All-optical delay of images by backward four-wave mixing in metal-nanoparticle composites

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We theoretically study a novel method for all-optical delay of images based on backward four-wave mixing in composites containing metal nanoparticles. In this approach a delayed phase conjugate probe pulse is generated by the interaction of two counter-propagating pump beams and a non-collinear shaped probe pulse in the nanocomposite. The fractional delay and the reflectivity for the phase-conjugate signal pulses are studied as a function of the input pump intensity. It is shown that this scheme can be used for delayed imaging combined with the elimination of optical diffraction. The advantages of this method include miniaturized design, tunable wavelength range up to the telecommunication range and wide bandwidth. © 2014 Optical Society of America

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Recent progress in the development of slow light devices has paved the way towards promising applications for storing, switching and processing of optical signals, increased resolution of spectroscopic interferometers, as well as in many other fields, e.g. nonlinear optics and quantum optics (see e.g. [1]). Recently it has been demonstrated that instead of single light pulses two- or three-dimensional images may also be slowed and stored. All-optical methods for delaying images can find applications in holography and optical pattern correlation measurements or all-optical image routers. The ability to slow images and delay them was demonstrated by using the effect of electromagnetically-induced transparency (EIT) [2–6] and coupled image resonators [7,8]. Applications in this field require methods for all-optical delay of images at arbitrary wavelengths (in particular at telecommunication wavelengths), large delay-bandwidth products, short (picosecond) pulse durations, and design suitable for on-chip intergration. Recently the authors of this letter have proposed a new approach for a slow light device based on the coherent interaction between pump and probe pulses in composite materials doped with metal nanoparticles [9]. In such materials near the plasmon resonance the absorption becomes saturated [10]. Therefore in a pump-probe regime an absorption dip in the homogenous plasmonic absorption profile and a steep change of the effective refraction index near the probe wavelength are created, leading to a significantly lower group velocity of the probe. In a non-collinear configuration with a TiO2 film doped with silver nanorods a large optical delay with a delay-bandwidth product of more than 60 for picosecond pulses at 1550 nm has been predicted.

Besides diverse other methods, backward degenerate four-wave mixing (DFWM) has also been studied for the realization of slow light in photorefractive crystals [11, 12]. The main drawback of slow light by using photorefractive materials is the very large response time, leading to a severely limited bandwidth. Backward DFWM results in phase conjugation [13] which is an important technique for imaging applications. In this Letter, we study an approach for all-optical delay applicable also for the delay of images based on composite materials doped with metal nanoparticles (NPs) and backward DFWM. In this scheme two counter-propagating pump pulses and an initial non-collinear pulse-shaped probe pulse excite the nanoparticle composite (see Fig. 1). As the result of the nonlinear interaction, a signal wave which is phase-conjugate with respect to the probe wave is generated. Due to the retarded nonlinear response of the metal nanoparticles in the ps-scale time range an absorption dip and a corresponding steep change of the refraction index lead to a delay of the signal wave. This process can be used for delayed imaging.

