

## Refractive-index dispersion of gases measured by optical harmonic phase matching

D. P. Shelton and Victor Mizrahi

*Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7*

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A new method for accurately measuring the dispersion of the refractive index of gases is presented. The technique makes use of the phase-matching condition which applies to a sample subjected to a spatially periodic electric field in an electric-field-induced second-harmonic generation experiment. The attainable accuracy is comparable to that of interferometric methods, and this method may be extended to measure the refractive index in the vacuum-ultraviolet range. Results are given for the refractive index at  $\lambda \geq 229$  nm for He, Ar, H<sub>2</sub>, D<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub>.

### I. INTRODUCTION

The refractive index of an atomic or molecular gas may be measured by long-established and highly accurate techniques, and is directly related to the linear polarizability of the constituent atoms or molecules.<sup>1-4</sup> The linear polarizability mediates most of the phenomena of linear optics.<sup>1</sup> The refractive index in the vacuum-ultraviolet (vuv) range would provide information about the electronic structure of simple atoms and molecules through their oscillator strengths,<sup>4,5</sup> but it is difficult to extend the interferometric techniques which are most accurate in the visible into the vuv range. A number of workers have used the phase-matching condition for nonlinear optical sum frequency generation as a basis for measurement of the refractive index in the uv or vuv region.<sup>6,7</sup> These experiments require several lasers, and either balance the dispersion of two near resonances,<sup>6</sup> or the negative dispersion of one gas component near resonance against the positive dispersion of a buffer gas.<sup>7</sup> Electric-field-induced second-harmonic generation (ESHG) is a particular case of sum frequency generation in which two photons of frequency  $\omega$  are combined with a zero frequency photon (i.e., static field) to produce a photon at frequency  $2\omega$ .<sup>8</sup> In this case, the use of a spatially periodic static electric field admits the possibility of a periodic phase-matching condition which may be satisfied for any gas at any frequency.<sup>9</sup> In what follows, we will describe the method for measuring the dispersion of the refractive index of a gas by periodic phase matching, and give the results of such an experiment done using visible lasers. In passing, we note that two-wavelength one-path interferometers which share some of the characteristics of the present method have also been demonstrated in the uv region.<sup>10,11</sup>

### II. EXPERIMENTAL METHOD

The experimental apparatus, which was principally designed to accurately measure the third-order nonlinear susceptibilities of gases, has been described in detail elsewhere.<sup>9,12-15</sup> The results presented here were obtained in the course of those experiments. A cw laser beam of about 1 W from an Ar<sup>+</sup>-pumped Rhodamine 6G or DCM dye laser, or from the Ar<sup>+</sup> laser directly, is weakly focused through a sample cell containing the gas in which second-harmonic generation takes place. This is made possible by a symmetry-breaking dc field. By arranging the electrodes so that the field direction alternates in space every coherence length (adjusted by varying the gas density) periodic phase-matching results, strongly enhancing the second-harmonic signal generated.<sup>9</sup> The particular electrode spacing used results in optimum pressures of order 1 atm, except for He (a low-dispersion medium) where phase match occurs at of order 100 atm. A double prism spectrometer serves to separate the second harmonic from the fundamental, and a photomultiplier detects the signal photons which are then counted. A measurement consists of filling the cell with a high purity gas sample and determining the count rate and precise density at which optimum generation occurs.

