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OPTICAL NUCLEAR POLARIZATION AS A CONSEQUENCE OF THE NON-CROSSING RULE (LEVEL-ANTI-CROSSING)

III. Experimental results and evidence for guest–host complexes in doped fluorene crystals

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Fluorene crystals doped with the guest molecules acridine, phenazine and anthracene have been studied before due to their strong Optical Nuclear Polarization (ONP) in low magnetic fields around 100 gauss. In this paper, these results together with supporting new data are interpreted in terms of ONP as a consequence of Level-Anti-Crossing (LAC) within the excited triplet state of a guest–host complex.

The results permit a first detailed check of the theoretical model of ONP by LAC including the influence of electronic relaxation. Furthermore, they render the spectroscopic parameters needed for the characterization of the triplet-state complexes:

– The zero-field splitting tensor and the orientation of its principal axes system with respect to the crystalline axes.

– The proton hyperfine tensor, which is essentially due to one-proton spin in the CH_2-group of the fluorene host molecule contributing to the complex.

1. Introduction

Optical Nuclear Polarization (ONP) has been discovered in 1967 [1] and has been observed since in a number of aromatic molecular crystals. Most challenging for the interpretation were the high absolute polarization which have been observed at room temperature in doped fluorene crystals [2] for rather low polarizing fields up to 200 G.

Although the essential features of the ONP could be understood with a general mechanism as developed in [3,4] and reviewed in [5,6], it was not possible to assign the striking low-field results to the well characterized triplet states of either the guest or host molecules. The breakthrough came with the postulation of a new type of triplet state in doped fluorene crystals, assigned to a complex formation among guest and host molecules. Experimental evidence was obtained for this interpretation with the direct EPR-detection of the triplet complexes in fluorene crystals doped with acridine and phenazine [7]. Alternatively [8], the spectroscopic parameters of the triplet state complexes could be obtained by assigning the low-field ONP-results to the polarization effects predicted by the general mechanism in the region of level-crossing among the spin sublevels of the triplet state complexes.

Strong ONP associated with level-anti-crossing due to the hyperfine coupling has been mentioned already in the first treatment of the general ONP-mechanism [3] as well as in the detailed analysis of the LAC-effects observed in the phosphorescence emission [9]. However, this idea has not been pursued due to the lack of experimental evidence.

Given the new situation, a thorough theory of the ONP by LAC has been worked out. A qualitative analysis based on the general ONP-mechanism [4] is given in [6]. A detailed analytical treatment [10] offers the most convenient access to the underlying physical concepts. In addition, the crucial role of electronic relaxation has been considered [11]. In particular, the influence of selective relaxation with significantly

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different relaxation rates for the transitions among the three triplet spin states turns out to permit a quantitative understanding of the experimental observations.

In this paper, we want to summarize the experimental results concerning the ONP by LAC in doped fluorene crystals. They allow a detailed check of the theoretical treatment [10,11]. Moreover, they provide further evidence for the guest–host complex formation and they extend the characterization of the relevant spectroscopic parameters, mainly the zero-field splitting and hyperfine tensor elements.

2. Methods and materials

The optical nuclear polarization has been measured by the two-field technique as described in [2] and reviewed in [5,6]. The ONP can be obtained conveniently as a function of the polarizing field H_p and of its orientation with respect to the crystalline axes. It has been shown [2a,6] that it is sufficient to measure the initial polarization rate $\dot{p}(0)$ starting with an unpolarized sample. $\dot{p}(0)$ is defined by the initial slope of the ONP as a function of the polarization time.

The absolute value of the polarization is obtained by normalizing to the total number of proton spins in the sample via the known population distribution in the Boltzmann equilibrium. The experimentally obtained ONP rate $\dot{p}(0)$ is proportional to the theoretically calculated rate $\dot{p}_T(0)$ normalized to one created triplet state. The proportionality factor contains the number of absorbed light quanta per unit time as well as the quantum efficiency of the intersystem crossing process populating the triplet state. Its experimental determination is known to be rather involved and inaccurate. This is of no importance for the present paper; note, however, that the proportionality factor can create large scale differences between the experimental polarization rates given here and the theoretical ones referred to.

