Comparison of XYZ Model Fitting Functions for ^{111}Cd in In_3La

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Abstract.
The XYZ model describes the interaction between nuclear probes and an electric field gradient that fluctuates among three orthogonal directions. The model presents a means to calculate the perturbation function that represents spectra obtained using perturbed angular correlation spectroscopy. Three analytic approximations of the perturbation function have been developed previously, and they are evaluated in the present paper in the context of Cd jumping among In-lattice sites in In_3La.

Keywords: PAC, XYZ Model, Relaxation

Recently, the jump frequency of ^{111}Cd tracers on the In sublattice in In_3La was measured via nuclear quadrupole relaxation using the method of perturbed angular correlation of gamma rays (PAC) [1]. In_3La has the Cu_3Au, or $L1_2$, structure, for which an In-lattice site has an axially symmetric electric field gradient (EFG). Principal axes of EFGs at neighboring In sites are orthogonal, so that the EFG at the nucleus of a PAC probe reorients by 90° in each jump. A model for such a fluctuating field interacting with a PAC probe has been termed the XYZ model [2].

When EFGs reorient in the XYZ model, there is a relaxation of the perturbation function $G_2(t)$ due to a loss of coherence in gamma radiation emitted by the ensemble of probes. Measurements of the relaxation can be used to determine the frequency of reorientation w_{EFG} of an EFG from one orientation to one other orientation. For the XYZ model, approximate perturbation functions [2–4] of various degrees of sophistication have been developed for spin-5/2 PAC probes. The present work compares the effectiveness of fitting using approximate functions to the results of full numerical fits as were done in Ref. [1].

The approximate perturbation functions considered here are those by Baudry and Boyer [3], Evenson *et al.* [2], and Guan [4]. In each work, two functions were given: one for the slow fluctuation regime ($w_{\mathrm{EFG}} \ll \omega_{\mathrm{Q}}$) and one for the fast fluctuation regime ($w_{\mathrm{EFG}} \gg \omega_{\mathrm{Q}}$), where ω_{Q} is the quadrupole interaction frequency.

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Baudry and Boyer found

$$G_2(t) \approx \mathrm{e}^{-\lambda t}\left[\frac{1}{5}+\frac{13}{35}\cos(6\omega_{\mathrm{Q}}t)+\frac{2}{7}\cos(12\omega_{\mathrm{Q}}t)+\frac{1}{7}\cos(18\omega_{\mathrm{Q}}t)\right] , \tag{1}$$

with relaxation parameter $\lambda = 2w_{\mathrm{EFG}}$, for $w_{\mathrm{EFG}} \ll \omega_{\mathrm{Q}}$, and

$$G_2(t) \approx \mathrm{e}^{-\lambda t} , \tag{2}$$

with relaxation parameter $\lambda = 100\omega_{\mathrm{Q}}^2/3w_{\mathrm{EFG}}$, for $w_{\mathrm{EFG}} \gg \omega_{\mathrm{Q}}$ [3].

In the approximation by Evenson *et al.*,

$$\begin{aligned} G_2(t) \approx\ & (1/5)\mathrm{e}^{-\lambda_0 t} + (13/35)\mathrm{e}^{-\lambda_1 t}\cos(6\omega_{\mathrm{Q}}t) \\ & +(2/7)\mathrm{e}^{-\lambda_2 t}\cos(12\omega_{\mathrm{Q}}t) + (1/7)\mathrm{e}^{-\lambda_3 t}\cos(18\omega_{\mathrm{Q}}t) , \end{aligned} \tag{3}$$

with relaxation parameters $\lambda_0 = 1.4896w_{\mathrm{EFG}}$, $\lambda_1 = 1.6237w_{\mathrm{EFG}}$, $\lambda_2 = 1.8173w_{\mathrm{EFG}}$, and $\lambda_3 = 2.1301w_{\mathrm{EFG}}$, for $w_{\mathrm{EFG}} \ll \omega_{\mathrm{Q}}$ [5], and

$$G_2(t) \approx (4/7)\mathrm{e}^{-\lambda_0 t} + 0.3032\mathrm{e}^{-\lambda_1 t} + 0.0968\mathrm{e}^{-\lambda_2 t} + (1/35)\mathrm{e}^{-\lambda_3 t} , \tag{4}$$

with relaxation parameters $\lambda_0 = 38\omega_{\mathrm{Q}}^2/w_{\mathrm{EFG}}$, $\lambda_1 = 10.15\omega_{\mathrm{Q}}^2 t/w_{\mathrm{EFG}}$, $\lambda_2 = 63.85\omega_{\mathrm{Q}}^2/w_{\mathrm{EFG}}$, and $\lambda_3 = 92\omega_{\mathrm{Q}}^2/w_{\mathrm{EFG}}$, for $w_{\mathrm{EFG}} \gg \omega_{\mathrm{Q}}$ [2].

