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## The effect of pressure on the phase transitions in MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub> : an ESR study

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**Résumé.** — Le sel d'ion-radical MTPP(TCNQ)<sub>2</sub> subit une transition de phase du 1<sup>er</sup> ordre tandis que le sel isomorphe MTPA(TCNQ)<sub>2</sub> ne subit qu'une transition induite par la pression hydrostatique. Ces transitions de phase se manifestent par des modifications des spectres RPE. Nous avons observé et discuté les changements des paramètres des spectres ainsi que des paramètres caractérisant la dynamique des excitons en fonction de la pression et de la température.

**Abstract.** — The ion-radical salt MTPP(TCNQ)<sub>2</sub> undergoes a first-order phase transition whereas the isomorphous salt MTPA(TCNQ)<sub>2</sub> undergoes only a pressure-induced transition. These phase transitions have a pronounced influence on the ESR spectra. We have observed and discussed the changes in the spectral parameters and those describing the exciton dynamics as a function of pressure and temperature.

### 1. Introduction.

A systematic investigation of the pressure effect on the physical properties of organic semiconductors began about twenty years ago [1]. Up to now the transport [2], spectral [2, 3], and spin [4-6] effects were studied. In particular, the anion radical salts of methyltriphenylphosphonium (MTPP<sup>+</sup>) and methyltriphenylarsonium (MTPA<sup>+</sup>) with tetracyanoquinodimethane (TCNQ) have raised interest [1, 4, 6, 8, 9]. Merkl *et al.* [4] have studied the triplet-exciton magnetic resonance and have found a pressure-induced phase transition in MTPA(TCNQ)<sub>2</sub> by ESR studies. They have explained the phase diagram for the salt in terms of the elementary theory of non-interacting triplet excitons. Nuclear relaxation and exciton dynamics in MTPA(TCNQ)<sub>2</sub> have been studied by Devreux and Nechtschein [6], whereas the effect of pressure on the phase transition of MTPP(TCNQ)<sub>2</sub> has been studied by differential thermal analysis by Iida *et al.* [7]. Although many studies were devoted to the effect of pressure on the phase transitions in crystalline salts of MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub>, the situation is still not clear.

In our opinion, the results concerning the phase transitions obtained from electron spin resonance are not entirely clear and require a more extended discussion based on additional crystal structure data.

The above was the motivation for an electron spin resonance study of the effect of pressure on MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub> salts. We focus our attention on the pressure dependence of the ESR spectra. Some of our preliminary results have been published elsewhere [8, 9].

### 2. Experimental.

Methyltriphenylphosphonium (MTPP) and its isostructural arsonium analogue (MTPA) form the complex anion radical salts with 7,7,8,8-tetracyanoquinodimethane (TCNQ). Close similarities have been noticed between these two salts.

The only essential difference between them concerns the nature of the phase transitions : MTPP(TCNQ)<sub>2</sub> undergoes a first-order structural phase transition at ambient pressure at 315.7 K [4, 7], whereas the similar salt MTPA(TCNQ)<sub>2</sub> undergoes only a pressure-induced transition [4]. This difference has stimulated us to reconsider the ESR results for these salts.

For the ambient pressure phase transition in MTPP(TCNQ)<sub>2</sub>, sharp discontinuities were found in the temperature dependences of the electrical conductivity by Iida *et al.* [10, 11] and Croteau *et al.* [12], magnetic susceptibility by Chesnut *et al.* [13], visible absorption by Yakushi *et al.* [14], infrared

reflectivity by Świetlik and Graja [15] and in parameters of the triplet-excitons magnetic resonance by Merkl *et al.* [4]. The enthalpy and the total entropy change associated with the phase transition were experimentally determined by Kosaki *et al.* [16].

According to the results of crystal structure analysis given by Konno and Saito [17], TCNQ molecules in  $\text{MTPP}(\text{TCNQ})_2$  are forming tetramers and a zigzag TCNQ column is produced by the stacks of these tetramers along the  $b$ -axis. This molecular arrangement remains the same in both phases for both salts. The distances of TCNQ molecules between tetramers are shortened by about 0.02 Å, whereas the average interplanar distances in the tetrad are longer by 0.08 and 0.04 Å than the corresponding distances in the low-temperature modification. The overlapping mode of the TCNQ moieties between the tetrad is also changed. The conformation of  $\text{MTPP}^+$  ion is different from that of the low-temperature form : two of the phenyl groups are rotated by about  $56^\circ$  and  $45^\circ$  from the positions observed in the low-temperature state [17].