We report measurements on fluorene crystals doped with acridine, phenazine and anthracene. The molecular structures with the chosen definition of the molecular axes as well as the fluorene crystal structure have been summarized in [6] and [7]. The light from a xenon arc was filtered to excite only the lowest energy singlet absorption of the guest molecules. In all cases, the ONP was found to depend exponentially on the polarization time. Furthermore, the initial polarization rate $\dot{p}(0)$ was proportional to the light intensity within experimental accuracy.

Table 1
List of doped fluorene crystals investigated in this paper

No.	ppm guest I	ppm guest II
Host:	fluorene-h_{10}	
1	1000 acridine-h_9	
2	200 acridine-h_9	+ 2000 acenaphthene-h_{10}
3	1000 acridine-d_9	
4	1000 phenazine-h_8	
5	200 phenazine-h_8	+ 2000 anthracene-h_{10}
6	1000 anthracene-h_{10}	
Host:	fluorene-d_8h_2	
7	1000 acridine-h_9	
8	1000 acridine-d_9	
9	1000 phenazine-d_8	
10	1000 anthracene-d_{10}	

The single crystals investigated in this paper are collected in table 1 together with the guest concentrations as prepared in the melt before crystal growing.

The synthesis of the host materials fluorene-h_{10} and fluorene-d_8h_2 (9,9′-H_2-octadeuterofluorene) was carried out in this laboratory [12]. The host material was extensively zone refined and checked for impurities which are at least below the 10 ppm level if not stated specifically. The crystals were grown from the melt, usually giving excellent cleavage planes which coincide with the crystalline *ab*-plane. Thus, it was possible to orient the crystal accurately (±0.5°), in particular when the magnetic field had to be oriented in the *ab*-plane or perpendicular to it. The positions of the crystalline *a*- and *b*-axes have been checked routinely using X-ray analysis. Independently, the orientation dependence of the ONP renders the same information. Agreement has been obtained in all cases within the experimental accuracy.

3. Analysis and discussion of the experimental results

Doped fluorene crystals have been investigated extensively [2] because the largest absolute polarizations by ONP were obtained in these systems. Now, these results become accessible to a detailed interpretation on the basis of the working hypothesis mentioned in the introduction:

(a) The observed effects are due to ONP as a consequence of LAC as described theoretically in refs. [10,11].

(b) On that basis the analysis of the ONP data renders the spectroscopic properties of a new type of triplet state, assigned to complex formation between guest and host molecules in agreement with the direct EPR-results [7].

Consequently, in this section, the presentation of the results will be combined with the analysis of the spectroscopic parameters of the triplet state complexes.

3.1. The principal axes system of the zero-field splitting tensor

As in ref. [7] an asterix will be used to indicate specific properties of the complexes. The first task is the assignment of the principal axes system x^*, y^*, z^* of the zero-field splitting tensor with respect to the crystalline axes. We will follow the EPR-convention that the z^*-axis is defined by the field orientation with the largest energy difference between the two $|\Delta m|=1$ transitions. This situation corresponds to the level-crossing case with the higher field, termed H^{z*}_{LC}.

The assignment of the other axes has been fixed with the following choice: One of the principal axes was found to be parallel to the long in-plane axis x of the fluorene molecules which coincides with the crystalline c-axis for all molecules of the unit cell. Thus, the corresponding axis of the complex has been termed x^* [7]. Note, that this definition determines also the relative signs of D^* and E^*. It is also helpful for a comparison with the zero-field splitting parameters of other molecular triplet states where the x-axis often coincides with the long in-plane axis.

The ONP-theory predicts the largest polarizations if the magnetic field is close to the LC-fields and if the field is aligned to the principal axes, where level-crossing occurs. Hence, the principal axes x^*, y^*, z^* should

