

hadrons (including neutrals) and 6 hadrons, in increasing order of steepness of the plot. On the basis of the semi-inclusive data and an independent-cluster-emission model, the larger estimate (5–6 hadrons per cluster) is strongly favored by the fluctuation analysis.³ However, we find inadequate sensitivity of the gap distribution to distinguish these two cases on the basis of kinematical configuration alone.

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Temperature Dependence of Fluorescence Linewidths in PrF_3 : A Comment on "Excited-State Exchange Broadening of Optical Transitions in PrCl_3 "

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Data are reported which show that the temperature dependence of fluorescence linewidths in PrF_3 are qualitatively like those in $\text{LaF}_3:\text{Pr}^{3+}$. This result is interpreted as supporting the explanation of German and Kiel for the anomalous temperature dependence of fluorescence linewidths in PrCl_3 .

Recently German and Kiel¹ have measured the temperature dependence of the fluorescence linewidths of the $^3P_0 \rightarrow ^3F_2$ and $^3P_0 \rightarrow ^3H_6$ transitions of the Pr^{3+} ion in PrCl_3 . They found an anomalous broadening with decreasing temperature of two of the fluorescence lines. The effect was observed only when the terminal state was a magnetic doublet and only at temperatures below 100 K. It was not observed for the same lines in $\text{LaCl}_3:\text{Pr}^{3+}$. German and Kiel ascribed the effect to a small temperature-dependent exchange splitting of the terminal state due to exchange interactions with neighboring ions in their ground states. The temperature dependence was due to the combined effects of differing interaction strengths between the terminal state and the various levels of the ground 3H_4 manifold, and the changing thermal populations of the various levels of the ground 3H_4 manifold.

This Comment reports measurements of the temperature dependence of the fluorescence linewidths in PrF_3 . These measurements were made to test the explanation advanced by German and Kiel for their observations. Pr^{3+} has an even number of electrons so that Kramers's theorem does not apply and all the degeneracy of the ion's energy levels may be lifted in an environment of sufficiently low symmetry. For PrCl_3 the site

symmetry² is sufficiently high (C_{3h}) that some doublets remain and these states have a magnetic moment. However for PrF_3 the site symme-

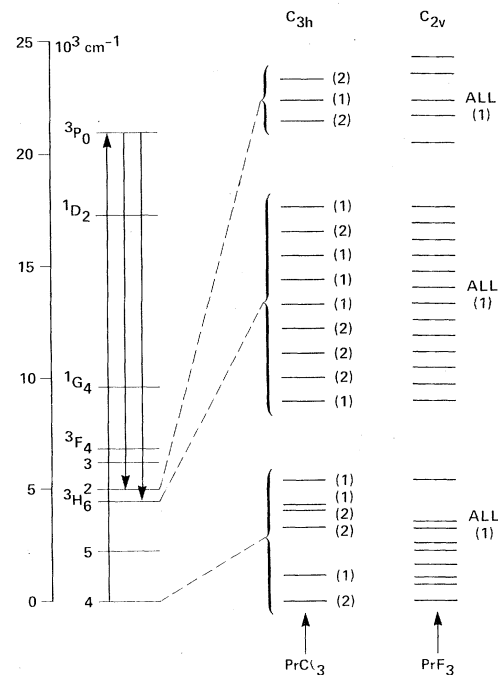


FIG. 1. Pr^{3+} energy levels— PrCl_3 and PrF_3 .

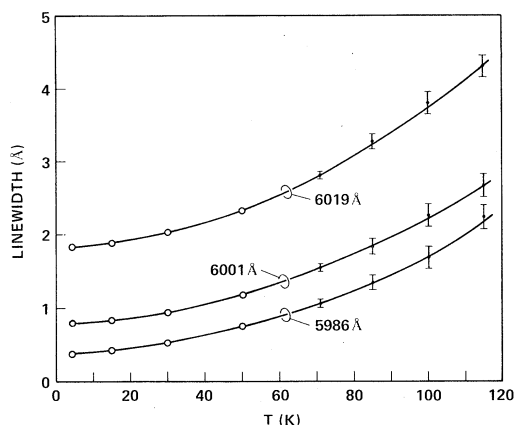


FIG. 2. ${}^3P_0 \rightarrow {}^3H_6$ fluorescence linewidth temperature dependence— PrF_3 .

try² is lower (C_2) so that there are only singlets with no magnetic moment. Therefore in PrF_3 there should be no exchange interactions between Pr^{3+} ions, and the anomalous broadening should not be observed as it is in PrCl_3 if the exchange-splitting mechanism advanced by German and Kiel is correct. Figure 1 shows the pertinent energy levels for the two compounds.

To the author's knowledge fluorescence measurements for PrF_3 have not previously been reported, although the Pr^{3+} energy levels have been studied by other means.³⁻⁸ The fluorescence was excited by the 476-nm line of a krypton laser, which leads to population of the 3P_0 level of the Pr^{3+} ions. The intensity of the fluorescence decreased rapidly with increasing temperature and was difficult to detect above 140 K. Figures 2 and 3 show the observed temperature dependence of the fluorescence linewidths of several of the ${}^3P_0 \rightarrow {}^3F_2$ and ${}^3P_0 \rightarrow {}^3H_6$ transitions. Qualitatively these results greatly resemble what is observed⁹ for $\text{LaF}_3:\text{Pr}^{3+}$, and it is evident that the anomalous behavior of PrCl_3 does not occur in PrF_3 . As explained above this result supports the hypothesis of German and Kiel regarding PrCl_3 .

German and Kiel made the point that the small exchange splittings were observed at temperatures many times that of any ordering temperature, since PrCl_3 is known not to be magnetically ordered down to 0.8 K, but they gave no further discussion. Several years ago Leask¹⁰ reviewed the temperature dependence of exchange splittings in a number of magnetic rare-earth compounds. He presented evidence that when exchange split-

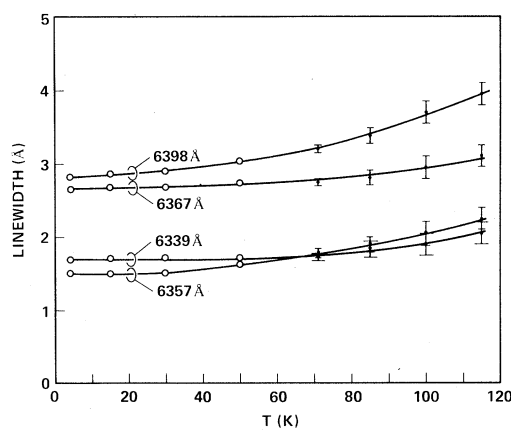


FIG. 3. ${}^3P_0 \rightarrow {}^3F_2$ fluorescence linewidth temperature dependence— PrF_3 .

tings persist at temperatures far above the ordering temperature they are due largely to short-range inter-ion interactions, such as exchange interactions, and that when exchange splittings fall to zero rapidly as the temperature is increased above the ordering temperature they are due largely to long-range inter-ion interactions such as dipolar interactions. German and Kiel estimate that dipolar interactions are negligible for PrCl_3 , so that the behavior of PrCl_3 fits the pattern described by Leask.

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