# Engineering ultracold polar molecules by using optical lattices and microwave fields.

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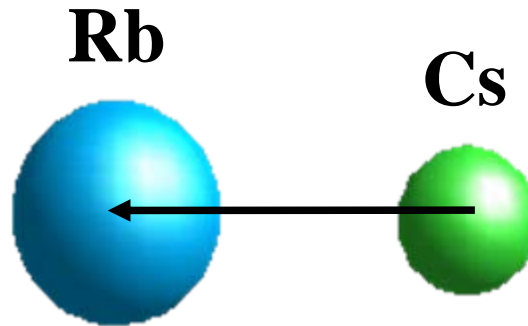
Collaborator:      Eite Tiesinga, NIST

# Polar molecules

The interest in polar molecules stems from the fact that polar molecules have permanent dipole moments. This allows them to strongly interact with an external electric fields and with each other by longe-range dipole-dipole forces.

**Rb**

**Cs**

Long-range **dipole-dipole** interactions **between** molecules can create new types of highly-correlated many-body states.

Polar molecules are considered to be promising candidates for implementation of quantum computing schemes.

# More about polar molecules

In all interesting applications polar molecules have to be **ultracold**
(T<~10-100 μK)

Historically, such cold molecules were obtained from laser-cooled atoms. First experiments produced dimers from the same atomic species.

Among first molecules created from different atomic species were KRb and RbCs.

They are the focus of our research.

# Approaches

We combine electronic and ro-vibrational structure calculations to obtain observable characteristics of polar molecules.

We use an *ab initio* relativistic configuration-interaction valence bond method, which I have developed over the last decade, to determine the electronic structures of small molecules. The method is well suited for description molecules formed from colliding atoms and calculates relativistic effects nonperturbatively.

This allows a realistic description of the avoiding crossings between potentials and determine **non-zero** dipole moments for transitions between the singlet and triplet states.
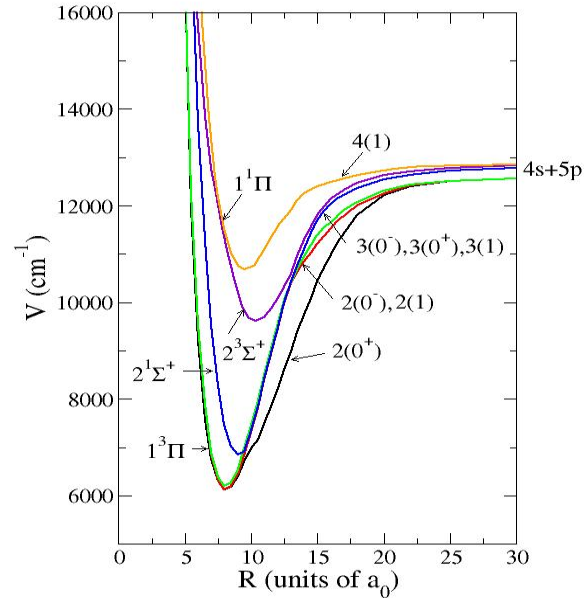
Ro-vibrational motion of molecules was described by solving the Schrödinger equation for potential surfaces using a discrete variable representation.
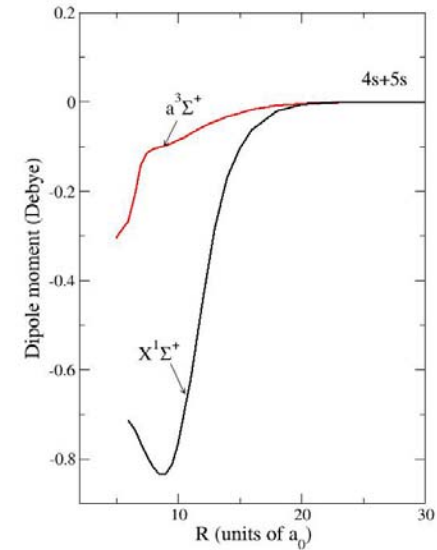
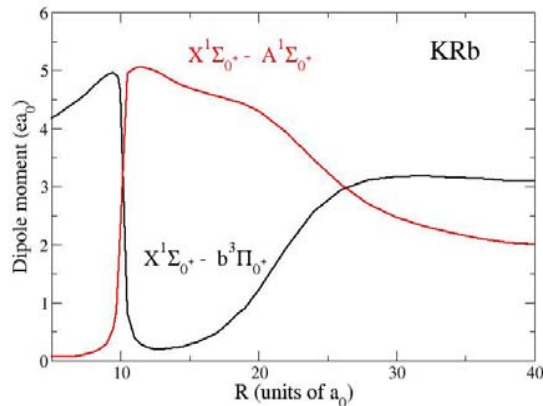# *Ab initio* data for KRb
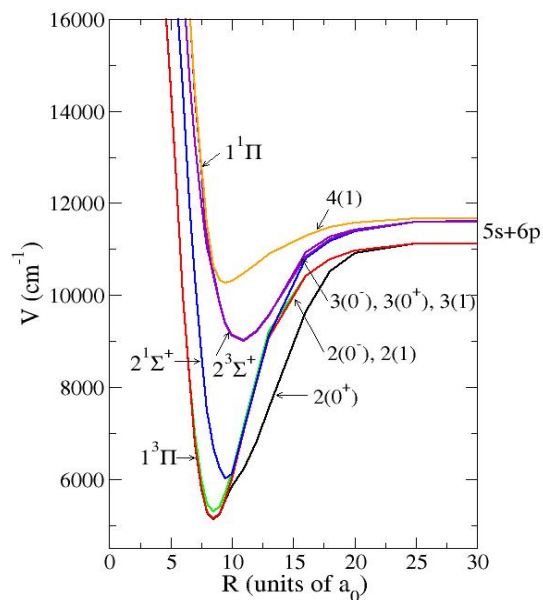
Ground state potentials
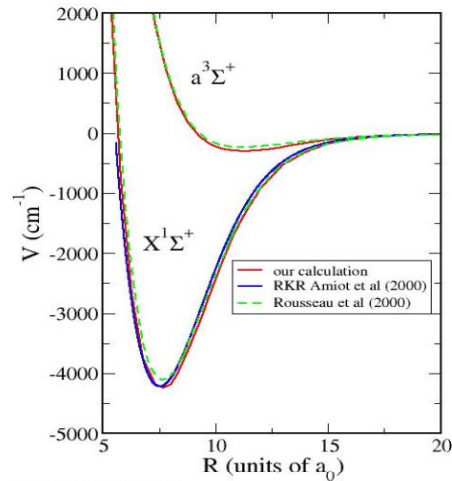
Permanent dipole moments
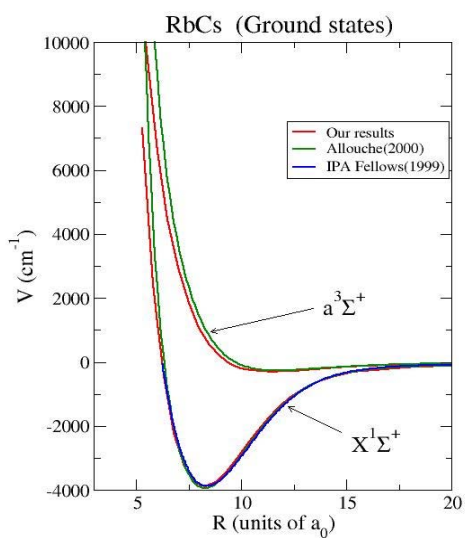
Excited state potentials

