6.6 Excitation of Coherent Raman Beats in Rare-Earth Solids with a Bichromatic Laser Field

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Coherent Raman beats represent an attractive technique for optical detection of nuclear magnetic resonance (NMR). The experiment consists of two parts: The excitation of a coherent superposition between two nuclear spin substates of one electronic state and subsequent coherent Raman scattering of a resonant test laser field, which couples one of the nuclear spin states to a different electronic state. The Raman signal, which is observed as a beat signal between the test laser and the scattered Raman field, provides information on energy level splittings and dephasing rates. To perform purely optical magnetic resonance experiments, we have developed a new excitation scheme [1], which uses a bichromatic laser field, whose two frequency components couple two different nuclear spin substates of the same electronic state to a different electronic state.

To identify those experimental parameters, which determine the efficiency of the bichromatic excitation of sublevel coherence, we applied a density-matrix formalism to calculate the evolution of the sublevel coherence during the excitation period analytically. The calculations showed that as long as the frequency difference between the two frequency components of the bichromatic pump laser field matches the splitting between two spin states, arbitrarily wide sublevel spectra can be excited with this method, even if the oscillator strengths of the optical transitions are small. However, a large excitation efficiency can only be obtained, if the coupling strengths of the two frequency components are of the same order of magnitude. In systems, where the ratio of the two dipole matrix elements is unfavorable, our experimental setup offers the possibility to choose different amplitudes for the two frequency components, thus optimizing the efficiency for arbitrary ratios of the oscillator strengths. The comparison between a calculation of the time evolution of the sublevel coherence and experiments performed on Pr^{3+}:$YAlO_3$ at cryogenic temperatures were in excellent agreement [2]. We observed that up to 7 % of the incident test laser photons were coherently Raman scattered.

[1] T. Blasberg and D. Suter, 'Excitation of Coherent Raman Beats in Rare Earth Solids with a Bichromatic Laser Field', Optics Communications **109**, 133-138 (1994)

[2] T. Blasberg and D. Suter, 'Bichromatic Excitation of Coherent Raman Beats in Rare Earth Solids', accepted by Phys. Rev. B, to be published in the March 1995 issue.