Windowless, geometry-independent, phase-matched method for the measurement of the oscillator strengths of Xe, Kr, and Ar

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We have developed a windowless, geometry-independent method for measuring the relative oscillator strengths of gases by observing phase matching in the sum-mixing process in a gas jet. The fundamental beams are focused into the gas jet. There is an optimum pressure for gas behind the inlet valve. The inverse of the optimum pressure is proved to change linearly with the inverse of the energy detuning from the measured level, and the slope of the linearity is proportional to the oscillator strength of the level. Using this method we have measured the relative oscillator strengths of some resonance levels of Xe, Kr, and Ar in the vicinity of the cutoff limit of LiF.

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After the first demonstration of the third-harmonic generation (THG) in a pulsed supersonic jet of xenon [1], the windowless technique of pulsed gas jet has been extensively employed in the generation of broadly tunable vuv and xuv coherent radiations and in high-harmonic generation [2,3]. This technique, furthermore, has led to an alternative method for the measurement of the oscillator strengths below the cutoff limit of LiF (104.5 nm), as well as above the limit, of nonlinear media by nonlinear optical phase matching.

Measurement of atomic oscillator strengths (f values) by nonlinear optical frequency mixing has been demonstrated by a number of authors. These measurements are based on the fact that maximum sum-frequency generation is observed when certain restrictions on the refractive indices are met. The gaseous media are usually contained in a heat-pipe oven [4–7] (HPO) or a gas cell [8,9]. The incident beams are unfocused in order that the intensity of the generated beam is maximum when the wave-vector mismatch is $\Delta k=0$, except that of Kramer, Chen, and Payne [8]. In the work of Kramer, Chen, and Payne, the incident beams are focused into the cell of an Xe-Ar mixture, the output of the four-wave sum mixing (FWSM) maximizes at a certain point of indices mismatch $b\Delta k_{\text{opt}}$ where $b\Delta k_{\text{opt}}<0$. Hence, high gas pressures (>10 Torr) are required to neglect $b\Delta k_{\text{opt}}$, thus satisfying the geometry-independent condition of their method.

For measuring the oscillator strength below 104.5 nm, the methods mentioned above are not easily applied, except that of Ferrell, Payne, and Garrett [9]. The most important reason for this is that windowless environment is required for measurement of FWSM or THG intensities, and neither the HPO nor the gas cell technique works well any more, especially for the case of the gas mixture. Ferrell, Payne, and Garrett have developed a phase-matching method which can measure the absolute f values of atomic resonance levels below 104.5 nm. They adjusted the phase matching in THG by varying the partial pressures of the target gas and the buffer gas, and addressed the position and the width of the phase matching by measuring the multiphotonionization (MPI) signal instead of measuring the output of THG itself. Thus they can measure f values of resonance levels without a window cutoff limit restriction; however, in their work the unfocused fundamental beam is required, and it is not easy for them to measure the levels with small oscillator strengths. Third harmonics of collimated waves often have lower efficiencies near levels with small oscillator strengths, and the MPI signal cannot easily be accurately measured. In this paper we present an alternative method for measuring the oscillator strengths of gas media by observing the phase matching of a focused wave-mixing process in a pulsed gas jet. We will show below that this windowless method is geometry independent and can be used to measure the levels with very small f values.

In order to generate coherent radiation by nonlinear frequency mixing in a gas jet, the fundamental waves must be focused because of the short dimension of the gas jet. For the sum-mixing process $\omega_g=\sum_{i=1}^{n}\omega_i$, the intensity of the generated radiation can be described as [10]

$$I_g \propto N^2|\chi^{(n)}|^2\left[\prod_{i=1}^{n}k_iI_i\right]F^{(n)}, \tag{1}$$

where N is the gas density to which gas distribution is normalized. $\chi^{(n)}$ is the nonlinear susceptibility of the sum-mixing process of the gas. $F^{(n)}$ is the geometric factor, which is dependent on the gas distribution, the confocal parameter, the focus of each fundamental wave, and the wave-vector mismatch between the generated radiation and the induced polarization $\Delta k=k_g-\sum_{i=1}^{n}k_i$. Under fixed experimental configuration, $F^{(n)}$ depends only on $b\Delta k$, with b as the normalized confocal parameter.

