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All-optical multiphoton absorption figures of merit: Polydiacetylene poly (bis para-toluene sulfonate) of 2,4-hexadiyne-1,6 diol

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The all-optical switching figures of merit are reported for the single crystal PTS (polydiacetylene poly (bis para-toluene sulfonate) of 2,4-hexadiyne-1,6 diol). Included are the effects of the absorption mechanisms from linear up to four-photon over the wavelength range 1.2–2.2 μm. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1767276]

One of the fundamental limitations to the implementation of passive all-optical switching devices is absorption, both linear and multiphoton. If absorption is too high at the device operating wavelength and input intensity, it becomes impossible to accumulate sufficient nonlinear phase shift to operate an all-optical device. This problem has been analyzed for linear, two and three-photon absorption, and formulas for the respective figures of merit (FOM) exist.[1–3] Unfortunately, there are very few cases in which the required nonlinear parameters have been measured, especially in nonresonant regimes where linear absorption is sufficiently small. Notable exceptions have been the two and three-photon figures of merit in the semiconductor AlGaAs for photon energies near and below half their band gap,[2,3] and the two-photon figures of merit for a number of organic materials including PPV (*p*phenylenevinylene), MEH-PPV [poly 2-methoxy-5-(2-ethylhexyloxy)-1, 4-phenylene-vinylene], and the single crystal polydiacetylene PTS [polydiacetylene poly (bis para-toluene sulfonate) of 2,4-hexadiyne-1, 6 diol] in their low linear absorption spectral regions.[4–6]

PTS in particular was one of the first polymeric materials to exhibit large refractive nonlinearities and it has been extensively studied at wavelengths at and below 1 μm.[6–8] The most recent has shown that the nonlinear loss in PTS is more complex than just two-photon in origin, as assumed in previous work.[8] In fact, we have now measured the two-, three-, and four-photon loss of PTS, as well as the dispersion in its intensity-dependent refractive index coefficient n_2 in the region 1.2–2.2 μm.[9] This makes possible the evaluation of figures of merit in this wavelength range based on all the active nonlinear absorption mechanisms present there. These results are reported in this letter.

The concept involved in all-optical figures of merit is well known.[1–3] All-optical devices require a nonlinear phase shift given by $\Delta\phi^{NL}=k_{vac}n_2 IL_{eff}$ for operation where $k_{vac}(=2\pi/\lambda_{vac})$ is the propagation wave vector in vacuum, L_{eff} is the effective length of the device, and the intensity (I)-dependent index change is given by $\Delta n=n_2I$. Most devices require $\Delta\phi^{NL}>2\pi$ so that the $n_2 IL_{eff}/\lambda_{vac}>1$ is

needed.[1] Although both scattering losses and absorption mechanisms can limit L_{eff}, especially in waveguides, the inherent material limitation to L_{eff} is absorption. In the highly nonlinear materials needed for all-optical switching, $L_{eff} \approx 1/\alpha_{total}$ where the total absorption is given by $\Sigma_{n=1}^{\infty}\alpha_n I^{n-1}$ with $n=1$ corresponding to linear absorption, $n=2$ two-photon absorption, $n=3$ three-photon absorption, etc. Thus, a general FOM can be defined as

$$\text{FOM} = \frac{n_2 I}{\lambda_{vac}\displaystyle\sum_{n=1}^{\infty} \alpha_n I^{n-1}} \qquad (1)$$

with an FOM >1 required for a material to be useful.

