Intrinsic Optical Damage in KBr at 532 nm

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The nonlinear interaction of 532-nm, 100-psec laser pulses with KBr crystals is monitored up to damage by measurement of the σ component of the self-trapped exciton luminescence versus photon flux. The temperature rise in the interaction volume is obtained from the relation between σ luminescence and flux with use of the known temperature dependence of the luminous efficiency. The mechanism of energy deposition is four-photon free-carrier generation and free-carrier heating with other small contributions. Damage occurs at a temperature very close to the melting point.

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Evidence is mounting that the interaction of intrinsic wide-gap optical solids with intense photon fields in the visible region is not necessarily governed—as hitherto widely believed[1]—by electron-avalanche impact ionization. Earlier theoretical work by Epifanov, Manenkov, and Prokhorov[2] predicted that avalanche is less effective when the photon energy, $\hbar\omega$, is greater than $E_g/4$ (E_g is the band gap) and more effective when $\hbar\omega$ is less than $E_g/5$. Measurements by Manenkov[3] of the temperature dependence of laser-induced damage thresholds first indicated basic discrepancies with the electron-avalanche theory in NaCl, KCl, and KBr exposed to 8-nsec pulses at 532 nm. Recently, Jones *et al.*[4] presented proof that significant amounts of energy can be deposited into NaCl from an intense 80-psec pulse at the same wavelength via four-photon absorption, and the temperature rise at the focal point can be as high as several hundred degrees. However, prior to the results reported below, experimental difficulties prevented a clear observation of intrinsic damage for the case where free-carrier generation is shown to be a high-order multiphoton process.

We exploit the temperature dependence of self-trapped exciton luminescence to determine the temperature rise in the interaction volume during the exposure of a reactive atmosphere-processed (to reduce OH^- contamination) ultrapure single crystal of KBr to intense 532-nm laser pulses. In these experiments the σ-luminescence band of self-trapped excitons (the self-trapped hole—or V_k center—plus captured electron resulting from interband excitation) is measured with a calibrated optical detection system[5] as a function of peak photon flux of the 100-ps (half-width at $1/e$ intensity) pulse. The resulting spatially and temporally integrated σ-luminescence signal, $L^{(1)}$, monitored with an optical multichannel analyzer, is plotted, initially in arbitrary units, as a function of laser peak flux F (Fig. 1). The laser-beam parameters are carefully measured (see Ref. 5) in order to determine precisely the flux distribution in the interaction volume. The results show that, below $F=2.2\times10^{29}$ photons/cm^2 sec, the slope of the $L^{(1)}$ vs F curve is nearly 4 on a log-log scale and the interaction of the photon field with the material does not result in a

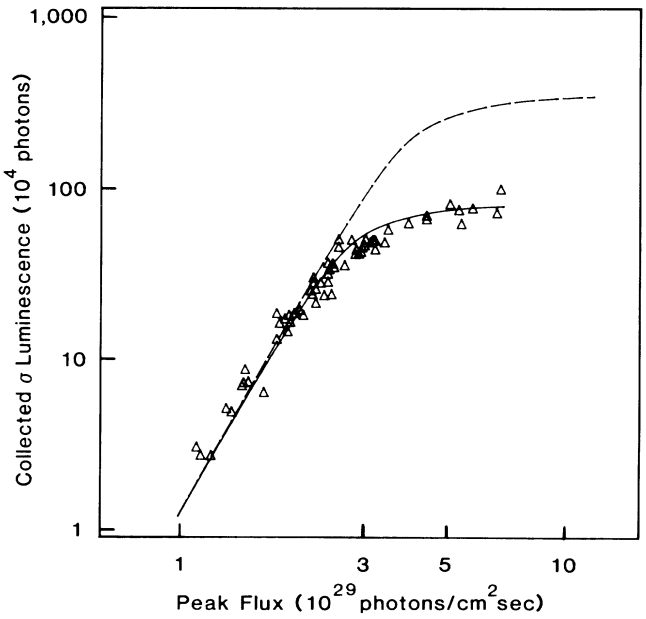

FIG. 1. Calculated (solid and dashed lines) and measured (triangle) spatially and temporally integrated emission from KBr as a function of the peak photon flux in 532-nm laser pulses of 100-psec duration ($1/e$ intensity half-width). The solid line is obtained with use of the free-electron heating model, while the dashed line is based on the polaron heating model. This luminescence is induced by four-photon exciton generation with subsequent radiative recombination of electrons and V_k centers.

discernible temperature increase. In this flux region the four-photon absorption cross section was found to be $\sigma^{(4)}=2\times10^{-112}$ cm^8 sec^3 (Ref. 5). The increase of the photon flux causes the local temperature, T, to rise, and the slope of the curve decreases according to the σ-luminescence efficiency, $\eta(T)$:

$$\eta(T)=[1+A\exp(-E_a/kT)]^{-1}. \quad (1)$$

Here $A=3.8\times10^9$ and $E_a=0.124$ eV.[6,7]

Analysis of the measured data presented in Fig. 1 shows that the temperature rise at the center of the interaction volume can be as high as 868° (from the initial value of 50 K to 918 K at the highest flux, $F=6.3\times10^{29}$ photons/cm^2 sec. Even at this flux, no catastrophic damage of the material can be detected. This is remarkable for four reasons:

(a) Electron-avalanche formation does not contribute significantly to energy deposition up to this point, since a considerable avalanche should lead to damage (discussed below).

