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Implementing NRF Physics in Geant4

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Executive Summary

Nuclear resonance fluorescence (NRF) refers to resonant absorption of a gamma photon on an atomic nucleus, followed by nuclear deexcitation with emission of one or more photons. The Geant4 radiation transport Monte Carlo code toolkit currently does not support NRF. This document describes plans for implementing this physics process in Geant4 for use in simulations of active interrogation of cargo. These plans will be executed as Task 3 of Pacific Northwest National Laboratory's project 50799, "Nuclear Resonance Fluorescence Signatures (NuRFS)."

1. Introduction

The Geant4 radiation transport Monte Carlo code framework [1] currently does not support nuclear resonance fluorescence (NRF). Of the many physics processes incorporated into Geant4, the suite of photonuclear processes [2] is the logical location to address NRF. The photonuclear interaction physics implemented in the latest release of the framework (version 8.0) is primarily of relevance to the high-energy physics community, emphasizing theoretical descriptions of nuclear response at photon energies (~100 MeV to hundreds of GeV) well above those of relevance to the practical use of NRF in active interrogation.[1] This document briefly describes plans for augmenting the Geant4 framework to include NRF.

2. NRF Physics in Brief

As its name implies, NRF entails resonant nuclear absorption of a gamma photon, followed by deexcitation with emission of one or more photons. The resonance cross section is of Breit-Wigner shape [3],

$$\sigma(E) = \pi\left(\frac{\lambda}{2\pi}\right)^2 \frac{2J_1+1}{2J_0+1} \frac{\Gamma_0(\Gamma/2)}{(E-E_r)^2+(\Gamma/2)^2} \qquad (1)$$

where λ is the wavelength of the incident photon of energy E, E_r is the resonance energy, J_0 and J_1 are the nuclear spins of the ground state and excited state, respectively, Γ is the total decay width of the excited state, and Γ_0 is the partial width for deexcitation to the ground state.

Thermal motion of the target nuclei leads to significant Doppler broadening of Eq. (1) at room temperature. In the thin-target limit that is the customary application domain for

[1] The lowest-energy photonuclear process supported in Geant4 is giant dipole resonance, a collective nuclear excitation mode relevant at energies above ~10 MeV.

nuclear physics experiments investigating NRF, it can be shown [3] that this broadening does not change the energy-integrated cross section (and thus, the resonance signal rate). This is not strictly true in the thick-target case of relevance to cargo inspection, however, due to the fact that the portion of the incident photon beam interacting with a thick slab of material varies exponentially with the interaction cross section, rather than linearly as in the thin-target approximation. The impact of the non-linear dependence of the resonance signal rate on the interrogated sample thickness for scenarios of relevance to broad-spectrum photon beam cargo inspection will be investigated in the course of this task. Prescriptions for describing thermal broadening of the resonance lineshape exist [3], valid under the assumption that thermal motion of nuclei in a solid can be reasonably well-described by a Maxwell distribution at absolute temperature, T.

The angular distribution of the emitted fluorescence photons is described by a correlation function, $W(\theta)$, defined with respect to the direction of the incident photon [4]. The correlation function depends upon the multipolarities of the electromagnetic radiation involved in the emission transition. Participating multipolarities in turn are constrained by selection rules determined by conservation of angular momentum and parity in the emission process [5]. Analytic expressions for the correlation function can be found in e.g. [4]. Here we merely note one particular case, corresponding to pure dipole transitions ($J_0 = 0, J_1 = 1$) :

$$W(\theta)_{\text{dipole}} \propto 1 + \cos^2(\theta) \qquad (2)$$

3. Planned Approach to Incorporating NRF Physics in Geant4

The object-oriented (OO) Geant4 code framework [1] is intended to facilitate convenient extension of the suite of physics models describing various radiation transport, nuclear physics and high-energy particle physics processes. Implementing a new physics process requires supplying a C++ class with member functions specifying two essential ingredients[2]: (1) the mean free path for occurrence of the process (derived in turn from the cross section and atomic number density within a particular material), and (2) the final state (i.e., particle species and kinematics formed at an interaction vertex) once the process has occurred. The new physics process class “plugs in” to the Geant4 OO physics framework by adhering to the class inheritance-based interface expected for these member functions. Once the physics has been appropriately defined within the code framework, it is available for “registration” with the Geant4 physics process manager in the initialization phase of the user’s application at run-time. The Geant4 framework then handles coordination of physics process sampling for all particles (primary and secondaries) involved in a given tracking history.