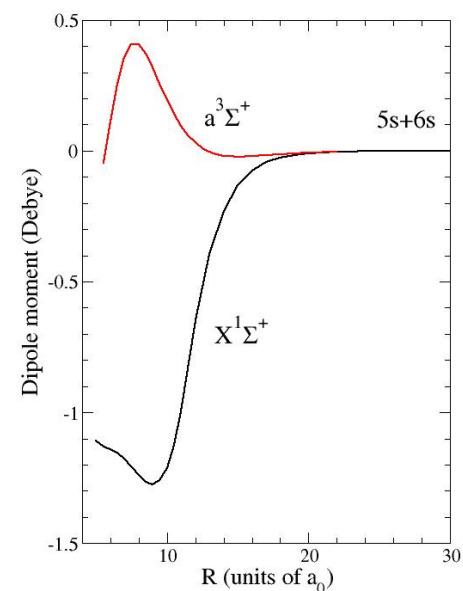Transition dipole moments

# Ab initio data for RbCs

Ground state potentials

Permanent dipole moments



Excited state potentials
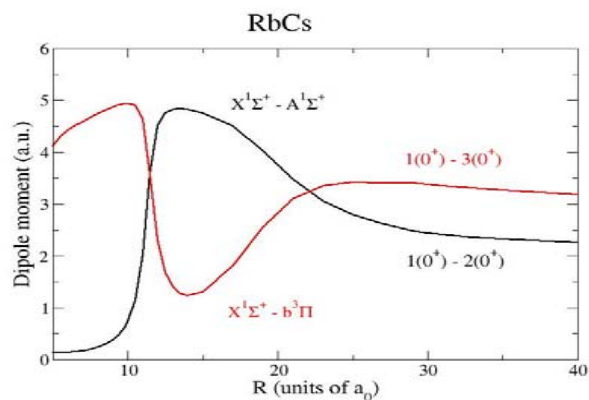
Transition dipole moments

# Production of polar molecules

Methods to associate two cold atoms into one cold molecule:

- Using magnetic Feshbach resonances
- Photoassociation
- Radio-frequency magnetic-dipole association

In most cases molecules are created in highly-excited vibrational levels of the ground state. However, it is often preferable for molecules to be vibrationally cold.

This was achieved in photoassociation experiments by DeMille's group [PRL 94, 203001 (2005)]. They formed polar RbCs in the v=0 level of the ground potential, although the number of vibrationally cold molecules was not large.

New developments in photoassociation [Rom *et al.* PRL 93, 073002 (2004)] show great promise to increase the molecular production rate by forming molecules in an optical lattice.

In optical lattices atoms and molecules are isolated from destructive perturbations

Tight confinement of atoms in a lattice is expected to increase production rates.

# Molecules in optical lattices

Futhermore, most of applications of polar molecules are based on optical lattices to hold molecules in fixed spatial locations.

For example, polar molecules confined in a periodic array of optical potential can simulate solid state systems with well defined degrees of motion.
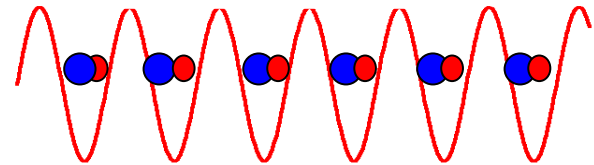
Parameters of the lattice can be externally controlled by tuning frequency and intensity of the lasers.

The interaction between polar molecules can also be tuned by external electric and microwave fields.

# Association in optical lattice

We suggest an association scheme to create polar molecules in an optical lattice with a combination of microwave and optical fields.

In particular, we will use the unique property of polar molecules, the non-zero permanent dipole moment. It allows dipole transitions between ro-vibrational levels of the ground potentials.
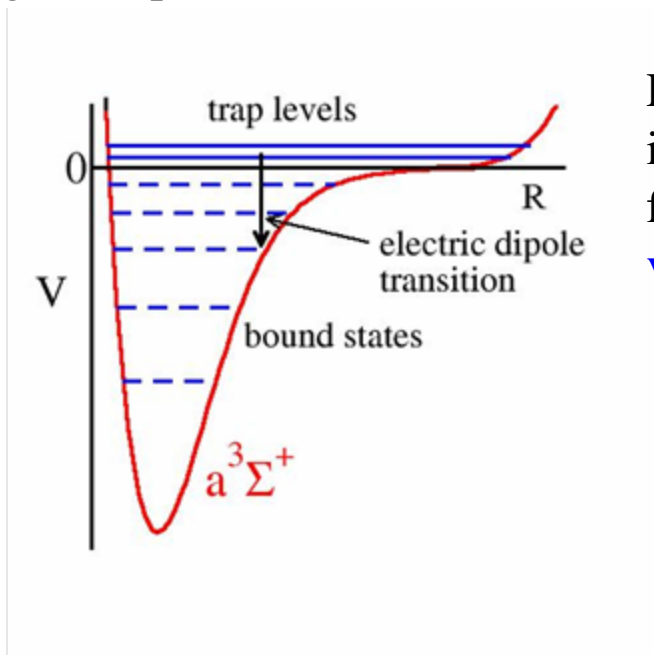


