### On the Feasibility of Laser-Driven Control of Nuclear Dynamics

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# Nuclear excitation experiments in the past

- Nuclear physics has had a long, colorful history (starting from the beginning of the 1900s)
- Largest body of laboratory work has focused on spectroscopy, determining energy spectra and multipole moments, and nuclear reactions.
- Theoretical interest has been focused mostly on the creation of numerous models of nuclear structure, nuclear forces, and describing the many types of nuclear reactions.
- Until now, the sole method of nuclear excitation has been indirect, i.e., with the use of secondary excited particles.
- Reasons:
  - □ Large transition energies compared to atomic transitions
  - □ Laser technology

#### **Direct laser-nucleus interactions**

- The increasing power of current-day/near-future lasers has opened new doors to the study of such interactions.
- Salient features:
  - □ More control and flexibility with types of radiation
  - Analogous to direct laser-atom/laser-molecule experiments
  - Relatively simple interaction
  - Allows for direct manipulation, preparation, and measurement of many nuclear properties

#### **Direct laser-nucleus interactions**

- While there are clear parallels with laser-atom interactions, there are also important differences:
  - □ Length scale: ~ 1-100 fm
  - □ Energy scale: ~ 10 keV 30 MeV
  - While for atoms, E1 transitions dominate, for nuclei, both E1 and M1 (as well as higher orders) become important
  - □ Complex mixing between various degrees of freedom
  - Often no clear cutoff between dipole allowed and dipole forbidden transitions

#### Nuclei meet Quantum Control

- Quantum control seeks not only to observe, but actively manipulate the course of physical and chemical processes governed by the laws of quantum dynamics.
- Exploits *direct* interactions of target with laser pulses, tailored specifically for optimizing a particular objective.
- Large body of work in the atomic/molecular realm focuses on achieving quantum control of excited-state transitions and populations (almost exclusively within dipole approximation)—E1 transitions
- Fundamental control problem at hand:

 $P_{i \to f}[\epsilon(t)] = 1.0$ 

- Do the principles governing controllability of transitions extend down to the nuclear realm? Differences and similarities?
- Focus: Both E1 and M1 transitions

#### **Research objectives**

#### Nuclear dynamics

- Choose an appropriate nuclear model, Hamiltonian, eigenfunctions, eigenvalues, excited state spectra
- □ Elucidate laser-nucleus interaction mechanics, dipole approximation?
- Transition control
  - Develop necessary formalism for describing E1 and M1 transitions, selection rules, dipole matrix elements
  - Formally evaluate the controllability of nuclear transitions ("in principle" calculation)
- Feasibility/ practicality
  - What kind of lasers (intensity, frequency, etc.) does it take to demonstrate this in a lab? Possible experimental set-ups
  - □ Which nuclei are applicable?
  - □ Possible applications, further methods of study....

#### Nuclear Models

- A large number of models, which can best be described as analogies with macroscopic objects, attempting to capture key properties of particular types of nuclei. The vast majority of these theoretical models are "semirigorous", mostly due to the following fact:
  - Nuclear lie in a no-man's land (too small for statistical mechanics, too large for effective bottom-up theories => very complex many-body problem)
- A completely formal and foundational model for nuclear structure has yet to be developed.

#### **Collective Models**

- The earliest models of nuclear structure -- collective models, based on a liquid-drop analogy, first proposed in the 1930s.
  - Surface often defined by the surface coordinate R through expansion by spherical harmonics. Good for describing nuclear vibrations and complex deformations (high multipole order)
- A second type of collective model assumed the nucleus to be a rigid rotor, a model very prevalent in the study of atomic and molecular dynamics.
  - Spectroscopic data showed that the relative spacing and location of certain nuclear excited states matched rigid-rotor model predictions.

#### Shell Models

- A separate kind of model was developed for the description of single-particle phenomena – the shell models
- The concept of nucleons occupying in well-defined shells is analogous to the well-known s-p-d-f shell structure in atomic physics
- Neutrons and protons separately fill up their own set of shells as dictated by the Fermi exclusion principle
- Experimental support: notable discontinuities seen in binding energies, that occur at the so-called "magic numbers" – shell closures.
- +Spin-orbit coupling correction

#### Particle-plus-rotor Model

- In this research, the particle-plus-rotor model (PRM) was used as the model of nuclear structure.
- Within this model, the nucleus is envisioned as follows: a small number of valence nucleons (typically only 1) are coupled to an even-even rigid rotor core that encompasses all remaining nucleons.
- The Hamiltonian used to describe the nucleus is correspondingly divided into two parts: the rotor Hamiltonian, and the intrinsic/valence Hamiltonian.