All of the materials constants, n_2, α_n vary with wavelength. For materials like semiconductors it is possible, to within some approximations, to calculate these parameters.[10] In general for organic materials, however, it is necessary to experimentally measure the wavelength dispersion in n_2 and the α_n. There are very few measurements of the spectral dispersion in α_n for $n>2$. Fortunately the importance of
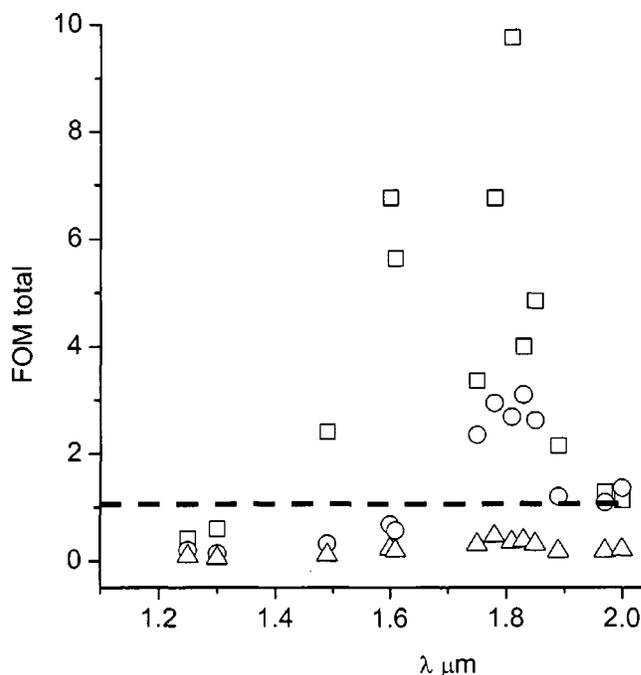

FIG. 1. Wavelength dispersion of the overall figure of merit at three different input intensities. ($\square$) 1 GW/cm²; ($\bigcirc$) 10 GW/cm²; ($\triangle$) 30 GW/cm².

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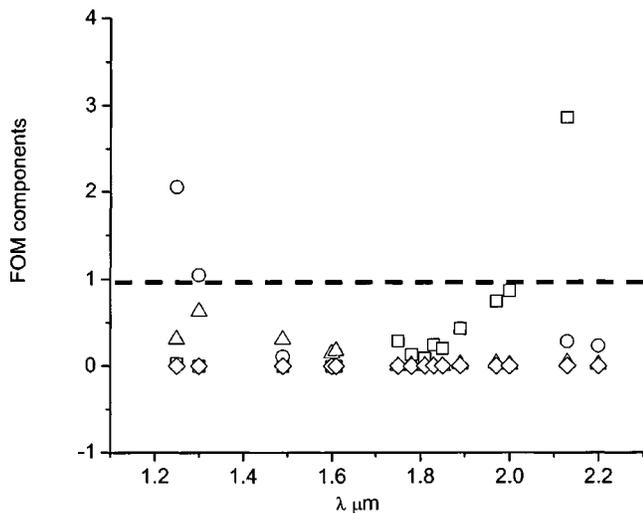
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FIG. 2. Wavelength dispersion of the figures of merit W^{-1} ($\square$), T ($\bigcirc$), V ($\triangle$), and U ($\diamond$) at 1 GW/cm^2.

FIG. 4. Wavelength dispersion of the figures of merit W^{-1} ($\square$), T ($\bigcirc$), V ($\triangle$), and U ($\diamond$) at 30 GW/cm^2.

multiphoton absorption decreases with increasing n so that usually only two-photon absorption plays an important role. However, as the wavelength increases, higher order absorption processes can play progressively more important roles because the nonlinear coefficient n_2 also decreases with increasing wavelength. To the best of our knowledge, the only experiment to date on the wavelength dispersion in n_2, α_2, α_3, and α_4 in a nominally transparent medium was reported recently for single crystal PTS.[9] The data can now be used to estimate the inherent material limitations in the wavelength range 1.2–2.2 μm, and to identify the relative contributions of the different multiphoton processes.

The overall FOM based on our measurements is shown in Fig. 1 for three different input intensities. Up to I = 1 GW/cm^2, the material is useful between 1.4 and 2.0 μm. (For $\lambda_{vac} > 2$ μm, linear absorption from vibrational overtones is responsible for the FOM falling below unity.) As the input intensity is increased, the spectral window for which the FOM > 1 decreases. For example, by I = 10 GW/cm^2 this window is reduced to 1.7–2.2 μm and for 30 GW/cm^2 the FOM < 1 over the full wavelength range. This means that