First, we use microwave fields to create molecules in high-vibrational states of the ground configuration followed by one optical Raman transition to form the vibrationally cold molecules.
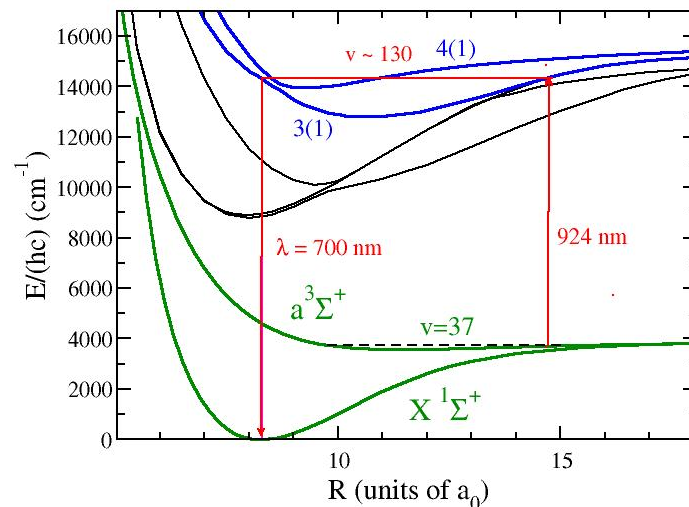


**RbCs**

# Trapping potential

Rb and Cs atoms are confined in sites of an optical lattice. This confinement can be described by a spherically symmetric harmonic trap. This also leads to a harmonic trapping potential for the relative motion between the atoms.
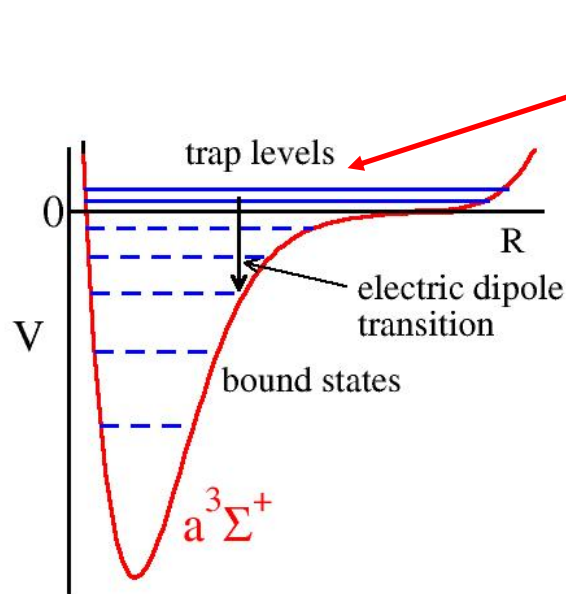


Condition: both atoms are double polarized. Then only $a^3\Sigma^+$ potential is important

The bottleneck in any molecule forming process from its constituent atoms is the first transition from the two free atoms to weakly-bound ``floppy'' molecule.

# More about association

Dipole moment for microwave transitions from lowest trap level of a harmonic potential with a frequency of 200 kHz (this is a tightly confining lattice).



Binding energy $a^3\Sigma^+(cm^{-1})$
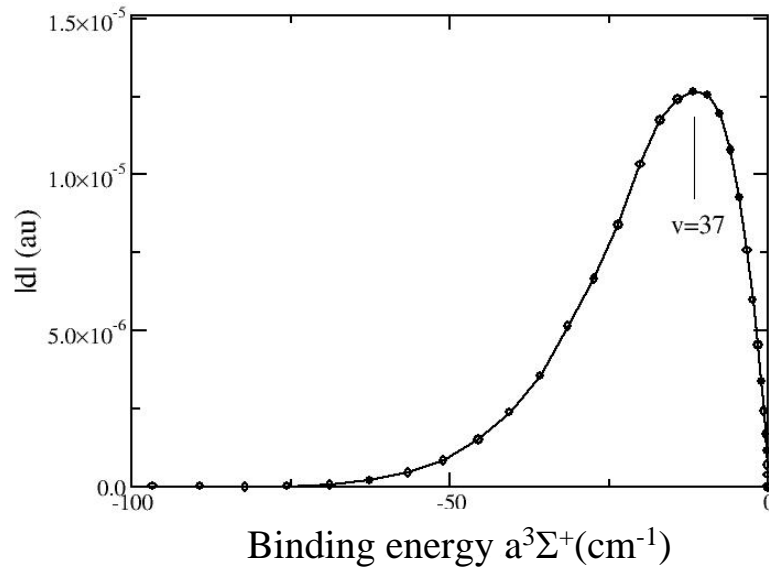
This data is for N=0 to N=1 rotational transitions within the $a^3\Sigma^+$ potential

Maximum dipole moment for v=37 with a binding energy of 10 $cm^{-1}$ . This is the same intermediate level as used in DeMille's experiment
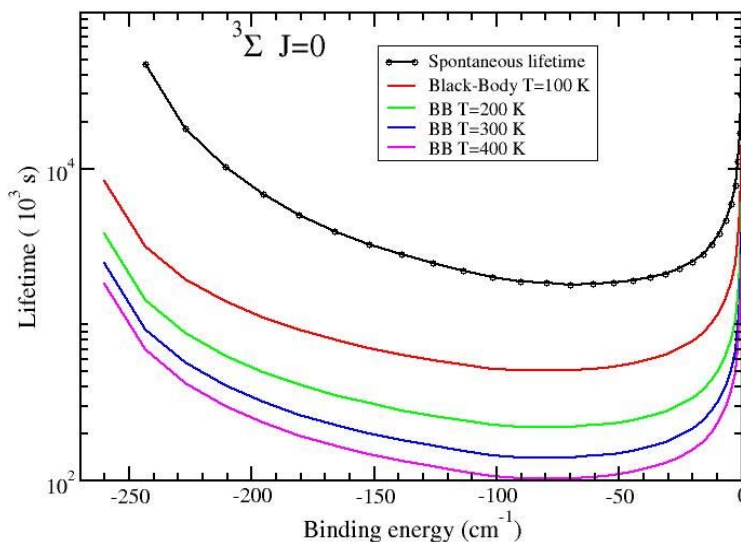
$B_{v=37} = 200$ MHz

In SI units this maximum corresponds to ~200 Hz/(W/cm$^2$).
In other words, oscillation periods < 1 ms

