Section 3
ADVANCED TECHNOLOGY DEVELOPMENTS

3.A Time-Resolved, Laser-Induced Phase Transition in Aluminum

Phase transformation in condensed matter is an important area of study in solid-state physics and material science, since it relates to the microstructural evolution of matter, yet the mechanism and time history of such a transformation is still not known. Several theories have been proposed to describe this phenomenon, but serious flaws are evident in all of them. A basic problem has been the lack of experimental information upon which to base a model. The laser has become a valuable tool in the study of melting processes in the past few years. However, only indirect inferences have been obtained concerning any time-resolved structural evolution, since the probes have measured changes in surface reflection,[1,2] surface second-harmonic generation,[3] electrical conductivity,[4] and lattice strain[5] during laser-induced phase transitions.

Recently, a technique has been developed and implemented at LLE that enables laser-induced order-disorder transformations to be directly monitored on the picosecond timescale.[6] The technique is based upon observing the electron diffraction pattern of the phase of the system under study. By using a short pulse of monoenergetic electrons to probe a thin specimen, it is now possible to take a snapshot of the system's ordered (or disordered) structure. The duration of the electron pulse is ~30 ps. Synchronized to this electron pulse is a laser pulse which is used to stimulate the phase transition (i.e., melt the solid). By probing the sample with the electron pulse at known times after the laser stimulus, information can be obtained on the evolution of the melting process.

For example, the actual time required for a given system to undergo a laser-induced phase transformation can now be determined. Conflicting theories placed this number between a few picoseconds,[7] where the cloud of laser-generated hot electrons rapidly transfers energy to the lattice, and the lattice dissolves; and >200 ns,[8] where the lattice only "softens" due to the increased temperature, and the ions remain relatively stationary. The picosecond electron-diffraction technique is well-suited for taking this measurement.

The Experiment

The experimental arrangement used to time-resolve the phase transformation is shown in Fig. 17.37. The electron-pulse generator is a modified streak camera tube. In its normal configuration, this tube is a diagnostic tool capable of time-resolving short optical pulses. The tube converts an optical pulse to an electron pulse through the photoelectric effect while preserving the temporal profile of the pulse. The electrons are then accelerated and deflected or "streaked" to map the temporal information of the pulse across the phosphor screen. The key features of this tube are the following: (1) It is capable of generating short electron bursts down to single-picosecond durations. (2) The electrons are monoenergetic to one part in 10^4. (3) The electron pulse can be synchronized to the original optical pulse with picosecond accuracy. For the purpose of this experiment, the deflection plates are of no use and have been removed. The remaining components are the photocathode, grid, focus cone, and anode. Once generated, the electron pulse accelerates through the tube to the anode and then "drifts" at a constant velocity $\sim 10^8$ m/s. The specimen to be studied is positioned in this drift region. The electron pulse passes through the specimen (aluminum in this case) that is of a proper thickness ($\sim$ 240 Å) such that each electron in the pulse sustains, on the average, one elastic collision. The aluminum films were made by first depositing aluminum onto a Formvar substrate and then vapor-etching to dissolve the Formvar. The singly scattered (diffracted) electrons then travel to the phosphor screen where they are visualized. The diffraction ring pattern produced, shown in Fig. 17.38(a), is a result of the wave-like properties of the electrons and the ordered polycrystalline nature of the aluminum specimen. By careful measurement of the ring diameters and by use of the Bragg relation, the lattice constant for the crystal is determined to be 4.03±0.08 Å, in close agreement with the known value of 4.05 Å.

The signal-to-noise ratio of the pattern is rather low, due to the low electron flux required to maintain a 30-ps pulse width. Increasing the electron flux causes space-charge broadening to occur, which stretches the pulse duration. To make the best possible use of the available signal, circular signal averaging about the center of the diffraction pattern is performed. Circular averaging smooths the randomly generated noise to a roughly constant background level, thus greatly enhancing the signal-to-noise ratio. Figure 17.38(b) shows the result of circular averaging, which is accomplished by simply spinning a photograph of the diffraction pattern.

