1. Introduction

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The importance of lasers to modern scientific research, and their resulting technological applications, has been highlighted in earlier volumes of this *Topics in Applied Physics* series. From initial experiments based on the high intensity of laser radiation emerged studies of the nonlinear properties of materials and useful spectroscopic information on the nature of solids, liquids, and gases. With the development of *tunable* lasers, important advances have been made in the field of high-resolution spectroscopy, as highlighted in Volume 2.

The interaction between electromagnetic radiation and atoms and molecules serves as the basis for using lasers to detect and continuously monitor atmospheric constituents and properties. With the steady increase in industrial activity, power generation, transportation, and other potential sources of air pollution, new techniques for atmospheric monitoring are clearly needed to augment those already in use [1.1, 2]. Most of the present instrumentation is based on sample-extraction methods; there are some notable exceptions, however, involving correlation [1.3], dispersive [1.4], and multi-spectral [1.5] techniques, which involve incoherent (non-laser) electromagnetic radiation. With lasers, however, one generally has greater flexibility of operation and can monitor a wider variety of pollutants due to higher resolution.

All of the basic laser techniques which had been proposed for monitoring atmospheric gases and particles have now been shown to work experimentally. The purpose of this volume is to present a unified, tutorial discussion of these techniques, their applications, and their limitations. Included are examples of results obtained so far (to 1976), so that the reader can evaluate the present capabilities of laser monitoring for specific applications and make extrapolations to the future when improved equipment is available. The reader is also referred to a monograph edited by DERR [1.6] which contains a large number of papers on remote sensing up to 1972, and a recent study by WRIGHT et al. [1.7].

This volume covers the application of laser techniques to the detection and continuous monitoring of particulate matter, aerosols, atoms, and molecules in the atmosphere. The term "atmosphere" is taken to encompass the troposphere (or lower atmosphere) which is between ground level and the tropopause at 10–15 km altitude, and the stratosphere (lower portion of the upper atmosphere) which is bounded below by the tropopause, and extends upward to 28–30 km. Monitoring of pollutants near the ground level is necessary in order to determine the quality of the air we breathe; and both ground-level and higher-level monitoring (at least to the inversion layer height) must be performed in order to develop mathematical models for predicting air quality for varying circumstances. Stratospheric gases and particles affect us in a less direct, but equally-important way; and laser techniques should prove to be especially useful for continuous surveillance of the stratosphere [1.8]. Other important atmospheric parameters, such as temperature and wind velocity, can also be measured remotely by lasers.

Considering the many types of monitoring instruments now available commercially, why should laser instrumentation be developed for atmospheric monitoring? This question is addressed in Chapter 2, which begins with a discussion of the present structure of the atmosphere and continues with an overview of the present capabilities of laser monitoring instrumentation. Remote sensing is not generally considered to be a substitute for point sampling, but an adjunct to it. Nevertheless, in some cases, remote sensing represents the only economical or technically feasible technique. An important application of laser monitoring is expected to be in the area of surveillance to check compliance with source emissions regulations. Chapter 2 considers this application and others which appear most promising for the future.

Chapter 3 describes in detail the transmission of laser radiation through the atmosphere. Not all wavelengths which would be optimal for detecting certain pollutants can be used due to strong absorption by normal atmospheric gases (this restriction is not as severe at high altitudes); and since the laser beam must sometimes travel distances of several km or more, careful consideration must be given to absorption, scattering, nonlinear effects, turbulence, and scintillation. For some laser techniques the monitoring process depends upon one or more of these interactions, and Chapter 3 can provide useful information for optimizing them.

The most advanced laser monitoring technique has been lidar (laser radar), which involves the detection of particles and aerosols by measurement of the laser radiation scattered by them. Lidar systems are currently in use in many countries, and applications include the mapping of particles for operational meteorology, atmospheric research, as well as air pollution studies. Chapter 4 concentrates on these lidar applications involving backscattered laser radiation, and also covers the newly-emerging technique using tunable lasers in a differential-absorption lidar system to remotely measure gaseous pollutants.

