Vacuum ultraviolet measurements on c-SiO_2 and a SiO_2

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VACUUM ULTRAVIOLET MEASUREMENTS ON c-SiO$_2$ and a-SiO$_2$

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Abstract: The reflectivity spectra of c-SiO$_2$ and a-SiO$_2$ show close similarity between 8 eV and 13 eV. We discuss these results in relation to band structure calculations.

The most studied forms of SiO$_2$ are silica (a-SiO$_2$) and alpha-quartz (c-SiO$_2$) which are stable at room temperature. We want to investigate and compare the two forms od SiO$_2$ at photon energies higher than 7 eV. We took vacuum ultra-violet (VUV) reflectivity spectra at 300 K between 7 and 13 eV for both c-SiO$_2$ and a-SiO$_2$ (Fig. 1). The absorption edge of a-SiO$_2$ was found to be at 7.8 eV and that of c-SiO2 at 8.6 eV. Two main peaks appear in the spectrum: one at 10.3 eV and one at 11.7 eV. The intensities of these reflectivity peaks and their widths agree, for both forms, with those of the literature [1,2,3]. New features in our spectra are: an additional structure at 9.5 eV and the fine structure of the peak around 11.7 eV for both a-SiO$_2$ and c-SiO$_2$. A 77 K spectrum taken on c-SiO$_2$ shows a shift of about 70 meV for the peak at 10.3 eV to higher energies. There is still controversy in the attribution of the structures observed in the reflectivity spectrum. For the band gap of a-SiO$_2$ data values of 9.0 eV and 9.3 eV have been given by photoconductivity. These values are 1.2-1.5 eV higher than the absorption edge we find for a-SiO$_2$. Many authors agree on the existence of an Urbach tail which extends down to the absorption edge and which is due to localised states. There are several calculations for the electronic band structure of alpha-quartz. The maximum in the valence band has been determined to be either in K or in M. The top of the

Fig. 1. Reflectivity of c-SiO$_2$ and a- SiO$_2$ at 300 K
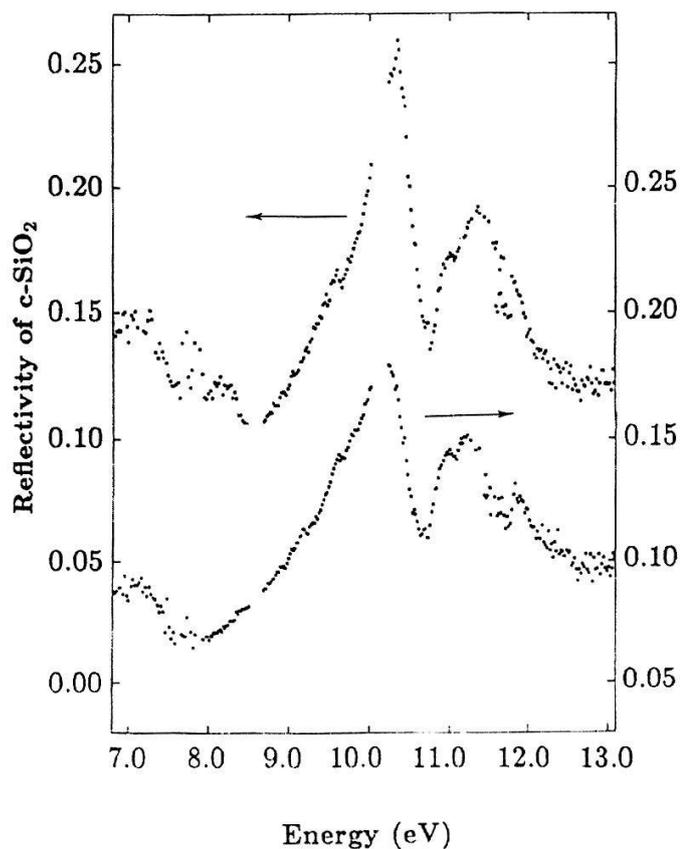
valence band is mainly composed of oxygen 2p non-bonding orbitals. The two main structures at 10.3 eV and 11.7 eV have been interpreted differently. Many authors agree that the first structure at 10.3 eV is of excitonic origin. The asymmetric shape of the peak could be due to a Fano resonance of an exciton with the continuum below.

