Lattice Stability of Solids Under intense irradiation

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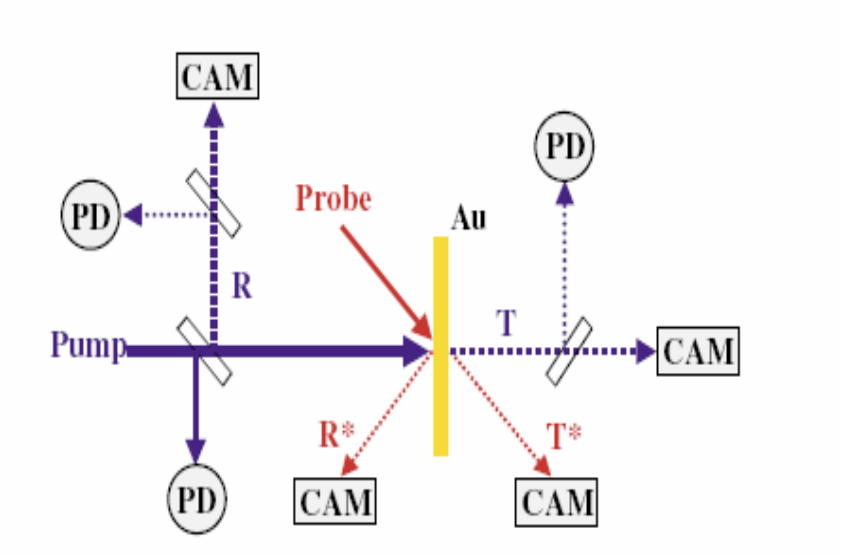
Femtosecond light sources

- Lasers
  - Operate in the visible (up to 400 nm), with a very short pulse (150 fs)
  - Pump laser 65 *microns* (FWHM) with a maximum irradiance of about 1013 W/cm2
  - Can heat a gold sample with thicknesses ranging from 280–320 A up to 6-8 eV (1ev=11000K)
  - Can also be utilized as probe in pump/probe experiments (measure e.g. reflectivity) with large beam diameter

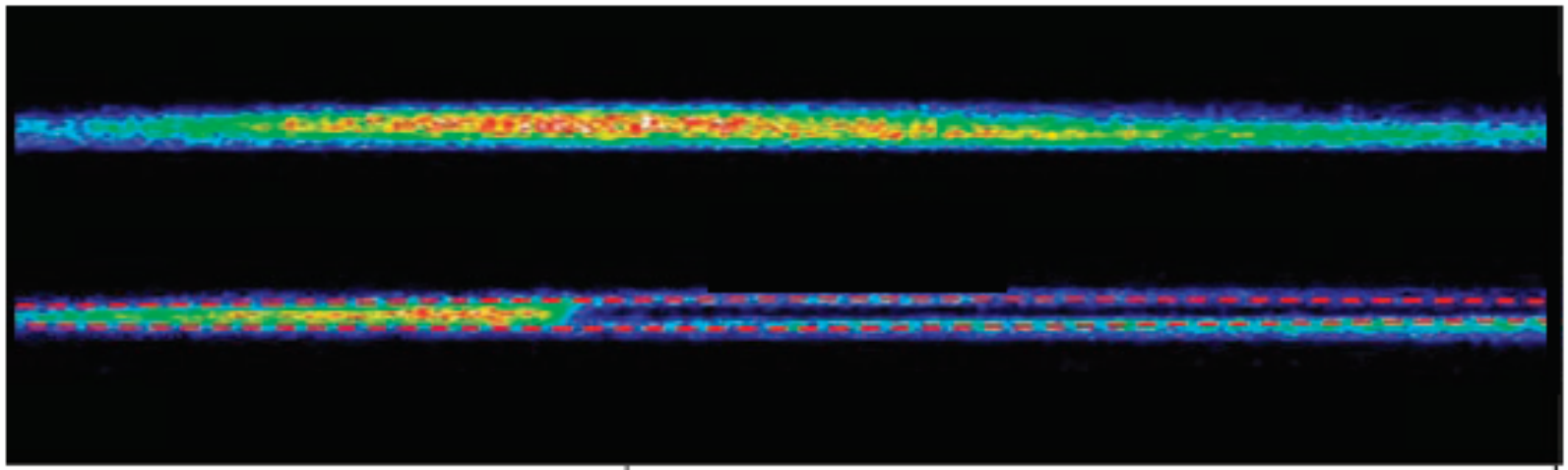
- X-rays pulse
  - As part of the effort to build free electrons lasers in the X-ray range (Stanford and Hamburg) in 2010
  - Intense bursts of X-rays in the 150fs range
  - Can be used as probe for real-time diffraction experiments

Schematic of the experimental setup (pump-probe)

- Free-standing gold foils with thicknesses ranging from 280–320 A
- Heated by a 400 nm, 150 fs (FWHM) pump laser 65 *microns* (FWHM) with a maximum irradiance of about 1013 W/cm2
- Reflectivity *R,* transmission *T* of the pump laser are monitored
- Optical property of the heated foil: collimated 800 nm, 150 fs probe laser with a beam diameter of 650 microns at 45 degrees
- K.Widmann, et al., PRL, 125002, 2004