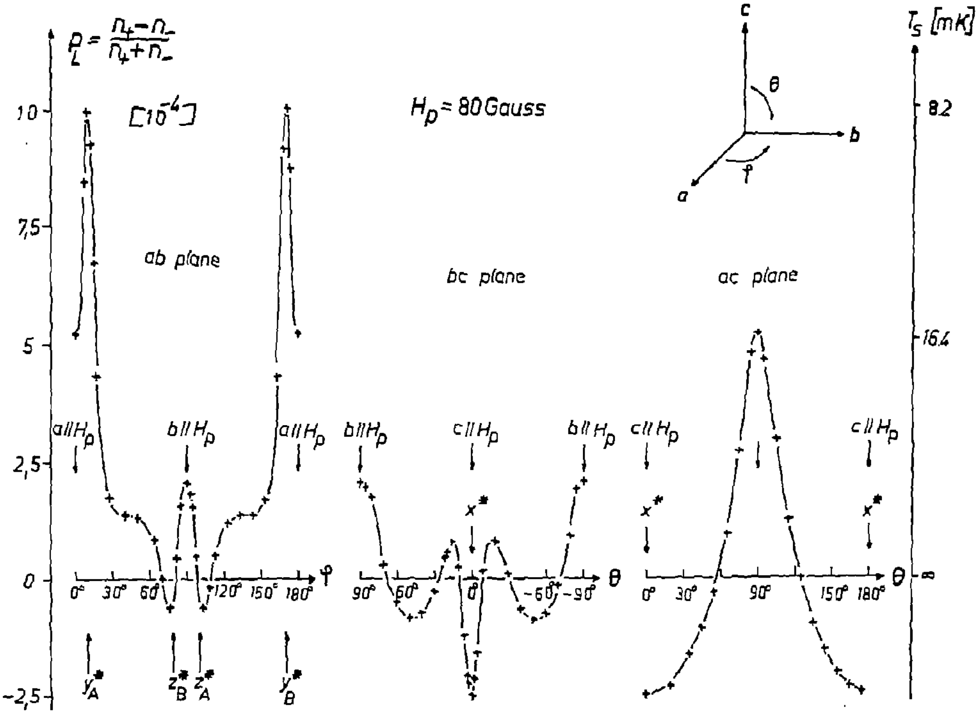


Fig. 1. Optical Nuclear Polarization in a polarizing field H_p = 80 G as a function of the field orientation in the three crystalline planes (taken from ref. [2b]).

The arrows mark the orientation of the crystalline axes a, b, c and the principal axes x^*, y^*, z^* of the zero-field splitting tensor of the triplet state complex.

be assigned with respect to the crystalline axes a, b, c by finding those field regions and orientations where ONP-extrema are observed as a function of both, the field strength and its orientation with respect to the crystalline axes.

This assignment has been carried out in fig. 1 for results reported earlier (see fig. 5 of [2b]) in crystals of partly deuterated fluorene-d_8h_2 doped with acridine-h_9 (system 7 in table 1). The orientation dependence in the ab-plane exhibits two sets (A and B) of a polarization maximum and minimum each. A deviation of the field orientation in planes perpendicular to the ab-plane reduces the ONP-extremes; thus the orientations characterized by the ONP-extremes are located precisely in the ab-plane.

The ONP-minima reach their extreme values in fields around 150 G while for all other orientations the extreme polarizations are observed in lower fields. Hence, the former orientations can be assigned to the up-field level-crossing case and they have been marked $z^*_{A,B}$ in fig. 1. The precise orientation of the z^*-axes was determined in fields near the ONP-extreme around 150 G.

The ONP-maxima are highest in fields near 80 G (see fig. 1) and the corresponding field orientations are found to be perpendicular to the $z^*_{A,B}$-axes within experimental accuracy. They define the second principal axis $y^*_{A,B}$ according to the axis definitions described above.

The fact that two principal axes are located in the ab-plane requires the third one to be parallel to the crystalline c-axis. Indeed, another ONP-extreme is observed at just this orientation. Note that the x^*-axis coincides for the two sets A and B of the principal axes. Thus, the two sets of principal axes exhibit the general features of the two magnetically inequivalent sites in the fluorene matrix, although the orientations of the y^*, z^*-axes in the ab-plane are completely different from the molecular y, z-axes of the fluorene molecules (see table 2).

For the comparison of the various complex and molecular axes with respect to the crystalline lattice it is sufficient to specify just one angle in the (a,b)-plane. For example, we choose here

$$\sphericalangle(y^*, a) \equiv \sphericalangle(z^*, b)\,, \tag{1a}$$

and compare it with the angle

$$\sphericalangle(y, a) \equiv \sphericalangle(z, b)\,, \tag{1b}$$

known experimentally for the fluorene and guest molecules. All data are collected in table 2. The values given for the ONP have been averaged over the values obtained for all the systems in table 1 as well as over the A and B sites. The average has been taken because the values for the individual systems and the two molecular sites never differ by more than ±1°. Consequently, the deuteration does not affect the orientations of the principal axes within experimental error.

