U.S. ARMY RESEARCH, DEVELOPMENT AND ENGINEERING COMMAND

New Process for Efficient Laser Pumping for IRCM: Three-for-One Cross-Relaxation

Larry Merkle[1] and Mark Dubinskii[2]

[1] General Technical Services

[2] Army Research Laboratory

22 Aug 2018

New Process for Efficient Laser Pumping for IRCM: Three-for-One Cross-Relaxation

Part of Advanced Solid-State Lasers Program

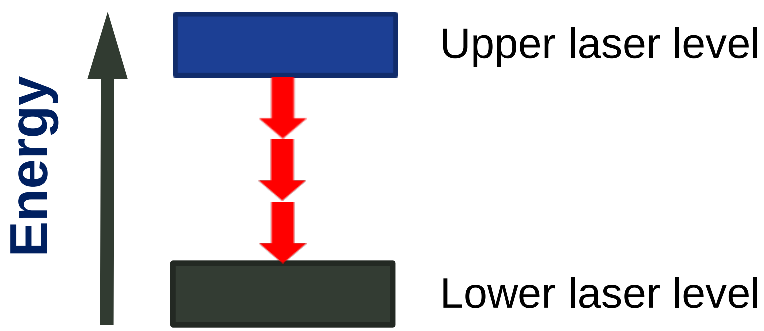
CFT Supported	Technology	Projects	Mission
AMD	Scalable Approaches to HEL/DEW	• Laser Engine for Master Laser • Beam Control Lasers • Novel Beam Control Concepts	- C-RAM - C-Sensors - C-UAS
FVL	Mid/Longwave IR Lasers	• MWIR Laser Engine • Conversion to LWIR	- IRCM/ - Illuminator lasers for DVE

MOTIVATION FOR THIS STUDY

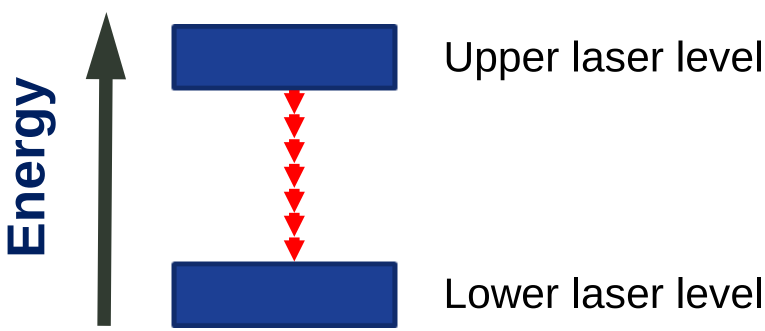
- Goal: Countermeasures against IR sensors (IRCM) – to protect Future Vertical Lift platforms and other airborne assets
- Our part: Materials for pulsed, mid-infrared (mid-IR) lasers
- Challenge: Significantly higher pulse energies to deal with future threat sensors
- SWaP considerations favor:
  - Single laser (rather than laser plus wavelength shifting device)
  - Solid-state laser (rather than gas or liquid)
  - Pumping (getting energy into the laser gain material) by laser diodes (for high efficiency)
- Pulsed operation favors this over direct use of laser diodes

TYPE OF LASER MATERIAL FAVORED

- **To emit the laser light: Trivalent rare-earth ion as a deliberate impurity**
  - Long upper state lifetime – more efficient use of pump power
  - Narrow emission peaks – higher gain at those peaks
- **Host crystals with low maximum phonon energies**
  - Minimize loss of energy to phonons (essentially to heat)



High-energy phonons:
Few-step process can cross gap – high probability
Serious "quenching" of the light emission



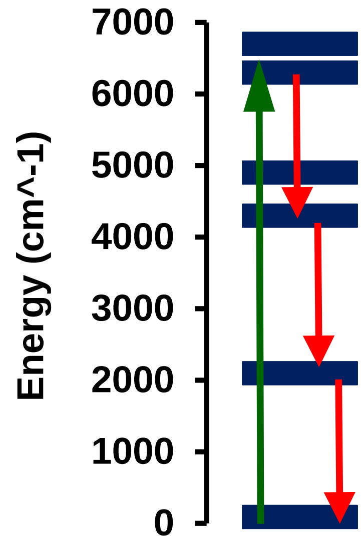
Low-energy phonons:
Many-step process needed to cross gap – low probability
Far less "quenching" of the light emission

