

Inelastic Electron-Phonon Scattering and Time-Resolved Laser-Induced Phase Transformation in Aluminum

In a beautiful experiment Williamson, Mourou, and Li¹ have measured directly the time it takes for the solid-liquid transition of a free-standing aluminum film. This was done by a direct measurement of the disappearance of electron diffraction patterns after the application of a short laser pulse. The results are interpreted¹ as due to an expansion of a critical nucleus of the melt throughout the solid sample thus leading to complete melting. One may doubt the validity of this model for melting through nucleation and, in addition, the model depends on parameters which are not independently measured. It is the purpose of this comment to propose a different explanation of the results of Williamson, Mourou, and Li; this alternative explanation relies on the inelastic electron-phonon scattering in the system.

In order to determine whether Fig. 3 of Ref. 1 could be related to the time for the homogeneous melting of the sample, we have performed a molecular-dynamics simulation of the melting of a Lennard-Jones crystal.² In this simulation a set of 4000 particles with periodic boundaries interact via Lennard-Jones potentials at constant pressure and some prescribed starting temperature T_s . This system, at T_s and zero pressure, forms a stable fcc crystal at values of T_s below ~ 0.8 in the usual molecular-dynamics reduced units. An fcc crystal was equilibrated at zero pressure and $T_s = 0.75$, and at a time, conveniently referred to as $t = 0$, the temperature of the system was raised from T_s to $T = 0.8$, i.e., slightly above the melting temperature. The equations of motion of all particles were solved simultaneously and followed in time, as is usual in molecular-dynamics simulations. Figure 1 shows the intensity of the (100) fcc peak as a function of time. Notice that after $t \sim 8$ psec (for argon) this intensity drops essentially to zero, indicating a melting of the Lennard-Jones crystal. After the melting process is over the diffusion constant was found to be $D = 4.8 \times 10^{-5}$ cm²/sec, characteristic of self-diffusion in liquids; before heating, i.e., before $t = 0$, the solid in equilibrium showed mean square atomic displacement $\langle r^2 \rangle \cong 0.76 \text{ \AA}^2$ with no diffusion. Another simulation with $T_s = 0.7$ (i.e., the system at a temperature further down in the solid range) gave $\langle r^2 \rangle \cong 0.48 \text{ \AA}^2$, $t = 27$ psec, and $D = 4.7 \times 10^{-5}$ cm²/sec when the temperature was raised to $T = 0.8$. An earlier calculation,³ with constant volume (not constant pressure), gave essentially the same results. Since these times are orders of magnitude shorter than the measurements of Ref. 1, it is clear that homogeneous melting cannot explain the results of Ref. 1. Changes in the potentials are not expected to change this qualitative

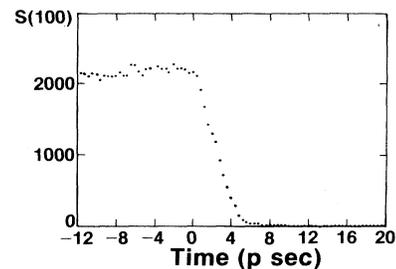


FIG. 1. X-ray intensity as a function of time for the (100) fcc peak. Note that this intensity decays much faster (~ 8 psec) than the measurements shown in Ref. 1, Fig. 3 (~ 10 nsec) at around the point marked as F_{melt} in that figure.

conclusion.

The absorption mechanism of laser radiation in a metal occurs mostly through the excitation of free or nearly free carriers.⁴ For melting to occur, energy has to be exchanged between the electron and phonon system through inelastic electron-phonon collisions. The inelastic electron collision time has been measured earlier to be $\tau_{\text{in}} \sim 10$ nsec and is thought to be mainly due to inelastic electron-phonon collisions.⁵ This time (τ_{in}) is quite close to the extrapolation of data in Fig. 3 of Ref. 1 to F_{melt} (referred to in Ref. 1 as being close to equilibrium). We suggest therefore that Fig. 3 of Ref. 1 is a measurement of the inelastic electron-phonon collision time and that the power dependence is due to nonlinear effects arising from the high-energy input from laser irradiation.

We would like to thank K. Meyer for calling our attention to Ref. 1 and for useful discussions. The authors express their appreciation for computing time made available by the U.S. Department of Energy on the ER-Cray. This work was supported by the U.S. Department of Energy.

A. Rahman and I. K. Schuller
Materials Science and Technology Division
Argonne National Laboratory
Argonne, Illinois 60439

(Received 1 August 1985)

PACS numbers: 64.70.Dv, 61.14.Fe, 61.14.Hg, 61.15.Cj

¹S. Williamson, G. Mourou, and J. C. M. Li, Phys. Rev. Lett. **52**, 2364 (1984).

²H. C. Andersen, J. Chem. Phys. **72**, 2384 (1980); M. Parrinello and A. Rahman, J. Appl. Phys. **52**, 7182 (1982).

³A. Rahman, unpublished.

⁴See, for instance, F. Abeles, *Optical Properties of Solids* (North-Holland, Amsterdam, 1972), Chap. 3.

⁵Ivan K. Schuller and K. E. Gray, Phys. Rev. Lett. **36**, 429 (1976), and Solid State Commun. **23**, 337 (1977).