STRUCTURAL SCIENCE
CRYSTAL ENGINEERING
MATERIALS

Volume 71 (2015)

Supporting information for article:

Acentric crystals of new *N*-benzylideneaniline derivatives as potential materials for non-linear optics

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During the revision process of this paper it was suggested by reviewers that new SHG measurements should be taken at different wavelengths of the laser sources, specifically measurements using a 1907 nm incident radiation coming from a Raman shifted YAG laser. Unfortunately the suggested measurements could not be done. The main reason is that we do not have access to 1907 nm radiation in Brazil or to another one around this wavelength, specifically with the experimental setup for hyper-Rayleigh scattering experiment. Nevertheless, we have extensively searched researchers in our country owing this technology. However, all that we found were only able to measure SHG from sample excitation with a 1064 nm laser or near it. Unfortunately, SHG at 532 nm would be strongly absorbed, as occurred using the lasers at 804 nm and 980 nm which were available in our laboratories with the appropriate setup for NLO response measurements in solid state. Even if 1907 nm radiation was readily available, the experiment would be conducted in solution in order to obtain an estimate of the molecular hyperpolarizabilities and so, would not reflect exactly the main outcome of our study that is the NLO response on the acentric crystal. Another hindrance in using 1907 nm is the fact that compounds absorb around this near-infrared wavelength range, as can be seen in Fig. S1. These spectra are the same as Fig. 8 from our article, but going up to 1800 nm as measured originally. In addition, for isolated molecules, mainly those with electron donor and acceptor substituents, our computational protocol, extensively tested in our previous paper (Paschoal D. & Dos Santos, H. F. (2013). J. Mol. Model. 19, 2079-2090), is reliable enough to predict electric properties up to the first hyperpolarizability. Even though we were not able to measure SHG as suggested, this study does open this perspective through providing knowledge of these acentric materials, which will now be available in the literature. In this sense we not only describe the full compound synthesis and crystals preparation, but our samples are also available upon request by anyone interested in measuring SHG with laser technology.

Figure S1 Diffuse-reflectance spectra of derivatives I–III dispersed in $BaSO_4$ (10 mg of sample in 365 mg of $BaSO_4$). The absorption-band positions are indicated. These spectra are the same as Figure 8 in the article, but going up to 1800 nm as measured originally.