With more sophisticated instrumentation it is possible to detect characteristic shifts in the wavelength of the backscattered radiation due to specific scattering molecules. This phenomenon, known as Raman scattering, can be used to monitor a variety of gases using a single, fixed-frequency laser, as discussed in Chapter 5. The Raman-scattering cross sections are not usually large, however, so this technique will probably be limited to major atmospheric constituents and source monitoring. Chapter 5 also covers detection of atoms and molecules by induced fluorescence using a tunable laser, where the cross sections are typically higher than for Raman scattering, but where application is generally limited to higher altitudes (lower pressures) where quenching of the fluorescent signal is reduced.

The most sensitive laser technique is based on the principle of resonance absorption, which occurs when the laser radiation is at the same wavelength as a major absorbing transition of the molecular species to be detected. This technique has already provided very high specificity in point sampling applications. It has also been the basis for instrumentation for *in situ* source monitoring (without taking a sample) and for long-path (to several km) ambient-air monitoring. The wide variety of applications of the absorption technique, involving detection of laser power returned from remote retroreflectors, buildings, and even natural foliage, is described in Chapter 6.

One unique characteristic of narrow-linewidth laser radiation is that it can be very sensitively detected by heterodyne techniques. Heterodyne detection permits the monitoring of backscattered radiation from non-cooperative targets and the passive, single-ended, remote monitoring of gases using their own emission lines. Chapter 7 presents a thorough treatment of all phases of heterodyne detection, with several examples from experiments already performed. It concludes with a discussion of expected future developments.

Thoughout this book an attempt has been made to achieve uniformity both in the text and in the symbols used. Following conventional usage, the terms "wavenumber" and "frequency" are used interchangeably, but the individual symbols are quite explicit in their meaning. A complete list of the symbols used, their definitions, and typical units, follows:

Glossary of Symbols (Typical Units)

a	Particle radius [μm], laser beam radius [mm]
$\hat{a}$	Anisotropic part of polarizability tensor
a_{ij}	Oscillator strength between states i and j
A	Area [cm^2, m^2]

A_{ij}	Einstein coefficient (transition probability) between states i and j [s^{-1}]
$A(\nu)$	True spectral absorption
$A'(\nu)$	Measured spectral absorption
$\mathscr{A}$	Absorption function
b_j	Zero-point amplitude of j-th vibrational mode [$erg^{1/2}/s$]
B	Bandwidth [Hz]
$\boldsymbol{B}$	Molecular rotational constant [cm^{-1}]
$\boldsymbol{B}'$	Combined scattering parameter in differential absorption equation
$B(\nu, T)$	Brightness [W/cm^2-Hz-sr]
c	Speed of light in vacuum (2.998×10^{10} cm/s)
c_j	Relative line strength
c_p	Pressure-broadening coefficient [cm^{-1}/atm]
$C_E(\tau)$	Autocorrelation function for constant-amplitude field with random phase fluctuations [W/cm^2]
$C_i(\tau)$	Autocorrelation function for current [A^2]
C_n^2	Atmospheric structure constant [$m^{-2/3}$]
C_v, C_p	Specific heat at constant volume, pressure [cal/gm-K]
C_D	Photodiode capacitance [F]
d_c	Coherence diameter for heterodyne detection [mm]
D	Diameter [cm]
D_c	Diameter of collecting aperture [cm]
D_p	Particle diameter ($=2a$) [μm]
D_{LO}	Diameter of local oscillator beam [mm]
D_{aa}	Sum diameter for optical collisions $a \leftrightarrow a$ [μm]
D_{ab}	Sum diameter for optical collisions $a \leftrightarrow b$ [μm]
$D_s(d)$	Variance of phase difference over diameter of transmitting aperture
D_λ^*	Detectivity [$cm\text{-}Hz^{1/2}\text{-}W^{-1}$]
e	Electronic charge (1.602×10^{-19} C)
$\mathscr{E}$	Electrical field strength [V/cm]
$E(t)$	Complex optical field [$W^{1/2}$ cm^{-1}]
$E_S(t)$	Signal optical field [$W^{1/2}$ cm^{-1}]
$E_{LO}(t)$	Local oscillator optical field [$W^{1/2}$ cm^{-1}]
E_i	Energy level of quantum state i [eV]
E_t	Threshold illuminance, eye detectability [W/cm^2]
E_N	Noise energy per quantum mode [erg]
f	Frequency [Hz]
Δf	Width (FWHM) of optical power spectrum [Hz]
f_p	Laser pulse repetition frequency [Hz]
f_t	Turbulence fluctuation frequency [Hz]
f_{LO}	Local oscillator frequency ($=c\nu_{LO}$) [Hz]
$f(a)$	Particle size distribution function [cm^{-1}]
F	Coefficient in photocurrent equation [cm^2/W-s]
g	Gravitational force on unit mass [dyne]
g_i	Degeneracy of i-th vibrational mode
g_M	Transmission factor for scanning Michelson interferometer
$g^{(1)}(\tau)$	Normalized first-order correlation function
$g^{(2)}(\tau)$	Normalized second-order correlation function
G	Gain
G_p	Geometrical cross-section per unit volume for particles [cm^2/cm^3]
G_q	Density of quantum modes [cm^{-3}]
G_D	Incremental shunt conductance of photodiode [mho]
h	Planck's constant (6.625×10^{-34} J-s)

