Identifying the role of the local density of optical states in frequency conversion of light in a microcavity

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We have reversibly switched the resonance of a GaAs–AlAs microcavity in the near-infrared near λ = 1300 nm within 300 fs by the electronic Kerr effect. We reveal by pump-probe spectroscopy a remarkable red shift or blue shift of the light confined inside the cavity for small pulse delays, depending on their temporal ordering. The color-converted light is efficiently generated in a broad frequency continuum that differs markedly from the instantaneous cavity resonance in terms of the central frequency and bandwidth. From observations on cavities with different quality factors, we identify the role of the local density of optical states (LDOS) available to the newly generated light frequencies. In particular, we distinguish the effect of the LDOS related to the cavity resonance itself, and the LDOS continuum that leaks in from the vacuum surrounding the cavity. Our new insights provide a unified picture for seemingly disparate results in traditional and nanophotonic nonlinear optics.

The generation of light with a controllable frequency is a longstanding challenge in physics that draws continued attention, driven by novel emerging applications,1 and by an ongoing miniaturization of devices.2–7 The generation of optical frequencies starting from incident light with a particular frequency is widely pursued in nonlinear optics.8 In well-known self-phase modulation, an effect [9]. Since the frequency shift of the output light equal to rate of change of phase δω = −∂δφ(t)/∂t. Since the frequency shift increases with incident light intensity and with interaction length, long fibers have become popular media to obtain frequency shifts by means of the electronic Kerr effect. Large supercontinuum frequency shifts have been obtained with Raman or soliton mechanisms, which are being applied in commercial white-light sources for ultrastable clocks or advanced microscopy.1

A modern approach to frequency conversion employs the confinement of light in nanophotonic structures, such as waveguides and especially microcavities.2–12 The frequency of the confined light is converted by changing the optical properties of the nanostructures in time by a separate pump pulse. The nanoscale confinement leads to strong field-enhancements, hence efficient conversion is feasible with low power. Even though the devices have a very small footprint, a similar phase shift is achieved as in many meters of fiber, hence they possess a great potential for on-chip integration.2–7 The physics of nanophotonic frequency conversion is considered to differ from usual nonlinear optics: (i) the frequency shift δω(t) is independent of the rate of change of the phase (∂δω(t)/∂t), (ii) the output spectrum reveals discrete cavity resonances instead of a continuum.10

In case of a single-mode microcavity, the input light is adiabatically converted to output light at a frequency equal to the instantaneous cavity resonance.2 In the case the cavity has multiple resonances, the output light is non-adiabatically distributed over many cavity resonances.10

The central question we address in this manuscript is how to reconcile these apparently different views in traditional and nanophotonic nonlinear optics. To this end we consider a nanophotonic system—a planar microcavity—that sustains both a cavity resonance and a flat continuum of modes. We study the frequency conversion of confined light that occurs when the cavity is switched in an ultrafast way via the electronic Kerr effect. We observe either a red- or a blue-shift of the confined light, depending on the timing of the pulses in our pump-probe experiments. We study color- conversion for different quality factors, which allows us to identify the role of the local density of optical states available to the generated light. We propose a unified framework to describe nonlinear frequency-conversion in open and nanophotonic systems, which also enables to discriminate regimes where adiabatic or non-adiabatic conversion occurs.

We have performed experiments on planar microcavities as shown in Fig.1(a) that are made from GaAs and AlAs by molecular beam epitaxy. The cavities consist of a GaAs λ-layer (d = 376 nm) sandwiched between two Bragg stacks made of λ/4-thick layers of GaAs (dGaAs = 94 nm) and AlAs (dAlAs = 110 nm), respectively. The bottom Bragg mirror of the cavities consists of 19 pairs of GaAs and AlAs. We varied the thickness of the top Bragg mirror to 7, 9, and 15 pairs of layers, which results in cavities with increased confinement with quality factors of Q = 390 ± 60, Q = 550 ± 60, and Q = 890 ± 60, respectively, corresponding to storage...
times between 265 ± 45 fs and 605 ± 45 fs.

