Lawrence Berkeley National Laboratory

Recent Work

Title

COHERENCE IN THE EXCITED STATES OF MULTIDIMENSIONAL SYSTEMS: DIMER AND EXCITON DYNAMICS IN CRYSTALLINE PHENAZINE

Permalink

<https://escholarship.org/uc/item/2223r02j>

Author

Zewail, Ahmed H.

Publication Date

1974-12-01

Submitted to Chemical Physics Letters

LBL-3521
Preprint c.1

COHERENCE IN THE EXCITED STATES OF
MULTIDIMENSIONAL SYSTEMS: DIMER AND
EXCITON DYNAMICS IN CRYSTALLINE PHENAZINE

Ahmed H. Zewail

December, 1974

RECEIVED
LAWRENCE
RADIATION LABORATORY

FEB 20 1975

LIBRARY AND
DOCUMENTS SECTION

Prepared for the U. S. Atomic Energy Commission
under Contract W-7405-ENG-48

For Reference

Not to be taken from this room

LBL-3521
c.1

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

-iii-

COHERENCE IN THE EXCITED STATES OF MULTIDIMENSIONAL SYSTEMS:

DIMER AND EXCITON DYNAMICS IN CRYSTALLINE PHENAZINE

Ahmed H. Zewail

Department of Chemistry; and Inorganic Materials Research Division,
Lawrence Berkeley Laboratory; University of California
Berkeley, California 94720

ABSTRACT

Coherence in multidimensional excited systems was established using optical and *zerofield* magnetic resonance spectroscopy. *High resolution* phosphorescence microwave double resonance at the Larmor frequency of the excited phenazine dimer gave the dynamics of optical coherence while the magnetic resonance spectra at 1.6°K gave a coherence time which was shown to be at least 10^5 times that of the stochastic-Markoffian limit and approximately 1/1000 of the radiative lifetime. These results indicate that the cross-chain hopping does not destroy the in-chain spin correlation.

INTRODUCTION

The migration of currentless excitons in molecular crystals is determined by the velocity which is derived from the exciton dispersion in the k -domain, and the coherence time, τ . The latter is the lifetime of the state in the rigid lattice approximation. In a vibrating solid, however, τ depends on the extent to which these excitons couple to the lattice (*e.g.*, exciton-phonon interaction) by a relaxation Hamiltonian. This coupling¹⁻¹¹ results in the modulation of the energy of the stationary states as well as inducing transition fluctuations between them. The net result is that the well-defined phases of the eigenstates become ill-defined and ultimately average out due to these ensemble fluctuations; the system becomes completely incoherent and is best described by a random walk process. Since these processes depend on the property of the whole ensemble, a density operator can easily describe the different limits of exciton migration and hence the loss of coherence in the k -states of the band. In fact, all the coherence information is contained in the off-diagonal elements of the time-dependent density matrix.

The relative magnitudes of τ and the exciton transfer time, which is related to the band dispersion, determine the dynamics of the migration. For isotropic scatterings, the diffusion of the excitons can be described by the following equation:

$$D_{ij} = \tau \langle V_i V_j \rangle = \ell \langle V_i V_j / |V(\vec{k})| \rangle \quad (1)$$

where V_i is the i th component of the velocity tensor of a *free* exciton; ℓ is the coherence length, and D_{ij} are the diffusion tensor elements which

not only depend on τ and V , but also on the statistical average (denoted by the brackets). Therefore, to fully characterize the exciton dynamics, the coherence parameters of the exciton must be determined.

The coherent properties of exciton states are related¹¹⁻¹³ to those of dimers (two-molecule chain) if the physics governing the scattering is not drastically different. As in the exciton case, coherence in dimers is determined by the rate at which the well-defined phases of the two states become random owing to the ensemble fluctuations. The comparison between the coherence time and the dimer transfer time thus establishes whether or not the excited states are coherent. Consequently, two limits must be recognized; the coherent limit where the resonance transfer time is much less than $\tau(\pm)$ of the two dimer states, and the incoherent limit where the transfer time is longer or comparable with τ . The same description can be used to characterize coherence in exciton states. In fact, there will be a one-to-one correspondence between the coherent properties of excitons and those of dimers, particularly if the band dispersion is simple, as in linear chain systems.

