

Laser Technology

LASER TECHNIQUES OF ATMOSPHERIC PROBING

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A review of laser techniques of atmospheric probing is presented. The various interactions of laser energy with the atmospheric medium, namely, Rayleigh scattering, Mie scattering, Resonance scattering, Raman scattering and absorption, are described. An outline of the basic lidar technique for the measurement of atmospheric density and aerosol characteristics is given. The DASE technique for the measurement of atmospheric minor constituents and the Raman scattering method for the determination of atmospheric temperature are discussed.

INTRODUCTION

THE invention of laser by Maiman in 1960 and the giant pulse laser by McClung and Hellwarth (1962) revolutionized the optical exploration of the atmosphere. Laser essentially provides high energy in a very narrow spectral bandwidth and giant pulse laser gives very narrow pulses of the order of nanoseconds. These characteristics give rise to a great improvement in signal to noise ratio and a high resolution in range compared to those possible by other optical sources for atmospheric probing. Also, laser makes the spectroscopic methods like Raman scattering, resonance absorption readily applicable to atmospheric probing.

Basically, laser techniques of atmospheric probing use laser as the transmitter either in a pulsed or C.W. mode. The transmitted energy is directed into the atmosphere by a suitable optical system. The atmospheric molecules and aerosols cause scattering and absorption of the laser energy. A small fraction of the energy scattered back is collected by a suitable optical system and is detected by a photo-device. The detected signal strength contains information on the presence, range and concentration of the atmospheric scatterers and absorbers. The laser system is generally referred to as LIDAR, acronym for Light Detection and Ranging. Lidars are used in monostatic or bistatic mode depending upon the application.

Laser techniques have been successfully applied for the determination of atmospheric densities, temperature, densities of atmospheric minor constituents, aerosol characteristics and lower tropospheric wind velocities. Some of these techniques which are well established are discussed in this review.

ATMOSPHERIC INTERACTIONS WITH LASER ENERGY

The atmospheric interactions of importance in determining the received lidar signal are : (i) Mie scattering; (ii) Rayleigh scattering; (iii) Resonance scattering; (iv) Raman scattering; and (v) Resonance absorption. The first two scattering processes are elastic processes in which no appreciable energy exchange takes place

between the scattering particle and the incident photons. Thus, both the Mie and Rayleigh scattering occur at the incident wavelength. Mie scattering takes place when the dimensions of the scattering particle are close to or larger than the wavelength of the incident radiation, whereas Rayleigh scattering takes place when the size of the particle is much less than the wavelength. Thus Mie and Rayleigh scattering apply to aerosols and atmospheric gas molecules respectively. Resonance scattering is a process in which excitation of an atom or a molecule at a wavelength of strong absorption produces simultaneous emission of a photon at a wavelength equal to or in close proximity with the exciting wavelength. This results in a significant enhancement in the scattering cross section compared to the Rayleigh scattering cross section. In the Raman scattering, the frequency of scattered radiation is shifted with respect to the excitation frequency, the amount of shift being characteristic of the scattering molecule. The scattered radiation shifted to the lower frequency side of the excitation frequency is called the Stokes scattering and the one to the higher frequency side is called the anti-Stokes scattering. But the probability of occurrence of anti-Stokes scattering is far less than Stokes scattering at normal atmospheric temperature. So, in general, Stokes-Raman scattering is used in atmospheric probing by lasers. In resonance absorption, when the excitation wavelength is within the absorption line of any particular molecular constituent, the excitation radiation is absorbed, reducing its strength.

Typical scattering cross sections per particle for various scattering mechanisms are given below.

<i>Scattering mechanism</i>	<i>Cross section</i> (cm ² /ster)
Mie	10 ⁻²⁷ to 10 ⁻⁸ (depending on the size)
Rayleigh	10 ⁻²⁷
Resonance	10 ⁻¹⁵ (for Na)
Raman	10 ⁻²⁹ to 10 ⁻³⁰

BASIC LIDAR EQUATION

In a lidar operated in monostatic pulsed mode, the number of photoelectrons converted from the received back-scattered photons from an altitude interval z and $z + \Delta z$ is given by

$$N_r = \frac{N_i \cdot n \sigma \cdot \Delta z \cdot E \cdot A_r \exp \left[-2 \int_0^z \alpha dz \right]}{z^2} \quad \dots(1)$$

where N_i = number of photons in the transmitted pulse,

n = number density of the scatterers,

σ = backscatter cross section,

$\Delta z = C \tau/2$,

C = velocity of light,

τ = transmitted pulse duration,

E = efficiency of the optics and the quantum efficiency of the photomultiplier,

A_r = effective area of the receiving mirror,

α = volume extinction coefficient,

$$= \alpha_R + \alpha_M + \alpha_{ab}$$

α_R = extinction due to Rayleigh scattering,

α_M = extinction due to Mie scattering,

and α_{ab} = extinction due to absorption.