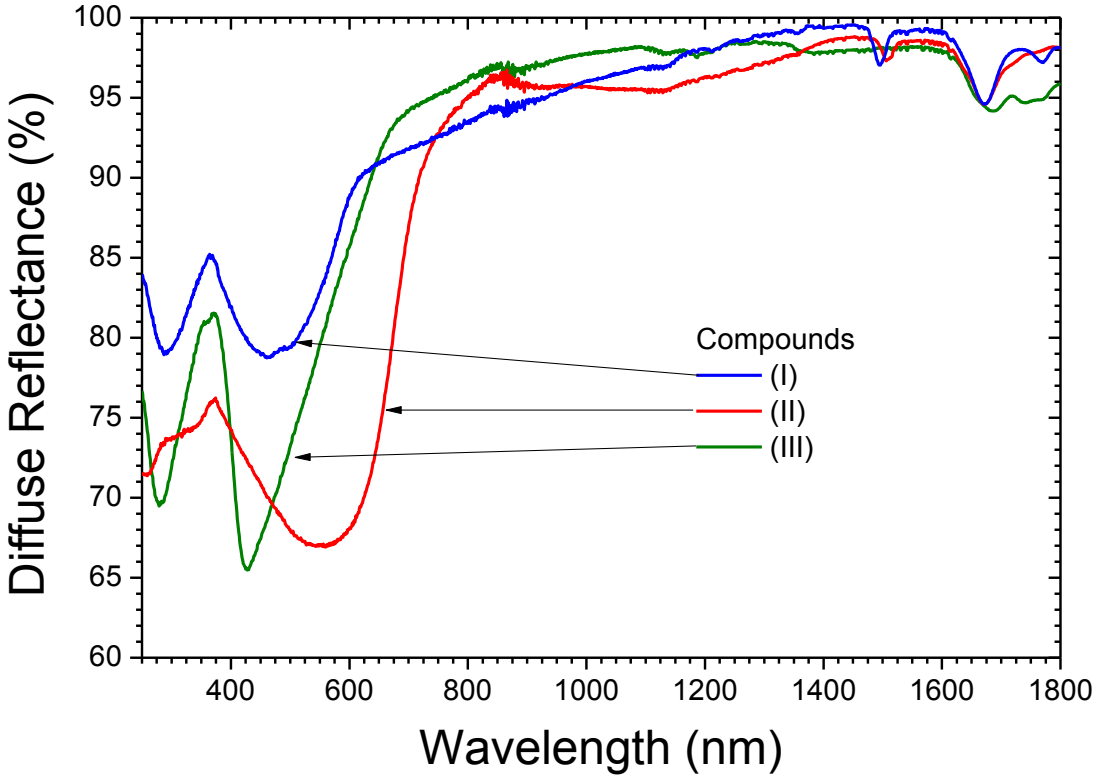

Figure S2 Single crystal of derivative I used in the single crystal X-ray analysis.

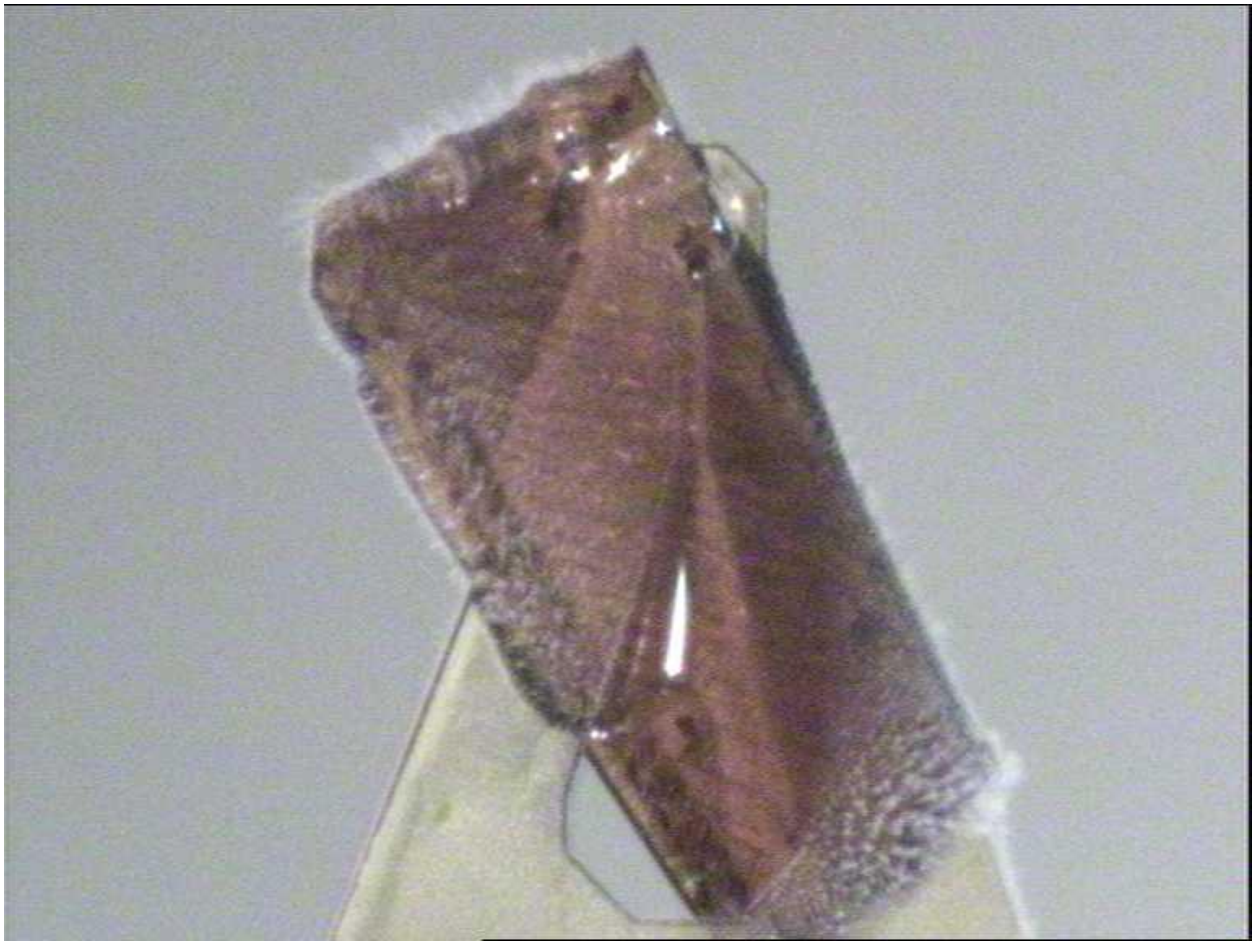

Figure S3 Single crystal of derivative II used in the single crystal X-ray analysis.