Cross section calculation

The cross section for resonance photon absorption in nuclear species (A,Z) of a given material will be computed from equation (1) based upon data extracted from an existing

[2] The description here is simplified to describe the important special case of “discrete” interactions that are conveniently modeled in terms of individual, spatially-separated scattering events. Other interactions, such as ionization energy loss of charged particles, are more readily simulated in terms of a continuous process occurring throughout a particle tracking step, and require implementing a somewhat more extensive set of member functions in the physics class definition.

Geant4 database, derived in turn from the Evaluated Nuclear Structure Data (ENSDF) [7] database of experimentally measured nuclear data. The Geant4 nuclear level database, which was constructed to support one facet of the framework's photon evaporation model (describing particle emissions following high-energy hadronic excitation of nuclei), consists of a set of files labeled by isotope (A,Z). Each file contains conveniently-formatted records with the following standard nuclear level information:

- Energies, spins, parities, and natural widths of nuclear states;
- The energies and relative intensities of gammas emitted in decay to levels of lower energy;
- Internal conversion coefficients, for use in calculating the probability of conversion electron emission (rather than photon emission) in a given transition.

A new database with information relevant to NRF cross section calculations will be built to augment the existing Geant4 database. The intention of the new database will be to extend the existing database for states not originally included and to correct possible discrepancies. The separation of the new and old databases, rather than modifying the old database, will maintain the integrity of the existing Geant4 processes that access the old database.

The natural widths listed in the database correspond to the total linewidth, Γ, appearing in eq. (1). In order to calculate the cross section for resonance absorption from the ground state to a particular level, the partial width Γ_0 must be known as well. This width can be calculated from the information in the Geant4 photon evaporation database assuming that decay channels that change the nuclear species (e.g. alpha or beta decay, or neutron emission) can be neglected. With this assumption in hand, knowledge of the relative gamma emission intensities and conversion coefficients suffices to determine Γ_0. This calculation need only be performed once, during run-time initialization of the class that will implement the NRF physics. The relevant nuclear level data for each material appearing in the detector geometry are stored internally in data structures ("material data tables", in Geant4 parlance) at run initialization, and are then made available to the physics class at each tracking step by the Geant4 tracking framework.

The algorithm for cross section calculation for a photon of energy E within a given material containing N distinct nuclear isotopes, (A_i, Z_i), i = 1, …, N, then consists of calculating (1) for each isotope's set of nuclear levels. In practice, because nuclear levels are widely separated compared to the intrinsic widths (of order 10^{-3} eV) of the levels, the cross section will be effectively zero for all but (at most) a single isotope. This cross section (or, rather, the interaction mean free path derived from it) is then returned to the tracking framework for use in its stochastic sampling of the next discrete interaction of the photon.

Note that the shift in resonant energy introduced by nuclear recoil ($\Delta E = E^2 / 2M$ for a nucleus of mass M) is large compared to both intrinsic level widths and Doppler broadening for all gamma energies of relevance to applications of NRF [3], and thus this kinematic shift essentially precludes subsequent resonant absorption of a nuclear deexcitation gamma. This physical condition will be enforced in the cross section

calculation by ensuring that the resonance energy, E_r, reflects conservation of energy and momentum during gamma absorption.[3]