In eqn. (1), the term $\exp[-2 \int_0^z \alpha dz]$ gives the two-way transmission where $\int_0^z \alpha dz$ is the optical depth. σ , the backscatter cross section depends on the nature of the received signal, i.e., whether the signal received is Rayleigh or Mie, or Raman or resonance scattered signal. $n\sigma$ is generally referred to as the volume backscattering (β). The product of N_t , E and A_r can be termed as system constant 'S'.

RAYLEIGH AND MIE SCATTERING METHOD

In the lidar, for signals received at the exploring wavelength, the return signal is due to Rayleigh and Mie backscattering from the atmospheric gas molecules and aerosols respectively. In this case, the term $n\sigma$ in eqn. (1) is given by

$$n\sigma = n_a \sigma_M + n_g \sigma_R \quad \dots(2)$$

where n_a and n_g are the aerosol and the total atmospheric gas number densities respectively and σ_M and σ_R are the Mie and Rayleigh backscattering cross sections respectively.

The received number of photoelectron counts are calculated using eqns. (1) and (2) as a function of height and is shown in Fig. 1. The results shown are for the Lidar under fabrication in the Space Physics Division, VSSC. The laser transmitter is ruby laser operated at $.6943\mu$ which is outside the absorption line/band of the atmospheric constituents so that the total extinction coefficient (eqn. 1) consists of only two terms α_R and α_M . The other parameters used are as follows:

Laser energy	5J
τ	10/ μ sec
E	0.0077
A_r	0.0706 m ²
Photomultiplier	EMI 9659

For aerosols the model given by Elterman (1964) for clear air condition is used and for atmospheric densities, the model by Sasi and Gupta (1979) is used.

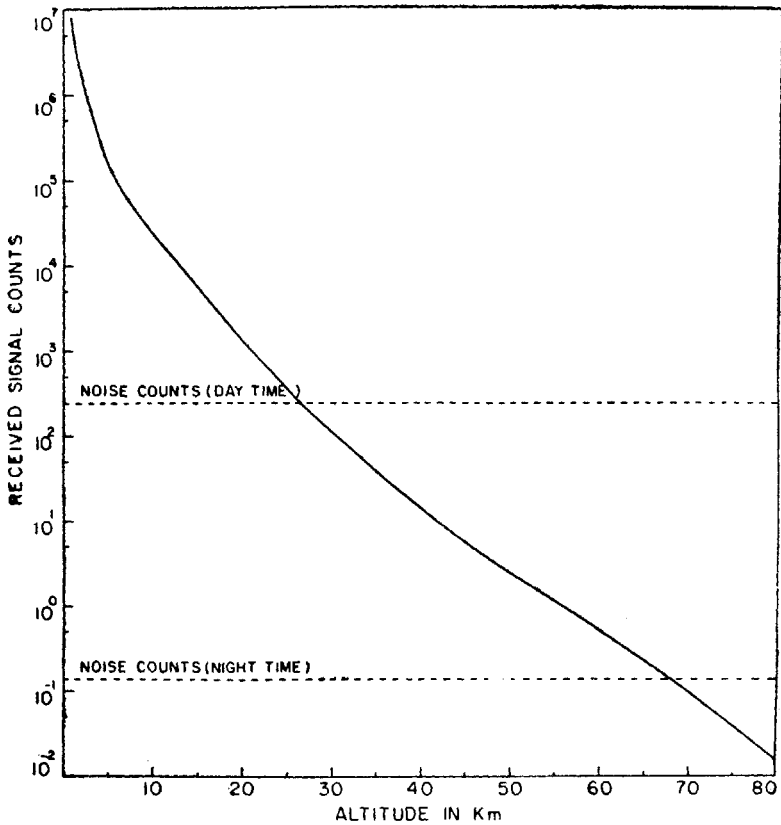


FIG. 1. Ruby lidar received signal counts vs altitude for Mie and Rayleigh scattering.