# Black-body radiation

Thermal radiation from the environment can cause a polar molecule to change ro-vibrational level.
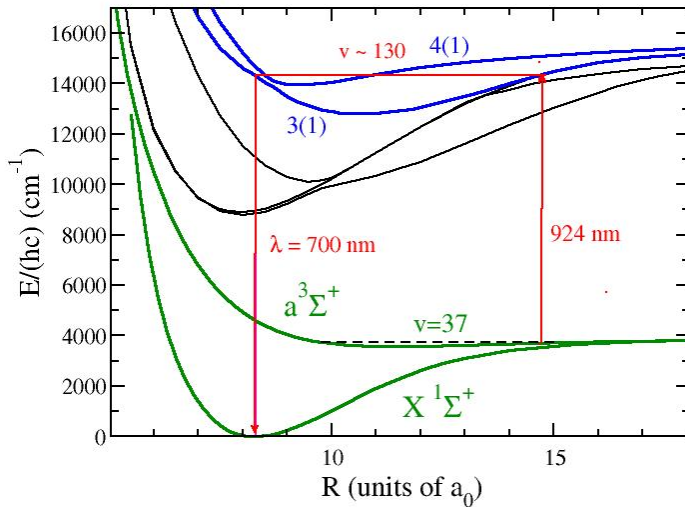
However, from our calculations we find that this lifetime is very long ( $> 10^5$ s)



This large lifetime allows us to perform the association in two steps.
  1. Microwaves create a sufficient number of v=37 $a^3\Sigma^+$ molecules.
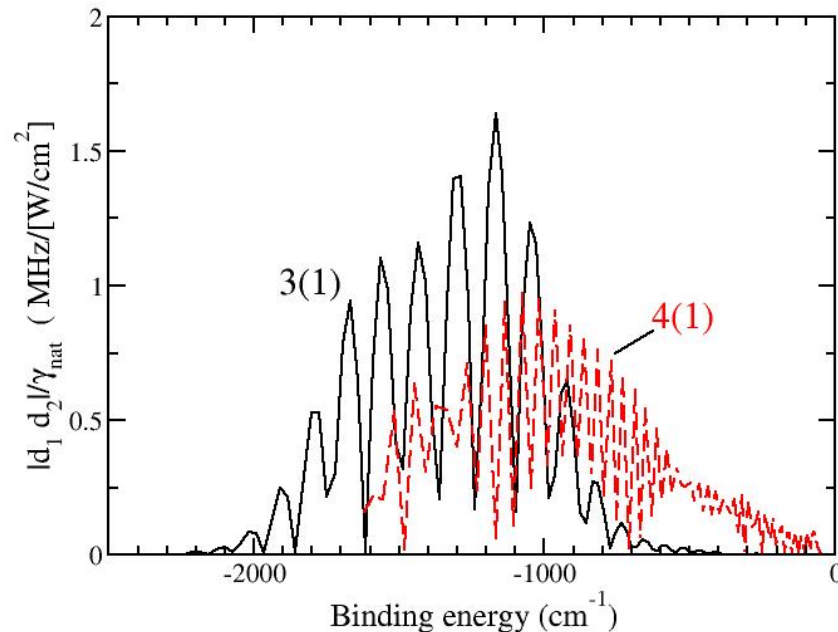  2. Apply optical field to convert to vibrationally cold molecules

# Optical Raman transition



Transition from v=37 $a^3\Sigma^+$ to v=0 $X^1\Sigma^+$
via vibrational levels of the excited 3(1) or 4(1)
potentials

Maximum corresponds to
$10^6 (I_1 I_2)^{1/2}$ 1/s
with intensities $I_1$ and $I_2$ in
$W/cm^2$

This Raman transition is the
same as used by DeMille's
group.

# Molecular Polarizability

We determine lattice parameters that can insure confinement of both molecules and individual atoms from which molecules were created.

Main property of an atom or a molecule relevant for trapping by an optical lattice is the atomic or molecular dynamic polarizability $\alpha$ as a function of laser frequency $\omega$.

The optical potential $V_0$ seen by the atoms or the molecules is

$$V_0(x) = -\alpha(\omega)\ I(x) = -\alpha(\omega)\ I_0\ \sin^2(kx) \sim \alpha(\omega)\ I_0\ k^2x^2$$

I.e. the molecular polarizability determines the depth of the lattice potential and contributes to laser-induced decoherences.

# Conditions on atomic and molecular $\alpha(\omega)$

During the photoassociation process and afterwards we would like the atoms and molecules to be tightly trapped in the lattice sites

A natural choice is that they see the same trapping potential, ie have the same polarizibility at a certain laser frequency.