We performed the ESR measurements of oriented single crystals of  $\text{MTPP}(\text{TCNQ})_2$  and  $\text{MTPA}(\text{TCNQ})_2$ , for two perpendicular orientations. The « Radiopan » X-band microwave spectrometer SE/X2542, and a special pressure appliance were used. A pressure corundum resonator, disposed within a beryllium bronze high-pressure cylinder permitted us to measure the ESR spectra as a function of hydrostatic pressure up to 500 MPa and at temperatures between 80 K and 340 K [18, 19]. The temperature of the system was controlled by a nitrogen vapour flux pumped through a heat exchanger disposed outside the high-pressure vessel. We applied petroleum ether as the pressure conveying liquid.

### 3. Results.

As opposed to Chesnut and Phillips [13], we measured the ESR spectra as a function of temperature and pressure for two single crystal orientations, i.e. for the stacking-axis parallel and perpendicular to magnetic field direction. The line evolutions for two perpendicular orientations of  $\text{MTPP}(\text{TCNQ})_2$ , at ambient pressure, together with the line evolution of  $\text{MTPA}(\text{TCNQ})_2$  have been shown elsewhere [9]. This evolution is characteristic of the exchange interaction, a process which first removes fine structure (the exchange broadening) and then sharpens up the spectrum to a single resonance line (the exchange narrowing). The character of the line evolutions for  $\text{MTPA}(\text{TCNQ})_2$  and both salts under the hydrostatic pressure is the same as at ambient pressure. The spin susceptibility and the exchange parameters were determined from the Lorentz-shaped curves only.

The pressure and temperature evolution of the ESR spectra can be described by a pressure or temperature dependent spectroscopic coefficient of the splitting,

where  $a'$  is defined as the distance between the line and a centre of the spectrum. The temperature dependences of the  $a'$  parameters for both salts at different pressures are shown in figure 1. At high temperatures single lines are observed. As the temperature decreases below about 200 K (at ambient pressure), the line splits. The doublet components narrow and draw aside as the temperature is decreasing; the value of the splitting coefficient depends also on pressure. This dependence is very weak in high temperature phases and becomes stronger in low temperature phases; the  $\text{MTPP}(\text{TCNQ})_2$  is a good illustration of that.

The results of the linewidth studies, defined as the peak-to-peak width of the first-derivative ESR curve, as a function of pressure and temperature are summarized in figures 2a and 2b, respectively, for both salts. The linewidth displays hysteresis typical of a first-order phase transition. Similar, but temperature dependent hysteresis was found for  $\text{MTPP}(\text{TCNQ})_2$  e.g. by Iida *et al.* [10, 11] in the electrical conductivity and by Świetlik and Graja [15] in the infrared reflectivity. The pressure dependent hysteresis broadens with the increase of the transition temperature. The similar character of the linewidth evolution of  $\text{MTPA}(\text{TCNQ})_2$  against the pressure testifies to the occurrence of the pressure-induced phase transition in the salt. The appropriate phase transition diagrams are shown in figure 3.

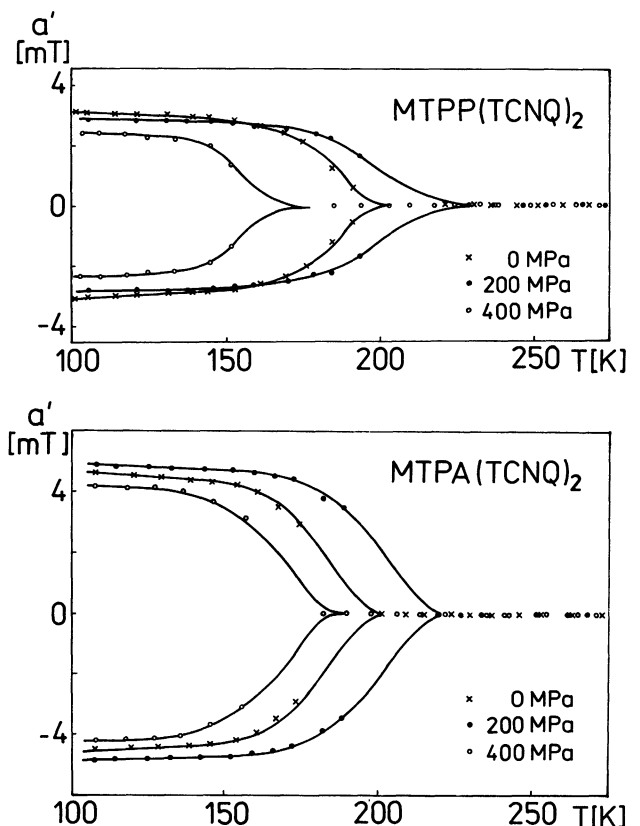


Fig. 1. — Splitting coefficients  $a'$  versus temperature at different pressures. The lines are only guides for the eye.

