

## 4.2 Frequency conversion in gases and liquids

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### 4.2.1 Fundamentals of nonlinear optics in gases and liquids

This chapter covers the properties of a nonlinear medium having spherical symmetry like gases and liquids. They therefore clearly differ from the properties of most solids (see Chap. 4.1).

Lasers have become so powerful these days that one can easily generate various kinds of optical overtones

$$\omega_o = \sum_{i,q} n_i \cdot \omega_i \pm k_q \cdot \omega_{\text{res},q} > 0, \quad (4.2.1)$$

where  $n_i$  and  $k_q$  are some integer (including  $n_i = 0$  or  $k_q = 0$ ), using a suitable nonlinear medium with eigenfrequencies  $\omega_{\text{res},q}$  and an incident laser frequency  $\omega_i$  (conservation of energy).

In case of frequency conversion in gases one generally has  $k_q = 0$  and deals with sum or difference frequency mixing

$$\omega_s = \omega_i \pm \sum_j \omega_j > 0 \quad (4.2.2)$$

which may be enhanced by exploiting suitable resonances of the atomic or molecular gas.

In case of stimulated scattering one generally has  $n_i = 1$ . Then  $\omega_{\text{res},q}$  is a suitable manifold of atomic or molecular (rotational or vibrational) resonances of the gaseous or liquid scattering medium numbered by the index  $q$ . Like in classical spectroscopy the plus sign stands for Stokes processes, whereas the minus sign is responsible for Anti-Stokes processes.

#### 4.2.1.1 Linear and nonlinear susceptibilities

Linear and nonlinear susceptibilities are discussed in [87Vid].

The complex linear susceptibility is given by

$$\chi^{(1)} = \bar{\chi}^{(1)} + i\tilde{\chi}^{(1)} = \frac{1}{\hbar} \sum_a \frac{|\mu_{ag}|^2}{(\Omega_{ag} - \omega)} \quad (4.2.3)$$

with the complex transition frequency

$$\Omega_{ag} = \omega_{ag} - i\Gamma_{ag} \quad (4.2.4)$$

and the dipole moment matrix elements  $\mu_{ag}$  between the states  $|a\rangle$  and  $|g\rangle$ . The nonlinear polarization is

$$\mathbf{P}^{\text{NL}} = \sum_{n=2}^{\infty} \mathbf{P}^{(n)}. \quad (4.2.5)$$

The definition of the electric field amplitude is given by

$$\mathbf{E}(r, t) = \frac{1}{2} \sum_j \mathbf{e}_j \hat{E}(r, \omega_j) \exp(i k_j r - i \omega_j t) + c.c. , \quad (4.2.6)$$

resulting in the definition of the total polarization

$$\mathbf{P}(r, t) = \frac{1}{2} \sum_j \mathbf{e}_j P(r, \omega_j) \exp(-i \omega_j t) + c.c. , \quad (4.2.7)$$

where the  $n$ th-order polarization is given by

$$\mathbf{P}_{\alpha_s}^{(n)}(r, \omega_s) = \frac{n! N}{2^{n-1}} \epsilon_0 \sum_{\alpha_1 \dots \alpha_n} \chi_{\alpha_s \alpha_1 \dots \alpha_n}^{(n)}(-\omega_s; \omega_1 \dots \omega_n) \mathbf{E}_{\alpha_1}(r, \omega_1) \dots \mathbf{E}_{\alpha_n}(r, \omega_n) . \quad (4.2.8)$$

The  $\alpha_s$  are the unit vectors of the spatial coordinates, which may be cartesian, cylindrical, or spherical. The polarization can be expressed in terms of the density matrix [71Han]

$$\langle \mathbf{P}(t) \rangle = N \text{Tr} [\rho(t) \boldsymbol{\mu}] = N \sum_{mn} \rho_{mn}(t) \mu_{mn} , \quad (4.2.9)$$

whose elements are given by  $i \hbar \dot{\rho}_{mn} = [H, \rho]_{mn}$ , where the Hamiltonian  $H = H^0 + H'$  contains  $H' = -\boldsymbol{\mu} \mathbf{E}(t)$ . From a perturbation approach one obtains

$$\chi_{\alpha_1, \alpha_2 \dots \alpha_n}^{(n)}(-\omega_s; \omega_1 \dots \omega_n) = \frac{1}{n! \hbar^n} \sum_{g b_1 \dots b_n} \rho(g) \frac{\langle g | \mathbf{e}_s \boldsymbol{\mu} | b_1 \rangle \langle b_1 | \mathbf{e}_1 \boldsymbol{\mu} | b_2 \rangle \dots \langle b_n | \mathbf{e}_n \boldsymbol{\mu} | g \rangle}{(\Omega_{b_1 g} - \omega_1 - \dots - \omega_n)(\Omega_{b_2 g} - \omega_2 - \dots - \omega_n) \dots (\Omega_{b_n g} - \omega_n)} . \quad (4.2.10)$$