The total noise level which comprises of the background sky brightness and photomultiplier dark current noise is also shown in Fig. 1 for both night and daytime conditions. In the night time, background noise is not a problem even upto altitudes of 50km. As the signal goes down rapidly with altitude, in order to reduce the errors at these altitudes signal counts from a number of successive transmitted pulses are to be summed up so as to reduce the error which is proportional to square root of the number of counts. For daytime conditions, the signal to noise ratio becomes unity at an altitude of about 26km. For the aerosol model assumed, the aerosol contribution to the total backscattered signal is significant upto 4km.

The lidar equation contains two unknowns namely $(n\sigma)$ and $\exp[-2 \int_0^z \alpha dz]$ for a return signal from a particular altitude. So, both n and the transmission term cannot be determined uniquely without the aid of supplemental information or some *a priori* knowledge about the relation between $n\sigma$ and α . One of the most direct methods of solving the problem is by slant path technique (Reagan &

Schotland, 1975). This technique is based on the assumption that the atmosphere is horizontally homogeneous over the region probed by the lidar. In this method, lidar returns from the same horizontal level at different zenith angles are recorded. From an analysis of these observations, the transmission term and the product of the system constant and $n\sigma$ can be determined. The system constant can be evaluated by carrying out the slant path method for an altitude at which the contribution to $n\sigma$ due to aerosols is negligible compared to that due to the molecules; or, it can be obtained by the direct target method (Hall & Ageno, 1970). For the determination of aerosol characteristics, an assumption that the ratio of the volume backscatter coefficient to the extinction coefficient is a constant in the altitude range of interest is to be made (Fernald *et al.*, 1972). Experimental results on aerosols reveal that this assumption is fairly valid (Blifford, 1970; and Blifford & Ringer, 1969). More information on aerosols can be obtained by using a multiwavelength lidar (Iwasaka *et al.*, 1976). Lidar, operated in a bistatic mode, permits angular scattering measurements to be made on aerosols (Reagan & Herman, 1970). This provides additional information leading to a better determination of the aerosol size distribution and refractive index than is possible with a monostatic lidar (Ward *et al.*, 1973).

The frequency spectrum of the Rayleigh backscattered signal from molecules will be broad because of the wide range of Doppler shifts due to random, high velocity molecular motions. On the other hand, the spectrum of the Mie scattering signal from aerosols will be narrow owing to their larger size. As the aerosol scatter signal will be stronger than the molecular scatter signal in the first few kms, from the ground (depending on the aerosol number densities), the aerosol scatter component can be used to measure the Doppler shift of the return signal which gives the line of sight wind component (Fiocco *et al.*, 1971). The doppler shift can be measured by heterodyne detection or by spectrometric approach at ranges of several hundred metres. Using spectrometric approach, for night time a wind accuracy of 0.27m/s has been reported (Beneditti-Michelangeli *et al.*, 1972).

Atmospheric density measurements from stratospheric altitudes to 80–100km have been carried out using very high power ruby lidars (Kent *et al.*, 1967; Sandford, 1967a, b; Clemesha *et al.*, 1967; and Kent & Wright, 1970). Atmospheric temperature has been determined from density measurements using hydrostatic equation (Sandford, 1967a, b).

Using resonant scattering technique, with Rhodamine 6G dye laser radar system (Bowman *et al.*, 1969), density of sodium was obtained at mesospheric altitudes. This technique is well suited for determining the concentrations of atomic minor constituents because of the much greater cross sections compared to the Rayleigh scattering. The resonant scattering cross section for molecules is not usually expected to be as large as it is for atoms; thus this method is not advantageous for detection of molecular minor constituents.

