Comment on "Photoalignment of Liquid Crystals By Liquid Crystals" - Reply

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Reznikov et al. Reply: In our Letter [1] we suggested near homeotropic alignment ($\theta_0 = 12^\circ$) of an adsorbed 4-4′-n-pentylcyanobiphenyl (5CB) monolayer using polarization and angle dependence of second-harmonic generation (SHG) and assuming a $\delta$-function polar distribution. In their Comment, Park et al. [2] claim that a planar alignment ($\theta_0 \approx 89^\circ$) of the adsorbed first aligning layer is more reasonable. They suggest the simultaneous analysis of birefringence and SHG results using the modified maximum entropy method (MMEM) [3]. However, as we explain below, (i) the use of MMEM here is inappropriate since it shows extreme sensitivity to the measured birefringence, and only slight sensitivity to SHG, (ii) in the Comment, the values for the nonlinear susceptibility $\chi^{(2)}$ components of 5CB monolayer were calculated using $\chi_{xyz}$ of fused quartz substrate, irrelevant for our experimental geometry, and (iii) these values were 100 times smaller than we calculated from our data. Thus, the model suggested by Park et al. is inconsistent with our experimental results.

In our model we considered the irradiated 5CB layer as possessing $C_{2v}$ symmetry. For this symmetry the nonzero $\chi^{(2)}$ components are $\chi_{zzz}$, $\chi_{xxx} = \chi_{zzz}$ and $\chi_{xyy} = \chi_{zzz}$. Then, assuming a $\delta$-function polar distribution and using our measured SHG ratio $\frac{\chi_{zzz}^2}{\chi_{xxx}^2} = 46$, we obtain the average molecular tilt angle $\theta_0$ from the normal to the substrate $\theta_0 = 12^\circ$. We also measured the magnitude of $\chi^{(2)}$ by referencing the sample SHG signal to both a quartz reference crystal and a previously studied Langmuir-Blodgett film ($\chi_{zzz} = 3 \times 10^{-14}$ esu/cm$^2$). These measurements both yielded a value of $\chi_{zzz} = 1.5 \times 10^{-14}$ esu/cm$^2$. Given a value of $\beta_{\xi\xi\xi} = 10^{-29}$ esu and a surface density of $N = 5 \times 10^{14}$ cm$^{-2}$ calculated from the packing of the molecules on the surface, this suggests an order parameter $\langle \cos^2 \theta \rangle \sim 1$. This is consistent with $\theta_0 = 12^\circ$ obtained from our measurements.

Park et al. use the MMEM [3] simultaneously analyzing birefringence $\Delta n$ and SHG results. They obtain the distribution function $f(\theta, \varphi)$ of 5CB molecules on the substrate that is claimed to reflect the ratios of nonlinear susceptibilities $\chi_{zzz} : \chi_{xxx} : \chi_{xyy} = 1 : 0.04 : 0.03$ and birefringence $\Delta n = 0.10$. We studied how $f(\theta, \varphi)$ changes when its parameters ($\lambda$s) are varied and found that as long as $\Delta n$ is fixed at 0.10, $f(\theta, \varphi)$ is insensitive to ratios of the nonlinear susceptibilities. On the other hand, changes in $\Delta n$ of a few percent result in noticeable changes in $f(\theta, \varphi)$ (see Fig. 1). In our Letter we provide an estimate for the birefringence value $\Delta n \sim 0.1$, but due to extreme sensitivity of the MMEM to $\Delta n$, the accuracy of this estimate is not adequate for reliably constructing the distribution function.

In addition, MMEM was shown to yield results consistent with a simple conventional $\delta$-function analysis [3]. Since this is not true here, it is inappropriate to use MMEM in our case.

Further, Park et al. calculated the values for $\chi_{zzz}$ and $\chi_{xyy}$ that are 100 times smaller than those we obtained and so are inconsistent with our data. Finally, Park et al. claim that their values for $\chi^{(2)}$ components are consistent with our ratios of SH intensities for the 5CB film to the bare substrate for $pp$ and $sp$ polarizations and the $\chi_{xyy}$ value for bare fused quartz substrate from the literature [4]. In this case the use of $\chi_{xyy}$ from [4] is irrelevant because a mixture of other substrate components would contribute in our SHG experimental geometry [4,5].

Finally, given the strong adsorption of the aligning layer on the substrate, it is reasonable that the polar cyano end of the molecule is attracted to the polar quartz substrate, and the hydrophobic alkyl tail of the 5CB repelled. Thus, a near normal orientation is physically reasonable, as well as consistent with our data.

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