For nonlinear pump and probe experiments, we employed two independently tunable optical parametric amplifiers, see Fig. 1(b), both with \( \tau = 140 \pm 10 \) fs pulse duration. The frequency of the probe light \( \omega_{pr} \) is tuned to the cavity resonance frequency \( \omega_{res} \). The probe bandwidth is broader than the cavity linewidth since the pulse duration is shorter than the cavity storage time. To avoid excitation of free carriers by two pump photons or by one pump and one probe photon, the pump pulses are tuned to \( \omega_{pu} = 4165 \) cm\(^{-1} \) (\( \lambda_{pu} = 2400\)nm). The incident pump and probe pulse energies are set to \( I_{pu} = 22 \pm 2 \) pJ/\( \mu \)m\(^2\) and \( I_{pr} \approx 0.2 \pm 0.02 \) pJ/\( \mu \)m\(^2\). At every delay setting \( \Delta t \), probe light impinges on the cavity, takes about 100 fs to charge the cavity, and circulates during the storage time on average. While the microcavity is switched, frequency-converted light escapes and is collected in reflection geometry, where it interferes with probe light that has directly reflected from the top mirror. The signal is frequency-resolved and integrated on an InGaAs detector array, yielding a transient reflectivity \( R^2 \). The interference is effectively a phase-sensitive heterodyne detection of light from the cavity, which allows us to sensitively detect frequency-converted light outside the instantaneous resonance.

In Fig. 2(a) we present transient reflectivity spectra measured at several pump-probe delays \( \Delta t \) of the cavity with \( Q = 390 \). \( \tau_{cav} = 265 \pm 45 \) fs. The reference spectrum reveals the unswitched cavity resonance at \( \omega_{res} = 7807.5 \) cm\(^{-1} \) (\( \lambda = 1280.8 \) nm) and maximum 100% transient reflectivity. During the switching event, the cavity resonance quickly shifts by 5.7 cm\(^{-1} \) to a lower frequency at maximum pump-probe overlap (\( \Delta t = -100 \) fs), and returns to the starting frequency immediately after the pump pulse has gone (\( \Delta t = +100 \) fs), see Fig. 2(b). The overlap between the probe pulse that resonates in the cavity and the pump pulse is maximal at a delay \( \Delta t = -100 \) fs, due to the charging time of the cavity. The shift to a lower frequency is caused by the increased refractive index, due to the positive non-degenerate Kerr coefficient of GaAs. In Fig. 2(a) we observe a remarkable transient reflectivity in excess of 100% at frequencies between 7813 cm\(^{-1} \) and 7851 cm\(^{-1} \), which unambiguously demonstrates generation of light, blue-shifted from the unswitched cavity resonance frequency. Simultaneously at frequencies between 7753 cm\(^{-1} \) and 7808 cm\(^{-1} \), below the cavity resonance, the signal is depleted compared to the reference spectrum. These observations highlight the major modification of the spectral distribution of the light confined in the cavity that is induced by the switch. As a net result, a significant part of the probe beam is blue-shifted by as much as 90 cm\(^{-1} \), from below to above the unswitched cavity resonance.

Fig. 2(b) shows a map of measured transient reflectivity spectra at each pump-probe delay \( \Delta t \). We observe that blue-shifted light occurs at time delays between \(-100 \) fs and \(+100 \) fs. Here the cavity resonance frequency \( \omega_{res} \) increases, hence the confined light experiences a decreasing refractive index, and thus a phase modulation \( \partial \omega(t)/\partial t < 0 \). Red-shifted light appears at time delays between \(-900 \) fs and \(-100 \) fs, where \( \omega_{res} \) decreases, hence the confined light experiences an increasing refractive index, and thus a phase modulation \( \partial \omega(t)/\partial t > 0 \). At earlier delay (\( \Delta t < -500 \) fs), both blue- and red-shifted light appear, since both an increase and decrease of the refractive index occur while the probe light recirculates in the cavity. The magnitude of the red-shifted light is larger than that of the blue-shifted light, since the pump pulse first induces an increasing and then a decreasing refractive index while the stored light intensity decreases. The fringes observed at \( \Delta t < 0 \) are the interference of light that directly reflects from the front Bragg mirror and light that escapes from the microcavity while it is being switched. Compared to the shift and width of the cavity resonance, the frequency shift of the stored light is much greater and has a much broader bandwidth.

To interpret our observations that contradict the standard view of frequency conversion in a nanocavity, we have calculated the nonlinear frequency conversion in reflection geometry, with only the pump fluence as an adjustable parameter. We employ a dynamic model de-
FIG. 2. (a) Measured and (c) calculated transient reflectivity ($R_t$) spectra at time delays $\Delta t = -500$ fs, $-100$ fs, and an unswitched reference taken at $\Delta t = 2.8$ ps where pump and probe have no temporal overlap. The high minimum reflectivity on resonance ($R_{\text{min}} = 85\%$) is caused by the asymmetric cavity design. Filled regions in (a) and (c) highlight how the new frequencies exit the switched cavity in reflection geometry. Panels (b) and (d) are maps of measured and calculated transient reflectivity versus time delay. The arrows indicate the direction of frequency conversion. The white symbols in (b) and the solid curve in (d) mark the resonance frequency of the cavity.

