DOE/ER/45376--2

DE91 000978

DYNAMICS OF SURFACE MELTING

Progress Report

for Period 1 January 1990 – 31 December 1990

Hani E. Elsayed-Ali

LABORATORY FOR LASER ENERGETICS
University of Rochester
250 East River Road
Rochester, NY 14623-1299
(716) 275-5101

OCT 2 2 1990

August 1990

Prepared for
The U.S. Department of Energy
Contract No. DE-FG02-88ER45376

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Notice

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed or represents that its use would not infringe privately-owned rights.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

The objectives of this program is to study the phenomenon of surface melting of single crystals of metals, to test for its existence, and to investigate its dynamics. Both conventional static electron diffraction and dynamic ultrafast electron diffraction are used in our study.

This year, the ultrahigh vacuum facility containing the picosecond electron reflection high-energy electron diffraction system was equipped with a cylindrical mirror analyzer and a static electron gun for Auger spectroscopy. An image analysis system capable of acquiring the pulsed diffraction patterns was assembled and used in analysis of picosecond laser heated surfaces.

A large set of time-resolved experiments were conducted to study the thermal response of Pb(110) to picosecond laser heating. The surface Debye-Waller effect was used to time-resolve the evolution of surface temperature. This provided us with a picosecond time-resolved surface lattice temperature probe. Results for laser fluences below surface melting show agreement with a heat-diffusion model.

The temperature dependence of the Pb(100) along the [110] and the [001] azimuths using x-ray photoelectron forward scattering of the $4f_{7/2}$ core-level photoelectrons confirmed, for the first time, surface melting of Pb(100) at temperatures as low as 560 K.

I. STATEMENT OF PROGRESS

The objective of this program is to study the phenomenon of surface melting of single crystals of metals. This involves testing for the existence of surface melting (the melting of the first few monolayers at a temperature below that of the bulk melting temperature) and possibly observing the dynamics (nucleation and growth) of the molten surface layer. For the past year our effort has been concentrated on the study of Pb(110) and Pb(100) surfaces. Surface melting of the Pb(110) surface was well demonstrated by ion channelling several years ago. This was also confirmed by a RHEED study we performed in the first year of this project. The Pb(110) is an open surface for which the phenomenon of surface melting is theoretically favored. Since this surface is relatively well studied, we are using it for our more complicated picosecond time-resolved study. We have also chosen to study surface melting of the Pb(100) since its atomic packedness falls between the open (110) surface for which surface melting is well demonstrated and the closed packed (111) for which others have shown that the surface melts very close to or at the bulk melting point.

This year, the ultrahigh vacuum facility containing the picosecond electron reflection high-energy electron diffraction system was equipped with a cylindrical mirror analyzer and a static electron gun for Auger spectroscopy. An image analysis system capable of acquiring the pulsed diffraction patterns was assembled and used in analysis of picosecond laser heated surfaces.

The thermal response of Pb(110) to picosecond laser heating was studied. The surface Debye-Waller effect was used to time-resolve the evolution of surface temperature. This provided us with a picosecond time-resolved surface lattice temperature probe. Results for laser fluences below that required for surface melting show agreement with a heat diffusion model. The development of our ability to monitor the surface temperature with up to ~100 ps time resolution and with better than 10 K accuracy is a major step to probe the kinetics of the surface melting phase transformation. A summary of our results can be found in the enclosed papers and manuscripts. Experiments utilizing this technique near the surface melting temperature are currently in progress.

Collaborative experiments with Professor Yongli Gao of the Department of Physics at Rochester were conducted to investigate the temperature dependence of the Pb(100) surface using angle-resolved photoelectron forward scattering. This study is unique in that, to our knowledge, it is the first to investigate surface melting in a (100) surface. While the phenomenon of surface melting is well established for the Pb(110) surface, previous experiments on Pb(111) have not confirmed the disordering of the surface below the bulk melting temperature. Our results show the existence of a surface melting phase transformation in Pb(100) at temperatures as low as 540 K.