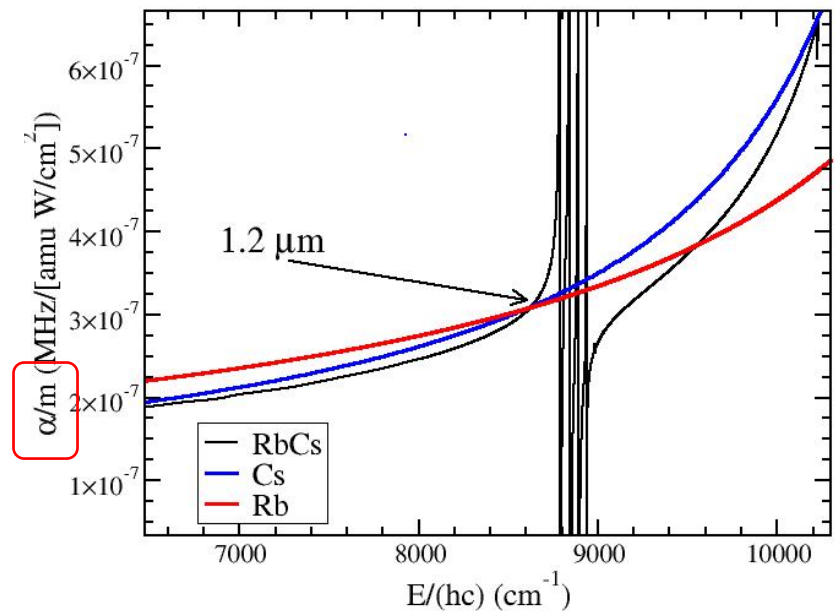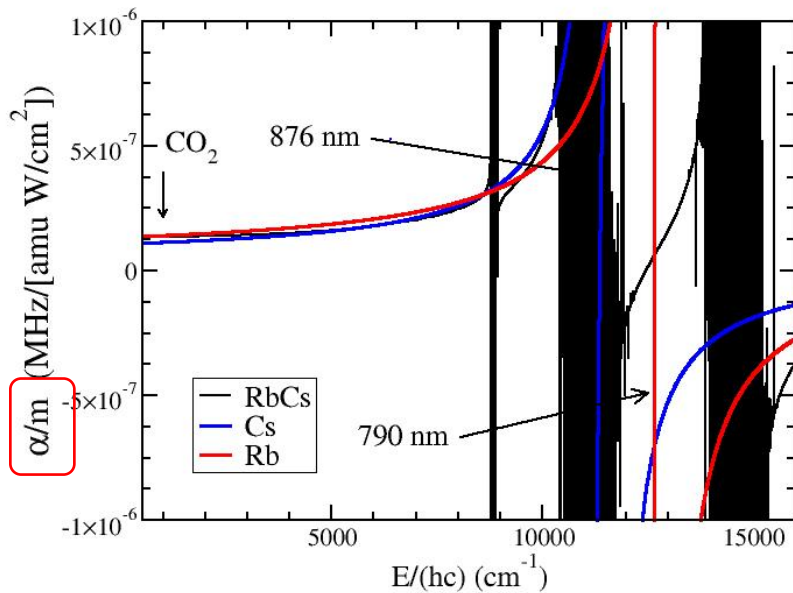The molecule is v=0 J=0 X $^1\Sigma_+$
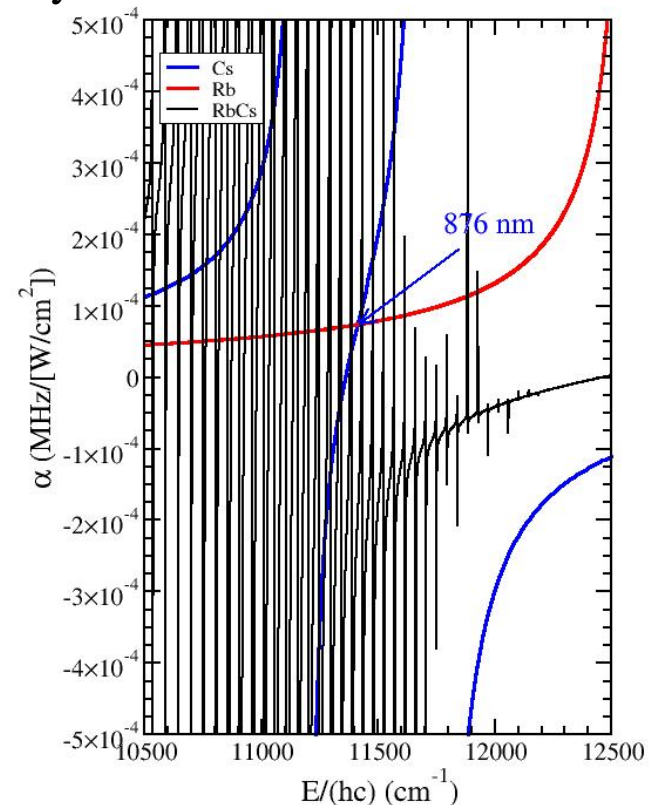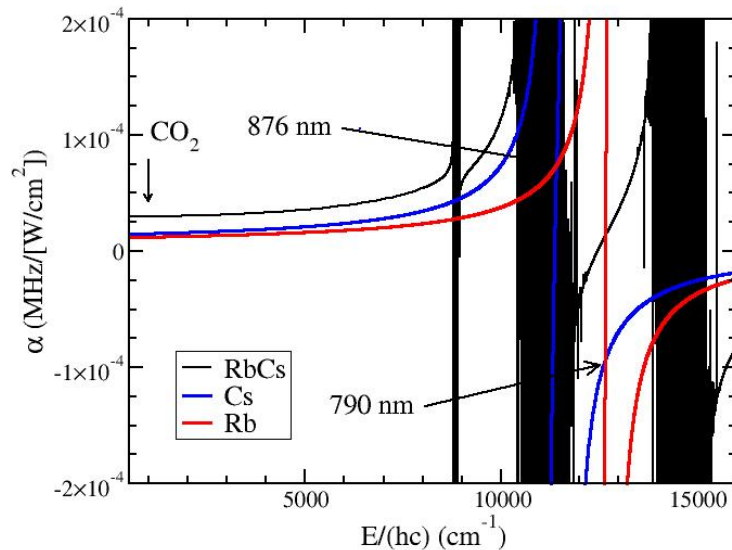


We looked and could not find such a point.

# More conditions II

Another choice is that the atoms and molecules have the same trapping frequency. The condition is:
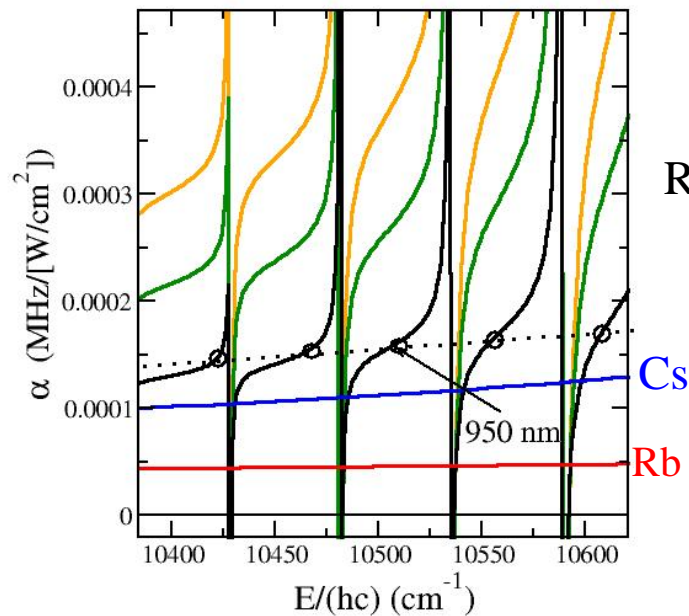
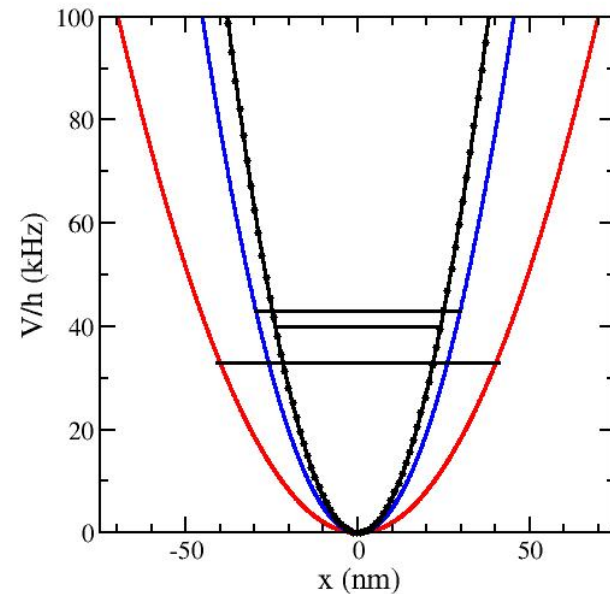$$\alpha_{Rb}/m_{Rb} = \alpha_{Cs}/m_{Cs} = \alpha_{RbCs}/m_{RbCs}$$

