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High resolution polarized Raman scattering study of multiferroic MnWO₄

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Multiferroic materials with magnetic and electric ordering within a single phase gained attention during the last years due to their potential for industrial applications. Materials with a cycloidal spin arrangement are of particular interest because the electrical polarisation is directly caused by the complex magnetic order. Hence, both ordering phenomena are tightly coupled in this category of compounds. One example is MnWO₄, which passes successively three antiferromagnetic phase transitions on cooling. The phase AF2 between 12.5 K and 7 K exhibits a cycloidal spin arrangement with an incommensurate magnetic propagation vector. This ordering induces a spontaneous polarisation by the inverse Dzyaloshinski-Moriya-interaction. As a consequence, the interaction between spin and polarisation could possibly lead to a change in the spectrum of lattice vibrations.

Due to the tiny polarization, it was believed up to now that this effect was too small to be observed experimentally. Using high resolution polarized Raman scattering, however, we were able to detect clear signatures of this magnetoelectric interaction. The Raman spectra were taken from a MnWO₄ single crystal in the temperature range from 100 K to 6 K covering the three magnetic phase transitions. A careful analysis of the experimental data allowed the precise determination of both, the wave numbers and intrinsic linewidths.

Most of the 18 Raman-active phonons show a significant shift in wave number below the magnetic ordering temperature that cannot be explained merely on the basis of lattice dynamics, but should be attributed to spin-phonon-interaction. The most striking effect is observed for the A_g mode with the highest frequency (about 884 cm⁻¹), which corresponds to an oxygen stretching vibration and exhibits a pronounced softening on cooling.

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