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INFRARED AND FAR INFRARED PROPERTIES OF SOME β-(BEDT-TTF)$_2$X COMPOUNDS

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ABSTRACT

Infrared and far infrared polarized reflectance spectra of β-(BEDT-TTF)$_2$X ($X = I_2Br^-$, AuI_2^-) are presented. Both have, as in the case of $X = I_3^-$, the strongest metallic character along the stacking axis, but two-dimensional plasmon behavior is found at low temperatures in all the materials. The distribution of oscillator strength is discussed with special emphasis on the non-Drude features.

INTRODUCTION

The class of organics denoted β-(BEDT-TTF)$_2$X (in short (ET)$_2$X) comprises several ambient pressure superconductors (see, e.g., [1,2] and these proc.) The crystal structure is characterized by strongly interacting, dimerized stacks of ET-molecules organized in sheets in the a-b plane with layers of inorganic ions in between [3]. High conductivity is found in the a-b plane ($\sigma_{DC}(300K) \simeq 30\Omega^{-1}cm^{-1}$) [1]. The 2D nature of ET-compounds has been confirmed by a number of infrared studies [4-9]. It is the purpose of this paper to present and discuss data on β-(ET)$_2$X, $X = AuI_2^-$ and I_2Br^-. The former material is an ambient pressure superconductor with $T_c \sim 5K$ [2], while the latter appears to have a metallic ground state (possibly because of counterion disorder) [10].

RESULTS AND DISCUSSION

The results presented here are based on measurements on hexagon shaped crystals in the ranges 60-700 cm^{-1} (Fourier transform spectroscopy) and 400-20000 cm^{-1} (dispersive spectroscopy).

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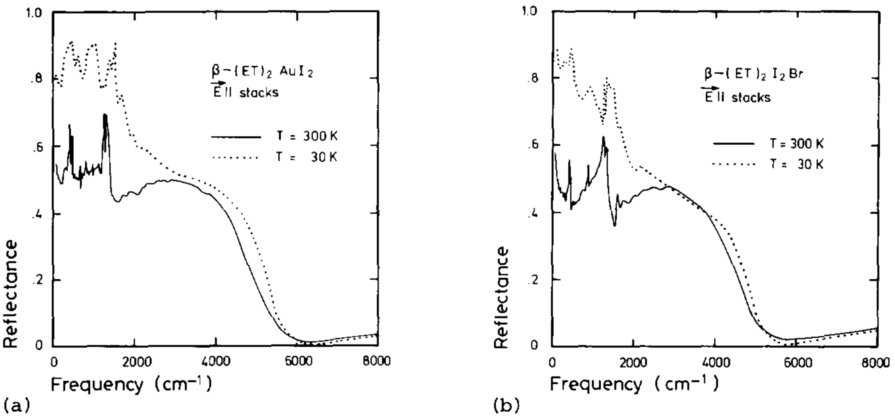
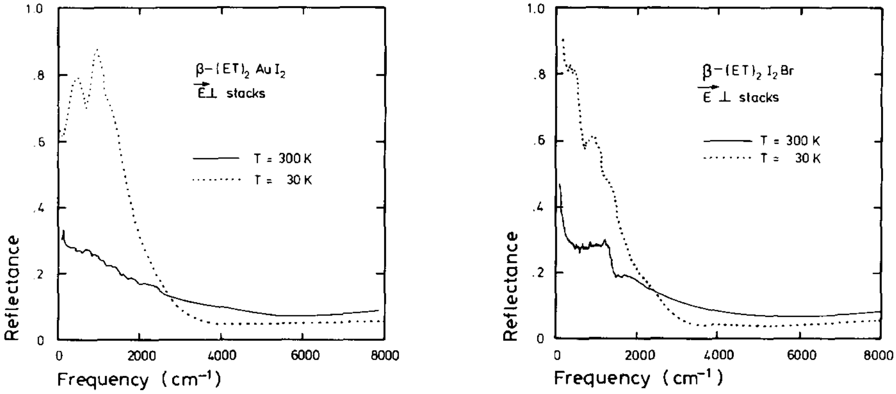


(a) (b)

Fig. 1. Stacking axis reflectance of (a) β-$(ET)_2AuI_2$, and (b) β-$(ET)_2I_2Br$ at two temperatures: T = 300K and 30K.

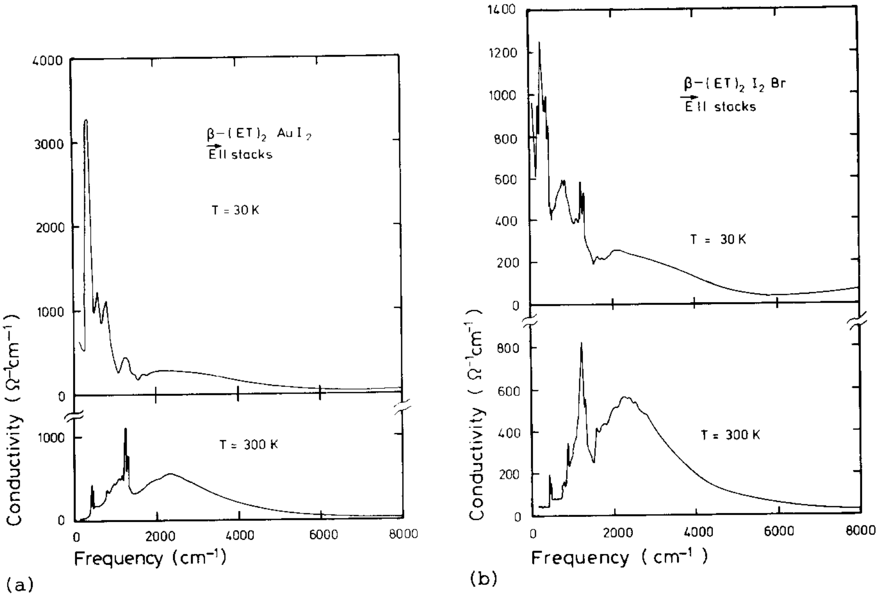
Fig. 1 presents stacking axis reflectance in the infrared for the two materials at T = 300K and 30K. Common features include a plasma edge at 5000 cm^{-1}, which sharpens on cooling, a mid-infrared reflectance level which shows a strong increase on cooling, and pronounced non-Drude structure in the range of molecular vibrations. Differences between the two materials are 1) a higher position for the plasma edge in the AuI_2^- material, and 2) in the same compound a higher level below 2000 cm^{-1} at 30K. In Fig. 2 we show corresponding results for $\bar{E}\perp$ stacks in the a-b plane. The 300K data show overdamped plasmon behavior, while at 30K a fairly well-defined plasma edge appears near 2000 cm^{-1}. Again non-Drude features are evident, being most pronounced in $(ET)_2I_2Br$ at 300K, but in $(ET)_2AuI_2$ at 30K.

