Supporting Information

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SI Text

I. Experimental Technique. Large single crystals of $PuCoGa_5$ with exposed crystallographic facets were grown by the self-flux method as described in the work by Sarrao et al. (1). A single crystal was polished to the dimensions of $2.208 \times 2.240 \times 0.641$ mm, with the tetragonal long axis being 2.208 mm. The drive transducer of the resonant ultrasound spectroscopy apparatus was driven well below its own first compressional resonance (from 100 kHz to 2.5 MHz). The response voltage generated on the pickup transducer—maximum whenever the drive frequency coincides with a sample resonance—was measured with a custom-built heterodyne amplifier (2).

The resonant ultrasound spectroscopy apparatus was custom-built for the experiment, comprising two 2-mm-diameter compressional-mode lithium-niobate piezoelectric transducers with vibration isolation from the rest of the apparatus. The top transducer was mounted on a freely pivoting arm, ensuring weak coupling and linear response. Temperature control was provided by an He^4 flow cryostat. The temperature was swept over 28 h from 295 to 13 K, sweeping at one-half the rate in the 50- to 13-K region. During the temperature sweep, the positions of 33 resonances were tracked. By sweeping frequency in nonlinear steps and taking data only near the resonances, temperature steps of 0.2 K (and 0.1 K from 13 to 50 K) were achieved in the 28-h period (3).

II. Data Analysis. If the elastic moduli, the sample dimensions, and the density are known, then the resonances can be calculated by solving the 3D elastic equation (4). However, given the experimentally determined resonance frequencies, the inverse problem must be solved to obtain the elastic moduli. If approximate values of the moduli are known a priori, then a standard gradient-based minimization scheme can be used to fit the moduli using the measured resonances (4). This method relies on the assumption that there are no missing resonances in the dataset, which is not always valid, because some modes may not be excited for a given sample transducer geometry.

To overcome the unknown initial conditions and the possibility of missing resonances, a data analysis procedure was developed using a genetic algorithm, which allows for loose bounds on the initial conditions (5–8). This algorithm has the following structure.

i) N sets of randomly generated elastic moduli (six elastic moduli per set generated within broad initial ranges) are created, where N is ∼10 times the number of elastic moduli. These sets are the parent sets, each denoted x_i^{parent}.

ii) N mutant sets of moduli are generated by $x_i^{mutant} = x_k^{parent} + s(x_l^{parent} - x_m^{parent})$, where k, l, and m are random integers between 1 and N, and s is a user-defined scaling parameter that controls the speed and accuracy of convergence; s is typically small, such that the mutant vectors are relatively small perturbations from the parent vectors.

iii) Crossing is performed by taking the ith vector from the parent set, taking the ith vector from the mutant set, and creating a third child vector by taking a modulus from the parent with probability p or the mutant with probability $1-p$ (repeated individually for each moduli in the set).

iv) For each parameter set in the parent and child pools, the resonance frequencies are calculated by numerically solving the elastic wave equation (4). The sum-of-squares residuals between the measured and computed frequencies are then calculated. For each x_i^{parent} and x_i^{child} pair, the parameter set with the lowest residual is selected for the next generation of evolution.

v) If the lowest residual is below the specified tolerance, then the algorithm terminates, or it returns to step *ii* for the next generation.

The computationally expensive step—numerically solving the elastic equation for each parameter set—can be farmed out to multiple processors, giving this algorithm very good scaling properties as the number of moduli and resonances increases. Because each resonance is primary related to two or three, rather than all five, elastic strains, a genetic algorithm is particularly effective—the level of independence between the moduli in a given parameter set results in faster convergence (7, 8).

When it is suspected that not all resonance modes have been measured in a given frequency range, a modification is made to step *iv*, which proceeds as follows.

i) The user inputs the maximum number of resonances that they think have been missed.

ii) The residuals are computed for each set as normal. Then, one missing peak is inserted between the first and second resonances, and the residual is recomputed. The missing resonance has a weight of zero for purposes of the residual calculation, because the missing frequency is unknown, but it has the effect of shifting all higher frequency resonances by one. The resonance is then inserted between the second and third resonances, and the residual is recomputed, continuing until the missing resonance has been inserted into every possible position. This procedure is then repeated for all possible combinations of two missing resonances and then, three, etc. until the maximum number of missing resonances is reached. The residual used for competition between parent and child sets is the lowest residual found during this process.

iii) At the end of each generation, the user can see whether resonances may be missing or not and where to look in the experimental frequency range; if there are no missing resonances, then all of the missing resonances will be placed at the end of the list of measured resonances.

This procedure, although crude, turns out to be remarkably effective at locating missing resonances: additional inspection of the data taken with a denser frequency scan and higher ultrasonic power almost always resulted in the identification of a previously missing resonance. However, if the numbers of missing and total resonances are large, then this procedure becomes prohibitively expensive: 60 resonances with 5 missing require $60!/5!55! = 5,461,512$ residual calculations per parameter set; 15 missing peaks would require over 53 trillion—in which case, a more elegant solution would be required.