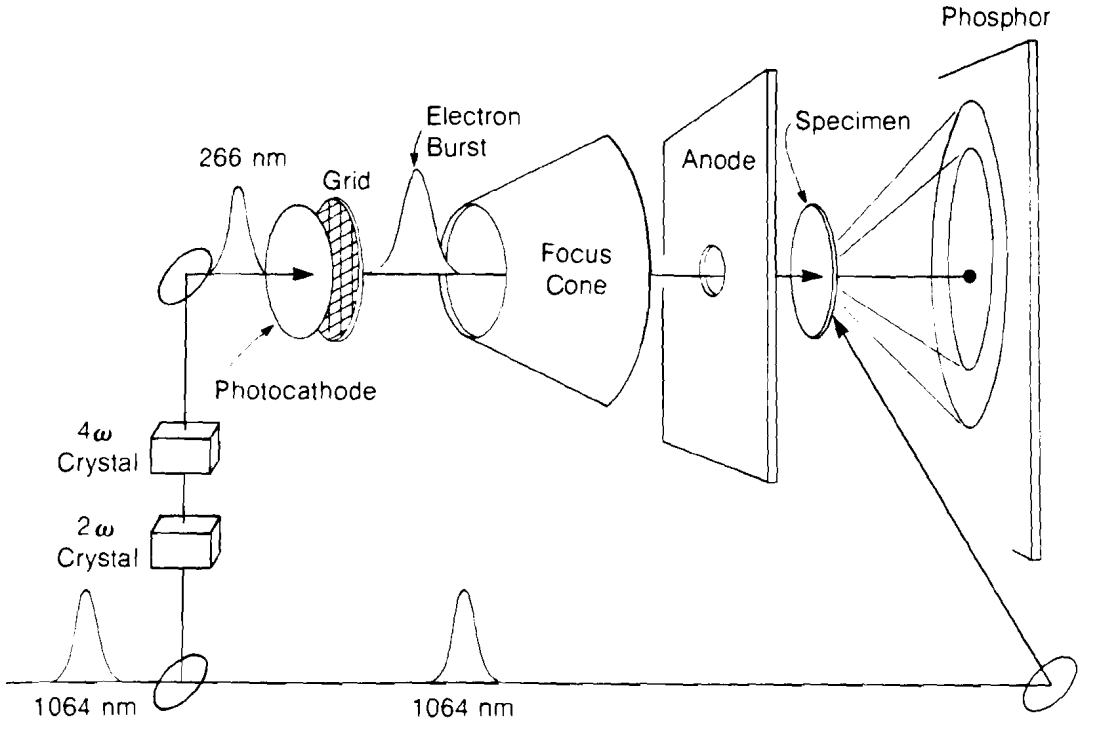

Fig. 17.37
Experimental layout for monitoring the laser-induced phase transition occurring in aluminum. The electron-pulse generator is comprised of a photocathode (at -25 kV), grid, focus cone, and anode (at ground potential). The specimen is located in the path of the electron beam. A variable-delay arm was placed in the 1064-nm beam in order to allow for laser-stimulus/electron-probe synchronization.

It is important that the laser stimulus be uniform in space over the probed region of the specimen. Therefore, the laser-beam diameter was expanded to ~6 mm to overfill the 2-mm specimen diameter, and the electron-probe diameter was reduced to ~1 mm. In this configuration, the probed region of the aluminum has a variation in fluence from the stimulus of no more than 15% from center to edge. The absorption of the laser stimulus (λ=1064 nm) by the specimens is 13 ±1%.

Synchronization between the laser stimulus and the probe pulse is accomplished by means of a laser-activated electron-deflection technique.[9] A fast deflection-plate assembly through which the electron pulse travels is positioned in place of the specimen. A photoconductive switch, activated with picosecond accuracy by the laser stimulus, charges the plates, establishing an electric field in a time on the order of the laser-pulse duration (~ 30 ps). If the laser stimulus arrives early, relative to the electron pulse, the electrons deflected by the electric field are detected at a new position on the phosphor screen. Proper synchronization between the two pulses is achieved when the electron pulse is evenly streaked between the undeflected and fully deflected positions on the phosphor screen.