The probe field is calculated in the time domain at every position in a one-dimensional planar microcavity, that experiences a time-dependent refractive index using the positive non-degenerate Kerr coefficient of GaAs. The field is Fourier transformed to yield spectra, as shown in Figure 2(c) at fixed time delays. Our calculations yield blue- and red-shifted probe light at the same frequencies as in our measurements. Fig. 2(d) shows a map of frequency-conversion spectra, in excellent agreement with the measured transient reflectivity (Fig. 2(b)) regarding frequency shift, and timing. Since the timing of the up- or down-shifting corresponds with the rate of change of the phase, one might naively conclude that the consideration of phase modulation accounts for the present results. This is, however, not the case, as shown in the next sections.

FIG. 3. Measured integrated blue-converted light, averaged over time delays between $\Delta t = -100$ and 0 fs as a function of inverse quality factor $Q^{-1}$. Data are shown for different pump pulse fluence 13, 168, 220 pJ/µm$^2$ (circles, triangles, squares), and lines are linear fits to the data.
$N(\omega \neq \omega_{res})$ that can be identified with $1/Q$ \cite{17, 18}. This contribution is the LDOS-continuum from the vacuum surrounding the cavity, that leaks into the cavity through the Bragg mirrors. The decreasing intensity of the frequency converted light with $Q$ cannot simply be explained by only considering the rate of change of phase.

Figure 3 also shows that the amount of frequencyconverted light increases with pump pulse fluence. This is understood from the third-order electronic-Kerr nonlinearity. The induced nonlinear polarization is expanded in nonlinear index $n = n_0 + n_2 I_{pu}$ to increase linearly with pump fluence $I_{pu}$. At constant pump pulse duration, an increased refractive index corresponds to an increased rate of change of the phase $\partial \delta \phi(t)/\partial t$, and thus an increased frequency shift. This trend agrees with traditional nonlinear optics \cite{8}.

At this point, we have shown that the frequencyconverted intensity is related to both the LDOS and the nonlinear polarization. This is further demonstrated by considering calculations for various experimental conditions, as shown in Figure 4. To distinguish frequency conversion of light stored inside the cavity from elastic scattering of a time-dependent cavity resonance, we calculate the transient property $1 - T^r - R^r$. In absence of frequency conversion, $1 - T^r - R^r = 0$. For a microcavity with a low quality factor ($Q = 400$), Fig. 4(a) shows a broad blue conversion up to 7880 cm$^{-1}$, with a concomitant red depletion as far as 7720 cm$^{-1}$, similar to our observations. The bandwidth of frequency conversion clearly exceeds the cavity linewidth ($\Delta \omega = 20$ cm$^{-1}$), revealing again the remarkable notion that light is generated in a continuum outside the cavity resonance. With the LDOS and the nonlinear polarization, we rationalize the broad frequency continuum: Fig. 4a) shows that for the low-$Q$ microcavity the LDOS in the central $\lambda$-layer has a maximum of 10 ns/m on resonance, decreasing to $3.10^{-2}$ ns/m in the far off-resonance wings \cite{19}. In the whole frequency range the LDOS is dominated by the cavity resonance, and the vacuum LDOS that leaks into the cavity is at least $3x$ less. Hence the role of this LDOS-contribution is limited. The nonlinear polarization $P_{NL}(\omega_{pr})$ induced by the switch pulse is calculated in the central $\lambda$-layer \cite{20}. Fig. 4(a) shows that the nonlinear polarization spans a broad range of frequencies from about 7640 cm$^{-1}$ to 7980 cm$^{-1}$, even broader than the LDOS. Together, the broad LDOS and the even broader polarization conspire to yield a broad frequency conversion. While one naturally expects a cavity to be active over one linewidth, Fig. 4(a) shows that this intuition is erroneous: it appears that a much broader extent of the cavity LDOS spectrum is significant to frequency conversion.

