

Shelton Replies: As pointed out in the preceding Comment,¹ there is a clear discrepancy between theory and the results of the experimental measurements^{2,3} of the hyperpolarizability (γ) of the Ne atom. Bishop's argument is persuasive and fairly demands a reconsideration of the γ_{Ne} values determined from the electric-field-induced second-harmonic-generation (ESHG) experiments. While care has been exercised to foresee and eliminate possible systematic errors, there remains in our analysis an untested assumption which could lead to serious errors. The assumption is that the contributions to $\chi^{(3)}$ due to interacting pairs of atoms are negligible at the gas densities employed in our experiments. This assumption is supported by the results of a theoretical calculation⁴ which indicates that the corrections to $\chi^{(3)}$ due to pair contributions are negative and less than 1% of the total $\chi^{(3)}$ under the conditions of our experiments. However, since the phase-match density of the sample varies strongly with the laser wavelength employed in our measurements ($\rho \propto \lambda^3$), significant distortion of the measured γ_{Ne} dispersion curve would result if the pair contributions to $\chi^{(3)}$ were 10 times larger than expected. Electrode arrays of different periodicity were used for the near-infrared and the visible measurements, and so the actual Ne phase-match pressures were 54, 48, 46, 29, and 24 atm for the γ_{Ne} measurements given in order of increasing laser frequency. The effect of pair-interaction corrections would be to straighten the dispersion curve

while leaving the static limiting value of γ_{Ne} approximately unchanged. The initial slope of the dispersion curve could become less negative or possibly change sign depending on the magnitude and frequency dependence of the pair hyperpolarizability. ESHG experiments employing several different electrode arrays are now under way in our laboratory with the aim of measuring the density dependence of $\chi^{(3)}$ and the pair contribution to $\chi^{(3)}$. An alternative, independent determination of γ_{Ne} by means of electric-field-induced birefringence (dc Kerr effect) measurements is also under way. However, unless rather large systematic errors in the previous experiments come to light, an observed but impossibly large negative dispersion of γ_{Ne} may well remain.

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