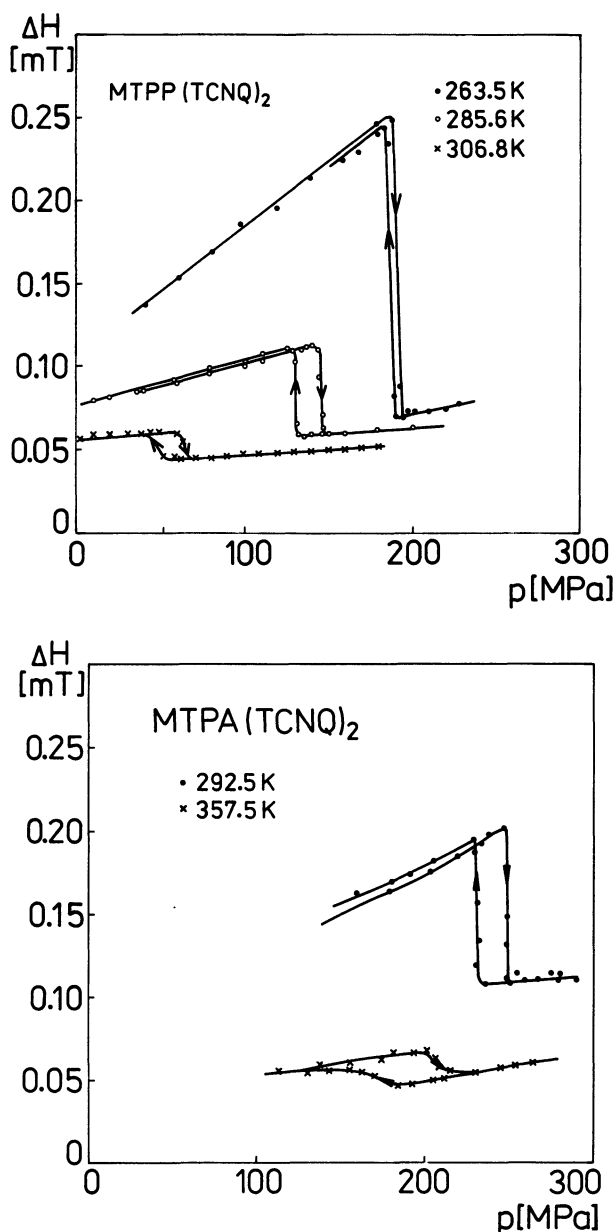


Fig. 2. — Linewidth *versus* pressure for selected temperatures. The pressure dependent hysteresis is visible.

The temperature dependences of the linewidth of both salts for two pressures : 500 MPa (above the phase transition) and 200 MPa (near or below the phase transition) are shown in figure 4. The intersections of various curves represent the points at which the line separation without exchange equals to the exchange frequency  $\nu$  [20]. To the left of this point (lower temperature), a two-line spectrum, and to the right (higher temperature) a single-line spectrum are observed. A sudden change in the linewidth for MTPP(TCNQ)<sub>2</sub> under pressure of 200 MPa, at about 255 K, is connected with the phase transition. The lack of experimental points near the intersection of the curves is due to the line deformation.