Experimental set-up (X-rays) A.M. Lindenberg et al., Science, 2005



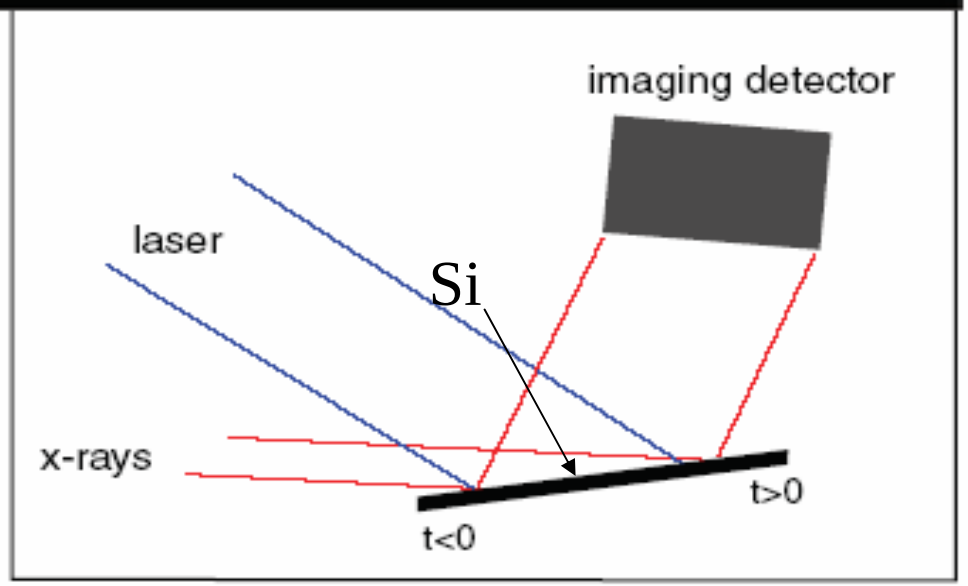


Fig. 1. (**Top**) A single-shot image of scattered x-rays from unperturbed sample above a single-shot image of perturbed sample. Dashed curves show region excited by laser pulse. (**Bottom**) Experimental setup showing cross-beam topography technique. By crossing the pump and probe beams on the sample and imaging the diffracted x-rays, we mapped temporal information into spatial information, enabling collection of the complete time history around time zero in a single shot. Time runs from left to right. The time window shown is ~8 ps.

