Anomalous fluctuations of $s$-wave reduced neutron widths of $^{192,194}$Pt resonances

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We obtained an unprecedentedly large number of $s$-wave neutron widths through $\mathcal{R}$-matrix analysis of neutron cross-section measurements on enriched Pt samples. Careful analysis of these data rejects the validity of the Porter-Thomas distribution with a statistical significance of at least 99.997%.

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Neutron resonance parameters remain some of the most important information for testing random matrix theory (RMT) [1], even more than fifty years after such data served as the original impetus for its creation. Today, RMT pervades the physics of virtually all complex systems and, in the nuclear physics arena, is most often invoked in studies of quantum chaos [2]. Given the conserved symmetries involved, RMT for the Gaussian orthogonal ensemble (GOE) of matrices is expected to correctly describe fluctuation properties of nuclear levels at relatively high excitation such as near the neutron threshold. It implicitly assumes that reduced neutron widths $\Gamma_{\lambda n}^0$ of $s$-wave resonances $\lambda$ follow a Porter-Thomas distribution (PTD) [3], which had been anticipated before RMT emerged. Currently, the overwhelming consensus is that $\Gamma_{\lambda n}^0$ data agree with the PTD. However, there are problems with both the data and analysis techniques used in reportedly the best test of the PTD to date [4] that call these results into question [5]. Measurement and analysis techniques have improved considerably since then, so it is worthwhile to perform new tests of the PTD.

In this Letter, we show that rich $\Gamma_{\lambda n}^0$ data extracted from high resolution neutron total and capture cross sections of $^{192,194}$Pt measured at the Oak Ridge Electron Linear Accelerator (ORELA) facility display a significant departure from the PTD. To our knowledge, this result represents the most stringent test of the PTD to date, and the observed disagreement could have far-reaching consequences.

Loosely stated, the PTD is based on the assumptions that $s$-wave neutron scattering is a single-channel process, the widths are statistical, and time-reversal invariance holds; hence, an observed departure from the PTD implies that one or more of these assumptions is violated, and so could be very interesting.

To make reliable conclusions regarding the validity of the PTD, it is important that the data set be as pure, complete, and large as possible. Perennial problems with neutron resonance data have been (i) contamination of $s$-by $p$-wave resonances and/or resonances of neighboring isotopes, (ii) obtaining enough resonances with known spin $J$ to perform statistically meaningful tests, and (iii) missed resonances due to finite experimental threshold.

In the present case, problem (i) was minimized because Pt is near the peak of the $s$- and minimum of the $p$-wave neutron strength functions ($S_0/S_1 \approx 10$), and because we made high resolution cross-section measurements on natural Pt and four samples enriched in $^{192}$Pt, $^{194}$Pt, $^{195}$Pt and $^{196}$Pt.

Problem (ii) was addressed by combining data for two target nuclei $^{192}$Pt and $^{194}$Pt, containing the largest number of resonances (158 and 411, respectively), and by the fact that all $s$-wave resonances have spin $J = 1/2$ for these even-even nuclides.

Finally, as described below, the novel approach of using an energy-dependent threshold in the analysis helps to solve all three problems. As a result, our $^{192,194}$Pt data are at least as pure, complete, and large as all previous $\Gamma_{\lambda n}^0$ data which have been used for testing the PTD.

Details of the measurements can be found in Ref. [6]. The ORELA was operated at a pulse rate of 525 Hz, a pulse width of 8 ns, and a power of 7-8 kW. Capture measurements were made at a source-to-sample distance of 40.12 m with a pair of $\text{CsI} \delta \text{D} \gamma$ detectors using the pulse-height-weighting technique, and were normalized via the saturated 4.9-eV resonance in the $^{197}$Au($\gamma,\gamma$) reaction. Total neutron cross sections were measured on a separate flight path via transmission using a $^6\text{Li}$-loaded glass scintillator at a source-to-detector distance of 79.83 m.

