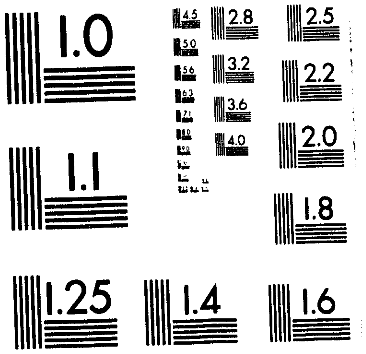
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1 of 1

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Microreflectance Infrared Study of Electron-Molecular Vibrational Coupling and Lattice Softness in BEDT-TTF Salts

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Abstract

Microreflectance infrared (MR-IR) spectra were obtained for several β- and κ-phase salts of bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET) and its deuterium analog d_8-ET by employing polarized and unpolarized light. The vibronic region of these MR-IR spectra provides information about the electron-molecular vibration (EMV) coupling, and the highest-frequency C–C–H bending vibration observed for an ET salt reflects the extent of its lattice softness. Under polarized light, the EMV coupling for an ET salt depends strongly upon how the light polarization vector is aligned with respect to the sample crystallographic axes. The lowest vibron frequency is indicative of increased EMV coupling.

Introduction

Reflection infrared spectroscopy is an important tool for characterizing organic conducting salts.[1-4] The samples of such salts are small, for the most part, and opaque. Therefore, infrared properties of organic salts are best obtained by use of a microreflectance infrared (MR-IR) technique, in which a microscope is interfaced with an FT-IR spectrometer. In the salts of bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET, see Table 1), the C-H bonds of ET form numerous C–H···anion contacts as shown in Figure 1. The C–H···anion interactions of ET salts have been studied with the MR-IR technique.[5-7] The MR-IR spectra of ET salts provide "fingerprints" of their C-H···anion contact environments, and have been employed to distinguish between α- and β-phase salts of ET[6] and to follow the thermal conversion of α-$(ET)_2I_3$ to α_t-$(ET)_2I_3$.[7]

-Figure 1 here-

In the present work we obtain room-temperature MR-IR spectra of several β- and κ-phase ET salts by employing polarized and unpolarized light. On the basis of the MR-IR spectra we discuss the electron-molecular vibration (EMV) coupling[8-16] and the lattice softness[17-21] of these salts.

Experimental

Room temperature MR-IR measurements on ET salts were obtained with a Digilab FTS-40 purged spectrometer interfaced (typically 256 scans per spectrum) with a UMA-300-A microscope and a Cadmium-Mercury-Telluride detector (resolution at 4 cm^{-1}). The reflectance mode was used because the single crystals are opaque. A Kramers-Kronig transformation was applied to all reflectance spectra, and the results are reported in absorbance units. Care is taken to use a crystal that demonstrates predominately reflectance characteristics and little, if any, transmission.

Structure of Donor-Molecule Layer

Figures 2 a-c show the packing patterns of ET molecules in the donor molecule layers of β- and κ-phase salts. In both phases, the π-framework of each ET molecule is not perpendicular but inclined to the donor layer. In β-$(ET)_2X$ (X = I_3, AuI_2, IBr_2) the ET molecules cant along the donor-stack direction (i.e., approximately along the a+c direction of Figure 2a).[22] In κ-$(ET)_2Cu[N(CN)_2]X$ (X = Cl, Br, I)[21,23,24] the donor layer defines the *ac*-plane (Figure 2b), and in κ-$(ET)_2Cu(NCS)_2$ the *bc*-plane (Figure 2c).[25] The donor molecules of κ-$(ET)_2Cu[N(CN)_2]X$ cant along the *a*-direction, which is parallel to the $Cu[N(CN)_2]X^-$ anion chain direction,[19,20,23,24] while those of κ-$(ET)_2Cu(NCS)_2$ cant along the *c*-direction, which is perpendicular to the $Cu(NCS)_2^-$ chain direction.[19,25] As far as donor-molecule layers are concerned, the *b*- and *c*-axes of κ-$(ET)_2Cu(NCS)_2$ correspond to the *c*- and *a*-axes of κ-$(ET)_2Cu[N(CN)_2]X$, respectively.

In our MR-IR study of ET salts with polarized light, it is necessary to specify the alignment of the light polarization vector with respect to the crystallographic axes of the salts. For β-$(ET)_2X$, κ-$(ET)_2Cu[N(CN)_2]X$, and κ-$(ET)_2Cu(NCS)_2$ we define the alignment angle θ to be zero when the light polarization vector is parallel to the *a*-, *c*-, and *b*-axis directions, respectively. Therefore, the donor molecules cant approximately along the $\theta=90°$ direction in β-$(ET)_2X$, and along the $\theta=0°$ direction in κ-$(ET)_2Cu[N(CN)_2]X$ and κ-$(ET)_2Cu(NCS)_2$.

