

Iwakuni *et al.* Reply: In our Letter [1] we reported that the pressure-broadening coefficients of $^{12}\text{C}_2\text{H}_2$ alternate between the *ortho* and *para* transitions. The preceding Comment [2] claims that the alternation is due to our use of the Voigt profile (VP) and recommends to use a more advanced profile, such as a Galatry profile (GP). We argue here that the analysis with VP does not result in false values and that the width parameters in [1] are reliable within the given uncertainties.

We first point out that an additional broadening parameter cannot be well determined with our spectra. Figure 1 shows the transmittance spectra of the $R(9)$ transition for the four highest pressures. In the VP fitting, the Lorentz width is adjusted while the Gauss width is fixed at the theoretical value [1]. We refer to this as the L fit. As claimed in the Comment, the M-shaped residual appears for several intense transitions, but *only* at the highest pressures. This suggests that the narrowing effect becomes significant at this pressure but that the L fit is adequate for the other pressures.

We have therefore also fitted the transmittance spectra to a generalized VP in which both the Lorentz and Gauss widths are adjusted. We denote this as the LG fit. This fit takes account of the collisional narrowing in a simple manner, and is essentially equivalent to the GP. It has already been applied to the line profile analysis in continuous-wave-laser spectroscopy for the same $^{13}\text{C}_2\text{H}_2$ band [3]. In contrast, the LG fit did not appreciably reduce the standard deviations of the present spectra except for a part of the highest-pressure spectra; i.e., the standard deviations for the $R(9)$ profiles are reduced by 0%, 5%, 7%, and 27% for pressures of 396, 1047, 1962, and 2654 Pa, respectively. Furthermore, the

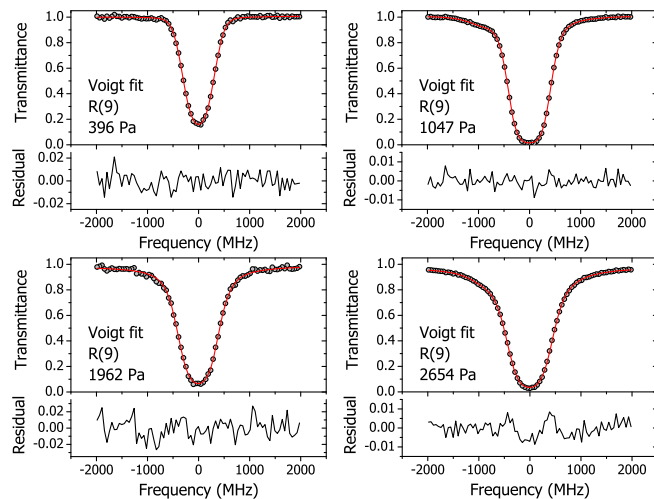


FIG. 1. Transmittance spectra of the $R(9)$ transition with sample pressures of 396, 1047, 1962, and 2654 Pa (open circles), fitted VP (red curves), and residuals (line graphs).

TABLE I. Comparison of pressure-broadening coefficients obtained by the L fit of the five lowest-pressure spectra, by the L fit of all spectra, and by the LG fit (in kHz/Pa). Numbers in parentheses are the uncertainties in units of the last digit.

Line	≤ 1962 Pa	All [1]	LG fit all
$R(7)$	47.7(8)	47.1(5)	51.9(6)
$R(8)$	42.4(7)	42.2(4)	52.1(9)
$R(9)$	46.4(6)	45.7(4)	49.4(6)
$R(10)$	47.1(16) ^a	44.1(11) ^a	50.9(14) ^a
$R(11)$	44.2(8)	44.1(4)	48.3(4)
$R(12)$	37.9(8)	39.3(5)	48.3(15)

^aUnreliable because of an accidental overlap with two weak lines [4].

determined Lorentz widths were less than the associated uncertainties for 73% spectra at pressures of 24.8 and 59.9 Pa because the signal-to-noise ratios were too low to extract the small pressure-broadening effect from the Doppler-broadened line.

Next we point out that the data at the highest pressure do not affect the reported *ortho-para* dependence. Table I lists some of the pressure-broadening coefficients determined without any highest-pressure data. The values agree with those in the Letter within the fitting uncertainties. The Lorentz width obtained by the L fit might deviate from the true value at the highest pressure, but the magnitude of the deviation is too small to wash out the *ortho-para* alternation.

We also list the pressure-broadening coefficients by the LG fit. The *ortho-para* alternations are obscured because the data quality does not allow us to determine both the Lorentz and Gaussian widths simultaneously, as mentioned above.

We thus concluded that the L fit was most appropriate in the present case. To fix the indeterminable parameters in the fit is a standard technique. Kusaba and Henningsen also employed the L fit to analyze the same $^{13}\text{C}_2\text{H}_2$ band [5] for similar reasons.

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- [1] K. Iwakuni, S. Okubo, K. M. T. Yamada, H. Inaba, A. Onae, F.-L. Hong, and H. Sasada, *Phys. Rev. Lett.* **117**, 143902 (2016).
- [2] J.-M. Hartmann and H. Tran, preceding Comment, *Phys. Rev. Lett.* **119**, 069401 (2017).
- [3] K. M. T. Yamada, A. Onae, F.-L. Hong, H. Inaba, and T. Shimizu, *C.R. Phys.* **10**, 907 (2009).
- [4] L. S. Rothman *et al.*, *J. Quant. Spectrosc. Radiat. Transfer* **130**, 4 (2013).
- [5] M. Kusaba and J. Henningsen, *J. Mol. Spectrosc.* **209**, 216 (2001).