Mechanical Doppler Compensation for Electron Excitation of NRF Photons

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Abstract—Identification of Special Nuclear Material (SNM) by Nuclear Resonance Fluorescence is developing as a promising method but is currently limited by available photon sources. A very limited source spectrum (< 1 eV width at MeV energies) is ideal. One method of providing this source is by electron excitation of a target containing the nucleus of interest. In this method, however, the photon energy mismatch caused by the nuclear recoil must be accounted for. We propose to provide this Doppler compensation by directing an MeV-range electron beam at a rotating target loaded with the nucleus of interest. Rotation rates are in excess of 200,000 RPM for the NRF lines of interest in SNM detection. Some design scenarios will be discussed, along with issues pertaining to nuclear physics and materials science.

Index Terms—Nuclear Resonance Fluorescence, SNM Detection, Photon Sources

I. INTRODUCTION

Nuclear resonance fluorescence (NRF) is a promising method for detection of special nuclear materials in cargo containers. The source of gamma radiation for an NRF inspection tool is a daunting task: resonance widths at several MeV tend to be in the tens of milli-electron volts due to the thermal broadening of even narrower natural linewidths. The T-REX program at Lawrence Livermore National Laboratory (LLNL) [1] is an example of one approach to this problem, using the Doppler-shifted photons from a short-pulse laser beam impinging on a relativistic electron beam from a linac. The e-beam energy required is in the hundreds of MeV, and very careful control of beam energies, beam quality, and laser-to-electron beam timing are required.

A more simple-minded approach might be called the “hair of the dog” approach: making the relevant gamma for detection of a particular material (such as 235U) by exciting the same material with an electron beam. However, the photons generated in this way share the energy level of the excited state with the recoiling nucleus, which has equal and opposite momentum with the exiting photon. The energy of an emitted photon from state with excitation energy $E_{ex}$ is less than $E_{ex}$ due to recoil:

$$E_{\gamma} = E_{ex} \frac{E_{2}}{2M c^2}$$

This energy differential can be regarded as a Doppler shift which can be compensated for by motion of the emitting source nucleus towards the absorbing nucleus by an amount

$$v \approx c \left( \frac{E_{2}}{E_{\gamma}} \right)$$

II. KINEMATICS WITH ELECTRON EXCITATION

For the case of an electron beam impinging on a target nucleus for electron excitation, the possibility of momentum being carried in by the electron must be accounted for and the simple formula given above must be modified. The proper accounting for this effect depends on the lifetime of the NRF state being utilized vis-a-vis the time for the target nucleus to lose its forward momentum in the atomic lattice of the material.

As an example, we note the recently reported 1733 keV resonance in 235U [3] which the authors measure with a resonance integral cross-section of $\sigma(\Gamma) = 29.8$ barn-eV. Following [1], we take the NRF peak cross section as

$$\sigma_{0} = \frac{25006}{E(MeV)^{2} J \Gamma} \left( \frac{2J + 1}{\Gamma + 1} \right)$$

Since the J* of the excited state is unknown, we assume $J = J_{v}$ and obtain $\Gamma \approx 35$ meV and thus the state has an estimated lifetime $\tau \approx 18$ fs. We estimate the time for the recoil nucleus to lose its forward momentum as $\tau_{recoil} \approx a_{0}/V$, where $a_{0} \approx n^{-1/3}$ is the lattice parameter and V is the forward velocity.

For a recoil velocity $V \approx 3000$ m/s, and $a_{0}^{-1/3} \approx 2.7$ A we have a momentum exchange time of $\approx 90$ fs. Thus $\tau_{\gamma} < \tau_{recoil}$ and excitation momentum should be included at the time of de-excitation.

For the case of electron excitation with $\tau_{\gamma} < \tau_{recoil}$, the momentum gained by the target nucleus is more than the amount which would make the resultant photon resonant with the same nucleus in the interrogated object, since electron momenta are greater than photon momenta at the
same energy. For $E$, just above threshold, the electron momentum reduces and reverses the required rotational speed. We again use the example of the $E_{ex} = 1.733$ MeV NRF line in $^{235}$U. Assume that the electron energy is just above threshold so that final electron momentum $p_f^* \approx 0$. Then the momentum carried into target U nucleus is $P_U = MV = P_f^* = \sqrt{(1.733 + 0.511)^2 - 0.511^2} = 2.185$ MeV/c or $V_{ne} = 2967 \text{ m s}^{-1}$. We want the $U^*$ to be moving at the time of de-excitation at a speed $V_{res} = c (E_{ex}/M) = c(2E_E/E_{ex}) = 2353$ m s$^{-1}$. This gives a target rotation rate $\Omega R = V_{res} - V_{ne} = -614$ m s$^{-1}$, or $n \approx 58,000$ RPM for a 0.1 m radius target. The negative sign indicates a reversal of the spin direction from that shown in Fig. 1.