On completion of the algorithm, the logarithmic derivatives of each resonance frequency with respect to each modulus is computed numerically:

$$\frac{\partial f_\mu}{\partial c_i} \times \frac{c_i^0}{f_\mu^0} \equiv \alpha_{i,\mu}, \qquad \text{[S1]}$$

where f_μ is a particular resonance frequency, c_i is one of six elastic moduli, and f_μ^0 and c_i^0 are the values at a reference temperature (usually 300 K). These α-coefficients are essentially geometric factors: all of the temperature dependences of each resonance f_μ are built from linear combinations of the

temperature-dependent c_i values, and the α-coefficients change by, at most, a couple of percentages from 300 K down to 4.2 K. In this way, we can write

$$\frac{2\Delta f_\mu(T)}{f_\mu^0} = \sum_i \alpha_{i,\mu} \frac{\Delta c_i(T)}{c_i^0}. \quad \text{[S2]}$$

The α-values defined in this way have the property that $\sum_i \alpha_i = 1$, and using these coefficients, the different resonances can be identified as predominantly shear or scalar. In addition, because the $\alpha_{i,\mu}$ values are essentially temperature-independent, after a fit has been performed at one temperature and the $\alpha_{i,\mu}$ and c_i^0 values are obtained, at each temperature, the frequencies $\Delta f_\mu(T)$ and their associated coefficients $\alpha_{i,\mu}$ form an overdetermined set of linear equations that can be solved for the elastic moduli at that temperature. This method provides a second route to computing the temperature dependence of the elastic moduli (as opposed to performing the fit at each temperature) and is an important consistency check.

III. Symmetry and Coupling. Elastic strain breaks into five irreducible representations in a tetragonal lattice: two scalar strains transforming as the A_{1g} representation and three shear strains transforming as the B_{1g}, B_{2g}, and E_g representations. Specifically, the strains and associated moduli are

Two scalar strains are in-plane $\epsilon_{A_{1g},x} \equiv (\epsilon_{xx} + \epsilon_{yy})/2$ and out-of-plane $\epsilon_{A_{1g},z} \equiv \epsilon_{zz}$, with respective moduli $(c_{11} + c_{12})/2$ and c_{33}.

Three shear strains are $\epsilon_{B_{1g}} \equiv (\epsilon_{xx} - \epsilon_{yy})/2$, $\epsilon_{B_{2g}} \equiv \epsilon_{xy}$, and $\epsilon_{E_g} \equiv \{\epsilon_{xz}, \epsilon_{yz}\}$, with respective moduli $(c_{11} + c_{12})/2$, c_{66}, and c_{44}.

The elastic free energy includes all bilinear combinations of these strains, allowing for a cross-term between the two A_{1g} strains that defines the sixth modulus c_{13}. The free energy is then

$$F_{elastic} = \sum_\mu \frac{1}{2} c_\mu \epsilon_\mu^2 + c_{13}\left(\epsilon_{A_{1g},x}\right) \times \left(\epsilon_{A_{1g},z}\right), \quad \text{[S3]}$$

where ϵ_μ is one of five strains, and c_μ is the corresponding modulus. Within Landau theory, an order parameter-η is introduced that vanishes linearly at temperature T_0, and its contribution to the free energy is

$$F_{o.p.} = \frac{1}{2}\alpha(T - T_0)\eta^2 + \frac{1}{4}\gamma\eta^4, \quad \text{[S4]}$$

where α and γ are constants. (The inclusion of cubic terms, which are allowed for A_{1g}-order parameters, does not change the qualitative behavior of the elastic moduli above T_0.) When the order parameter and the strain have the same symmetry, they can couple linearly:

$$F_{coupling} = \beta\epsilon_\mu\eta, \quad \text{[S5]}$$

where β is a coupling parameter. The total free energy is then

$$F = F_{elastic} + F_{o.p.} + F_{coupling}. \quad \text{[S6]}$$

We eliminate η by minimizing the free energy at fixed strain, $\partial F/\partial\eta|_\epsilon = 0$, which gives an elastic modulus that diverges as $1/(T - T_0)$ as T_0 is approached from above (9):

$$\begin{aligned} c'_\mu = \frac{\partial^2 F}{\partial \epsilon_\mu^2} &\approx c_\mu - \frac{\beta^2}{\alpha(T - T_0)} && T > T_0 \\ &\approx c_\mu - \frac{\beta^2}{2\alpha(T_0 - T)} && T < T_0. \end{aligned} \quad \text{[S7]}$$

This linear coupling between strain and order parameter is only possible when the order parameter shares all symmetries with the strain and not just the point-group symmetry. This constraint means that the order parameter must be symmetric under time reversal [no linear coupling of magnetic order parameters to strain (10)], preserve gauge symmetry (no linear coupling of the superconducting order parameter), etc.

When the strain couples to the square of the order parameter, $F_{coupling} = \beta\epsilon_\mu\eta^2$, the same exercise can be carried using equilibrium conditions $\eta = 0$ for $T > T_c$ and $\eta = \sqrt{\alpha(T_c - T) - \beta\epsilon/\gamma}$ for $T < T_c$, which gives

$$\begin{aligned} c'_\mu &= c_\mu && T > T_c \\ &= c_\mu - \frac{\beta^2}{2\gamma} && T < T_c. \end{aligned} \quad \text{[S8]}$$

In this case, there is no softening of the elastic modulus above T_c and only a drop in elastic modulus across the transition. Because the square of any representation contains a scalar (A_{1g} in a tetragonal lattice), the scalar moduli can always couple to the square of an order parameter—for example, superconductivity. A more interesting case would be a magnetic order parameter that breaks inversion with symmetry E_u. The square of this order parameter contains A_{1g}, A_{2g}, B_{1g}, and B_{2g} objects, which means that all moduli except for c_{44} (with E_g symmetry) should show a jump across the magnetic ordering transition.

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