First, we study the mechanism of slow light in metal nanocomposites by using backward degenerate four-wave mixing. As presented schematically in Fig. 1 we consider a slab with a metal-NP composite illuminated by two counter-propagating quasi-cw pump beams E1 and E2 and a non-collinear probe pulse Epr, which has an arbitrary spatial amplitude and phase distribution. The three beams have the same central frequency and are assumed to be s-polarized. Then a s-polarized signal pulse Eγ is generated through the DFWM process. Since the two pump beams propagate along the opposite directions (k1 = −k2), and the signal wave is phase conjugate with respect to the probe (k3 = −k4), the phase-matching condition is automatically satisfied. A main feature in the optical response of the metal composite in the spectral range of plasmon resonance is the enhancement of the local electric field in the vicinity of the NPs. In this work we consider the excitation by picosecond pulses, therefore the transient response of the metal NPs is mainly influenced by electron-phonon processes and determined by the electron thermalization and the cooling of the hot electrons through the thermal exchange with the lattices in the metal. Using the two-temperature model, the nonlinear change of the dielectric function of
the metal can be described as [14]
\[
\Delta \varepsilon_m (t) = \frac{\chi_m ^{(3)} }{\tau_{ep}} \int_{-\infty}^{t} |E(t')|^2 e^{-\frac{i\omega t'}{\tau_{ep}}} dt',
\]
where \( \tau_{ep} \) is the electron-phonon coupling time in the range of 1-3 ps [15]. \( \chi_m ^{(3)} = \chi_m (\omega_0; \omega_0, -\omega_0, \omega_0) \) is the inherent third-order nonlinear susceptibility of the metal NPs at the pump wavelength \( \omega_0 \) and \( E(t) \) is local total field at the nanoparticles enhanced by the plasmon resonance. With the slowly varying amplitudes of each beams \( A_1, A_2, A_{pr}, \) and \( A_s \) the total electric field is given by \( E(r, t) = A_1 e^{i k_1 r - i \omega t} + A_2 e^{i k_2 r - i \omega t} + A_{pr}(z, t) e^{i k_{pr} z - i \omega t} + A_s(z, t) e^{i k_3 r - i \omega t} + c.c. \). We assume that the intensity of both quasi-cw pump beams is much higher than those of the probe and signal waves. Neglecting dispersion and using the slowly-varying envelope approximation and Eq. (1), the propagation of signal and probe pulses can be described in the frequency domain as follows:
\[
\begin{align*}
\partial A_s (\Omega, z) / \partial z &= a A_s + b A^*_{pr} \\
\partial A_{pr} (\Omega, z) / \partial z &= b A^*_s + a A_{pr}
\end{align*}
\]
where \( A_s (\Omega, z) \) and \( A_{pr} (\Omega, z) \) are amplitudes of signal and probe pulses, \( z \) is the coordinate along \( k_4 \),
\[
b = 2i \delta A_1 A_2 / (1 - i \Omega \tau_{ep}) \quad \text{and} \quad a = i \delta \left( |A_1|^2 + |A_2|^2 \right) \left( 1 + \frac{1}{1 + i \Omega \tau_{ep}} \right) - \alpha / 2,
\]
Here \( A_1 \) and \( A_2 \) are the input amplitudes of the two pump waves, \( \delta = \omega_0 \chi_m ^{(3)} / 2c \Re \sqrt{\varepsilon_{in} (\omega_0)} \cos \theta \), \( \alpha = 2 \Im \sqrt{\varepsilon_{in} (\omega_0)} \omega_0 / c \) is the linear absorption coefficient, \( \theta \) is a half intersecting angle between vectors \( k_1 \) and \( k_3 \), and \( \varepsilon_{in} (\omega_0) \) is the linear effective dielectric function of the composite. For spherical particles with diameters smaller than 10 nm the nonlinear field enhancement factor \( x \) in the frequency domain is given by the implicit relation (see Ref. [9])
\[
x = \frac{3 \varepsilon_h}{\varepsilon_m + 2 \varepsilon_h + \chi_m ^{(3)} |x E(\omega_0)|^2)
\]
which is solved numerically, and the effective third-order susceptibility of metal nanocomposite is calculated by 
\[
\chi_{eff} ^{(3)} = f \chi_m ^{(3)} |x|^2 x^2 ,
\]
where \( \chi_m ^{(3)} \) is that of the metal and \( f \) is the volume filling factor of NPs. Here \( \varepsilon_{m0} \) and \( \varepsilon_h \) are the permittivities of the metal and the host, respectively and \( \varepsilon_{in} (\omega_0) \) is determined by the generalized Maxwell-Garnett formula including the intensity dependence of \( x \) given by Eq. (4). For spherical NPs with large diameter and nonspherical NPs we calculate the enhancement factor \( x \) and \( \varepsilon_{in} (\omega_0) \) by using the generalized discrete dipole approximation modified to include nonlinear saturation effects (see Ref. [10]).

In order to characterize the pulse shape change we use the distortion function \( D \) of the probe pulse defined by [16]
\[
D = \sqrt{\frac{\int_{-\infty}^{t} |I_{out}(t + \Delta t) - I_{in}(t)| dt}{\int_{-\infty}^{t} I_{out}(t + \Delta t) dt}}, \quad (4)
\]
where \( I_{in} \) and \( I_{out} \) are normalized input and output probe pulse intensities, respectively, and \( \Delta t \) is the peak delay time.

