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Phonon generation by carrier recombination in a-Si:H

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Abstract

We present results of pump-probe anti-Stokes Raman experiments at 1.8 K on the generation of nonequilibrium phonons in a-Si:H following intense optical excitation. We obtained a decay time of ~ 70 ns for the TO phonons in all samples studied. In plasma deposited samples we observed an additional slowly decaying ($\geqslant 100$ ns) contribution to the Raman signal, which was not seen in hot-wire deposited samples. We propose a model to explain the additional phonons as resulting from fast nonradiative recombination of free carriers with carriers localized in the tail states.

Keywords: Raman scattering; Amorphous silicon; Nonradiative recombination

During the last few decades, recombination processes of photoexcited carriers in hydrogenated amorphous silicon (a-Si:H) have been extensively studied [1,2]. It is well established that at high optical densities, fast nonradiative recombination (FNR) dominates the carrier dynamics [3]. To explain this, several models of FNR were proposed: Auger recombination [4], distant-pair recombination [5], and spin-dependent and excitonic recombination [6]. However, the role that carriers localized in the tail states play in the FNR process is still not clear. In this paper we present results of pulsed Raman experiments on the generation of nonequilibrium phonons in a-Si:H following intense optical excitation. These phonons are the product of fast (< 1 ps) carrier relaxation and FNR.

We used two 1.0 μm thick a-Si:H films, one grown by plasma enhanced CVD (PE) and one by hot-wire assisted CVD (HW), both on c-Si substrates. The samples were immersed in superfluid helium (1.8 K). Interband excitation was produced by the 10-ns pulses of two synchronized frequency-doubled Nd:YAG lasers with average intensity (P) in the focus of 0.5 W cm^{-2}. The pulses of one laser (probe) had been electronically delayed with respect to those of the other (pump). By changing the delay Δt between pump and probe, and monitoring the anti-Stokes intensity, dynamics of nonequilibrium phonons were investigated in the wide range of 10 ns up to 15 ms.

In Fig. 1 we present the time-integrated anti-Stokes Raman spectra for both samples, each for $\Delta t = 100$ ns and 15 ms. All spectra show the familiar broad lines centered at 300 and 480 cm^{-1}, typical for LA- and TO-like vibrations, respectively. The most striking feature of Fig. 1 is that in the PE sample $I_{AS}(\omega)$ significantly drops going from

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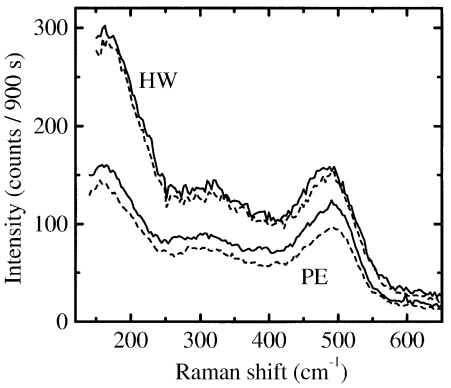

Fig. 1. Measured anti-Stokes spectra of the HW and PE sample for $\Delta t = 100$ ns (solid lines) and 15 ms (dashed lines).

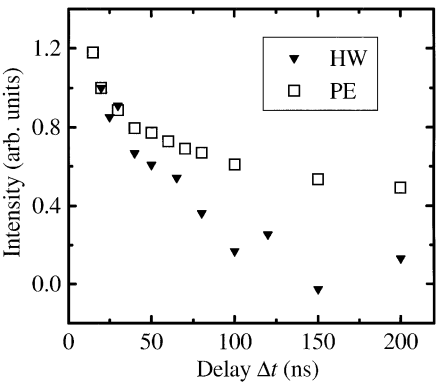

Fig. 2. Normalized anti-Stokes signal of the TO phonons as a function of delay Δt in the HW and PE sample.

$\Delta t = 100$ ns to 15 ms, whereas $I_{AS}(\omega)$ remains nearly the same in the HW sample. To show this to better advantage, in Fig. 2 the normalized anti-Stokes signal of the TO phonons is plotted as a function of Δt for both samples. To focus the analysis on the time-dependent contribution of $I_{AS}(\omega)$, we substracted $I_{AS}(\omega)$ measured at $\Delta t = 15$ ms. In both films a relatively fast decay time $\tau \sim 70$ ns is observed for the TO phonons. This fast decay is in quantitative agreement with earlier results of Scholten et al. [7] that were interpreted as phonon lifetimes. The new observation is that in the PE sample, in contrast to the HW sample, $I_{AS}(\omega)$ does not decay to zero, but instead to a slowly ($\gg 100$ ns) decaying background. This slow contribution is also observed for the TA and LA modes.