If we fix the frequency and the intensity of each incident wave, the output intensity depends only on the gas density in the active region, and will maximize at the optimum point of wave-vector mismatch $b\Delta k_{\text{opt}}$, which makes the optimum gas density different at different wavelengths given by

$$\Delta k_{\text{opt}} = C(\lambda) N_{\text{opt}} , \tag{2}$$

where $C(\lambda)$ is the wave-vector mismatch caused per atom at the wavelength of the generated radiation λ. If λ is close to a resonance level, but still far enough away so that absorption is not important, and much farther from the other resonances of the atom, $C(\lambda)$ can be defined as follows [6]:

$$C(\lambda) = \frac{r_e}{\lambda} \frac{f_{0j}}{\frac{1}{\lambda_{0j}^2} - \frac{1}{\lambda^2}} + C_{\text{others}} , \tag{3}$$

where λ_{0j} and f_{0j} are, respectively, the wavelength and oscillator strength of the resonance level; r_e is the classical radius of the electron; and C_{others} is the contributions to the wave-vector mismatch of both the other discrete levels and the continuum at the generated wavelength minus those of both all the discrete levels and the continuum at the fundamental wavelengths.

Applying Eq. (3) into (2), we get

$$\begin{aligned} \Delta k_{\text{opt}} &= N_{\text{opt}} \left[\frac{r_e}{\lambda} \frac{f_{0j}}{\frac{1}{\lambda_{0j}^2} - \frac{1}{\lambda^2}} + C_{\text{others}} \right] \\ &= N_{\text{opt}} \left[\frac{r_e}{2} \frac{f_{0j}}{\frac{1}{\lambda_{0j}} - \frac{1}{\lambda}} + C_{\text{others}} \right] . \end{aligned} \tag{4}$$

In the derivation an approximation of $\lambda/\lambda_{0j} \sim 1$ has been made, i.e., the generated radiation is close to the resonance level. Given that $b\Delta k_{\text{opt}}$ is constant, and fundamental waves are far below the first resonance level, C_{others} can be regarded as a constant when fundamental waves are tuned in narrow ranges, so that the subsequent tuning of the generated wavelength is in a narrow range near the resonance level and has negligible effect on the photoionization and the population of each level. Under these circumstances we derived

$$\frac{1}{N_{\text{opt}}} = \frac{r_e}{2\Delta k_{\text{opt}}} \frac{f_{0j}}{\frac{1}{\lambda_{0j}} - \frac{1}{\lambda}} + \frac{C_{\text{others}}}{\Delta k_{\text{opt}}} . \tag{5}$$

In the gas jet we cannot know the exact gas density in the active region, but we can measure and control precisely the pressure behind the gas jet valve. The dependence of gas density along the axis of the gas jet on the backing pressure can be described as [11]

$$N = \frac{N_0}{\{1 + [M(x)]^{2/3}\}^{3/2}} , \tag{6}$$

where N_0 is the density of gas in the valve and $M(x)$ is the Mach number, which depends on γ, the ratio of principal specific heats of the gas. For inert gases, $\gamma = \frac{5}{3}$,

$$M(x) = 3.26 \left[\frac{x}{D} - 0.075 \right]^{2/3} - \frac{2}{3.26 \left[\frac{x}{D} - 0.075 \right]^{2/3}} ; \tag{7}$$

for diatomic molecules, $\gamma = \frac{7}{5}$, and

$$M(x) = 3.65 \left[\frac{x}{D} - 0.4 \right]^{2/5} - 0.82 \left[\frac{x}{D} - 0.4 \right]^{-2/5} , \tag{8}$$

where D is the nozzle diameter and x is the distance from the nozzle. For gas outside of the jet axis, the density is also proportional to density behind the gas jet valve with an extra coefficient of $A(x,d)$, where d is the distance of the laser beams from the axis of the gas jet [11]. Substituting Eq. (6) into Eq. (5), we have

$$\frac{1}{N_0} = \frac{A(x,d)}{\{1 + [M(x)]^{2/3}\}^{3/2}} \left[\frac{r_e}{2\Delta k_{\text{opt}}} \frac{f_{0j}}{\frac{1}{\lambda_{0j}} - \frac{1}{\lambda}} + \frac{C_{\text{others}}}{\Delta k_{\text{opt}}} \right] . \tag{9}$$

