Supplemental Material to the article

"Cubic nonlinearity enhancement in ENZ media: non-degenerate optical Kerr effect"

Nonlinear absorption in the ENZ regime. The nonlinear index is a complex quantity, $n_2 = n_{2r} + in_{2i}$, whose real part defines the nonlinear phase shift, and the imaginary part is associated with nonlinear absorption. According to the work by Caspani et al. [28], n_{2r}, experience enhancement when the probe wavelength approaches the ENZ wavelength λ_{ENZ}. In contrast to n_{2r}, the dependence $n_{2i}(\lambda)$ demonstrates more complex behavior in ENZ materials. The magnitude of n_{2i} changes the sign at λ_{ENZ}, which indicates the transition from positive to negative (saturable) nonlinear absorption [28]. Thus, at the ENZ wavelength, an enhanced nonlinear phase shift can be achieved with zero nonlinear losses. This is a unique feature of ENZ materials. However, as we have shown in the main text of the article, n_{2r} is generally enhanced at a wavelength λ', which is shifted from λ_{ENZ}. We attributed this anomalous shift to the spectral dispersion of both the linear permittivity and the cubic susceptibility $\chi^{(3)}$. Below we discuss the properties of n_{2i}, in the ENZ regime, especially at the wavelength λ'.

For the case of a non-degenerate nonlinear interaction between an intense pump beam and a weak probe beam, n_{2i} is defined by [28]

$$n_{2i} = \frac{3}{2\varepsilon_0 n_r^{\mathrm{pump}}} \operatorname{Im}\left[\frac{\chi^{(3)}}{n}\right] = \frac{3}{2\varepsilon_0 n_r^{\mathrm{pump}}} \frac{\chi_i^{(3)} n_r - \chi_r^{(3)} n_i}{|n|^2}, \tag{S1}$$

where ε_0 is the vacuum permittivity, c is the speed of light, n_r^{pump} is the real part of the linear refractive index at the pump wavelength, $\chi_r^{(3)}$ and $\chi_i^{(3)}$ are the real and imaginary parts of the third-order nonlinear susceptibility at the probe wavelength, n_r and n_i are the real and imaginary parts of the linear refractive index at the probe wavelength.

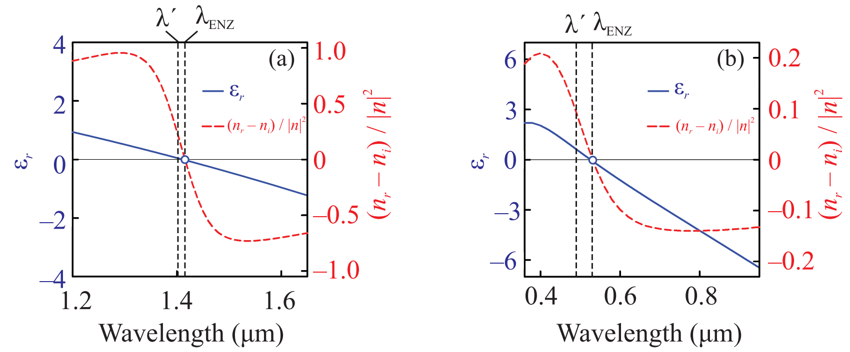

Fig. S1. (Color online) The real part of the linear permittivity as a function of wavelength for (a) – AZO and (b) – TiN. Experimental data on the permittivity of AZO and TiN are taken from [29]. Red curves show the trend of n_{2i} vs probe wavelength, which is calculated using Eq. (S1) and assuming nondispersive $\chi^{(3)}$ with equal real and imaginary parts. The vertical dashed lines indicate the ENZ wavelength (λ_{ENZ}) and the maximum of n_{2r} (λ')

In work [28], the simple case $\chi_r^{(3)} = \chi_i^{(3)} = \mathrm{const}$ was considered, that is, the spectral dispersion of $\chi^{(3)}$ was neglected. Under this assumption, the frequency dependence of n_{2i} is determined by the term $(n_r - n_i)/|n|^2$. The magnitude of n_{2i} is equal to zero when $n_r = n_i$. In turn, this condition is fulfilled when $\varepsilon_r = 0$. Thus, when $\chi_r^{(3)} = \chi_i^{(3)} = \mathrm{const}$, the value of n_{2i} is equal to zero at λ_{ENZ}. In order to illustrate this, we plotted ε_r and n_{2i} as a function of the probe wavelength for two ENZ materials: AZO (Fig. S1a) and TiN (Fig. S1b). Importantly, the wavelength corresponding to $n_{2i} = 0$ is not affected by the spectral dispersion of the linear permittivity.

Thus, the enhancement of n_{2r} is achieved at λ', whereas nonlinear absorption vanishes at λ_{ENZ}. Interestingly, λ' does not match with λ_{ENZ}. As a result, the enhanced nonlinear phase modulation in the ENZ media can be accompanied by either nonlinear losses ($n_{2i}(\lambda') > 0$) or saturable ($n_{2i}(\lambda') < 0$) absorption. This can be appreciated from Fig. S1, where the wavelength λ' indicates the maximum of n_{2r}. As seen, for both AZO and TiN λ' is blue-shifted from λ_{ENZ} towards the spectral range, where nonlinear losses take place ($n_{2i} \sim (n_r - n_i)/|n|^2 < 0$).