To study the role of $Q$, Fig. 4(b) describes a high-$Q$ cavity ($Q = 7000$) with similar nonlinear polarization and mode volume as in Fig. 4(a). The LDOS is strongly pronounced: on resonance the LDOS averaged over $\lambda$-layer is nearly an order of magnitude larger ($N(\omega_{res}) = 6 \times 10^3$ ns/m), and off-resonance an order of magnitude lower ($N(\omega \neq \omega_{res}) = 10^{-3}$ ns/m), in agreement with the $Q$-related LDOS scalings above. The frequency conversion spectrum is narrow, with blue conversion up to 7820 cm$^{-1}$, and red depletion down to 7795 cm$^{-1}$. This situation is similar to earlier work in nanophotonic nonlinear optics \cite{2} \cite{11}: Since converted light exits the cavity only in the narrow range of the LDOS, this explains why high-$Q$ cavities typically yield adiabatic frequency conversion. Similarly, in case of a multimode cavity, the output will be distributed over many resonances corresponding to non-adiabatic conversion. It is clear that the frequency-integrated conversion is much smaller than for the low-$Q$ cavity (Fig. 4a), in agreement with our $1/Q$ results in Fig. 3. It appears that the cavities’ LDOS spectrum is so narrow that it has little overlap with the nonlinear polarization, which explains the small frequency-integrated conversion. In the time-domain, this result implies the cavity storage time.
is not matched to the durations’ of both pump and probe pulses. Hence ultrafast frequency conversion of light is best done in a cavity with a short storage time matched to the pulse duration, rather than with ultrahigh-$Q$ cavities.

To address the role of the nonlinear polarization, Fig. 3(c) reveals a broad frequency conversion spectrum, with blue conversion extending beyond 8000 cm$^{-1}$, and red depletion below 7800 cm$^{-1}$. In the limiting case of a broadband LDOS typical of a homogeneous material, the output spectrum will be completely determined by the nonlinear polarization, a situation typical for traditional nonlinear optics.

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To the best of our knowledge, the identification of the essential role of the LDOS in nonlinear frequency conversion has not been made to date. As our study concerns planar microcavities, the relevant LDOS is obtained by summing over zero transversal wavevectors. For the study on 2D slab photonic crystal cavities as in Ref. [11], the relevant LDOS must be summed over all wave vectors in the plane of the slab. In case of a 3D cavity, the relevant LDOS must be summed over all wave vectors in 3-dimensional space.

In Ref. [14], red-converted light was observed in a microcavity whose resonance was switched by free carriers, while the interpretation was qualitative at best. This result shows that frequency-conversion in a cavity can also be induced with time-dependent material excitations.

The inverse relation between off-resonant LDOS and quality factor $Q$ is readily derived for a 1D Bragg resonator: Since $Q$ is proportional to the finesse $F = \pi/\delta\omega$, with $n$ the order, the well-known relation for the finesse $F \sim 1/\omega$ can be rewritten as $Q = \pi \sqrt{1 - (\omega_{rad}/\omega)}$, using the standard relation between transmission and reflection $T(\omega) = R(\omega) = 1$. At frequencies outside the cavity resonance, but in the stopband of a Bragg mirror, the transmission decreases with depth $z$ as $T(\omega) = e^{-z/\ell_B}$, with $\ell_B$ the Bragg attenuation length. For a thick Bragg mirror $T(\omega) << 1$, we can invert $Q$ in terms of $T$ for a low-order resonance to $T(\omega) = \pi/Q$. In 1D the LDOS is proportional to $T$, giving the final result $N_{rad}(\omega) \propto 1/Q$.

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The LDOS in the central $\lambda$–layer was calculated with a static transfer matrix method, where we accounted for switching by increasing the refractive index of all GaAs layers by 0.06% [13]. The off-resonant LDOS was approximated by calculating the one for only Bragg stacks with a central $\lambda/4$–layer.

The nonlinear polarization $P_{NL}(\omega_{pr})$ induced by the third order effective susceptibility $\chi_{eff}^{(3)}$ is calculated from the stored probe field $E(\omega_{pr})$ in the middle of the $\lambda$–layer, that is windowed with pump field squared $|E(\omega_{ps})|^2$ (using the slowly varying envelope approxima-
tion) to yield \( P^{NL}(\omega_{pr}) = 3\varepsilon_0 \chi_{eff}^{(3)}(\omega_{pr}) E(\omega_{pr}) |E(\omega_{pu})|^2 \).

[21] The broadband \( \omega^2 \) LDOS typical of homogeneous media can be identified in the theory for second harmonic generation in Ref. [23].

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