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A Comparative Investigation of Doppler broadening of Neutron absorption and Photon-induced Nuclear Resonance Fluorescence Reactions

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1. Introduction

The accuracy of the computational analysis in nuclear industry depends on the nuclear cross section data. At elevated temperatures, the evaluation of such nuclear cross section is strongly affected by the Doppler broadening which is due to thermal motion of the targeted nucleus. Therefore, the accurate calculation of Doppler broadened cross section data for both neutron and photon-induced reactions is necessary. As neutron-induced reaction is common in nuclear analysis, many data processing codes which consider Doppler broadening and its effects, are widely available. Conversely, such data for the photon-induced reactions is very rare. Recently, some of the photon-induced reactions such as nuclear resonance fluorescence (NRF) has gained attention due to its wide utilization, from study of nuclear structure to material assay and isomer production such as Tc-99m [1]. Therefore, it is important to give more attention towards the accurate compilation of nuclear data for photon-induced reaction especially the Doppler broadened cross section.

The Doppler broadening process reflects particle-nucleus interaction in the resonance energy region which is difficult to determine accurately. Parameters for modeling the cross section for such process can be calculated with integrated values over the distribution of target nucleus velocity. This paper presents a brief derivation of the Doppler broadening process and some quantitative cross sections for both neutron- and photon-induced resonance phenomena.

2. Doppler Broadening of Resonances

Neutron and photon-induced reactions have different resonance mechanisms. The difference comes from the type of incident particle because the major forces for interactions with target nucleus in both processes are distinct. When a neutron is in the vicinity of target nucleus, a strong nuclear force combines them together, which results in so-called compound nucleus formation. Therefore, the imparted energy of the compound nucleus (i.e. $^{A+1}Z^{*}$) is summation of the neutron's energy and its binding energy per nucleon. When the imparted energy is very similar to any of the excited states of compound nucleus, a resonance reaction can happen for the corresponding excited state.

In the process of photon-induced nuclear reactions, compound nucleus is not formed as the major driving force is a 'weak electromagnetic one' in nature. Therefore, the only way to change the target nucleus configuration is to knock out a nucleon from the nucleus. However, to do so, the wavelength of incident photon should be as small as a size of nucleon which corresponds to the energy in the upper region of giant dipole resonance (GDR) reaction [2]. The GDR reaction emits at least one nucleon because incident photon's energy is higher than the neutron separation energy, which corresponds to the GDR's threshold energy. In case the incident photon energy is less than the binding energy of the nucleons then NRF reaction will occur. The NRF reaction just excites the target nucleus depending on the energy of incident photon. In most of the NRF reactions, excited nucleus stays at the perturbed state for an extremely short period and a photon is re-emitted with a slightly modified energy and the nucleus decays back to the original state. The interaction probability is very low unless the incident photon's energy is very close to an allowable excited state of target nucleus, resulting in very narrow resonance phenomenon.

If target nucleus is under thermal motion, the aforementioned resonance cross section should undergo the Doppler broadening phenomenon due to change in the relative velocity of the incident particle. In the following sections, Doppler broadening was investigated using the single-level Breit-Wigner (SLBW) formula for both capture resonance of neutrons and photon-induced NRF resonance reactions. The Maxwell-Boltzmann distribution was used to represent the nuclear thermal motion depending on temperature.

2.1 Broadening of Neutron-induced Reactions

With regard to the radiative capture resonance of neutron by a stationary nucleus, the SLBW formula is given as follows:

$$\sigma_{\gamma}(E) = \sigma_0 \frac{\Gamma_{\gamma}}{\Gamma}\sqrt{\frac{E_0}{E_c}}\left[4\left(\frac{E_c - E_0}{\Gamma}\right)^2 + 1\right]^{-1}, \qquad (1)$$

where Γ is the total decay width of the resonance energy, Γ_{γ} is the radiative decay width, E_0 is the resonance energy and E_c is the center of mass energy. For this calculation, the Maxwell-Boltzmann distribution is considered for modeling nucleus velocity in 3-dimensional space and the temperature-dependent cross section can be evaluated in the following way [3]:

$$\bar{\sigma}_{\gamma}(y, \mathrm{T}) = \frac{1}{y^2\sqrt{\pi}}\int_0^{\infty} dx x^2 \sigma(x, T_0)\left[e^{-(x-y)^2} - e^{-(x+y)^2}\right], (2)$$