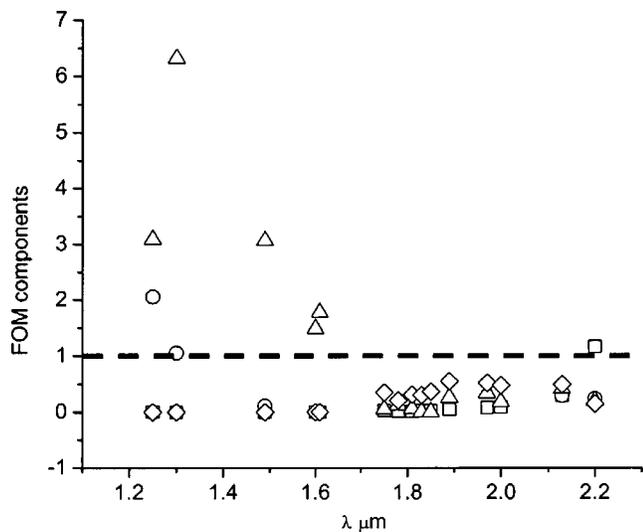FIG. 3. Wavelength dispersion of the figures of merit W^{-1} ($\square$), T ($\bigcirc$), V ($\triangle$), and U ($\diamond$) at 10 GW/cm^2.
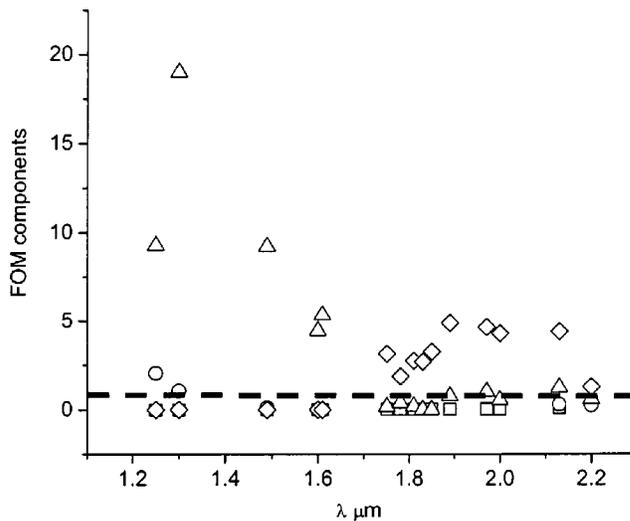
insufficient nonlinear phase shift can be accumulated over one absorption length at this input intensity for an all-optical switch. This variation in the values of the FOM corresponds to the dramatic difference in scenarios of nonlinear propagation in PTS depending on the input intensity.[11] That is, at I = 1 GW/cm^2, one observes strong self-focusing in the bulk of PTS, versus at I = 30 GW/cm^2, the dynamics of nonlinear propagation is largely dominated by higher-order multiphoton absorption.

The different multiphoton contributions to the FOM can be obtained by re-writing the FOM as

$$\text{FOM} = \frac{1}{\dfrac{\lambda_{vac}}{n_2} \displaystyle\sum_{n=1}^{\infty} \alpha_n I^{n-2}} = \frac{1}{W^{-1} + T + V + U}, \tag{2}$$

where the W, T, and V are the previously defined values of the one-photon, two-photon, and three-photon absorption figures of merit respectively.[1–3] (By analogy, we define the four-photon FOM as $U = \lambda_{vac} \alpha_4 I^2 / n_2$.) The multiphoton contributions are shown in Figs. 2–4 at the intensities 1, 10, and 30 GW/cm^2. Since $W^{-1} > 1$ above 2 μm due to vibrational overtones, its contribution is only shown in that wavelength range. At 1 GW/cm^2, the short wavelength limit is a consequence of two and three-photon absorption. In fact, the two-photon absorption is the long wavelength tail of the large two-photon absorption line centered at 0.93 μm so that for wavelengths shorter than 1.2 μm the overall FOM remains smaller than unity right into the visible region of the spectrum.[8] As mentioned previously, it is the linear absorption due to vibrational overtones that blocks the region $\lambda_{vac} > 2$ μm. At 10 GW/cm^2, it is the strong three-photon absorption peak of unknown origin that limits the useful spectral window below 1.7 μm.[9] At the highest intensity investigated, 30 GW/cm^2, it is four-photon absorption that now blocks out the region 1.7 μm to beyond 2.2 μm. This four-photon absorption is enhanced via an accidental three-photon resonant enhancement as discussed in detail in Ref. 9.

In summary, we have evaluated the all-optical figures of merit for single crystal PTS and found them to be limited by different absorption processes at different input intensities over the spectral window 1.2–2.2 μm. This material is defi-

nitely useful for all-optical switching at intensities up to 1 GW/cm^2 over the spectral window of 1.4–2.0 μm.

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