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SYMMETRY SELECTION RULES IN RAMAN HETERODYNE DETECTION OF NUCLEAR RESONANCE


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Abstract - A general discussion of symmetry-based selection rules for the Raman heterodyne detection of nuclear resonance is given. Detailed measurements for Pr³⁺ in a cubic host YAG confirm selection rule predictions in zero magnetic field. Significant departures are observed in the Earth's field which result from symmetry reduction and partial removal of site cancellation effects.

1 Introduction

Raman heterodyne detection [1] is one of several recent laser-based techniques for selective nuclear resonance measurements of optical impurities in solids. These involve resonant excitation either using simultaneous laser and radio-frequency fields [2] or laser radiation alone [3]. In this paper we give a general discussion of selection rules for the Raman heterodyne effect based on symmetry arguments. Using an adaptation of earlier techniques [1], we have made detailed experimental tests of the predictions for the dilute impurity Pr³⁺ in a cubic host, YAG. We find significant departures from predicted zero-field selection rules in the Earth's magnetic field (61 gauss) which are associated with partial removal, by the field, of site interference effects [4,5].

2 Symmetry effects

In the Raman heterodyne method [1], nuclear resonance induced by an applied radio frequency magnetic field, produces Raman side bands at the nuclear frequency on a c.w. laser beam transmitted through the sample (see Figure 1). The laser frequency must be resonant with a zero-phonon transition of the impurity under study, providing optical selection. The transmitted beam is incident on a photodiode and the Raman side bands are detected as a heterodyne beat signal in its output.

As first pointed out by Taylor [5], the Raman side bands are generated by a nonlinear response described by

\[ P_j(\omega) = x_{ijk} E_j(\omega) H_k(\omega_f) \]  

where \( \omega = \omega_l \pm \omega_f \) and \( i,j \) and \( k \) denote cartesian components. \( P_j \) is an optical polarisation generated by incident laser field \( E_j \) at frequency \( \omega_l \) and radio frequency
magnetic field $H_k$ at $\omega_{\text{rf}}$. The nonlinear susceptibility, $\chi$, is a property of the crystal. It is a third rank axial tensor and must be invariant under the point symmetry operations of the crystal. Consequently certain components $\chi_{ijk}$ may be zero and relations exist among the remainder. A full tabulation of $\chi_{ijk}$ for the 32 point groups is given by Birss [6]. In particular for spherical and cubic symmetry $\chi$ has a single non-zero component, $\alpha$, and Eq (1) becomes

$$P = \alpha E \times H$$

which is similar in form to an a.c. Faraday effect.

Heterodyne detection of the Raman side bands requires a 'local oscillator' optical field at the photodetector having the same polarisation. In many cases of low symmetry the side bands and incident laser do have components of the same polarisation ($i=j$ in Eq (1)) so that the latter may be used as the local oscillator giving a heterodyne signal proportional to $E_0 P_0$ (1). However for higher symmetries, particularly spherical and cubic (Eq (2)), the side band and laser polarisations are orthogonal ($i \neq j$). In these cases a suitable local oscillator field may be obtained for example using a linear polariser between the sample and the detector [5]. The heterodyne signal is then proportional to $E_0 P_0 d$ where $d$ denotes the polariser axis.

Figure 1: Experimental arrangement for Raman heterodyne detection of nuclear resonance in cubic crystals. Labels are explained in Section 3.

3 Measurements

Figure 1 indicates our experimental arrangement for investigation of Raman heterodyne nuclear resonance signals of $\text{Pr}^{3+}:\text{YAG}$ at 1.5K. The crystal symmetry is $O_h$ and there are six rare earth sites of $D_2$ symmetry per unit cell. The apparatus is similar to that used previously for studies of $\text{Pr}^{3+}$ in $\text{LaF}_3$ [1] and $\text{YAlO}_3$ [4] but has the important addition (as discussed above) of the polariser (P) between sample (C) and detector (D) to allow observation of signals from cubic crystals. About 20 mW of linearly polarised light from a C.W. dye laser of bandwidth <2 MHz (L) and tuned to the zero-phonon absorption line at 6069 Å was focussed along the (001) direction to a diameter of ~100 μm. A radio frequency magnetic field of amplitude ~0.1 to 1 gauss was applied either along (100) or (001) using a broad band travelling wave coil driven by amplified output from a swept frequency oscillator (O) and surrounding the sample immersed in superfluid helium. The heterodyne signal in the photodiode output was demodulated in a double balanced mixer (M) with reference derived from the oscillator. The radio frequency was repetitively swept through resonance at ~25 Hz and signals (S) were averaged over up to 1000 sweeps.
Figure 2: Energy levels of Pr$^{3+}$: YAG showing hyperfine splittings of lowest $^3H_4$ and $^1D_2$ crystal field components (3). The arrows indicate one set of possible optical and radio-frequency transitions for Raman heterodyne detection of \( \pm \frac{1}{2} \) to \( \pm \frac{3}{2} \) transitions in $^3H_4$ at 33.4 MHz. \( E, P \) and \( H \) refer to the fields in Eq (1).

