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

(T. Blasberg and D. Suter)

Raman heterodyne detection of nuclear magnetic resonance (NMR) is a sensitive technique for optical detection of magnetic resonance. The technique uses a resonant radio frequency field to excite a coherent superposition between two nuclear spin sublevels of the same electronic state. For the optical detection of the sublevel coherence a resonant laser field couples one of the nuclear spin states to a different electronic state and is coherently Raman scattered from the sublevel coherence. The magnetic resonance is observed as the interference between the laser field and the scattered Raman field. The observed signal is linear in the transition matrix elements of the magnetic resonance transition and the two optical transitions, which are involved in the Raman scattering. This linear dependence is the reason that signal contributions from different sites or different rf transitions can interfere, in many cases destructively.

In Ref. [1] we discuss 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 multilevel atoms with different positions within an inhomogeneously broadened optical resonance line. In conventional Raman heterodyne experiments that are performed with a single laser beam, 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. We show that 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 calculate 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 the solid Pr^{3+}:$YAlO_3$ at cryogenic temperatures. Our results show that the interference reduces the signal amplitude of conventional experiments but can be eliminated in the new experiment.

[1] T. Blasberg and D. Suter, 'Interference of Scattering Pathways in Raman Heterodyne Spectroscopy of Multilevel Atoms', submitted to Phys. Rev. B (1994)