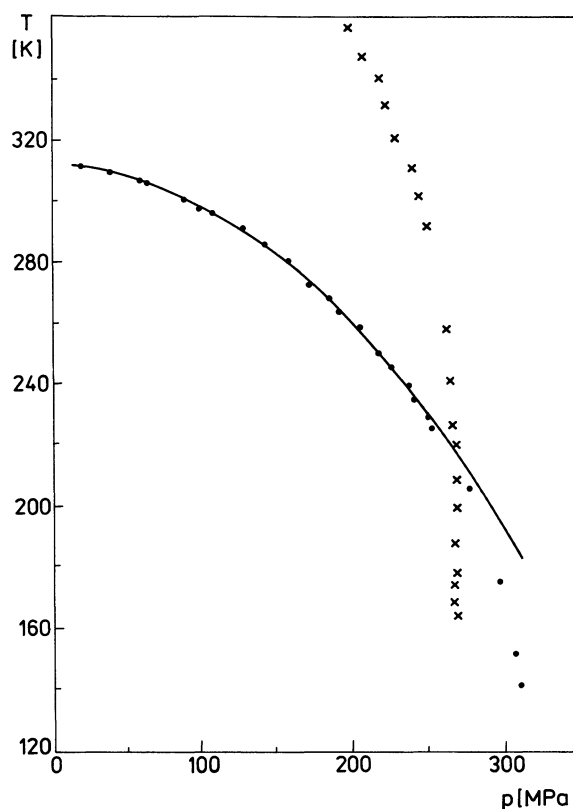


Fig. 3. — The  $p$ - $T$  phase diagrams of the salts : ● MTPP(TCNQ)<sub>2</sub>, × MTPA(TCNQ)<sub>2</sub>.

From Chesnut and Phillips [13] it is known, in the case of pairwise spin correlation with a ground singlet state and when an associated excited triplet states are separated by the energy  $E_a$ , that

$$\chi(T) \sim \frac{1}{T} (\exp(E_a/kT) + 3)^{-1} \approx \frac{1}{T} \exp(-E_a/kT) \quad (1)$$

The singlet-triplet separation energy  $E_a$  is an activation energy for triplet excitons in TCNQ salts. In figure 5 the plots  $\ln \chi.T$  vs.  $1/T$  give the activation energy values for both systems at selected pressures. At low temperatures (slow exchange) the intensity of the line is determined by equation (1). As the presence of the exchange is strongly felt by the system, the intensity goes through a maximum as can be seen in figure 5 for the samples under the pressure.

#### 4. Discussion.

Apart from the central line at the free electron resonance field, which is always observed in TCNQ salts, and which originates from structural defects or impurities, in the less conducting salts a number of lines were observed which must be due to thermally accessible triplet states. The lines are very narrow at low temperatures indicating that the excitations

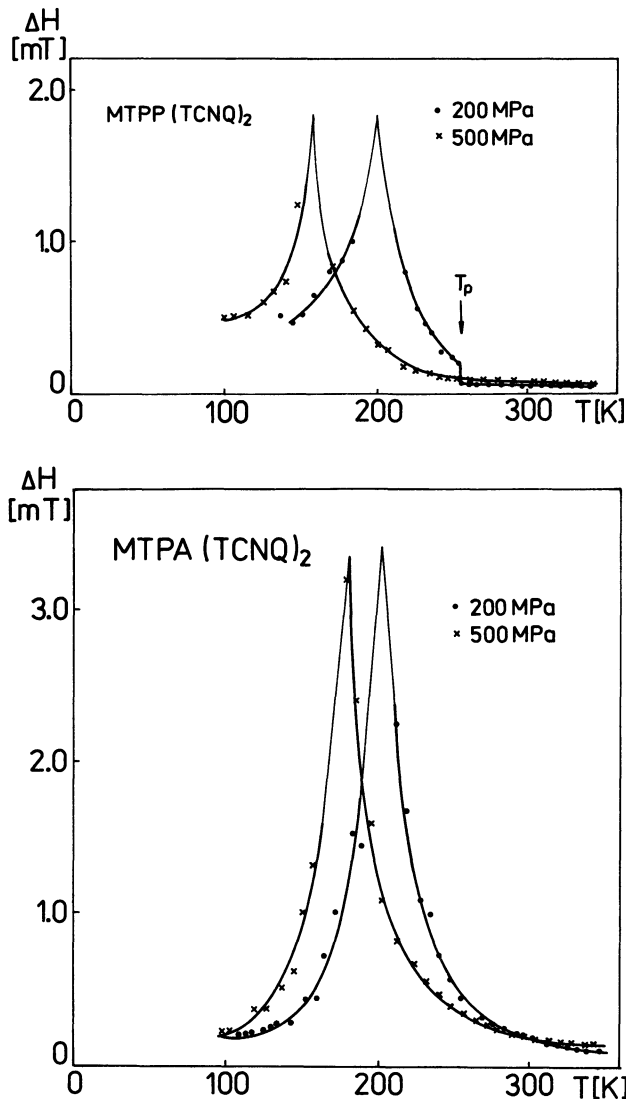


Fig. 4. — Linewidth *versus* temperature for two pressures : 500 MPa (above the phase transition) and 200 MPa (near or below the phase transition). The phase transition temperature of MTPP(TCNQ)<sub>2</sub> under the pressure of 200 MPa is marked.