Results and Discussion

A. Vibronic Envelope and Electron-Molecular Vibration Coupling

As representative examples, Figures 3a and 3b show the MR-IR spectra of κ-$(ET)_2Cu(NCS)_2$ and κ-$(d_8$-$ET)_2Cu(NCS)_2$, respectively. The MR-IR spectra of ET salts have two characteristic features. One is a broad band extending from the infrared to ~3000 cm^{-1} region (i.e., plasma region), and is attributed to inter- and intramolecular electronic transitions[16,26-28] associated with the ET molecules. The other feature is an envelope in the 1200-1450 cm^{-1} region (i.e., vibronic region), which is attributed to the coupling of conduction electrons to totally symmetrical vibrations of ET molecules.[8-16] The vibronic region is more diagnostic than the plasma region in characterizing ET salts.

The C=C stretching ($\upsilon_{c=c}$) vibrations of ET, being totally symmetric, are Raman-active but infrared-silent. For symmetrical neutral donor molecules with a central C=C bond, the $\upsilon_{c=c}$ vibrations are observed at 1500-1550 cm^{-1}. Upon forming salts, the $\upsilon_{c=c}$ modes of the donor molecules, being lowered in symmetry, interact with the conduction electrons so that the $\upsilon_{c=c}$ modes become infrared-active and are shifted to lower frequencies thereby leading to the vibronic envelopes. Table 1 lists the $\upsilon_{c=c}$ frequencies observed for several neutral donor molecules and the center positions of the vibronic envelopes (hereafter referred to as the vibronic frequencies) observed for their salts. With respect to the $\upsilon_{c=c}$ values, the vibronic frequencies are considerably shifted to lower values in the salts. The magnitude of this shift is a measure of the strength of the electron-molecular vibration (EMV) coupling in a given salt.[13-15]

When an ET salt has C-C-H bending vibrations (see below) that overlap with the vibronic envelope, it is difficult to determine the vibronic frequency. In such a case, the latter can be determined from the MR-IR spectrum of the corresponding d_8-ET salt, because the C-C-D bending vibrations shift beyond the vibronic envelope (see Figures 3a and 3b).[29] Table 2 lists the vibronic frequencies observed for several β- and κ-phase salts of d_8-ET with the use of unpolarized light.

Figure 4 shows the vibronic envelopes of β-$(d_8\text{-ET})_2I_3$ observed by use of polarized light with the θ=0°, 45° and 90° alignments. The vibronic frequency is lowest, and the vibronic absorption is strongest, when θ=90°. As already noted, the light polarization vector is parallel to the donor-molecule canting direction in β-$(ET)_2I_3$ when θ=90°. Therefore, the stretching vectors of the central C=C bond vibration are contained approximately in the θ=90° plane (Figure 2a). This provides an optimum arrangement of the donor molecules for their EMV coupling, which leads to the lowest vibronic frequency and the strongest vibronic absorption. Figure 5 shows the vibronic envelopes of κ–$(d_8\text{-}ET)_2Cu[N(CN)_2]Br$ observed by use of polarized light. The vibronic frequency is lowest, and the vibronic absorption is strongest, when θ = 0°. This observation is common to all other κ-$(d_8\text{-}ET)_2Cu[N(CN)_2]X$ and κ-$(d_8\text{-}ET)_2Cu(NCS)_2$ salts examined in the present work. This finding is understandable, because the light polarization vector is parallel to the donor-molecule canting direction for the κ-phase salts when θ = 0°.

Table 3 summarizes the vibronic frequencies of several β- and κ-phase salts of d_8-ET obtained by use of polarized light with the θ = 0°, 45° and 90° alignments. The polarization dependence of the vibronic frequency is substantial, and this arises from the fact that the EMV coupling is optimum when the light polarization vector is parallel to the stretching vector of the donor-molecule central C=C bond, and that the donor molecules of the β- and κ-phase salts are inclined with respect to their donor-layer planes and hence cant along a certain direction.

B. C-C -H Bending Vibrations and Lattice Softness

In the vibronic envelopes of ET salts, vibrational structure often occurs due to the ethylene group C-C-H bending vibrations (See Figure 3a). Normally, the C-H stretching ($\upsilon_{C\text{-}H}$) vibrations are more intense than the C-C-H bending vibrations.[30] However, the bending vibrations occur at the same energy as the vibronic envelope and are resonance-enhanced,[31,32] so that they are more intense than the υ_{C-H} vibrations. In the MR-IR spectra of ET salts, the υ_{C-H} vibrations are very weak and obscured by the broad band of the plasma region.

The C-C-H bending vibrational features may be absent when the C-H···anion contact interactions are such that the bending frequencies occur outside the vibronic envelope[33] or when the bending frequencies are lowered below the vibronic region by isotope substitution as in d_8-ET salts.[29] In such a case, there is no resonance-enhancement of the C-C-H bending vibrational modes, and thus their intensities are weak.

Because the C-C-H bending vibrations in ET salts occur under the constraint of the C-H···anion and the C-H···donor contacts[22] (See Figure 1), they may reflect the lattice softness.[17-21] When a C-H···anion interaction is less strained (i.e., softer), its C-C-H bending frequency is expected to be lower. Since each ET molecule has 8 C-H bonds, there are a number of C-C-H bending frequencies which must be considered. Table 4 lists the C-C-H bending frequencies of β- and κ-phase hydrogenic salts of ET observed from their vibronic envelopes by use of unpolarized light at room temperature. Of several C-C-H bending frequencies observed for a given ET salt, the most relevant with respect to the lattice softness is the highest-frequency vibration because it represents the "most-strained" C-C-H bending mode and hence corresponds to the shortest C-H···anion distance. The absence of superconductivity in β-$(ET)_2I_2Br$[22] and κ-$(ET)_2Cu[N(CN)_2]I$[20] is attributed to the random potentials associated with their structural disorder. For the ET salts without such random potentials, Table 4 shows that the highest C-C-H bending frequency is lower than 1320 cm^{-1} for the superconductors, but higher than 1320 cm^{-1} for the nonsuperconductors. Thus, the superconductors can be considered to have a softer lattice than do the nonsuperconductors.