$\hat{i}$	Isotropic part of polarizability tensor
$i(t)$	Photocurrent [A]
$i(\omega)$	IF photocurrent [A]
i_{LO}	Local oscillator current [A]
$I(\nu, t)$	Laser beam intensity [W/cm^2]
$\bar{I}$	Time average of optical field intensity [W/cm^2]
$I_s(\lambda, \theta)$	Intensity of scattered radiation of wavelength λ, at angle θ from direction of propagation [W/cm^2]
$I_S(\nu)$	Radiation intensity at top of atmosphere [W/cm^2]
$(\delta I_F)^2$	Relative variance of intensity fluctuations in the Fresnel zone of diffraction of a collimated beam
$(\delta I_R)^2$	Relative variance of intensity fluctuations due to random refraction
$I'(\nu)$	Intensity per unit spectral interval [W/cm^{-1}]
J	Rainfall rate [mm/h]
J	Rotational quantum number
k	Boltzmann's constant (1.380×10^{-23} J/K)
$k(\nu)$	Differential absorption coefficient [cm^{-1} ppm^{-1}]
$\mathbf{k}$	Wave vector ($k = 2\pi/\lambda$) [cm^{-1}]
K'	Optical system efficiency
$K(\nu, \psi)$	Total atmospheric absorption at wavenumber ν and angle ψ with respect to zenith
ℓ	Inter-electrode detector lead spacing [mm]
ℓ_c	Coherence length of laser radiation [m]
L	Depth of pollutant layer or cloud [m]
L	Cell length [cm]
L_p	Spatial pulse length ($= c\tau_p/2$) [m]
m	Mass of molecule or particle [gm]
m	Complex refractive index ($= n - i\kappa$)
m	Temperature exponent of Lorentzian linewidth
m_e	Mass of electron (9.109×10^{-31} gm)
M	Molecular weight [gm]
$\hat{n}$	Relative refractive index ($= n_1/n_2$)
n_e, n_B	Effective transducer noise due to electronics, Brownian molecular motion [atm/Hz$^{1/2}$]
n_0, n_r	Number of photons emitted, received per pulse
n_b, n_d	Number of photoelectrons due to background, dark current
$\bar{n}_k$	Average number of photons in quantum state k
N	Number density of molecules or particles [cm^{-3}]
$N'_p(a)da$	Concentration of particles with radii between a and $a + da$ [cm^{-3}]
N_i	Concentration of molecules in state i [cm^{-3}]
N_0	Number density of air molecules per atmosphere pressure at 15° C (2.55×10^{19} cm^{-3} atm^{-1})
N_A	Avogadro's number (6.022×10^{23} mol^{-1})
NEP	Noise-equivalent power [W/Hz$^{1/2}$]
p	Pressure [atm, Torr]
$p(I)$	Intensity probability distribution function
P_0, P_r	Transmitted, received laser power [W]
$P(\theta)$	Phase function for angular dependence of scattered radiation
P_s	Scattered power [W]
$P_i(\omega)$	Power spectrum of photocurrent [A^2/Hz]
P_{LO}	Local-oscillator power [W]
P_S	Signal power [W]

