Selection Rules for an Electronic Raman Effect in a Situation where the Appropriate Point Group is a Double Group*

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The low-temperature Raman spectrum of the compound ytterbium gallium garnet is reported. The $^2F_{7/2}$ ground state of the ytterbium ion in this compound gives rise to four crystal-field components, all of them Kramers pairs. Raman scattering to two of these levels situated at 554 and 539 cm^{-1} above the ground state has been found. These levels belong to the Γ_5 species of the double group D_2 ,and in order to find the Raman selection rules, the transformation properties of both the symmetric and antisymmetric parts of the scattering tensor for the double group have to be considered. This is in contrast to the *first-order* Raman spectrum of the garnet host lattice, where all phonon modes have to be labeled in the factor group $O_h(10)$ and where the scattering tensor is *symmetric*.

RESULTS AND DISCUSSION

THE electronic Raman spectrum of the compound ytterbium gallium garnet (YbGaG) has some interesting properties which are not encountered in the vibrational spectrum associated with the phonon modes of this crystal. The electronic Raman transition here originates in the lowest-lying Stark component of the crystal-field-split manifold of the trivalent rare-earth ion, and terminates on the three other Stark levels of this manifold, which are anticipated to have an energy less than 1000 cm^{-1} above the ground state.[1–5] On the other hand, we have the phonon spectrum of this crystal, which is associated with the 240° of freedom of the 80 atoms which occupy the primitive cell.[6] All these degrees of freedom can be labeled according to the irreducible representations of the factor group $O_h(10)$ of the garnets, and the result is that the phonon Raman spectrum should give rise to only seven E_g modes, 11 T_{2g} modes, and three A_{1g} modes.

The intensities of Raman spectra are related to components of the Raman scattering tensor, and the polarization spectra can thus in principle be assigned by relating the laboratory axes to the macroscopic axis of the crystal. In Fig. 1, part of the laser-excited low-temperature Raman spectra of the compound YbGaG is shown, together with the spectrum in the same wave-number interval of thulium gallium garnet. The orientation of the crystals was such that the direction of propagation of the incident He–Ne laser beam was approximately perpendicular to [110] and parallel to [111]. An analysis[6] of the scattering tensor for the A_{1g}, E_g, and T_{2g} vibrational modes reveals that a deviation of the incident radiation from a direction which is exactly parallel to (say) the z axis of the crystal results in experimental tensors which are different from the above-mentioned tensors. The T_{2g} modes, which were originally associated with the tensor components $\alpha_{xz}+\alpha_{zx}$, $\alpha_{zy}+\alpha_{yz}$ and $\alpha_{xy}+\alpha_{yx}$, will now in the experimental case have, besides these nondiagonal components, components on the diagonal of the matrix as well. On the other hand, the E_g modes, which are related to the components $\alpha_{xx}-\alpha_{yy}$ and $2\alpha_{zz}-\alpha_{xx}-\alpha_{yy}$, are now characterized by a scattering matrix which has nondiagonal as well as diagonal elements. The A_{1g} modes,

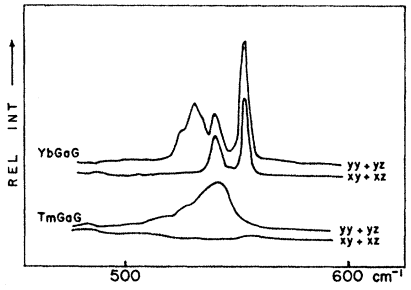

FIG. 1. Part of the 77°K Raman spectrum of TmGaG and YbGaG excited with the 6328-Å radiation of a He–Ne gas laser. The labeling xz, etc., refers to laboratory axis. The Raman spectrum was detected with a noncooled 9558 B.E.M.I. phototube. Only a small part of the photocathode was used; the photoelectrons of the nonilluminated part of the cathode were prevented from reaching the first dynode by a magnetic defocusing field.

however, are not affected, and the scattering tensor for these modes remains a pure case of trace scattering only.

The rather broad and asymmetric phonon band of TmGaG centered at ~542 cm^{-1} has vanishing intensity if the experimental scattering-tensor elements α_{xy} and α_{xz} are recorded with a suitable direction of propagation and polarization of the laser beam and direction of the scattered radiation. The components α_{yy} and α_{yz} were also recorded by placing an analyzer in front of the slit of the monochromator, and it was found that $\alpha_{yz}=0$. It is clear from the above discussion that this phonon mode is of species A_{1g}.