Concluding Remarks

Our study suggests that, on a qualitative level, the extent of the EMV coupling and the lattice softness of ET salts can be deduced from their MR-IR spectra by utilizing the vibronic region. Under polarized light, the EMV coupling depends sensitively upon how the light polarization vector is aligned with respect to the crystallographic axes of the crystals under study. This is due to the fact that, in a given ET salt, the donor molecules cant along a certain direction and hence the EMV coupling is optimum when the light polarization vector is parallel to that direction. The C-C-H bending vibrations found in ET salts occur under the constraint of the C-H···anion and C-H···donor interactions present in the crystals. Thus, the highest-frequency C-C-H bending mode observed to ET salts in the vibronic region reflects the extent of their lattice softness: the lower the frequency, the softer the lattice. For ET salts which do not possess structural disorder, the highest C-C-H bending frequency is lower than 1320 cm^{-1} for the superconductors, but higher than 1320 cm^{-1} for the nonsuperconductors.

Acknowledgments

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Table 1 Comparison of the $\upsilon_{c=c}$ frequencies of neutral donor molecules[a] with the vibronic frequencies of their salts (cm^{-1})

Donor	$\upsilon_{c=c}$	Salt	Vibronic frequency
TTF[b]	1518	(TTF)Br[c]	1368
TMTSF[d]	1539	$(TMTST)_2ReO_4$[e]	1415
TMTTF[d]	1538	(TMTTF)Br[d]	1340
ET	1511	κ-$(ET)_2Cu(NCS)_2$	1290

TMTTF is the
sulfur analogue
of TMTSF

[a]

[b] Ref. 34

[c] Ref. 35

[d] Ref. 36

[e] Ref. 37

Table 2 Vibronic frequencies of several β- and k-phase salts of d_8-ET obtained from their MR-IR spectra with unpolarized light[a]

Salt	Vibronic frequency (cm^{-1})
β-(d_8-ET)$_2$AuI$_2$	1290
β-(d_8-ET)$_2$I$_3$	1263
β-(d_8-ET)$_2$I$_2$Br	1288
β-(d_8-ET)$_2$ICl$_2$	1317
κ-(d_8-ET)$_2$Cu[N(CN)$_2$]Cl	1278
κ-(d_8-ET)$_2$Cu[N(CN)$_2$]Br	1288
κ-(d_8-ET)$_2$Cu[N(CN)$_2$]I	1276
κ-(d_8-ET)$_2$Cu(NCS)$_2$	1283

[a] To obtain accurate vibronic frequencies, unpolarized light was used because the reflectance of these salts is sensitive to polarized light and frequency.

Table 3 Polarization dependence of the vibronic frequencies observed for β-and κ-phase salts of d_8-ET

	Vibronic frequency (cm^{-1})		
Salt	$\theta = 0°$	$\theta = 45°$	$\theta = 90°$
β-$(d_8\text{-ET})_2I_3$	1291 (1.15)[a]	1288 (1.62)	1275 (14.58)
β-$(d_8\text{-ET})_2IBr_2$	1302 (0.955)	1281 (1.24)	1275 (23.91)
κ-$(d_8\text{-ET})_2Cu[N(CN)_2]Cl$	1285 (1.72)	1309 (1.31)	1322 (1.53)
κ-$(d_8\text{-ET})_2Cu[N(CN)_2]Br$	1224 (8.33)	1261 (5.39)	1295 (5.17)
κ-$(d_8\text{-ET})_2Cu[N(CN)_2]I$	1218 (3.71)	1255 (3.46)	1308 (2.84)
κ-$(d_8\text{-ET})_2Cu(NCS)_2$	1214 (7.43)	1238 (5.22)	1297 (5.70)

[a] Numbers in parenthesis are in absorption units.

