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Search for the Anderson transition in ruby

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A new diagnostic to detect the occurrence of an Anderson transition, using photon echoes, is proposed and experimentally tested for the R_1 line in ruby. A search for mobility edges in samples up to 0.1 wt.% Cr_2O_3 concentration was unsuccessful.

I. INTRODUCTION

The concept of the Anderson transition (AT) and mobility edges associated with inhomogeneously broadened optical transitions in crystals[1,2] is, at present, a highly controversial topic both experimentally and theoretically. In inorganic systems, observation of an AT has been reported for two systems[3,4]; however, further work[5] with one of them (ruby) found no compelling evidence for a mobility edge. Controversy also exists for organic systems with Ahlgren and Kopelman[6] favoring a percolation model to explain their observed critical concentration behavior of energy transfer versus the AT model advanced by Klafter and Jortner,[7] along with the experimental support reported by Smith *et al.*[8]

So far, the only diagnostic used in attempts to observe the AT in optical solids has been based on energy-transfer effects on trap excitation or grating decay.[9] The expectation is that the transfer properties and, hence, trap excitation or grating decay will undergo a sudden change at the AT. Since the interpretation of energy-transfer behavior for AT diagnostics seems to be in doubt, we propose and describe new experiments which attempt to identify the AT using another technique.

Specifically, we consider the possibility that a change in homogeneous linewidth or coherence time T_2' will occur as the system switches from a localized to an extended state. Now, in the original idealized problem considered by Anderson,[10] zero bath temperature was assumed and, hence, as argued by Holstein *et al.*,[2] no change in T_2' is expected for an AT. On the other hand, for real systems at finite temperature this may no longer be true. We suggest that T_2' might be different for a spatially extended state, which can be scattered by defects, compared to a localized state. Indeed, recent photon-echo experiments[11] with a 100% stoichiometric system are consistent with this idea in that a coherence time, an order of magnitude shorter, was observed compared with a weakly doped (localized) crystal.

II. EXPERIMENTAL

Since coherent transient methods provide about two-orders-of-magnitude-higher-resolution capability[12] (<10 kHz) than frequency domain methods,[13,14] we used photon echoes to sensitively probe for possible small changes in T_2' associated with an AT. Our S/N was such that a change in homogeneous linewidth of 20 kHz could have been detected out of a total of 660 kHz for 0.1% ruby.

A new photon-echo technique, shown in Fig. 1, was used. A single frequency (1-MHz linewidth) scanning cw dye laser is tuned to the Cr^{3+} ${}^4A_2(-\frac{1}{2}) \rightarrow \bar{E}(-\frac{1}{2})$ $[R_1(-\frac{1}{2})]$ transition at 693.4 nm. The laser power into the 1-mm-thick sample is 30 mW, focused to a diameter of ~0.05 mm. The laser beam is gated by an acousto-optic modulator

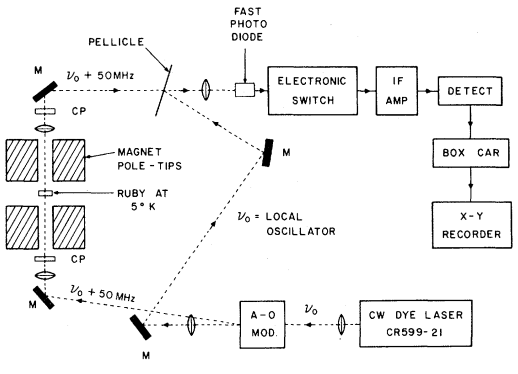

FIG. 1. Schematic of experimental apparatus used for photon-echo studies in ruby with heterodyne detection.

(AOM) to provide two pulses, each 200 nsec long. The undeflected zero-order beam bypasses the sample and a portion of it is wave-front matched to the echo signal to allow heterodyne detection. The beat frequency is 50 MHz, which is the AOM operation frequency. This technique has a number of advantages over methods which use (a) Stark shifting,[15] (b) nonseparated signal and local oscillator beams with AOM shifting,[12,16] or (c) direct detection.[17] These are as follows: (1) No coating is required on the sample as it is for Stark shifting. (2) Optical pumping of the ground-state levels (and consequent reduction of signal,[12]) due to the local oscillator, is eliminated. (3) No spurious power-dependent local oscillator effects can occur.[16] (4) The large beat frequency of 50 MHz allows observation of shorter decays versus method (b), which is limited to beat frequencies <5 MHz because of wave-front matching considerations. (5) No expensive and slow repetition rate optical switch is required as in (c); the switching is now in the electronics. There is one disadvantage to the method which may markedly reduce the sensitivity. This is that the optical wave front, after passing through the cryostat windows and sample, must not be excessively deformed compared to the reference beam. Also, fluctuations due to turbulence in air paths and in the cryostat can be of concern, since what Fig. 1 depicts is, in essence, a Mach-Zehnder interferometer.