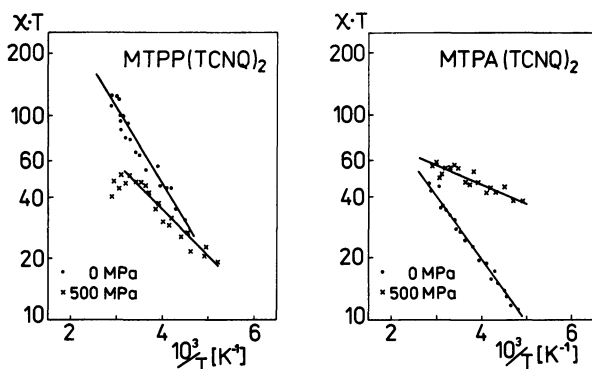


Fig. 5. — Product of triplet spin exciton intensity  $\chi$  and temperature  $T$  vs.  $1/T$  for selected pressures. Straight lines yield the singlet-triplet activation energy.

move so fast through the crystal that the hyperfine structure is averaged out. This is the case which can be seen in figure 4 at temperatures around 100 K. At higher temperatures the lines broaden and merge into a single narrow line (Figs. 1 and 4). This was explained by Jones and Chesnut [20] as an exchange broadening caused by the exciton-exciton collisions. They showed that in general, the exchange frequencies of the ion-radical salts follow the temperature dependence given by

$$\nu = \nu_0 \exp(-E_{\text{ex}}/kT) \quad (2)$$

where  $E_{\text{ex}}$  is the activation energy for spin exchange. Although the general expression describing the effects of exchange is complicated even for a simple two-line case, the limiting expressions for the linewidth  $\Delta H$  and the line separation (or twofold splitting coefficient)  $2a'$ , given in terms of the exchange frequency  $\nu$ , are quite simple. The « slow » and « fast » exchange limits are defined by the following inequalities :

$$2a'_0 \gg 2\nu/\gamma \quad (3)$$

$$2a'_0 \ll 2\nu/\gamma \quad (4)$$

where  $2a'_0$  is the line separation without exchange, and  $\gamma$  is the gyromagnetic ratio of the electron. The limiting exchange frequencies of the salts, below and above the phase transition temperatures, calculated from experimental data at ambient pressure are listed in table I.

Table I. — Limiting exchange frequencies.

Salt	Frequencies $\nu_{\text{lim}}$ [Hz]	
	$T < T_p$	$T > T_p$
MTPP(TCNQ) <sub>2</sub>	$8.4 \times 10^7$	$7.0 \times 10^7$
MTPA(TCNQ) <sub>2</sub>	$1.4 \times 10^8$	$1.3 \times 10^8$

Following the procedure proposed by Jones and Chesnut [21], the different values for the exchange frequencies are generally found to depend upon the limiting approximation employed. One can say the same about the exchange energy  $E_{\text{ex}}$ . This is a reason for the indispensable modification of the usual procedure which we propose here.

Combining certain elements of the solutions [20] one obtains the following formula :

$$\nu = \frac{\gamma}{2} [\sqrt{3}(\Delta H - \Delta H_0) + \sqrt{2\sqrt{a_0'^2 - a'^2}}] \quad (5)$$

describing the exchange frequency  $\nu$  as a function the experimental parameters  $a'$ ,  $a'_0$ ,  $\Delta H$ , and  $\Delta H_0$  where  $\Delta H_0$  is the linewidth without exchange. Equation (5) is valid at low temperatures, i.e. it is a slow exchange limit.

For the fast exchange one can use the formula [20] :

$$\nu = \frac{2\gamma}{\sqrt{3}} \frac{a_0^2}{\Delta H - \Delta H_0} \quad (6)$$

As one can see from the experimental data summarized in figures 6 and 7, the exchange frequencies are pressure and temperature dependent. According to equation (6) the exchange frequency is inversely proportional to the line broadening  $\Delta H - \Delta H_0$ , in the fast exchange limit [20]. The  $\ln(\Delta H - \Delta H_0)$  vs.  $1/T$  for both salts at selected pressure are plotted in figure 6. Jumps in the values of  $\Delta H - \Delta H_0$  for the MTPP(TCNQ)<sub>2</sub> salt, observed at temperatures 315 K and 262 K at the ambient pressure and at 200 MPa, are caused by the phase transitions. The activation energy for spin exchange  $E_{ex}$  can be calculated from the slope of the lines.

The exchange frequencies measured from the slow-exchange line collapsing and broadening (Eq. (5)) and from the fast-exchange line narrowing (Eq. (6)), are shown in figure 7 for selected temperatures as a function of pressure.