where $x^2 = \alpha E_r$, $y^2 = \alpha E$, $\alpha = \frac{A}{k(T-T_0)}$, A is the mass ratio between target nucleus and neutron, k is the Boltzmann constant, and $\sigma(T_0)$ is reference cross section at the base temperature T_0. The effective or average cross section can be separated into two parts because the latter exponential term is so small that it is negligible when the energy of incident neutron is high enough ($y \geq 4$). In this study, the effective cross section can be determined using the following equations:

$$\bar{\sigma}_\gamma(y,\mathrm{T}) = \bar{\sigma}^*_\gamma(y,\mathrm{T}) - \bar{\sigma}^*_\gamma(-y,\mathrm{T}) \quad (3)$$

$$\bar{\sigma}^*_\gamma(y,\mathrm{T}) = \frac{1}{y^2\sqrt{\pi}}\int_0^\infty dx x^2 \sigma(x,T_0) e^{-(x-y)^2} \quad (4)$$

2.2 Broadening of Photon-induced NRF Reaction

For the NRF resonance reaction for a stationary nucleus, the SLBW formula is given as follows:

$$\sigma(E) = \frac{\pi}{2}\frac{2J+1}{2J_0+1}\frac{(\hbar c)^2}{E^2}\frac{\Gamma\Gamma_0}{(E-E_r)^2+(\Gamma/2)^2}, \quad (5)$$

where J is the angular momentum of excited nucleus, J_0 is the angular momentum of its ground state, $\hbar$ is the reduced Planck constant, c is the speed of light, Γ is the total decay width of an excited state, Γ_0 is the partial decay width to the ground state, and E_r is the resonance energy of nucleus. The Maxwell-Boltzmann distribution is considered for only single-direction of nucleus motion and the broadening is reflected with Doppler width Δ, which represents a relative change of photon's frequency to target velocity for temperature variation. Then, the effective cross section can be described as follows:

$$\sigma_{avg}(E) = \int_0^\infty dE'\sigma(E')\frac{1}{\sqrt{2\pi}\Delta}\exp\left(\frac{-(E'-E)^2}{2\Delta^2}\right), \quad (6)$$

where $\Delta = \frac{E_0}{c}\sqrt{\frac{kT}{M}}$ and M is the mass of target nucleus. The integral in Eq. (6) can be performed with the delta approximation in vicinity of resonance energy. The approximation is quite acceptable because full width at half maximum (FWHM) in Eq. (5) is usually three orders of magnitude smaller than the Doppler width. Therefore, Eq. (6) is usually approximated in the following way:

$$\sigma_{eff}(E) \approx \frac{\pi}{E_r^2}\sqrt{\frac{\pi}{2}}\frac{2J+1}{2J_0+1}\frac{(\hbar c)^2\Gamma_0}{\Delta}\exp\left(-\frac{(E_r-E)^2}{2\Delta^2}\right) \quad (7)$$

Above two models help to calculate Doppler-broadened resonance cross section easily with only parameterized values such as the energy width and resonance energy from experiment. In addition, they can also be worthwhile when the experimental results are insufficient, as such with the NRF reactions for many nuclei.

3. Doppler Broadening of U-238 Resonances

The temperature dependent cross sections for a certain isotope are usually obtained from experimental results and corresponding models including the Doppler broadening effect are developed to interpret these results. Generally, for the nuclear reactions, such cross section data is widely available and reported in various data libraries (e.g. ENDF, JENDL, and ENSDF etc.). However, for NRF reaction only a few experiments have been carried and its cross section data is scarcely available in the ENSDF library. With the increasing NRF reaction applications, the evaluation of accurate temperature-dependent NRF cross sections is needed. This can be used for the development of corresponding Doppler-broadened cross section model. Fortunately, the high intensity gamma-ray source (HIGS) facility recently performed NRF experiments for U-238 and reported the cross section data even for unknown energy levels [4]. In this study, a method to calculate the Doppler-broadened NRF cross section has been proposed using the parameters derived from experiments performed at the HIGS facility. The method is based on the mathematical model developed in previous section.