However, in systems where the interaction between the chains is not negligible, the coherent properties depend on both the in-chain dynamics and the rate of cross-chain interaction. Thus the study of dimers in such systems not only establishes the relationship between the topology of the crystal and dimer coherence, but also shines some light on the problem of exciton dynamics.

In this paper we will (a) report for the first time on the measurements of these coherence parameters in a multidimensional system (*i.e.*, the resonance interactions are not confined to one direction) at low temperatures ($< 4.2^\circ\text{K}$) using optical and magnetic resonance spectroscopy in zerofield.

(b) Illustrate the use of *high resolution* phosphorescence microwave double resonance (PMDR)¹⁴ in studying band structure of molecular solids; (c) show the importance of spin-orbital coupling in describing the spin transitions in molecular aggregates formed in a two-dimensional lattice and finally (d) suggest a way for describing optical coherence in band-to-band transitions.

EXPERIMENTAL

Isotopically mixed phenazine crystals were grown from the melt under vacuum with guest composition ranging from 0.1% m/m - 6% m/m (by weight). The spectrometer for the optical detection of magnetic resonance is described in detail elsewhere.^{13,15} Crystals were excited by a 100 W. mercury lamp and the appropriate filters. A 3/4-meter Jarrell-Ash spectrometer (resolution $0.2 \text{ \AA}$) was used to isolate the monomer from dimers emission, and the photomultiplier (EMI 9558) output was phase detected to monitor the microwave induced phosphorescence. All experiments were recorded under c.w. optical excitation.

PHENAZINE CRYSTAL STRUCTURE

Phenazine crystallizes¹⁶ in two forms, α and β . Crystals grown from the melt have the properties of the α -form.¹⁷ The space group of α -phenazine is $P2_1/a$ with two molecules in the unit cell: $a = 13.22 \pm 0.01$, $b = 5.06 \pm 0.005$, and $c = 7.088 \pm 0.007 \text{ \AA}$; $\beta = 109^\circ 13' \pm 15'$. The arrangement of the molecules in the crystal and the molecular axes adopted here are shown in Figure 1. One of the main features of the crystal packing is the stacking of the molecules along the shortest translational axis, b .

TRIPLET EXCITONS OF CRYSTALLINE PHENAZINE: GROUP VELOCITY AND BAND DISPERSION

The anisotropy of intermolecular interactions in the triplet state of phenazine crystal has been established from optical absorption^{18,19} and emission.²⁰ The ratio²⁰ of translationally equivalent (TE) to translationally inequivalent (TI) splitting is 4.2. Figure 2 gives the resonance multiplet emission of the isotopic mixed crystal at 1.7°K . One fact is evident from the spectrum: an emission at energy of $-\Delta$ from the monomer energy was not seen (2Δ is the dimer splitting which is simply twice the resonance interaction if the cluster eigenfunctions do not project onto the host states). This indicates that one dimer state has a destructive moment while the observed one has a constructive moment to the ground state of the crystal. Hence, the $17 \pm 1 \text{ cm}^{-1}$ splitting²⁰ is the contribution of the TE interaction to the band structure (cf. Fig. 2).²¹ The other contribution is from the interaction,¹⁸ β_{ab} , between interchange

equivalent molecules of phenazine unit cell. Assuming that the b-axis is the principal axis²² for the TE interactions (cf. Figure 1), neglecting the small interactions, and utilizing only near neighbor forces, the complete band dispersion is:

$$\epsilon_{\pm}(\vec{k}) = 2\beta_b \cos(\vec{k} \cdot \vec{b}) \pm 2\beta_{ab} [\cos \vec{k} \cdot \frac{1}{2}(\vec{a} + \vec{b}) + \cos \vec{k} \cdot \frac{1}{2}(\vec{a} - \vec{b})] \quad (2)$$