Final state sampling

The cross section (1) refers to resonance absorption from the ground state to a specific nuclear level, summed over all possible photon emission modes. Note that if decay to the ground state is the only possible deexcitation channel, then $\Gamma = \Gamma_0$ in (1). In the more general case where gamma emission to states lying between the ground and resonant excited state is possible, then

$$\Gamma = \Gamma_0 + \Gamma_1 + \Gamma_2 + \cdots \qquad (3)$$

where the Γ_i's are the partial widths for decay to the i'th level of energy less than that of the resonant state. Each of these levels (above the first excited state) can in general decay by gamma emission to various lower-energy levels as well, and so on. The probability distribution governing the frequency of occurrence of these sequences of gamma emissions, or cascades, can be calculated (at run initialization) from the same nuclear level data read from the Geant4 database and used to calculate the absorption cross sections. Similar techniques for gamma cascade calculation have been developed in the past at PNNL [6], so that some opportunity for software re-use may present itself here. The first stage of final state preparation given excitation of a particular nuclear level will be sampling of the gamma cascade on the basis of this probability distribution.

The angular distribution of the outgoing photon will be sampled from correlation functions, $W(\theta)$, appropriate to the multipolarities of the electromagnetic radiation as discussed briefly at the end of section (2) above. In general, more than one electric and magnetic multipole satisfy the selection rules governing a particular transition and can thus contribute [5], with relative intensities that depend upon the detailed overlap of the initial and final state nuclear wave functions. Thus the resulting angular distribution is a superposition of the distributions for the contributing multipoles. It is anticipated that in many cases where both electric and magnetic multipoles can contribute to the same transition, the mixing ratio will be unknown (and, in any event, this information is not tabulated in the existing Geant4 nuclear level database files developed for its hadron physics photon evaporation model). In these cases, at least two options can be pursued for determining a reasonable angular distribution: (1) Supplement the Geant4 photon evaporation database with multipole mixing ratios extracted from the (more extensive) ENSDF database, if this information is available for the transition; or (2) select the dominant multipole contribution based upon the semi-phenomenological Weisskopf estimates [5] of multipole intensity.

No matter what approach is ultimately adopted for implementing realistic angular distributions, it will be useful to have one or more approximate (or "baseline") angular distribution sampling algorithms available for purposes of estimating the contribution of uncertainties in the angular emission to systematic uncertainties in detected signal rate calculations. Simple isotropic emission, for example, will be included as an angular

[3] In principle, the experimentally measured gamma ray energies drawn from Geant4's re-casting of the ENSDF database already include this kinematic recoil effect (i.e., the measured gamma energy is slightly smaller than the difference in energies assigned to the two levels involved in the transition).

distribution model option, user-selectable during run-time initialization of the NRF physics class.

4. Validation

The implementation of the Geant4 NRF physics process will be compared to existing data for validation. Three different types of measurements will be compared. Measurements of two different thicknesses of the same material will be examined, as well one with very large NRF cross sections. PNNL and Passport collected well-controlled and high statistics spectra for aluminum and lead in June, 2006 at the 5.3 MeV facility at the University of California at Santa Barbara that will serve well for these purposes.

The validation will compare the resonant photon flux incident on the detector observed in the NRF spectra with those of the simulations. The incident flux from the measurements will be determined from the strength of the observed peaks divided by the detector efficiency. The detector efficiency will be determined from simulations. Comparing the flux incident on the detector reduces the computational time of the simulation. Modeling the background distribution is not within the scope for this project, but the project does plan to discuss a modeling of the NRF peaks and background with Passport Systems.

The photon energy distribution for the validation simulations will consist of a collection of narrow-band width groups, each group centered around a resonance. The relative strength of these groups will be weighed by the relative strength of the bremsstrahlung distribution. To test that the possible contributions from higher-energy photons can be ignored, the project will conduct simulations to test if higher energy photons can be downscattered into the resonance region in quantities to significantly affect the observed resonant photon rate at the detector.

The measure of success of the NRF implementation of the NRF process into Geant4 will be if the data and simulations agree to within 100% of the observed rates. This is a reasonable level of agreement considering uncertainties in the detector efficiencies at high energies, 30%, in the experimental setup, 20%, in the bremsstrahlung distribution 10-20%. If a higher level of agreement is required for specific applications, the cross section database and/or the processing of the final states can be fine-tuned.

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