# Laser Heating of Free Electrons in Wide-Gap Optical Materials at 1064 nm

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Free-electron absorption of 1064-nm photons is measured photoacoustically in NaCl and $SiO_2$. The electrons are generated with a 266-nm pump pulse by two- or three-photon transitions. For a given pump pulse energy, the photoacoustic signal and the rise in lattice temperature are observed as a function of the energy of the 1064-nm pulse and found to be in agreement with the theory of free-carrier heating. Temperatures approaching the melting point are produced in these materials under prebreakdown conditions. No evidence of electron-impact ionization and ensuing avalanche formation is found up to the intrinsic damage threshold.

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Generation of free electrons and subsequent lattice heating via electron-photon-phonon interactions are the two major processes leading to laser-induced intrinsic damage in transparent optical materials.[1-4] We have shown recently that in KBr and NaCl, exposed to intense laser pulses at 532 nm, these two processes are free-electron formation by four-photon transitions across the band gap and free-electron photon absorption,[5,6] resulting in a significant rise of lattice temperature. Damage occurs when the peak temperature in the interaction volume approaches the melting point. The experiments provide conclusive proof that under these conditions impact ionization does not measurably contribute to free-carrier formation.

In this Letter we show, by observation of the electron-photon-phonon interaction and the resulting increase in lattice temperature, that even at 1064 nm electron avalanche formation is conspicuously absent up to the intrinsic damage threshold. By employing two laser pulses of different wavelengths, one in the ultraviolet and one in the near infrared, we efficiently generate free electrons with the former and heat them with the latter, thus separating these two processes for independent study. The experiments were also performed with the intention to verify the validity of the so-called multiphoton-assisted avalanche mechanism of laser damage.[7] In a low-order multiphoton process (here two- and three-photon absorption), a large free-electron density can be generated which would represent the starting density for avalanche formation during the interaction with a 1064-nm pulse.

To assure that indeed intrinsic effects are studied and not energy absorption due to material imperfections, it is essential to measure the energy deposited by the two laser pulses in the prebreakdown regime rather than monitoring only damage thresholds. The materials chosen in our investigation are NaCl and $SiO_2$ (fused silica). For electron pump pulses, we use the fourth harmonic of a Nd:YAlG (Nd-doped yttrium-aluminum-garnet) laser (266 nm) and its fundamental wavelength (1064 nm) to heat the electrons. The carrier generation process is two-photon absorption in NaCl and three-photon absorption in $SiO_2$. It is efficient and, therefore, does not require very high intensity to create a large density of electron-hole pairs ($10^{16}$ cm$^{-3}$) for the measurement of free-electron heating. Furthermore, absorption by free electrons is much less efficient at 266 nm than it is at 1064 nm.[8] Thus, as confirmed by our measurements, we can neglect the contribution of pump pulses to lattice heating. Similarly, we do not have to consider carrier generation by 1064-nm light because it would require valence electrons to simultaneously absorb at least seven photons, an unlikely process at the intensities used in our experiments.

The theory of free-electron absorption yields the following expression for the change of lattice temperature in the interaction volume:[8]

$$c\rho\frac{dT(\mathbf{r},t)}{dt}=AT^{1/2}(\mathbf{r},t)n_c(\mathbf{r},t)F^{3/2}(\mathbf{r},t)\,, \tag{1}$$

where $c$ is the specific heat, $\rho$ is the mass density, $A$ is a combination of known material parameters,[8] $F$ is the photon flux, and $n_c$ is the density of conduction electrons. From this equation, the total energy, $U$, absorbed from a single heating pulse is calculated by temporal and spatial integrations with $n_c(\mathbf{r},t)=n_0f(\mathbf{r})f_1(t)$ and $F(\mathbf{r},t)=F_0\times g(\mathbf{r})g_1(t)$:

$$U=c\rho\left(2T_0^{1/2}V_1Dn_0F_0^{3/2}+V_2D^2n_0^2F_0^3\right)\,, \tag{2}$$

where $V_1=\int_V f(\mathbf{r})g^{3/2}(\mathbf{r})dV$ and $V_2=\int_V f^2(\mathbf{r})g^3(\mathbf{r})dV$, $f(\mathbf{r})$ and $f_1(t)$ are the spatial and temporal distributions of the free-electron density, and $g(\mathbf{r})$ and $g_1(t)$ are those of the 1064-nm laser pulse. Neglecting free-electron diffusion,[6] $f(\mathbf{r})$ and $f_1(t)$ are directly related to the spatial and temporal shapes of the 266-nm pump pulse.

The total absorbed energy can be measured photoacoustically by the technique of Jones *et al.*[6] In a two-wavelength experiment, the photoacoustic signal is comprised of two parts: (a) a small contribution, $U_0$, of

 

the energy deposited via multiphoton carrier generation, and (b) the energy, $U$, of the 1064-nm light absorbed by free electrons. The signal is proportional to the sum of the two parts, i.e., $S=A_c(U_0+U)$. Under the condition of constant pump energy, $A_c$, $U_0$, and $D$ are constants that can be determined from a measurement of $S$ as a function of $F_0$. Having achieved this, we can then determine the peak temperature in the interaction volume for a given peak flux of the heating pulse. Note that for Eq. (2) to be valid, it is important to precisely determine the spatial and temporal characteristics of both pump and heating pulses and to assure their spatial and temporal coincidence in the interaction volume.

The Nd:YAlG laser system emits 100-ps pulses at 1064 nm which are converted to 48-ps 266-nm pulses by harmonic generators. In order to overlap the focal planes of both wavelengths with only one focusing lens, a beam compressor consisting of a positive and a negative lens is placed in the infrared-beam path. By varying the distance between the two lenses, we can change the effective focal length for the 1064-nm pulse independently. A movable prism is installed to vary the path length of the 1064-nm pulse for temporal overlap with the 266-nm pulse.