PR:RPC

Particular material chosen for this study: $Pr:RbPb_2Cl_5$

Why Pr^{3+} as the laser ion:

- Absorption peaks ~1.5 microns – Can use diode lasers developed for telecom
- Three emissions in desired wavelength range
- Improves chance that at least one will be efficient

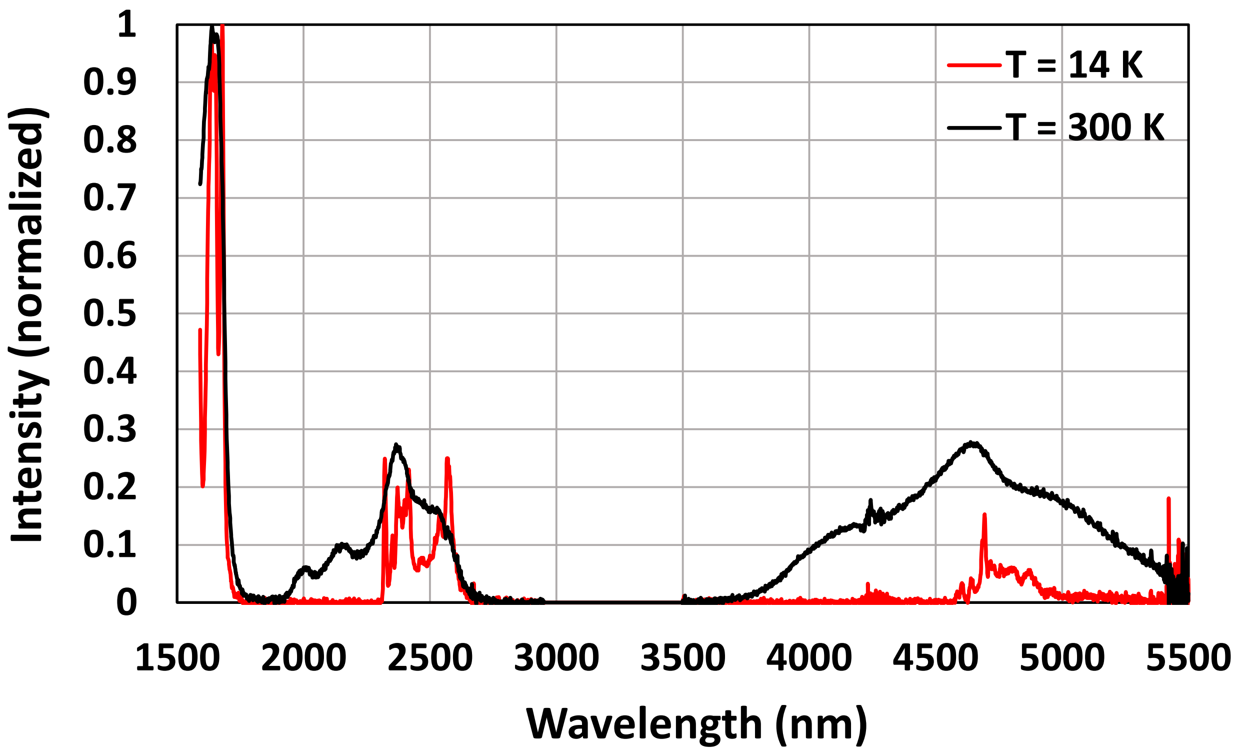


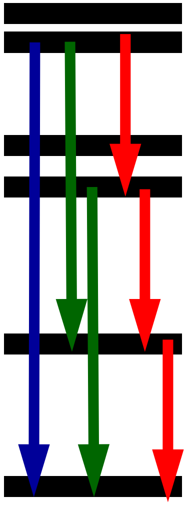
Why RPC as the host crystal:

- Very low maximum phonon energy – 203 cm^{-1} (so need ~10-12 phonons to quench by multiphonon)
- Fairly good resistance to moisture, compared to many low-phonon-energy crystals