Diffraction observation: decay of the (111) and (220) peaks starting immediately

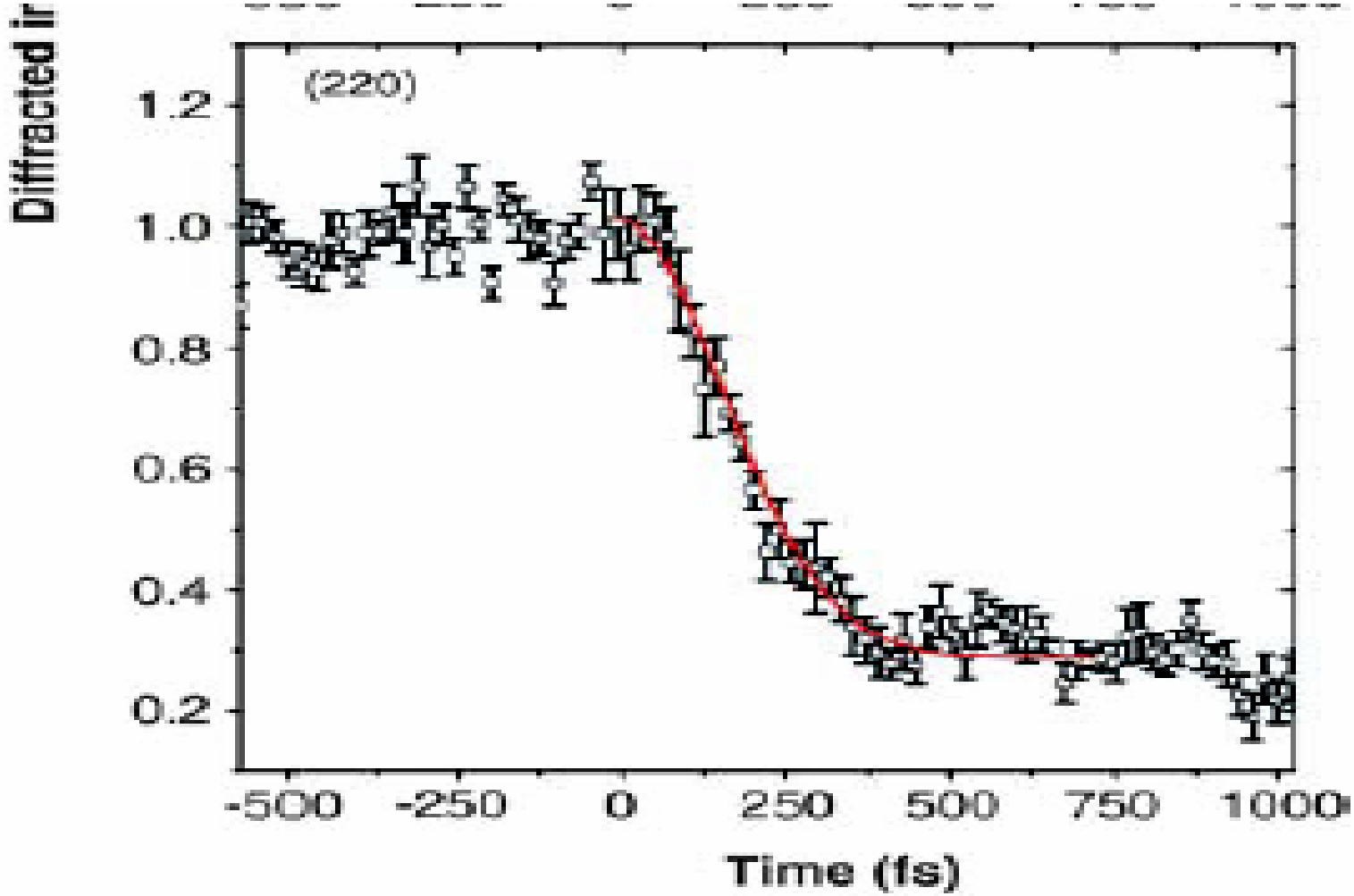


Fig. 2. Time-dependent diffracted intensity for (111) (top) and (220) (bottom) reflections.

Ab initio calculations

- The laser couples directly only to the electrons (optical frequency)
- The electron thermalization time is 10-100 fs
  - We suppose the electrons are at a given temperature
- The electron-phonon coupling term yields thermalization times in the 10 ps range
  - Just after irradiation, there is a small transfer of energy to the ions
- One can suppose that the ions couple through an electron gas at finite temperature
- Depending on the laser and the target the temperatures are estimated to be ~2eV (22 000K) for the insulators, and 6eV (66 000 K) for metals
- Therefore, use a simulation using a Fermi-Dirac smearing of the electrons .

Molecular dynamics for silicon (Silvestrelli et al., PRL, 1996)

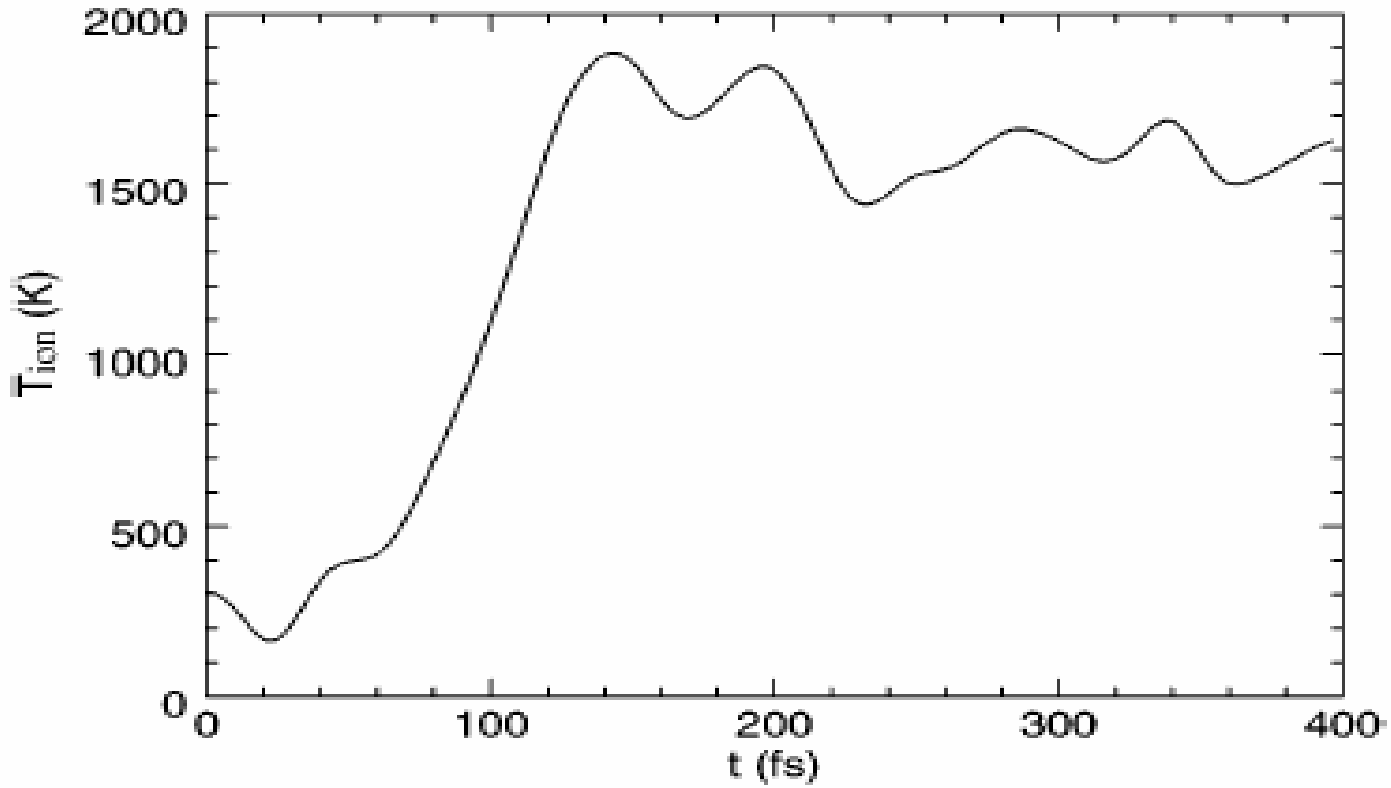


FIG. 1. Time dependence of the instantaneous ionic temperature, defined as $T_{\text{ion}}(t) = [M/(3N-3)k_B]\sum_{I=1}^{N} v_I^2(t)$, where k_B is the Boltzmann constant, M is the Si ion mass, and $v_I(t)$ the ionic velocity at time t.