The $\mathcal{R}$-matrix code SAMMY [7] was used to fit both our transmission and capture data and extract resonance parameters. Resonance energies $E_\lambda$ and neutron widths $g_1 \Gamma_{\lambda n}$, where $g_1$ is the statistical factor for resonances with spin $J$ and $\Gamma_{\lambda n}$, were used in the subsequent analysis described below. For even-even targets, $s$-wave resonances have $g_J \equiv g_{1/2} = 1$, and hence, $g_1 \Gamma_{\lambda n} = \Gamma_{\lambda n}$.

An asymmetrical shape in the transmission data could be used to assign $\ell_n = 0$ resonances [8], see Fig. 1. However, there remained many weak resonances, most of which are $p$ wave, for which we could not unambiguously determine the $\ell_n$ value. As shown below, the potential problem posed by these resonances has been surmounted.

The need to use a threshold on observables $g_1 \Gamma_{\lambda n}$, to guard against possible systematic errors due to in-
struminal effects and \( p \)-wave contamination, was realized very early on \cite{3} in using such data to test theory. Use of an \( E_n \)-dependent \( s \)-wave threshold of the form 
\[
T_0 = a_0 E_n^{3/2},
\]
where \( a_0 \) is a constant factor, is a key improvement in our method compared to previous analyses (in which only energy-independent thresholds were used) for at least three reasons. First, \( p \)-wave contamination is eliminated equally effectively at all energies. Second, statistical precision of the analysis is maximized by allowing instrumented equally effectively at all energies. Third, statistical precision of the instrumental threshold can be surmounted equally effectively at all energies. Third, statistical precision of the analysis is maximized by allowing the largest number of \( s \)-wave resonances to be included.

The PTD is a special case (degrees-of-freedom \( \nu = 1 \)) of the family of \( \chi^2 \) distributions, the probability density function (PDF) of which is denoted hereafter as \( f(x|\nu) \). Because theory predicts fluctuations, but not average values, it is necessary to scale the data to their average, or expectation, value: \( x \rightarrow \Gamma_0^\nu/E[\Gamma_0^\nu] \), where \( E[\bullet] \) denotes the expectation value operator.

We began our fluctuation analysis by employing the maximum-likelihood (ML) method, which has been used since the earliest tests of the PTD, to estimate \( \nu \) and \( E[\Gamma_0^\nu] \). We used threshold \( T_0 \) shown in Fig. 1 (with factor \( a_0 \) given in Table I), which was chosen to reduce \( p \)-wave contamination to very low levels.

The joint PDF for statistical variables \( \Gamma_0^\nu \) and \( E_\lambda \) is defined in a 2D region \( \mathcal{I} \) given by inequalities \( E_\lambda < E_{\max} \) and \( \Gamma_\lambda > T_0(E_\lambda) \), where \( E_{\max} \) is an upper limit of energies \( E_\lambda \). The expression for this PDF reads
\[
\hat{h}^\nu(E_\lambda, \Gamma_\lambda^\nu | \nu, E[\Gamma_\lambda^\nu]) = Cf\left(\frac{\Gamma_\lambda^0}{E[\Gamma_\lambda^0]} | \nu\right).
\] (1)

The factor \( C \), ensuring a unit norm of \( \hat{h}^\nu \), is \( \nu \)- and \( E[\Gamma_\lambda^0] \)-dependent. The ML function was calculated from all \( n_0 \) pairs \([E_\lambda^{\exp}, \Gamma_\lambda^{\exp}]\) obtained from the experiment, which fall into the region \( \mathcal{I} \). Specifically,
\[
L(\nu, E[\Gamma_\lambda^\nu]) = \prod_{i=1}^{n_0} \hat{h}^\nu(E_\lambda^{\exp}, \Gamma_\lambda^{\exp} | \nu, E[\Gamma_\lambda^0]).
\] (2)

A contour plot of this ML function in the form
\[
z(\nu, E[\Gamma_\lambda^\nu]) = 2^{\nu} \left[ \ln L_{\max} - \ln L(\nu, E[\Gamma_\lambda^\nu]) \right]^{\frac{1}{\nu}}
\] (3)
is depicted in Fig. 2. Here, \( L_{\max} \) is the maximum of the ML function. Results of the ML analysis are listed in Table I.