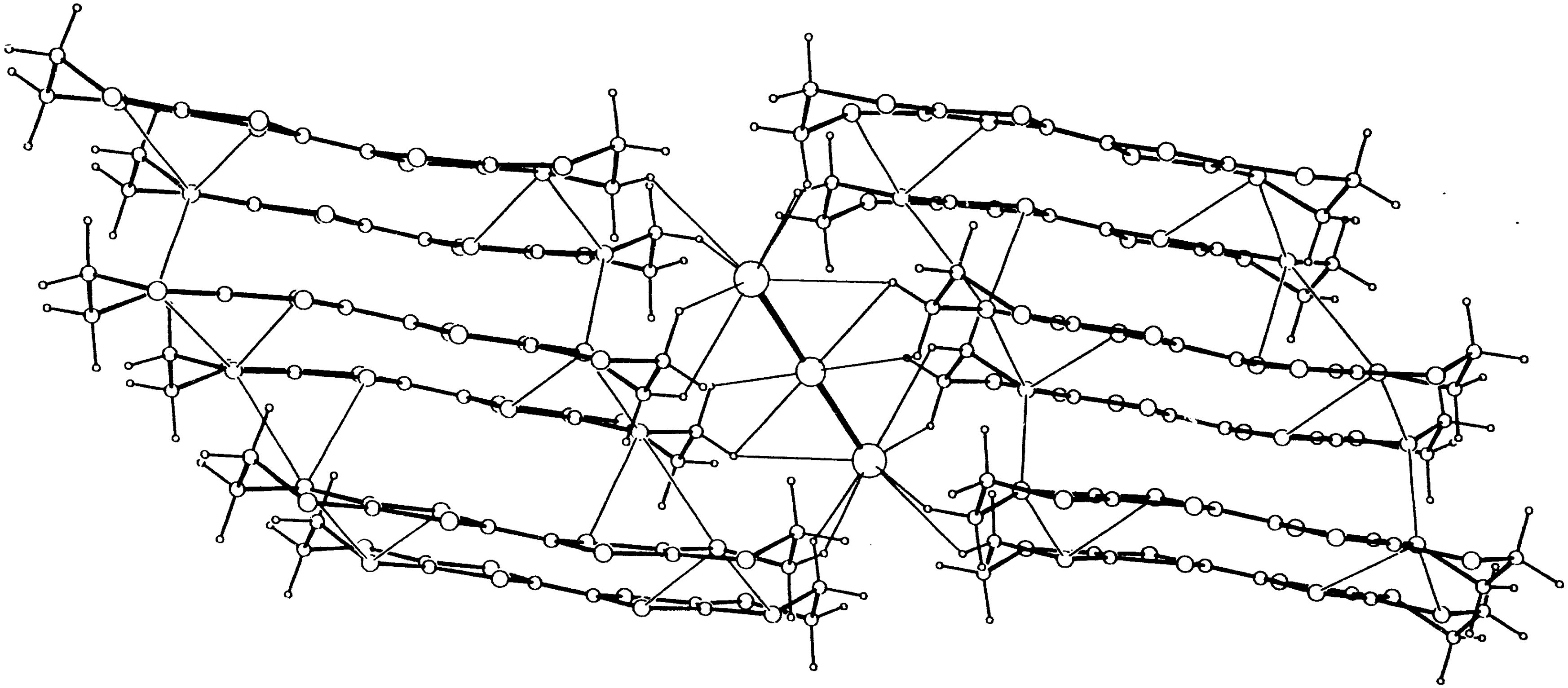
Table 4 C-C-H bending frequencies of some hydrogenic ET salts observed from their MR-IR spectra by employing unpolarized light and their superconductivity behavior

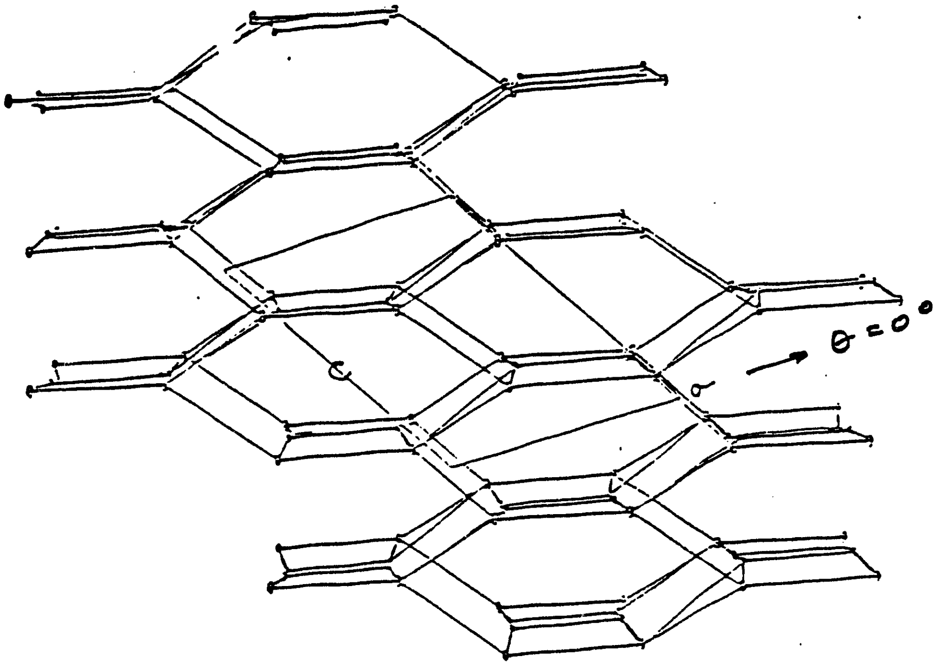
Salt	C-C-H bending frequency (cm^{-1})	Superconductor at Liquid H_e Temperatures
β'-$(ET)_2ICl_2$	1343, 1269	No
β-$(ET)_2I_2Br$	1315, 1255	No
β-$(ET)_2IBr_2$	1317, 1260	Yes
β-$(ET)_2I_3$	1300, 1260	Yes
β-$(ET)_2AuCl_2$	1338, 1265	No
β-$(ET)_2AuBr_2$	1324, 1271	No
β-$(ET)_2AuI_2$	1310, 1265	Yes
κ-$(ET)_2Cu(NCS)_2$	1316, 1267, 1250	Yes
κ-$(ET)_2Cu[N(CN)_2]Cl$	1313, 1268, 1250	Yes (under 0.3 kbar)
κ-$(ET)_2Cu[N(CN)_2]Br$	1297, 1259, 1247	Yes
κ–$(ET)_2Cu_2(CN)_3$	1297, 1258, 1229	Yes (1.5 kbar)
κ-$(ET)_2Cu[N(CN)_2]I$	Not available[a]	No