(b) The dependence of the σ-luminescence yield on photon flux in Fig. 1, which can be explained by the microscopic processes in KBr exposed to 532-nm laser pulses,[5] reveals the intrinsic nature of the higher-order nonlinear photon-material interaction and laser-induced damage for this case.

(c) The heat mechanism required to explain the dependence at higher fluxes is the free-electron heating proposed by Epifanov[4,8] and not polaron heating.[4,9]

(d) The highest temperature reached, which is only 90 K below the melting point at normal pressure, demonstrates for the first time that melting is indeed directly indicated as the failure mode at least for the small effective interaction volumina used in our experiments (typically 5×10^{-8} cm^3).

The calculations of the flux dependence of the total luminescence emission are based on the known four-photon absorption cross section, photon collection efficiency of the detection system, and the rate equations for the interaction of the photon field with the material.[5] Now, however, the temperature dependence of the σ-luminescence efficiency and the heating rates cannot be neglected. They are crucial for analysis of the measurements. The spatial distributions of time-integrated luminescence emission and temperature must be calculated for each pulse. To achieve this, the mechanism of lattice heating must be known. As we have shown before[4] in NaCl exposed to intense 80-psec pulses at 532 nm, polaron or the free-electron heating proposed by Epifanov may occur with small contributions from absorption by laser-generated primary defects. Yet, prior to the present work it has not been possible to experimentally prove in any wide-gap optical material which of the two mechanisms actually prevails at visible wavelengths. It turns out that the σ-luminescence yield dependence on incident flux permits discrimination between the two possible heating processes. In the calculation, polaron heating (see Ref. 4) is given by a temperature-dependent absorption cross section,[10]

$$\sigma_p\approx(4\pi e^2/nc_l m\omega_0)(2\alpha/3)\times(\omega_0/\omega)^{5/2}\{1+[2/\exp(\hbar\omega_0/kT)-1]\}. \quad (2)$$

According to Epifanov,[8,11] free-electron heating can be expressed as

$$c(dT/dt)=1.09(mkT/2\pi)^{1/2}(n_c/l_{ac}v_s)\times(eE/m_\omega)^3. \quad (3)$$

The symbols in Eqs. (2) and (3) are $\omega_0=3.13\times10^{13}$ sec^{-1} (optical-phonon frequency), $m=0.388m_e$ (band mass), $\alpha=3.16$ (polaron coupling constant), ω the laser frequency, n the refractive index, c_l the speed of light, T the lattice temperature, $c(T)$ the Debye specific heat calculated with a Debye temperature of 173 K, $l_{ac}=3.0\times10^{-7}$ cm (mean free path of conduction electrons), $v_s=3.15\times10^5$ cm/sec (sound velocity), n_c the conduction-electron density, E the peak electric-field strength, e the electron charge, and k the Boltzmann constant.

We do not consider beam deformation caused by self-focusing or defocusing, or free-carrier and thermal diffusion. Recent investigations of beam deformation in alkali halides[4,5,12] show that for a tightly focused laser beam (less than 10 μm in radius) it is a much smaller effect than previously believed[13] at photon fluxes up to the damage threshold. This is confirmed by our observation of an undisturbed far-field beam pattern emerging from the sample. In addition, the calculations show that the free-carrier density is only 1.9×10^{18} cm^{-3} at the highest flux used in these experiments. This is considerably less than that required to cause a significant decrease in the refractive index.[9] Simple calculations[6,11] show that free carrier and thermal diffusion are negligible for a laser pulse of 100 psec.

The calculations described above provide the temperature distribution immediately after the laser pulse has passed through the interaction volume as well as the time-integrated luminescence intensity distribution as a function of laser peak flux for the two mechanisms of charger-carrier energy absorption. Examples of temperature and luminescence intensity distributions are shown in Fig. 2. The temperature distribution is approximately the same as the fourth power of the energy density of a Gaussian beam (because the absorbed energy is approximately proportional to the incident intensity to the fourth power) and the luminescence distribution is that of the free-carrier density modulated by thermal quenching according to Eq. (1).

