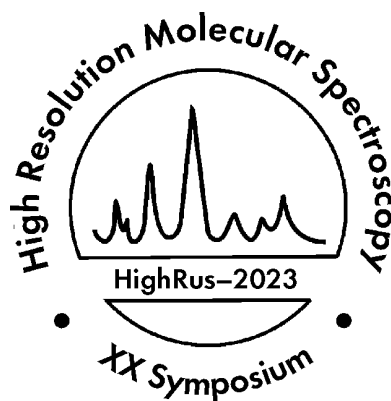




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# Abstracts of Reports

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## Collisional transformation of the complex vibrational susceptibility of a linear molecule: the $\nu_3$ band of $\text{CO}_2$ as a guiding example

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We report results of a joint analysis of pressure effects on both parts of the complex vibrational susceptibility of an ensemble of linear vibrating rotators. While the imaginary part of susceptibility which determines the absorption band shape was numerously studied for different systems, the transformation of the real part remains - to the best of our knowledge - unexplored. Its comprehension, especially for frequencies close to anomalous dispersion, might be useful to control phase-matching conditions when coherent optical responses are generated.

The concept of the infinite-order relaxation matrix  $\Gamma$ , which couples different rotational transitions (lines) forming a vibrational band, allows us to simultaneously calculate both terms of susceptibility by applying a certain model of  $\Gamma$ . The results are reported for compressed  $\text{CO}_2$ , for which the quantum strong-collision model (SCM) [1, 2] was successively tested both for IR absorption and Raman spectra [3] as well as for the time-domain responses [4]. Moreover, the use of SCM leads to compact susceptibility expressions [1] that do not require matrix inversion.

At low pressures, the SCM exactly reproduces the resolved line structure of a vibration-rotation absorption band which, as pressure is raised, transforms into a smoothed Bjerrum doublet and then coalesces into a quasi-lorentzian line exhibiting the pressure narrowing effect.

The transformations of the real susceptibility are following. Initially, it consists of resolved dispersion terms, but, as the line structure is smoothed by pressure, an unexpected triplet is formed with a hump near the band origin at  $\nu_0$ . This feature survives till the doublet absorption band structure remains resolved. As the gas density is further increased, the line mixing manifests itself in the appearance of a single dispersion curve centred at  $\nu_0$ . The line mixing reduces both the absorption band width and the spectral interval occupied by anomalous dispersion. However, the far periphery of normal dispersion remains unaffected and continues to scale as  $|\nu - \nu_0|^{-1}$ .

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