#### 4.2.1.2 Third-order nonlinear susceptibilities

These processes are responsible for the lowest-order frequency conversion in gases such as sum or difference frequency mixing, stimulated scattering processes and photorefractive. For the *degenerate* case the dominant terms in a system of spherical symmetry are [71Han]:

$$\chi^{(3)}(-3\omega; \omega, \omega, \omega) = \chi_T^{(3)}(3\omega) = \hbar^{-3} \sum_{abc} \frac{\langle g | \mathbf{e}_s \boldsymbol{\mu} | a \rangle \langle a | \mathbf{e}_1 \boldsymbol{\mu} | b \rangle \langle b | \mathbf{e}_2 \boldsymbol{\mu} | c \rangle \langle c | \mathbf{e}_3 \boldsymbol{\mu} | g \rangle}{(\Omega_{ag} - \omega)(\Omega_{bg} - 2\omega)(\Omega_{cg} - 3\omega)} , \quad (4.2.11)$$

where the index T stands for the third harmonic generation.

For the *nondegenerate* case we have the general third-order nonlinear susceptibility [62Arm, 71Han]

$$\chi_{\alpha_1, \alpha_2, \alpha_3, \alpha_s}^{(3)}(-\omega_s; \omega_1, \omega_2, \omega_3) = \frac{1}{6 \hbar^3} \sum_{gabc} \rho(g) \frac{\langle g | \mathbf{e}_s \boldsymbol{\mu} | a \rangle \langle a | \mathbf{e}_1 \boldsymbol{\mu} | b \rangle \langle b | \mathbf{e}_2 \boldsymbol{\mu} | c \rangle \langle c | \mathbf{e}_3 \boldsymbol{\mu} | g \rangle}{(\Omega_{ag} - \omega_1 - \omega_2 - \omega_3)(\Omega_{bg} - \omega_2 - \omega_3)(\Omega_{cg} - \omega_3)} \quad (4.2.12)$$

obeying the conservation of energy

$$\omega_s = \omega_1 + \omega_2 + \omega_3 . \quad (4.2.13)$$

### 4.2.1.3 Fundamental equations of nonlinear optics

Maxwell's equations in SI units [62Jac, 87Vid] are given by (1.1.4)–(1.1.7) and the material equations (1.1.8) and (1.1.9), see Chap. 1.1.

With the following three approximations

1. magnetization  $\mathbf{M} = 0$ :  $\mu_0 \mathbf{H} = \mathbf{B} \rightarrow \mu = 1$ ,
2. source-free medium:  $\rho = 0$ ,
3. currentless medium:  $\mathbf{j} = 0$

we get the simplified Maxwell equations

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t} \quad (4.2.14)$$

$$\nabla \times \mathbf{H} = \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}}{\partial t} \quad (4.2.15)$$

resulting in the wave equation

$$\Delta \mathbf{E} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (4.2.16)$$

with the polarization  $\mathbf{P} = \mathbf{P}^L + \mathbf{P}^{\text{NL}}$ . This gives the driven wave equation

$$\Delta \mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} - \mathrm{i} \frac{\epsilon_0}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathbf{P}^{\text{NL}}}{\partial t^2} . \quad (4.2.17)$$

With the plane-wave approximation  $\hat{E}(r, \omega) = \hat{E}(z, \omega)$  and the slow-amplitude approximation

$$\frac{\partial \hat{E}_j}{\partial t} \ll \omega \hat{E}_j, \quad \frac{\partial \hat{E}_j}{\partial z} \ll k \hat{E}_j, \quad (4.2.18)$$

we get the fundamental equations of nonlinear optics

$$\frac{d \hat{E}_j}{dz} = \mathrm{i} \frac{\omega_j}{2 \epsilon_0 c n_j} P_j^{\text{NL}} \exp(-\mathrm{i} k_j z) - \frac{\kappa_j}{2} \hat{E}_j, \quad (4.2.19)$$

where  $\kappa$  is the absorption coefficient and where the total derivative is given by the partial derivatives

$$\frac{d \hat{E}_j}{dz} = \frac{\partial \hat{E}_j}{\partial z} + \frac{n_j}{c} \frac{\partial \hat{E}_j}{\partial t}, \quad (4.2.20)$$

and  $\hat{E}_j$  is a slowly-varying-envelope function in space and time.

