Dewitz and Hübner Reply: In the preceding Comment [1] the authors claim that the surface charge model applied in our investigation of the angular dependences of third harmonic generation (THG) from microdroplets [2] is inadequate.

In [2] we reported three major results: (i) The THG scattering profile of droplets having a radius of $a \gtrsim 8\ \mu$m is dominated by only a few intensity maxima at magic angles close to the forward and backward directions; (ii) the intensity and the angular distribution of the THG signal depend only weakly on the particle size and the central wavelength of the laser; (iii) both theory and experiment find intensity of comparable size in the forward *and backward* direction.

Carroll and Zheng (CZ) do not prove that these findings are incorrect. On the contrary, their results confirm ours. As a consequence the surface charge model, although it contains severe approximations, includes the relevant physics of the three effects (i)–(iii) mentioned above.

We do not believe that our model is able to yield exact angular dependences. For this purpose the model in [1] should be better suited, since it describes the source more accurately. Nevertheless, the results presented in [1] do not at all agree better with the experiment. This is true in the forward, but not in the backward direction, where our data are much closer (experiment: 26°/155°; Green function method [1]: 29°/141°; surface charge model [2]: 10°/158°). Thus in the end the deviations in both models are of the same magnitude. Nevertheless, we think that detailed comparisons of the angular dependencies obtained by the experiment and both theories are questionable at this stage of the investigations.

Carroll and Zheng mention that the range $79 < ka < 83$ is difficult to cover either analytically or numerically. In fact, this was one major reason why we applied the surface charge model, since it can deal with the full range numerically. We cannot agree with the statement "The derivation is very involved." Furthermore, the authors do not show how they obtained the generated field. In addition, when taking into account only a few fixed ka values, CZ did not cover the experimental situation. Because of the large spectral width and the uncertainties in the droplet radii, always a wide range of size parameters are excited ($\Delta ka \geq 1$) in the experiment [2]. Thus the results must be averaged in this range.

Figure 1 shows the total intensities obtained within our model in the range $77 < ka < 81$. The nonlinearity strongly enhances a few peaks to become dominant in height and weight. Because of the broad excitations, one of these or more will contribute. As a consequence, only the size parameter *ka in these peaks* has to be considered for the investigation of the angular dependence. The stabilization of the angular dependences is then caused by the nearly identical angular distribution of the intensities in the four dominant peaks. Thus a spectrally broad femtosecond (fs)-laser excitation is beneficial, since it always covers at least one of the most dominant peaks to get a stable angular dependence. Figure 1 also shows that we find many more than four resonances besides the four dominating ones. Since in the surface charge model only TE but no TM modes are included, the total number of resonances is reduced by a factor of 2.

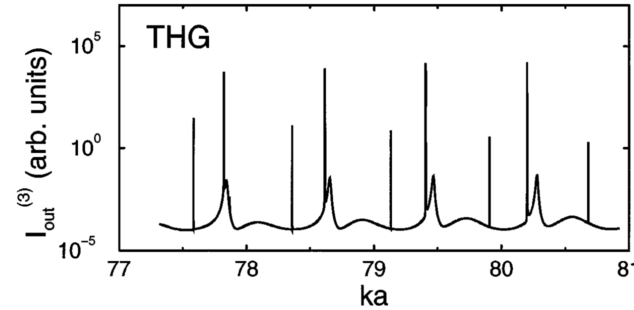

FIG. 1. Total THG intensities (logarithmic scale) of water droplets as a function of the size parameter ka.

The treatment presented in [1] in Eq. (1) cannot be called exact. The dependence of $\chi^{(3)}$ on the droplet size and the frequency is not included. Also, the reduction of $\chi^{(3)}$ to one element is based on the symmetry classification of $\chi^{(3)}$ in homogeneous and isotropic space. This does not apply to a general point in the droplet, since the symmetry is reduced due to the finite size of the droplet. That is, the tensors at the origin or at the surface will be different independent of the chosen coordinate system.

In the mentioned experimental work [3] a droplet train setup was used. The fluctuations in the droplet radii were reduced very effectively due to a very precise adjustment of some resonator frequency. A cw laser was used. The applicability of this setup to fs experiments is questionable, because the frequency of the resonator and the laser have to be synchronized. This will reduce the precision of the resonator frequency. Furthermore, the laser frequency cannot be varied in a large range and the spectral width of the fs pulse will always give a broadened excitation.

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