

Anderson localization in ruby

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The localization of the wave functions of the fluorescing levels in ruby and other exchange-coupled systems is discussed. It is pointed out that when the range of the interaction is less than the minimum distance between ions, as happens in ruby, the Anderson transition occurs near or above the nearest-neighbor critical percolation concentration. The role of off-diagonal dipolar interactions in establishing extended states is assessed. Dipolar effects are unimportant when the homogeneous linewidth exceeds the dipolar coupling between neighboring spins.

The purpose of this paper is to comment on the localization of the wave functions of the fluorescing states in ruby and other optically active systems with exchange-coupled ions. In an early theoretical investigation Lyo¹ concluded that the extended states associated with the ²E levels of ruby would disappear at a critical concentration of 0.3–0.4 at. % Cr³⁺. Following his prediction, experimental evidence of a mobility edge in this concentration range was reported by Koo, Walker, and Geschwind.² However, subsequent measurements indicated that there were no extended states in the region where Lyo predicted the Anderson transition would occur. Recently, in an experimental study similar to that of Ref. 2 but carried out in much greater detail, Chu *et al.* found no compelling evidence of a mobility edge.³ At present there appears to be a general consensus that the ²E states are localized for Cr concentrations below 1 at. %.

In the theory outlined in Ref. 1 the localization arose from the strain broadening associated with a random distribution of single-ion levels (diagonal disorder). The criterion for localization was given approximately as

$$\frac{\Delta W}{V(\bar{r}_{NN})} \approx 3-5 \quad (1)$$

where ΔW is the width of the inhomogeneous line and $V(\bar{r}_{NN})$ is the off-diagonal exchange interaction evaluated at the average nearest-neighbor separation \bar{r}_{NN} . The latter parameter is approximately equal to $0.6n^{-1/3}$, n being the concentration.

Recently it has been shown that Anderson locali-

zation can occur in dilute systems even in the absence of diagonal disorder, provided the off-diagonal interactions are short ranged (e.g., nearest neighbor or exponential) and have a (predominantly) common sign.⁴⁻⁷ In such systems the Anderson transition is driven by fluctuations in the configuration of near neighbors. The analysis of Ref. 6 shows that in dilute three-dimensional arrays with nearest-neighbor interactions all of the states become localized before the critical percolation concentration is reached. In the case of exponential interactions, i.e., $V_0 \exp(-\alpha r_{ij})$, the numerical studies of Ref. 7 indicated that Anderson localization occurred at a critical concentration n_c such that

$$\frac{\alpha}{n_c^{1/3}} \approx 2.7 \quad (2)$$

provided \bar{r}_{NN} was much greater than the lattice constant. Using a value of α appropriate to ruby, 1 \AA^{-1} (Ref. 2) $\alpha/n^{1/3}$ was found to be on the order of 17 for 0.4 at. % Cr. Since this value was greatly in excess of 2.7 it was concluded that the Anderson transition occurs at a much higher Cr concentration than predicted in Ref. 1.

Since (2) applies only to situations where the critical value of \bar{r}_{NN} is much greater than the lattice constant, it can not be used to give a quantitative estimate of n_c in ruby. In order to gain some insight into the problem, we have carried out a series of calculations analogous to the ones outlined in Ref. 7 but which utilized values of the exchange similar to those given in Ref. 1 along with a random distribution of site energies which had a

width comparable to the inhomogeneous line width. Varying n we found a mobility edge when the Cr concentration was equal to 10 at. %. Although a lack of detailed knowledge about the diagonal and off-diagonal exchange in ruby makes quantitative comparisons not very meaningful, we can conclude from our analysis along with those of Refs. 6 and 7 that the Anderson transition in dilute exchange-coupled systems like ruby, where the range of the interaction is less than the minimum nearest-neighbor distance ($r_{\text{NN}} = 2.7 \text{ \AA}$ in ruby), occurs for concentrations comparable to or greater than the nearest-neighbor critical percolation concentration. We emphasize that the transition will occur even in the absence of strain broadening. In the presence of diagonal disorder the transition is likely to occur at a higher concentration since Anderson localization can take place in a periodic array if ΔW is sufficiently large.

Our second point pertains to the role of dipolar interactions. In his paper on localization in periodic arrays with diagonal disorder Anderson⁸ pointed out that localization would not occur unless the off-diagonal interaction fell off more rapidly than r_{ij}^{-3} . More recently Elyutin⁹ concluded there would be no localized states in a *dilute* system with dipolar off-diagonal interactions. In view of the latter result it would appear that all states would be extended in systems where there are off-diagonal dipole moments (e.g., ruby).

We argue that this is not always the case. Because of thermal disorder, states which would have been extended as a consequence of the dipolar coupling become localized at experimentally accessible temperatures. The point here is that the long-range polarization fields arising from the interactions between the dipoles are weakened by the fluctuations in the relative phases of the moments on the various sites. This dephasing, which comes from fluctuations in the hyperfine fields as well as from direct and Raman processes involving the phonons, is the cause of the homogeneous line

width. Since in random systems the inverse homogeneous line width T_2 , is a measure of the single-ion coherence time we can use $d^2 n T_2 \hbar^{-1}$, where d is the off-diagonal dipole moment, as a measure of the relative importance of dipolar interactions. When $d^2 n T_2 \hbar^{-1} \gg 1$ the thermal disorder is insufficient to destroy the coherence of the dipolar fields, whereas when $d^2 n T_2 \hbar^{-1} \ll 1$ dipolar interactions have a negligible effect.

In the case of ruby we can infer a value of d from the radiative lifetime of 3 ms. We obtain $d^2 n \approx 10 \text{ MHz}$ for 0.4 at. % Cr, which is near the lower limit of the 5–100-MHz homogeneous line width observed experimentally at liquid-helium temperatures.^{10,11} Thus it is possible that the dipolar interactions can give rise to extended states at low temperatures. However, since the homogeneous line width is a rapidly increasing function of temperature,¹² the dipolar forces can probably be neglected when $T \gtrsim 5 \text{ K}$. We note that in systems with weak ion-ion interactions \hbar/T_2 will exceed the maximum inter-ion interaction at high temperatures. When this happens the states become localized entirely on individual ions in the sense that energy transfer between corresponding levels of adjacent ions takes place incoherently.

In conclusion we would like to suggest that the best candidates for observing the Anderson localization of Frenkel excitons are likely to be exchange-coupled systems where the concentration of optically active ions can be varied continuously between 0% and 100%. In addition, to sort out the relative effects of diagonal and off-diagonal disorder it is important that the substitution of inactive ions have a minimal effect on the inhomogeneous linewidth. Moreover, in order to suppress the effects of the dipolar interaction the temperature must be high enough so that $d^2 n T_2 \hbar^{-1} < 1$.

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