The second-harmonic power generated by focusing a fundamental Gaussian mode laser beam through a sample, in a region of length  $2L$  subjected to a spatially periodic electrostatic field of the form

$$E^{(0)}(z) = E_0^{(0)} \cos(Kz) \quad (1)$$

is given by<sup>9</sup>

$$P^{(2\omega)} = \frac{\omega^3}{z_0 \pi c n_{2\omega}^2} \left( \frac{\mu_0}{\epsilon_0} \right)^{3/2} \chi_{\text{NL}}^2 P^{(\omega)^2} E_0^{(0)^2} \left[ \int_{-L}^L dz \cos(Kz) \frac{\cos\{\Delta k(z-z_w) - \arctan[(z-z_w)/z_0]\}}{[1+(z-z_w)^2/z_0^2]^{1/2}} \right]^2, \quad (2)$$

where  $z_0$  is the confocal parameter of the focused beam,<sup>16</sup>  $z_w$  is the position of the beam waist with respect to the center of the static field region, and  $\chi_{\text{NL}}$  is the macroscopic nonlinear susceptibility of the medium. The wave vec-

tor mismatch  $\Delta k$  is determined by the difference in refractive index of the sample between frequencies  $\omega$  and  $2\omega$ , and is given by<sup>9</sup>

$$\begin{aligned} \Delta k &= 2k_\omega - k_{2\omega} = (4\pi/\lambda_\omega)(n_\omega - n_{2\omega}) \\ &= \frac{-2\pi}{\lambda_\omega \epsilon_0} (\alpha_{2\omega} - \alpha_\omega) \rho + (\text{terms in } \rho^2) + \dots, \end{aligned} \quad (3)$$

where  $n_\omega$  is the refractive index of the gas,  $\alpha_\omega$  is the linear polarizability of the constituent molecules, and  $\lambda_\omega$  is the vacuum wavelength for light of frequency  $\omega$ . At the gas pressures employed in these experiments (maximum 130 bars for He, maximum 10 bars for all other gases) the terms of order  $\rho^2$  and higher, the refractive index virial corrections, are less than 0.05% of the linear term and may be neglected.<sup>17-19</sup>

The spatially periodic electrostatic field is produced by an array of  $2N$  electrode pairs with spacing  $l'$ .<sup>9,15</sup> For an electrostatic field with a large number of periods  $N = KL/\pi$ , the integral in Eq. (2) is sharply peaked near  $|\Delta k| = K$ , with peak width and height proportional to  $1/N$  and  $N^2$ , respectively. The variation of harmonic intensity with gas density is illustrated in Fig. 1 of Ref. 9. By measuring the density  $\rho$  at which peak second-harmonic generation occurs, the refractive-index dispersion may be obtained from Eq. (3). The array used in these experiments consists of 150 pairs of wires ( $N = 75$ ) of diameter 1.59 mm, with average spacing  $l' = 2.6924$  mm. This geometry produces a field distribution described by Eq. (1) over the volume sampled by the laser beam, plus higher spatial frequency components of smaller amplitude which may be shown to have a negligible effect on the precise phase-match condition. In order to evaluate the precise value of  $\Delta k$  for which periodic phase matching occurs, we have collected all the factors with  $\Delta k$  dependence in Eq. (2), including those with implicit dependence through  $\rho$ , into a single factor such that  $P^{(2\omega)} \propto I^2(\Delta k)$ . The factor  $I(\Delta k)$  may be approximated to sufficient accuracy by the following sum over electrode pairs:

$$\begin{aligned} I(\Delta k) &= \frac{\Delta k}{K} \frac{1}{2N} \sum_{n=-N}^{N-1} (1+u^2)^{-1/2} \\ &\quad \times \{ (1 - \Delta k/K) \cos \phi \\ &\quad + (\cos \phi + u \sin \phi) [x_0(1+u^2)]^{-1} \}, \end{aligned} \quad (4a)$$

where

$$\phi = \frac{K + \Delta k}{K} (x' - x_w) - \arctan u, \quad (4b)$$

$$u = (x' - x_w)/x_0, \quad (4c)$$

$$x' = (n + \frac{1}{2})\pi, \quad (4d)$$

and  $\Delta k/K$  is the wave vector mismatch in units of the array spatial frequency  $K = \pi/l'$ , and  $x_0 = Kz_0$  and  $x_w = Kz_w$  are the normalized laser beam confocal parameter and waist position. The laser beam parameters, measured using a beam scanner, were about  $z_0 = 14$  cm and