### 4.2.1.4 Small-signal limit

In this case the only nonlinear polarization for a medium of density  $N$  is given by

$$P_s^{(3)}(\omega_s) = \frac{3}{2} \epsilon_0 N \chi_T^{(3)}(-\omega_s; \omega_1, \omega_2, \omega_3) E_1 E_2 E_3. \quad (4.2.21)$$

Within the plane-wave approximation one obtains

$$\frac{d \hat{E}_s}{dz} = \mathrm{i} \frac{3 \pi \omega_s}{c n_s} N \chi_T^{(3)} E_{10} E_{20} E_{30} \exp \left\{ \left( -\frac{\kappa_1 + \kappa_2 + \kappa_3}{2} - \mathrm{i} \Delta k \right) z \right\}, \quad (4.2.22)$$

where the wave-vector mismatch is given by the conservation of momenta

$$\Delta k = k_s - k_1 - k_2 - k_3 . \quad (4.2.23)$$

The wave vector  $k_j$  of the  $j$ th wave is given by the refractive index  $n_j$

$$k_j = \frac{\omega_j n_j}{c} . \quad (4.2.24)$$

With the optical depth  $\tau_j = \kappa_j L = \sigma_j^{(1)}(\omega_j) N L$  and the length  $L$  of the nonlinear medium we have

$$\hat{E}_s(L) = i \frac{3\pi\omega_s}{c n_s} N L \epsilon_0 \chi_T^{(3)} E_{10} E_{20} E_{30} \frac{\exp\left(-\frac{\tau_s}{2}\right)}{\frac{\tau_s - \tau_0}{2} - i \Delta k L} \left\{ \exp\left[\frac{\tau_s - \tau_0}{2} - i \Delta k L\right] - 1 \right\} , \quad (4.2.25)$$

where the total optical depth  $\tau_0 = \tau_1 + \tau_2 + \tau_3$ . With the intensity

$$\Phi_j = \frac{\epsilon_0 n_j c}{2} |E_j|^2 \quad (4.2.26)$$

the intensity conversion is given by

$$\frac{\Phi_s}{n_s} = \left[ \frac{6\pi\omega_s}{c^2 n_s} N L \chi_T^{(3)}(-\omega_s; \omega_1, \omega_2, \omega_3) \right]^2 \frac{\Phi_{10}\Phi_{20}\Phi_{30}}{n_1 n_2 n_3} F(\Delta k L, \tau_0, \tau_s) , \quad (4.2.27)$$

containing the general phase-matching factor

$$F(\Delta k L, \tau_0, \tau_s) = \frac{\exp(-\tau_0) + \exp(-\tau_s) - 2 \exp\left(-\frac{\tau_0 + \tau_s}{2}\right) \cos(\Delta k L)}{\left(\frac{\tau_s - \tau_0}{2}\right)^2 + (\Delta k L)^2} < 1 . \quad (4.2.28)$$

#### 4.2.1.5 Phase-matching condition

Maximum conversion efficiency is achieved for conservation of momenta  $k_j$  where  $\Delta k = 0$

$$\omega_1 n_1 + \omega_2 n_2 + \omega_3 n_3 = \omega_s n_s . \quad (4.2.29)$$

In case of the third harmonic generation this gives  $n_1 = n_s$ . Frequency mixing in a two-component system results in

$$\frac{N_a}{N_b} = \frac{\omega_s \bar{\chi}_b^{(1)}(\omega_s) - \sum_{j=1}^3 \omega_j \bar{\chi}_b^{(1)}(\omega_j)}{\sum_{j=1}^3 \omega_j \bar{\chi}_a^{(1)}(\omega_j) - \omega_s \bar{\chi}_a^{(1)}(\omega_s)} . \quad (4.2.30)$$

For the third harmonic generation in a two-component system we have:

$$\frac{N_a}{N_b} = \frac{\bar{\chi}_b^{(1)}(3\omega) - \bar{\chi}_b^{(1)}(\omega)}{\bar{\chi}_a^{(1)}(\omega) - \bar{\chi}_a^{(1)}(3\omega)} . \quad (4.2.31)$$

The frequency mixing in a one-component system is given by:

$$\omega_s \bar{\chi}^{(1)}(\omega_s) = \sum_{j=1}^3 \omega_j \bar{\chi}^{(1)}(\omega_j) . \quad (4.2.32)$$

## 4.2.2 Frequency conversion in gases

The following conditions have to be met for large conversion efficiencies:

1. a large nonlinear susceptibility  $\chi_T^{(3)}$  which may be enhanced by a proper two-photon resonance,
2. large column densities with a proper phase matching,
3. small optical depths for the incident and generated waves to avoid reabsorption.