In the approximation by Guan, [1]

$$\begin{aligned} G_2(t) \approx (1/5)\ & \mathrm{e}^{-\lambda_0 t} + (13/35)\mathrm{e}^{-\lambda_1 t}\cos(6\omega_{\mathrm{Q}}t - \phi_1)/\cos(\phi_1) \\ & +(2/7)\mathrm{e}^{-\lambda_2 t}\cos(12\omega_{\mathrm{Q}}t - \phi_2)/\cos(\phi_2) \\ & +(1/7)\mathrm{e}^{-\lambda_3 t}\cos(18\omega_{\mathrm{Q}}t - \phi_3)/\cos(\phi_3) , \end{aligned} \tag{5}$$

with relaxation parameters $\lambda_0 = 1.5w_{\mathrm{EFG}}$, $\lambda_1 = 1.62w_{\mathrm{EFG}}$, $\lambda_2 = 1.8w_{\mathrm{EFG}}$, and $\lambda_3 = 2.05w_{\mathrm{EFG}}$ and phase factors $\phi_1 = 0.18w/\omega_{\mathrm{Q}}$, $\phi_2 = 0.27w/\omega_{\mathrm{Q}}$, and $\phi_3 = 0.154w/\omega_{\mathrm{Q}}$, for $w_{\mathrm{EFG}} \ll \omega_{\mathrm{Q}}$, and

$$G_2(t) \approx (1/5)\mathrm{e}^{-\lambda_0 t} + (13/35)\mathrm{e}^{-\lambda_1 t} + (2/7)\mathrm{e}^{-\lambda_2 t} + (1/7)\mathrm{e}^{-\lambda_3 t} , \tag{6}$$

with relaxation parameters $\lambda_0 = 207\omega_{\mathrm{Q}}^2/7w_{\mathrm{EFG}}$, $\lambda_1 = 29\omega_{\mathrm{Q}}^2 t/w_{\mathrm{EFG}}$, $\lambda_2 = 497\omega_{\mathrm{Q}}^2/10w_{\mathrm{EFG}}$, and $\lambda_3 = 19\omega_{\mathrm{Q}}^2/w_{\mathrm{EFG}}$, for $w_{\mathrm{EFG}} \gg \omega_{\mathrm{Q}}$ [4]. Note that λ_n and w_{EFG} are in MHz when ω_{Q} is in Mrad/s.

PAC spectra obtained for ^{111}Cd in In_3La at different measurement temperatures can be found in Ref. [1]. w_{EFG}, reported in Ref. [1], and ω_{Q}, reported in Ref. [6], were determined by full numerical fits, *i.e.* minimizing the difference between spectra and test-$G_2(t)$ functions generated numerically from the XYZ model as described in reference [2]

[1] Our Eq. 5 has the correct signs for the phase shifts, and has corrected the expressions for ϕ_n in terms of ω_{Q}.