$z_w = 0$  cm. The value  $|\Delta k/K|_{\text{opt}}$  which maximizes  $I^2(\Delta k)$  is a weak function of the parameters  $N$ ,  $l'$ ,  $z_0$ , and  $z_w$ . The variation of  $|\Delta k/K|_{\text{opt}}$  in the region of parameter space relevant to the present experiment is illustrated in Fig. 1. For this experiment  $|\Delta k/K|_{\text{opt}} = 0.9954$ , with the corresponding value of  $|\Delta k|_{\text{opt}} = 1161.5 \text{ m}^{-1}$ . Allowing for variations in the beam focusing and admixture of higher-order modes, the value of  $|\Delta k|_{\text{opt}}$  is probably accurate to  $\pm 0.05\%$ .

Having established  $|\Delta k|_{\text{opt}}$  for periodic phase matching in our apparatus, the experiment simply consists of varying the wave vector mismatch of the sample by varying its density until  $|\Delta k| = |\Delta k|_{\text{opt}}$ , and of measuring the density  $\rho_{\text{opt}}$  at which this occurs. Sample density is varied by slowly leaking out gas. A parabola is fitted to the measured second-harmonic signal near the top of the phase-matching peak, and in this way the pressure for peak signal may be determined with a relative precision of order  $10^{-4}$  for typical signals of 1000 photons/second. The temperature is measured by a thermistor thermometer in direct contact with the gas (accuracy 0.2°C). The density is obtained from the measured pressure and temperature of the gas by means of the virial equation of state.<sup>20</sup> The accuracy of the measured density is chiefly determined by the accuracy of the pressure gauges employed (recently calibrated MKS Baratron capacitance manometer,  $\pm 0.15\%$  of reading; Heise Bourdon tube gauge,  $\pm 0.1\%$  of 160 bar full scale). High purity gases were used in the measurements (He 99.999%; Ar 99.998%; H<sub>2</sub> 99.999%; D<sub>2</sub> 99.5 at.%, chief impurity HD; N<sub>2</sub> 99.9995%; O<sub>2</sub> 99.98%). The laser wavelengths spanned the range  $\lambda = 458\text{--}700$  nm; dye laser wavelengths were measured with a 0.75-m grating spectrometer.

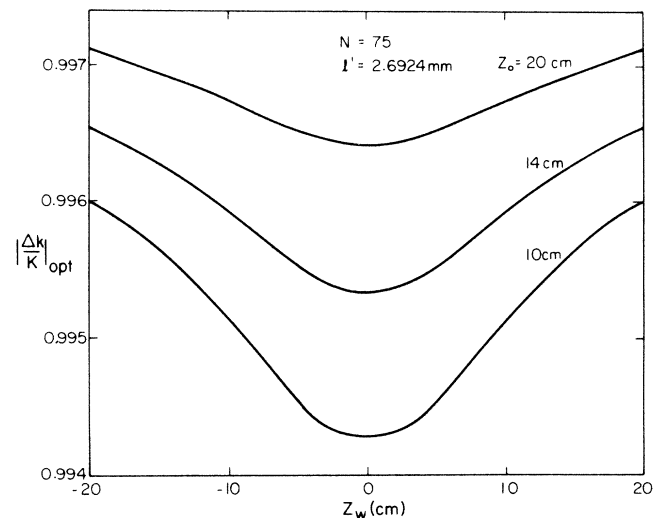


FIG. 1. Variation of the optimum wave vector mismatch  $|\Delta k/K|_{\text{opt}}$  with laser beam confocal parameter  $z_0$  and waist position  $z_w$  (measured from the center of the electrode array).