 $H = H_{val} + H_{rot}$ 

- The former describes the rotation of the rotor as well as demonstrates the strength of coupling between the valence particles and the core.
- The latter can take on many different forms, ranging from the most foundational shell model Hamiltonian to the Nilsson potential.

### Why the PRM?

- Experiments show that many important nuclear properties can be determined by just considering the socalled "valence" nucleons.
- Both rigid-rotor type and single-particle contributions to the energy spectra.
- While the total angular momentum and spin of a nucleus can be determined largely by the valence nucleons, only collective, rotational models of nuclei can explain the relatively low excitation energies of many nuclear transitions.
- Flexibility with degree of coupling and valence nucleon wavefunctions

#### Focus nuclei

- Which nuclei are most appropriate for our application of PRM?
  - □ Heavy metals, rare-earths (actinide series, in particular)
  - □ Odd-A nuclei (only one valence nucleon)
  - □ Strong-coupling axially-symmetric case: deformation alignment.
- These nuclei tend to show good agreement between experiment and theory.
- Relatively low-energy excited states good for setting up feasible laboratory experiments with current/near-future lasers facilities.

### PRM Hamiltonian (1)

#### The rotor Hamiltonian:

$$H_{rot} = \frac{\hbar^2}{2\mathcal{I}} [\mathbf{I}^2 + \mathbf{j}^2 - \mathbf{I}_3^2 - \mathbf{j}_3^2] - \frac{\hbar^2}{2\mathcal{I}} (\mathbf{I}_+ \mathbf{j}_+ + \mathbf{I}_- \mathbf{j}_-)$$

- This represents an axially symmetric rotor with a particle strongly coupled to the rotor's deformation axis (includes Coriolis term)
- I=total nuclear angular momentum
- j= particle' s total intrinsic angular momentum
- R = rotor angular momentum

$$I = R + j$$

### PRM Hamiltonian (2)

The intrinsic Hamiltonian (Nilsson model):

$$-\frac{\hbar^2}{2m}\nabla^2 + \frac{m}{2}\omega_{\perp}^2(x^2 + y^2) + \frac{m}{2}\omega_z^2 z^2 + C\mathbf{l} \cdot \mathbf{s} + D\mathbf{l}^2$$

There are numerous parameters, which are defined as follows:

 $C = -2\hbar \mathring{\omega}_0 \kappa, \qquad D = -\hbar \mathring{\omega}_0 \kappa \mu, \qquad \mathring{\omega}_0^3 = \omega_\perp^2 \omega_z$ 

- Values for these parameters can be looked up in various textbooks on nuclear physics are in the literature.
- Eigenfunctions are typically represented as a superposition of isotropic harmonic oscillator functions, with expansion coefficients found in tabulated compilations in the literature.



#### Eigenfunctions/ eigenvalues

While in the Hamiltonian consists of a sum of rotor and valence parts, the eigenfunctions for PRM involve a product of the two parts:

$$IMK\rho\rangle = \left[\frac{2I+1}{16\pi}\right]^{\frac{1}{2}} (D_{MK}^{I}|K,\rho\rangle + (-1)^{I-j}D_{M,-K}^{I}|-K,\rho\rangle)$$

The corresponding energy eigenvalues are:

$$E^{i}(I,K) = \frac{\hbar^{2}}{2\mathcal{I}}[I(I+1) - K^{2} + \delta_{K,\frac{1}{2}}(-1)^{I+\frac{1}{2}}(I+\frac{1}{2})a^{i}] + E^{i,(val)}_{\rho}(K)$$

