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INTERLEVEL TRANSFER MECHANISMS AND THEIR APPLICATION TO GRASERS

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ABSTRACT

Within the gamma-ray laser (GRASER) research community, much attention is being given to two-step schemes that store energy in a long-lived isomeric state and achieve lasing by transferring population to a short-lived state. Because the electron system exhibits large multipole moments and is in the near field of the nucleus, it can be used as an intermediate mechanism for transferring energy, angular momentum, and parity change. Two distinct electron-nucleus interaction mechanisms are discussed: (1) resonant electronic transitions and (2) collective outer-shell excitations.

INTRODUCTION

For nearly two decades, researchers¹ have been proposing GRASERS using interlevel transfer. The long-lived isomer would be separated and concentrated by laser² or radio-chemical means³ and implanted in a host crystal. If rapid interlevel transfer can be accomplished, superradiance will develop and produce an intense, highly directed burst of semicoherent gamma radiation. Calculations suggest that a GRASER using the Mössbauer and Borrmann effects in a perfect crystal can be superradiant with as few as 10^{13} active nuclei¹⁰.

Direct interlevel transfer can clearly be accomplished: it is the basis of Mössbauer spectroscopy. However, it becomes difficult at long wave lengths because linewidths are very narrow and we cannot transfer rapidly enough for superradiance. Assuming they obey the Weisskopf formula, low-energy transition

rates go like $(\Delta E)^{2L+1}$, where L is the multipolarity. For instance, an M2 transition at $\Delta E = 100\text{eV}$, would have a linewidth of about 10^{-11}Hz .

Electronic transitions, on the other hand, have much larger multipole moments and much broader resonances. Therefore, we propose to excite mixed nuclear-electronic transitions made possible by the near-field interaction.

RESONANT ELECTRONIC TRANSITIONS

This technique uses a laser to eject an electron selected so the most probable transition to fill its hole is resonant with the nuclear interlevel-transfer transition and has the correct multipolarity and parity. The process is identical to NEET¹⁸. The technique lacks energy efficiency because the energy to create the hole will generally exceed the interlevel transfer

energy. A substantial fraction of the wasted energy will be deposited in the lattice and impair the Mössbauer and Borrmann effects. Furthermore, Auger and Coster-Kronig transitions are also near-field effects and compete with the interlevel transfer.

To assess the feasibility of this mechanism, we obtained analytic solutions for interlevel transfer probability assuming hydrogenic electron wavefunctions, and also obtained numerical results using a Hartree-Fock algorithm for the electrons and a modified muonic atom code⁶ for the electron-nucleus interaction.

Comparison of analytic and numerical results show that the order of magnitude of the interlevel transfer probability is

$$P \approx K_1 \frac{(K_2 A)^{2L/3}}{\Gamma_i (\Gamma_i + \Gamma_f)} B^{L+1} \quad (1)$$

where B is the binding energy of the electron hole, A is atomic weight, Γ_i and Γ_f are the initial and final electronic linewidths,

and the constants $K_1 = 1\text{eV}$ and $K_2 = 10^{-15} \text{eV}^{-3/2}$. Some salient features of Eq. (1)¹ are: (1) transitions with higher binding energy are favored, but such transitions of higher Z atoms are greatly broadened owing to the abundance of electrons in the near field; and (2) lower multipolarities are strongly favored owing to the magnitude of K_2 . The first feature would favor light nuclei, which unfortunately would be unlikely to display isomerism or closely spaced levels. The second feature places some limitation on the ratio of lifetimes between the storage and the upper lasing state. For a dipole transition, the lifetime ratio does not exceed 10^6 , except for the unlikely circumstance of a forbidden transition between the storage and lower lasing state. An alternative is to arrange for an intermediate transfer state between the storage and upper lasing level and thereby effect a quadrupole transition by two successive dipole transitions. The likelihood of such a favorable constellation of states is also small.

