1. INTRODUCTION

The fundamental process responsible for initiating the melting of condensed matter has remained a mysterious and, as yet, unsolved problem. The long-standing interest in this most basic of phase transitions has nevertheless resulted in a great number of theoretical interpretations. The key question still to be answered is, what happens on the atomic scale when a crystal melts?

Theories are available that deal with the actual microscopic mechanism of melting (1-4). However, as is often the case, progress on experimental verification has lagged behind. The difficulty that arises is in making a microscopic observation on a time scale appropriate to the ongoing process.

In the past ten years the short-pulse laser has evolved as an ideal tool for studying phase transitions. In addition to supplying energy densities large enough to create a melt in a time much shorter than the transition time, the optical pulse can also be used to probe the melting substance. An important consequence of this feature is that we can now drive a solid to a superheated temperature faster than the system can respond. Figure 1 shows a conventional phase diagram where path 1 is followed under normal equilibrium conditions. If a quantity of energy, $E_a$, is rapidly deposited into the system, the path taken is that along the equipartition energy line, path 2. Depending on the peak power of the laser pulse and the system in question, the range over which superheating occurs is several thousand degrees. The time $\tau_{\text{melt}}$, is then the actual material-dependent melting time. The departure of path 1 from the equipartition line is indicative of defect formation. With the short-pulse laser and a suitable probe, the rate of defect formation even below the melt threshold can be time-resolved.

Several probe techniques have now been developed to time-resolve phase transformations in semiconductors during laser annealing. However, most of these probes (e.g., electrical conductivity (5,6), optical reflection (7,8), optical transmission (9), Raman scattering (10), and time of flight mass spectrometry (11)) supply no direct information about the atomic structure nor the temperature of the material. Probing the structure can reveal when and to what degree a system melts as it is defined by degradation in the long-range order of the lattice. True structural probes based on X-ray (12) and low-energy electron diffraction (13), and EXAFS (14) with nanosecond time resolution, have been developed offering fresh insight into both the bulk and surface dynamics of material structure. Also, a subpicosecond probe based on structural dependent second
FIGURE 1. Caloric diagram of the solid and liquid phases for a simple metal. Path 1 is followed when the energy is deposited on a time scale slower than the defect formation time. Path 2 is followed when the energy is deposited instantaneously.

harmonic generation (15) has been demonstrated. But at present, only the technique of picosecond electron diffraction (16) can produce an unambiguous picture of the atomic structure on the picosecond time scale.

In this paper we describe an experiment utilizing this instrument to time-resolve the laser-induced phase transition in aluminum. The results are then interpreted in the context of the defect theory of melting.

2. EXPERIMENT AND RESULTS

The technique takes advantage of the strong scattering efficiency of 25 keV electrons in transmission mode to produce and record a diffraction pattern with as few as $10^4$ electrons in a pulse of 20 ps duration. The burst of electrons is generated from a modified streak camera that, via the photoelectric effect, converts an optical pulse to an electron pulse of equal duration (17). Also of importance is the fact that the electron pulse can be synchronized with picosecond resolution to the laser pulse (18). The experimental arrangement is illustrated in Fig. 2. A single pulse from an active-passive modelocked Nd$^{3+}$:YAG laser is spatially filtered and amplified to yield energies up to 10 mJ. The streak tube (deflection plates removed), specimen, and phosphor screen are placed in vacuum at $10^{-6}$ mm Hg. The electron tube is comprised of the photocathode, extraction grid, focusing cone, and anode. A gold photocathode is used to permit the vacuum chamber to be opened to air. The photocathode is held at the maximum voltage (-25 kV) so that space charge, which can cause significant temporal broadening, is minimized. The portion of the laser irradiating the photocathode is first upconverted to the fourth harmonic of the fundamental wavelength in order to