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Electroabsorption spectroscopy of DAST microcrystals in solution

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[Introduction] The organic crystal 4-(*N*,*N*-dimethylamino)-*N*-methyl-4′-toluene sulfonate (DAST) has been widely studied as an organic nonlinear optical material. In the present study, we aim at clarification of electro-optical properties of DAST microcrystals in solution. Using electroabsorption (EA) spectroscopy, we measured electric field-induced change in absorption spectrum (EA spectrum). EA spectroscopy is a powerful technique to probe the electronic structure straightforwardly because the EA spectra can be related to the differences in the sample's dipole moment and polarizability between the electronically ground and excited states and the field-induced change in the transition dipole moment. For an ensemble of mobile polar molecules, the parameters characterizing the field-induced orientation/alignment can be also obtained. These molecular parameters are determined by fitting the EA spectra to the theoretical model [1].

[Experimental Section] DAST microcrystal was synthesized by the reprecipitation method [2]. 0.5 ml Mixture solution of DAST ethanol solution (5mM) and dodecyltrimethylammonium was injected into stirred acrydic A-138 decaline (50ml) solution. The resulting mixture was then filtrated with a Millipore filter (type: JH; pore size: 0.45μm). The sample solution was flowed in a home-made cell which consisted of two quartz plates coated with ITO conductive film and insulating layer. The cell was assembled with the polymer spacer to form a sandwiched structure. EA spectra were measured using electric-field modulation spectroscopy technique [3]. Sinusoidal voltage with a frequency of 4 Hz was applied.

[Results] The absorption spectrum of DAST microcrystals is shown in Figure 1. The DAST microcrystals show absorption peak at 553 nm and a shoulder below 500 nm. The absorption band can be decomposed into three components.

EA spectra are shown in Figure 2. The EA spectra remarkably depend on χ, which is the angle between the direction of the applied electric field and the direction of the electric field vector of the incident light inside the cell. The spectra could be simulated by a linear combination of the absorption and its derivative spectra (Figure 3, 4). With the theoretical model fitting, we obtained the ground state dipole moment to be

~3×10^4 D. Difference of the dipole moments between the ground and excited states was obtained to be ~3.5×10^3 D, which is one order of magnitude smaller than the dipole moment in the ground state. Electrophotoluminescence measurements have been also done for DAST crystals.

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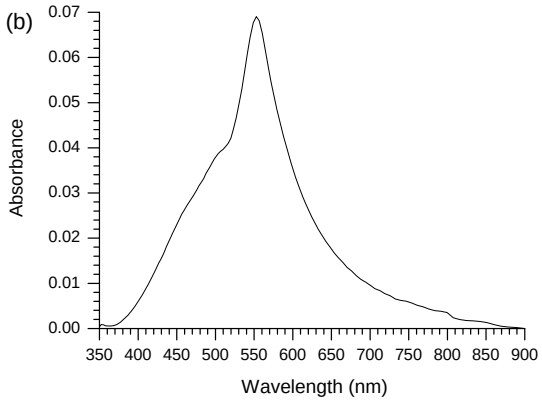

FIG.1 The absorption spectrum of DAST microcrystals

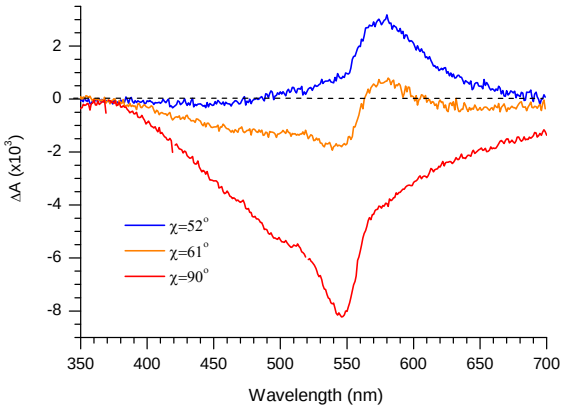

FIG.2 EA spectra of DAST microcrystals observed with different angles of χ

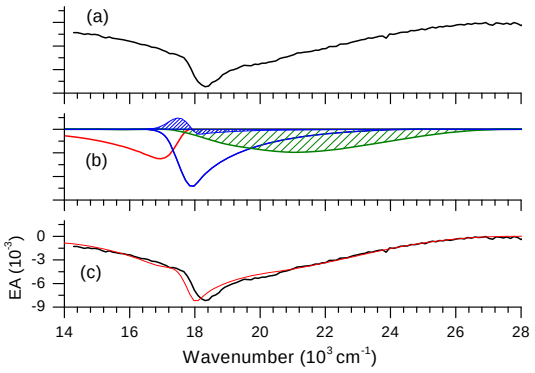

FIG.3 (a). EA spectra observed at χ= 90° (b). The first and the second derivative components of the EA spectra (b). EA spectra of DAST microcrystals with simulations

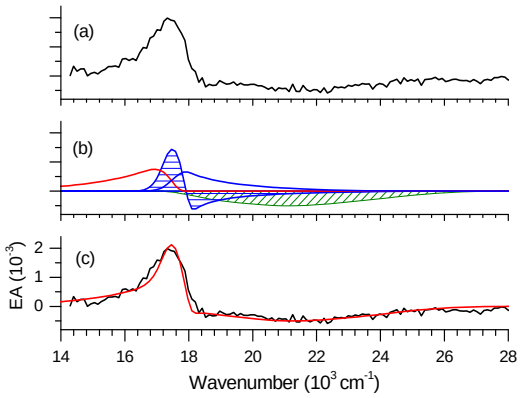

FIG.4 (a). EA spectra observed at χ= 54.7° (b). The first and the second derivative components of the EA spectra (b). EA spectra of DAST microcrystals with simulations