At high temperatures, e.g. at 333 K, the exchange frequencies of both salts are of the order of  $10^{11}$  Hz and decrease by a factor near 2.5 at 500 MPa. A curve  $\nu(p)$  for MTPP(TCNQ)<sub>2</sub> is smooth above 315 K because the salt is in the high-temperature phase over the whole range of pressure. The discontinuity found at MTPA(TCNQ)<sub>2</sub> near 200 MPa testifies to the pressure-induced phase transition. Really, the transition temperature at 200 MPa is 358 K [9].

For both procedures used for the evaluation of the slow-exchange frequencies, in spite of some inaccuracies caused by the nearness to the limiting exchange frequencies, large jumps are observed at critical points. This suggests a relationship between the exciton dynamics and the mechanism of the phase transformation.

From the linear dependence of the  $\ln \nu$  vs.  $p$ , shown in figure 7, it is concluded that the pressure

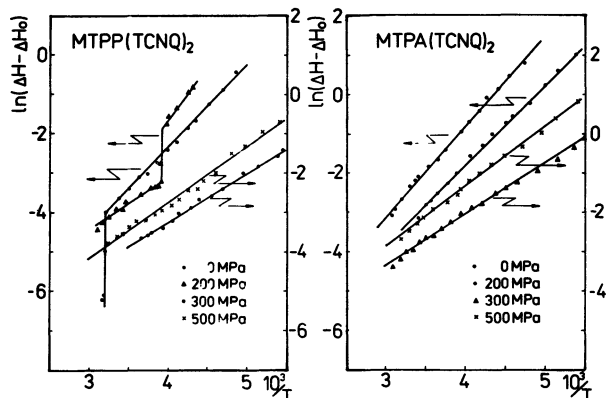


Fig. 6. — Exchange narrowing  $\Delta H - \Delta H_0$  vs.  $1/T$  for selected pressures. The lines yield the activation energy of the exchange.

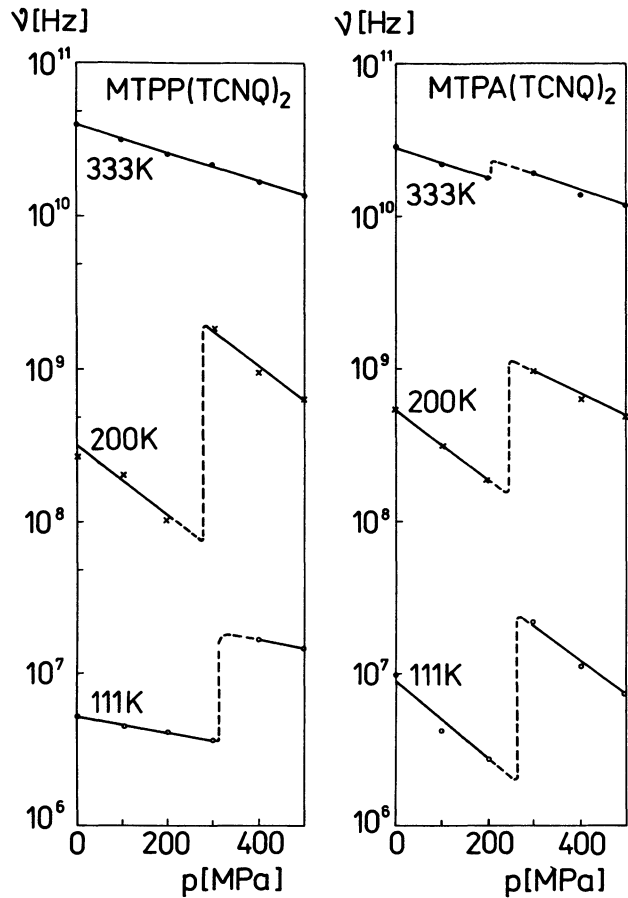


Fig. 7. — Exchange frequency  $\nu$  calculated from the fast-exchange approximation (at 333 K and 200 K) and from the slow-exchange approximation (111 K) as a function of pressure. The frequency jumps are seen at the critical pressures.

dependence of the exchange frequency can be described by the formula

$$\nu = \nu_0 \exp(-\alpha p), \quad (7)$$

where  $\nu_0$  is the exchange frequency at ambient pressure and  $\alpha$  is a temperature dependent coefficient with typical values between  $0.001 \text{ MPa}^{-1}$  and  $0.006 \text{ MPa}^{-1}$ .