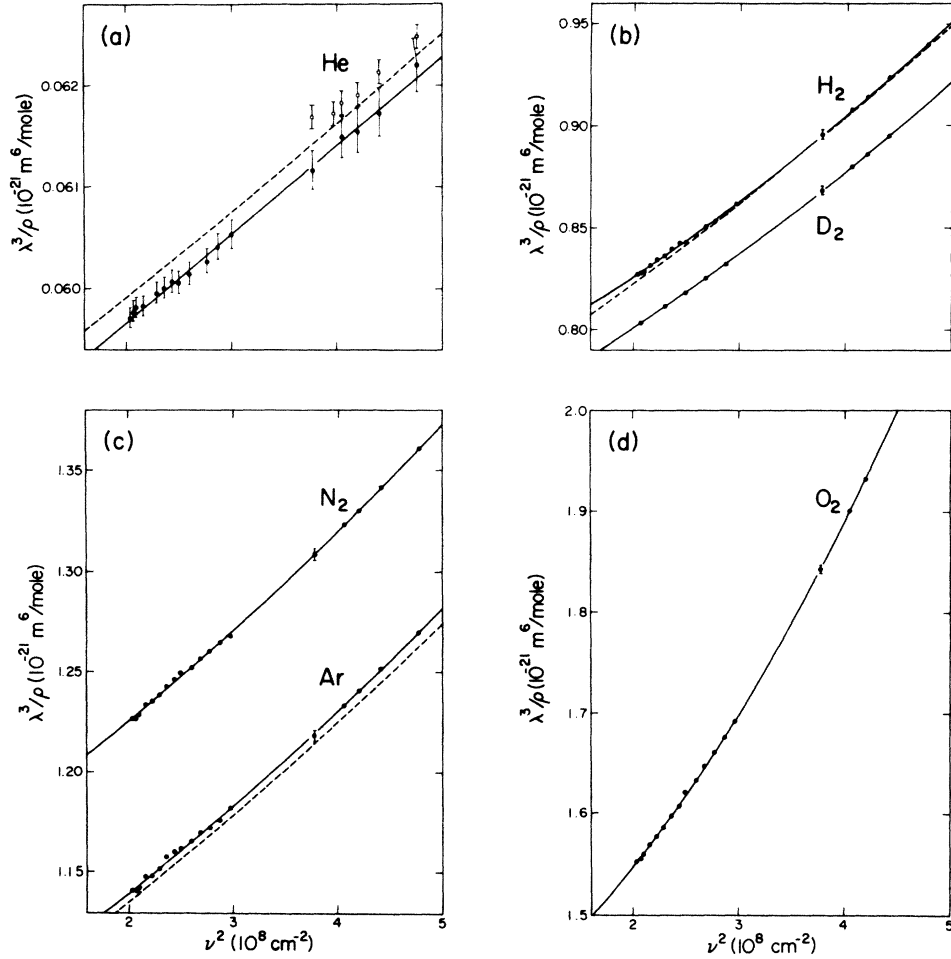


FIG. 2. Results of periodic phase match density measurements for He, Ar, H<sub>2</sub>, D<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub>, plotted as  $\lambda^3/\rho$  versus  $\nu^2$ , where  $\rho$  is the gas density for optimum phase match. The error bars are uniformly  $\pm 0.2\%$  except for He, so a single representative error bar has been indicated. The solid lines are least-squares fits of Eq. (6) to the data. The dashed curves in (a), (b), and (c) were obtained from the literature for the purpose of comparison (see the text). For He the open circles are the results from a previous similar experiment (Ref. 9) (with error bars enlarged to  $\pm 0.2\%$  to account for temperature uncertainties arising because the thermometer was not in direct contact with the gas).