The slow background cannot be accounted for by bare phonon-dynamics. We therefore search for a clue by considering FNR processes in photoexcited a-Si:H that involve tail states. The tail states become populated following the fast relaxation of hot carriers under the emission of phonons. In the absence of optical pumping, the lifetime of carriers in the tail states is known to be as long as ~ 1 ms. We propose a model in which the carriers that populate the tail states during the pump pulse cannot escape nor decay by recombination until the probe pulse arrives. Then, in the presence of the probe pulse, the abundance of free carriers gives rise to *additional* phonon generation as FNR becomes possible. To investigate this, we resort to a simple rate-equation model that connects the concentrations of free carriers (N_f), carriers trapped in the tail states (N_t) and the phonons ($N(\omega)$). For simplicity, both the relaxation rate of free carriers to the tail states, w, and the bimolecular FNR rate, C, are taken equal for electrons and holes. The kinetic equations then read

$$\frac{dN_f}{dt} = g(t) - wN_f - CN_f(N_f + N_t), \tag{1a}$$

$$\frac{dN_t}{dt} = wN_f - CN_fN_t. \tag{1b}$$

The term $g(t)$ is proportional to P and describes the generation of free carriers by pulsed optical excitation. The terms wN_f relate to the trapping of free carriers at the tail states. The terms involving C correspond to the FNR of free carriers with free and trapped carries. The kinetic equation for the concentration of phonons is

$$\frac{dN(\omega,t)}{dt} = G(t) - \frac{N(\omega,t)}{\tau}. \tag{2}$$

Here τ is the phonon lifetime and $G(t)$ the total phonon generation rate, proportional to a weighted sum of free-carrier relaxation to the tail states, wN_f, and the carrier recombination, $CN_t(N_t + N_f)$. The Raman intensity is taken equal to the time average of the product of the laser intensity and $N(\omega,t)$ as obtained from solving Eqs. (1a), (1b) and (2). With the above ingredients, we

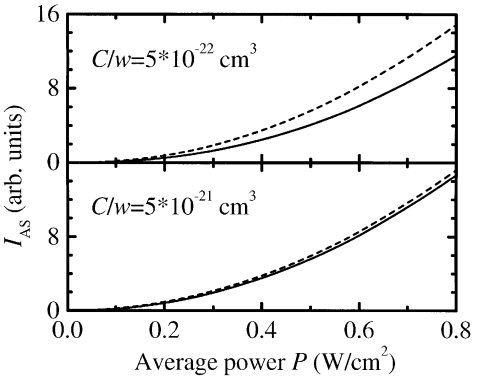

Fig. 3. Calculated anti-Stokes Raman intensity of the TO phonons for pump and probe pulses (solid and dashed curves respectively) as a function of power P, for $C/w = 5 \times 10^{-22}\,\mathrm{cm}^3$ (upper two curves) and $C/w = 5 \times 10^{-21}\,\mathrm{cm}^3$ (lower two curves).

calculated the anti-Stokes Raman intensity of the TO phonons for the pulses of the two lasers as a function of P. The solid curves in Fig. 3 give the result for the first laser (pump). The dashed curves represent the Raman signal of the second laser (probe) with the delay Δt choosen such, that the population of nonequilibrium phonons generated by the pump has decayed. (The signals we measured for $\Delta t \geqslant 100\,\mathrm{ns}$ equal the sum of the two contributions.) The solutions are shown for two values of C/w. It can be seen that for $C/w = 5 \times 10^{-22}\,\mathrm{cm}^3$ (upper two curves) the signal of the probe exceeds that of the pump by $\sim 30\%$ for $P = 0.5\,\mathrm{W/cm}^{-2}$. This corresponds to the situation in the PE sample. For $C/w = 5 \times 10^{-21}\,\mathrm{cm}^3$ (lower two curves) and the same P, this effect is virtually absent, as was found in the HW sample. Closer examination of the solutions shows that the additional phonon generation is only significant in a relatively narrow range of P, depending on the parameter $P \cdot C/w$. Apparently, in our PE sample this range of P coincides with the range used in the Raman experiments, which is not the case for the HW sample. Despite the roughness of our analysis, we conclude that anti-Stokes Raman scattering experiments can give new insight into the relative speeds of nonradiative hot carrier relaxation and trap-mediated hot carrier recombination in amorphous semiconductors.

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