With a, often called the decoupling parameter, given by:

$$a^{i} = -\sum_{nj} |C_{nj}^{i}|^{2} (-1)^{j+\frac{1}{2}} (j+\frac{1}{2})$$

#### Laser-nucleus interaction Hamiltonian

- A system comprised of a nucleus inside of an external electromagnetic field characterized by its fourdimensional potential,  $A_{\mu} = (\Phi, A)$  and corresponding electric and magnetic fields is described by the Hamiltonian:  $H = H_{nucl} + H_{field} + H_{int}$
- The first term represents the PRM Hamiltonian, while the second term describes the field itself:

$$H_{field} = \frac{1}{8\pi} \int (\mathbf{E}^2(\mathbf{r}, t) + \mathbf{B}^2(\mathbf{r}, t)) d^3r$$

The last term is the most important, representing the interaction of the external EM radiation with the nucleus:

$$H_{int} = -\frac{1}{c} \int j_{\mu} A^{\mu} d^3 r$$

#### **Dipole approximation**

#### Dipole approximation is valid in this research:

 Laser types considered all have wavelengths greater than that of hard x-ray beams (λ>10 pm)

Coherent radiation source

Largest nuclei have r~15 fm

Within the dipole approximation, the interaction Hamiltonian can be greatly simplified to:

$$H_{int} = \int \left( \rho(\mathbf{r}, t) \Phi(\mathbf{r}, t) - \frac{1}{c} \mathbf{j}(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}, t) \right) d^3r = -\mathbf{P}(t) \cdot \mathbf{E}_0(t) - \mu(t) \cdot \mathbf{B}_0(t) + \cdots$$

#### **Dipole approximation**

• The electric dipole operator, P, is given by:

$$\mathbf{P}(t) = -\int d^3r \mathbf{r} \rho(\mathbf{r}, t)$$

In our analysis of transitions, the important quantities are the transition dipole moments, given by the appropriate diagonal matrix elements of the dipole operator, so for the case of E1 transitions:

$$\langle f|H_{int}(E1)|i\rangle = \langle f| - \mathbf{P}(t) \cdot \mathbf{E}_0(t)|i\rangle = -\mathbf{E}_0(t) \cdot \langle f|\mathbf{P}(t)|i\rangle$$

• Within PRM, the magnetic dipole operator,  $\mu$ , has the special form:

$$\mu = \sum_{i=1}^{n} \mu_N [g_l^{(i)} \mathbf{l}^{(i)} + g_s^{(i)} \mathbf{s}^{(i)}] + \mu_N g_R \mathbf{R}$$

• The transition dipole moments corresponding to M1 transitions:

$$\langle f|H_{int}(M1)|i\rangle = \langle f| - \mu(t) \cdot \mathbf{B}_0(t)|i\rangle = -\mathbf{B}_0(t) \cdot \langle f|\mu(t)|i\rangle$$

#### Selection rules

#### E1 transitions:

- □ π<sub>i</sub> π<sub>f</sub>=-1
- □ |∆K|≤ 1
- *□* Δj≤ 1
- □ ΔI=0,+1,-1
- M1 transitions:
  - □ π<sub>i</sub> π<sub>f</sub>=+1
  - □ |∆K|≤ 1
  - □ ∆j≤ 1 (no 0->0)
  - □ ΔI=0,+1,-1

# More on nuclear excitation by lasers

- Difficulty of atomic/molecular control: accessible states close together at times, lying within bandwidth of laser pulse.
- In contrast, closest nuclear excited states typically lie >> 10 keV apart. Very narrow linewidth: ~ meV
- Taking into account potential sources of pulse broadening:
  - □ Power broadening (at most ~ 1-2 keV for ultra-high intensity)

$$\Delta E \approx R_0 E_0 = (10^{-12} \sim 10^{-13})\sqrt{I} \quad eV$$

□ Femtosecond pulse length:

$$\Delta E \Delta t \ge \frac{\hbar}{2} \Rightarrow \Delta E \ge \frac{\hbar}{2(10^{-15} \, s)} = 0.052 \, eV$$

□ Laboratory bandwidth of laser pulse (< 5 eV)

 All together, this amounts to a total possible broadening of at most a couple keV.