### III. RESULTS AND DISCUSSION

Far below resonance, as is the case here, the linear polarizability to which  $n_\omega - 1$  is proportional may be expanded<sup>1,3</sup> as a power series in even powers of the frequency  $\omega$ . Using this result, the phase-match density may be expanded in the following power series:<sup>9</sup>

$$\rho^{-1} = a\lambda^{-3} + b\lambda^{-5} + \dots, \quad (5)$$

where this and all subsequent occurrences of  $\rho$  are understood to refer to the gas density at optimum phase match. Accordingly, we have presented our data in the form of a plot of  $\lambda^3/\rho$  versus  $\lambda^{-2}$  in Fig. 2. A function of the form

$$\lambda^3/\rho = A' + B'\lambda^{-2} + C'\lambda^{-4} \quad (6)$$

has been fitted to the data, as shown by the solid curves in Fig. 2. The coefficients  $A'$ ,  $B'$ , and  $C'$  of this fit are given in Table I. Making use of Eq. (3), the dispersion of

TABLE I. Coefficients of the least-squares fit of the function  $\lambda^3/\rho = A' + B'\lambda^{-2} + C'\lambda^{-4}$  to the data in Fig. 2, where  $\rho$  is the gas density for optimum phase match. The accuracy of  $\lambda^3/\rho$  calculated from the tabulated coefficients is  $\pm 0.2\%$  over the range  $458 \leq \lambda \leq 700$  nm (except for He; see the text). Extrapolation outside this interval will be less accurate. Units are  $\rho$  (mol/m<sup>3</sup>) and  $\lambda$  (m).

Gas	$A'$ ( $10^{-21}$ m <sup>6</sup> /mol)	$B'$ ( $10^{-35}$ m <sup>8</sup> /mol)	$C'$ ( $10^{-48}$ m <sup>10</sup> /mol)
He	0.057 91	0.0875	
Ar	1.063 9	3.386	1.96
H <sub>2</sub>	0.766 2	2.477	2.48
D <sub>2</sub>	0.741 9	2.513	2.20
N <sub>2</sub>	1.146 9	3.528	2.02
O <sub>2</sub>	1.358 8	5.469	19.49

the refractive index may be obtained for standard conditions from the measured  $\lambda^3/\rho$ , through the relation

$$(n_{2\omega} - n_{\omega})_{\text{STP},i} = \frac{\rho_{\text{STP},i} |\Delta k|_{\text{opt}} \lambda^3}{4\pi\lambda^2 \rho} \quad (7)$$

The refractive-index dispersion  $(n_{2\omega} - n_{\omega})_{\text{STP},i}$  is the value that would be obtained for a gas whose density is equal to the density of an ideal gas at standard temperature and pressure of 273.15 K and 1 atm (the subscript  $i$  denotes ideal gas). Finally, making use of the power series expansion for  $n_{\omega} - 1$  one obtains

$$\begin{aligned} (n_{\omega} - n_0)_{\text{STP},i} &= \frac{\rho_{\text{STP},i} |\Delta k|_{\text{opt}}}{4\pi} \left( \frac{1}{3} A' \lambda^{-2} + \frac{1}{15} B' \lambda^{-4} \right. \\ &\quad \left. + \frac{1}{63} C' \lambda^{-6} + \dots \right) \\ &= A \lambda^{-2} + B \lambda^{-4} + C \lambda^{-6} + \dots, \end{aligned} \quad (8)$$

where  $n_0$  is the zero frequency limit of  $n_{\omega}$ . The coefficients  $A, B, C$  have been evaluated using the results in Table I, and the values of  $A, B$ , and  $C$  so obtained are given in Table II. Note that experimental data for  $229 \leq \lambda \leq 700$  nm in effect contributes to the dispersion functions given in Table II, even though the laser frequencies employed were all in the visible. The absolute values of  $n_{\omega}$  may be obtained from Eq. (8) if the refractive index is known at a single frequency.