In this section, the Doppler broadening of both neutron and photon-induced reactions is compared for U-238's resonance cross sections using Eqs. (4) and (7). The comparison is performed over certain energy range including several resonance peaks to assess the broadening effect when the temperature changes

Although U-238 has 2126 resonance peaks in its resonance region, ranging from 4.40 eV to 20.6 keV, the lower part of resonance region is considered in this work, since the low-energy resonances have relatively high peak cross section. Table 1 summarizes the parameters for the several resonance peaks that are included in the low energy range. The low-energy capture resonances are shown in Fig. 1 for several temperatures.

Table 1. Neutron-induced resonance data for U-238 [5]

E_0(eV)	Γ_n (meV)	Γ_γ (meV)	σ_0 (barn)
6.67	1.52	26	2.16e+5
20.90	8.7	25	3.19e+4
36.80	32	25	3.98e+4
66.54	26	22	2.14e+4
102.47	70	26	1.86e+4
116.85	30	22	1.30e+4
165.27	3.2	18	2.41e+3
208.46	53	22	8.86e+3

For the photon-induced reactions, the number of resolved NRF peaks are relatively small. Therefore, noticeable peaks are selected at energies above 2000 keV with an interval of 200 keV. Table 2 shows the integrated cross section I_s for some excited states of U-238. The partial energy width was derived from the experimental results and the associated NRF model, assuming the cross-sections were measured at 300 K. The corresponding NRF resonances between 2.332 MeV and 2.529 MeV are shown in Fig. 2 for several temperatures. The Doppler broadenings are compared in terms of peak frequency, maximum cross section, and variation of FWHM.

Table 2. NRF resonance data for U-238

E_γ (keV)	J_π	I_s (eV·barn)	Γ_0 (meV)
2332.7	1-	10	4.736
2365.6	1-	44	21.43
2410.0	1+	18	9.098
2422.8	1-	12	6.130
2467.8	1+	80	42.40
2491.5	1-	9	4.862
2499.4	1+	32	17.40
2529.0	1-	12	6.679

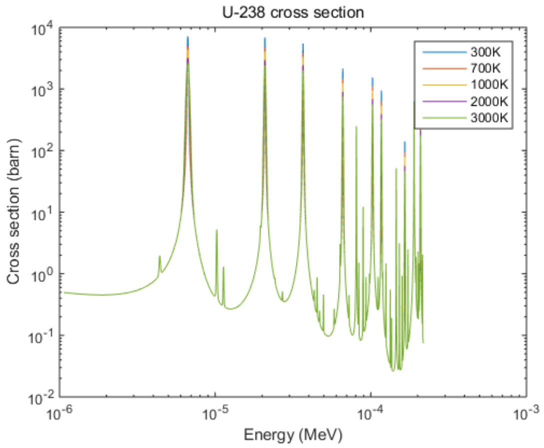

Fig. 1. Neutron- induced resonance cross sections at low energy range.

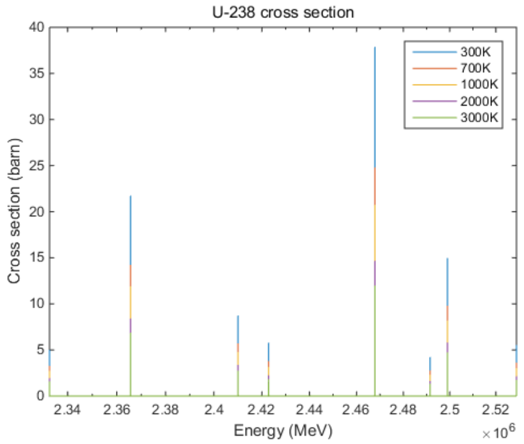

Fig. 2. NRF resonance reaction near 2.4 MeV.

Figures 3 and 4 show Doppler- broadened neutron capture resonance at E=6.67 eV and NRF resonance at E=2.468 MeV, respectively. Comparing the two temperature-dependent resonances, one can clearly note that the Doppler broadening is more effective in the NRF resonance reaction. Table 3 summarizes the impacts of Doppler broadening of the two types of resonances. It is clear that The FWHM is only about 0.25 eV for the neutron capture resonance, while it is about 5 eV for the NRF reaction at 2000 K. It is noteworthy that ratio of the FWHM values for 300 K and 2000 K is quite similar for the two resonance reactions.

Table 3. Resonance parameters for various temperatures.