Figure 3: Raman heterodyne resonance signals for Pr$^{3+}$: YAG for various combinations of polarisations of optical and radio-frequency fields. Traces a, c and e zero magnetic field; b, d and f in Earth's field. a and b, \( \chi \text{yyz} \); c and d, \( \chi \text{yyz} + \chi \text{xyz} \); e and f, \( \chi \text{xxx} + \chi \text{xyy} \).

The transition at 6069 Å is between the ground state component of $^3H_4$ and the lowest component of $^1D_2$. It has inhomogeneous width of several cm$^{-1}$ and homogeneous width ~16 kHz [3]. The low rare-earth site symmetry (D$_2$), gives electronically non-degenerate levels, each of which is split into three components by second-order magnetic hyperfine (pseudoquadrupole) interactions with the I=$\frac{3}{2}$ nucleus of $^{141}$Pr. These splittings, indicated in Figure 2, are typically 5-100 MHz [3].

We have made measurements of the ground state $I_\text{z} = \pm \frac{1}{2} \text{ to } \pm \frac{3}{2} \text{ to } \pm \frac{5}{2}$ transitions in crystals containing 0.5% and 5% Pr$^{3+}$. Figure 3 shows results of measurements on a 0.5% sample for different combinations of polarisations i, j and k in Eq (1). Traces a, c and e were recorded under conditions where care was taken to screen the sample from the ambient magnetic field in the laboratory using \( \mu \)-metal. The measured magnitude of this field was <1 gauss, essentially that of the Earth. Traces b, d and f show signals observed when the sample was exposed to this field. For a and b, the axis of the polariser was oriented parallel to the incident laser polarisation whereas in c to f it was set at 45° to it.

4 Discussion

It is seen from Figure 3 that in zero field \( \chi_{ijk} = 0 \) unless \( i \neq j \neq k \). Further in the case of trace c the phase of the signal was inverted upon reversal of the phase of the magnetic field, indicating that \( \chi_{ijk} = -\chi_{jik} \). In YAG the low rare earth site symmetry (D$_2$) imposes no restriction on allowed optical or radio frequency transitions among the levels. Therefore the measurements illustrated in Figure 3 verify in detail the selection rules (Eq (2)) based on the overall cubic symmetry.
The signals observed in the Earth's field arise from partial removal of site interference effects [4]. The nonlinear susceptibility $\chi$ is given by a sum of components from different rare earth sites ($S$) each of which is proportional to the product of matrix elements $\mu_{ab}$ for the individual resonant optical and radio frequency transitions

$$\chi_{ijk} = \sum_s \chi_{s,ijk} \sim \sum_s \mu_{s,12} \mu_{s,13} \mu_{s,23}$$

where 1 and 2 denote the two nuclear levels and 3 the excited optical level. Since the site symmetry ($D_2$) is lower than the overall crystal symmetry ($O_h$), it follows that many of the $\chi_{s,ijk}$ must be non zero although their sum $\chi_{ijk}$ is zero. Cancellations arise because the contribution from one site is sensitive to the phase of the matrix elements as well as to their magnitude (see Eq (3)). It may therefore cancel with the contribution from another site of the same local symmetry but related by an operation of the crystal point group. The cancellation may be lifted by application of a magnetic field which reduces the overall symmetry. Experimentally this appears either as separation of Zeeman components from one site, which produce signals of opposite signs (Zeeman interference) or as separation of cancelling signals from different sites by virtue of different g-factors (site interference). Thus the magnetic field required to lift the site or Zeeman cancellation is extremely small, being only of order the quasilent of Raman heterodyne line width and g-factor. Substantial signals may also be observed in even smaller fields due to partial removal of cancellation. For an arbitrary applied field orientation the overall symmetry becomes $C_i$ and all components of $\chi$ are non zero. This is consistent with observed signals for YAG in the Earth's magnetic field as illustrated in traces b,d and f of Figure 3.

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References

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