A numerical search of electronic configurations revealed a few atoms that may support interlevel transfer if there were an appropriate coincidence with nuclear levels. One of these is ^{24}Mg , which has no known nuclear-level coincidence, but is highly deformed. We considered a $2S_{1/2}$ hole filled by the $2P_{1/2}$ electron with transition of the nucleus from 1^- to 0^+ at the same energy. The dominant broadening for both states in Auger effect, which gives 8.8mV for the $2S_{1/2}$ and 0.6mV for the $2P_{1/2}$. The transition energy is only 38eV . The matrix element for the transition from numerical calculation is 1.4mV for one Weisskopf unit, which leads to an interlevel transfer probability of about 3%. This may be sufficient for gamma-ray lasing if all other conditions were favorable.

COLLECTIVE ELECTRONIC EXCITATION

Transition rates can be greatly enhanced if several electrons act collectively to drive a nuclear interlevel transfer. For example, if 10 electrons were involved, the matrix element would be 10 times larger, and the transition rate would be 100 times larger. Furthermore, if collective excitations are generated by multiple photon absorption, a low-quantum-energy laser can be used to drive a high-quantum-energy interlevel transfer^{7,8}. The simultaneous absorption of as many as 99 photons have been reported for 6.4eV radiation in uranium⁹. Because reasonably low energy quadrupole transitions are known in nuclei and the quadrupole transition rates generally exceed those expected from the single particle model, it would be advantageous if the collective outershell excitations had a substantial quadrupole component. This should be straightforward to calculate by standard numerical techniques. Lacking such calculations, but judging from experience with the liquid-drop model, we would conjecture that as much as half the excitation energy could be quadrupole oscillations.

To estimate the matrix elements for coupling between the collective excitations and the nucleus, we simply sum over the N electrons that are acting coherently. The near-field interaction Hamiltonian is:

$$H' = -\sum_e \frac{Ze^2}{|\vec{r}_n - \vec{r}_e|}, \quad (2)$$

assuming the magnetic interaction is small.

If we knew the initial and final electron and nucleus wavefunctions ψ_{ei} , ψ_{ef} , ψ_{ni} , ψ_{nf} , we could find the interaction matrix element by expanding the Hamiltonian in spherical harmonics.

$$\begin{aligned} E' &= \langle \psi_{ni} \psi_{ei} | H' | \psi_{nf} \psi_{ef} \rangle \\ &= 4\pi e^2 Z \sum_e \sum_{lm} \frac{1}{2L+1} \int_0^\infty \psi_{ni}^* Y_{lm}(\theta_n, \phi_n) r_n^L \psi_{nf} d\Omega_n r_n dr_n \quad (3) \\ &\quad \times \int_0^\infty \psi_{ei}^* Y_{lm}^*(\theta_e, \phi_e) r_e^{-L-1} \psi_{ef} d\Omega_e r_e^2 dr_e, \end{aligned}$$

where we have ignored the contribution for the electron wavefunctions inside the nucleus. A rough estimate can be obtained by guessing the radial matrix elements and assuming the N electrons act in perfect coherence.

$$E' = - \frac{4\pi NZe}{2L+1} \langle r_n^L \rangle \langle r_e^{-L-1} \rangle \quad (4)$$

For a uranium nucleus, $r \cong 7.5 \times 10^{-13}$ cm, but it is highly deformed, so we could estimate $\langle r_n^2 \rangle \cong 7.5 \times 10^{-24}$. An estimate of the radius of the collectively excited electron shell might be 5×10^{-12} . If we say that 10 electrons act collectively, the quadrupole matrix element is about 40eV. Furthermore, if we found an appropriate dipole transition for the same nucleus, the matrix element would be about 2KeV. These are large interaction energies. At this time we have no estimates of the corresponding linewidths, but it seems reasonable that interlevel transfer probabilities will be substantial. The most serious question is whether Mössbauer and Borrmann effect can be maintained with the laser intensities required for collective excitation. Certainly a traveling wave transfer scheme will be necessary.

The strong coupling suggests that nuclear transitions may provide a sensitive probe of collective electron excitation, especially for determining their total energy and multipolarity. The broad range of possible experiments may usher in a new era of whole-atom physics.

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