Thus the inverse of the optimum backing pressure, $1/P$, changes linearly with the inverse of the energy detuning from the resonance, $1/\Delta E$, in wave numbers $\Delta E = 1/\lambda_{0j} - 1/\lambda$; the slope of the linearity is proportional to the oscillator strength of the resonance level. If we have measured the dependence of $1/P$ on $1/\Delta E$ near different resonance levels, by comparing the slopes we can get the relative oscillator strengths. In the comparison all geometry-dependent factors, such as $A(x,d)$, $M(X)$, and Δk_{opt} are canceled; therefore, the method is geometry independent.

Of the many methods previously reported in the literature, this one, together with that of Ferrell, Payne, and Garrett and that of Kramer, Chen, and Payne, is independent of any knowledge of fundamental beams, the gas distribution, and the focal conditions. In our method, furthermore, we do not necessarily neglect C_{others}, as Wynne and Beigang [4] did; thus the sensitivity and accuracy are improved. No window is needed in the experiment; therefore, with our method it is possible to measure the oscillator strengths below 104.5 nm without any special difficulty. We do not require a high enough gas density, as Kramer, Chen, and Payne did, which is more difficult to achieve in the gas jet case. The calibration of $1/\Delta E^2$, which is caused by three-photon resonance of $\chi^{(3)}$, is unnecessary because we address the optimum point by fixing the frequency and adjusting the pressure. This method may be applied to different gases: if γ is the same for those gases, no calibration is needed; if γ is different, a calibration of Mach number should be done. Compared with the method of Ferrell, Payne, and Garrett, the wave-mixing process in our method limits us to work in the region where absorption of the measured level can be neglected. In contrast to their method, THG occurs only when the frequency is close enough to the measured level, thus ionizing the atoms. The fundamental beams in our method are focused; thus the wave-mixing process near the levels with small oscillator strengths is enhanced, and small f value levels can thus also be measured. Moreover, our method holds not only in the third-order wave-mixing process, but also in fifth- and even higher-order processes, but that of Ferrell, Payne, and Garrett can only work in THG. For other

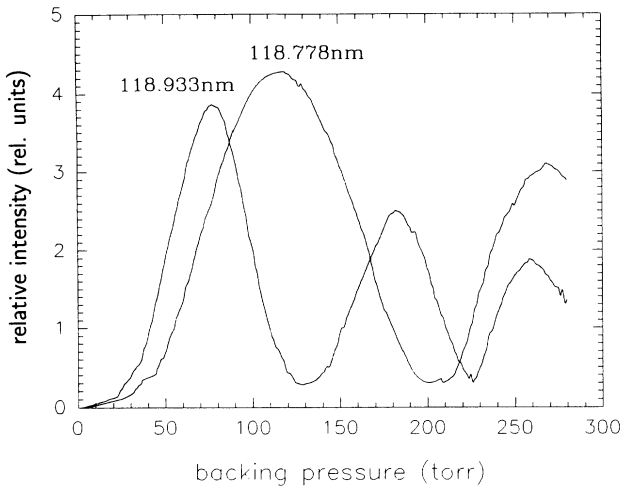

FIG. 1. The typical dependence of vuv intensities on the backing pressure at different wavelengths. The curves were measured in the FWSM near Xe $5d[3/2]_1$ (119.2 nm). The vuv wavelengths of the curves are, respectively, 118.933 and 118.778 nm.

wave-mixing process a theoretical extension should be done. Our method is thus applicable over a greater region; however, because we cannot measure the absolute gas density in the gas jet, we can only get a relative f value, rather than an absolute one.