We have studied the temperature dependence of Pb(100) along the [110] and [001] azimuths using x-ray photoelectron forward scattering of the $4f_{7/2}$ core-level electrons. Polar scans of the intensity of the backscattered photoelectrons as observed by our energy analyzer are shown in Fig. 1 for three different temperatures. One can readily see that the intensity of the scattered peaks decreases more rapidly as the bulk melting temperature is approached. Also the scattered peaks at larger angles with the normal seem to be more affected since at these steeper angles we are observing fewer surface monolayers. In Fig. 2 the intensity of the normal to the surface electrons was plotted for the two azimuths, for temperatures ranging from just above room temperature to 2 K below the bulk melting temperature. We observe that the Pb(100) surface shows characteristics of surface melting which are manifested as a deviation of the scattering peak intensities from Debye-Waller predictions at or above 540 K along both azimuths (bulk melting temperature of lead is 600.4 K). Comparison of the data for the [001] and the [110] azimuths indicates that the [001] azimuth begins to deviate from Debye-Waller prediction at a slightly lower temperature (less than 10 K) than that for the [110] azimuth. Extensive analysis is currently underway to better quantify the degree of surface disorder of Pb(100) with temperature and that of the anisotropy between the [110] and the [001] azimuths.

II. WORK STATEMENT FOR NEXT YEAR

Next year we plan to continue our main efforts on lead since we have been working on it and have developed a good understanding of its surface characteristics. The following tasks will be accomplished.

Task 1:

Our picosecond time-resolved surface temperature measurements of the laser heated surfaces will be continued. We will concentrate on the range of temperatures where we observed surface melting in Pb(110) and Pb(100). The RHEED patterns tend to be noisy at these high temperatures due to the large inelastic background. Therefore, our electron detection method using microchannel plates, which has an inherent background noise level due to x-ray generation by the high-energy electrons and due to some UV scattering from the photocathode, are replaced by a simple phosphor screen. To compensate for sensitivity loss a high repetition rate laser system is currently in its final stage of assembly and will be used in our experiments. External image intensifiers are also used, although these also have a background noise level. Our goal is to understand the surface melting phase transformation from an atomistic point of view by looking for its nucleation and growth stages in real time.

Task 2:

Our static surface melting studies will continue using RHEED and, in collaboration with Prof. Gao of Rochester, using angle-resolved photoemission. An image analysis system for quantitative RHEED studies is currently operational and will continue to be utilized for the analysis of the temperature dependence of the diffraction patterns. The scientific issues we will continue to investigate include the temperature dependence of the onset of surface disorder, the nature (melting versus roughening) of the disorder, the depth of the disordered layer with temperature, and the degree of anisotropy, if any, in the temperature dependence of the diffraction intensity.

IV. PERSONNEL ASSOCIATED WITH PROGRAM ON DYNAMICS OF SURFACE MELTING

PERSON	PERCENT SUPPORT ON CONTRACT (DE-FG02-88ER45376)
Hani Elsayed-Ali (Scientist) Principal Investigator	60%
John Herman (Graduate Student) (Physics)	50%
Elizabeth Murphy (Graduate Student) (Physics)	0% (Supported by a University of Rochester Fellowship)

REFEREED PUBLICATIONS

"Picosecond Time-Resolved Lattice Surface Temperature Probe," **H. E. Elsayed-Ali** and J. W. Herman, accepted for publication in Appl. Phys. Lett.

"Ultrahigh Vacuum Picosecond Laser-Driven Electron Diffraction System," **H. E. Elsayed-Ali** and J. W. Herman, Rev. Sci. Instrum. 61, 1636–1647 (1990).

"Comment on Thermal Response of Metals to Ultrashort-Pulse Laser Excitation," **H. E. Elsayed-Ali**, Phys. Rev. Lett. 64, 1846 (1990).