Experimental scans showing the dependence of photon echo amplitude (intensity)$^{1/2}$ on position within the $R_1(-\frac{1}{2})$ inhomogeneously broadened line are shown in Fig. 2. Circularly polarized light was used to allow discrimination from the nearby $R_1(+\frac{1}{2})$ line. No sharp breaks in the photon-echo

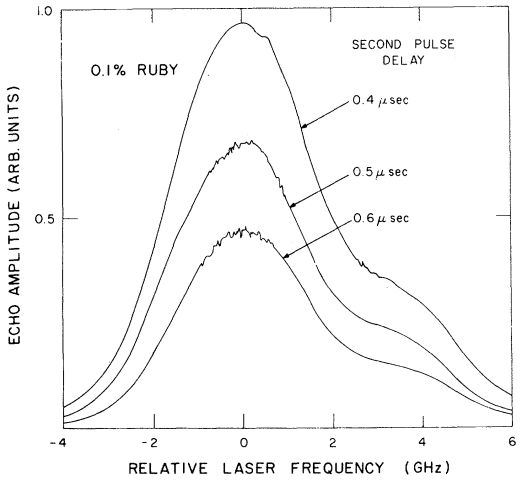

FIG. 2. Photon-echo signal in ruby vs laser tuning over the inhomogeneous ${}^4A_2(-\frac{1}{2}) \rightarrow \bar{E}(-\frac{1}{2})$ line in a field of 1 kG along the c_3 axis with the pulse separation as parameter. The sample temperature is ~ 5 K.

signals corresponding to the suggestion of a mobility edge were observed. The feature on the high-frequency side of the line is due to the ^{53}Cr isotope. Analysis of the data shows that for frequencies up to 4 GHz on either side of line center, T_2' was independent of frequency, 480 ± 40 nsec for 0.1% ruby and 2000 ± 200 nsec for 0.03%. These values are in good agreement with previous measurements.[15,18]

III. DISCUSSION

Although our concentration range is lower than that studied by Koo, Walker, and Geschwind (KWG),[4] our inhomogeneous linewidths (2–5 GHz) are smaller than theirs (24–78 GHz) so that we are covering a similar spectral density range, and therefore the results should be comparable. We estimate from our S/N that a break of 5% in echo amplitude could have been seen at the spectral density where a break in the transfer rate was seen by KWG. The echo amplitude equation $A = A_0 \exp(-2t_p/T_2')$, where t_p is the pulse separation, gives the linewidth sensitivity of 20 kHz mentioned earlier. The conclusion is that the change in linewidth due to an AT is less than 20 kHz or, alternatively, the "breaks" seen by KWG are not due to an AT. In view of recent energy-transfer studies,[19] the latter possibility seems more likely.

An important factor which needs further consideration in AT theoretical studies is the effect of homogeneous broadening. Criticism of Anderson's theory regarding the neglect of this factor has been made by Ziman.[20] Anderson's opinion[21] is that inclusion of homogeneous broadening would modify but not destroy the AT. However, if the homogeneous linewidth is comparable to the interaction V responsible for the AT (e.g., phonon broadening at higher temperatures), then it is reasonable to expect that an AT will not occur.[22] For ruby, a fundamental contribution to Cr^{3+} homogeneous broadening arises from the Cr-Al interaction via Al-Al nuclear-spin flips,[23] and it is of interest to compare this with V.