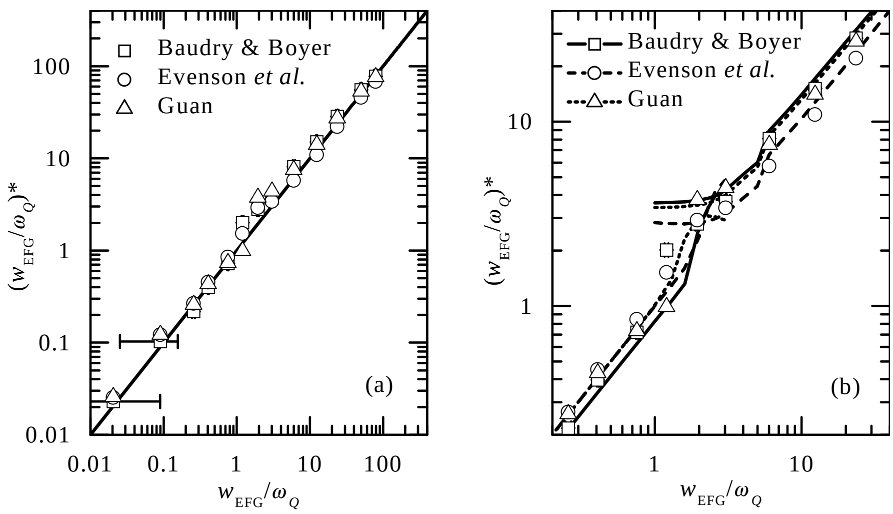

Figure 1. (a) Comparison of results of approximate fits $(w_{\rm EFG}/\omega_{\rm Q})^*$ plotted vs. results of full numerical fits $w_{\rm EFG}/\omega_{\rm Q}$. (b) Enlargement of central portion of (a).

by adjusting $w_{\rm EFG}$ and $\omega_{\rm Q}$. Spectra were refitted in the present study to Eqs. 1–6. Results $(w_{\rm EFG}/\omega_{\rm Q})^*$ obtained from fits here are plotted vs. results $w_{\rm EFG}/\omega_{\rm Q}$ obtained from full numerical fits as data points in Fig. 1. Note that $\omega_{\rm Q}$ can only be fitted in the slow fluctuation regime, so that values of $\omega_{\rm Q}$ used in the fast fluctuation regime were determined by extrapolating results from the slow fluctuation regime.

Fig. 1(a) shows results over the range of $w_{\rm EFG}/\omega_{\rm Q}$ that is accessible to measurement. The line indicates values for which $(w_{\rm EFG}/\omega_{\rm Q})^*$ would agree exactly with $w_{\rm EFG}/\omega_{\rm Q}$. As can be seen, results from approximate fits agree well with those of the full numerical fits across the whole range, with the most notable deviation located at the intermediate fluctuation regime ($w_{\rm EFG}/\omega_{\rm Q} \approx 1$). The deviation arises because the discrepancy between the approximate functions and the XYZ model and correlation between $w_{\rm EFG}$ and $\omega_{\rm Q}$ are greatest in this range.

Fig. 1(b) focuses on a narrower range of $w_{\rm EFG}/\omega_{\rm Q}$ in order to better compare the different fitting methods. Here, curves indicate results of approximate fits to $G_2(t)$ generated by simulations of the XYZ model. These results suggest that one should use the approximations for the slow fluctuation limit up to $w_{\rm EFG}/\omega_{\rm Q} \approx 2$ and approximations for the fast fluctuation regime above. Some important points regarding fitting PAC spectra using approximate functions for $G_2(t)$ are as follows.

– *Best approximate form.* At least in the case of Cd in In_3La, fits using the approximate forms by Evenson *et al.* (our Eqs. 3 and 4) work best.

- *Range of available data affects results.* Fitting with approximate functions is expected to work well when data are available across the full range of slow and fast fluctuation regimes. When data are fitted only for $w_{\mathrm{EFG}}/\omega_{\mathrm{Q}} \leq 2$, systematic errors can be expected in the determination of the dependence of fluctuation frequency on temperature or other experimental parameters. The degree of error is expected to increase as the lower limit of the range of fitted data increases.
- *Experimental effects on results.* Measured PAC spectra include data uncertainties that depend on time as well as experimental artifacts coming from the experimental geometry and other aspects of the experimental setup. These experimental factors affect the results of fitting with approximate functions, as can be seen in Fig. 1(b) by the deviations between data points and curves.
- *Cubic offset problem in fast fluctuation limit.* If a cubic offset is fitted for spectra, the relaxation parameters and the cubic offset are highly correlated in the fast fluctuation regime. So, the cubic offset must be held fixed at a known value in order to get reliable values of the fluctuation frequencies in the fast regime. This problem applies to full numerical fits as well as approximate fits.

It is now computationally practicable to fit PAC spectra by generating perturbation functions numerically from an appropriate dynamical model, providing the model is sufficiently simple. We advocate fitting spectra with such full numerical fits in order to avoid uncertainties in interpetation of data caused by the above issues.

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