IR FLUORESCENCE OF PR:RPC

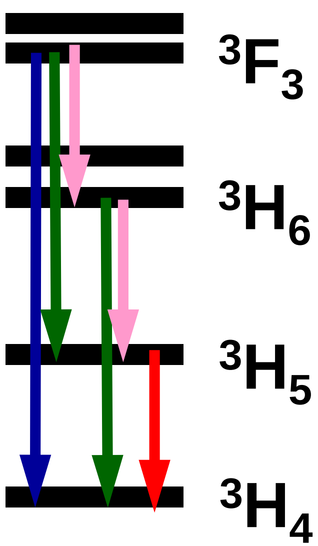
- At low temperatures: Three emission bands with similar total intensity
- By room temperature: Desired band (mid-IR) is stronger
- ~3/4 of all photons emitted are in the 3600-5500 nm band!

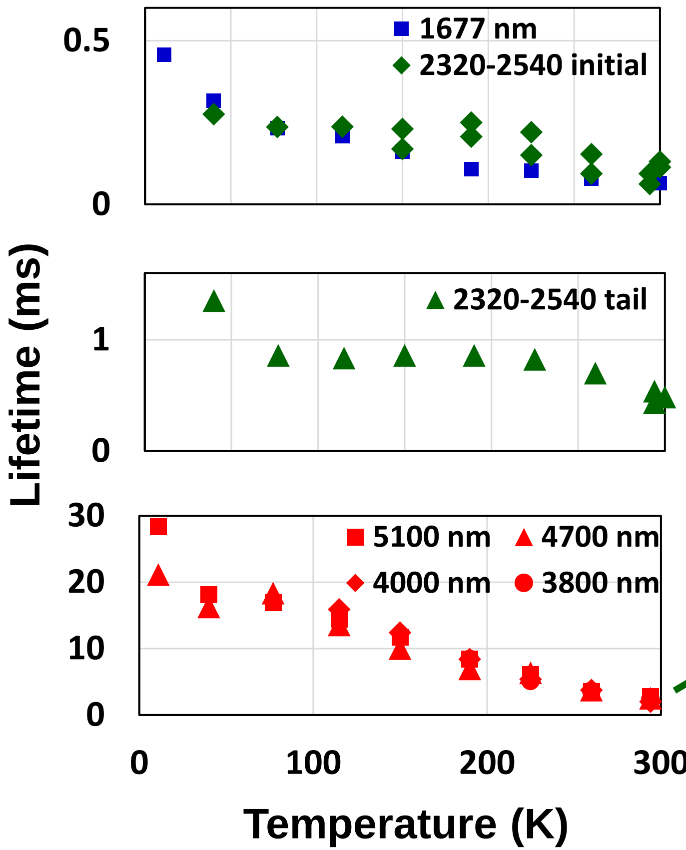




LIFETIMES

- Mid-IR fluorescence encouraging – but what does it mean?
  Are the three different transitions all strong?
- Lifetime data say: No

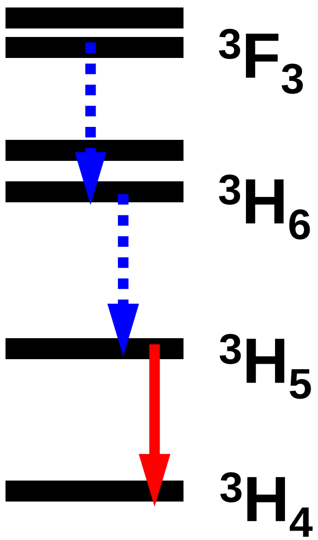




- **3600-5500 nm range: Only small traces of the fast decays**
- **So, that band <u>mostly</u> due to $^3H_5$ emission**
- **Why is it so strong?**