Pair correlation function

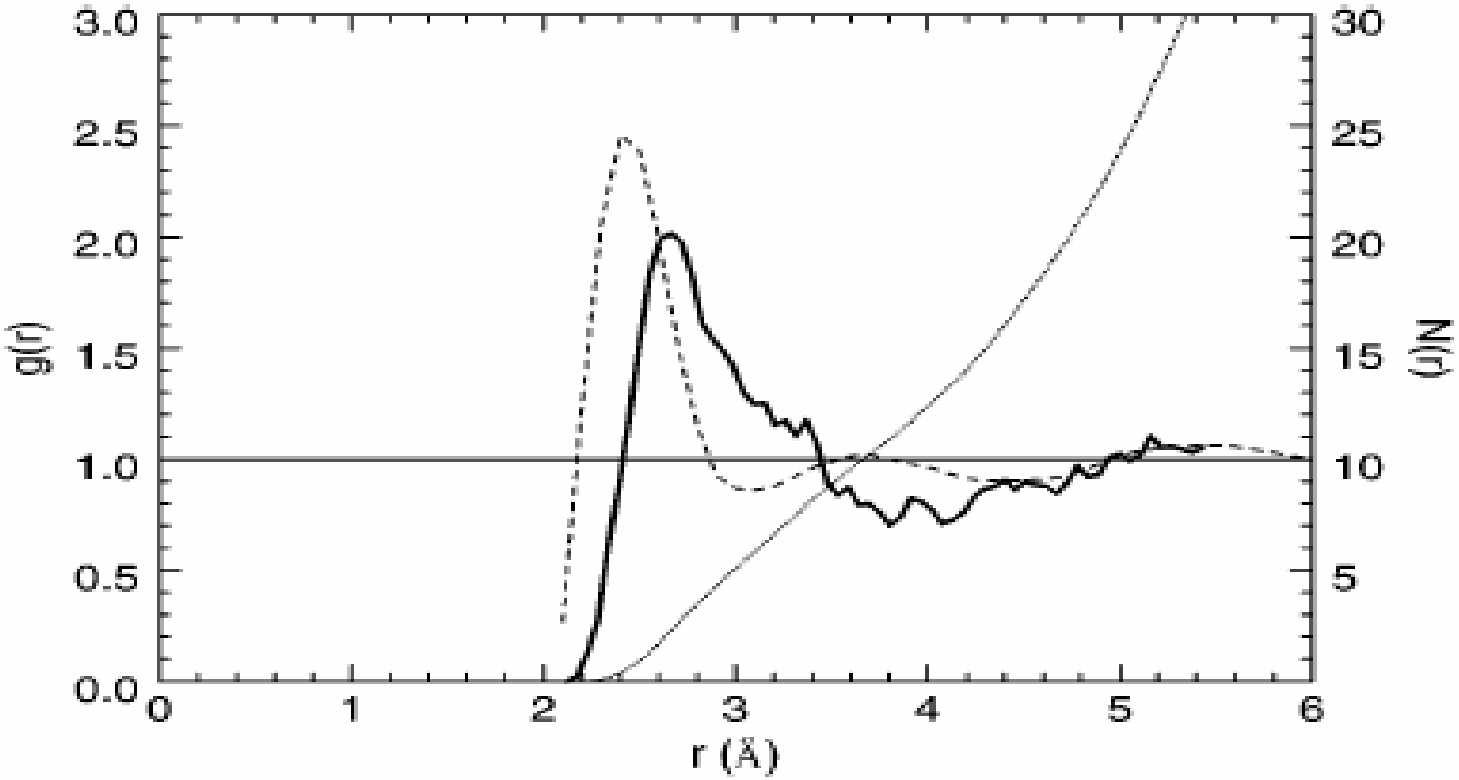


FIG. 3. Pair correlation function $g(r)$. Solid line: MD simulation. Dashed line: experimental result [19] for l-Si. Dotted line: coordination number $N(r)$.