# More conditions III

A final choice is that the center-of-mass motion of the atoms and molecules have the same trapping frequency and their spatial extent are the same . The condition is:

$$\alpha_{Rb} + \alpha_{Cs} = \alpha_{RbCs}$$

RbCs v=0, J=1, $X^1\Sigma^+$

# Controlling molecules in an optical lattice

Another goal is to determine lattice parameters that ensure strong trapping forces and simultaneously small decoherent rates.

Localization of the molecule in a lattice site requires a deep lattice potential (tens of kHz) to prevent tunneling. This can be achieved by increasing laser intensity. Higher laser intensities increase the possibility of exciting a molecule leading to larger decoherence processes.

We analyze the frequency dependence of polarizability for KRb and RbCs and demonstrate the existance of frequency windows where ro-vibrational spectrum of molecule has negligible contribution to polarizability. In such frequency windows the Franck-Condon factors with the ground state are small and light-induced decoherences are kept to a minimum.

# Real and Imaginary Polarizability

Windows interspersed with with regions of multiple resonancies. The resonancies are due to bound states of excited potentials and related to large dipole moments when inner- and outer-turning points of vibrational levels of initial and final states agree.
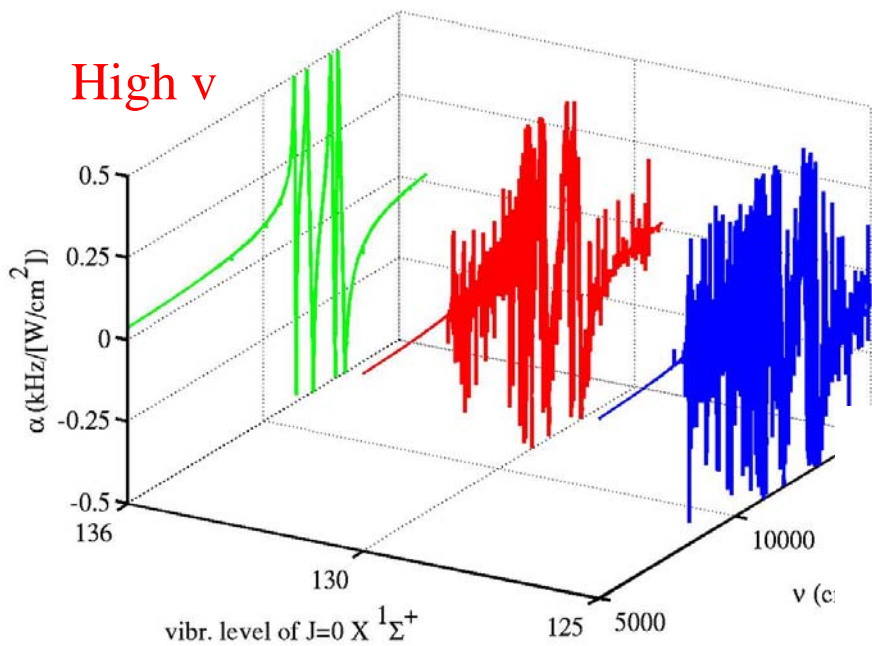


**KRb**

The ratio between real and imaginary parts is $10^6$ -$10^7$ and decoherence is expected to be small.