Table 2
Summary of the orientation of the complex (x^*, y^*, z^*) and molecular (x, y, z) axes with respect to the crystalline axes (a, b, c)

Guest	Fluorene–guest complex $\sphericalangle(y^*, a) = \sphericalangle(z^*, b)$			Isolated molecule $\sphericalangle(y, a) = \sphericalangle(z, b)$	
	ONP	EPR	Mol. site	EPR	X-ray
Phenazine	14°	14°	(A)	57.5°	pure fluorene matrix
		15.5°	(B)	57.0°	
Acridine	10.5°	11°	(A)	59°	
		13.5°	(B)	56°	
Anthracene	11°	–	–	55.5° 56.9°	55° 10′
Ref.	this work	[7]		[7,13,14]	[6] of [7]

3.2. The zero-field splitting: D^*, E^*

The zero-field splitting parameters can be extracted from the ONP as a function of the polarizing field H_p oriented along the principal axes x^*, y^*, z^*.

The results of some of the systems of table 1 are presented in figs. 2–4. The curves have been assigned already to the individual level-crossing cases. The procedure will be discussed now in more detail and is based on the theoretical interpretation as outlined in [6,10,11].

Quite obvious is the assignment of the curve $H \| z^*$ (fig. 2, bottom). This orientation is defined to have the higher LC-field, i.e., $H^{z^*}_{LC}$. Indeed, only for the experimental curve, designated $H \| z^*$, sharp field dependencies are observed around 150 G while the ONP behaves smoothly for the other orientations of the field H_p in this field range.

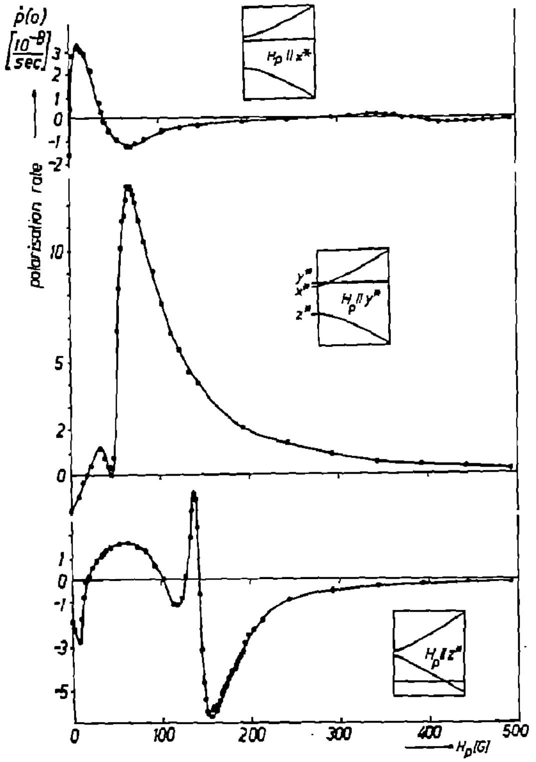


Fig. 2. Initial ONP-rate $\dot{p}(0)$ as a function of the polarizing field H_p oriented along the principal axes x^*, y^*, z^*.

The assignment of the axes to the individual curves is described in the main text.

System 1 of table 1: fluorene-h_{10} + acridine-h_9.

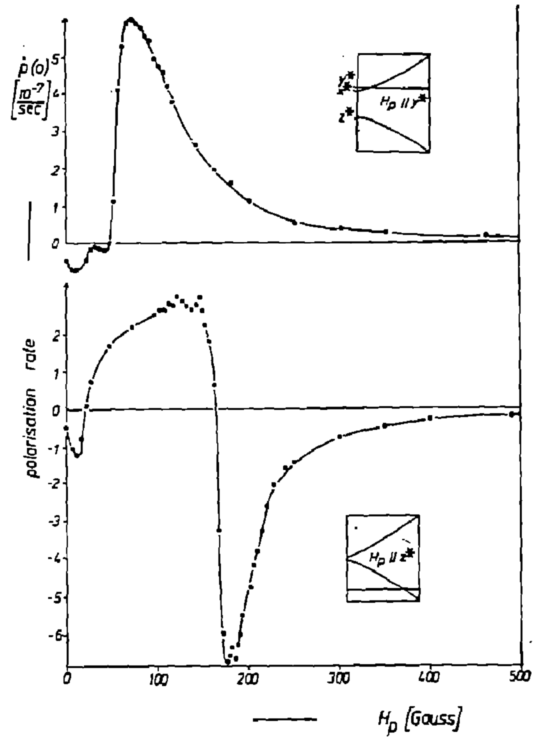


Fig. 3. Initial ONP-rate $\dot{p}(0)$ as a function of the polarizing field H_p oriented along y^* and z^*, the two level-crossing cases.