Alternatively one can exploit the excess momentum in this case to find a “magic angle” at which zero rotation is required for the resonant match-up to occur. For full momentum carryover from the incoming electron, this angle is given by:

$$\cos[\theta] = \frac{\sqrt{(E_e(e^{-} - m_e c^2)^2 - m_e c^4) - \sqrt{(E_e(e^{-}) - E_{res} + m_e c^2)^2 - m_e c^4}}}{E_{ex}}.$$

The above analysis assumes that the initial electron and final electron are moving in the same direction, which is reasonable because inelastic Coulomb excitations of this type are strongly peaked in the forward-scattering direction. A plot of this angle as a function of initial electron energy is shown as Fig. 2.

III. EXCITATION CROSS SECTIONS

Little experimental data is available for electronic excitation of NRF states in U and Pu. We give some estimates of the cross sections based on the virtual-photon method of Weizsäcker and Williams. The virtual photon method can be employed to estimate the inelastic electron photon cross section from the resonance-integral NRF data. Details are summarized in [4]. To obtain the electron excitation cross section $\sigma(E_0)$ for a projectile of energy $E_0$ to produce a resonant state in the nucleus at energies around $E$, one uses the equation:

$$\sigma(E_0) = \sum_{n, \lambda} \int \sigma_{\gamma, \lambda}(E) n_{n, \lambda}(E, E_0) dE/E.$$

Here $\pi$ and $\lambda$ are the transition type ($E$ or $M$) and the multipole order of the transition. The virtual photon numbers for the lowest order transitions $E1, M1,$ and $E2$ are given by:

$$n_{e, E1} = \frac{\alpha}{\pi} \frac{E_e^2 + E_f^2}{(\hbar c k_f^2)^2} \ln (2E_1^2 - 2k_f k_e),$$

$$n_{e, E2} = \frac{\alpha}{\pi} \frac{E_e^2 + E_f^2 - 2m^2 c^4}{(\hbar c k_f^2)^2} \ln (2E_1^2) + \frac{\alpha}{\pi} \frac{8k_f m^2 c^4 + E_f}{(\hbar c k_f)^2},$$

$$n_{e, M1} = \frac{\alpha}{\pi} \frac{E_e^2 + E_f^2 - 2m^2 c^4}{(\hbar c k_f^2)^2} \ln (2E_1^2).$$

Here $\xi^{-1} = (E_f E_i + (\hbar c) k_f k_i - m^2 c^4)/(2mc^2\hbar c)$, $\alpha = 1/137$, and the initial and final energies $E_f$ and $E_i$ include the rest masses. Cross sections are strongly forward peaked in the angle of the scattered electron. Emitted photons are uncorrelated with initial and final electron momenta.

Again we use the example of excitation of the 1.733 MeV resonance in $^{235}$U. The multipolarity of this state is currently unknown. Calculations for $E1, M1,$ and $E2$ at various electron kinetic energies are shown as Table 1 using the data from [3].

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>$\sigma(E1(\mu b))$</th>
<th>$\sigma(M1(\mu b))$</th>
<th>$\sigma(E2(\mu b))$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.007</td>
<td>0.011</td>
<td>0.24</td>
</tr>
<tr>
<td>3</td>
<td>0.12</td>
<td>0.17</td>
<td>0.51</td>
</tr>
<tr>
<td>5</td>
<td>0.18</td>
<td>0.24</td>
<td>1.15</td>
</tr>
<tr>
<td>10</td>
<td>0.29</td>
<td>0.35</td>
<td>3.9</td>
</tr>
</tbody>
</table>

Table 1. Estimate of the inelastic cross section for production of 1.733 MeV photons in $^{235}$U.

The cross section estimates indicate that an acceptable production of NRF photons can be obtained with $E2$-type resonances at electron energies 10 MeV and above. Because all of the photons generated by this technique are usable, the brightness of this source is very high compared to the bremsstrahlung. It may be desirable to use the mechanical Doppler method to “swing” the usable photon channel out of the forward bremsstrahlung cone, to minimize overall exposure for the detection process. As an example, at 180 degrees to the beam, a 100 $\mu$A beam of electrons on a target with a loading of 0.015 g cm$^{-2}$ (one gram total $^{235}$U inventory on a 10 cm radius target with a 1 cm width) will produce around $10^6$ usable NRF photons per second, for a brightness of around $2.5 \times 10^{11}$ photons per steradian per MeV. The same target will produce $2.5 \times 10^5$ bremsstrahlung photons per steradian per MeV in the backwards direction, resulting in a $10^7 : 1$ excitation photon-to-bremsstrahlung photon ratio in-band.

IV. CONCLUSION

The above arguments show that NRF photons can be produced by electron excitation with acceptable yields for 10 MeV energies and reasonable current levels. The recoil kinematics can be handled with rotating targets, but the lifetimes
of the excited states must be considered when designing a system. Molecular dynamics calculations will be done which will further quantify the recoil momentum dynamics in the atomic lattice for these conditions. High-yield systems may allow for separating the bremsstrahlung from the desired photons, resulting in a relatively low-dose detection scheme. More nuclear data is needed, however. The multipolarity of NRF lines is important for this scheme, and since $E2$ lines have substantially higher production by electrons, strong $E2$ resonances are desirable. Current efforts are underway to measure the multipolarity of the NRF lines in U and Pu, and hopefully some candidate lines will appear.

ACKNOWLEDGMENT

This work is supported by an ARI grant from the National Science Foundation and the Department of Homeland Security.

REFERENCES