"Femtosecond Thermoreflectivity and Thermotransmissivity of Polycrystalline and Single Crystalline Gold Films," **H. E. Elsayed-Ali**, T. Juhasz, G. O. Smith, and W. Bron, submitted to Phys. Rev. Lett.

PUBLICATIONS IN CONFERENCE PROCEEDINGS

"Picosecond Transient Surface Temperature Measurement by Reflection High-Energy Electron Diffraction," **H. E. Elsayed-Ali** and J. W. Herman, Ultrafast Phenomena VII, C. Harris and E. Ippen (eds.) (Springer-Verlag, Berlin, 1990).

"Transient Surface Debye-Waller Effect," **H. E. Elsayed-Ali** and J. W. Herman, Picosecond and Femtosecond Spectroscopy from Laboratory to Real Word (SPIE, Bellingham, WA, 1990) Vol. 1209, pp. 76–85. (invited)

"Hot Electron Relaxation In Metals," **H. E. Elsayed-Ali**, High Energy Density Physics With Subpicosecond Laser Pulses (Optical Society of America, Washington, DC, 1989), Vol. 17, pp. 58–65. (invited)

"Femtosecond Thermomodulation of Single-Crystalline and Polycrystalline Gold Films," **H. E. Elsayed-Ali**, T. Juhasz, G. O. Smith, and W. E. Bron, Ultrafast Phenomena VII, C. Harris and E. Ippen (eds.) (Springer-Verlag, Berlin, 1990).

ABSTRACTS

"Picosecond Reflection High-Energy Electron Diffraction," **H. E. Elsayed-Ali** and G. A. Mourou, Int. Quant. Elect. Conf. (IQEC), Tokyo, Japan, July 1988.

"Hot Electron Relaxation in Gold Films with Different Structures," T. Juhasz, G. O. Smith, W. E. Bron, and **H. E. Elsayed-Ali**, Bull. Amer. Phys. Soc. 35, 831 (March 1990).

"Femtosecond Transient Thermomodulation of Thin Gold Films With Different Crystal Structures," **H. E. Elsayed-Ali**, T. Juhasz, and G. O. Smith, Conf. on Lasers and Electro-Optics (CLEO), Anaheim, CA, May 1990, p. 128.

INVITED PRESENTATIONS

"Picosecond Time-Resolved Electron Diffraction Studies of Laser Heated Metals," **H. E. Elsayed-Ali,** to be presented in Int. Conf. on Lasers '90, San Diego, CA, December 1990.

"Transient Surface Debye-Waller Effect," **H. E. Elsayed-Ali** and J. W. Herman, Picosecond and Femtosecond Spectroscopy from Laboratory to Real Word (SPIE, Bellingham, WA, 1990), Los Angles, CA, January 1990. (Also listed above).

"Hot Electron Relaxation In Metals," **H. E. Elsayed-Ali**, High Energy Density Physics With Subpicosecond Laser Pulses (Optical Society of America, Washington, DC, 1989), Vol. 17, pp. 58–65, Snowbird, UT, September 1989. (Also listed above).

PATENT APPLICATION

"System for Surface Temperature Measurement with Picosecond Time Resolution," **H. E. Elsayed-Ali.**

