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Two-color pump–probe experiments on silicon inverse opals

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We present time-resolved pump–probe experiments in a transmission geometry using off-resonant excitation on very high-quality silicon inverse opals. We show that the nonlinear optical response can drastically be modified by tempering of the sample. The as-grown samples are dominated by an absorptive response with recovery times as short as one picosecond. For the tempered samples, both the relaxation and the scattering times increase, leading to a prominent dispersive response. The data reveal a transient blueshift of the fundamental stop band on the order of 150 nm with transmission changes as large as a factor of five. Based on simple calculations using the Drude model we estimate corresponding refractive index changes as large as $\Delta n = -0.5 + i0.07$.

1 Introduction

One of the visions in the field of three-dimensional photonic bandgap materials is to incorporate nonlinear optical switches into highly integrated photonic circuits. This requires a complete three-dimensional bandgap, i.e. a large refractive-index contrast. Therefore silicon-based structures are of particular interest. Here, we investigate silicon inverse opals [1] which can serve as a model system.

For photonic bandgap materials in general, theory has predicted interesting ultrafast all-optical switching devices, e.g. based on the optical Kerr effect [2]. The latter results from the real part of the third-order nonlinear optical susceptibility, the corresponding imaginary part is usually not considered. In the simplest case, the photonic band edge provides a sharp spectral feature whose position is shifted by a nonlinear optical index change. Corresponding ultrafast experiments with silicon-based photonic crystals and using the Kerr effect are very sparse [3, 4] and could reach transmission changes only below 1%. By free carrier generation much higher reflection changes can be achieved [5], but on cost of very long relaxation times. In this paper and in Ref. [6], we perform femtosecond two-color pump–probe experiments on high-quality silicon inverse opals in transmission geometry.

The linear transmission spectrum of one of our samples [7], consisting of 6 layers, is shown in Fig. 1. Note that the fundamental photonic stop band is well developed. The inset shows an SEM image of the sample surface. The silicon grown by CVD is known to be amorphous. Subsequent tempering for one

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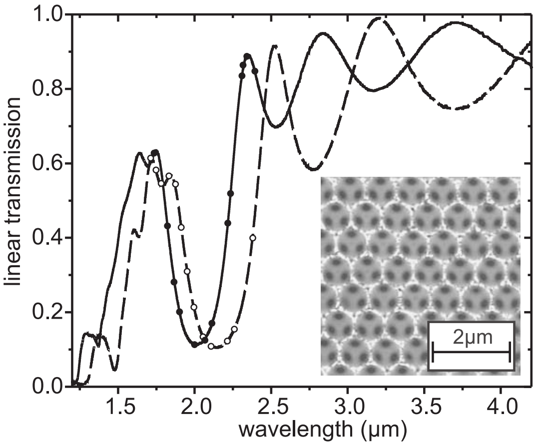

Fig. 1 Linear transmission spectra of the Silicon inverse opal in [111] direction, as-grown (dashed) and tempered (solid). The dots correspond to the probe wavelengths investigated in Fig. 3. The inset shows an SEM image of the sample surface.

day at 600 °C can bring it into a polycrystalline state. This reduces the silicon refractive index from 3.95 to 3.48, hence shifting the stop band towards the blue.

To confirm the assumption that tempering converts the silicon into the polycrystalline state, we have performed X-ray diffraction measurements on a 1 µm thick silicon film, also grown by CVD. Curve i) in Fig. 2 shows the angle resolved diffraction pattern of an untempered sample. No diffraction peaks are observable which indicates that the sample is amorphous. Curves ii) and iii) show the results for films that were tempered at 600 °C for 7 and 22 hours, respectively. Strong diffraction peaks arise as it is expected for polycrystalline silicon.

In our pump–probe experiments, we use two optical parametric amplifiers (OPAs) that can be tuned independently from 1.2 µm to 2.4 µm, both pumped by a regeneratively amplified Ti:sapphire laser system (Spectra-Physics Hurricane). For more experimental details see Ref. [6].