Phonon spectrum of silicon (with Abinit)

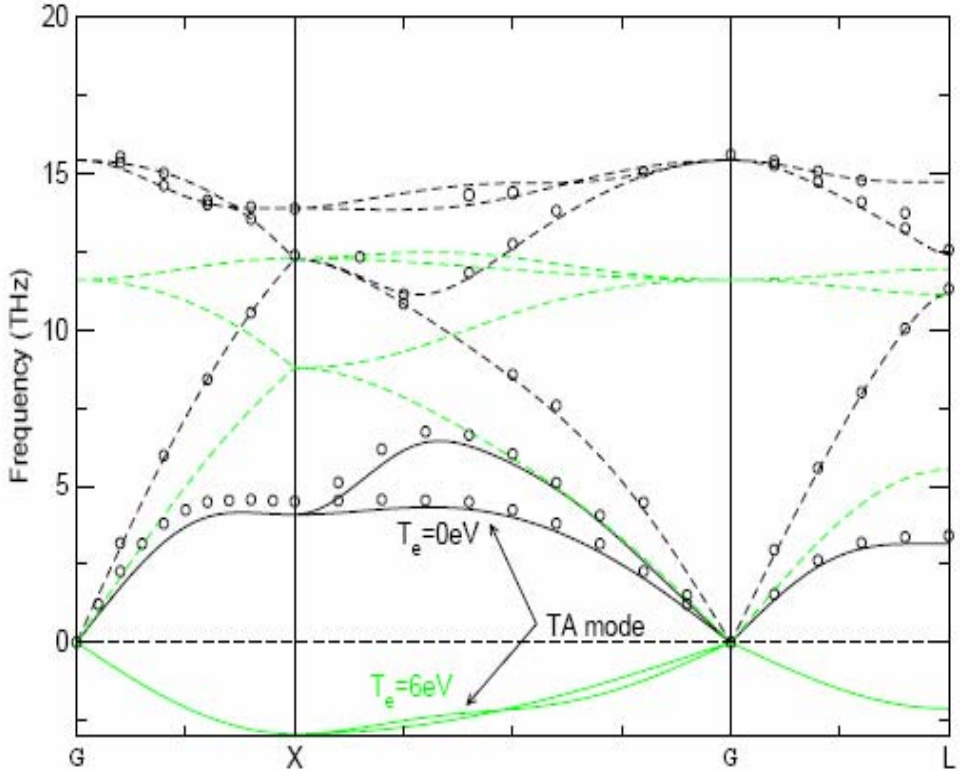


Figure 1: Phonon spectrum of Si at different electronic temperature. The black curves is the spectrum for $Te = 0$ eV. The green curves are for $Te = 2.15$ eV. Dots are experimental results from (*24*)

Analysis of the phonon spectrum

- The first “ab initio” calculation of the phonon spectrum of Si was done by R. Martin (1968)
- Use a (local) electron-ion pseudo potential, linearly screened by the dielectric function.
- And more simplifications:
  - Incomplete screening of ionic charge leads to the introduction of a “bond charge” always at the midpoint of two atoms
  - The dielectric function is very simplified (diagonal)
- Results:
  - If a metallic type complete screening is allowed, the TA modes are unstable
  - Taking a bond charge of 1/6 leads to a fair agreement with experimental data

The phonon spectrum of R. Martin

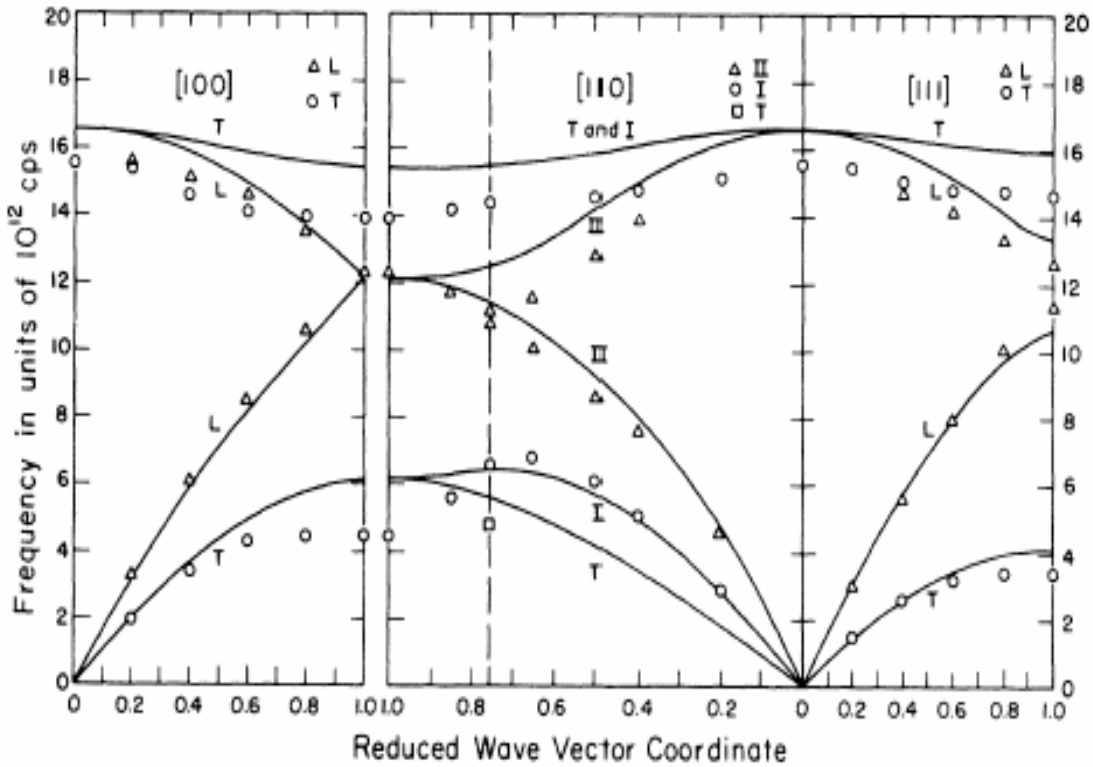


FIG. 2. Phonon dispersion curves for Si. The solid curves are the results of the present calculation and the points are the neutron-diffraction data of Dolling (Ref. 2) except for the single [110] TA point measured by Palevsky et al. (Ref. 2). The vertical dashed line at 0.75 in the [110] direction indicates the zone boundary.