The gradient of the above energy dispersion (cf. Figure 2) with respect to $\vec{k}$ in a given direction is the exciton group velocity in that direction:

$$V(\vec{k}) = (1/\hbar) \nabla_{\vec{k}} \epsilon(\vec{k}) \quad (3)$$

Thus, V_i and V_j of Equation (1) can be determined. However, the population distribution among the k-states must be known in order to perform the statistical averaging. Since $KT \ll$ bandwidth, we cannot assume equal distribution. For a thermalized band, the mean square group velocity can be expressed in terms of the partition function of the two exciton branches, the lattice spacing between the molecules in the different crystal directions, resonance integrals and modified Bessel functions for a fixed temperature T. For example, the diagonal element (ath component) of the velocity tensor is given by:

$$\langle V_a^2(T) \rangle = \frac{\frac{1}{\hbar^2} \int \sum_{\mu=+,-} (\partial \epsilon_{\mu} / \partial k_a)^2 Z(\epsilon_{\mu}) d^3k}{\int \sum_{\mu=+,-} Z(\epsilon_{\mu}) d^3k} \quad (4a)$$

$$= a^2 \beta_{ab}^2 / \hbar^2 f(I_0^{\beta_1}, I_1^{\beta_1}; \beta_1, \beta_2) \quad (4b)$$

f is a function which depends on $\beta_1 (= 2\beta_b/KT)$, $\beta_2 (= 2\beta_{ab}/KT)$ and on the Bessel functions²³ I_0 and I_1 . Furthermore, in the high temperature limit⁴ f goes to one. $Z(\epsilon_+)$ and $Z(\epsilon_-)$ are the Boltzmann factors of the two branches. Similar expressions can be obtained for $\langle V_b^2(T) \rangle$.

Since β_b is known from our optical spectra and β_{ab} from the neat crystal absorption,¹⁸ both $\langle V_b^2(T) \rangle^{1/2}$ and $\langle V_a^2(T) \rangle^{1/2}$ can be obtained using the above equations. For $T = 3^\circ K$, these velocities are 3.7×10^4 and 4.9×10^3 cm/sec, respectively. These results assume the perfection of the lattice and the absence of elastic scattering among the k -states.⁹

SPIN AND OPTICAL EXCHANGE IN PHENAZINE EXCITON STATES: COHERENCE TIME

The Larmor frequencies of the zerofield transitions of triplet state phenazine monomer, $\omega(M)$, and dimer, $\omega(D)$, were obtained from their optically detected magnetic resonance (ODMR)¹⁴ spectra. To extract the correlation time for scattering from these steady state measurements, the modified²⁴ Bloch equations in the *rotating frame* were solved for the thermalized pair states ($\pm$), and an extrapolation to the band states was made. These equations are given by:

$$\dot{G}_+ - i[n_+ \gamma H_1 M_0 - \Delta \hat{\omega}_+ G_+] = (G_- \tau_{-+}^{-1}) - (G_+ \tau_{+-}^{-1}) \quad (5a)$$

and

$$\dot{G}_- - i[n_- \gamma H_1 M_0 - \Delta \hat{\omega}_- G_-] = (G_+ \tau_{+-}^{-1}) - (G_- \tau_{-+}^{-1}) \quad (5b)$$

γ is the gyromagnetic ratio for the electron and $G = u + iv$; u and v are the dispersion and absorption components of the complex moment G whose time derivative ($\dot{G}$) depends on the spin population ($n_{\pm}$) in the two states,

-7-

the Larmor frequencies ($\omega_0^\pm$) and the scattering rates ($\tau_{\pm}^{-1}$) between the two exciton states due to the relaxation Hamiltonian. H_1 is the amplitude of the rf field of frequency ω , and M_0 is the equilibrium magnetization. $\Delta\hat{\omega} = (\omega_0 - iT_2^{-1}) - \omega$, where T_2 is the transverse relaxation time. Knowing the difference in Larmor frequencies, the coherence time can therefore be established; the correlation time for the photons at this single point of the *time-correlation-function* is suitable for such measurements.

To correlate the results of the optical measurements with those obtained from the ODMR spectra, the exchange theory^{25a} was used to relate the scattering in dimers to the optical splitting. For the exciton case, the optical analogue^{25b} of the above magnetic equations can be written for any excited k state (cf. Reference 9) of the band (say, in a band-to-band optical transition of one-dimensional excitons²⁶):

$$\dot{P}_k + [T_{2k}^{-1} - 2\pi i(\nu_k^0 - \nu)]P_k - \sum_{k'} P_{k'} \tau_{k'k}^{-1} - P_k \tau_{kk'}^{-1} = C_k \quad (6)$$

The macroscopic polarization for the transition between the ground state[†] and the excited state (energy = $h\nu_0$) has the components, r_1 and r_2 : $P_k = r_{1k} + ir_{2k}$. The transition moment, μ_k , and the strength of the oscillating field, ϵ (which couples the two levels of population, N_0 and N_k), are contained in C_k : $C_k = -\hbar^{-1}i\epsilon\mu_k^2(N_0 - N_k)$. The solution of these equations in different limits²⁷ of band width and dispersion gives the coherence time. Further, the comparison between theory and experiment can establish the thermalization regime (Boltzmann or non-Boltzmann) for scattering among band states.