NOTE: Room T lifetime ~2.5 ms – long enough for efficient laser diode pumping

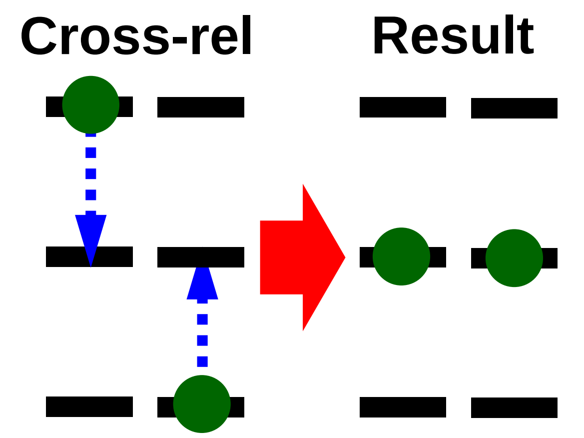
INTERPRETATION – WHAT IT IS NOT



- Simplest picture: Perhaps most Pr^{3+} ions decay by non-radiative processes (quenching) to the first excited state, $^3H_5$
- It fluoresces (and has the potential to lase)
- Lifetimes and theoretical methods allow estimates of these strengths
- Predicts $^3H_5 \rightarrow {}^3H_4$ mid-IR band ONLY ~10-20 % as strong as we observe
- Clearly something very different – and better – is happening!

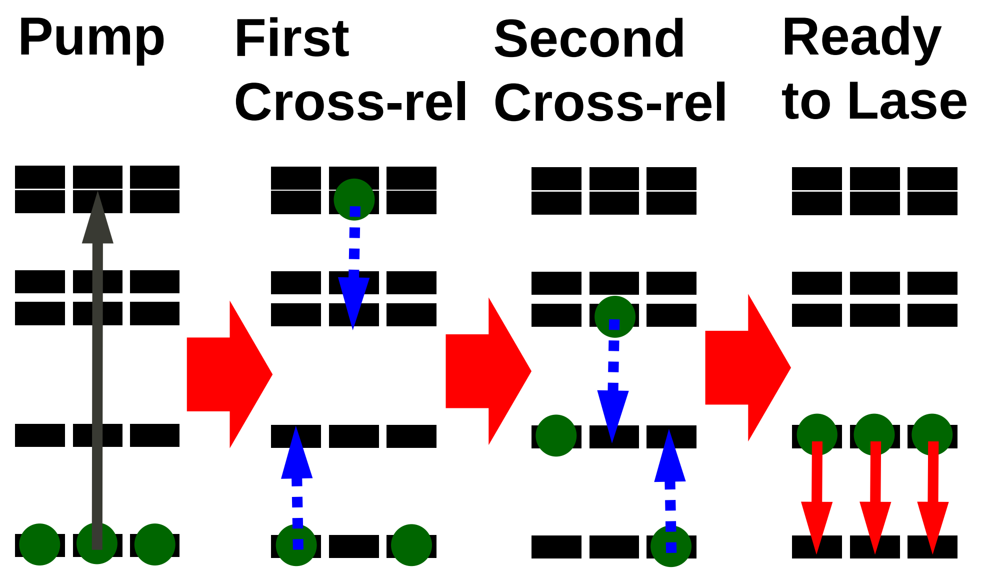
THE KEY: CROSS-RELAXATION

- Two ions close enough together
- Energy levels evenly spaced
- One can hand half its energy to the other – “cross relaxation”
- Thulium lasers use this to get two excited Tm^{3+} ions for each one pump photon – “2-for-1” excitation



To explain our strong mid-IR:

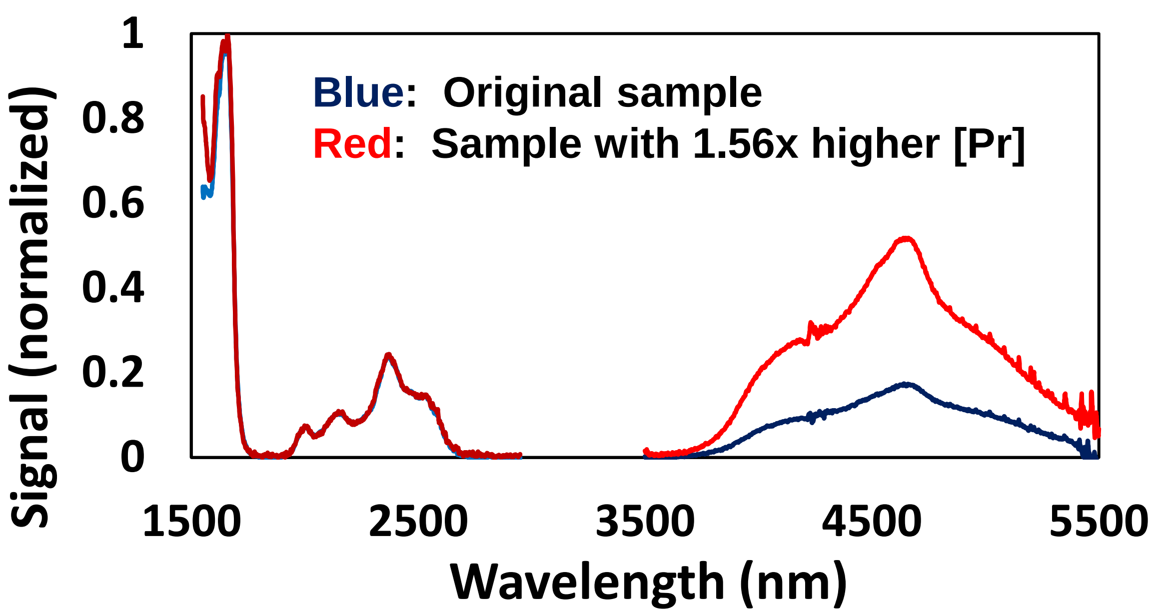
- **Must assume TWO similar processes**
- **Together: THREE ions excited for each ONE pump photon**