[a] Due to poor crystal quality.

Figure Captions

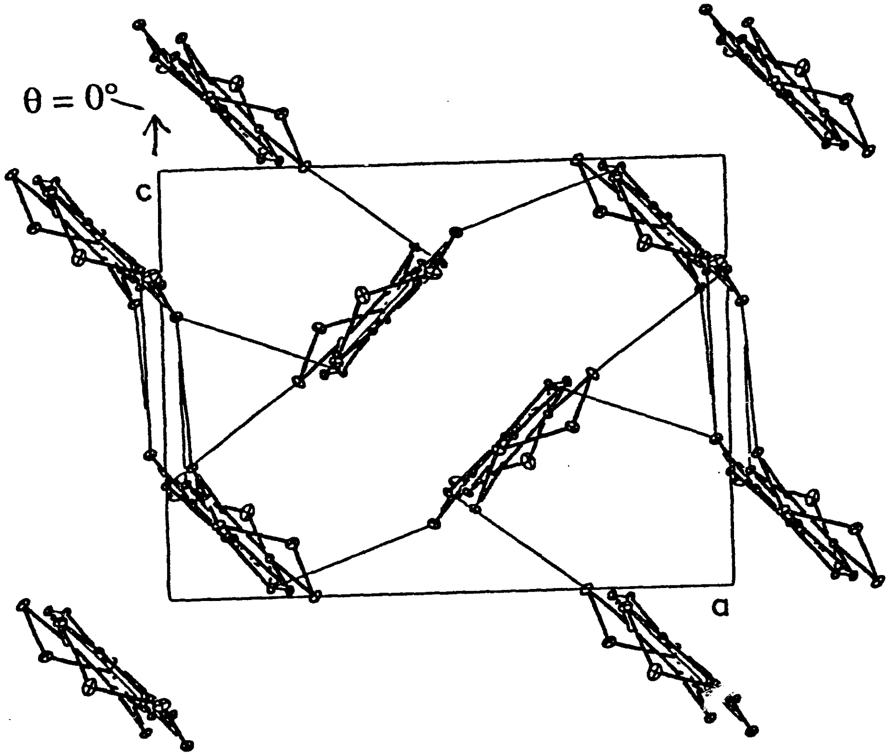
Figure 1	Arrangement of ET molecules around the IBr_2^- anions in β-$(ET)_2IBr_2$ (Taken from Ref. 38).
Figure 2	Packing pattern of ET molecules in (a) β-$(ET)_2X$ (X = I_3, AuI_2, IBr_2), b) κ-$(ET)_2Cu[N(CN)_2]X$ (X = Cl, Br, I), and (c) κ-$(ET)_2Cu(NCS)_2$. The definition for the θ = 0° alignment for the polarized light reflectance measurements is also shown.
Figure 3	MR-IR spectra of (a) κ-$(ET)_2Cu(NCS)_2$ and (b) κ-$(d_8\text{-}ET)_2Cu(NCS)_2$ obtained with unpolarized light.
Figure 4	MR-IR spectra of β-$(ET)_2I_3$ obtained by using polarized light with the alignment angles θ = 0°, 45° and 90°.
Figure 5	MR-IR spectra of κ-$(ET)_2Cu[N(CN)_2]Br$ obtained by using polarized lights with the alignment angles θ = 0°, 45° and 90°.





(a)