From our spectra we find:

- an absorption edge of 8.6 eV for c-SiO$_2$ and 7.8 eV for a-SiO$_2$;

- a not previously observed structure at 9.6 eV;

- a structure at 10.3 eV, which agrees with previous results in shape, width and absolute intensity;

- a structure at 11.7 eV which is split into three peaks. Our observation confirms a previous result found by Platzoeder [3] for alpha-quartz and establishes it also for a-SiO$_2$.

Further measurements are needed, in particular lower temperature reflectivity measurements, which are currently being performed could give new insight into the problem.

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4-(N,N-DIMETHYLAMINO)-3-ACETAMIDONITROBENZENE BULK CRYSTALS AND SINGLE CRYSTAL CORED FIBERS FOR NONLINEAR OPTICAL APPLICATIONS

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Abstract: 4-(N,N-dimethylamino)-3-acetamidonitrobenzene (DAN) bulk crystal cored fibers of good optical quality have been grown and characterized. The measured second-order nonlinear optical susceptibility coefficient d_{23} =50 pm/V was is one of the largest phase-matchable coefficients observed in dielectric crystals.

1. Introduction

Attempts have been made to grow high quality single crystals of 4-(N,N-dimethylamino)-3-acetamidonitrobenzene (DAN), since second-harmonic generation (SHG) powder tests indicated efficiencies in the range of 115-200 times the value of urea [1,2]. In this work we present techniques for producing high optical quality bulk crystals and single crystal cored fibers of DAN and their optical characterization.

2. Growth and optical characterization of DAN bulk crystals and single crystal cored fibers

DAN bulk crystals were grown from solutions. Dimethylsulfoxide was saturated with DAN, and crystals of high optical quality were obtained from excess feed material by a temperature difference method based on thermal convection [3]. Crystal (001) plates of dimensions up to 15x7x2 mm^3 could be produced.

The material exhibits a large birefringence (0.7-0.3) and strong dispersion in the spectral region from 496.5 nm to 1064 nm as expected by judging from the orientation of the DAN molecules in the crystal lattice. Birefringence and dispersion data were used to calculate propagation directions for phase-matched frequency doubling of Nd:YAG laser radiation as

well as phase-matching angles and on wavelengths for propagation directions within the x - z plane. Loci of directions for collinear phase-matched second-harmonic generation of type I and type II were predicted and experimentally verified. Effective nonlinear optical coefficients were measured at several phase-matching directions which allowed the determination of all second-order nonlinear susceptibility coefficients allowed by crystal symmetry under the assumption that Kleinman's symmetry holds [4]: $d_{21} = (1.5 \pm 2)$ pm/V, $d_{22} = (5.2 \pm 1)$ pm/V, $d_{23} = (50 \pm 15)$ pm/V and $d_{25} = (1.5 \pm 2)$ pm/V at the wavelength $\lambda = 1064$ nm. The Figure of merit d_{eff}^2/n^3 $=140$ $[pm/V]^2$ for second-harmonic generation was determined for type I phase-matching, that is one of the highest values so far observed in dielectric crystals.

DAN single crystal cored fibers were grown in quartz glass capillaries of 7 to 20 μm in diameter by means of the inverted Stockbarger technique [5]. The growth direction parallel to the fiber axis is the a-[100] crystallographic axis. This favorable orientation allows the full use of the second-order susceptibility coefficients d_{22}, d_{23} and d_{34}. For a fiber of 15 mm length and 7 μm in diameter a SHG conversion efficiency of 0.1% for the 1064 nm line of a pulsed Nd:YAG laser with a fundamental peak power 6 W was achieved.

3.   References

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