The LHS column of Fig. 3 shows a series of time traces for the as-grown sample (i.e., untempered) for fixed pump wavelength, fixed pump intensity, and varying probe wavelength as indicated. "Relative transmission" refers to the transmission with respect to the linear optical case. The RHS column shows corresponding results after tempering. Obviously, the nonlinear responses are completely different. For the untempered sample (LHS), we find induced absorption for all wavelengths and a recovery time of just one picosecond. In contrast, after tempering (RHS), the recovery times increase to about 5 ps and a shift of the fundamental stop band is observed: On the long-wavelength (short-wavelength) side of the stop band, we find increased (reduced) transmission – as expected for a reduction of the silicon refractive index after excitation with the pump. Such index reduction is expected from free carriers, which are generated via two-photon absorption. Indeed, in additional experiments, we find that the transmission change scales quadratically with the pump fluence (not shown).

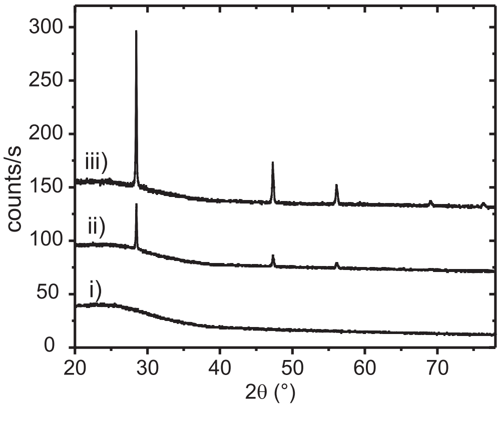

Fig. 2 X-ray diffraction measurements of a 1 µm thick silicon film on glass substrate, vertically displaced by 60 counts/s. i): untempered; ii): tempered for 7 hours at 600 °C; iii): tempered for 22 hours at 600 °C.

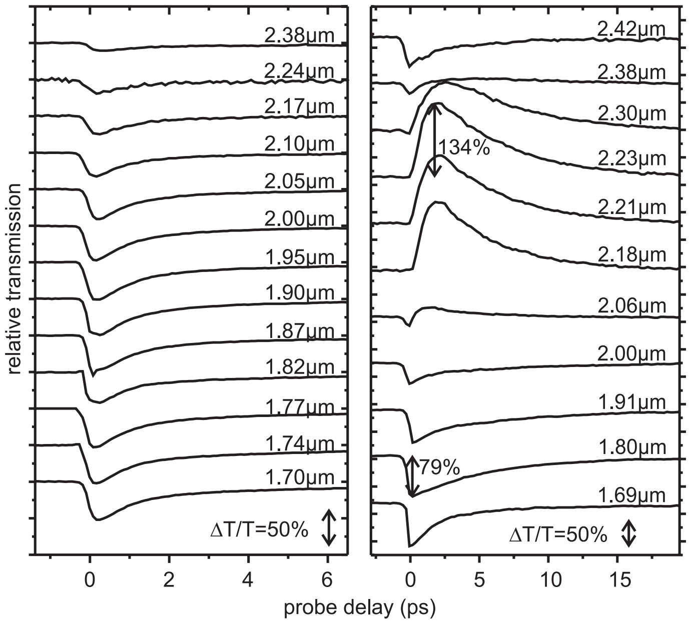

Fig. 3 Pump–probe traces for several probe wavelengths (as indicated), vertically displaced for clarity. LHS column: As-grown sample (a-Si), $\lambda_{\text{pump}} = 1.84$ µm, $I_{\text{pump}} = 66$ GW/cm^2; RHS column: Tempered sample (polycrystalline Si), $\lambda_{\text{pump}} = 1.73$ µm, $I_{\text{pump}} = 90$ GW/cm^2. All curves in one column are shown on the same scale of transmission relative to the unexcited sample (see arrows). Note the different time scales on the LHS and RHS, respectively.