$\lambda = 810$ nm; @ $I = 10^4$ W/cm$^2$; $V_0 \sim 120$ kHz
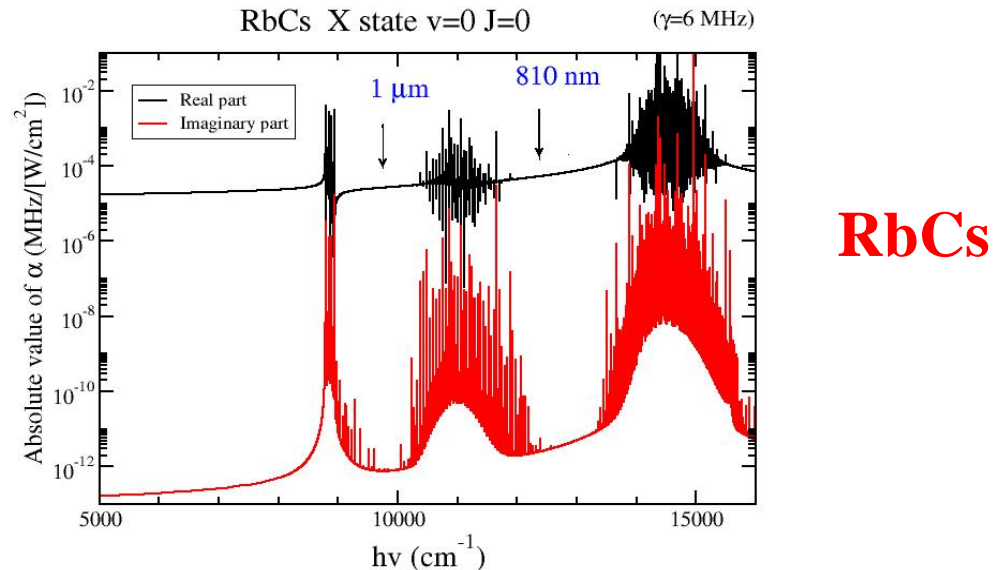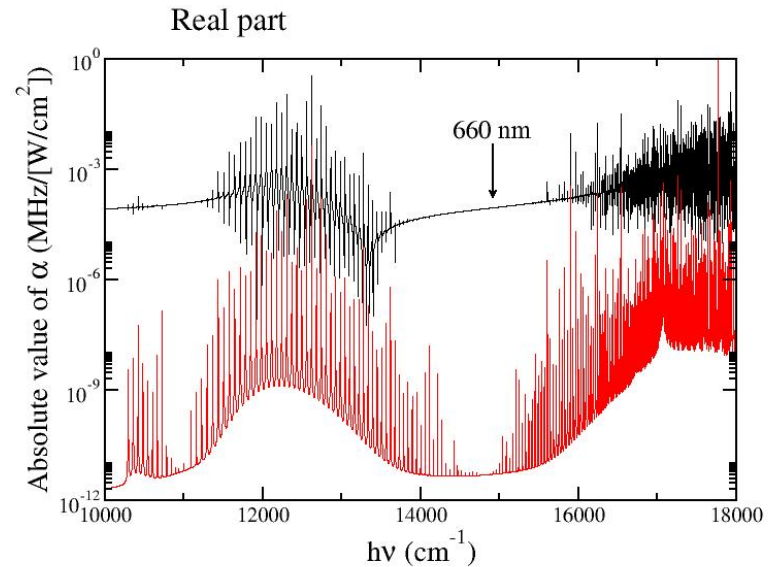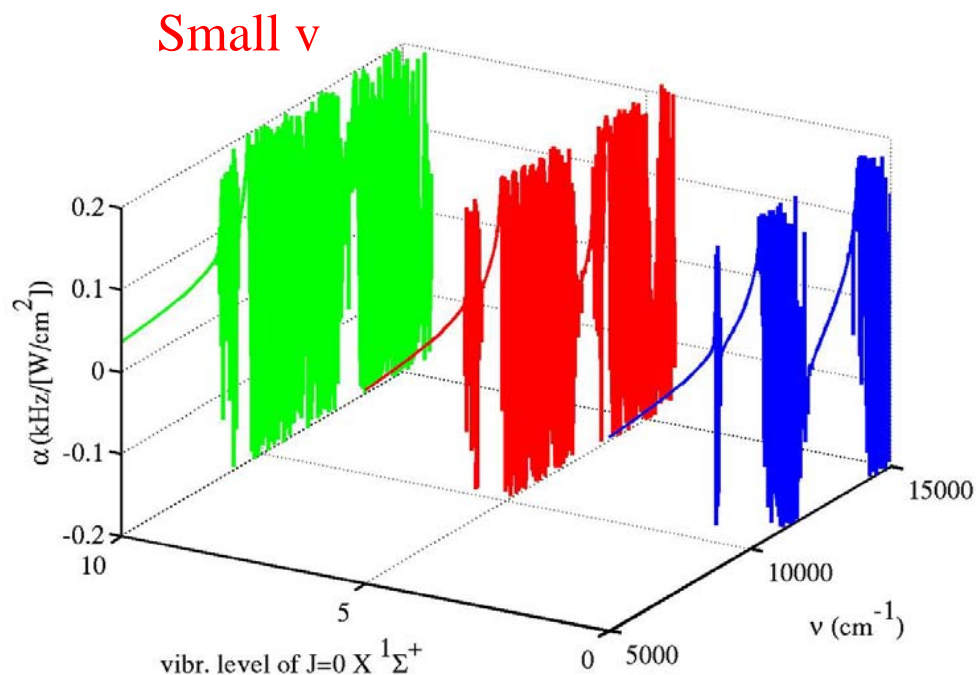


**RbCs**

# Vibrational dependence of $\alpha$

High v

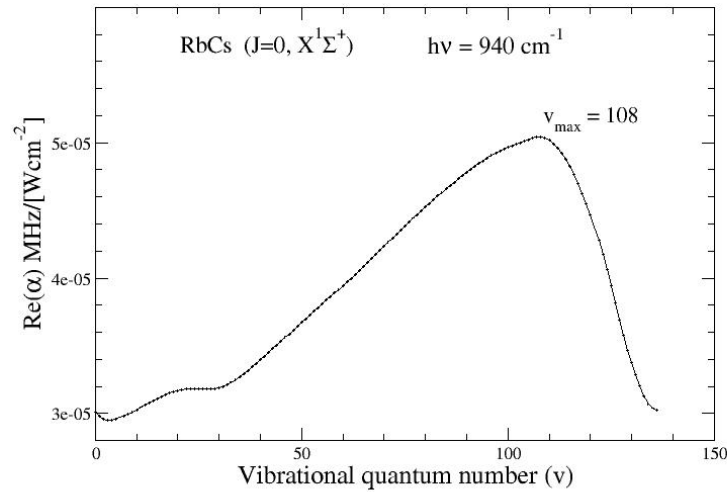For J=0 levels of the $X^1\Sigma^+$ state of RbCs

Small v

For increasing v windows disappear, and we see more resonant structure.

For weakly bound levels $\alpha$ becomes the sum of the atomic polarizabilities.
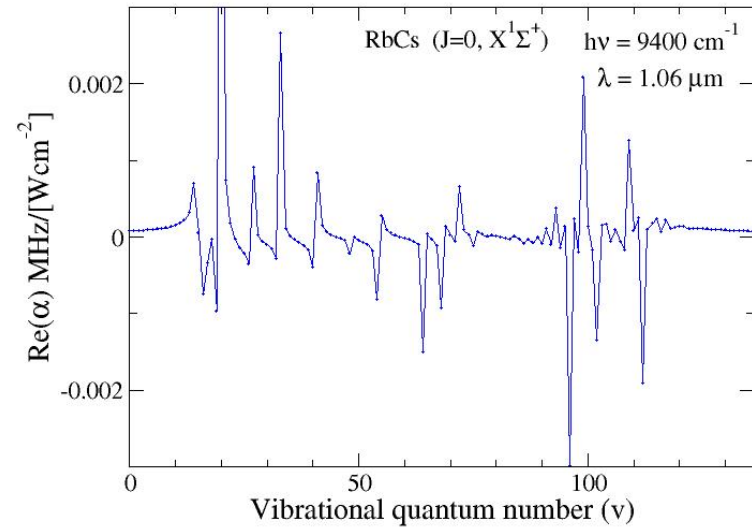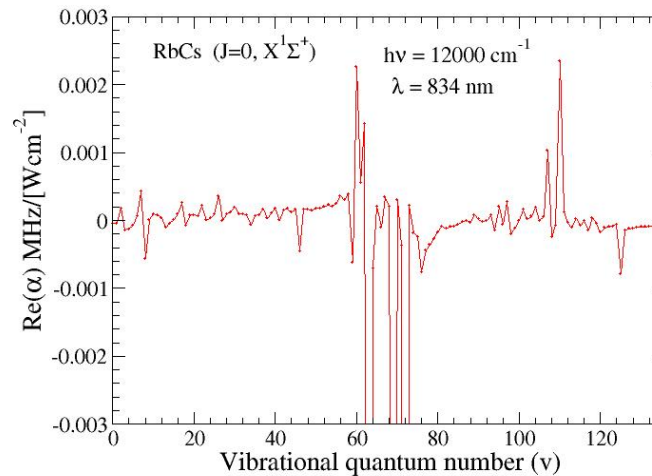
# Continued