To extract τ from the optical measurements, one needs the linewidth of the transitions as well as the separation between them. The dimer

[†]The transition is labeled only by the exciton momentum index.

emission is not completely resolved from the N-mers. *High resolution* PMDR spectra of isotopic phenazine crystals ($T = 1.7^\circ\text{K}$) at $\omega(\text{D})$ resonance frequency²⁸ isolates the dimer phosphorescence (cf. Figure 2), which is in excellent agreement with the spectrum extrapolated from the steady state phosphorescence. Approximating⁴ the lineshape of the transition, a coherence time of 6×10^{-13} sec is characteristic of $\psi(+)$ dimer state. Further, the broadening of the dimer transition ($1.9 \text{ \AA}$) implies that the modulation of the dimer stationary states by the scattering Hamiltonian is perhaps significant, but not very rapid. In fact, Equation (6) tells us that the optical exchange between phenazine states is not exceedingly fast. Further yet, the resonance transfer time is comparable with $\tau(+)$, which implies the incoherent nature of dimer states.

For the exciton case the situation is analogous. The linewidth²⁹ of the two Davydov components of the $k = 0$ level is 0.6 cm^{-1} , which gives a scattering time of 4.4×10^{-12} sec. Using the value of $\langle V_a^2 \rangle^{1/2}$, $\lambda = 2.2 \text{ \AA}$ which is less than the lattice spacing. In other words, it seems that coherence in the dimer and exciton states is lost at $T < 4.2^\circ\text{K}$ where one expects the phonon population to be relatively small. However, crystal field inhomogeneity is expected to influence the coherence time measurements. Further, the experimental measurements are limited by the correlation time at this point of the correlation function. Alternatively, the coherence time was obtained from the magnetic spectra of the excited state.

The ODMR spectra (cf. Figures 3 and 4) of the monomer and the dimer consist of an allowed (by symmetry) electron spin transition flanked by weaker satellites due to hyperfine and quadrupole interactions^{30,31} between the nuclei and the electron spin angular momentum of the excited triplet

state. The excitation distribution in the pair, obtained from the analysis of the monomer and the dimer spectra, will be published later. A typical spectrum for the dimer is shown in Figure 4. The Larmor frequency of the $|D| + |E|$ transition of the dimer differs by 1.3 MHz (δ_{xz}) from that of the monomer, while the 2E transition is different by 2.8 MHz (δ_{xy}).

These differences in Larmor frequencies can be explained by invoking the spin-orbital induced anisotropy in the dimer spin states¹¹ and consequently in the different k-states^{7,11} of the exciton band. Phenazine (9,10 diazaanthracene) is a prototype system for such interactions because of the large and selective coupling³² between its lowest $\pi\pi^*$ state and the nearby³³ $n\pi^*$ state. A simple group theoretical argument indicates that if the non-bonding orbitals are parallel to the y-axis (cf. Figure 1), T_x and T_z spin states will benefit from the coupling with the nearby $n\pi^*$ state (via spin orbital Hamiltonian) while T_y will not. Therefore one expects a larger anisotropy between the spin-orbital coupling routes of the $2|E|$ transition compared to the $|D| + |E|$ transition, as confirmed experimentally.

From the solution of Equation 5 one expects only one transition (cf. Figure 4) for the pair since the intensity of the other transition is very small (due to the large bandwidth) contrary to the observation¹² of both transitions in small band cases. We therefore obtain a coherence time of 10^{-6} sec that is clearly longer by orders of magnitude than that obtained from the optical measurement which is limited by the experimental correlation time and crystal field influences. This indicates that the system is coherent for 10^5 times longer than that of the

stochastic limit. Assuming that the dimer coherence time is of the same order of magnitude as that of $\tau(k)$ at this single point of the correlation function, and using the value of $\langle v_b^2 \rangle^{1/2}$, the coherence length and D_{bb} are 10^6 Å and $685 \text{ cm}^2/\text{sec}$ respectively. These values are larger by orders of magnitude than that of random walk processes.^{4,34} However, it should be mentioned here that these results represent an upper limit since the crystal is not necessarily free from dislocations, strains and impurities. We hope to report on similar experiments which can distinguish between coherence times in different directions of the Brillouin zone, so a map for the scattering processes could be drawn and a test for the assumption⁴ of "constant relaxation time" can be made. The details of the optical and magnetic resonance in zero and applied magnetic field will be published in a forthcoming paper.

