## **Resonant Raman scattering of CrCl<sub>3</sub>**

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Chromium trihalides belong to the family of the magnetic layered materials with general formula  $CrX_3$ , where X is I, Br and Cl. In bulk form, they exhibit the ferromagnetic ( $CrBr_3$ ) and antiferromagnetic ( $CrI_3$  and  $CrCl_3$ ) order of spins. However, a single layer of each material exhibits the ferromagnetic ordering [1, 2].



Figure 1: (a) Raman spectra of the bulk  $CrCl_3$  under different laser excitation: 633 nm, 561 nm, 515 nm, 488 nm, 405 nm. Six Raman-active modes are observed, with the mode around 120 cm<sup>-1</sup> beaing weak in intensity compared to the other modes. (b) Temperature evolution of the  $A_g^3$  mode with marked the  $T_C$ .

Here, we investigate the resonant conditions, polarization properties and temperature dependence of a bulk  $CrCl_3$  at low temperature (T = 5 K).

To achieve the best resonant conditions for Raman scattering, we compared the intensities of the Raman peaks measured on CrCl<sub>3</sub> under different excitations (633 nm, 561 nm, 515 nm, 488 nm, 405 nm). The most intense Raman signal was observed under 405 nm excitation at T=5 K, see Figure 1 (a). This excitation energy was then used to investigate the polarization dependence and temperature evolution of the bulk CrCl<sub>3</sub>. The polarization dependence measurements revealed that there are two types modes: three  $E_g$  and  $A_g$ , assigned as  $E_g^1$ ,  $A_g^1$ ,  $E_g^2$ ,  $A_g^2$ ,  $A_g^3$  and  $E_g^3$ . Note that the top indexes are additional numbering to resolve the peaks sharing the same notation. Additionally, the modes  $A_g^1$  and  $A_g^3$  appear to be doubled sharing the same symmetry as adjacent modes.

Furthermore, temperature-dependent measurements revealed that the Raman shifts of all modes decrease with increasing temperature. As shown in Figure 1 (b) the temperature evolution of the  $A_g^3$  mode exhibits distinct behavior in the range of 5 K - 30 K compared to temperatures above 30 K. These measurements suggest that at temperatures above 30 K we observe the vanishing of ferromagnetic interactions.

Our results confirmed the presence of six Raman-active modes with two distinct symmetries  $E_g$  and  $A_g$  described in the literature [1]. Furthermore, the results of the temperature-dependent measurements showed that the Raman scattering can be applied to determine the Cure temperature of the CrCl<sub>3</sub>.

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## References

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