Temperature (K)	Neutron, 6.67 eV		Photon,2468 keV	
	σ_{peak} (barn)	FWHM (eV)	σ_{peak} (barn)	FWHM (eV)
300	7114	0.103	37.876	2.4
700	5056	0.150	24.796	3.0
1000	4328	0.178	20.746	3.6
2000	3173	0.247	14.669	5.4
3000	2635	0.296	11.977	6.3

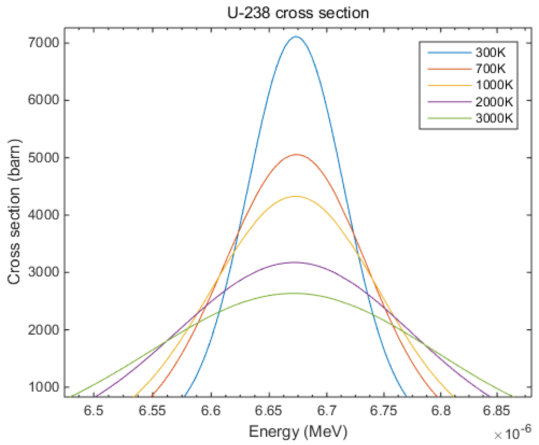

Fig. 3. Doppler broadening of neutron capture resonance at E=6.67 eV

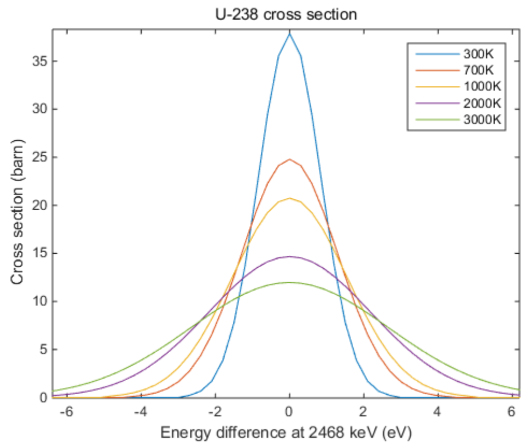

Fig. 4. Doppler broadening of NRF resonance at E=2.468 MeV.

As is well known, the total area under the resonances is rather independent of the material temperature. If temperature increases, the Doppler broadening expands possible energy range in which resonance reaction occurs effectively and it also reduces simultaneously the peak height of resonance with increasing temperature. Consequently, Doppler broadening of neutron capture resonance can result in a slightly enhanced neutron capture by the U-238 resonance in nuclear reactors, i.e., negative fuel temperature coefficient of reactivity, which is critically important for inherent safety of nuclear reactors. The enhanced neutron capture due to Doppler broadening is ascribed to reduced energy self-shielding caused by the lowered peak height in the resonance. Meanwhile, in the NRF resonance reaction, a temperature increase is not expected to enhance the NRF reaction itself since the peak cross section is quite small and the associated energy self-shielding will be very weak. However, a temperature change may affect the probability that a re-emitted photon can cause another NRF reaction, i.e., cascade NRF reactions.

When a photon is re-emitted after an NRF resonance reaction, its energy should be noticeably lower than that of the original incident photon due to recoiling energy of nucleus. The recoil energy loss can be evaluated using the following formula:

$$E_{recoil} = \frac{E^2}{2Mc^2}, \quad (8)$$

where M is the mass of target nuclei and E is the energy of an incident photon causing the NRF resonance reaction. When the energy of incident photon is near 2.468 MeV, recoiling energy is about 13.7 eV, which is similar to total width of the resonance at 3000 K. Therefore, it is clear that the re-emitted photon can hardly cause another NRF reaction, although the associated probability is not zero. Nevertheless, it should be noted that probability of a second NRF reaction by re-emitted photon can be increased significantly by a strong Doppler broadening, as shown in Fig. 4. In particular, if the incident photon energy is low, then the second NRF reaction can be probable for U-238 due to Doppler broadening at high temperature.

4. Conclusions

We compared the Doppler broadening for both neutron capture and photon-induced NRF resonance reactions with temperature variation. It has been found that the FWHM is quite smaller for neutron capture resonance than for the NRF reaction for the same temperature. However, the ratio of the resonance width for two difference temperatures is rather similar for the two types of resonances. In addition, we have shown that a cascade NRF reaction may occur due to a strong Doppler broadening of the NRF resonance for a heavy nuclide, although the associated probability is quite small.

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