The irradiation condition of the YbGaG was approximately the same as that of TmGaG, and we note a

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[1] D. L. Wood, J. Chem. Phys. **39**, 1671 (1963).
[2] R. Pappalardo and D. L. Wood, J. Chem. Phys. **33**, 1734 (1965).
[3] M. T. Hutchings and W. P. Wolfe, J. Chem. Phys. **41**, 617 (1964).
[4] J. A. Koningstein, Theoret. Chim. Acta **3**, 271 (1965).
[5] R. A. Buchanan *et al.*, Phys. Rev. **159**, 245 (1967).
[6] J. A. Koningstein and O. Sonnich Mortensen, J. Mol. Spectry. (to be published).

TABLE I. Activity table for the double group D_2.

D_2	E	$\bar{E}$	C_2 C_2	C_2' C_2'	C_2'' C_2''	Transformation properties of scattering tensor	${}_4F_{7/2}$ levels
Γ_1	1	1	1	1	1	α_{xx}, α_{yy}, α_{zz}	
Γ_2	1	1	−1	1	−1	$\alpha_{xz}\pm\alpha_{zx}$	
Γ_3	1	1	1	−1	−1	$\alpha_{yz}\pm\alpha_{zy}$	
Γ_4	1	1	−1	−1	1	$\alpha_{xy}\pm\alpha_{yx}$	
Γ_5	2	−2	0	0	0		All levels

shift of the A_{1g} mode towards lower energy. The Raman band is now peaked at 530 cm^{-1}, and a weak shoulder occurs at 523 cm^{-1}. Also present in the spectrum are two Raman lines whose intensity is dependent on the concentration of the Yb ion. We associate these bands with electronic Raman transitions of the Yb^{3+} ion.[7]

The scattering tensor for the electronic Raman transition is different from that for the vibrational transitions. First, the scattering tensor also contains antisymmetric combinations such as α_{xy} α_{yx}; and second, the interaction of the rare-earth ions is much weaker, and a "factor" group analysis does not have to be carried out. Instead, we can label the electronic levels according to the species of the D_2 point group which describes the symmetry of the environment of the lanthanide ion in the garnet crystal.[8] The electronic levels are, however, Kramers doublets, because they are the crystal-field components of a $J=\frac{7}{2}$ Russell-Saunders manifold; and in order to predict the scattering tensor the D_2 double group has to be employed. The character table and transformation properties of the components of the scattering tensor and the labeling of the electronic energy levels are shown in Table I. All electronic levels belong to the irreducible representation Γ_5, and an electronic Raman transition is allowed if in the representation product $\Gamma_5\times\Gamma_5$ a species occurs to which a component of the scattering tensor also belongs. Now $\Gamma_5\times\Gamma_5=\Gamma_1+\Gamma_2+\Gamma_3+\Gamma_4$, and consequently the Raman scattering tensor of all electronic transitions contains diagonal as well as off-diagonal elements. This is in perfect agreement with experiment. Separation of $\alpha_{yy}+\alpha_{yz}$ and $\alpha_{xy}+\alpha_{xz}$ into their individual components for the two electronic Raman transitions of Yb^+ in YbGaG reveals that of the various scattering components, $\alpha_{yy}\neq\alpha_{yz}\neq\alpha_{xy}\neq\alpha_{xz}$, and none is equal to zero.

[7] J. A. Koningstein, J. Chem. Phys. **46**, 2811 (1967).

[8] R. Pappalardo, Nuovo Cimento **26**, 748 (1962).

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Coupled Motion of Vortices in Superposed Superconducting Films*

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The coupling of vortices between two superconducting tin films electrically insulated from one another (i.e., in a dc transformer) has been studied as a function of the applied perpendicular magnetic field, of the current in the primary, and of the temperature. The results are qualitatively interpreted in terms of a vortex-slipping mechanism, and they support the picture of vortex flow as the dissipation process in superconducting films.

INTRODUCTION

IN 1965, Giaever showed[1] that it was possible to couple the motion of vortices present in two superposed superconducting films, electrically insulated from one another. This phenomen was detected by measuring the voltage across the secondary film, in which no external current was passing, induced by the motion of the vortices due to the passage of a dc current in the other film (the primary). Since then various authors[2–4] have investigated these dc transformers, driving currents simultaneously through the primary and secondary films, varying the thickness of the primary, and studying the influence of an external magnetic field or the impurity concentration on the coupling between the vortices in the two films. Similar experiments were

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[1] I. Giaever, Phys. Rev. Letters **15**, 825 (1965).

[2] I. Giaever, Phys. Rev. Letters **16**, 460 (1966).

[3] M. D. Sherrill, Phys. Letters **24A**, 312 (1967).

[4] P. E. Cladis and R. D. Parks, Bull. Am. Phys. Soc. **12**, 395 (1967).