Figure 1

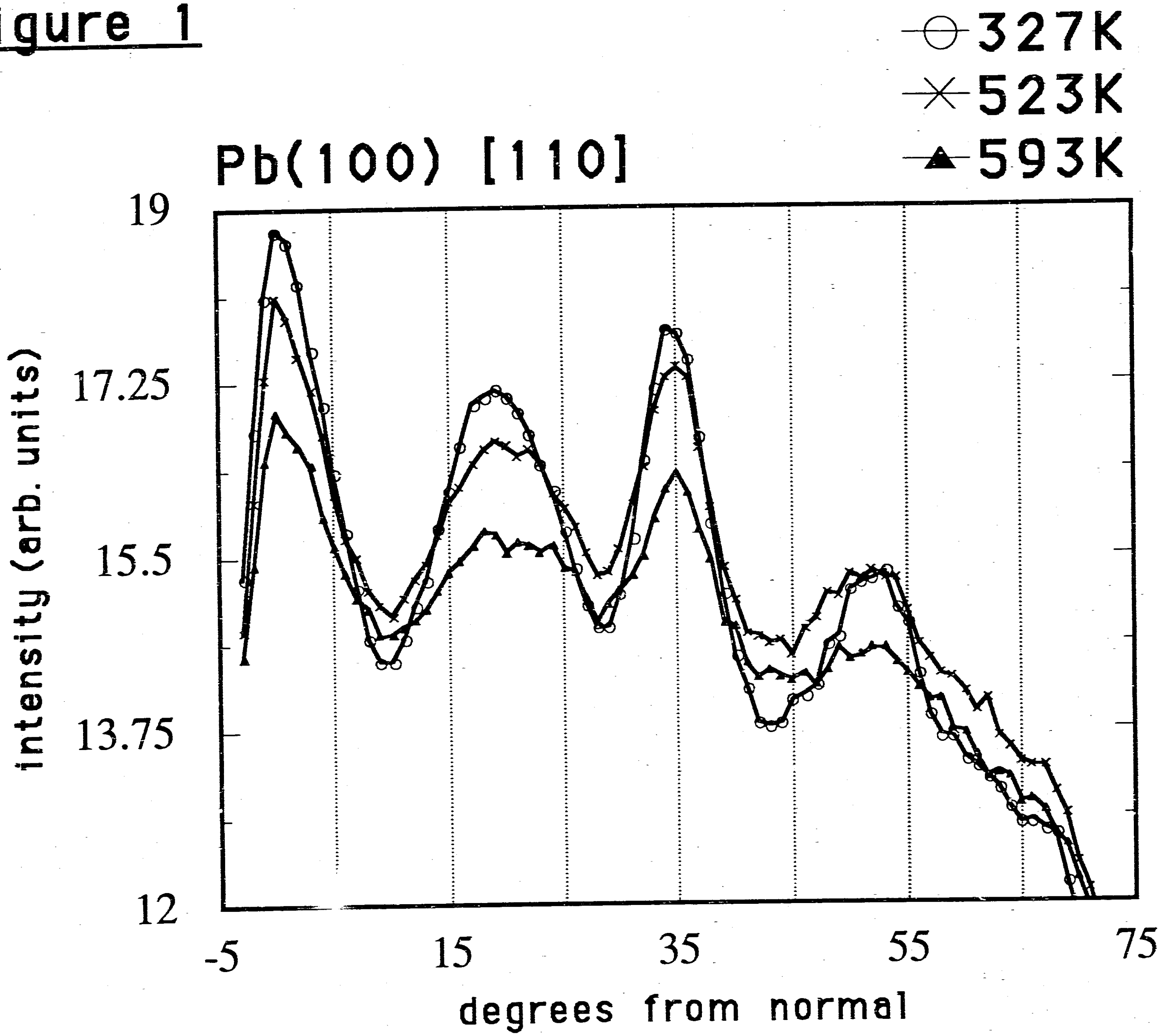

327K
523K
593K
Pb(100) [110]
19
17.25
15.5
13.75
12
intensity (arb. units)
-5
15
35
55
75
degrees from normal

