Dielectric Function of GaAs: a View into Electron and Lattice Dynamics

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Intense ultrafast optical excitation creates unique non-equilibrium conditions in a semiconductor, strongly modifies the bandstructure, and drives structural changes.[1-3] Our previous results have shown that measuring fundamental optical properties, such as the dielectric constant[1] and second-order susceptibility[2] is a tremendous improvement over ordinary reflectivity and second-harmonic measurements, and yields results qualitatively different from previous assumptions. We now move beyond single-frequency measurements using a new broadband spectroscopic technique to simultaneously monitor the response of the dielectric function $\varepsilon(\omega)$ of GaAs across the spectral range from 1.5 to 3.5 eV, with 100-fs temporal resolution.

The GaAs sample is driven by a 1.9-eV pump pulse and probed by a broadband probe pulse generated by strong self-phase modulation of a 70-fs 2.2-eV pulse in CaF_2. Reflectivities $R_1(\omega)$ and $R_2(\omega)$ are measured for p-polarized light at two incident angles, 60 and 76 degrees, chosen to give high sensitivity to changes in the dielectric function. The reflectivity is determined by measuring both the reflected probe spectrum and a reference spectrum for each shot using an imaging spectrometer. The real and imaginary parts of the dielectric function are then evaluated using an optimization algorithm which finds the $\varepsilon(\omega)$ which gives the experimental $R_1(\omega)$ and $R_2(\omega)$ using the Fresnel reflectivity formulae and a three layer model which takes account of an oxide layer of thickness 3.5 nm and ε=4 on the bulk GaAs sample. By varying the time delay between the pump and the probe, we observe the changes in the dielectric function in time as a result of the intense excitation. The broadband probe has a significant chirp, for which we compensate by temporally shifting the data based on a measured calibration of the dispersion.

The data in Fig. 1 show $\varepsilon(\omega)$ at 500 fs and 4 ps after excitation by a pulse of 40% of the threshold for observable permanent change. Also shown in Fig. 1a (solid and dashed curves) corresponding to the real and imaginary parts of the unperturbed $\varepsilon(\omega)$. The 500-fs data display a drop in the real part and a broadening in the imaginary part of the dielectric function which occur directly following the excitation. By 4 ps $\varepsilon(\omega)$ has changed considerably, and agrees closely with measurements on GaAs at elevated temperatures[4], shown at 400 °C (solid and dashed curves). While the initial effects are likely electronic in this fluence regime, by 4 ps the lattice has been heated by the electrons and this dominates the changes in $\varepsilon(\omega)$.

FIG 1. Dielectric function (● - Re[$\varepsilon(\omega)$], o - Im[$\varepsilon(\omega)$]) for a pump fluence of 0.40 F_{TH} after 500 fs and 4 ps. The lines drawn on the 500-fs plot show the real (solid line) and imaginary (dashed line) parts of the dielectric function for unpumped GaAs. The lines drawn of the 4-ps plot show the dielectric function for GaAs at 400 °C.[4]

At a higher fluence of 65% of the threshold, the changes in $\varepsilon(\omega)$ are much more pronounced, shown in Fig. 2 at 500 fs and 4 ps. At 500 fs the imaginary part of $\varepsilon(\omega)$ is very broadly spread across the entire spectral range, and the real part has decreased. By 4 ps, the dielectric function is quite similar to that of amorphous GaAs[5], shown by the curves, suggesting that the material has become disordered. It is interesting to note that this occurs below the threshold for permanent change, indicating that the amorphization is reversible.

Above the damage threshold, $\varepsilon(\omega)$ changes dramatically. Fig. 3 shows $\varepsilon(\omega)$ at 500 fs and 4 ps after an excitation of 160% of the threshold. Within 500 fs much of the oscillator strength has moved from the large E_2 peak (initially centered at 4.75 eV) down to our spectral range. The real part is negative above 2.7 eV because most of the oscillator strength is at lower frequencies. By 4 ps the real part is negative above 1.8 eV, and $\varepsilon(\omega)$ is metal-like above 2 eV. These changes are much more drastic than those due to disordering and indicate changes in local bonding.

FIG 2. Dielectric function (● - Re[$\varepsilon(\omega)$], o - Im[$\varepsilon(\omega)$]) for a pump fluence of 0.65 F_{TH} after 500-fs and 4-ps. The lines drawn on the 500-fs plot show the real (solid line) and imaginary (dashed line) parts of the dielectric function for unpumped GaAs. The lines drawn of the 4-ps plot show the dielectric function for amorphous GaAs.[5]

FIG 3. Dielectric function (● - Re[$\varepsilon(\omega)$], o - Im[$\varepsilon(\omega)$]) for a pump fluence of 1.60 F_{TH} after 500-fs and 4-ps. The lines drawn on the 500-fs plot show the real (solid line) and imaginary (dashed line) parts of the dielectric function for unpumped GaAs.

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