The procedure followed is to irradiate the specimen with the laser stimulus and, at various times thereafter, to probe the structure of the aluminum with the electrons. This process was repeated at several laser-pulse energy levels. Figure 17.39 displays two diffraction patterns of the same aluminum specimen (a) before and (b) during the laser-induced melt phase. The most distinct difference between the two figures is seen in the nearly complete loss of intensity of the two outer rings in Fig. 17.39(b). In this case, the ordered structure has been degraded as a consequence of the formation of a liquid phase. The specimens were used only once, although at fluence levels of 7-10 mJ/cm^2, the specimens frequently survived multiple shots.

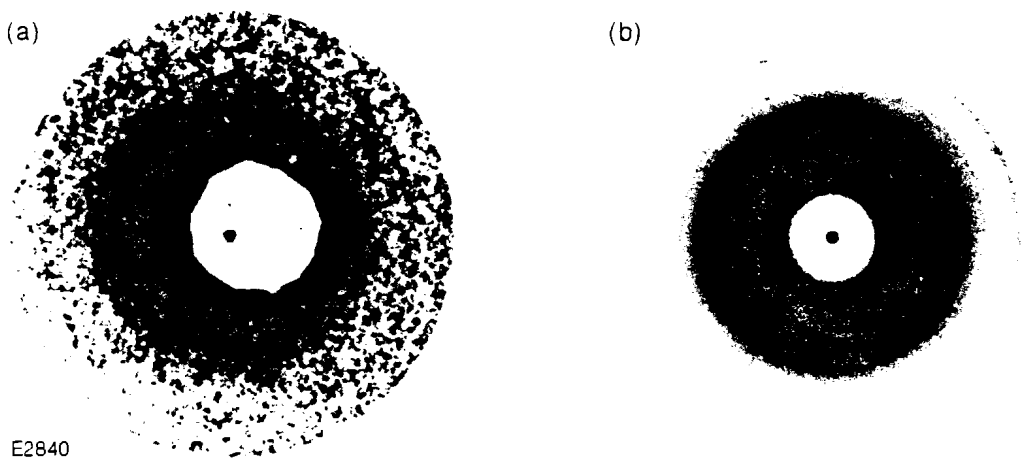

Fig. 17.38
(a) Photograph of the diffraction pattern of a 240-Å-thick film of aluminum as seen on image intensifier. The dark mask at the center (#96 wratten filter) acts to block the intense zero-order mode of the diffraction pattern. Only the intense center spot transmits through to the film.
(b) The center of the rings is the zero-order spot. Rotating the image about this point results in an appreciable enhancement in the signal-to-noise ratio. The background noise is smoothed to a constant background level, causing the rings to stand out more clearly.

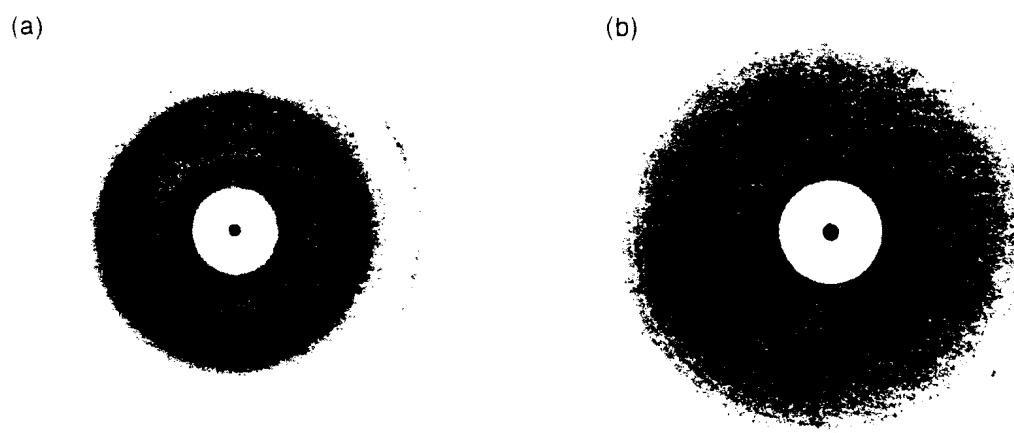

Fig. 17.39
(a) A diffraction pattern of aluminum prior to the arrival of the laser stimulus.
(b) The same specimen as used in (a), but in the liquid phase. The electron probe follows the laser stimulus by 1 ns.