DIFFERENTIAL ABSORPTION TECHNIQUE

This technique provides a range resolved measurement of specific gaseous constituents at appreciable ranges and with potentially high sensitivity. It makes

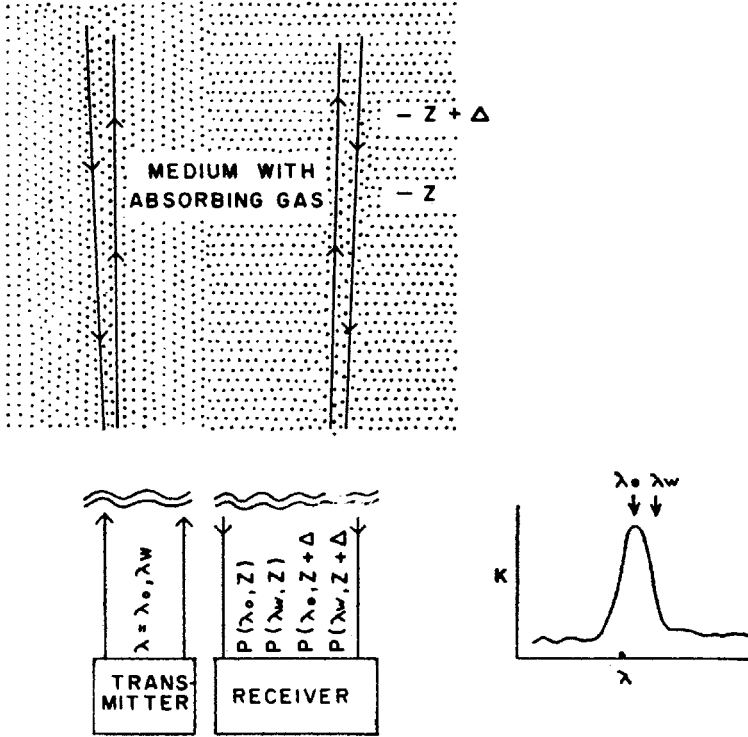


FIG. 2. Principle of DASE method.

use of the term $\exp \left[-2 \int_0^z \alpha dz \right]$ in the lidar equation. Fig. 2 illustrates the principle of this method (referred to as Differential Absorption of Scattered Energy, DASE technique). This method makes use of the very strong wavelength dependence of the gaseous adsorption cross section in the vicinity of an absorption line or within an absorption band, together with the elastic backscatter return from the aerosols and atmospheric gas. With a tunable laser, near simultaneous measurements are made within and on the wing of the absorption line of the gas being measured at two close-by ranges. The return signals are then processed to isolate the gaseous absorption from the other extinction components. An advantage of the DASE technique is that no absolute calibration of the system is required as it depends only on comparison of signals.

The number density $\bar{N}(z)$ of the gas at altitude z averaged over an interval Δ can be expressed in terms of the power P_r received at the two ranges z and $z + \Delta$ for the wavelengths λ_0 within an absorption line of the gas and λ_w outside the absorption line. The number density is given by

$$N(z) = \frac{1}{2k\Delta} \left[\ln \left\{ \frac{P_r(\lambda_0, z)}{P_r(\lambda_0, z + \Delta)} \right\} - \ln \left\{ \frac{P_r(\lambda_w, z)}{P_r(\lambda_w, z + \Delta)} \right\} + T + B \right] \quad \dots (2)$$

where $T = -2 [\bar{\alpha}_A(\lambda_0, z) - \bar{\alpha}_A(\lambda_w, z)] \Delta,$

$$\bar{\alpha}_A = \bar{\alpha}_R + \bar{\alpha}_M \text{ (bar indicates average over the interval } \Delta),$$

$$B = \ln [\beta(\lambda_0, z + \Delta)/\beta(\lambda_0, z)] - \ln [\beta(\lambda_w, z + \Delta)/\beta(\lambda_w, z)],$$

$$K = K(\lambda_0) - K(\lambda_w),$$

and $K(\lambda_0)$ and $K(\lambda_w)$ are the absorption cross sections at λ_0 and λ_w .