System 5 of table 1: fluorene-h_{10} doped with 200 ppm phenazine-h_8 and 2000 ppm anthracene-h_{10}.

The LC-field $H_{LC}^{z^*}$ can be determined from the field position of the turning point of the slope between the positive and negative ONP-contribution. Since the slope is rather steep, this field position and hence $H_{LC}^{z^*}$ can be extracted with an accuracy of better than 1 G and the values are collected in table 3.

The assignment of the second LC-case is more involved. The following arguments, however, support the choice $H \parallel y^*$ made in the figures.

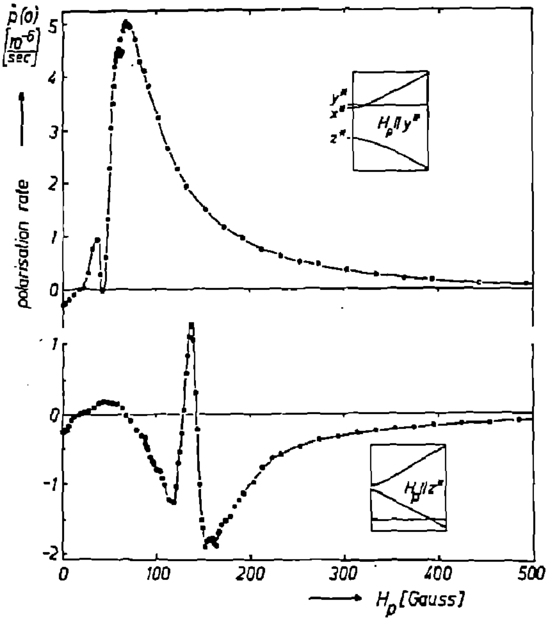
(1) When the influence of selective electronic relaxation is considered, the theory [11] produces excellent agreement with the experimental curve designated $H \parallel y^*$.

(2) From the EPR-results [7], $H \parallel y^*$ is obtained as the second LC-case, in agreement with the ONP-assignment.

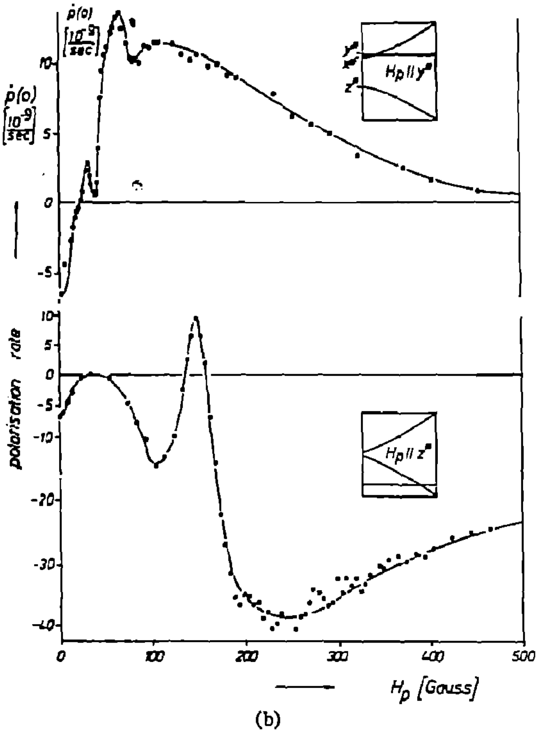
With the choice of the x^*-axis being parallel to the crystalline c-axis the assignment of the two level-crossing cases requires the same sign of the zero-field splitting parameters D^* and E^*. The absolute sign cannot be concluded from the present data. However, arguments have been given in [6] supporting the positive sign. Therefore the inserts in figs. 2–4 use this sign for the field dependence of the electronic energy levels.

As pointed out in [11] the influence of relaxation hampers a simple evaluation of a precise value of the LC-field $H_{LC}^{y^*}$ from the experimental curves.