Figure 17.40 shows the results of this preliminary experiment. The curve represents the fluence required to transform the aluminum specimen from the solid to the liquid phase at a given delay time. We see that the curve falls exponentially to an asymptotic value corresponding to the latent heat of the liquid state. At short delay times (<20 ps), the curve becomes indistinct due to the limited temporal resolution of the probe.

Though the actual phase transition has not been time-resolved, it clearly occurs on a timescale shorter than 20 ps. The fact that an increase in fluence is required to cause the phase transition for short delays (~ 100 ps) implies that for lower fluences at such delays, the specimen must be in a super-heated solid state.

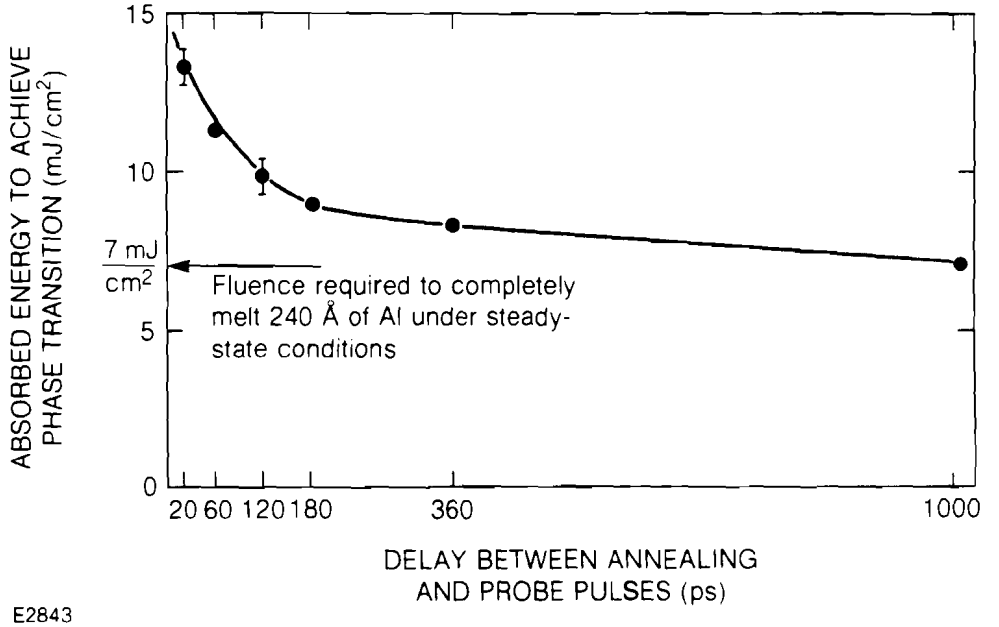

Fig. 17.40
This plot shows the absorbed energy per unit area required to achieve a phase transition after a given delay time. The error bars span the two discrete stimulus levels used that were found to be immediately above and below the level required to cause the phase transition at the indicated time delay.

Summary

We have demonstrated an application of the picosecond electron-diffraction technique by resolving the delay between the laser stimulus of a polycrystalline solid and the moment the melt occurs. True time resolution of the solid-liquid transition will require that the probe duration be much shorter, perhaps down to the single-picosecond timescale. With the present system, however, a great deal of information can be collected about laser-induced phase transitions. In addition to monitoring changes in phase, the electrons, under optimal conditions, can measure the change in the lattice constant resulting from an increase in temperature. This information will become most valuable in determining how far the melt temperature is exceeded before the system collapses into the liquid (or vapor) phase.

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