If \( \ln L \) displays near its maximum a shape close to a paraboloid, a contour for a fixed value \( z = k \) will encircle \( \nu \)- and \( E[\Gamma_\lambda^\nu] \)-dependent. The ML function was calculated from all \( n_0 \) pairs \([E_\lambda^{\exp}, \Gamma_\lambda^{\exp}]\) obtained from the experiment, which fall into the region \( \mathcal{I} \). Specifically,
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In the spirit of classical statistics, parameters \( \nu \) and \( E[\Gamma_\lambda] \) are not random variables. Consequently, the function \( z \) refers in ideal conditions to a distribution of estimates of these parameters, not to the parameters themselves. Further, as can be deduced from Fig. 2, the shape of function \( \ln L(\nu, E[\Gamma_\lambda]) \) differs strongly from a paraboloid, which is also the case for 192Pt. So, at this point it is premature to draw a reliable conclusion from the values of \( \tilde{v}_{\exp} \) for 192,194Pt.
To understand how this was achieved, consider a set of \( g_i \) values distributed normally with zero mean and unit variance. For such a set, the statistics

\[
Z = n_0 \sum_{i=1}^{n_0} g_i \quad \text{and} \quad Z^2 = \sum_{i=1}^{n_0} (g_i)^2,
\]

will be governed by the same normal distribution, and by a \( \chi^2 \) distribution with \( \nu = n_0 \) degrees of freedom, respectively. Therefore, for a set of values \( \{g_i^{\exp}\} \) deduced in an appropriate manner from experiment, it is straightforward to use \( Z \) and \( Z^2 \) to calculate probabilities for rejecting the null hypothesis that this set is consistent with the mentioned normal distribution.

To employ these statistics, variable \( \Gamma_\nu \) needs to be transformed to variable \( y \) obeying the considered normal distribution. This was accomplished in two steps. First, the marginal CDF for the above-threshold widths

\[
H_{\Gamma_\nu}^0 (\Gamma_\nu^0 | E[\Gamma_\nu^0]) = \int_0^{E_{\text{max}}} \int_0^1 I^0 (E, \tau_\nu | 1, E[\Gamma_\nu^0]) \, dE \, d\Gamma_\nu^0,
\]

was used to transform \( \Gamma_\nu^0 \) to the variable \( r \), which follows a uniform distribution. This was achieved by substitution \( r = H_{\Gamma_\nu}^0 (\Gamma_\nu^0 | E[\Gamma_\nu^0]) \). In the second step, with the aid of the inverse CDF of the normal distribution with zero mean and unit variance, \( G^{-1}(r) \), we made a transformation \( r \to g \), specifically by \( g = G^{-1}(r) \). Then, quantities

\[
r_i = H_{\Gamma_\nu}^0 (\Gamma_\nu^{\exp} | E[\Gamma_\nu^0]) \quad \text{for} \quad i = 1, 2, \ldots, n_0,
\]

and sets \( \{g_i^{\exp}\} \) were calculated using values \( \Gamma_\nu^{\exp} > T_0(E_{\lambda_i}) \) from the experiments. Following this procedure, we easily determined probabilities \( p_Z = P(Z > Z_{\exp} | E[\Gamma_\nu^0]) \) and \( p_{Z^2} = P(Z^2 > Z_{\exp}^2 | E[\Gamma_\nu^0]) \). Here, for a fixed value \( E[\Gamma_\nu^0] \), values of \( Z_{\exp} \) and \( Z_{\exp}^2 \) are given by Eqs. (4). After replacement \( g_i \to g_i^{\exp} \). Hence, probabilities \( p_Z \) and \( p_{Z^2} \) represent separate statistical significances for rejecting the PTD at various values of \( E[\Gamma_\nu^0] \). Both probabilities, calculated from \( 192,194^{\text{Pt}} \) data, are plotted in Fig. 3.

As seen, the critical values of \( E[\Gamma_\nu^0] \) for testing the validity of the PTD range from 5.3 to 6.9 meV and from 15.0 to 18.3 meV for \( 192 \text{Pt} \) and \( 194^{\text{Pt}} \), respectively.