Looking at old references again

- Biswas and Ambegaokar (1982) reconsidered the model, by modifying the dielectric function to include excited electrons and a subsequent increase of screening.

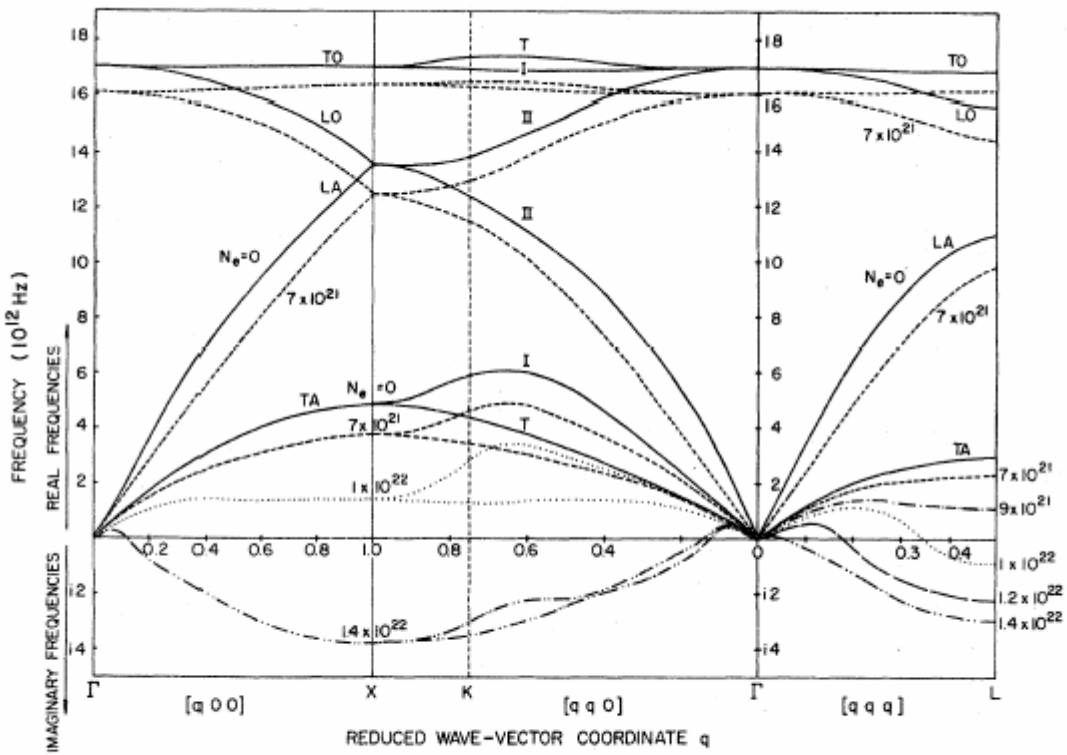


FIG. 3. Phonon spectrum at different excited electronic densities N_e. The solid curves are for the unexcited system and the dashed curves for the different excited densities indicated. The longitudinal and the optic modes for 1×10^{22} are very similar to those at 7×10^{21} and have been omitted.

Experimental results for Gold

- Measurement of the optical conductivity
- Observation of a very long steady state where ion motion seems negligible
- Ab initio calculation of the optical conductivity (using Kubo-Greenwood formula, as in the module conducti) and assuming a liquid state yields results which are far away from experiments (S. Mazevet et al.)
- Seems to be different from silicon