For systems obeying singlet-triplet statistics, the exchange frequency varies according to equation (2). Our data confirm that  $\nu$  is activated, with pressure-dependent activation energy  $E_{ex}$ . For the fast exchange limit,  $E_{ex}$  can be determined directly from the slope of line broadening  $\Delta H - \Delta H_0$  vs.  $1/T$  (see Fig. 6). For the low exchange limit the activation energy can be determined from a slope of  $F(\Delta H, a')$  vs.  $1/T$ , where

$$F(\Delta H, a') = F_0 \exp(-E_{ex}/kT) \quad (8)$$

and  $F(\Delta H, a') \equiv \nu$  from equation (5). This procedure should eliminate any ambiguity resulting from equations given by Jones and Chesnut [20].

Table II. — Activation energies.

Pressure [MPa]	MTPP(TCNQ) <sub>2</sub> Activation energies for				MTPA(TCNQ) <sub>2</sub> Activation energies for			
	spin exchange fast $E_{ex}$ [eV]	spin exchange slow $E_{ex}$ [eV]	S-T separation $E_a$ [eV]	propagation $E_v$ [eV]	spin exchange fast $E_{ex}$ [eV]	spin exchange slow $E_{ex}$ [eV]	S-T separation $E_a$ [eV]	propagation $E_v$ [eV]
0	0.181	0.067	0.087	0.094	0.166	0.055	0.074	0.092
100	0.188	0.064	0.093	0.132	0.182	0.076	0.042	0.140
200	0.210	0.062	0.064	0.146	0.201	0.089	0.039	0.162
300	0.103	0.060	0.049	0.064	0.128	0.036	0.022	0.106
400	0.125	0.043	0.031	0.094	0.130	0.057	0.019	0.111
500	0.133	0.038	0.034	0.099	0.135	0.074	0.021	0.114

The activation energies for spin exchange  $E_{ex}$ , as determined in both regimes, as well as the singlet-triplet separation energies  $E_a$ , found from equation (1) for MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub>, are collected in table II, and shown in figures 8 and 9 as a function of pressure. We would like to emphasize that  $E_a$  was estimated from the integral absorption of Lorentz-shaped ESR curve at temperatures above 200 K. This means that comparison of  $E_a$  and  $E_{ex}$  found for the fast exchange limit, is reasonable.

If one will accept the general assumption that the propagation of the triplet exciton can involve an activated process, than the velocity  $v$  of the exciton propagation is :

$$v = v_0 \exp(-E_v/kT) \quad (9)$$

where  $E_v$  is activation energy for exciton propagation connected with left activation energies by

$$E_{ex} = E_a + E_v \quad (10)$$

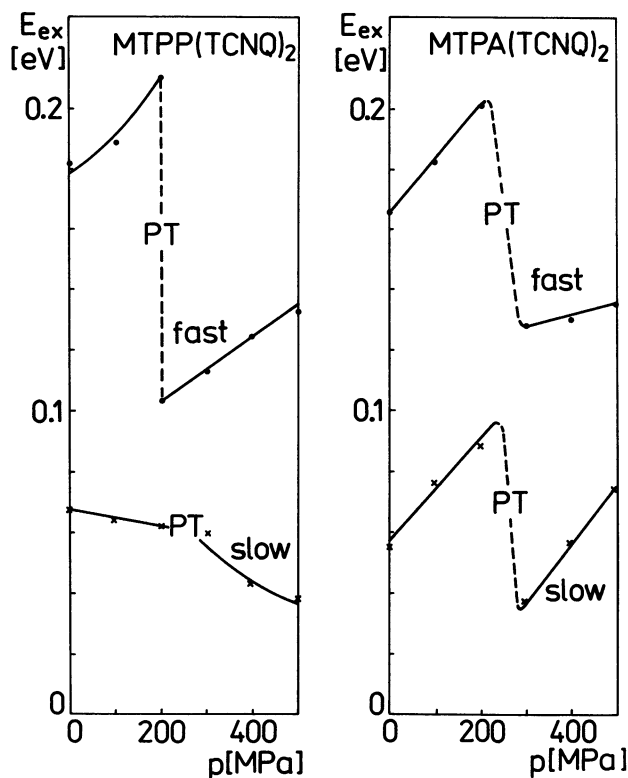


Fig. 8. — Pressure dependence of the activation energies of spin exchange  $E_{ex}$  calculated for the fast- and slow-exchange. The critical pressures are marked.

Energies  $E_v$  are also collected in table II, and are shown in figure 9 vs. pressure.