Fig. 2. Slow light by DFWM in TiO2 layer containing Ag NPs smaller than 10 nm with filling factor of 10^-4. (a) fractional delay \( F \), (b) conjugate reflectivity \( R \), and (c) pulse distortion.
By using the above described formalism we now demonstrate the slow light mechanism by using backward FWM and metal nanocomposites. For a simplified numerical approach using the Maxwell-Garnett model we consider first a TiO$_2$ composite containing spherical silver NPs with diameters of 10 nm which exhibit a plasmon resonance at around 610 nm. In Fig. 2 the fractional delay (a), the phase conjugate reflectivity (b), and the pulse distortion (c) are shown as functions of the medium length normalized by the effective length $L_{\text{eff}} = 1/\alpha$ and the pump intensity at 625 nm. The probe pulse duration is 1 ps. We assume the same intensity of the both counterpropagating pump beams and an electron-phonon response time of $\tau_{ep} = 1$ ps. In this case, the effective length is $L_{\text{eff}} = 90.2 \mu$m. Figure 2 shows that by using this configuration the maximum fractional delay can be in the range of 1 with reflectivity up to more than 0.5 for a pump intensity lower than 8 MW/cm$^2$. The distortion $D$ is about 0.6.

Let us now study the possibility to use backward FWM in metal nanocomposites for the purpose of image delay. To demonstrate this mechanism at telecommunication wavelengths we have chosen a 4.1 $\mu$m-thick TiO$_2$ film containing gold nanorods with a diameter of 22 nm and a length of 64 nm with a filling factor of $10^{-3}$. For these non spherical NPs we use for the numerical simulations the discrete dipole approximation modified to include nonlinear saturation effects [10]. The effective nonlinear susceptibility for gold at the wavelength of 1550 nm is $\chi_3 = -1.5 \times 10^{-11}$ m$^2$V$^{-2}$ [17]. Due to field enhancement and the extremely large inherent nonlinearity of gold at 1550 nm, a pump intensity of only 30 kW/cm$^2$ is sufficient. Figure 3 shows for an object with a total area of 0.32x0.32 mm$^2$ shown in (a) the delayed image obtained by the backward DFWM (c) as well as the distorted image after propagation over the same distance of 0.5 mm in free space without phase conjugation (b).

One of the main advantages of this process is that it provides optical phase conjugation [13]. A peculiar property of the phase conjugation in the metal-dielectric composite is that it permits both image reconstruction and optical delay, a combination not commonly found in other schemes for image delay. As can be seen from the figure without phase conjugation diffraction effects are noticeable already after 0.5 mm of propagation, since the characteristic size of the source features is only 10 nm. However, as shown in Fig. 3(c) phase conjugation reverses the curvature of the wavefront, resulting in an almost-perfect reconstruction of the object after propagation over 0.25 mm. The resultant fractional delay of the image in (c) is about 0.75.

To conclude, we have investigated all-optical delay by backward DFWM in metal nanocomposite materials. A fractional delay of a signal pulse has been predicted in the range of unity in both visible and telecommunication wavelength range. In particular, at telecommunication wavelength a weak pump intensity in the order of 30 kW/cm$^2$ can be applied for slowing down of optical pulses. Due to the optically phase conjugation property of the signal beam with regard to the probe, all-optical delay of images can be realized. The advantage of the proposed technique comprises miniaturized design, wide bandwidth reaching up to THz, and control of the central wavelength by changing the sizes and shapes of metal nanoparticles. The optical delay can be further increased by a thicker nanocomposite with lateral pump incidence as proposed in [9] or by several parallel composite slabs.

Fig. 3. Delayed image (c) by optical phase-conjugation with DFWM and diffracted image (b) in free-space with the same propagation length at telecommunication wavelength 1550 nm for a object shown in (a). The nonlinear material is TiO$_2$ layer doped with Ag nanorods with a diameter of 22 nm and a length of 64 nm.

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