6.5 Interference of Scattering Pathways in Raman Heterodyne Spectroscopy of Multilevel Atoms

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Raman heterodyne detection of nuclear magnetic resonance is a sensitive technique for optical detection of an NMR transition, which is driven by a resonant radio frequency field. The observed signal is linear in the transition matrix elements of the magnetic resonance transition and two optical transitions. This linearity is the reason that signal contributions from different sites or different rf transitions can interfere, in many cases destructively. We discovered such an interference effect, which appears to have been overlooked in the past. It occurs between signal contributions that originate from the same magnetic resonance transition of atoms with different positions within the inhomogeneously broadened optical resonance line. These atoms contribute to the coherent Raman scattering through different scattering paths, which involve optical transitions to different nuclear spin states of the same electronic state. The interference between all possible scattering paths leads to complete signal cancellation, if the atoms, that are involved in the different scattering paths, are equally polarized. To study the effect and to eliminate the interference, we used a novel pump-and-probe technique with two laser beams. With two independent laser frequencies, it becomes possible to separate the individual scattering paths. We calculated the dependence of the signal on the frequency of both laser beams as well as on the radio frequency and compare the results to experimental data from $Pr^{3+}:YAlO_3$. Our results show that the interference reduces the signal amplitude of the conventional Raman heterodyne experiment but can be eliminated in the new experiment.