SUPPORTING EVIDENCE

- Modeling this process: Predicts correct fraction of the ions emit in the mid-IR band
- Also makes sense of temperature dependences of the upper state lifetimes, (much harder to explain without cross-relaxation)
- Recently: Sample of Pr:RPC with a higher Pr concentration
- Pr's closer together, so cross-relaxation should be even stronger

- Result: TRUE



SIGNIFICANCE FOR MID-IR LASERS

- MOST of the excited Pr ions seem to participate in this “3-for-1” process.
- So, nearly three Pr ions available to emit in the mid-IR for every one originally excited by the laser diode
- In laser terminology: nearly 300 % quantum efficiency
- Average photon in that band only about 1/3 the energy of pump photon – Normally would limit any laser efficiency to ~33 %
  (Even if all else were perfect)
- With this process, that ideal limit becomes ~100 %!
  (Of course, all else won’t be perfect, but this eliminates one huge limitation.)
- “2-for-1” process widely used to pump Tm lasers
- This “3-for-1” process potentially helps efficiency even more.

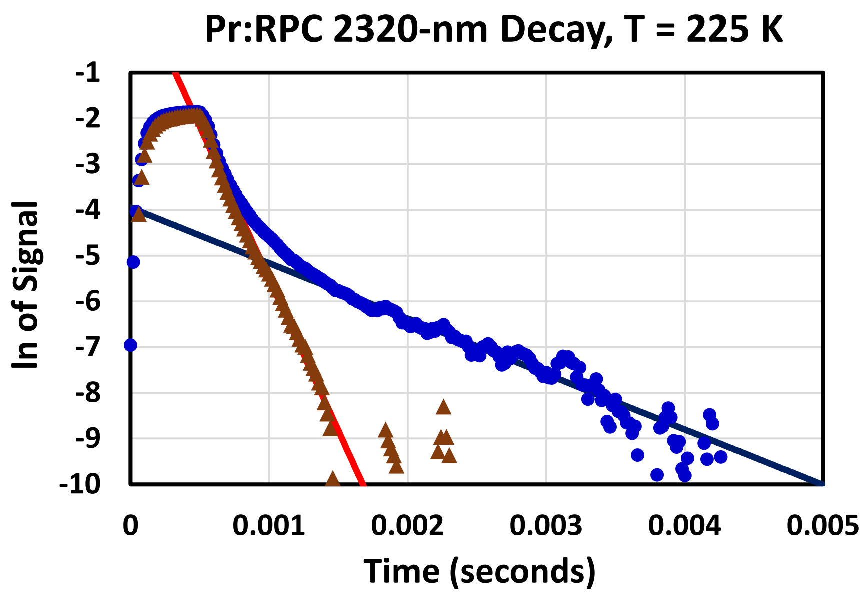
SUMMARY / CONCLUSION

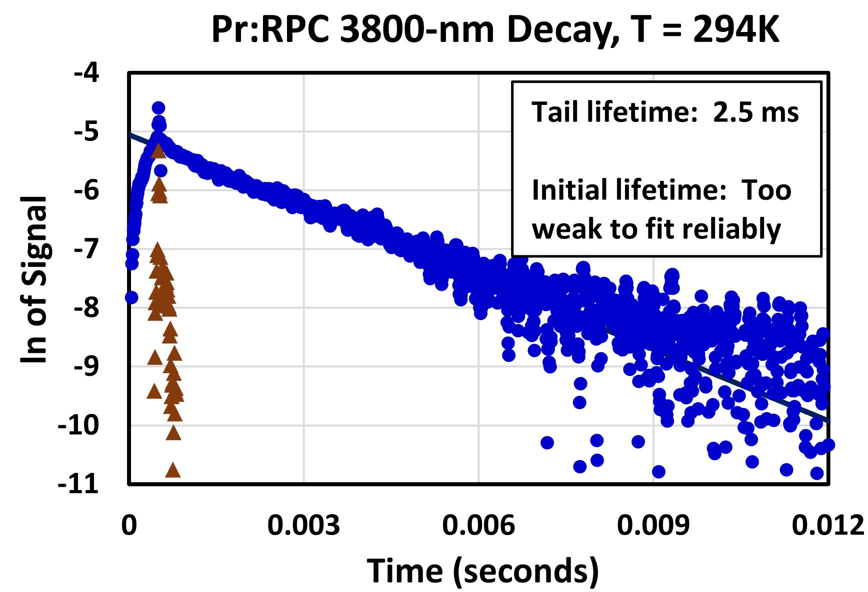
- Observed new process for pumping Pr^{3+} mid-IR laser ions
