



An investigation of the near-infrared collision induced absorption bands of oxygen with SCIAMACHY solar occultation data

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ABSTRACT

The present work investigates the Collision Induced Absorption (CIA) bands of oxygen that occur at 1065 and 1270 nm from solar occultation data obtained with the spaceborne spectrometer SCIAMACHY. The effort is motivated by the interest in these two strong CIA bands in atmospheric research and the paucity of data on them under realistic atmospheric conditions. The observing geometry provides long integration paths and in turn easily measurable absorption signals. Our analysis method relies on the accurate separation of the CIA band signature from the rest of the continuum and structured components over selected spectral intervals. We show that the shapes of the two CIA bands seem well described by specific laboratory determinations over a broad range of tropospheric and low-stratospheric conditions. By analyzing a full month of solar occultation data we find that the ratio of peak binary cross sections in air, $\sigma_{1270 \text{ nm}}^{\text{peak}}/\sigma_{1065 \text{ nm}}^{\text{peak}}$, is ~ 4 and seemingly independent of tangent height from 6 to 18 km. We tentatively estimate the absolute binary cross sections and compare them to existent measurements. A better characterization of the near-infrared CIA bands of oxygen should facilitate their implementation in remote sensing applications in the way it is often done with the CIA bands that occur in the visible.

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1. Introduction

Oxygen molecules interact with the atmospheric gas to form more complex compounds often referred to as oxygen collision, or $\text{O}_2 \cdot \text{X}$, complexes even though they comprise a combination of bound, metastable and purely collisional states [1,2]. The $\text{O}_2 \cdot \text{X}$ complexes become apparent in Earth's spectrum of transmitted/reflected sunlight through a number of Collision Induced Absorption (CIA) bands from the ultraviolet to the infrared. The CIA bands of oxygen have bandwidths of up to a few tens of nm and intensities that scale in accordance with the product of densities of the O_2

and X collision partners. In the terrestrial atmosphere, X is typically another O_2 or N_2 molecule.

The CIA bands of oxygen are used for the remote sensing of sunlight photon pathlengths through clouds [3] and the characterization of aerosol properties [4], are essential to the accurate retrieval of some trace atmospheric constituents [5] and play a minor though non-negligible role in the energy budget of the atmosphere [6]. Recently, they were also used to test the classical theory of lunar eclipses [7,8] and were postulated as observables in the atmospheres of transiting Earth-like extrasolar planets [8]. In perspective, much of the interest in the CIA bands of oxygen stems from the fact that their integrated intensities may be comparable to the intensities of some O_2 monomer bands but because their band widths are broader their optical depths remain linear over much longer paths.

The first detections of the CIA bands of oxygen in laboratory and atmospheric observations date back from

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the 19th century [9,10]. The spectroscopy of the $O_2 \cdot X$ complex in the frame of atmospheric research was reviewed in 1998 [6], and it was noted that major deficiencies remained in the characterization of some bands. Since then, a number of solar transmission measurements and laboratory experiments [11–26] have significantly contributed to a better description of the absorption properties of the $O_2 \cdot X$ complex.

Solar transmission measurements have a reduced control over the column-integrated conditions of temperature and pressure in the gas, especially if the measurements are carried out from the ground. In addition, they must deal with other molecular bands partially masking the CIA bands [15,22]. In contrast, laboratory experiments have some control over both parameters and the masking effect of nearby bands can usually be avoided or minimized. Laboratory experiments do, however, often involve relatively short pathlengths, which makes it necessary to work at super-atmospheric pressures to obtain acceptable signals [14,20,21].

Since the CIA band properties are sensitive to the conditions in the medium [13,21,27], it is not always clear whether the binary cross sections inferred from ground-based solar transmission measurements and laboratory experiments are directly applicable to the full range of atmospheric conditions. This uncertainty is particularly evident at 1065 and 1270 nm [11,13,14], the two CIA bands that jointly absorb as much of the solar energy input as the other shorter-wavelength CIA bands put together [6].