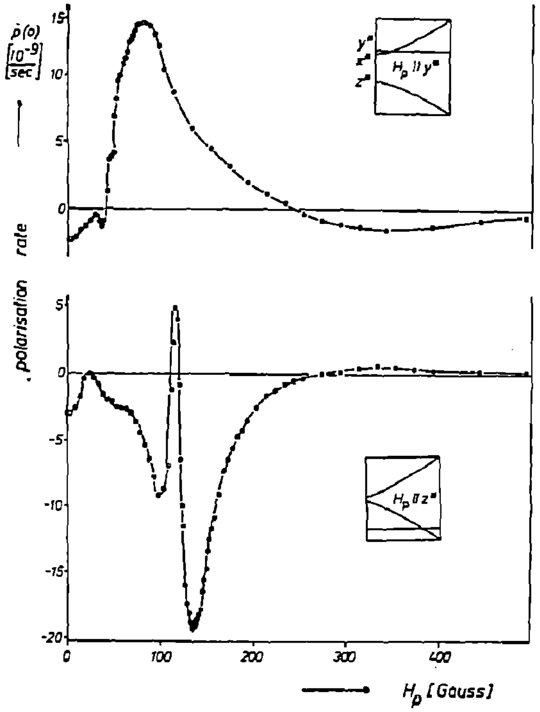
However, in a first approximation, $H_{LC}^{y^*}$ can be identified with the field position of the turning point in the downfield slope of the ONP-maximum observed for $H \parallel y^*$. This assignment would be analogous to that used for $H \parallel z^*$. It is also obvious that this field position renders an upper limit for $H_{LC}^{y^*}$ and thus for



(a)



(b)



(c)

Fig. 4. Initial ONP-rate $\dot{p}(0)$ as a function of H_p oriented along y^* and z^* for the partly deuterated host fluorene-d_8h_2 doped with:

(a) Acridine-h_9, system 7 of table 1.
(b) Phenazine-d_8, system 9 of table 1.
(c) Anthracene-d_{10}, system 10 of table 1.

E^*. The experimental values are again collected in table 3 and marked with the term "upper limit". The comparison with the EPR-values shows that they are definitely smaller than the EPR-results, but relatively close, which supports this assignment. Note, however, that the EPR-parameters result from measurements at lower temperatures around 230 K while the ONP results have been obtained at or above room temperature.

An alternative approach to the determination of $H^{y^*}_{\mathrm{LC}}$ is based on the following sequence of arguments. The ONP-curves for $H \| y^*$ exhibit one rather accurately defined feature: The field position of the sharp ONP-minimum on the downfield side of the broad ONP-maximum. The theoretical interpretation given in [11] specifies this field position to be within experimental error identical to the LAC-field of the

Table 3
LC-fields for the various systems of table 1. For $H \parallel y^*$, two fields are given as described in the text. The zfs-parameters D^* and E^* are calculated from the LC-fields given in the second and third column. The EPR-values are taken from [7]. Standard errors in the last decimal are given in parentheses

Guest	Crystal no. (table 1)	$H_{\mathrm{LC}}^{z^*}$ (G)	$H_{\mathrm{LC}}^{y^*}$ (G) upper limit	$H_{\mathrm{LAC}}^{(n),y^*}$ (G) minimum	D^* (cm^{-1})	E^* (cm^{-1})
Acridine	1	143	52	45		
	2	142	52	44		
	3	144	–	–		
	7	143	50	43		
	Average	143(1)	51(2)	44	0.0134	0.00080
	EPR	141	55		0.0132(1)	0.00093(10)
Phenazine	5	166(1)	53(3)	45	0.0155	0.00075
	9	165(1)	47(3)	38	0.0154	0.00060
	EPR	168	57		0.0157(1)	0.00085(10)
Anthracene	6	129	48		0.0121	0.00078
	10	123(1)	47(3)		0.0115	0.00078

narrow ONP-contribution, i.e.,

$$H_{\mathrm{LAC}}^{(n),y^*} = H_{\mathrm{LC}}^{y^*} \pm \tfrac{1}{2} A_{y^*y^*}/g\beta\,. \qquad (2)$$

The experimentally obtained values for $H_{\mathrm{LAC}}^{(n),y^*}$ are collected in table 3. Depending on the sign of the hyperfine splitting element $A_{y^*y^*}$, $H_{\mathrm{LC}}^{y^*}$ is upfield or downfield from the minimum position. Although both the sign and the absolute value of the hyperfine tensor elements can be obtained in principle from the complete set of ONP-results, at present, the choice of the sign in (2) will be left open. In any case, the following conclusions can be correlated with the two field positions given in table 3 for $H \parallel y^*$:

(a) The difference between the fields represents an estimate of the value of the hyperfine coupling element given in (2).