Figure 2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c
a

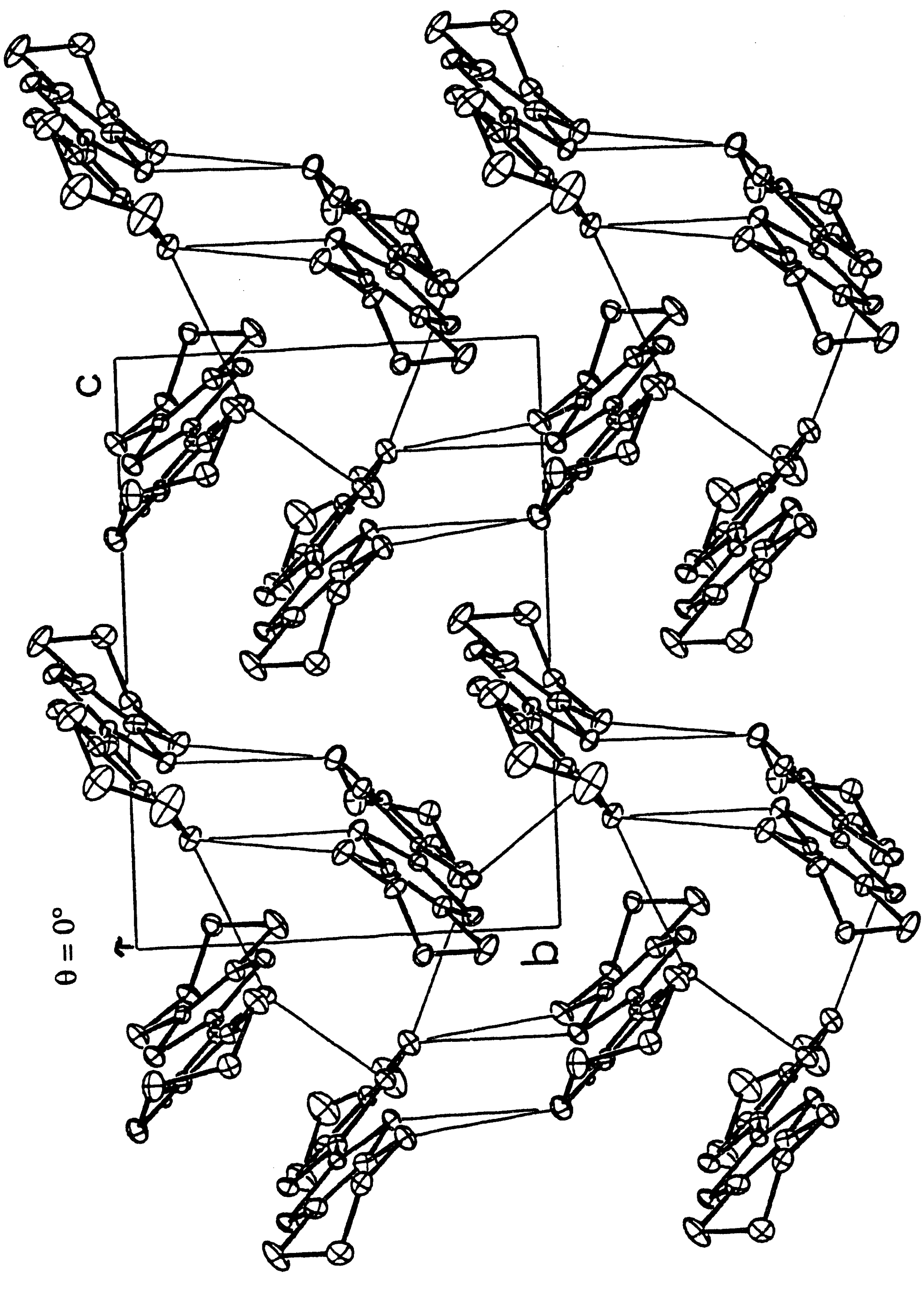
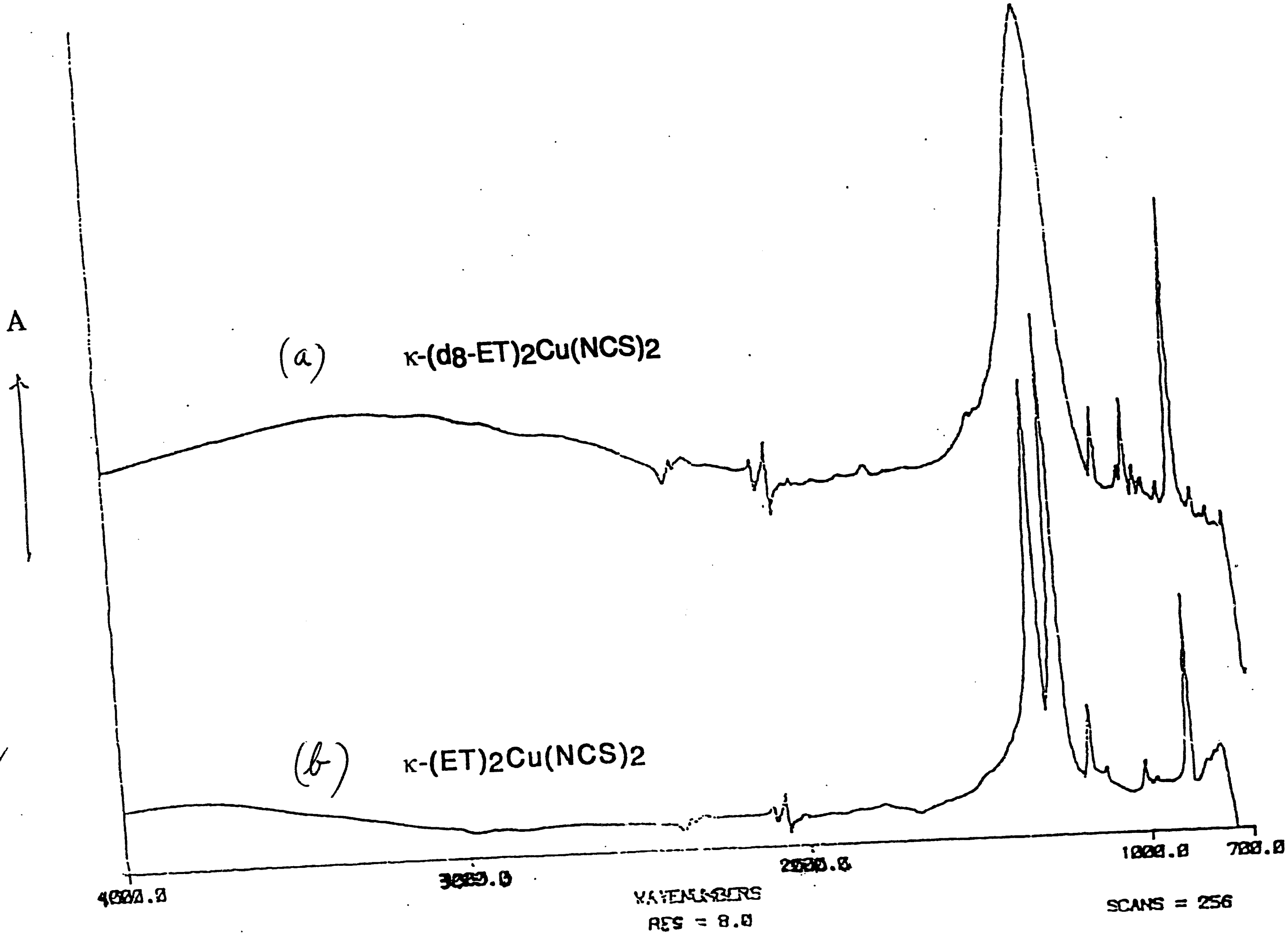
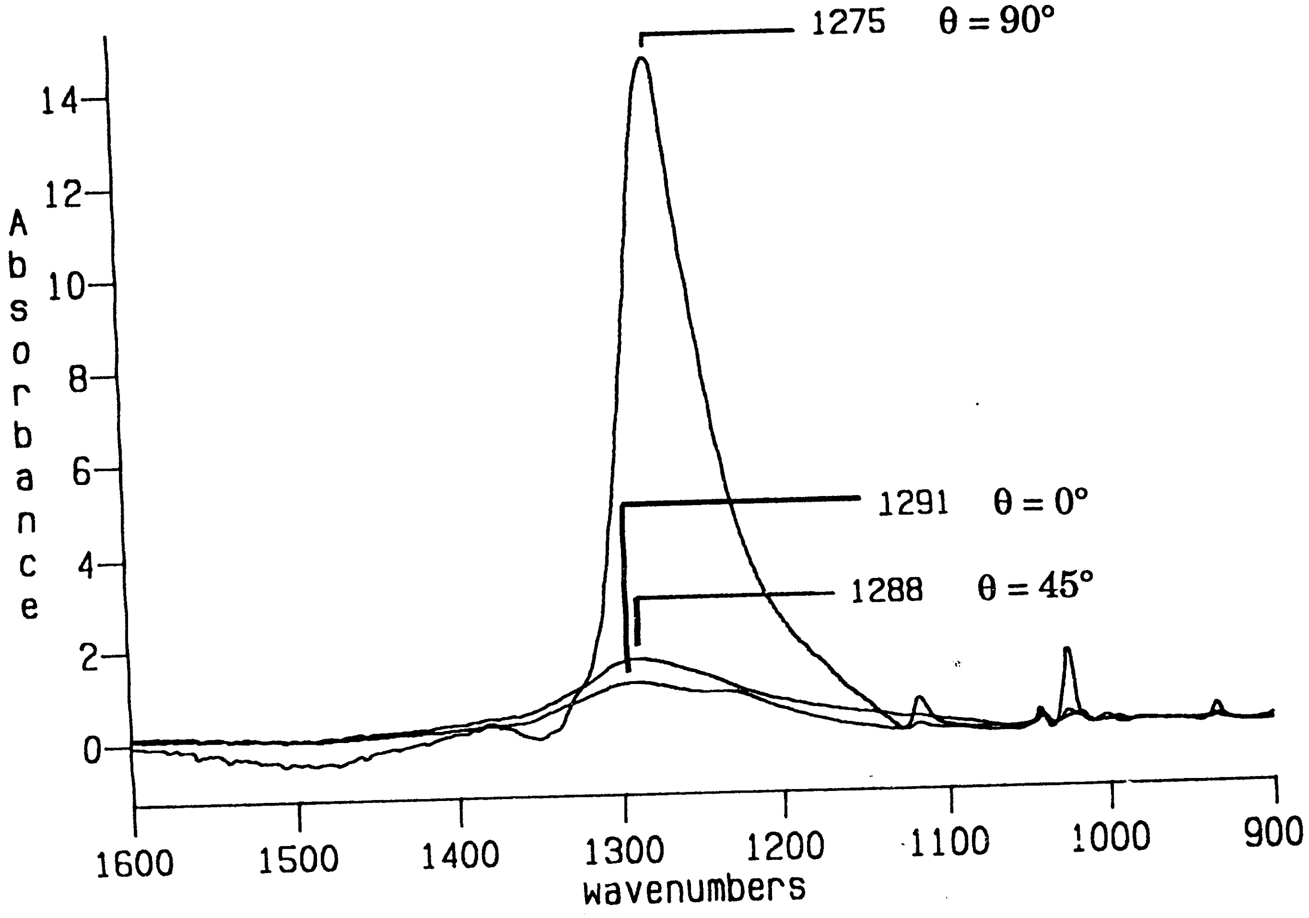