Despite the works mentioned above, a systematic effort to thoroughly characterize the CIA bands of oxygen is still lacking. A new addition [28] to the HITRAN spectroscopic database [29] compiles selected sets of binary cross sections for the $O_2 \cdot O_2$ and $O_2 \cdot N_2$ complexes (and other complexes as well). To facilitate their correct implementation, the data are accompanied by the ranges of temperature and pressure employed in their determinations. Thus, we find it timely to assess the binary cross sections for the near-infrared CIA bands of oxygen appeared in the last two decades against atmospheric observations, especially if the measurements probe atmospheric layers that are not separately accessible in ground-based measurements.

To that end, we investigate spectra of the direct solar light transmitted through the atmosphere measured by the space-borne spectrometer SCIAMACHY [30,31]. The broad spectral coverage of the instrument and long integration paths in the solar occultation geometry enable the simultaneous identification of multiple CIA bands. By focusing on tangent heights from 6 to 18 km we probe pressures that vary by more than one order of magnitude and temperatures within a range of tropospheric and low-stratospheric conditions. To the best of our knowledge, this is the first investigation to include the near-infrared CIA bands of oxygen over such a broad range of atmospheric conditions.

2. Data

The SCIAMACHY spectrometer is a payload instrument on the European Space Agency's ENVISAT satellite [30,31]. The satellite is in a Sun-synchronous near-polar orbit at

98° inclination observing the terrestrial atmosphere in nadir, limb and solar/lunar occultation modes. The instrument covers the spectral range from 240 to 2380 nm using eight separate channels with nominal resolutions between 0.24 and 1.48 nm. The satellite's orbit restricts the operation in the solar occultation mode to tangent point latitudes 49–69°N. In this mode the estimated vertical resolution is 3 km.

For the present investigation, we used solar occultation spectra measured in March 2003 at tangent heights between 6 and 18 km. This range of tangent heights was selected because the lower-altitude spectra are severely absorbed, whereas higher up the CIA bands become exceedingly faint for an accurate analysis. We used data collected in Channels 3–6, which span wavelengths 391–605, 597–789, 776–1056 and 990–1750 nm. Level 1b products were obtained by application of the SciaL1C tool available on the ESA/SCIAMACHY website. Channel 6 suffers from bad/dead pixels, which were removed from the full set of spectra. To enhance the signal-to-noise ratios, reduce the amount of data and average the signal over the solar disk, we formed daily means over 2-km altitude bins. Reference spectra to remove the solar spectrum and the instrument's response function were formed by co-adding all individual spectra from each day between 100 and 120 km altitude. The spectra from Channels 5 and 6 were carefully merged to ensure an optimal baseline at short wavelengths for the CIA band at 1065 nm.

The spectral resolutions of the channels differ from the pre-flight specifications. We derived in-flight resolutions from the comparison of SCIAMACHY and synthetic spectra assuming a Gaussian shape for the instrumental slit function. For Channels 4 and 6, the derived full widths at half maximum in the intervals that contained the O_2 and $O_2 \cdot X$ features were 0.36 and 1.1 nm, respectively. For Channels 3 and 5, we did not need to derive the full widths at half maximum.

Our analysis relies on the Differential Optical Absorption Spectroscopy (DOAS) technique [32]. Its application to solar occultations is straightforward because the extinction of direct solar light in the atmosphere follows the Beer–Lambert law of exponential attenuation. We denote by τ the absorbance (or optical depth) of the path-integrated gas column at a specified tangent height. The basis of the DOAS technique is to fit the empirical absorbance with a synthetic function that includes each possible contributing absorber. Its success depends on the availability of good representations for all terms contained in the synthetic spectrum. Conversely, one might use the systematic mismatch between observations and synthetic spectra to draw valuable conclusions on the likely band shapes.

3. The CIA bands of oxygen in the near infrared

The $X(0)–a(0)$ band of O_2 at 1270 nm is one of the strongest oxygen features in the near-infrared spectrum of the terrestrial atmosphere. The $X^3\Sigma_g^-–a^1\Delta_g$ transition is electric-dipole forbidden but occurs following both magnetic dipole and electric quadrupole selection rules. The associated $X(0)–a(1)$ band at 1065 nm involves the transition into the

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