(b) Alternatively, the E^*-value can be even smaller than given by $H_{\mathrm{LC}}^{y^*} = H_{\mathrm{LAC}}^{(n),y^*}$, which then represents a new upper limit for $H_{\mathrm{LC}}^{y^*}$. It should be emphasized that the absolute E^*-value is certainly very small, more than an order of magnitude smaller than D^*. Hence, it is not surprising that its determination is quite critical. On the other hand, the exact position of $H_{\mathrm{LC}}^{y^*}$ hardly affects the determination of the D^*-value from $H_{\mathrm{LC}}^{z^*}$, because the approximation

$$D^* \approx g\beta\, H_{\mathrm{LC}}^{z^*}\,,$$

is applicable within experimental accuracy as can be checked quickly with the values given in table 3.

Table 3 demonstrates that the zfs-parameter D^* agrees within experimental accuracy for the completely independent methods. Hence we conclude:

(1) Both EPR and ONP establish the existence of the complex triplet states in doped fluorene crystals with the main feature of an unusually small zero-field splitting. Note that the complexes are clearly detectable by ONP even in fluorene crystals doped with anthracene where no EPR-signals could be observed.

(2) With their successful interpretation the ONP-results represent the first experimental verification of ONP by LAC.

3.3. Further information on the triplet-state complexes

A set of experiments has been performed with fluorene crystals doped with a second guest (systems 2 and 5 of table 1). The guest was chosen in order to provide a deep energy trap which is able to take over any excitation energy in the triplet exciton band of the host or energy provided by detrapping processes concerning the triplet-state complex. However, the ONP-results remain unchanged with respect to the LAC-effects and rule out the conceivable possibilities mentioned above.

According to the theory [10,11] the ONP-results can render in principle the hyperfine tensor which would be an important piece of information concerning the characterization of the complexes. First, however, the question should be answered, which of the many protons of the guest and host molecules make up the essential part of the hyperfine tensor.

So far, the ONP-results have been interpreted with the assumption that only one proton spin interacts with the electronic triplet state. In order to check this hypothesis experimentally, the influence of the majority of the protons can be excluded by selective deuteration of the guest and host molecules. Although the complete set of deuteration possibilities has not been investigated yet, the results obtained so far allow important conclusions. Fig. 5 presents three ONP curves ($H \| z^*$) for the progressively deuterated system of fluorene doped with acridine.

Comparing fig. 5(a) and (b), we note that the deuteration of the guest hardly changes the result at all. Hence we conclude that the hyperfine tensor due to the protons of the guest in the triplet state complex is negligible with respect to the predominant hyperfine tensor of the other protons.

The result for the partly deuterated host molecule in fig. 5(c) indicates more significant changes:

(a) The whole positive region of ONP below 100 G is considerably reduced.

(b) The ONP-minimum near zero-field is absent.

These changes are therefore expected to be due to the hyperfine tensor of the aromatic fluorene protons within the complex.

Most important, however, seems the fact that the predominant field dependence of the ONP around the LC-region centered at 142 G remains essentially unchanged with respect to the results of fig. 5(a) and (b). As a consequence, the by far predominant part of the proton hyperfine tensor is due to the fluorene CH_2-protons within the triplet state complex.

Going one step further, we may even conclude that only one of the CH_2-protons of a fluorene molecule can be responsible for the predominant part of the hyperfine tensor. The following arguments support this statement:

