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DYNAMICS OF SURFACE MELTING

Progress Report

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ABSTRACT

The objectives of this program is to study the phenomena of surface melting of single crystals of metals, to test for its existence, and to investigate its dynamics using picosecond reflection high-energy electron diffraction (RHEED).

In this year, the UHV facility containing picosecond RHEED has become fully operational with a dedicated laser system and image acquisition capabilities. Details of this facility are described. This system was utilized to study surface heating and cooling of a Pb(110) crystal subjected to picosecond laser irradiation. Diffraction pattern sensitivity to temperature is due to the surface Debye-Waller factor which results in a reduced number of elastically scattered electrons with lattice heating. Development of a picosecond time-resolved surface temperature probe is a key step to study the dynamics of surface melting that was well demonstrated in our present work. A heat diffusion model of surface heating and cooling has also been constructed.

Results on static RHEED experiments on surface melting of Pb(110) crystal are presented. The existence of a surface melting (disordering) transformation at temperatures at least 50°K below the bulk melting point was observed in our experiments. Quantitative RHEED study of this phenomena are in progress.

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 conditions necessary 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. Our effort has been concentrated on the study of Pb(110) surface mainly because it is currently the standard surface that is used for surface melting studies (Pb has a relatively low bulk melting point which facilitates the experiments and the 110 surface is an open surface for which the phenomenon of surface melting is theoretically favored in addition to other reasons discussed in our proposal.)

Our approach is to use conventional cw reflection high-energy electron diffraction (RHEED) in an ultrahigh vacuum system in order to monitor the surface structure as a function of temperature. The novel technique of picosecond time-resolved laser-driven RHEED is used to probe the temperature (through the surface Debye-Waller factor) and the structure of the Pb(110) after picosecond laser heating.

Our cw RHEED study on Pb(110) has confirmed surface disordering at temperatures significantly below (more than 50°K) the bulk melting point. An interesting observation is the total disappearance of diffraction rods due to elastically scattered electrons at ~570°K while the Kikuchi lines which originate from deeper layers remained well observable. Quantitative analysis of these patterns using our recently developed image analysis techniques is currently underway. One key development of the present work is the demonstration and use of picosecond RHEED as a time-resolved surface temperature probe with ~200 picoseconds time resolution. This capability is very important for the study of the dynamics of the surface melting process.

The basic idea of the technique of picosecond RHEED is the utilization of a picosecond laser pulse to create an electron pulse with equal time duration. These photogenerated electrons can be collimated and focussed to make them suitable to obtain a good RHEED pattern from the surface of the studied crystal. Typically we use a Nd:YAG laser ($\lambda = 1.06\ \mu m$) to irradiate the sample while the electron probing pulse is generated by irradiation of a photocathode with a frequency quadrupled Nd:YAG ($\lambda = 0.266\ \mu m$). Frequency quadrupling allows us to use a rugged metallic photocathode. Only a very small part of the Nd:YAG fundamental is converted to the ultraviolet, thus, most of the laser energy is available to irradiate the sample. By spatially delaying the ultraviolet laser pulse from the fundamental it is possible to obtain RHEED patterns of the studied sample from a few hundred picoseconds to up to tens of nanoseconds after laser irradiation.

The sensitivity of the diffraction pattern to temperature comes from the fact that at temperatures below surface melting, as the temperature is raised, there is an increased atomic vibrational amplitude and thus an increased dephasing of the atomic scattering centers. The effect of this dephasing on the diffraction pattern is to reduce the number of electrons elastically scattered (i.e., to reduce the diffraction rod intensity). The reduced rod intensity shows up as increased background which is due to inelastically scattered electrons. This is known as the Debye-Waller effect.

We have conducted a measurement of the surface Debye-Waller effect with a few hundred picosecond time resolution on a Pb(110) crystal. The 1.06 μm fundamental was used to heat the surface of the sample while the electron pulses were used to probe the surface in the manner described previously. The infrared heating pulse passed through a delay line before hitting the sample. Adjustments of the delay line provided the means by which the electron pulse arriving to the sample was delayed a set amount of time from the time the heating pulse strikes the sample.

The diffraction pattern was masked except the 02 diffraction rod which was monitored with a photomultiplier connected to a gated integrator. Results on the percentage change in the diffraction rod intensity as a function of the position of the delay line (arrival time of the laser pulse) showed quantitative agreement with that of the heat diffusion model. Further experiments aimed at minimizing the standard of deviation of our data by more averaging, increase the uniformity of the laser irradiation, and better quantify the change in diffraction rod intensity by image analysis techniques are currently in progress.

II. WORK STATEMENT FOR NEXT YEAR

For next year we plan to continue to concentrate our efforts on lead, particularly the 100 surface, although the 100 and 111 surfaces will also be investigated. The following tasks will be accomplished.

Task 1:

The cw RHEED studies that we performed this year on the Pb(110) will be continued with the goal of performing a quantitative analysis on the diffraction pattern. The image analysis techniques we developed this year will be used to accomplish this goal. The change in the intensity of the diffraction rods with temperature will be quantitatively recorded and compared to that predicted from the surface Debye-Waller temperature. Deviation from that predicted by a Debye-Waller theory is expected at the onset of the occurrence of surface melting. These deviations will be analyzed and interpreted in view of the disordering of the first few monolayers with temperature. The intensity of the background and that of Kikuchi lines will also be quantitatively investigated.

Task 2:

The picosecond time-resolved RHEED studies will be continued. Our goal is to quantitatively compare the surface transient temperature measurements in response to picosecond laser heating to our numerical heat diffusion model. Ultrafast laser heating will be accomplished for the sample set at a particular temperature using the resistively heated stage in the UHV chamber. Agreements and deviations from the heating/cooling response predicted by the heat diffusion model will be pointed out. Since the heat diffusion model is sensitive to the heat capacity and conductivity of the laser irradiated material, a change in the properties of the surface layer by melting could be detectable depending on the magnitude of the effect and the accuracy of our temperature measurements. Our target is to reach a temperature measurement accuracy of better than 50°K with a time resolution better than 200 ps.

Task 3:

We will continue our picosecond time-resolved RHEED studies with the goal of observing the structure of the surface in response to a fast heating laser pulse. These measurements will be similar to those in Task 2, except that the complete RHEED pattern will be recorded with ~200 ps time resolution. analysis of the diffraction patterns will include looking for changes in the lattice spacing by measuring the diffraction rod spacings which are inversely proportional to that of the lattice. Correlation of these measurements with the transient Debye-Waller factor measurements will be made.

III. PUBLICATIONS

H. E. Elsayed-Ali and J. W. Herman, "An Ultrahigh Vacuum Picosecond Laser Driven Electron Diffraction System," to be submitted to Review of Scientific Instruments, August 1989.

H. E. Elsayed-Ali and J. W. Herman, "Transient Surface Debye-Waller Effect," Invited paper to be presented at Picosecond and Femtosecond Spectroscopy, The International Society for Optical Engineering, Los Angeles, January 1990.