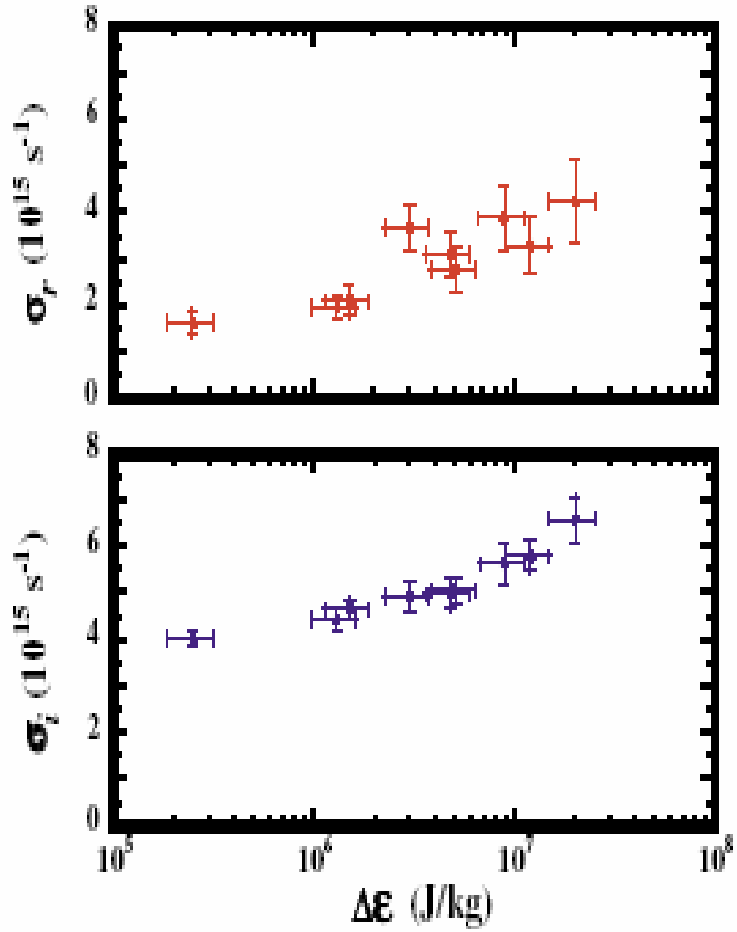


FIG. 3 (color online). The real and imaginary parts of the ac conductivity as a function of increase in energy density.

Phonon spectrum of gold

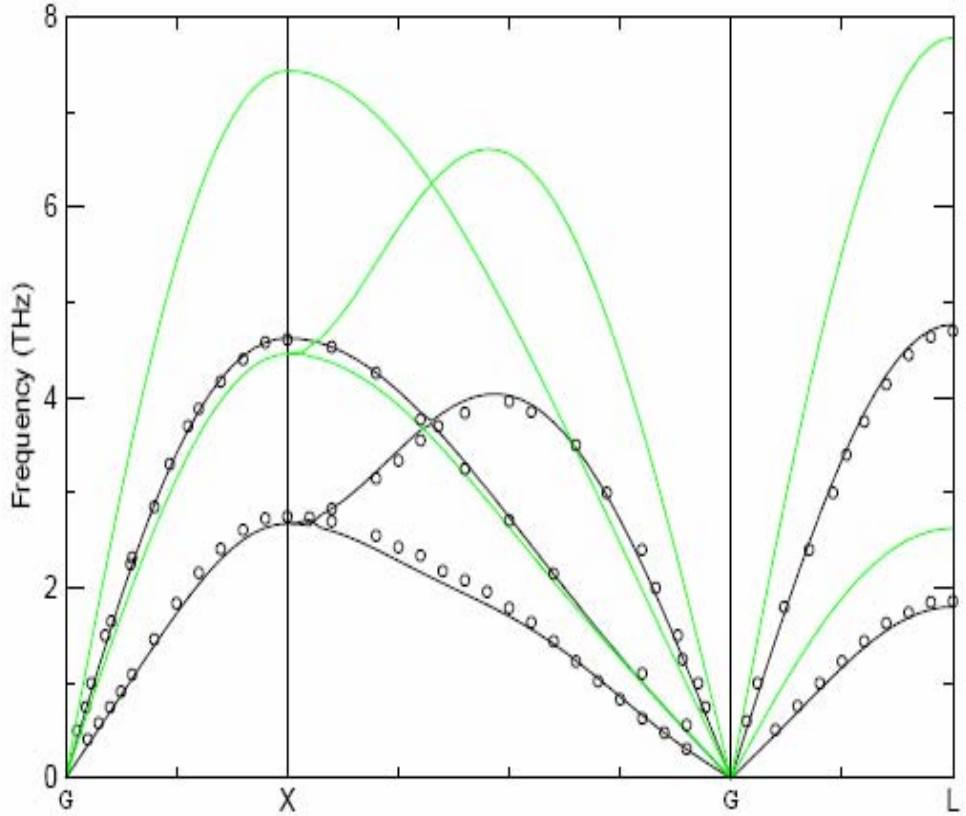


Figure 2: Phonon spectrum of Au at different electronic temperatures. The black curves are the spectrum for $Te = 1$ eV. The green curves are for $Te = 6$ eV. Dots are experimental results from (*25*).

Lindemann's criterion of melting

- Melting will occur if the mean square displacement is larger than some fraction of the lattice spacing (e.g. 0.2)

$$\sqrt{< x^2 >_{Tm}} \approx Aa$$

- The simple Debye model represents the phonon DOS by a single parameter, the Debye temperature.

- This parameter can be extracted from the heat capacity whose high temperature part is given by the Debye form: C(T)=3Mk D(T/thetaD)

- From Θ_D one can extract the mean square displacement, which is proportional to Θ_D^2
- Hypothese: $T_m(T)/T_m(0) \propto \Theta_D^2\ (T)/\Theta_D^2\ (0)$

Variation of the Debye temperature of gold

$$C_V(T_i) = 3nNk_B \int_0^{\omega_L} \left(\frac{\hbar\omega}{2k_BT_i}\right)^2 \mathrm{csch}^2\left(\frac{\hbar\omega}{2\mathrm{k_B T_i}}\right) \mathrm{g}(\omega)\mathrm{d}\omega$$

