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Research paper

# Line shape in a free-jet hypersonic expansion investigated by cavity ring-down spectroscopy and computational fluid dynamics

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ABSTRACT

Experiments are carried out for spectroscopic studies using hypersonic jet of carbon monoxide seeded in argon as a carrier gas. Probing of this jet using cavity ring-down spectroscopy revealed a double peak structure for various absorption lines. Flow field simulation using computational fluid dynamics is used to understand the shape of such lines integrated over line of sight. Absorption contribution from warmer non-isentropic part of the jet, owing to its transverse velocity variation, is found responsible for those line shapes.



## 1. Introduction

Experimental facilities with supersonic or hypersonic free expansion of gases are widely used as tools for molecular spectroscopic studies. Such jets are characterized by supersaturated gaseous conditions with low rotational temperatures which simplify the structure of molecular spectra. Therefore, steady expanded jets or pulsed jets are conventionally used to attain desired levels of cooling. Understanding the line shape for a particular transition has been one of the major objectives of free-jet based spectroscopic studies. Among such fundamental studies, Veeken and Reuss considered free expansion of ammonia initially to determine the rotational temperature and onwards to estimate the number density [1]. There, anomalous line shape with double peak structure was attributed to condensation occurring preferentially in the allegedly denser axial region. Similar line shape with central shallow dip was also noticed by Mizugai et al. [2]. Snels and Baldacchini [3] conducted experiments with ammonia using diode laser and found non-Gaussian line shape with central dip for rotation-vibration lines of $\nu_2$ band. Clustering at the axis was invoked for those line shapes showing dependence with probing distance from the nozzle exit and the stagnation pressure of the jet. In a similar study, apparent double peak line structure was attributed to clustering by Bajaj et al. [4]. Possibility of clustering was also reported in the supersonic jet of $N_2O$ [5] and acetylene [6].

While one of the explanations for anomalous line shapes is the formation of clusters in the neighborhood of jet axis, other explanations are also available in the literature. Gaveau et al. numerically predicted shape and width of absorption lines in case of CO-He free-jet for P(1) and P(2) lines [7]. In their study, shape of the absorption lines was found to be dependent on the variation of density and temperature in the jet. However the effect of jet boundary was not considered in that study. Campbell et al. found that 'Doppler dephasing' of rotational lines is responsible for the peculiar line shape observed in their microwave study of OCS and rare gas/OCS complexes [8]. This explanation was found non-unique by Lovas and Suenram for the supersonic jet of OCS seeded in various rare gases [9]. Besides, 'doubling' of line shape was accounted for loss of density due to clustering of OCS with carrier gas. Nevertheless, in their collaborative efforts, Campbell and Lovas finally concluded that density and temperature gradients along the laser beam are responsible for double peaked lines [10]. Ramos et al. disregarded condensation of nitrogen and attributed the observed double peak structure of lines to inhomogeneous line broadening due to strong temperature and density gradients within the silent zone, along the line of sight [11].

Thus diversity can be seen in the literature for explaining line shape with two peaks. In some of the reported findings, it has been noticed that, the observations used for justification of clustering do not exclusively conclude the clustering. Besides, flow field gradients can also be led responsible for such observations. However, it has also been felt that the studies, which account for flow field along the laser beam for the anomalous line shape, need detailed investigations to mark specific region of the flow and the flow

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property responsible for such line shape. In view of this, spectroscopic measurements are carried out for hypersonic jet of carbon monoxide and argon using cavity ring-down spectroscopy (CRDS). These studies are continuation of the earlier findings of Louviot et al. [12] where as well double peak structure of absorption lines had been observed for carbon monoxide in CRDS based measurements. Therefore present investigations specifically deal with analysis of absorption line shapes for line of sight spectroscopic measurements. Numerical simulations are also carried out to get the simulated lines. Efforts are then extended to correlate the experimental and simulated lines with structure of the free-jet. Details of the experimental setup, numerical simulations and reasoning for the anomalous line shape are given in the following sections.

## 2. Experimental and numerical methodology

Experiments are carried out in an experimental facility having a High Enthalpy Source (HES) connected to a vacuum chamber [12,13]. In the present studies, carbon monoxide (Air Liquide, 99.998% purity) is used as the seeding gas (5% by mass) and argon (Air Liquide, 99.998% purity) as the carrier gas (95% by mass). Mass flow rate of this mixture is regulated while flowing through HES where it is heated by passing 80 A current through graphite. A circular nozzle of 2 mm diameter is used as a throat for expansion of the gas in the vacuum chamber. This chamber is evacuated to the pressure of 11 ± 1 Pa so as to establish flow field known as a Campargue-type expansion [14]. Expansion of jet is probed using high sensitive cw-CRD spectrometer. Telecom grade fibered Distributed Feedback (DFB) diode lasers of power 10–20 mW are used to act as light source. Each of these lasers covers about 7 nm of wavelength (~30 $cm^{-1}$). For this, their temperatures are adjusted between −10 and 60 °C with the help of in-house developed proportional integral differential (PID) stabilizer. From the total light emitted by the laser, 10% is transferred to wavemeter (Burleigh WA-1600) and 90% is introduced into the optical cavity. This high finesse cavity ($F > 200{,}000$) consists of two high reflectivity (>99.9987%), plano-concave (1000 mm radius) mirrors which are separated by about 800 mm. Mode matching between laser beam and $TEM_{00}$ mode of cavity is achieved using single lens and two steering mirrors. Cavity length is modulated by mounting the output mirror on a piezoelectrical transducer in order to set resonance with laser frequency. At the resonance, acousto-optical modulator (AOM) gets switched off, owing to which photons evolve freely inside the cavity. Ring-down time is then obtained using an exponential fit of the intensity measured by a photodiode. Spectroscopic measurements are carried out at locations which are 5 mm, 20 mm and 40 mm downstream of the nozzle exit. These locations would be referred here onwards as X1, X2 and X3 respectively. It should be noted here that, the location of Mach disk for present experimental conditions is about 16 cm from nozzle exit. Therefore these axial probing stations are about 1/32th, 1/8th and 1/4th of the Mach disk location. These line of sight measurements are always ensured in the direction perpendicular to the jet axis. Experimental data is recorded using a LabVIEW based data acquisition system for the spectral range of carbon monoxide as [6220–6320] $cm^{-1}$ corresponding to the 3 ← 0 vibrational band.