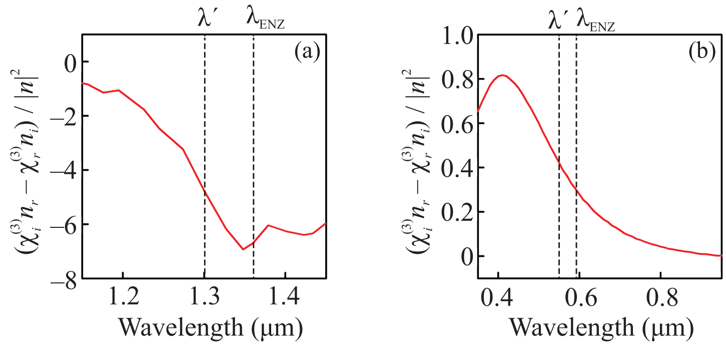

Fig. S2. (Color online) The trend of n_{2i} vs probe wavelength calculated using measured values of $\chi^{(3)}$ and n for (a) – AZO and (b) – TiN. The vertical dashed lines indicate the ENZ wavelength (λ_{ENZ}) and the maximum of n_{2r} (λ'). The wavelength λ'' is not shown since it is highly shifted from λ_{ENZ}

For the case of dispersive $\chi^{(3)}$, i.e. when λ_{ENZ} falls into the resonance of $\chi^{(3)}$, the wavelength dependence of n_{2i} is determined by the term $\left(\chi_i^{(3)} n_r - \chi_r^{(3)} n_i\right)/|n|^2$. Upon solving the equation $n_{2i} = 0$ we derive an expression for the wavelength λ'' at which nonlinear losses are zero:

$$\varepsilon_r(\lambda'') = \frac{\varepsilon_i(\lambda'')}{2} \left(\frac{\chi_r^{(3)}(\lambda'')}{\chi_i^{(3)}(\lambda'')} - \frac{\chi_i^{(3)}(\lambda'')}{\chi_r^{(3)}(\lambda'')} \right). \tag{S2}$$

The value of ε_r at λ'' is generally nonzero, therefore $\lambda'' \neq \lambda_{\mathrm{ENZ}}$. Thus, the enhancement of n_{2r} occurs at λ', whereas nonlinear losses vanish at λ'', and $\lambda' \neq \lambda'' \neq \lambda_{\mathrm{ENZ}}$. Figure S2 shows the wavelength dependence of n_{2i} plotted using measured values of $\chi^{(3)}$ and n for AZO (Fig. S2a) and TiN (Fig. S2b). Experimental data were taken from [26, 33]. The dispersion of $\chi^{(3)}$ significantly modifies the nonlinear optical response of an ENZ material. From Figure S2a we see that λ' lies in the region of negative n_{2i}. As a result, the ENZ-enhanced nonlinear phase modulation in AZO is accompanied by saturable absorption. In the case of TiN, n_{2i} is positive at λ', indicating the presence of nonlinear losses. The sign of n_{2i} is defined by the magnitudes of both, linear refractive index and nonlinear susceptibility of a particular material (see Eq. (S1)). It is important to note that $\varepsilon(\lambda)$ and $\chi^{(3)}(\lambda)$ of many ENZ materials depend on a variety of parameters, such as doping level, stoichiometry, crystallinity, etc. Therefore it is important to determine λ', λ'' and n_2 for each ENZ material of choice. We also note that n_{2r} and n_{2i} are independent. The ENZ enhancement of nonlinear phase shift is not affected by the nonlinear absorption occurring within the material.

The applicability limits of the condition $\varepsilon_r = 0$. According to Eq. (4), $\varepsilon_r^{\mathrm{opt}}$ at λ_{ENZ} is equal to zero only in the following cases: (1) $\varepsilon_i(\lambda_{\mathrm{ENZ}}) = 0$ and/or (2) $D(\lambda_{\mathrm{ENZ}}) = \pm 1$. We consider $D = 1$ only because $D = -1$ leads to negative n_r^{opt}. $D(\lambda_{\mathrm{ENZ}}) = 1$ when $dn_r/d\omega = -dn_i/d\omega$ at λ_{ENZ} (see Eq. (3)). Below we show, that the latter is fulfilled when $d\varepsilon_i/d\omega \ll d\varepsilon_r/d\omega$ at λ_{ENZ}. The real and imaginary parts of the linear refractive index are given as

$$n_r = \frac{1}{\sqrt{2}} \sqrt{\varepsilon_r + \sqrt{\varepsilon_r^2 + \varepsilon_i^2}}, \tag{S3}$$

$$n_i = \frac{\varepsilon_i}{2 n_r}. \tag{S4}$$

Upon inserting Eq. (S1) and Eq. (S2) into condition $dn_r/d\omega = -dn_i/d\omega$ one can obtain the following relation

$$\frac{d\varepsilon_i}{d\omega} \left(\frac{d\varepsilon_r}{d\omega} \right)^{-1} = \left(\frac{2|\varepsilon|}{\varepsilon_i - \varepsilon_r - |\varepsilon|} - \frac{\varepsilon_i}{\varepsilon_r + |\varepsilon|} \right)^{-1}. \tag{S5}$$

At the ENZ wavelength we have: $\varepsilon_r = 0$ and $|\varepsilon| = \varepsilon_i$. In this case, the right-hand side of Eq. (S5) is equal to zero. Thus, $D(\lambda_{\mathrm{ENZ}}) = 1$ when $d\varepsilon_i/d\omega \ll d\varepsilon_r/d\omega$ at λ_{ENZ}. It is important to note, that here we assumed $\chi_r^{(3)} = \chi_i^{(3)} = \mathrm{const}$.