ACKNOWLEDGEMENT

I wish to thank Professor C. B. Harris for his support and encouragement, and the very useful comments which were instructive. The discussions with R. Shelby and W. G. Breiland were very helpful and fruitful. This research was supported in part by the National Science Foundation and in part by the Inorganic Materials Research Division of Lawrence Berkeley Laboratory under the auspices of the U.S. Atomic Energy Commission.

REFERENCES

1. H. Sternlicht and H. McConnell, J. Chem. Phys. 35, 1793 (1961)
2. J. Jortner, S. A. Rice, J. L. Katz and S. I. Choi, J. Chem. Phys. 42, 309 (1965).
3. H. Haken and G. Strobl, in The Triplet State, edited by A. B. Zahlan, Cambridge University Press (1967).
4. P. Avakian, V. Ern, R. E. Merrifield, and A. Suna, Phys. Rev. 165, 974 (1968).
5. R. W. Munn and W. Siebrand, J. Chem. Phys. 52, 47 (1970).
6. M. Grover and R. Silbey, J. Chem. Phys. 54, 4843 (1971).
7. A. H. Francis and C. B. Harris, Chem. Phys. Lett. 9, 181 (1971).
8. R. P. Hemenger, K. L. Lindenberg and R. M. Pearlstein, J. Chem. Phys. 60, 3271 (1974).
9. C. B. Harris and M. D. Fayer, Phys. Rev. B 10, 1784 (1974).
10. R. M. Hochstrasser and P. N. Prasad, in Excited States, Vol. 1, edited by E. Lim, Academic Press (1974).
11. A. H. Zewail and C. B. Harris, Phys. Rev. B XX, XXX (1975).
12. A. H. Zewail and C. B. Harris, Chem. Phys. Lett. 28, 8 (1974).
13. A. H. Zewail and C. B. Harris, Phys. Rev. B XX, XXX (1975).
14. For a review, see M. A. El-Sayed, Acc. Chem. Res. 4, 23 (1971).
15. M. J. Buckley and C. B. Harris, J. Chem. Phys. 56, 137 (1971).
16. F. H. Herbstein and G. Schmidt, Acta Cryst. 8, 399 (1955); ibid., 406 (1955).
17. R. M. Hochstrasser, J. Chem. Phys. 36, 1808 (1962).
18. R. H. Clarke and R. M. Hochstrasser, J. Chem. Phys. 47, 1915 (1967).
19. R. M. Hochstrasser, M. T. P. Int. Rev. of Sci., in press. See also R. H. Clarke, Chem. Phys. Lett. 6, 413 (1970).
20. A. H. Zewail, Chem. Phys. Lett., in press (1974).