The detection system consists of a resonant PZT (lead zirconate titanate) transducer and a 5-in.-long fused silica rod. The transducer is glued onto one end of the rod, while the sample is attached to the other. This separation allows us to discriminate, via a time-of-flight difference, light scattered onto the transducer from the acoustic signal generated by the laser-solid interaction. The transducer is coupled to a pulse amplifier whose output is fed to a transient digitizer for analysis. The photoacoustic signal obtained is a modulated sine wave; its peak-to-peak amplitude is used as a measure for the absorbed energy. The NaCl crystals (ultrapure reactive atmosphere processed to reduce the $OH^-$ concentration) are obtained from the University of Utah, while the $SiO_2$ was purchased from Wale Apparatus, Co.

Precise alignment of both laser pulses turns out to be the major difficulty in the experiment because they are less than 100 ps long and are tightly focused with a typical focal waist of only 5 $\mu$m. The temporal overlap of the two pulses in the interaction volume is measured with a third-harmonic cross-correlation method, while the spatial alignment is ensured by optimizing the photoacoustic signal.

After proper alignment it is readily ascertained that both the pump and heating pulses do not individually produce a significant photoacoustic signal. Further, for constant heating pulse energy, by varying the power of the pump pulse the order of the multiphoton carrier-generation process is determined to be 2 for NaCl and 3 for $SiO_2$ at 266 nm.

The free-electron-photon interaction is studied as a function of heating pulse power under a constant pump condition. The results shown in Fig. 1 are in very good agreement with the predicted flux dependence of free-electron heating [Eq. (2)]. In these two measurements the free-electron densities are estimated to be on the order of $10^{16}$ cm$^{-3}$ using the known two- and three-photon absorption cross sections for wide-gap solids.[9] The estimate of the peak temperature (at $\mathbf{r}=0$) reached in the interaction volume is obtained from Eq. (1) after integration over time:

$$\Delta T(\mathbf{r}=0)=4T_0\left[\left(\frac{V_1}{V_2}\right)QE^{3/2}+\left(\frac{V_1}{V_2}\right)^2Q^2E^3\right]. \quad (3)$$

Here $E$ is the pulse energy and $Q$ is a constant determined from the fit of Eq. (2) with the experimental data in Fig. 1. Note Eq. (3) does not require knowledge of the free-electron density and pulse flux, provided the energy of the pump pulse remains constant and the measured photoacoustic signal depends on the flux of the heating pulse according to Eq. (2). Despite the complexity of the experiment, it is possible to show that the highest temperatures reached at the center of the interaction volume are above 1000 K in $SiO_2$ and well over 500 K in NaCl before damage. Further, Fig. 1 points to a complete absence of electron avalanche formation which would result in an explosive increase of the photoacoustic signal above a given photon flux of the heating pulse.

To ensure that our measurements do not pertain to absorption by laser-generated defects in the lattice,[6,10] we measured the photoacoustic signal as a function of time delay between pump and heating pulses under otherwise constant experimental conditions. With increasing delay, the signal decreases in accordance with the correla-

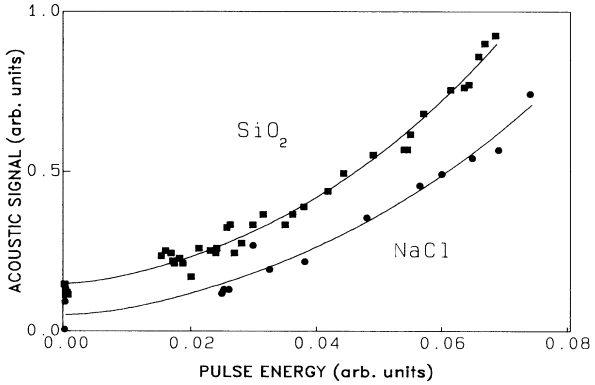


FIG. 1. Measured photoacoustic signal vs energy of 1064-nm pulses, obtained in NaCl (circles) and $SiO_2$ (squares) under the constant pump condition. It shows clearly a nonlinear dependence on the energy of the heating pulse. The solid line for each measurement is the fit of Eq. (2) with the experimental data, showing a good agreement between theory and measurement.

tion function of the two pulses in both materials, showing no evidence of photon absorption by long-lived ($\geq 150$ ps) defects. The identical dependence of the photoacoustic signals on the flux of the heating pulse in both samples is a further indication that defect absorption is negligible because one would expect it to behave differently in these two dissimilar materials.

In order to exclude multiphoton absorption processes resulting from a combination of photons at the two different wavelengths, we studied the polarization dependence of the photoacoustic signal for a fixed power of pump and heating pulses. No polarization dependence was detected, indicating that these processes can be neglected as well.

In conclusion, we have shown for the first time that the interaction of free electrons in NaCl and $SiO_2$ with photons in the near infrared (here at 1064 nm) is indeed governed by the free-electron heating mechanism proposed by Epifanov.[8] The estimated highest temperatures reached in our experiments are close to the melting points of the materials, confirming the earlier measurement in KBr by Shen *et al.*[5,10] We have further found no evidence for avalanche formation by electron-impact ionization at 1064 nm even under the condition of large density ($\approx 10^{16}$ cm$^{-3}$) of starting electrons. These results throw doubt on the prevailing opinion that avalanche formation is the mechanism of intrinsic damage in wide-gap materials at near-infrared wavelengths.

The two-wavelength approach reported here also provides a new tool for the investigation of the fundamental processes involved in laser-induced intrinsic damage of optical materials. It removes some of the difficulties associated with the previous single-wavelength approach.[5,6,10] Further application of this technique will permit the study of lattice heating as a function wavelength.

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