Before proceeding further, we performed an additional analysis to verify that \( p \)-wave contamination was negligibly small. To this end, we performed ML calculations based on a hybrid PDF for a mixture of \( s \) and \( p \) observables \( g_i \Gamma_{\lambda_i} \) obeying two separate PTDs:

\[
h^0(E_{\lambda_i}, g_i \Gamma_{\lambda_i} | \mu_0, \mu_1, \theta) = \int_0^{\frac{D}{\mu_0 E_{\lambda_i}^2}} f \left( \frac{g_i \Gamma_{\lambda_i}}{\mu_0} \frac{1}{E_{\lambda_i}^2} \right) \, d\frac{1}{E_{\lambda_i}^2}.
\]

TABLE I: Results of ML-based fluctuation analysis of \( s \)-wave reduced neutron widths of \( 192,194,196^{\text{Pt}} \) resonances.

| Sample | \( E_{\text{max}} \) (keV) | \( a_0 \) (eV⁻¹) | \( n_0 \) | \( \tilde{\nu}_{\text{exp}} \) (eV⁻¹) | \( a_{\text{ml}} \) (eV⁻¹) | \( \tilde{\mu}_0 \) (meV) | \( \tilde{\mu}_1 \) (meV) | \( R \) | \( S \) |
|--------|----------------|-------------|--------|----------------|-------------|-------------|-------------|----|----|
| \( 192^{\text{Pt}} \) | 4.98 | 7.00 × 10⁻⁶ | 153 | 0.57 | 0.06 | 0.16 | -0.15 | 5.91 ± 0.71 | (2.68 ± 0.47) × 10⁻⁶ | 26.1 ± 4.5 | 0.9970 |
| \( 194^{\text{Pt}} \) | 15.93 | 2.25 × 10⁻⁶ | 161 | 0.47 | 0.19 | 0.18 | -0.16 | 14.9 ± 1.5 | (8.84 ± 1.09) × 10⁻⁶ | 25.4 ± 3.1 | 0.9975 |
| \( 196^{\text{Pt}} \) | 15.99 | 3.19 × 10⁻⁶ | 68 | 0.60 | 0.28 | 0.36 | -0.26 | 39.7 ± 6.4 | (14.8 ± 1.6) × 10⁻⁶ | - | - |

*Due to a high instrumental threshold, \( \tilde{\mu}_1 \) was deduced assuming that \( 192,194,196^{\text{Pt}} \) share a common true value of \( \mu_0/\mu_1 \).
Here, $\mu_0$ and $\mu_1$ stand for expectation values $E[\Gamma_{\lambda n}]$ and $E[\mu_1^2 \frac{1eV}{E_\lambda^3}]$ referring to $s$- and $p$-wave resonances, respectively, while $\theta$ represents the $J$-independent resonance-density ratio $\rho_J/\rho_{J+}$. The PDF $h_0^0$ is defined in the region limited from below by threshold $T_{01} = a_0 E_\lambda^{3/2}$ with $a_0 \ll a_1$ which is at the same time safely higher than experimental threshold $T = a E_\lambda^{3/2}$. We assumed that $\mu_1$ is $J$-independent. For mass numbers $A \approx 190$ this is justified, as it holds with about 1% precision that $g_{1/2}/g_{3/2} = f(3/2)/f(1/2)$, where $f(J)$ is the resonance-density spin factor [4]. Factor $D$ ensures the unit norm of $h_0^0$.

Following a path analogous to that described above, we constructed ML function $L(\mu_0, \mu_1)$ and arrived at estimates $\hat{\mu}_0$ and $\hat{\mu}_1$ given in Table I. In case of $^{192}$Pt, $\hat{\mu}_1$ has been determined indirectly, see Table II. The method of its determination is supported by the fact that ratios $\hat{\mu}_0/\hat{\mu}_1$ for $^{194,196}$Pt are within their r.m.s uncertainties equal each other. In Table III values of the dimensionless, energy-independent quantity $R = (1 \text{ eV}^{3/2}) \frac{\hat{\mu}_0}{\hat{\mu}_1}$, which represent ratios of $T_0(E_\lambda)$ to local expectation values $E[\Gamma_{\lambda n}]$ for $p$-wave resonances, are listed for $^{192,194}$Pt targets. From these values and their r.m.s uncertainties, we calculated probabilities of 0.069% and 0.0047% for $^{192}$Pt and $^{194}$Pt, respectively, that among the observables $g_J \Gamma_{\lambda n} > T_0(E_\lambda)$ there occurs one which belongs to a $p$-wave resonance, thus verifying that $p$-wave contamination is negligible.