Two numerical techniques, computational fluid dynamics (CFD) and method of characteristics (MOC), are employed to predict flow properties in the jet. These predictions form inputs for obtaining two synthetic absorption lines. Among these numerical techniques, initially, commercial CFD solver Fluent of ANSYS-15 [15] is used to visualize and understand the structure of hypersonic free-jet. This viscous flow solver considers mass, momentum and energy conservation equations to simulate the flow from reservoir till 50 mm downstream of the orifice. Beyond this distance, the Knudsen number ($K_n$), which can be defined as the ratio between the mean free path of the expanding molecules and the nozzle diameter, becomes too high. As a result of this, Fluent solver remains invalid due to break down of continuum assumption necessary for using Navier–Stokes equation. It has also been noticed from the experimental conditions that the Knudsen number for present case varies between 0.2 (core) and 0.6 (shear layer/jet boundary) which is still far from the free molecular regime ($K_n > 10$). Thus, using this computational technique, it is possible to analyze the flow in all parts of the jet which would later be helpful to construct the absorption line along entire line of sight. In view of this, two dimensional (2D) axi-symmetric viscous simulations are carried using mixture properties and assumption of perfect gas. Density based solver and Sutherland's law for viscosity are opted for these computations. Reservoir conditions ($P_0$ = 133.32 kPa and $T_0$ = 1300 K) are provided at the inlet of the fluid domain. Supersonic flow boundary condition with chamber pressure is implemented at the outlet. Converged steady state results are considered for further analysis. After viscous flow simulations using CFD, MOC based fit has been initially used to get the axial variation of Mach number. Axial temperature of the flow is then recovered using law of energy conservation for adiabatic expansion. Other flow variables are also obtained from isentropic relations. Then Eq. (1) is used to estimate off-axial variation of density in the silent zone [16].

$$\rho(z,y) = \rho(z,0)\{cos^2[\theta(z,y)]\}\{cos^2[\pi\theta(z,y)/2\phi]\} \tag{1}$$

Here, $\rho(z,y)$ is off axial density evaluated from the axial counterpart, $\rho(z,0)$, and angle made by velocity vector with jet axis ($\theta$). Value of $\phi$, given by Ashkenas and Sherman [16] is used here. This equation is the best fit for MOC based prediction hence it is valid only in the isentropic core of jet [16]. Therefore, boundary of isentropic core is mandatory input for this approach to construct the computational domain. Results obtained from CFD simulations are considered here to provide this valuable input. Thus obtained off-axial density and known reservoir density are used to evaluate local temperature using isentropic relation. Mach number of any location is then obtained from isentropic relation with the help of local and stagnation temperatures. Velocity is computed using Mach number and temperature.

Flow field information received from CFD based 2D axi-symmetric simulations is used to build the entire three dimensional (3D) jet. Besides, only 3D inviscid core is constructed using similar information given by MOC. These transformations help to obtain the absorption line shape for entire line of sight (CFD) and for limited sight (MOC) at any laser probing station. At a given probing station, laser beam of 1 mm diameter with Gaussian intensity distribution and zero divergence is introduced in such 3D domains. Since the chosen mesh for computations has length scale of the order 0.02 mm, it ensures participation of sufficient number of cells (identified by $x$, $y$ and $z$ co-ordinates) in the laser beam for predicting the absorption line shape. Then, the flow properties viz. component of velocity in the line of sight, density and temperature are used to get the synthetic line from CFD and MOC. Eq. (2) is used for calculation of absorption coefficient integrated along the line of sight.

$$\alpha(\tilde{\upsilon}) = \sum_z \left\{ \sum_{xy} \left[ n_{xyz}\bar{\sigma}_{ij}(T_{xyz}) g_D(\bar{\upsilon} - \bar{\upsilon}'_{ij}) \frac{l(z)}{L} \left( \exp\left(-\frac{x^2+y^2}{w_0^2}\right)\right)^2 \times \frac{1}{\sum_{xy}\left(\exp\left(-\frac{x^2+y^2}{w_0^2}\right)\right)^2} \right] \right\} \tag{2}$$

Here $x$, $y$ and $z$ are Cartesian coordinates, $n_{xyz}$ is number density (molecule $cm^{-3}$) corresponding to the cell or elemental volume at $x$, $y$ and $z$, $T(x,y,z)$ is temperature (K) of the cell, $\bar{\sigma}_{ij}(T)$ is integrated