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Photon-recoil assisted rovibrational spectroscopy of the $^{24}MgH^+$ ion

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Precision spectroscopy of molecules promises a wealth of interesting applications e.g. for better determination of natural constants, for tests of fundamental particle theories, and for qubit realizations. Unfortunately, most molecules do not possess closed transitions for laser cooling, crucial for obtaining temperatures low enough for exploiting this novel realm of physics.
A way to circumvent this issue is to work with a trapped molecular ion, and sympathetically cool its translational degrees of freedom by Coulomb interaction with a co-trapped atomic ion possessing suitable transitions for laser cooling. Doppler laser cooling will lead to the formation of a two-ion Coulomb crystal, which can then be prepared in the ground state of motion via the sideband cooling technique.
Having prepared this system, it is possible to detect a change of the internal state of the molecular ion by monitoring the common motional state on the cooling ion, a technique called quantum logic spectroscopy (QLS), first proposed by Schmidt et al. [1]. In our group, an extension of QLS called photon-recoil spectroscopy (PRS) [2], where the target ion absorbs more than one photon, is used.
So far we have successfully performed PRS of a dipolar transition in the atomic ion $^{24}Mg^+$ of 280 nm, to verify that the technique works. The light-ion interaction has been modeled by rate equations, and my simulations are in good agreement with our experimental results [3].
The next goal is to perform PRS on the closed rovibrational transition v = 0, J = 1 $\leftrightarrow$ v = 1, J = 0 in $^{24}MgH^+$ of 6.17 μm, to show the applicability to transitions resulting in very low photon recoil. We have so far modeled this system and produced simulated spectra as guidelines for the ongoing experiments.
The ultimate goal is to drive purely rotational transitions in $^{24}MgH^+$, with energy splittings in the THz range, resulting in photon recoil too low for detection by PRS. The solution is to drive Raman transitions with counter-propagating laser beams, since the effective k-vector is then given by the difference of the two k-vectors driving the transition: $\vec{k}_{Raman} = \vec{k}_1 - \vec{k}_2 \approx 2\vec{k}_1$.
The ideal choice of laser for this is a femtosecond frequency comb for two reasons: 1) $\vec{k}_{Raman}$ can be made up of any two k-vectors with the right frequency difference, hence the different teeth of the comb can all work in pairs to drive the transition, and the full power of the comb is exploited. 2) The frequency difference between the teeth is easily changed by tuning the repetition rate, making the comb extremely versatile: Any transition within the spectral bandwidth of the comb ($\sim$ 8 THz) can be driven.
Frequency comb driven Raman transitions of 1.8 THz between D-finestructure levels in $^{40}Ca^+$ has recently been demonstrated in our group [4], laying a strong foundation for the future $^{24}MgH^+$ experiments.

[1] Schmidt et al., Science, 309, 5735 (2005)
[2] Wan et al., Nature Com., 5, 3096 (2014)
[3] In preparation (2019)
[4] Solaro et al., Phys. Rev. Lett., 120, 253601 (2018)

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