Comparison with the measured spatially integrated luminescence yields, $L^{(1)}$, shows that only free-electron heating (with up to 15% contribution by V_k-center absorption[4-6,11] to the total energy deposited in a single

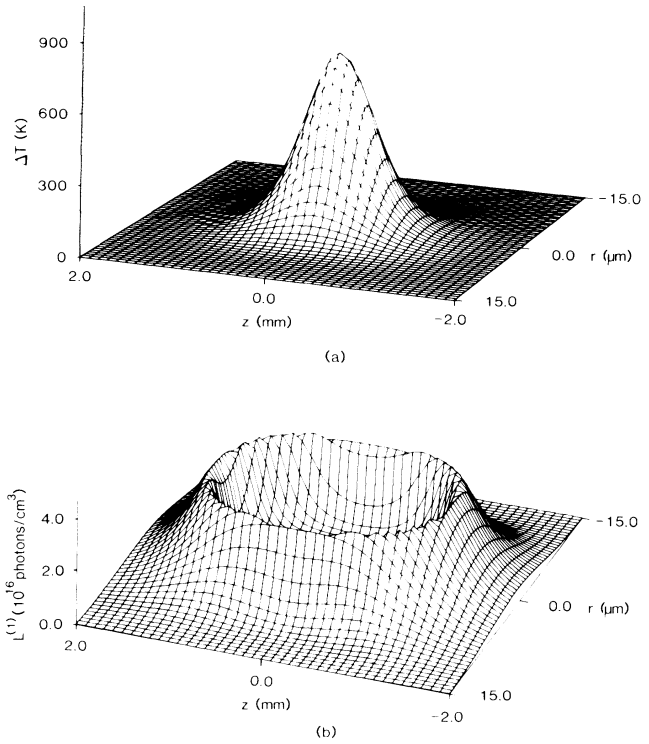

FIG. 2. (a) Temperature distribution and (b) spatially resolved time-integrated σ-luminescence emission generated in KBr by a single 532-nm pulse of peak flux 6.3×10^{29} photons/cm^2 sec. The beam axis is the z axis and r is the radius in cylindrical coordinates from the optical axis. The geometrical focal point is $(r,z)=(0,0)$.

laser pulse) can explain the experimental data (see Fig. 1). Polaron heating is not efficient enough to account for the onset of thermal quenching of $L^{(1)}$ at 2.2×10^{29} photons/cm^2 sec, even if the σ_p obtained from Eq. (2) is increased by two orders of magnitude. This finding is of considerable interest because it provides an answer to one of the main open questions in laser-induced damage: The mechanism of energy deposition from the laser photon field at visible wavelengths to the wide-gap insulator is indeed free-electron heating.

We further investigated any possible contributions to energy deposition by avalanche formation concurrent with four-photon absorption. If dominant, it would have resulted in an explosive increase in temperature[4] accompanied by catastrophic damage at a well-defined threshold flux, an event not observed up to $F=6.3\times10^{29}$ photons/cm^2 sec. We calculated the heating contribution due to avalanche formation using the expression for the ionization coefficient,[14] $\gamma=(aF)^5T^4$, and Eq. (3). Here a is a characteristic material parameter estimated to be between 10^{-30} and 10^{-28} cm^2 (sec/K)$^{4/5}$. To arrive at the maximum contribution that might be considered consistent with our experimental data, we choose $a=1.35\times10^{-30}$ cm^2 (sec/K)$^{4/5}$ such that sufficient additional energy is deposited to reach melting (an additional 89°) at 6.3×10^{29} photons/cm^2 sec. Any larger value for a would cause avalanche breakdown at lower fluxes, in disagreement with our measurements.

We conclude, therefore, that avalanche ionization is not dominant and, in fact, plays only a minor role in energy deposition up to the highest flux for which we did not yet observe damage but which in the absence of an electron avalanche corresponds to a maximum temperature of 918 K. However, we hasten to state that our experiments do not rule out avalanche formation at higher fluxes.

Note that the calculations of the temperature distribution and, for that matter, the maximum temperature reached in the center of the interaction volume are obtainable from the collected luminescence directly without reference to the magnitude of the flux or a model. They are obtained from the initial temperature (50 K), photon flux distribution, the known temperature dependence of the luminous efficiency and the calibrated measurement of the collected luminescence without the need for detailed knowledge of the mechanism of energy deposition. Only knowledge of the order of the primary multiphoton process, here four-photon electron-hole pair generation, is required. In that sense, monitoring the flux dependence of the σ-luminescent intensity up to the damage threshold is nothing less than a reliable means for temperature measurement in laser-induced damage. The damage criterion invoked is the onset of irreversible broadening of the σ-luminescence emission band. At that level, damage is not yet detectable by visual inspection with crossed polarizers or the occurrence of a hot spark. All data presented above pertain to prebreakdown events.

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