In eqn. (2), the terms B and T can be neglected if the spectral dependence of α_A and β can be neglected in the range λ_0 to λ_w and if the line and wing measurements are made nearly simultaneously to avoid significant temporal changes in α_A and β . Thus knowing K , $\bar{N}(z)$ can be estimated with this technique. Neglecting B and T , eqn. (2) can be written as

$$\bar{N}(z) = D/2K\Delta,$$

where D is the difference of the two logarithm terms in eqn. (2). Minimum detectable concentration of a minor atmospheric constituent can be obtained by setting a limit to the minimum value of D that can be detected. For a typical detection and digitization equipment the minimum value of D can be taken to be 0.02. With this value of D , the minimum detectable concentrations of water vapour and ozone, two constituents which are of great importance in atmospheric studies, are shown in Figs. 3 and 4 respectively. For water vapour, the absorption line at 0.69438μ is considered and for ozone the band around 0.29μ . Shown in the figures are the typical concentrations of water vapour and ozone for tropical conditions (McClatchey & D'Agati, 1978). For the concentration shown, water vapour measurements can be made upto 5km with a range resolution of 200 metres and ozone upto about 40km with a resolution of 1km.

The potential errors in this technique are investigated in detail by Schotland (1974). He showed that the neglect of the spectral dependence of β and α_A does not result in significant errors because of the close proximity of λ_0 and λ_w . But the requirement of near simultaneous measurement is very stringent because of the temporal variability of β and α_A due to fluctuations in aerosol concentrations. Schotland (1974) showed that if a relative accuracy in \bar{N} of 10 per cent is desired per observation, the measurements at the absorption line and wing should be made within a millisecond. The simultaneity requirement can be relaxed, by taking the average of \bar{N} over a number of observations. This, of course, results in loss of time resolution. This stringent requirement of simultaneity applies to measurements in troposphere. For measurements at stratospheric levels, the simultaneity requirement is not stringent because of the negligible aerosol backscatter.

Using this technique, Schotland (1971) has determined the water vapour concentration in the first few kilometers from the ground. He used a thermally tuned Q-switched ruby laser with an energy of 0.6J in a 20 n sec. pulse. Stratospheric ozone measurements have been made by Megi *et al.* (1977) using a

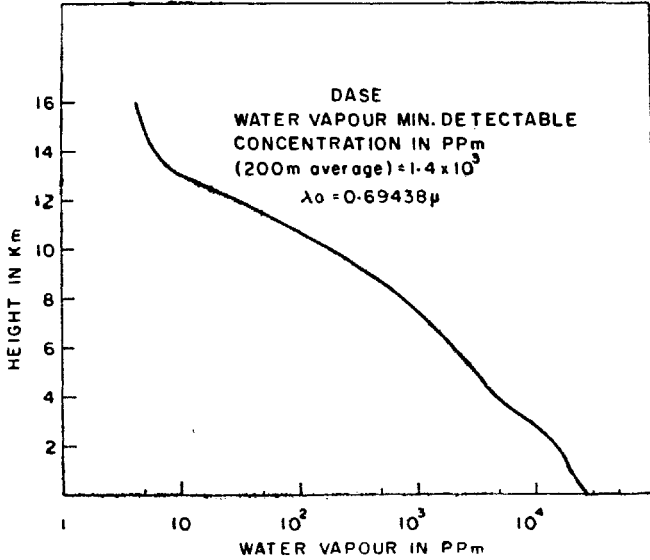


FIG. 3. Water vapour profile.

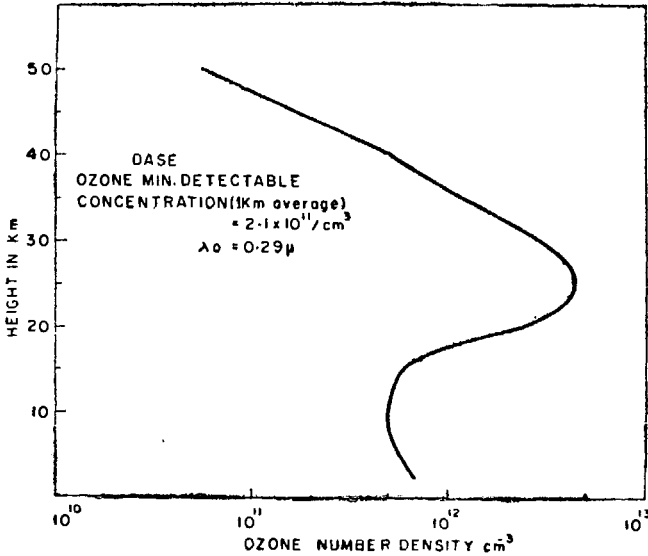


FIG. 4. Ozone profile.

frequency doubled Rhodamine 6G dye laser with an output energy of 20–40mJ in the wavelength range 300–310 μ .

