ELECTRONIC RAMAN EFFECT IN Pr^{3+} IONS IN SINGLE CRYSTALS OF $PrCl_3$

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The electronic energy levels of rare earth ions in crystals have been extensively studied using both absorption and fluorescence techniques. Additional useful information about these energy levels should be provided by Raman studies, because of the essentially different selection rules involved. In this laboratory we have succeeded in obtaining an electronic Raman effect in Pr^{3+} in pure $PrCl_3$. While preparing the work for publication, a letter by Elliott and Loudon[1] appeared suggesting the possibility of observing just such an effect. It therefore seemed appropriate to communicate our main results at this time.

The Pr^{3+} ion ($4f^2$) was chosen for study because the energy levels of this ion are very well established. Also, $PrCl_3$ crystals do not absorb the Hg 2537Å radiation, which has many technical advantages for the excitation of Raman spectra. For purposes of comparison, the spectrum of $LaCl_3$ crystals was also investigated: the La^{3+} ion (no $4f$ electrons) has no electronic states lower than 50 000 cm^{-1}, and also the isomorphic $PrCl_3$ and $LaCl_3$ crystals would be expected to have similar vibrational spectra. Polarization studies of the light scattered by single crystals of $PrCl_3$ and $LaCl_3$ were carried out at 300°K and 77°K. The Raman spectra were photographed with a 21-foot grating spectrograph (dispersion 0.8 Å/mm). Scattered 2537Å radiation was reabsorbed by mercury vapor before entering the spectrograph in order to reduce the background of stray light. The optical path length of the crystals was 0.5 cm, and exposure times were in the range of 4 to 20 hours.

At room temperature five Stokes and five anti-Stokes Raman lines with frequency shifts less than 250 cm^{-1} were observed. They were assigned to five (one A_g, one E_{1g}, three E_{2g}) of the six Raman-active vibrational modes of the $PrCl_3$ lattice. An analogous set of vibrational lines was observed in $LaCl_3$. No electronic Raman transitions were observed at room temperature, which is consistent with the known fact that the electronic energy levels of the rare earth ions are very broad at 300°K. At 77°K, however, additional lines appeared in the spectrum of $PrCl_3$ (but not in $LaCl_3$) which must be attributed to electronic Raman transitions. In all, ten electronic Raman lines were observed. These have

been assigned to transitions from the lowest Stark level of the $^3H_4$ state of the Pr^{3+} ion to (i) three other Stark levels of the $^3H_4$ state; (ii) five Stark levels of the $^3H_5$ state; and (iii) two Stark levels of the $^3F_2$ state. The frequencies and polarizations of these lines are given in Table I. The expected frequencies based on absorption and fluorescence studies are given for comparison. The agreement between the two sets of frequency data is good. Furthermore, the polarizations of the Raman lines are consistent with those calculated from the known crystal quantum numbers (μ) of the levels.

All of the observed electronic Raman lines have a width of several cm^{-1}, presumably because the undiluted salt was used. The lines corresponding to transitions from the lowest Stark level to the other Stark levels of the $^3H_4$ state are more intense than those corresponding to transitions to the Stark levels of the $^3F_2$ state. These latter, in turn, are more intense than lines corresponding to transitions to the Stark levels of the $^3H_5$ state. The intensities of the strongest electronic Raman lines of Pr^{3+} are comparable to the intensities of the strongest vibrational lines of the $PrCl_3$ crystal.

From our work it is clear that electronic Raman spectra can be obtained with dilute crystals.

Table I. Observed electronic Raman frequencies (cm^{-1}) in $PrCl_3$ at 77°K.

State	μ	Raman[a]	Absorption and fluorescence[b]	Polarization obs	Polarization cal
$^3H_4$	3	32	33.0	$\sigma\pi$	$\sigma\pi$
	2	100	96.0	σ	c
	1	139	133.0	$\sigma\pi$	$\sigma\pi$
$^3H_5$	3	2136	2134.5	$\sigma\pi$	$\sigma\pi$
	2	2169	2169.1	$\sigma\pi$	c
	1	2190	2187.7	$\sigma\pi$	$\sigma\pi$
	3	2204	2201.7	$\sigma\pi$	$\sigma\pi$
	2	2227	2222.0	$\sigma\pi$	c
$^3F_2$	1	4918	4922.1	$\sigma\pi$	$\sigma\pi$
	2	4949	4950.3	σ	c

[a]This work.

[b]From references 2 and 3.

[c]These transitions can have any polarization whatever.

If the lines became narrower by a factor of ten (i.e., if the width was reduced from several cm^{-1} to several tenths of a cm^{-1}), as might reasonably be expected in a 10% solution of $PrCl_3$ in $LaCl_3$, then no increase in path length or exposure time would be necessary to obtain lines of the same strength on the plate as those obtained by us for the pure material.

We wish to thank Dr. B. P. Stoicheff for his help throughout the course of this work.

LIFETIME AND BEAM SIZE IN A STORAGE RING

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We want to report on measurements of the lifetime of stored beams carried out with the Frascati e^+e^- storage ring (AdA) at the Laboratoire de l'Accelerateur Lineaire of the Science Faculty at Orsay. The design parameters of the ring have already been published[1] as well as preliminary results about operation at a low stored intensity.[2] The use of the Orsay linac as injector allowed the storage of a big enough number of electrons to observe the following effects.

With a small number of stored electrons (less than 10 000) the lifetime is of the order of 50 hours corresponding to a residual (air) pressure of ~5 $\times 10^{-10}$ Torr. The vacuometer reading was 3.5 $\times 10^{-10}$ Torr.

The lifetime τ depends on the number N of particles in the beam. It can be fitted by

$$1/\tau = \alpha(E)N + 1/\tau_0, \tag{1}$$

where $\alpha(E)$ is a strongly energy-dependent parameter. A best fit illustrating (1) is shown in Fig. 1.

The energy dependence of $\alpha(E)$ is shown in Fig. 2. In the interval $100 < E < 207$ MeV this curve can be fitted by

$$\alpha(E)E^{5.5} = \text{const}. \tag{2}$$

$\alpha(E)$ has a maximum at about 70 MeV. Below this energy the lifetimes increase very rapidly. Lifetime measurements below 50 MeV were not carried out because of the difficulties in revealing the synchrotron radiation at such low energies.

The lifetime of a given beam is independent of the presence of the other, so that the effect described must be interpreted as a "self-interaction" of the particles in each bunch.

A theoretical explanation of the effect can be given in the following manner. We observe that in a "natural beam," i.e., in a beam the dimensions of which are defined solely by the fluctuations of the synchrotron radiation, the momentum distribution is subject to the inequality

$$\delta q \gg \delta q_z \approx \delta q_e. \tag{3}$$

(The latter equality is one of order of magnitude.) Here δq is the rms of the radial momentum, δq_z the rms of the vertical momentum, and δq_e the rms of the longitudinal momentum, the latter being measured in the system in which the bunch is at rest. The inequality (3) holds above a certain critical energy (of the order of about 30 MeV in AdA). The Møller scattering between two elec-