- Potential to dramatically improve pumping efficiency: "Three-for-one" excitation
- Can literally triple efficiency of converting pump photons into excited Pr ions available for lasing
- Next: Investigate other Pr-doped, low-phonon-energy crystals
  - Determine whether conditions enabling this interaction are common, or rare
  - Identify laser materials to optimize "3-for-1" pumping and other needed laser properties
- Potential path to efficient, low-SWaP mid-wave IRCM laser

BACK-UP SLIDES

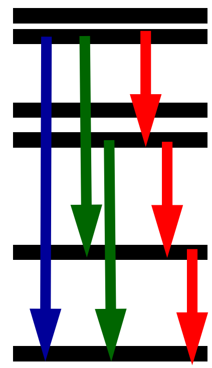
LIFETIMES FROM FLUORESCENCE DECAYS

Evidence for/against overlapping fluorescence transitions





- In this wavelength region, BOTH of the overlapping transitions contribute substantially.



- In this wavelength region, ONE of the three potentially overlapping transitions clearly dominates.

U.S. ARMY RDECOM ARL RESEARCH LABORATORY

FLUORESCENCE QUANTUM EFFICIENCY

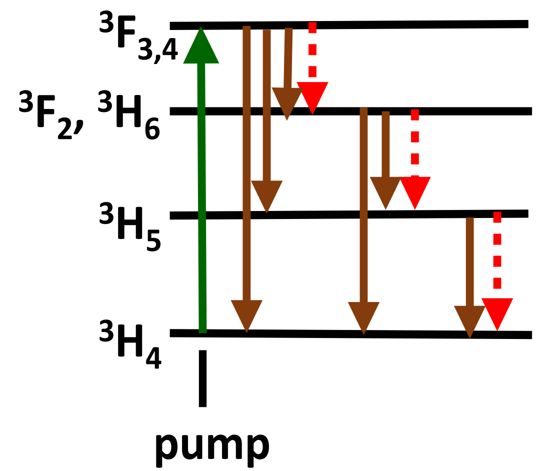
- Strength of absorption peaks, plus well-known theory, predicts the approximate "radiative" lifetime of each upper state
- Compare with observed lifetime: Estimates the quantum efficiency – fraction of the time each level decays radiatively