The range dependence of the exchange interaction used by Lyo is[1]

$$V = V_0 \exp(-0.724r) ,$$

where $V_0 = 111$ cm^{-1} and the ion spacing r is in Å. To estimate V, we use the ensemble average nearest-neighbor spacing[1] $r = (0.17/n)^{1/3}$, where n is the Cr density. For our 0.1% sample this gives $V \sim 10$ MHz. This may be compared with the measured Cr-Al homogeneous width of 65 MHz from hole-burning studies[14] in zero field (dilute sample) and <90 kHz for a field of 1 kG along the c axis as measured by photon echoes.[24] Since the experiments were performed in a magnetic field of 1 kG, the conclusion is that the Cr-Al homogeneous width was not a significant factor in the present studies. It is ap-

parent, however, that a magnetic field may be necessary in AT studies of ruby to reduce the Cr-Al homogeneous linewidth relative to the exchange interaction. An extension of current work to higher concentrations seems desirable, especially in view of a recent upward estimate by Huber[25] of the critical concentration to ~10% from the value of ~0.3% estimated earlier by Lyo.[1] However, complications will arise because of the rapidly decreasing coherence times due to Cr-Cr spin flipping.[26]

In conclusion, an attempt to observe an Anderson transition mobility edge in ruby using a photon-echo measure of homogeneous linewidth has been unsuccessful. While this by itself is not conclusive, recent energy-transfer studies[19] also argue against the occurrence of an AT in ruby for concentrations $\lesssim 1\%$.

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[1]S. K. Lyo, Phys. Rev. B 3, 3331 (1971).
[2]For a recent review, see T. Holstein, S. K. Lyo, and R. Orbach, Comments Solid State Phys. 8, 119 (1978).
[3]C. Hsu and R. C. Powell, Phys. Rev. Lett. 35, 734 (1975).
[4]J. Koo, L. R. Walker, and S. Geschwind, Phys. Rev. Lett. 35, 1669 (1975).
[5]S. Chu, H. M. Gibbs, S. L. McCall, and A. Passner, J. Opt. Soc. Am. 70, 633 (1980).
[6]See D. C. Ahlgren and R. Kopelman, J. Chem. Phys. 73, 1005 (1980), and references therein.
[7]See J. Klafter and J. Jortner, J. Chem. Phys. 75, 1004 (1980), and references therein.
[8]D. D. Smith, R. D. Mead, and A. H. Zewail, Chem. Phys. Lett. 50, 358 (1977).
[9]P. F. Liao, L. M. Humphrey, D. M. Bloom, and S. Geschwind, Phys. Rev. B 20, 4145 (1979).
[10]P. W. Anderson, Phys. Rev. 109, 1492 (1958).
[11]R. M. Shelby and R. M. Macfarlane, Phys. Rev. Lett. 45, 1098 (1980).
[12]R. G. De Voe, A. Szabo, S. C. Rand, and R. G. Brewer, Phys. Rev. Lett. 42, 1560 (1979).
[13]A. Szabo, Phys. Rev. Lett. 27, 323 (1971).
[14]P. E. Jessop, T. Muramoto, and A. Szabo, Phys. Rev. B 21, 926 (1980).
[15]A. Szabo and M. Kroll, Opt. Lett. 2, 10 (1978).
[16]R. M. Macfarlane, R. M. Shelby, and R. L. Shoemaker, Phys. Rev. Lett. 43, 1726 (1979).
[17]I. D. Abella, N. A. Kurnit, and S. R. Hartman, Phys. Rev. 141, 391 (1966).
[18]S. Meth, Ph.D. thesis (Columbia University, 1977) (unpublished), p. 57.
[19]P. E. Jessop and A. Szabo, Phys. Rev. Lett. 45, 1712 (1980); S. Chu, H. M. Gibbs, S. L. McCall, and A. Passner, *ibid.* 45, 1715 (1980).
[20]Private communication to P. W. Anderson by J. M. Ziman as noted in Ref. 21.
[21]P. W. Anderson, Comments Solid State Phys. 2, 193 (1970).
[22]R. Orbach, in *Optical Properties of Ions in Solids,* edited by B. Di Bartolo and D. Pacheco (Plenum, New York, 1975), p. 396.
[23]R. G. De Voe, A. Wokaun, S. C. Rand, and R. G. Brewer, Phys. Rev. B 23, 3125 (1981).
[24]P. F. Liao and S. R. Hartmann, Opt. Commun. 8, 310 (1973).
[25]D. Huber (unpublished).
[26]A. Compaan, Phys. Rev. B 5, 4450 (1972).