In the experiment we measured the relative oscillator strengths of Xe, Kr, and Ar by observing the phase matching in nonresonant THG or FWSM processes. The output of a dye laser [Quatel, Datachrome, operation in 4-(dicyanomethylene)-2-methyl-6-(*p*-dimethyl-aminostyryl)-4*H*-pyran (DCM)], which was pumped by the second harmonic of a *Q*-switched Nd:YAG (where YAG denotes yttrium aluminum garnet) laser, was frequency doubled and both the uv and the visible beams were focused by a quartz lens ($f=10$ cm) into a pulsed jet of inert gases. Coherent vuv radiation was generated by THG ($\omega_{vuv}=3\omega_{uv}$) or FWSM ($\omega_{vuv}=2\omega_{uv}+\omega_{dye}$). The power of the dye laser is typically 1–3 MW, and the efficiency of the frequency doubling in BBO (β-BaB_2O_4) is about 10%.

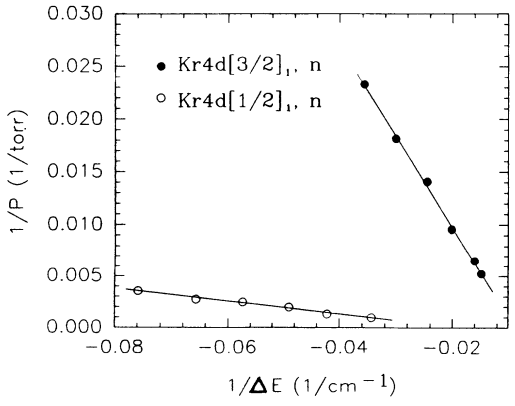

FIG. 2. The dependence of $1/P$ on $1/\Delta E$ for Kr levels of $4d[3/2]_1$ and $4d[1/2]_1$; "n" in this figure means that the data were obtained in the negatively dispersive regions.

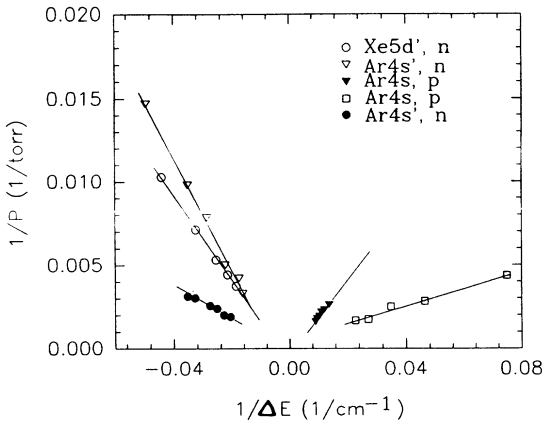

FIG. 3. The dependence of $1/P$ on $1/\Delta E$ for the levels of Xe $5d'[1/2]_1$, Ar $4s'[1/2]_1$, and Ar $4s[1/2]_1$. "n" and "p" mean that the data were measured in the negatively and the positively dispersive regions, respectively.

The vuv radiations were diffracted by a vuv monochromator (Acton Model VM-502) and detected by a windowless electron multiplier (Hamamatsu Model R595). The pulsed gas jet was provided by a homemade piezoelectric valve. The pressure in the valve was measured by using a resistance manometer (HLP03). The measurement had a ±0.5% accuracy in the range of 1–1000 Torr. We measured the dependence of the relative flux on the backing pressure using a flowmeter and found that the flux is precisely proportional to the backing pressure up to 5 atm. To guarantee that the geometry was unchanged between measurements, we traced the doubling crystal, thus eliminating the lateral displacement of the laser beams caused by wavelength tuning.

The typical dependences of vuv intensity on the backing pressure at different wavelengths are shown in Fig. 1, in which coherent vuv radiations were generated by FWSM in the high-energy side of Xe $5d[3/2]_1$, where $\frac{3}{2}$ and 1 are quantum numbers in *jl*-coupling notation. We measured the optimum pressures in THG at different energy detuning near a series of levels, respectively; the energy detuning near each level was scanned in a small range; thus C_{others} varied by less than 3%. The results near Kr $4d[3/2]_1$ and Kr $4d[1/2]_1$ are shown in Fig. 2, in which "n" means that the results are from the negatively dispersive region. The inverse of the optimum pressure shows good linear dependence on the inverse of energy detuning. Comparing the slopes of the two levels, we get the relative oscillator strength of

$$\frac{f_{4d[3/2]_1}}{f_{4d[1/2]_1}}=13.96\ .$$

TABLE I. Relative slopes of Xe $5d'$, Ar $4s$, and Ar $4s'$ measured in THG processes.

