Measurement of the Angular Dependence of Spontaneous Raman Scattering in Anisotropic Crystalline Materials Using Spherical Samples: Potassium Dihydrogen Phosphate as a Case Example

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A new and flexible experimental configuration has been developed and tested to measure the spontaneous Raman scattering for samples with any crystal cut, probed with any specific pump polarization, and for which the scatter signal in any direction and with any polarization can be measured. This experimental requirement stems from the challenges, arising from the complexity of light propagation, in obtaining accurate measurements of the angular dependence of the Raman scattering cross section in birefringent materials. The nonlinear optical material KH_2PO_4 (KDP) is used as the model medium. This study is motivated by the need to improve our understanding and management of transverse stimulated Raman scattering (TSRS) in KDP crystals typically used for frequency conversion and polarization control in large-aperture laser systems. Key to this experimental platform is the use of high-quality spherical samples that enable one to measure the Raman scattering cross section in a wide range of geometries using a single sample. The system demonstrated in this work is designed to enable experiments to (1) develop a better understanding of the Raman polarizability tensor and (2) directly measure the angular dependence of the spontaneous Raman scattering in a crystal cut suitable for polarization control.

The spontaneous Raman scattering is an experimentally measureable quantity from which the Raman polarizability tensor of vibrational modes of interest can be established using the theoretically expected formulation based on symmetry as a guide.[1] Given this tensor, the Raman-scattering cross section can be estimated at any orientation and the corresponding TSRS gain coefficient can be calculated. Efforts to develop an empirical description of the Raman tensor has provided an approximation for the off-diagonal elements, but its precise form remains incomplete,[2] while measurements for the Raman scattering cross section are limited.[3] An accurate Raman polarizability tensor would enable (1) the modeling of TSRS for multiple crystal-cut configurations to guide the design of KDP-based optical elements that provide minimum SRS gain, (2) the estimation of material limits in inertial confinement fusion laser designs, and (3) the optimization of hardware designs and operational conditions.

Based on the consideration discussed above, we developed an experimental Raman scattering spectroscopy system (Fig. 1) that uses spherical crystal (KDP) samples and facilitates the measurement of the angular dependence of the spontaneous Raman scattering cross section in directions orthogonal to beam propagation for any specific crystal orientation of interest. The optic axis of the sample sphere is oriented by two programmable rotation stages. Custom vacuum chucks, installed in the open aperture of the rotation stages, are used to "hold" the sphere and rotate it. Any position on the sphere (4π steradians) can be reached with a combination of, at most, three stage moves. Linear stages provide additional vertical and horizontal alignment capabilities in the x–y plane to overlap the sphere center and the excitation laser beam focus with the focal point of the signal collection optics.

The Raman scattering intensity of the totally symmetric mode (914 cm^{-1}) integrated over 100 cm^{-1} about the peak (860 to 960 cm^{-1}) as a function of the azimuthal angle ϕ is shown in the results presented in Fig. 2. The optic axis (OA) was aligned in the azimuthal (horizontal) plane, and for $\phi = 0°$, the OA was oriented along the laboratory z axis (beam propagation direction). Four combinations of polarization of the excitation laser and the analyzer were used, with the excitation polarization parallel to the laboratory x or y axis and the analyzer parallel to the laboratory z or y axis. Accordingly, the inset in Fig. 2 describes the ini-