# More on nuclear excitation by lasers

- Given the essentially negligible pulse broadening mechanisms, energy spectra to be understood as collection of relatively distantly spaced excited states.
- Considering selection rules, the number of accessible states from the ground state is very limited (typically no more than 3-4)
- Further constraints: maximum photon energy ~ 12.4 keV
  => often only one excited state under consideration.
- Thus, on-resonance laser excitation must be used in nuclear excitation, unlike in atomic/molecular excitation

#### Evaluation of controllability

- At this juncture, the next step is to formally evaluate the controllability of nuclear transitions:
  - This is typically done using computer algorithms that have been created by many leading researchers in the field of quantum control (Turinici, Schirmer, etc.)
  - Involves calculating nested commutators between the nuclear Hamiltonian matrix and the various dipole matrices – Lie algebra rank analysis
  - It has been theoretically shown, however, that the probability of finding a Hamiltonian (out of all possible Hermitian matrices) which is not controllable --- is essentially null.

#### Evaluation of controllability

- While formal controllability calculation awaits, an intuitive consideration based on the above mentioned selection rules and energy spectra can already suggest the outcome:
  - Only a handful of states (often only two) need even be considered for practical applications
  - Within an energy range ~ maximum laser pulse bandwidth/ broadening, never will there be two accessible states.
  - □ (2I+1)-fold degeneracy in M cannot be removed, irrelevant in the context of energy levels, which depend only on I and K.
  - Preliminary conclusion => full controllability...

#### Practicality of control experiments on nuclei

- Most important constraint in assessing excited states of nuclei: photon energy
  - □ Highest-energy laser photons achievable have E~ 12.4 keV
  - Higher energy gamma ray beams may not validate the dipole approximation, upon which controllability formalism/calculations hinge.
  - Only a small slice of nucleide chart has excited states that lie below this value (heavy metals, actinide/ lanthanide series)
- Both E1 and M1 transition dynamics depend strongly on the laser intensity, which for nuclei, must be enormous intensities.
  - Commonly cited intensity thresholds for complete population inversion lie in the neighborhood of I=10^27 W/cm^2

#### Practicality of control experiments on nuclei

Higher-energy excited states and larger transition matrix elements can be achieved though acceleration of nucleus into laser field:

$$\nu_N = (1+\beta)\gamma\nu_L \qquad I_{eff}^N = (1+\beta)^2\gamma^2 I_{eff}^L$$

 For example, at RHIC at Brookhaven, heavy ions are routinely accelerated to .99995 c, which leads to a frequency Doppler shift factor of ~200
 States with energies up to 1-2 MeV could potentially be accessed.

#### Current/near-future laser facilities

Name (Location)	Type, wavelength	Peak power	$\begin{array}{c} \text{Maximum} \\ \text{intensity} \\ (W/cm^2 ) \end{array}$	Maximum energy/ pulse	Typical pulse length
HERCULES (University of Michigan)	Ti:Sapphire, 800 nm	$500 \mathrm{~TW}+$	$\begin{array}{c} 5\times10^{22} \\ \rightarrow 10^{23} \end{array}$	20 J	30 fs
GEKKO PW (Osaka, Japan)	Nd:Glass, 1053 nm	$1 \mathrm{PW}$	$\sim 10^{21}$	700 J	$\sim 700 \text{ fs}$
FIREX I (Osaka, Japan)	Nd:Glass, 1053 nm	$1 \mathrm{PW}$	$\sim 10^{19}$	10 kJ	10 ps
TITAN (Livermore Labs)	Nd:Glass, 1053 nm	$1.25 \ \mathrm{PW}+$	$10^{19} - 10^{20}$	600 J	400 fs or long-pulse
VULCAN (Rutherford Appleton Labs, UK)	Nd:Glass, 1053 nm	$1 \mathrm{PW}$	$\sim 10^{21}$	2.6 kJ	500 fs
*VULCAN Upgrade (Rutherford Appleton Labs, UK)	Nd:Glass, 1053 nm	10 PW	$\sim 10^{23}$	300 J	30 fs
(*)PETAL (Aquitaine, France)	Nd:Glass, 1053 nm	7.2 PW	$\sim 10^{21}$	3.5 kJ	0.5-10 ps
*HIPER (Aquitaine, France)	Nd:Glass, 1053 nm	150  PW+(?)	$\sim 10^{24}(+)$	$30 \mathrm{~MJ}$	?
NIF (Livermore Labs)	Nd:Glass, 351 nm	500  TW	$2 imes 10^{15}(+)$	1.8 MJ	ns range



## Future prospects? Alternate methods?