T (K)	**3F3** Lifetime (ms)	Quantum Efficiency if Tau(rad) = 0.58 ms	**3H6** Lifetime (ms)	Quantum Efficiency if Tau(rad) = 13.4 ms	**3H5** Lifetime (ms)	Quantum Efficiency if Tau(rad) = 30.4 ms
11	0.456	0.786	11.054	0.825	24.750	0.814
40	0.317	0.547	1.353	0.101	17.235	0.567
77	0.233	0.402	0.859	0.064	17.630	0.580
115	0.208	0.359	0.835	0.062	14.630	0.481
150	0.161	0.278	0.859	0.064	11.407	0.375
190	0.107	0.184	0.859	0.064	8.060	0.265
225	0.103	0.178	0.825	0.062	5.753	0.189
260	0.078	0.134	0.699	0.052	3.683	0.121
300	0.064	0.110	0.485	0.036	2.449	0.081

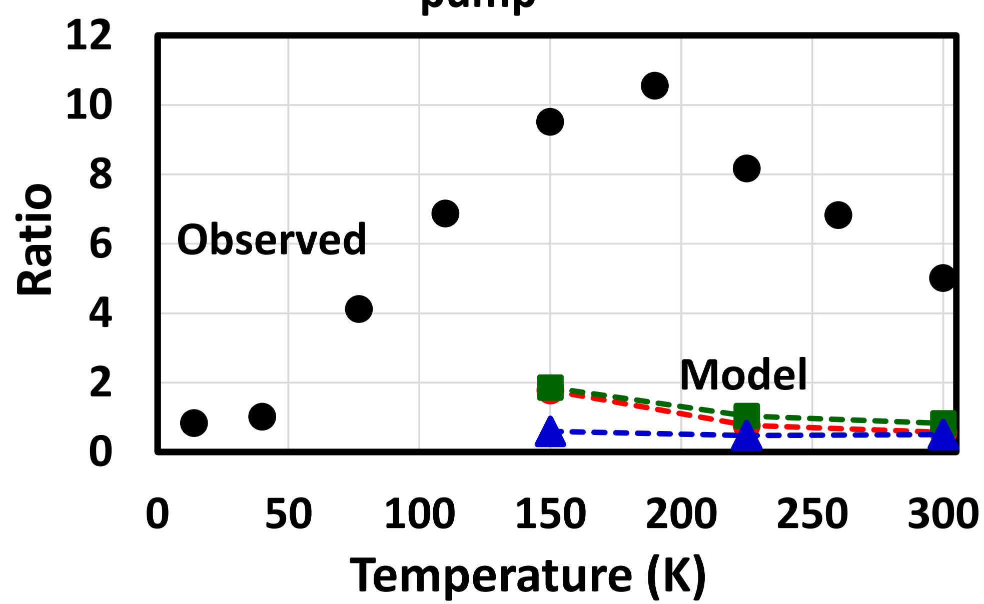
SIMPLE DECAY MODEL

(Does NOT include cross-relaxation – and does NOT fit the data)

- **Rate equations for each energy level identified and solved**
- **Solution arranged to give ratio of "photon flux" in the 3.6-5.5 micron band to that in the 1.5-1.8 micron band**

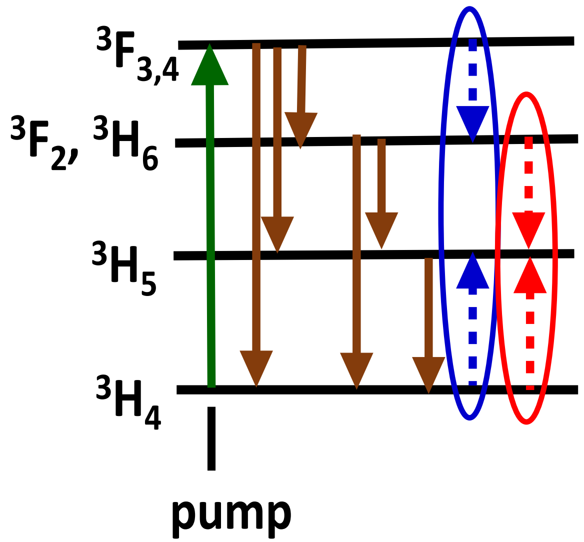


- **Different limiting cases calculated, then compared to the observed data**
- **NONE agrees with the observed data.**



CROSS-RELAXATION MODEL

- Rate equations for each energy level again identified and solved
- Solution arranged to give ratio of "photon flux" in the 3.6-5.5 micron band to that in the 1.5-1.8 micron band



- Different limiting cases calculated, again compared to the observed data
- Agreement with observed data now reasonable – and FAR better than in simpler model