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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 ($\theta_0 \approx 12°$) of an adsorbed 4-4′-*n*-pentylcyanobiphenyl (5CB) monolayer using polarization and angle dependence of second-harmonic generation (SHG) and assuming a δ-function polar distribution. In their Comment, Park *et al.* [2] claim that a planar alignment ($\theta_0 \approx 89°$) 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 χ_{yzy} 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 χ_{zzz}, $\chi_{zxx} = \chi_{xzx}$ and $\chi_{zyy} = \chi_{yzy}$. Then, assuming a δ-function polar distribution and using our measured SHG ratio $\frac{2\chi_{zzz}}{\chi_{zxx}+\chi_{zyy}} = 46$, we obtain the average molecular tilt angle θ_0 from the normal to the substrate $\theta_0 \approx 12°$. We also measured the magnitude of $\chi^{(2)}$ by referencing the sample SHG signal to both a quartz reference crystal ($d_{11} = 0.85 \times 10^{-9}$ esu) and a previously studied Langmuir-Blodgett film ($\chi_{zzz} \approx 3 \times 10^{-14}$ esu/cm^2). These measurements both yielded a value of $\chi_{zzz} \approx 1.5 \times 10^{-14}$ esu/cm^2. Given a value of $\beta_{\xi\xi\xi} \approx 10^{-29}$ esu and a surface density of $N \approx 5 \times 10^{14}$ cm^{-2} calculated from the packing of the molecules on the surface, this suggests an order parameter $\langle\cos^3\theta\rangle \sim 1$. This is consistent with $\theta_0 \approx 12°$ obtained from our measurements.

Park *et al.* use the MMEM [3]simultaneously analyzing birefringence Δn 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 χ_{zzz}:χ_{zxx}:χ_{zyy} = 1:0.04:0.03 and birefringence $\Delta n = 0.10$. We studied how $f(\theta, \varphi)$ changes when its parameters (λ′s) are varied and found that as long as Δn is fixed at 0.10, $f(\theta, \varphi)$ is insensitive to ratios of the nonlinear susceptibilities. On the other hand, changes in Δn 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 Δn, 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 δ-function analysis [3]. Since this is not true here, it is inappropriate to use MMEM in our case.

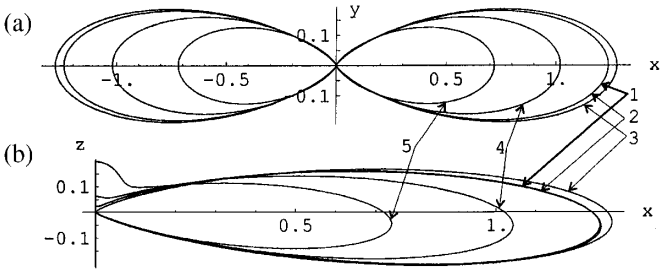

FIG. 1. Distribution function $f(\theta, \varphi)$ cross sections by (a) the x-y plane and (b) the x-z plane. (1) χ_{zzz}:χ_{zxx}:χ_{zyy} = 1:0.058:0.035, $S_x = 0.78$. (2) χ_{zzz}:χ_{zxx}:χ_{zyy} = 1:0.14:0.037, $S_x = 0.78$. (3) χ_{zzz}:χ_{zxx}:$\chi_{zyy} = 1{:}-3.58{:}-0.31$, $S_x = 0.81$. (4) χ_{zzz}:χ_{zxx}:χ_{zyy} = 1:0.18:0.052, $S_x = 0.72$. (5) χ_{zzz}:χ_{zxx}:χ_{zyy} = 1:0.2:0.062, $S_x = 0.57$.

Further, Park *et al.* calculated the values for χ_{zzz} and χ_{zyy} 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 χ_{yzy} value for bare fused quartz substrate from the literature [4]. In this case the use of χ_{yzy} 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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[1] Y. Reznikov *et al.,* Phys. Rev. Lett. **84**, 1930 (2000).
[2] B. Park, J. Wu, and H. Takezoe, preceding Comment, Phys. Rev. Lett. **87**, 249601 (2001).
[3] J.-G. Yoo *et al.,* J. Appl. Phys. **84**, 4079 (1998).
[4] P. Guyot-Sionnest and Y. R. Shen, Phys. Rev. B **35**, 4420 (1987).
[5] P. Guyot-Sionnest, W. Chen, and Y. R. Shen, Phys. Rev. B **33**, 8254 (1986).