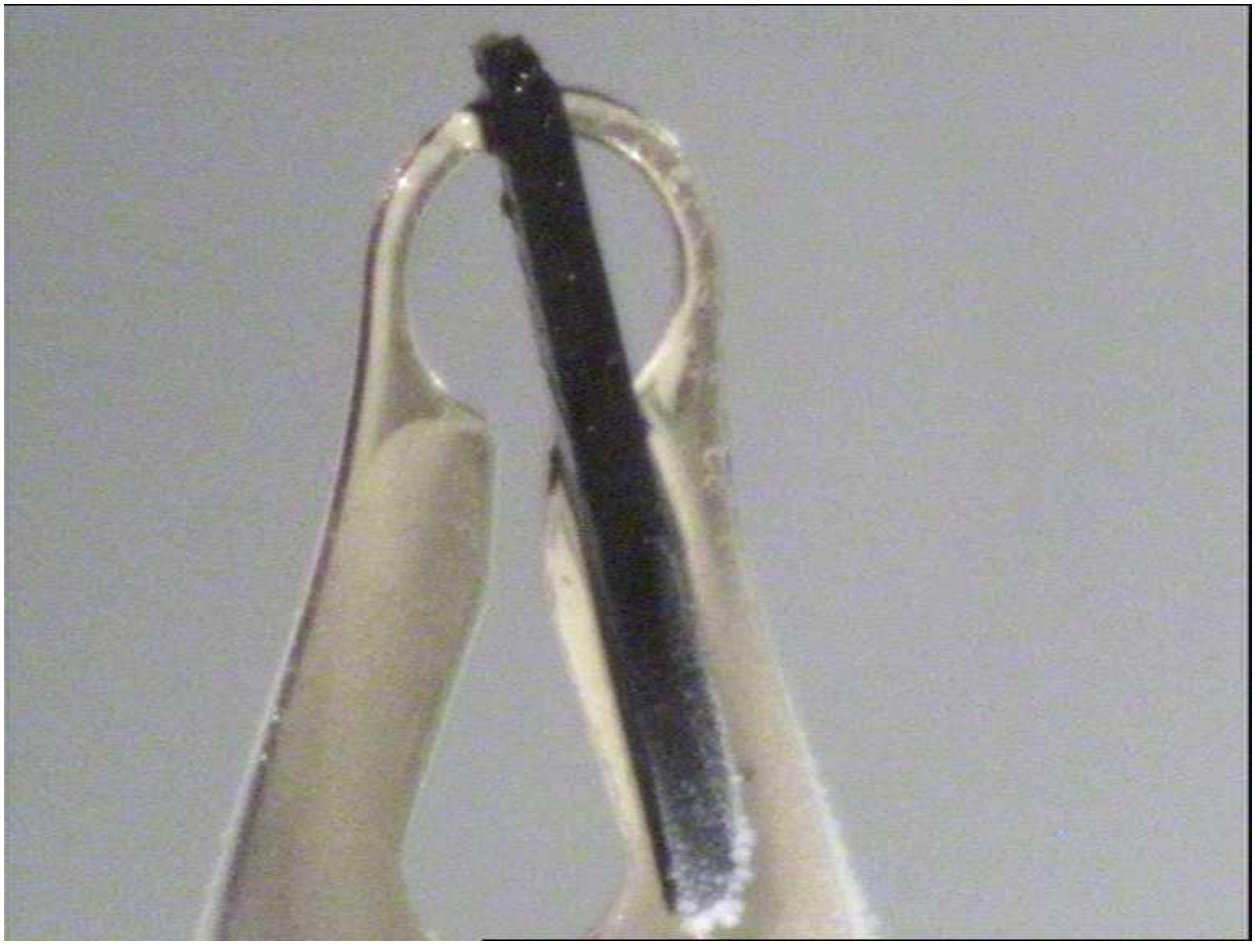

Figure S4 Single crystals of derivative III obtained by the slow cooling method in acetone.