Improper Ferroelectric Transition in $\alpha'$-(BEDT-TTF)$_2$IBr$_2$
Studied by Nonlinear Optical Microscope

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In previous studies, we have reported the activation of optical second-harmonic (SH) generation in $\alpha$-(ET)$_2$I$_3$ [1] and the growth of large-scale polar domains [2], and discussed the unconventional mechanism of the ferroelectric transition associated with the Wigner crystallization of electrons. In an attempt to expand the family of ferroelectric organic conductors, we performed SH microscope observations for $\alpha'$-(ET)$_2$IBr$_2$ in the present study. Figure 1 shows the transmission image of a film-like single crystal of the complex and the scanning microscope image of SH signal collected from the same region. The sample was embedded in a polymer matrix for heat dissipation of the tightly focused excitation light. As shown in the transmission image, the sample was cracked together with the matrix when cooled. The complex is reported to be centrosymmetric, but there appeared SH signal below the charge ordering temperature as shown in Fig. (b). Note that the observed SH signal was emanated only from the regions near the cracks. The uneven signal distribution, which is attributable to inhomogeneous pressure caused by the polymer matrix, is presumably an indication of high susceptibility of the ferroelectric phase to external perturbations. The ferroelectric state in the complex showed several improper features, which are discussed in conjunction with the pattern of the charge ordering.

Fig 1. (a) Transmission and (b) SH image measured at 20 K.