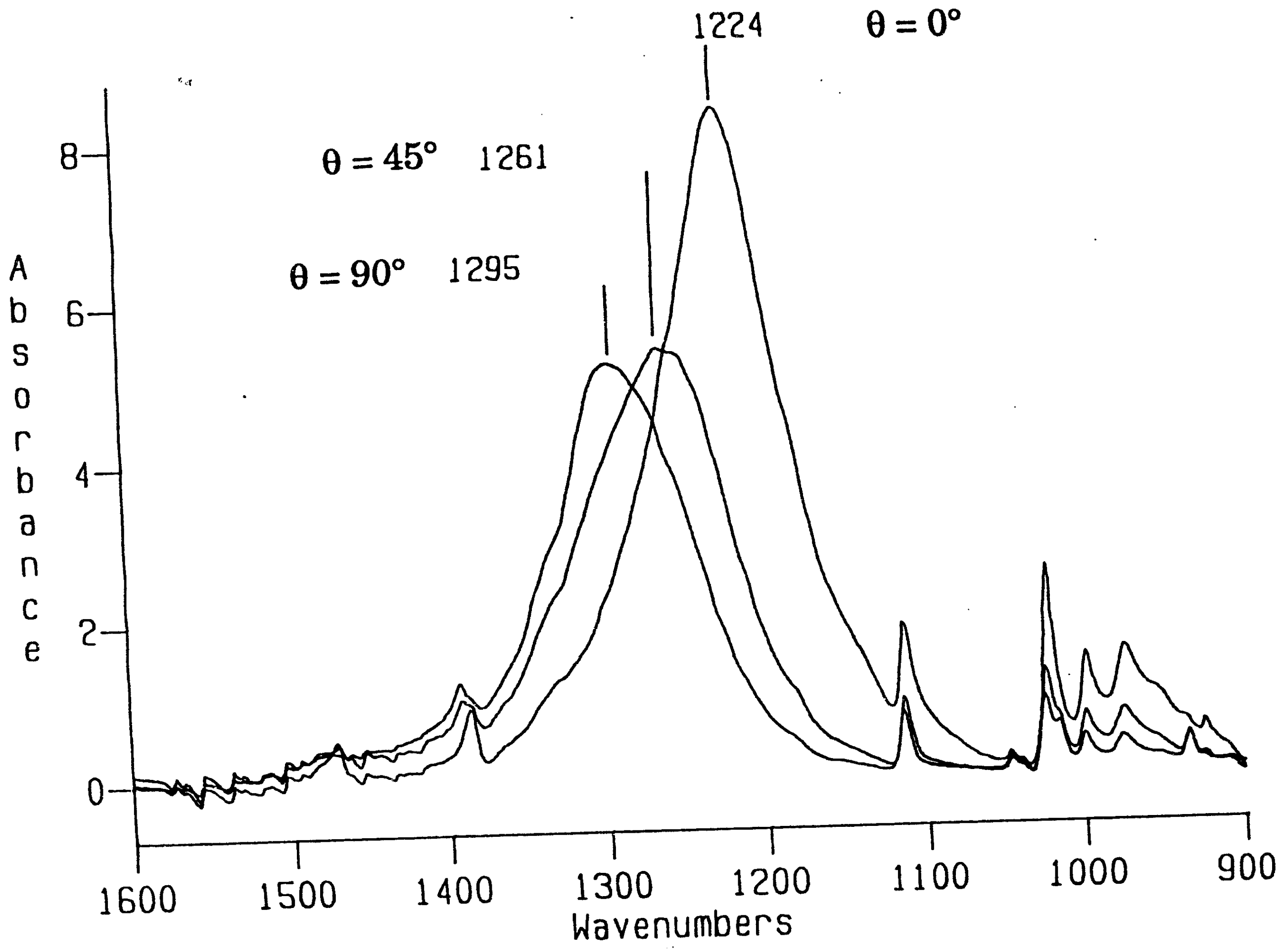


Fig 9

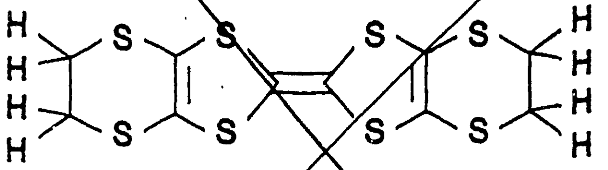

A
(a) κ-(d8-ET)2Cu(NCS)2
(b) κ-(ET)2Cu(NCS)2
4000.0
3000.0
2000.0
1000.0
700.0
WAVENUMBERS
RES = 8.0
SCANS = 256


1275 θ = 90°
1291 θ = 0°
1288 θ = 45°
Absorbance
0
2
4
6
8
10
12
14
1600
1500
1400
1300
1200
1100
1000
900
wavenumbers

β-(d8ET)2I3



κ-(d8ET)2Cu[N(CN)2]Br



~~BEDT-TTF (ET)~~

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