CO$_2$ laser light



YAG laser light



visible laser light

# Controllable dipole-dipole interactions

We show that microwave fields can induce a tunable dipole-dipole interaction between rotationally symmetric (J=0) molecules. Even though polar molecules have a permanent dipole moment, for any J=0 rotational state it averages to zero.

However, a dipole moment is easily induced by applying a microwave field with ω ~ the J=0 to J=1 resonance.



**KRb**
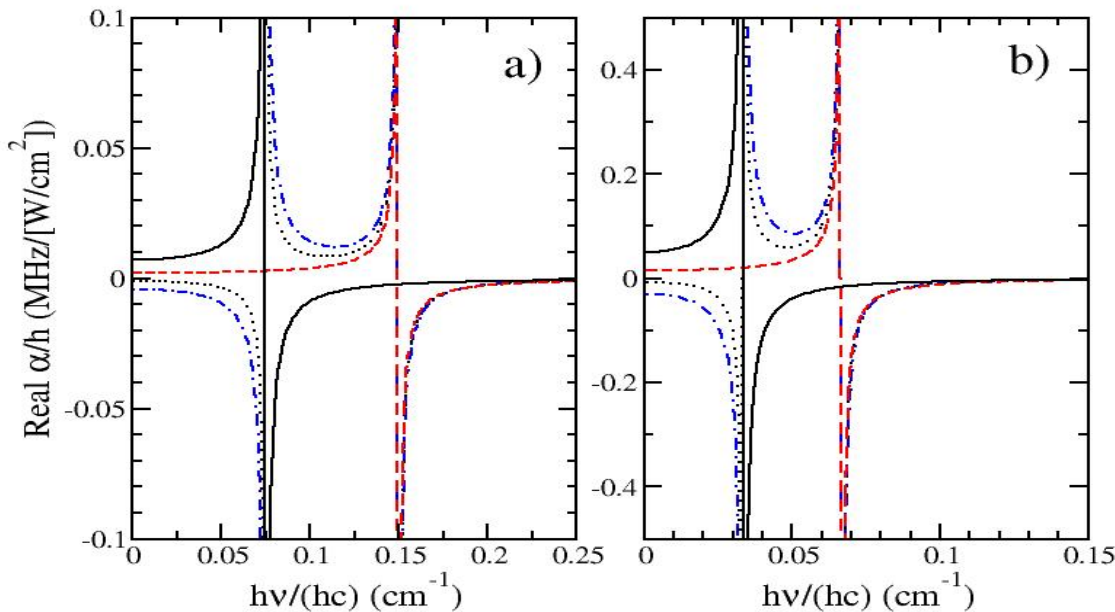
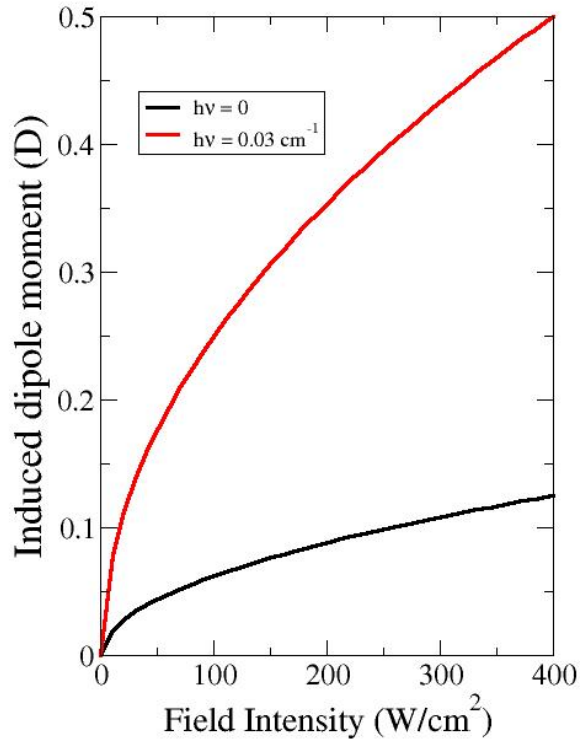**RbCs**

The J=0 and J=1 polarizabilities differ and even have opposite sign except ω = 0.3 cm$^{-1}$ for KRb and 0.1 cm$^{-1}$ for RbCs.

There microwave field creates the same Stark shift for a J=0 and J=1 level.

# Induced dipole moments



For a microwave field resonant with the J=0 to J=1, the dressed J=0 molecular state has a induced dipole moment of one half of the permanent dipole moment of the v=0, J=1 state.

Assuming that polar molecules interact via

$$V_{dd} \sim d_{ind}^2/(\lambda_L/2)^3$$

The characteristic interaction time is

$$h/V_{dd} \sim 2 \text{ ms}$$

This time is enough to establish entanglement of molecules during ultracold experiment with typical time scale of a few 100 ms.

# Conclusion

- Despite the fact that molecules have an internal structure that is more complex than atoms, this complexity can be used to good advantage or neutralized.

- Microwave fields can be used to create high-v molecules from ultracold atoms. A Raman transition converts these to vibrationally cold molecules.

- We compared atomic and molecular polarizability to find laser frequencies where they see the similar trapping potentials.

- For vibrationally cold molecules we found frequency windows in the optical domain where the decoherences are kept minimal.