilized within the lattice by an unidentified entity replacing the adjacent aluminium ion. The TSL work reported here is not inconsistent with this picture but suggests that slight modifications of such defects may exist and thermally decay at slightly different temperatures. The comparable spectral output (as visually observed) of peaks above 100 K might be interpreted as evidence that similar type recombination processes are occurring - presumably recombination at similar hole centers.

B. Suggestions for Further Study

The results reported by Dr. Halliburton et al [8] of ESR work showed only a B-7 center in the temperature range of 100 K to 300 K. However, the TSL work suggests that a more detailed ESR work might reveal more information in that region since there were five distinguishable glow peaks seen.

Also, it could be that a future TSL study of berlinite at temperatures below 96 K and beyond 520 K would give a clearer picture for correlation with the first H° center and the B-5, B-6 centers reported in the ESR study.

Specific spectral response data for different groups of TSL peaks could help to identify recombination processes. It would be particularly interesting to look at the TSL peaks above room temperature and see if their spectral response corresponds to that of peaks below room temperature. Because of relatively low emission, however, these measurements could prove difficult.

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VITA 2

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