Figure 2a

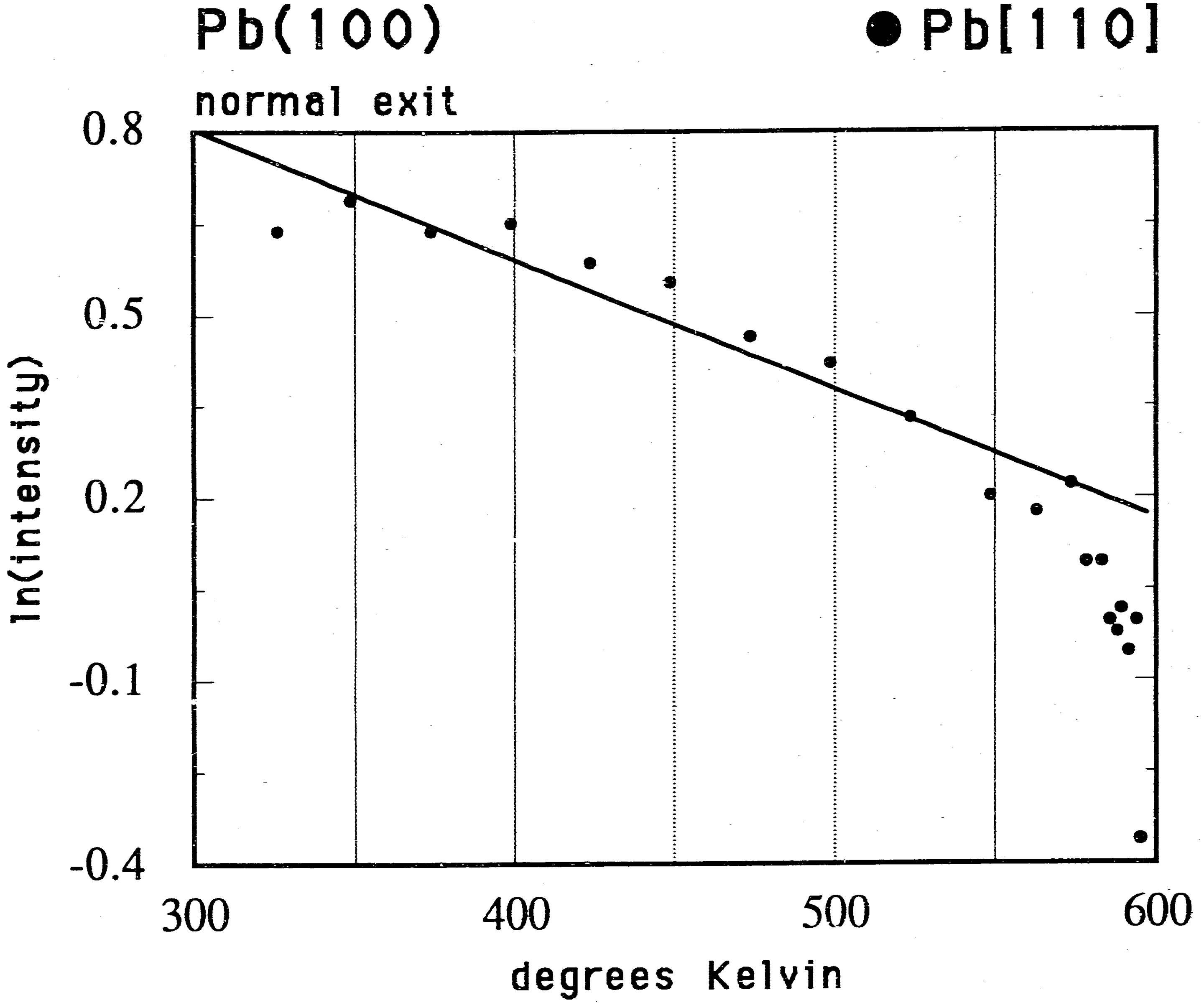
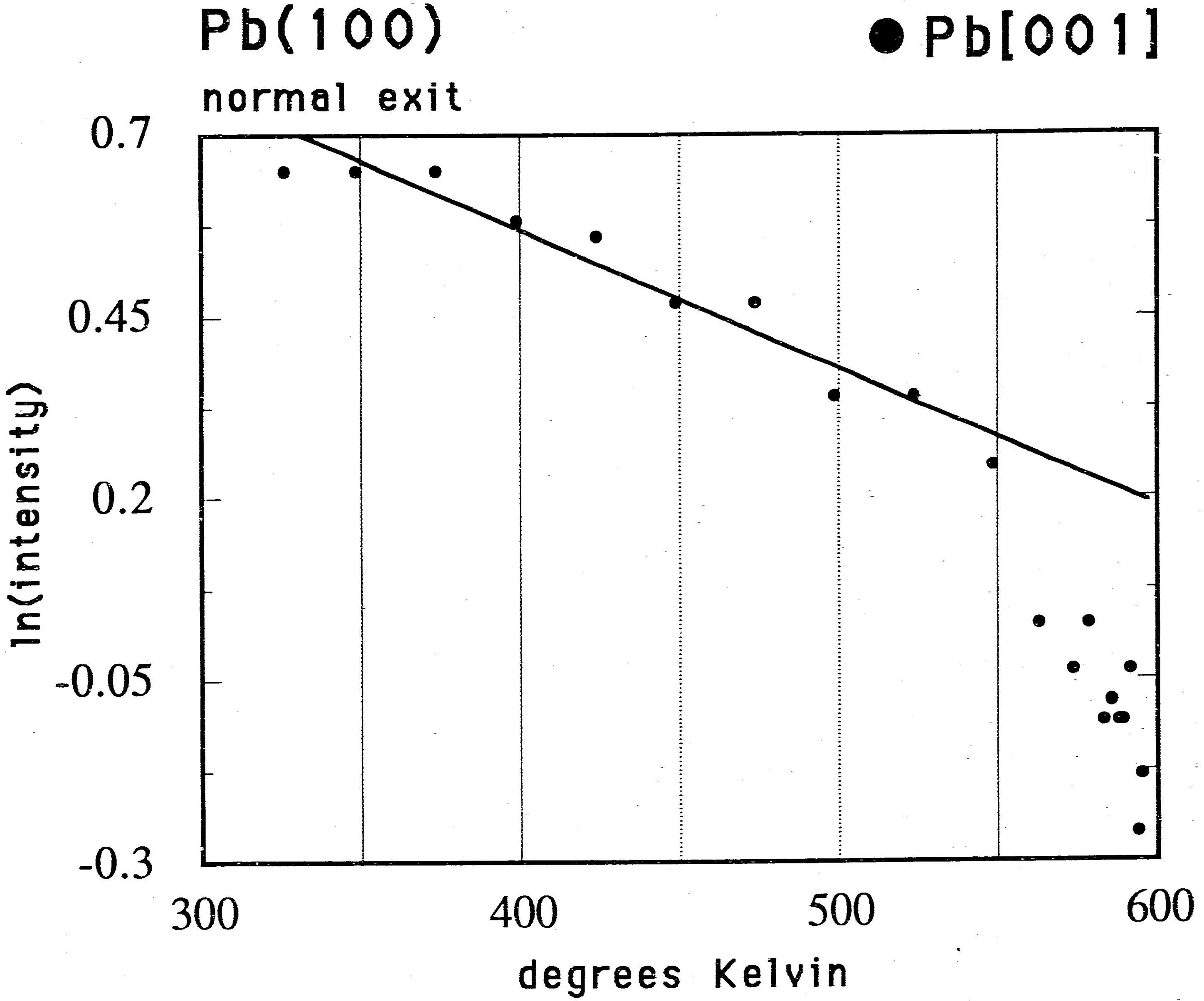

Pb(100)
normal exit
Pb[110]
0.8
0.5
0.2
-0.1
-0.4
ln(intensity)
300
400
500
600
degrees Kelvin

Figure 2b


Pb(100)
normal exit
Pb[001]
0.7
0.45
0.2
-0.05
-0.3
ln(intensity)
300
400
500
600
degrees Kelvin

- END -

DATE FILMED

11 / 05 / 90