RAMAN SCATTERING METHOD

Though the cross section for Raman scattering is usually smaller than that for Rayleigh scattering, it provides a very powerful lidar technique for identifying and

monitoring the atmospheric minor constituents and pollutants and for determining atmospheric temperature.

The pure rotational Raman scattering has a higher cross section than the vibrational-rotational Raman scattering. But the frequency shift of pure rotational scattering from the excitation frequency is much smaller than that of vibrational-rotational scattering. So, the optical filter specifications will be more stringent for isolating the pure rotational Raman scattering from the elastic scattering in the lidar return signal than for the vibrational-rotational Raman scattering.

It is advantageous to work with Raman scattering scheme at lower excitation wavelengths because of two reasons : 1) the Raman cross section is proportional to $1/\lambda^4$; and 2) at lower wavelengths photo-detectors are more efficient.

In the lidar eqn. (1), σ will correspond to Raman scattering cross section in this case. The transmission term in eqn. (1) should be replaced by two terms of the same exponential form, one for the upgoing excitation wavelength and the other for the downcoming Raman scattered signal.

Raman scattering from atmospheric water vapour, N_2 , O_2 and pollutants has been studied by many workers (Cooney, 1970, 1971, 1972*a, b*; Melfi *et al.*, 1969; Melfi, 1972; and Strauch *et al.*, 1971). Employing a frequency doubled Q-switched Ruby lidar, Melfi (1972) obtained the water vapour mixing ratio (defined as the ratio of mass of water vapour to the mass of dry air in a sample of the atmosphere) as a function of altitude upto about 2.5km using the Raman backscattered signals of H_2O and N_2 . Good agreement was found between the Raman laser data of H_2O and the conventional balloon-sonde measurements. Using H_2O and N_2 Raman scattered signals for the determination of mixing ratio has an advantage of requiring no absolute calibration of the lidar because the ratio of Raman scattered signals from H_2O and N_2 gives the mixing ratio (neglecting the wavelength dependence of α within the short range of H_2O and N_2 Raman scattering wavelengths). Using pure rotational and vibrational-rotational N_2 Raman signals, the elastic scattering (Mie and Rayleigh scattering) signals from the atmosphere can be normalized (Cooney, 1969, 1975).

Raman scattering scheme provides a powerful method of range resolved, remote analysis of atmospheric pollutants. Its advantage over other laser techniques for pollutant monitoring is that it requires processing of the received signal only without changing the wavelength of the transmitter for different pollutants whereas other techniques like resonance fluorescence or DASE require tuning of the laser which is more cumbersome and expensive. Several experimental studies on atmospheric pollutants using Raman scattering have been carried out (Inaba & Kabayasi, 1969; Kildal & Byer, 1971; Melfi *et al.*, 1973; and Garvey & Kent, 1974). One major difficulty in pollutant monitoring by Raman scattering technique, is to provide enough isolation between the comparatively very strong elastic scatter signal and the weak Raman signal from the different pollutants.

From the N_2 density obtained by Raman scattering technique, the atmospheric temperature can be determined by using the hydrostatic equation. The accuracy of N_2 density measurements is about 1 per cent while, by applying the hydrostatic

equation for clear air conditions, an accuracy of $\pm 2^\circ\text{K}$ in temperature can be obtained in the troposphere (Reagan & Schotland, 1975).

Garvey and Kent (1974) reported N_2 Raman scatter signal measurements to stratospheric height with their powerful Mark II lidar system. Thus, it is possible by this technique to determine temperatures upto stratospheric heights, above which temperature can be determined by Rayleigh scattering technique upto mesospheric heights as mentioned under Rayleigh and Mie scattering method.

A second method for the determination of temperature by Raman scattering technique, suggested by Cooney (1972) makes use of the shape of the envelope of the pure rotational Raman line which is temperature-dependent. If two interference filters are appropriately located within the envelope of the rotational lines, then the differences in the received signals at these wavelengths can be used as a measure of temperature. Cooney and Pina (1976) reported temperature measurements to an altitude of 1km using this method with an accuracy of about $\pm 2^\circ\text{K}$.

The sensitivity of Raman scattering technique can be improved by making use of the resonance Raman scattering. When a molecule or atom is excited at a frequency close to its resonance frequency, a great increase in the scattering cross section results.

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