### 4.2.2.1 Metal-vapor inert gas mixtures

Metal-vapor inert gas mixtures are generally generated in concentric heat pipes because for efficient frequency mixing the phase matching can be accurately and independently adjusted through the partial pressures in the heat pipe [71Vid, 87Vid, 96Vid].

Tables of the multi-wave mixing experiments in different gaseous nonlinear media are arranged according to the elements, Table 4.2.1. For every element the wavelength is given together with the method of generation. The method of generation is indicated where  $(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$ , for example, indicates a two-photon resonance of  $\omega_1$  in the particular atomic or molecular medium and the additional wave  $\omega_2$  can make the resulting radiation tunable.

### 4.2.2.2 Mixtures of different metal vapors

The modified concentric heat pipe [71Vid, 87Vid, 96Vid] is used for phase matching with small partial pressures avoiding strong homogeneous broadening.

In Table 4.2.2 mixtures of different metal vapors are listed.

### 4.2.2.3 Mixtures of gaseous media

For  $\lambda > 106$  nm one prefers gas cells with lithium fluoride windows [87Vid]. For  $\lambda < 106$  nm one should use either pulsed nozzle beams [87Bet] without windows or gas cells with a fast shutter [85Bon].

In Table 4.2.3 mixtures of gaseous media are given.

**Table 4.2.1.** Metal-vapor inert gas mixtures.

Vapor	Wavelength [nm]	Method	Ref.
Na	354.7	$3 \cdot \omega_1$	[75Blo1, 75Blo2, 76Oha]
Na	330.5	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[74Blo]
Na	268 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[84Bol]
Na	231	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[76Bjo]
Na	151.4	$7 \cdot \omega_1$	[77Gro2, 79Mit]
Na	117.7	$9 \cdot \omega_1$	[77Gro1, 79Mit]
Rb	354.7	$3 \cdot \omega_1$	[71You, 75Blo1, 76Pue, 76Oha]
Cs	213.4	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[74Leu, 75War]
Be	121–123	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[79Mah]
Mg	173.5	$2 \cdot \omega_1 + \omega_1 + \omega_1$	[85Hut]
Mg	140–160	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[76Wal, 80Jun, 96Ste]
Mg	143.6 (cw)	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[83Tim]
Mg	115, 121.2, 127	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[81Car, 85Car]
Mg	121–129	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[78McK]
Mg <sup>+</sup>	123.6	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[85Leb]
Ca	200	$3 \cdot \omega_1$	[76Fer]
Ca <sup>+</sup>	127.8	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[75Sor]
Zn	106–140	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[82Jam]
Cd	118.2, 152, 177.3	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2; 3 \cdot \omega_1$	[72Kun]
Cd	128.7–135.3	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[84Miy]
Cd	145.3–171.1	$(\omega_1 + \omega_1)_{\text{Res}} - \omega_2$	[86Miy]
Sr	155.3, 166.7, 169.7, 173.5, 183.5 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[90Nol1]
Sr	184.6, 185.7, 192.0, 195.8, 208.8, 217.9 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[90Nol1]
Sr	171.2, 168.3, 169.7, 190 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[77Bjo, 78Fre]
		$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[90Nol2]
Ca	153.0, 159.5, 161.3, 163.3, 167.0 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[90Nol1]
Ca	169.7, 170.9, 172.3, 173.1, 176.9 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[90Nol1]
Zn	134.5–141.6 (cw)	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[90Nol1]
Cd	138.1–140.3	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[88Sch]
Sr	177.8–195.7	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[74Hod, 75Sor, 76Sor]
Sr	165–166	$(\omega_1 + \omega_2)_{\text{Res}} + \omega_3$	[78Eco]
Sr	192.3	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[80Pue, 81Egg]
Sr	171.2	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[80Eco]
Ba	190–200	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[80Hei]
Hg	109–196	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[83Hil2]
Hg	184.9, 143.5, 140.1, 130.7, 125.9, 125.0	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[81Bok]
Hg	125.1, 183.3, 208.5	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[81Tom]
Hg	124.7–125.5, 122.8–123.5, 117.4–122	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[82Mah, 82Tom]
Hg	120.3	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[76Hsu]
Hg	87.5–105, 99.1–126.8	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[85Her, 83Hil2]
Hg	89.6	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[78Sla]
Hg	132–185	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[86Hil2]
Tl	195.1	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[75Wan]
Eu, Yb	185.5, 194	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[75Sor, 76Sor]