21. Corrections for exciton superexchange must be taken into consideration for accurate determination of the resonance interactions. However, the moment expansion method, applied to these types of lattices (H.-K. Hong and R. Kopelman, J. Chem. Phys. 55, 724 (1971)), indicate that the modulation of the energy of TE pairs by the TI interactions is relatively small (see ref. 28).
22. From Figure 1, one might expect the b-axis translationally equivalent interaction to be the largest if the optical transition is purely $\pi\pi^*$ in nature. The extension (R. H. Clarke, Ph.D. Thesis, University of Pennsylvania, 1969) of the Jortner, Rice, Katz and Choi (J. Chem. Phys. 42, 309 (1965)) π -electron exchange calculations to phenazine triplet state predicted a large translationally equivalent interaction. We plan to experimentally identify the principal axes of the exchange interactions by optical and magnetic techniques.
23. Handbook of Mathematical Functions, edited by M. Abramowitz and I. A. Segun, Dover Publications, Inc., New York, 1965.
24. H. McConnell, J. Chem. Phys. 28, 430 (1958).
25. (a) R. Kubo and K. Tomita, J. Phys. Soc. Japan 9, 888 (1954);
(b) I. D. Abella, N. A. Kurnit and S. R. Hartman, Phys. Rev. 141, 391 (1966).
26. R. M. Hochstrasser, T. Li, H.-N. Sung, J. E. Wessel and A. H. Zewail, Pure & Appl. Chem. 37, 85 (1974).
27. A. H. Zewail, to be published.
28. The high resolution PMDR spectra are very sensitive to the power level because of the proximity of the monomer and dimer Larmor frequencies. However, the spectra clearly show the characteristics of the pumped N-mer emission. These observations enabled us to identify the different kind of dimers and to directly measure the exciton superexchange. The details of these studies will be published later.
29. R. H. Clarke, Ph.D. Thesis, University of Pennsylvania (1969).
30. J. Schmidt and J. H. van der Waals, Chem. Phys. Lett. 3, 546 (1969).
31. For the analysis of zero-field ODMR spectra of dimers, see R. M. Hochstrasser and A. H. Zewail, Chem. Phys. 4, 142 (1974).
32. M. A. El-Sayed, J. Chem. Phys. 43, 2864 (1965).
33. R. M. Hochstrasser and C. Marzzacco, J. Chem. Phys. 49, 971 (1968).
34. In the coherent limit it is not possible to calculate the absolute magnitude of the diffusion coefficients, and hence the description of the exciton migration by this process is not meaningful. However, in the intermediate limit one could picture the exciton as walking randomly following its well-behaved steps.

35. J. Ph. Grivet and J. M. Lhoste, Chem. Phys. Lett. 3, 445 (1969).
36. D. A. Antheunis, J. Schmidt and J. H. van der Waals, Chem. Phys. Lett. 6, 255 (1970).
37. D. A. Antheunis, J. Schmidt and J. H. van der Waals, Mol. Phys. 27, 1521 (1974).

FIGURE CAPTIONS

Figure 1: The crystal structure (cf. reference 16) and the molecular symmetry of phenazine: (A) The projection of the unit cell down the c-axis. Both kinds of molecules, translationally equivalent and interchange equivalent, are depicted. (B) Molecular axes of the molecule. (C) The ordering of the spin substates taken from reference 35.

Figure 2: The left hand section of this figure gives the dispersion of the exciton band in two dimensions; the lengths of cross-hatched and slashed rectangles are the translationally equivalent and translationally inequivalent splittings, respectively. The top figure shows the phosphorescence zero-zero transition of 2% isotopic mixed phenazine crystal at 1.7°K. The slashed columns are located at the energy of the different N-mers of the b-axis chains. The heights of these columns do not represent the actual transition moments and the interchange equivalent pairs are not depicted. The bottom spectrum shows both the phosphorescence and the $2|E|$ -PMDR spectra at less resolution.

Figure 3: Zerofield ODMR spectra of isotopic mixed phenazine crystal (0.5%) at 1.6°K. The top spectrum was recorded with 0 db (8 mw, 50% modulation) output power while the bottom spectrum was taken at -10 db power level. The peak position of the allowed spin transition is referred to as the frequency of the $|D| + |E|$ transition. The zerofield transition frequencies are in good agreement with those reported for phenazine in a diphenyl host (cf. references 35-37).

Figure 4: The right hand section of this figure demonstrates the relationship between the monomer and the dimer microwave absorption for wide band cases. Only the slow exchange limit is shown. $2\delta_{ij}$ is the separation between $\psi(+)$ and $\psi(-)$ zero-field ODMR transitions in the ij th manifold ($i,j = x,y,z$). The left hand side of the figure shows the ODMR spectrum ($|2E|$ transition) of the dimer at 1.6°K. Transitions characteristic of the perdeutero host were observed on the perproto emission with opposite phase; the intensity is sensitive to both power and concentration. Detailed analysis of the shape of these transitions could establish the relationship between the dimer and exciton spread in Larmor frequencies, and the density of states in phenazine crystals.

(A)

(B)

(C)

Fig. 1

OPTICAL EMISSION AND BAND DISPERSION OF PHENAZINE CRYSTAL

Fig. 2

ZERO FIELD ODMR SPECTRA OF PHENAZENE MONOMER

Fig. 3

ODMR SPECTRUM AND COHERENCE PARAMETERS
OF CRYSTALLINE PHENAZENE

Fig. 4

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720