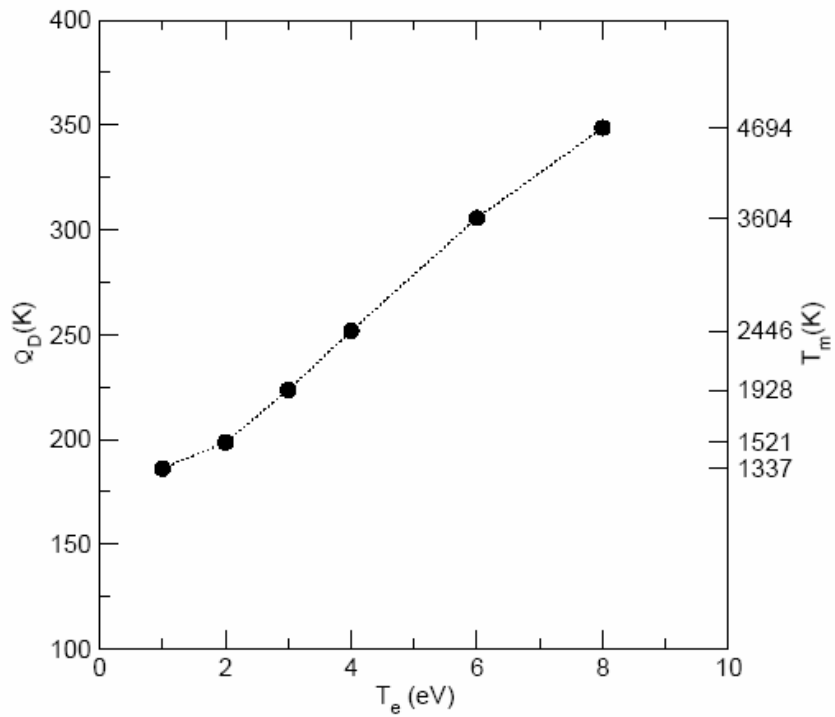


Figure 3: Debye temperature variation as a function of electronic temperature for gold. On the right hand side, corresponding melting temperature.

Comparison between gold and aluminum

- The phonon spectrum of Aluminum will be only slightly modified even up to 6eV
- The melting temperature, computed with the same technique will only increase of 10%
- The general form of interatomic potential for Al and Au are different.
- For Al, (complete) linear screening of a pseudo potential yields good values for the spectrum
- The electrons are quasi free, and dense: only slightly affected by temperature
- For Au, a noble metal, d electrons are localized and can get excited. Thight binding potential:

$$V(r) = Z_s^2 \cosh(\kappa r_c) \frac{\exp(-\kappa r)}{r} - Z_d(1 - \frac{Z_d}{10}) W_d (\frac{d}{r})^5$$

DOS of high temperature Aluminum and gold

- For aluminum, even at 6 eV, the density of states is not changed (this is a “degenerate” electron gas)
- For gold, the 3d states are somewhat localized and can get excited.

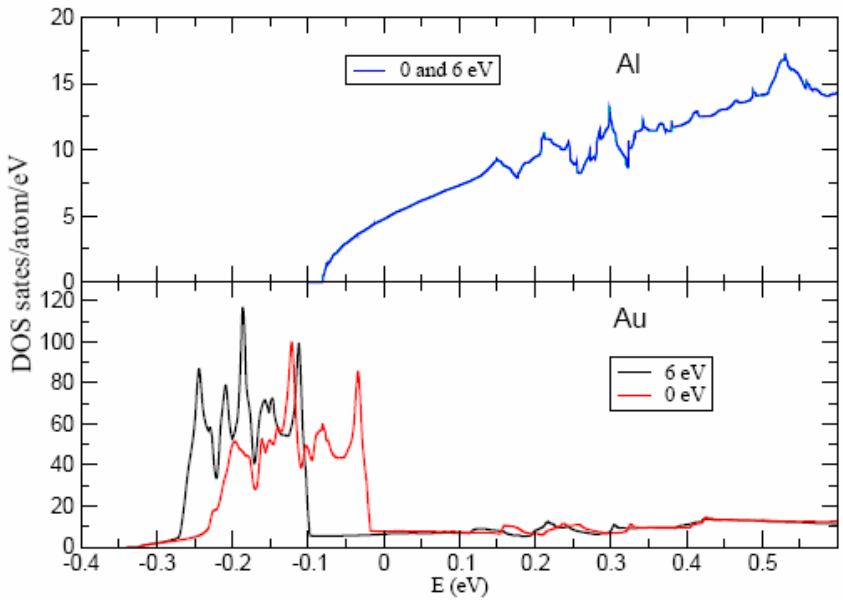


Figure 4: Density of state of aluminum and gold at 0K and 6eV.Whereas the DOS of Aluminum is practically unaffected, the DOS of Gold is shifted and shrink.

Conclusion

- Intense ultra fast laser irradiation changes the nature of chemical bonding of solids (and is used in many technological applications)
- As a general rule, semi-conductors and metals respond in a different way to these excitations.
- In many semi-conductors, a slight metallic character rapidly leads to a destabilization of the lattice
- In metals, a subdivision occurs whether localized electrons participate in the bonds.
- In e.g. Al, free electrons are not affected
- In Au, excitation of d electrons, increase the ion charge a decrease the band width leading to a strongly repulsive potential.
- A more systematic study is in order….