As may be seen from Fig. 2, the scatter of the data is small compared to the error bars estimated from the accuracy of the instruments employed in the measurements. The estimated overall uncertainty of the measurements varies between  $\pm 0.2\%$  and  $\pm 0.4\%$  for He, but has a nearly uniform value of  $\pm 0.2\%$  for all the other gases at all wavelengths. Thus, the values of  $\lambda^3/\rho$  and  $n_{\omega} - n_0$  evaluated from the coefficients given in Tables I and II should also be accurate to  $\pm 0.2\%$ . This is comparable to the accuracy with which  $n_{\omega} - n_0$  may be obtained from

the best interferometric measurements of  $n_{\omega}$ . The advantage of the present method is greatest where the dispersion is smallest.

The important question of whether there are unrecognized systematic errors in the measurements may be addressed in two ways. The first way is to compare the present results with previous measurements made with different apparatus using the same method.<sup>9</sup> Though  $|\Delta k|_{\text{opt}}$  was twice as large in the previous experiments, those results agree within error bars with the present measurements, with the exception of He where they differ by 0.5% [Fig. 2(a)]. Since the He measurements were made using a different pressure gauge than that used for the other gases, it is possible that this gauge was not meeting the manufacturer's accuracy specification. Another way of addressing the question of unrecognized systematic errors is by comparing the results of the present experiment with the best results of other experimental determinations or *ab initio* calculations. For helium the *ab initio* results of Glover and Weinhold<sup>21</sup> are probably definitive. Their Hylleraas estimate has been used to obtain the dashed curve shown in Fig. 2(a). The discrepancy of 0.3% between our measurements and the theory is consistent with a pressure scale error. Even so, our measurements appear to give the most accurate experimental value for the refractive index dispersion of He.<sup>2-4,26</sup>

For  $\text{H}_2$  the comparison is more satisfactory. Both the results of *ab initio* calculations using explicitly correlated wave functions,<sup>22,23</sup> and the results of accurate experimental measurements<sup>24,25</sup> fall within our error bars. The agreement is illustrated in Fig. 2(b), where the average of theory<sup>22</sup> and experiment<sup>25</sup> is shown by the dashed line. These two sets of results<sup>22,25</sup> are indistinguishable on the scale of Fig. 2(b) for  $\nu^2 < 3 \times 10^8 \text{ cm}^{-2}$ , but at higher frequencies they diverge. At  $\nu^2 = 5 \times 10^8 \text{ cm}^{-2}$  the results of calculation<sup>22</sup> and measurement<sup>25</sup> fall 0.4% above and below their average, respectively. A different aspect of our data also allows a stringent test of the theory.  $\text{H}_2$  and  $\text{D}_2$  were measured together under the same conditions, and because they are so similar, the linearity of the capacitance manometer is sufficient to ensure that the accuracy of the ratio  $\rho_{\text{D}_2}/\rho_{\text{H}_2}$  is set by the precision of the pres-

TABLE II. Coefficients of the refractive index dispersion function  $(n_{\omega} - n_0)_{\text{STP},i} = A\lambda^{-2} + B\lambda^{-4} + C\lambda^{-6}$ , obtained from the coefficients in Table I using Eq. (8) and the value of  $|\Delta k|_{\text{opt}}$  for the present apparatus. The accuracy of  $n_{\omega} - n_0$  calculated from the tabulated coefficients is  $\pm 0.2\%$  over the range  $229 \leq \lambda \leq 700$  nm (except for He; see the text). Extrapolation outside this interval will be less accurate. Units are  $\lambda(\mu\text{m})$ .

Gas	$A$ ( $10^{-6} \mu\text{m}^2$ )	$B$ ( $10^{-8} \mu\text{m}^4$ )	$C$ ( $10^{-10} \mu\text{m}^6$ )
He	0.079 60	0.024 06	
Ar	1.462 4	0.930 8	1.285
$\text{H}_2$	1.053 2	0.680 8	1.626
$\text{D}_2$	1.019 7	0.690 8	1.442
$\text{N}_2$	1.576 5	0.969 9	1.320
$\text{O}_2$	1.867 7	1.503 6	12.758

TABLE III. Comparison of the experimentally measured ratio of phase match densities for  $\text{H}_2$  and  $\text{D}_2$  with the results of an *ab initio* calculation (Ref. 23).