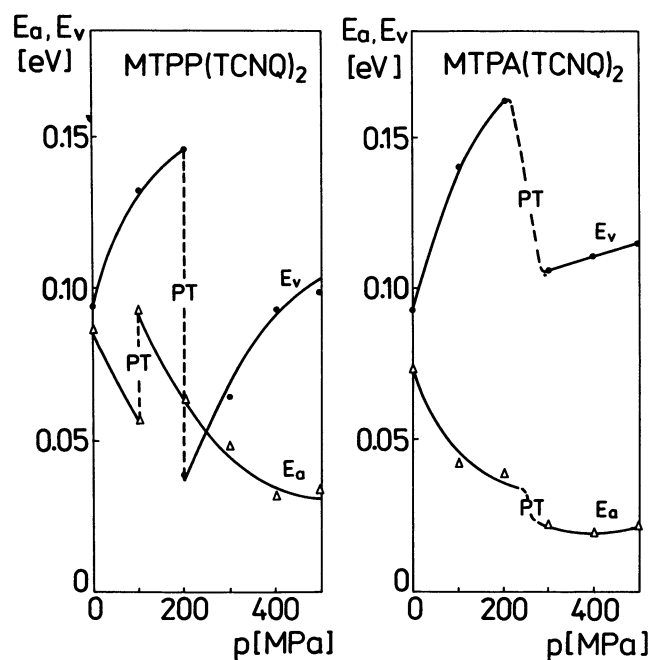


Fig. 9. — Pressure dependence of the singlet-triplet separation energies  $E_a$  and the activation energies for exciton propagation  $E_v$ . The critical pressures are marked.

All the parameters describing the ESR spectra of the salts under pressure, especially the exchange frequencies (Fig. 7) and the activation energies (Figs. 8 and 9), undergo distinct and sudden changes at phase transition points.

The activation energy for singlet-triplet separation  $E_a$  (Fig. 9) decreases with increasing pressure, and at the critical pressure undergoes a jump. The opposite direction of these jumps confirms the other nature of the phase transitions in both salts. An increase in  $E_a$  for MTPP(TCNQ)<sub>2</sub> becomes clear if one notices a decrease in the separation of TCNQ molecules between tetramers [17] above the phase transition. The interaction between the spins becomes stronger, and hence, more energy is needed to break the pairs in order to excite them into triplet states. The decreasing  $E_a$  with increasing pressure for both salts is reasonable since a decrease of the distances between the spins in the tetrads or diads reduces the energy needed to excite the triplet state. The pressure-induced phase transition in MTPA(TCNQ)<sub>2</sub> is caused by the decrease of all distances between the TCNQ molecules, both in tetrads and between them. This is why the separation between singlet-triplet states decreases not only with the increasing pressure, but also jumps above the transition point.

The dominant role of the exchange effects at the phase transition point is seen from the analysis of the activation energies for spin exchange  $E_{ex}$  given in figure 8 and for exciton propagation  $E_v$  given in figure 9.  $E_{ex}$  of both salts within the fast exchange region increases with the pressure, however, undergo large drops at critical pressures. The increase of  $E_{ex}$  and  $E_v$  and the decrease of  $\nu$  with increasing pressure

is caused by an increase of the interaction between the spins, resulting from the decreasing distances between the TCNQ molecules. One should expect that when the lattice contracts, the propagation of excitons along the chain becomes easier. Any hindrance to the propagation of excitons in both salts under pressure suggests that not only the intermolecular distances but also the intermolecular overlapping play an important role in the exciton propagation. An inconvenient overlapping affects the exciton transport along the TCNQ chain.

## 5. Conclusions.

The pressure ESR studies of MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub> salts led us to the following conclusions :

1) The exciton dynamics is considerably disturbed under pressure. The propagation of excitons is hindered, probably due to a change in overlapping of the TCNQ<sup>-</sup> orbitals. The singlet-triplet separation energy diminishes as a result of a reduction of intermolecular distances within the tetrads.

2) The phase transitions in these salts are characterized by a sudden change in the exciton parameters describing their dynamics :  $\nu$ ,  $E_{ex}$  and  $E_v$ . The observed changes suggest the delocalization of the excitons at the transition point ; i.e. establishing new molecular positions and conditions for the triplet excitons.

3) The diverse nature of the MTPP(TCNQ)<sub>2</sub> and MTPA(TCNQ)<sub>2</sub> phase transitions is probably caused by different modifications of distances between tetrads in these salts.

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