**Table 4.2.2.** Mixtures of different metal vapors.

Mixture	Wavelength [nm]	Method	Ref.
Na + K	2–25 $\mu\text{m}$	$(\omega_1 - \omega_2)_{\text{Res}} - \omega_3$	[74Wyn]
Na + Mg	354.7	$3 \cdot \omega_1$	[75Blo2]

**Table 4.2.3.** Mixtures of gaseous media.

Gas	Wavelength [nm]	Method	Ref.
He, Ne, Ar, Kr, Xe	231.4	$3 \cdot \omega_1$	[67New, 69War]
He	53.2	$5 \cdot \omega_1$	[76Rei, 77Rei, 77She, 78Rei2, 78Rei1]
He	38	$7 \cdot \omega_1$	[77Rei, 78Rei2]
He	82.8, 50, 35.5	$3 \cdot \omega_1$ ; $5 \cdot \omega_1$ ; $7 \cdot \omega_1$	[83Bok]
He, Ne	106.4	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[78Rei2]
He, Ne	88.7	$3 \cdot \omega_1$	[78Rei2, 78Rei1]
He, Ne	76, 70.9, 62.6, 59.1	$4 \cdot \omega_1 \pm \omega_2$	[77She, 78Rei2]
He, Xe	49.7, 35.5	$5 \cdot \omega_1$ ; $7 \cdot \omega_1$	[83Bok]
Ne	53.2, 118.2	$5 \cdot \omega_1$ ; $(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[76Rei, 77She, 78Rei2]
Ne	72.05–73.58, 74.3–74.36	$3 \cdot \omega_1$	[84Hil]
Ar	120.4	$(\omega_1 + \omega_1)_{\text{Res}} + 3 \cdot \omega_1$	[80Din]
Ar	85.7–87.0, 97.4–104.8	$3 \cdot \omega_1$	[83Hil3, 83Mar]
Ar	106.7	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[82Mil]
Ar	102.6–102.8	$3 \cdot \omega_1$	[79Rei, 80Rei]
Ar, Kr	61.6, 53.2	$5 \cdot \omega_1$	[78Rei2, 81Rei]
Ar	57	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[76Hut]
Kr	131.2, 92.3, 92.8, 94.2	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[85Bon]
Kr	121.6	$3 \cdot \omega_1$	[81Hil, 80Lan, 81Bat]
Kr	123.6	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[82Mil]
Kr	112.4, 120.3–123.6	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[79Cot1, 79Cot2]
Kr	131.2	$(\omega_1 + \omega_1)_{\text{Res}} - \omega_2$	[85Bon]
Kr	72.5–83.5; 127–180	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[87Hil]
Kr	121–200	$(\omega_1 + \omega_1)_{\text{Res}} - \omega_2$	[90Mar]
Kr, Xe	71–92	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[89Miy]
Kr, Xe	110–210	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[82Hil]
Xe	84.6–109.5, 155–220	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[82Hil, 82Hag, 83Hil1]
Xe	155	$(\omega_1 + \omega_1)_{\text{Res}} - \omega_2$	[83Hut]
Xe	147	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[82Mil]
Xe	140.3–146.9	$3 \cdot \omega_1$ ; $(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[81Hil, 83Val]
Xe	74.8, 75, 75.2	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_1$	[82Mui]
Xe	125.4, 125.9, 126.1	$(\omega_1 + \omega_1)_{\text{Res}} - \omega_2$	[82Mui]
Xe	101.5, 101.8, 13.0	$(\omega_1 + \omega_1)_{\text{Res}} + \omega_2$	[82Mui]
Xe	163.1–194.6	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[74Kun]
Xe	118.2	$3 \cdot \omega_1$	[83Kun, 76Kun, 82Gan, 83Bok]
Ar, Kr, Xe, CO, N <sub>2</sub>	72; 90.4–102.5	$3 \cdot \omega_1$	[87Pag]
Ne, Ar, Kr, Xe, Hg	60–200	$(\omega_1 + \omega_1)_{\text{Res}} \pm \omega_2$	[86Hil1]
Ar, Xe, CO	74.2, 80.4, 95.1, 98.2, 100.1, 116.5, 117.8, 123.6	$\omega_1 + \omega_1 + \omega_2$ ; $3 \cdot \omega_1$	[89Cro]