With the question of $p$-wave contamination settled then, the data in Fig. III indicate that for $s$-wave resonances in $^{192}$Pt and $^{194}$Pt, the validity of the PTD is rejected with statistical significance levels $S_{192} = 0.9970$ and $S_{194} = 0.9975$, respectively, in excellent agreement with our initial ML analysis. Because results for the two isotopes should be independent, the combined probability that the PTD is valid is less than $1.2 \times 10^{-5}$. Although we have shown above that it is very unlikely, if a $p$-wave intruder occurs among the $s$-wave $^{192}$Pt observables, we calculate that if the resonance having the smallest $g_J \Gamma_{\lambda n}$ value (relative to threshold $T_0$) is considered to be $p$ wave, this probability will increase in the worst case to $2.8 \times 10^{-5}$.

We conclude that our data reject the validity of the PTD with a statistical significance of at least 99.997%. This inescapable conclusion has been made thanks to rich experimental data obtained using state-of-the-art neutron spectroscopy, and the implementation of a novel approach for testing the PTD. On the other hand, equally convincing evidence that the PTD holds for some or the majority of heavy and intermediate-weight nuclei is still missing.

This result implies that at least one of the three assumptions behind the PTD is violated. For energies of the measurements reported herein, only elastic scattering is possible, so the single-channel assumption is valid. Also, addition of another channel would result in $\nu > 1$, which would disagree even more strongly with the data than the PTD does. Violation of time-reversal invariance also implies $\nu > 1$, and therefore also is excluded. Hence, our results indicate the assumption that the widths are statistical is violated. However, there are no indications of non-statistical effects such as doorway states in the data.

One possible explanation is suggested by the calculations of Ref. [17] in which it was found that transition strength distributions deviated further from the PTD, in the direction of smaller $\nu$, as model quantum-mechanical systems became more collective. Hence, our result that $\nu \approx 0.5$ for the $^{192,194}$Pt+n systems suggests the surprising conclusion that $^{193,195}$Pt display regular, rather than the expected chaotic, behavior at relatively high excitations near the neutron threshold.

Alternatively, our results also could be interpreted as indicating that the PDF for reduced neutron width amplitudes for the $^{192,194}$Pt+n systems are not form invariant. In Ref. [11] it was shown that this form-invariance assumption could replace the original somewhat qualitative “statistical” assumption as part of a more general derivation of the PTD. Violation of this assumption could have far-reaching consequences.

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[1] T. Guhr et al., Phys. Repts. 299, 189 (1998).
[2] H. A. Weidenmuller and G. E. Mitchell, Rev. Mod. Phys. 81, 539 (2009).
[3] C. E. Porter and G. E. Thomas, Phys. Rev. 104, 483 (1956).
[4] O. Bohigas, R. U. Haq, and A. Pandey, in Nuclear Data for Science and Technology, edited by K. H. Bockhoff (D. Reidel, Dodrecht, 1983), p. 899.
[5] P. E. Koehler, EPJ Web of Conferences 2, 05001 (2010).
[6] P. E. Koehler et al., J. Nucl. Sci. and Tech., Suppl. 2, 546 (2002).
[7] N. M. Larson, Oak Ridge National Laboratory Technical Report No. ORNL/TM-9179/R8, 2008.
[8] For the sake of brevity subscripts $\lambda$ are dropped.
[9] H. Zhongfu et al., Chin. J. Nucl. Phys. 13, 147 (1991).
[10] Y. Alhassid and A. Novoselsky, Phys. Rev. C 45, 1677 (1992).
[11] T. J. Krieger and C. E. Porter, J. Math. Phys. 4, 1272 (1963).