$\lambda$ (nm)	$(\rho_{\text{D}_2}/\rho_{\text{H}_2})_{\text{expt}}$	$(\rho_{\text{D}_2}/\rho_{\text{H}_2})_{\text{calc}}$
694.5	1.0308(7)	1.0310
660.2	1.0306(3)	1.0313
633.0	1.0301(5)	1.0316
610.2	1.0311(6)	1.0319
590.2	1.0304(4)	1.0322
514.7	1.0317(2)	1.0336
496.6	1.0318(3)	1.0341
488.1	1.0317(7)	1.0343
476.6	1.0319(4)	1.0347

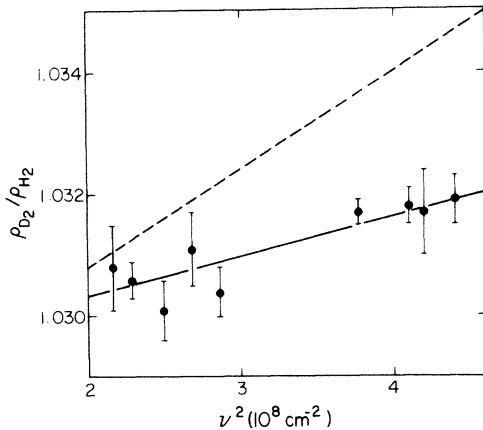


FIG. 3. Ratio of phase match densities for H<sub>2</sub> and D<sub>2</sub>. The solid line is a least-squares fit to the experimental data, while the dashed line is obtained from the results of an *ab initio* calculation (Ref. 23). The zero frequency limit of the line fitted to the measured density ratios is 1.0290. Considering the measured density ratios directly is more accurate than using the ratio of the fitted curves for H<sub>2</sub> and D<sub>2</sub> from Table I.

sure measurements and not by the absolute accuracy of the gauge. As a result we are able to obtain the ratio of phase-match densities with an accuracy of about  $\pm 0.05\%$ . Our experimental results for  $\rho_{D_2}/\rho_{H_2}$  are given in Table III and Fig. 3 along with the values of the phase-match-density ratio calculated from the *ab initio* results of Rychlewski.<sup>23</sup> The calculation is seen to overestimate the dispersion difference between H<sub>2</sub> and D<sub>2</sub> at high frequencies. Accounting for the HD impurity in the D<sub>2</sub> gas would shift the experimental points up by at most 0.02%, and so cannot account for the discrepancy.

For Ar the most accurate previous experimental results in this frequency range appear to be those of Larsén,<sup>3,26</sup> and have been used to obtain the dashed curve in Fig. 2(c). The accuracy of the values of  $\lambda^3/\rho$  calculated from Larsén's data is  $\pm 0.2\%$ , the same as ours and both results agree within the mutual error bars. These results show that the dispersion formula derived from vuv measurements by Chashchina *et al.*<sup>27</sup> does not accurately give the dispersion for Ar at low frequencies. Finally, it appears that the present measurements give the most accurate values of the dispersion of the refractive indices of N<sub>2</sub> (Refs. 2 and 28) and O<sub>2</sub> (Ref. 2) in this frequency range. With some straightforward experimental design changes, the accuracy of the measurements could be improved by a further order of magnitude.

An interesting possibility is that of extending these measurements to shorter wavelengths and into the vuv. In addition to the advantages shared by phase-matched-sum-frequency-generation techniques in general,<sup>6,7</sup> periodic phase matching offers the advantages of high accuracy absolute dispersion measurements using a single laser, with no requirement for favorably situated resonances. The periodic phase-matching condition is only weakly dependent on laser beam parameters and sample absorption. The method may be made insensitive to laser beam focusing and mode structure by making measurements relative to a reference gas.

#### ACKNOWLEDGMENTS

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