Temperature-Driven Spin–Phonon Coupling in Layered VOCl Revealed by Raman Spectroscopy

Chinmay Kumar Mohanty[1] , Igor Antoniazzi[1], Amit Pawbake[1],Adam Babiński[1], Clement Faugeras[2] , and Maciej R. Molas[1]

[1]*Faculty of Physics, University of Warsaw, Pasteura 5, Warsaw, Poland*
[2] *LNCMI, UPR 3228, CNRS, EMFL, Université Grenoble Alpes, 38000 Grenoble, France*

Email: c.mohanty@uw.edu.pl

Two-dimensional (2D) layered magnetic materials (LMMs) represent an emerging class of atomically thin van der Waals (vdW) systems with robust magnetic order persisting to the monolayer limit, making them promising candidates for opto-, spin-, and valleytronic applications. VOCl, an oxychloride LMM, exhibits easy-axis antiferromagnetic ordering along the a axis below 79 K [1]. Owing to strong magneto-optical coupling, magnetic phase transitions in VOCl can be efficiently investigated using optical techniques. In this work, we employ temperature-dependent Raman spectroscopy to investigate the evolution of phonon modes in VOCl flakes of varying thickness, revealing temperature-driven phonon anomalies associated with spin-phonon coupling. The temperature-dependent phonon behavior of 66-layer, 18-layer, 12-layer, and 4-layer VOCl flakes is comparatively analyzed across a wide temperature range. The Raman spectra of VOCl flakes consist of three phonon modes, denoted A_g^1, A_g^2, and A_g^3. At high temperatures (>150 K), all modes exhibit conventional anharmonic behavior, characterized by a nearly linear frequency evolution for all thicknesses. Below approximately 150 K, the A_g^1 and A_g^2 modes show pronounced deviations from this trend, consistent with a magnetic phase transition from the paramagnetic state to the incommensurate antiferromagnetic phase of VOCl [2]. Upon further cooling, the temperature-dependent Raman shifts of these modes become nearly constant as the Néel temperature (~80 K) is reached [3]. Notably, in the 4-layer sample, the A_g^1 mode exhibits an additional anomaly near 50 K, which may correspond to the magnetic compensation temperature proposed in Ref. [3].

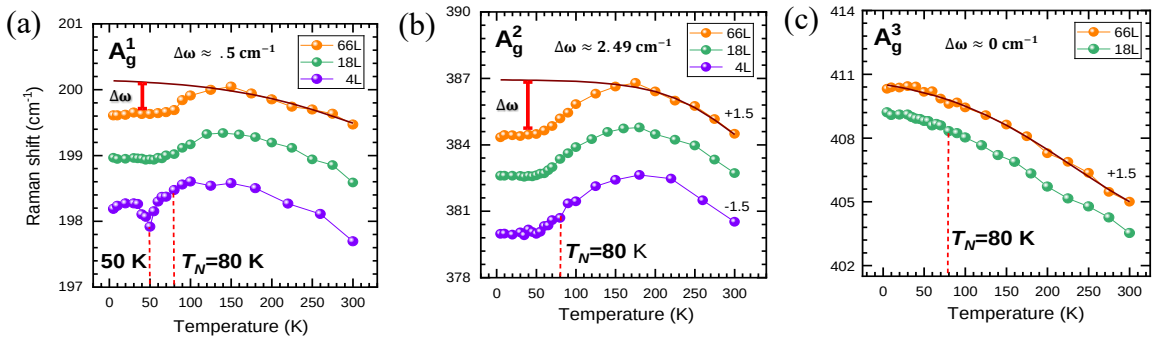

Fig. Temperature dependence of : (a) – (c) Raman shifts obtained for the A^1_g, A^2_g, and A^3_g phonon modes

Our measurements demonstrate that the A_g^1, A_g^2, and A_g^3 phonon modes in VOCl exhibit a pronounced mode- and thickness-dependent temperature response. This contrasting behavior highlights the crucial role of phonon symmetry in governing magnetoelastic interactions and provides insight into the influence of reduced dimensionality on lattice dynamics in layered VOCl.

[1] G. V. et al., ACS Nano 16 (2022) 13814.
[2] W. Wang et al., 2D Materials 9 (2021) 015027.
[3] W. Wang et al., Chinese Physics B 30 (2021) 107502.