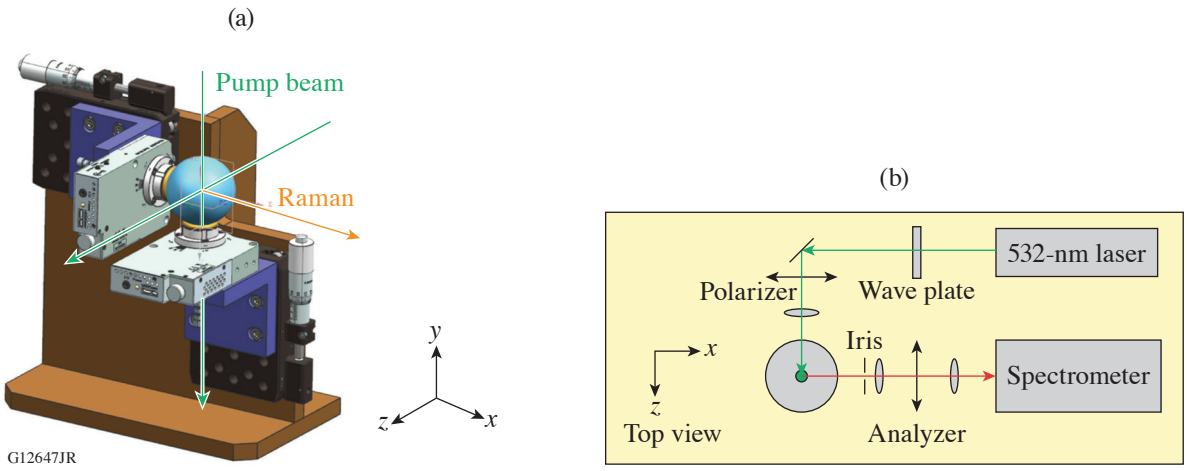
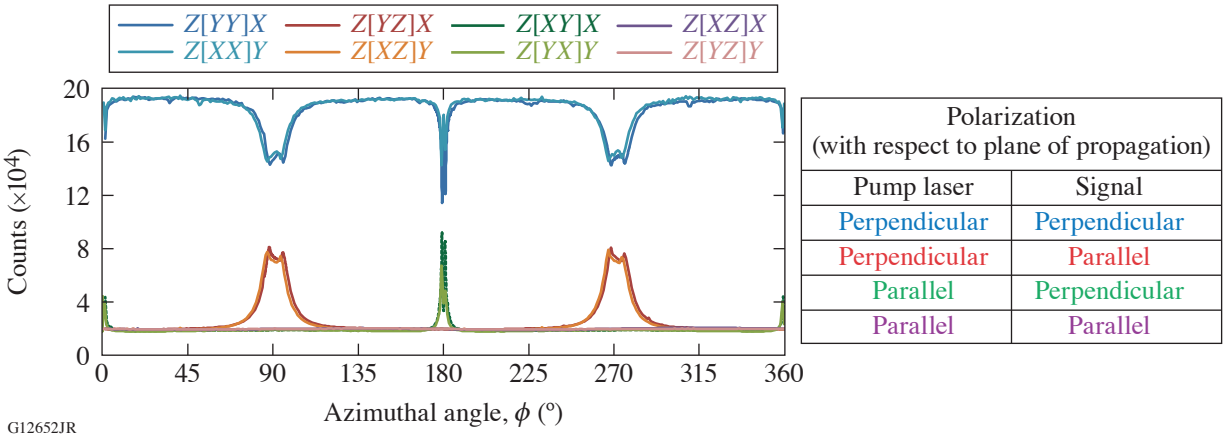


Figure 1
Schematic depiction of (a) the sample holder and (b) the Raman scattering spectroscopy system utilizing high-quality spherical samples to measure the Raman scattering signal orthogonal to the laser excitation beam. The excitation beam propagates along the horizontal laboratory z axis, while the Raman signal was measured at an orthogonal direction along the horizontal laboratory x axis.



Polarization (with respect to plane of propagation)	
Pump laser	Signal
Perpendicular	Perpendicular
Perpendicular	Parallel
Parallel	Perpendicular
Parallel	Parallel

Figure 2
The integrated spontaneous Raman scattering signal of the totally symmetric internal PO_4 mode is measured as a function of the azimuthal angle for four different initial configurations, which are given in the legend and follow Porto notation. The letters to the left and right of the brackets define the propagation direction of the incident and scattered light; the letters inside the brackets, left to right, are the polarization of the incident and scattered light. Additional information on measurement geometry is provided in the table.

tial setting of the Raman scattering measurement in the Porto notation.[4] The notation in the legend of Fig. 2 refers to the crystal axes (X, Y, Z) at the initial position $\phi = 0°$. Measurements were performed with the Raman signal initially measured along the X crystal axis and then after rotating the sample by 90° about the Z crystal axis to measure the Raman signal along the Y crystal axis (X or Y directions are indistinguishable during measurement and therefore arbitrarily assigned). Consequently, eight different measurements were performed with the above-described initial alignment conditions.

The blue traces in Fig. 2 represent data acquired when both the excitation and signal polarizations were perpendicular to the (horizontal) plane of propagation. Wide double valleys are found at $\phi = 90°$ and 270°, while narrow double valleys are found at $\phi = 0°$ and 180°. An inverse set (red trace) of wide (only) double peaks is seen when the excitation laser is perpendicularly

polarized. The presence of narrow double peaks (green trace) requires a parallel polarized excitation laser, but perpendicularly polarized Raman signal. The Raman signal is very low when both the excitation and the signal polarizations are parallel to the plane of propagation. Data sets were nearly indistinguishable when the sphere was rotated 90° about the optics axis to interchange to crystal *X* and *Y* axes.

The data acquired using a 30-mm polished KDP crystal in this new experimental configuration demonstrate that a more-precise measure of the Raman tensor can now be performed. This system was designed to help improve predictive capabilities in order to minimize TSRS-induced effects in large-aperture laser systems by enabling direct measurement to obtain information regarding the optimal crystal cut and crystal mounting configuration.

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