For the interpretation of the data it is instructive to have a look on the electron scattering times of amorphous and polycrystalline silicon. Random variations of the microscopic potential in amorphous silicon break momentum-conservation restrictions for carrier scattering, leading to electron scattering times in amorphous silicon even below one femtosecond [8]. This causes a broad and featureless nonlinear optical response and leads to a dominance of absorptive rather than dispersive effects. In crystalline silicon, much longer scattering times are expected.

To further analyze and interpret our experimental data, we have performed simple one-dimensional model calculations based on the scalar wave approximation [9]. We assume that 50% of the void vo lume in the opal template is filled with silicon and consider 6 layers. The dielectric function $\varepsilon(\omega)$ of silicon at frequency ω is taken as the sum of a constant term and the Drude free-carrier contribution, i.e., $\varepsilon(\omega) = (3.48)^2 - \frac{n_{\text{e}} e^2}{m_{\text{eff}} \varepsilon_0} \left(\frac{1}{\omega^2 + \tau^{-2}} + \mathrm{i} \frac{\tau^{-1}}{\omega^3 + \omega \tau^{-2}} \right)$. Here, n_{e} is the optically excited electron density, e the elementary charge, τ the Drude scattering time, and m_{eff} the effective electron mass. The latter is chosen to be 20% of the free electron mass. The calculated linear transmission spectrum as well as two nonlinear spectra are depicted in Fig. 4. The linear spectrum (solid curve) qualitatively resembles the measured transmission spectrum (Fig. 1). The shape of the nonlinear spectrum sensitively depends on the choice of τ. For $\tau = 0.5$ fs (dashed curve in Fig. 3), the calculated spectrum is dominated by a broad and featureless increase of absorption without any major stop-band shift (also compare LHS of Fig. 3). In contrast, for $\tau = 10$ fs (dotted curve in Fig. 4), a clear blue-shift of the fundamental stop band results (also compare RHS of Fig. 3), which corresponds to a calculated maximum change in refractive index of $\Delta n = -0.5 + \mathrm{i}0.07$ at 2.2 µm wavelength.

In conclusion, we have systematically studied the nonlinear optical response of state-of-the-art silicon inverse opals which can serve as a model system for three-dimensional silicon-based photonic bandgap materials. We have shown that the nonlinear optical response dramatically depends on the underlying

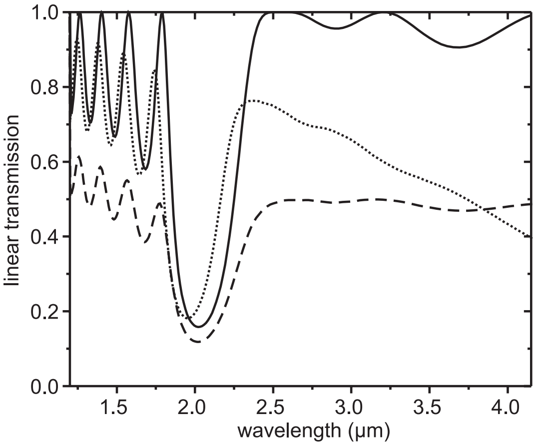

Fig. 4 Calculated transmission for three different parameter sets. The solid curve shows the linear spectrum (i.e., $n_e = 0$). The other two curves correspond to an optically excited electron density of $n_e = 1.5 \times 10^{20}$ cm^{-3} and $\tau = 0.5$ fs (dashed curve) and $\tau = 10$ fs (dotted curve), respectively.

electron scattering times. The latter can be tailored by sample tempering, initiating a transformation from amorphous to polycrystalline silicon. We achieve nonlinear optical transmission changes as large as 134% or stop band shifts on the order of 150 nm, corresponding to estimated refractive index changes of $\Delta n = -0.5 + i0.07$.

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