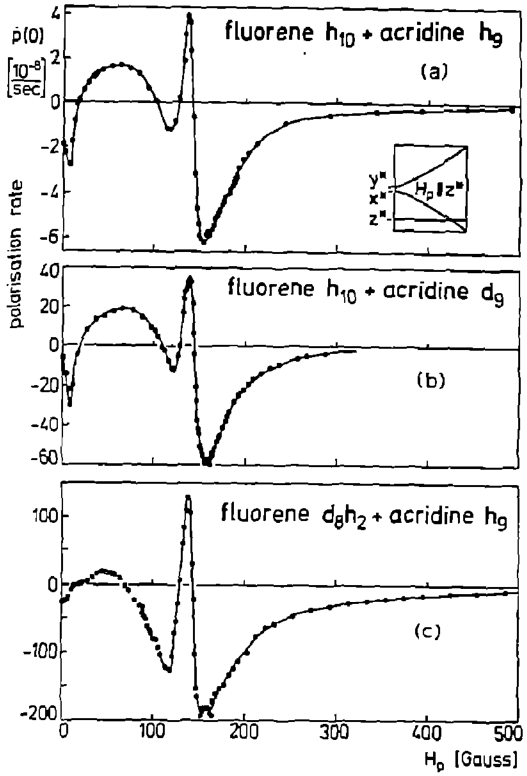


Fig. 5. Initial ONP-rate $\dot{p}(0)$ as a function of H_p oriented along z^*, the up-field level crossing case, for the progressively deuterated system fluorene doped with acridine.
(a) System 1 of table 1: fluorene-h_{10} + acridine-h_9.
(b) System 3 of table 1: fluorene-h_{10} + acridine-d_9.
(c) System 7 of table 1: fluorene-d_8h_2 + acridine-h_9.

(1) Inspection of the fluorene crystal structure (as reproduced in [6,7] shows that only one of the CH_2-protons of a fluorene molecule can be involved in the complex formation at a given instant of time. This does not exclude the possibility that more than one of fluorene host molecules contribute to the complex. Alternatively, two or more guest molecules could contribute to one complex. This possibility seems to be ruled out for guest concentrations below 1000 ppm used in the present experiments; in addition, the statistical distribution of the guest molecules in substitutional sites of the fluorene matrix is confirmed by the EPR-

[7] and ONP-results. Hence, it is plausible, that a single proton site of a fluorene CH_2-group plays the central role in the complex formation as characterized by a large hyperfine coupling.

(2) The EPR-results (see fig. 3 of ref. [7]) exhibit an uninterpreted doublet structure of the $|\Delta m| = 1$ transitions of the triplet state complexes if the magnetic field is aligned near $H \| x^*$. At the present state of knowledge it is tempting to interpret this splitting as due to the hyperfine coupling constant $A_{x^*x^*}$ of one proton in a fluorene CH_2-group within the complex.

Therefore, it seems likely that the triplet state complexes in doped fluorene crystals represent the rare example of a triplet state, the hyperfine coupling of which is mainly due to a single proton spin. It is needless to say that by accident this system is ideal to study the details of the ONP by LAC.

Although the present results do not permit the analyzis of the complete hyperfine tensor yet, we can draw some relevant conclusions: The proton hyperfine tensor responsible for the ONP-results has to have elements with absolute values comparable to or higher than those of the hyperfine tensor known for the CH_2-protons in the triplet state of a fluorene monomer. In the latter case both protons are equivalent with a nearly isotropic hyperfine tensor and a coupling constant of about 7 G [14].

In this context we should remark that the proton hyperfine tensor of the complex is totally different from that of the monomer triplet states of both the guest and the host molecule, at least with respect to the fluorene CH_2-protons. This is in contrast to recent observations of the hyperfine coupling in excited triplet states of charge transfer complexes [15].

A further analysis of the hyperfine tensor in the complex requires first an extended theoretical treatment of the ONP by LAC which can be worked out in continuation of [10,11]. In addition, further EPR- and ONP-investigations are in progress with the aim to determine the whole hyperfine tensor of the triplet state complex which is hoped to provide a clue to the understanding of the nature of the complex formation.

Finally, we would like to note that the theoretical interpretation of the ONP [11] specifies a selective electronic relaxation among the triplet zero-field sublevels of the complex as given by the inequality

$$W_{x^*z^*} \gg W_{x^*y^*}, W_{y^*z^*} \, . \tag{3}$$

Note that the selectivity (12) corresponds to the predominant relaxation rate W_{xz} observed for the triplet state of acridine doped in diphenyl [16] at low temperatures. Note, however, that the transition $x^* \leftrightarrow z^*$ is not the high energy transition in the complex as it is in the monomer ($x \leftrightarrow z$). Furthermore, the orientations of the z^*- and z-axis are rather different for the complex and monomer (see table 1).

Although the ONP-results contain also independent information on the selective population and decay rates of the excited triplet states, this information is more directly available from the EPR-measurements. Hence, they are better used as an independent information in order to distinguish between alternative possibilities for the assignment of spectroscopic parameters from ONP-results.

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