Levels	Xe $5d'$	Ar $4s$	Ar $4s'$
Wavelength (nm)	106.82	106.67	104.82
Slopes			
Negatively dispersive	−0.256	−0.088	−0.343
Positively dispersive		0.052	0.218

TABLE II. The relative oscillator strengths of Xe $5d'$ and Ar $4s$ compared to Ar $4s'$.

Authors	$f(\text{Xe } 5d')/f(\text{Ar } 4s')$	$f(\text{Ar } 4s)/(\text{Ar } 4s')$
Present work, negative phase matching	0.746	0.257
Present work, positive phase matching		0.239
Chan *et al.* [12] [high-resolution dipole (e,e)]	0.721	0.250
Geiger [13] (electron impact)	0.804	0.259
Natali, Kuyatt, and Miekzarek [14] (electron impact)	0.669	0.252

The oscillator strengths of these two levels have also been measured by other groups. The data of Chan *et al.* [12], which was obtained with the high-resolution dipole method, gives a ratio of 15.55. The results measured with electron impact by Geiger [13] and Natali, Kuyatt, and Miekzarak [14] give, respectively, 11.8 and 18.6. Noting that the absolute oscillator strength of Kr $4d[1/2]_1$ reported before is much smaller, in the vicinity of 0.0050 (Refs. [12–14]), our result is in reasonable agreement with the previous works [12–14], demonstrating that our method is rather sensitive.

As mentioned above, this method can also be applied to measure the relative oscillator strengths of different gases. Figure 3 shows the relative dependence of $1/P$ on $1/\Delta E$ for Xe $5d'[1/2]_1$, Ar $4s'[1/2]_1$, and Ar $4s[1/2]_1$. The relative slopes are listed in Table I. The derived relative strengths of Xe $5d'$ and Ar $4s$ to Ar $4s'$ are listed in Table II. The value measured in the negatively dispersive region and that in the positive region are consistent within an error of 10%. The ratio of the slopes in the two regions near the same level corresponds to the ratio of the positive to the negative optimum $b\Delta k$ value of the function $(b\Delta k)^2F^{(3)}(b\Delta k)$. In our experiment $b/l \approx 3$, so the calculation of the $(b\Delta k)^2F^{(3)}(b\Delta k)$ function gives a value of -0.47 for this ratio. This deviates from the values measured in the experiment (-0.63 for Ar $4s$ and -0.67 for Ar $4s'$, respectively), perhaps due to the non-Gaussian mode of laser beams and the displacement of the foci from the center of the gas get. Table II also shows the results of the previous measurements by other authors. Generally, ours are in good agreement with theirs.

In the experiment the absolute wavelength of the dye laser was calibrated to a precision of ± 0.02 nm, and the relative wavelength error is ± 0.002 nm. The accuracy in measuring the backing pressures at the vuv intensity maximum is within $\pm 5\%$; thus for the levels with larger f values, the relative slopes are accurate within an error of $\pm 10\%$. However, for the levels with smaller f, such as Kr $4d[3/2]_1$, the error is estimated within $\pm 30\%$. No contribution of dimers was observed to affect the accuracy of the experiments.

In conclusion, we have developed a windowless, geometry-independent technique for measuring the relative vuv and xuv oscillator strengths of gases by observing the phase matching in nonlinear optical processes. The focusing of the fundamental beams makes it possible to measure the levels with small f values. If we know the f value of one resonance level of one gas medium, it is possible for us to measure the oscillator strengths of other discrete levels of this or other gases. Using an achromatic lens to focus the incident beams, so that they are of the same confocal parameter and focused at the same point, by comparing the phase matching between the same order of sum-mixing processes, the oscillator strengths of high levels of atoms will become measurable by this nonlinear optical phase-matching method.

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