δP	Differential absorbed power [W]
P_{Ω}	Scattered power per unit solid angle [W/sr]
q	Halfwidth parameter in Γ-distribution of particle sizes
q_j	Fluorescence quenching coefficient [atm^{-1}]
q_n	Coefficient in intensity fluctuation equation
Q	Total partition function
Q_j	Normal coordinate of j-th vibrational mode
Q_A	Wave parameter of aperture ($=ka^2/z$)
Q_B	Total backscatter efficiency [sr^{-1}]
Q_F	Fluorescence quenching factor
Q_S	Total scattering efficiency
Q_W	Water content [$\mathrm{g/m^3}$]
r	Radial distance [m]
R	Range [m]
$\mathscr{R}$	Detector responsivity [V/W]
R_{ij}	Transition dipole matrix element between states i and j [$\mathrm{erg}^{1/2}\,\mathrm{cm}^{3/2}$]
R_o, R_s	Mixer IF output, series resistance [Ohm]
R_p	Radius of phase-front curvature [m]
R_{IF}	IF amplifier input impedance [Ohm]
S	Integrated spectral line intensity [cm]
S_{ij}	Transition intensity between states i and j [cm]
$S(R)$	"S-value" for lidar performance
$(S/N)_{PC}$	Signal-to-noise ratio for pulse-gated photon-counting system
$(S/N)_{BI}$	Signal-to-noise ratio for boxcar integration detection system
t	Time [s]
t_s, t_r	Sampling, transit time [s]
Δt	Lifetime for re-emission [s]
T	Absolute temperature [K]
$T(\nu)$	Transmittance
$\mathscr{T}$	Transmission function
T'	Combined absorption parameter in differential-absorption equation
T_s	System noise temperature [K]
T_A, T_M, T_{IF}	Antenna, mixer, IF-input noise temperature [K]
$(\Delta T)_m$	Minimum detectable temperature change for an ideal radiometer [K]
$(\Delta T')_m$	Minimum detectable temperature change for an actual radiometer [K]
$U(\nu)$	Apparatus spectral transmission function [cm]
v	Vibrational quantum number
v_r	Radial velocity of scatterers [m/s]
$v_{\perp}$	Wind velocity perpendicular to laser beam [m/s]
V	Volume [$\mathrm{m^3}$]
V_L	Visual range of lights (at night) [km]
V_M	Meteorological range [km]
w_j	Nuclear spin weight
$W^{(1)}(t)$	Rate of photocarrier generation [s^{-1}]
$W^{(2)}(t)$	Joint probability of photocarrier generation between t and $t+\tau$ [s^{-2}]
x	Dimensionless particle size parameter ($=2\pi a/\lambda$)
y	Altitude-pressure variable ($=-\ln p$)
$Y(R)$	Geometrical factor to account for overlap of transmitted and received beam paths
z	Pathlength variable [km]
Z	Specific distance [km]
α	Attenuation, extinction coefficient ($=4\pi\kappa/\lambda$) [cm^{-1}, km^{-1}]

α_s, α_a	Extinction coefficients due to scattering, absorption [cm^{-1}]
α_M, α_R	Extinction coefficients due to Mie, Rayleigh scattering [cm^{-1}]
α_p, α_g	Attenuation coefficient due to particles, gases [cm^{-1}, km^{-1}]
β	Volume backscattering coefficient [m^{-1} sr^{-1}]
β	Fraction of laser energy transferred into translational energy in an opto-acoustic cell [$=\frac{3}{2}(C_p/C_v-1)$]
β_R	Rayleigh volume backscattering coeffcient [m^{-1} sr^{-1}]
γ	Halfwidth (HWHM) of spectral line at half maximum absorption coefficient [cm^{-1}]
γ_D, γ_L	Doppler, Lorentzian halfwidths [cm^{-1}]
γ_0	Halfwidth characteristic of apparatus spectral resolution [cm^{-1}]
Γ	Full spectral width (FWHM) of line at half maximum absorption coefficient [cm^{-1}]
δ	Thermal diffusivity ($=\kappa/\varrho_m C_v$) [cm^2/s]
δ	Scattering depolarization ratio
δ_p	Aerosol scattering phase function asymmetry coefficient
$\delta^2_{\ln I}$	Variance of log intensity
δ^2_I	Variance of intensity
δ^2_d	Variance of shifts of laser beam position
Δ	Beam overlap parameter [cm]
ε	Emissivity
$\zeta(z)$	Scattering ratio
$\zeta_z(v)$	Fraction of radiation reaching top of atmosphere from lower altitude z
η	Detector quantum efficiency
η_F	Fluorescence efficiency
θ	Angular separation, commonly scattering angle [rad]
κ	Index of absorption (imaginary part of refractive index, m)
κ	Thermal conductivity [W/cm-K]
$\kappa(y)$	Weighting function [$=d\zeta(y)/dy$]
λ	Wavelength [µm, nm]
Λ	Junge model curve-fitting parameter
Λ	Resonance Raman enhancement factor
$\boldsymbol{\mu}$	Electric dipole moment vector [C-m]
μ	Chemical potential energy [erg]
μ_h	Hole mobility [cm^2/V-s]
μ_S, μ_N	Photomultiplier noise factors for signal, dark current pulses
v	Wavenumber (or "frequency") of electromagnetic radiation ($=f/c$) [cm^{-1}]
v_0	Wavenumber at line center [cm^{-1}]
v_{ij}	Transition wavenumber between states i and j ($\|E_i-E_j\|/hc$) [cm^{-1}]
v_j	Wavenumber corresponding to j-th vibrational mode [cm^{-1}]
δv	Laser frequency modulation amplitude [cm^{-1}]
ξ	Parameter in Junge scattering equation ($=\Lambda-2$)
ξ	Optical efficiency
ξ	Laser power distribution parameter
ξ_S, ξ_N	Counting efficiency for photoelectron signal, dark current pulses
ϱ	Reflectivity
$\boldsymbol{\varrho}$	Transverse coordinate vector [mm]
$\varrho(f)$	Spectral energy density [erg/cm^3-Hz]
ϱ_m	Mass density [gm/cm^3]
$\sigma(v)$	Absorption cross-section per molecule or atom [cm^2]
σ_s, σ_a	Cross section for scattering, absorption [cm^2]
σ_p, σ_g	Cross section due to particles, gases [cm^2]

σ_M, σ_R	Cross section due to Mie, Rayleigh scattering [cm^2]
$(\sigma_a)_T$	Total absorption cross section [cm^2]
$\sigma_B(a, m, \lambda)$	Backscattering cross-section for particles of radius a refractive index m, at wavelength λ [cm^2]
$d\sigma/d\Omega$	Differential cross section [$=\sigma(\theta, \phi)$] [cm^2/sr]
τ	Optical depth
τ	Lifetime, time constant [s]
τ_p	Pulse length [s]
τ_t	Thermal relaxation time [s]
ϕ	Polarization angle [rad]
ϕ	Generalized aerosol parameter
$\phi(t)$	Time-dependent phase angle [rad]
ψ	Angle of sun with respect to zenith [rad]
ψ_i, ψ_j	Eigenfunctions for i-th, j-th molecular states
ω	Circular frequency [rad/s]
ω_c	Photodiode rolloff frequency [rad/s]
Ω	Solid angle [sr]
Ω_r	Receiver field of view [sr]

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