Using data to about 20000 cm^{-1} and suitable extrapolation procedures, the complex dielectric function has been determined by Kramers-Kronig transformation.



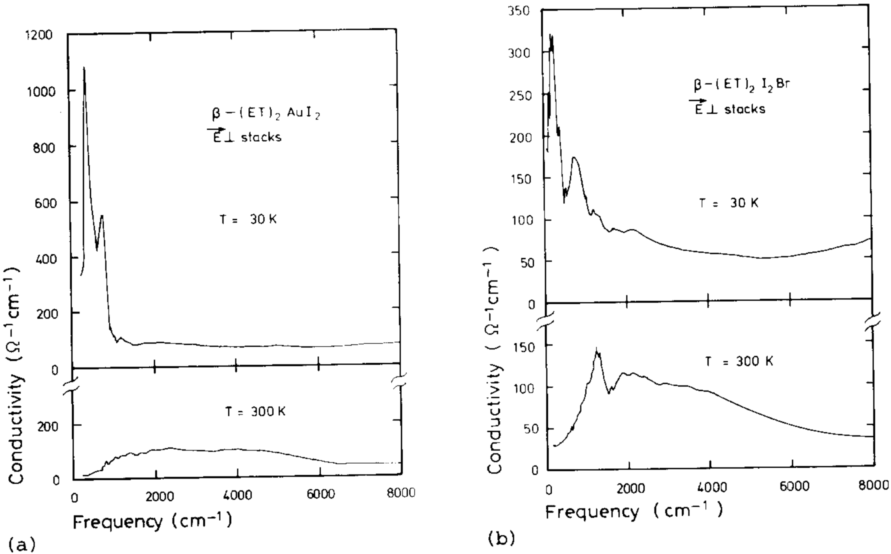
(a) (b)

Fig. 2. Reflectance perpendicular to the stacks of (a) β-$(ET)_2AuI_2$, and (b) β-$(ET)_2I_2Br$ at two temperatures: T = 300K and 30K.



(a) (b)

Fig. 3. Frequency dependent conductivity of (a) β-$(ET)_2AuI_2$, and (b) β-$(ET)_2I_2Br$ at two temperatures: T = 300K and 30K. E∥ stacks.



(a) (b)

Fig. 4. Frequency dependent conductivity of (a) β-$(ET)_2AuI_2$, and (b) β-$(ET)_2I_2Br$ at two temperatures: T = 300K and 30K. E⊥stacks.

The frequency dependent conductivity, σ, is shown in Figs. 3+4 for $\bar{E}$∥ and ⊥ to stacks respectively. A common feature is evident in all these results: At room temperature σ shows a broad peak around 2200 cm^{-1} with vibrational structure on the low frequency side. At low temperature the oscillator strength moves down in frequency so the spectra become more Drude-like. Although there is still sharp fine structure, σ peaks well below 500 cm^{-1}. The corresponding behavior in the real part of $\tilde{\varepsilon}$ is illustrated in Fig. 5. Note that despite the drastic changes at low frequencies, the high frequency zero crossing (i.e. the plasmon frequency) shows only a temperature dependence consistent with effects of thermal contraction. Thus we believe that the behavior at low frequency is irrelevant for a determination of one-electron band structure parameters from plasma frequencies. We have therefore performed Drude-model fits in limited spectral ranges around the plasma edges and parameterized the results in terms of average transfer integrals along and perpendicular to the stacks; these parameters are given in Table 1.

One conceivable explanation for the temperature dependence of σ is that the carriers may be localized on the dimers on the time scale of spectroscopy at 300K. The mean free path is indeed very short. Then the 2200 cm^{-1} band could be the intradimer excitation. This is a localized version of the interpretation by Tajima et al. [4,5]. At low temperature the self-screening and thermal contraction promotes a more Drude-like spectrum from the electron gas.

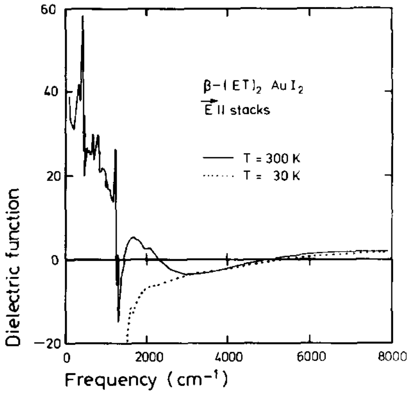


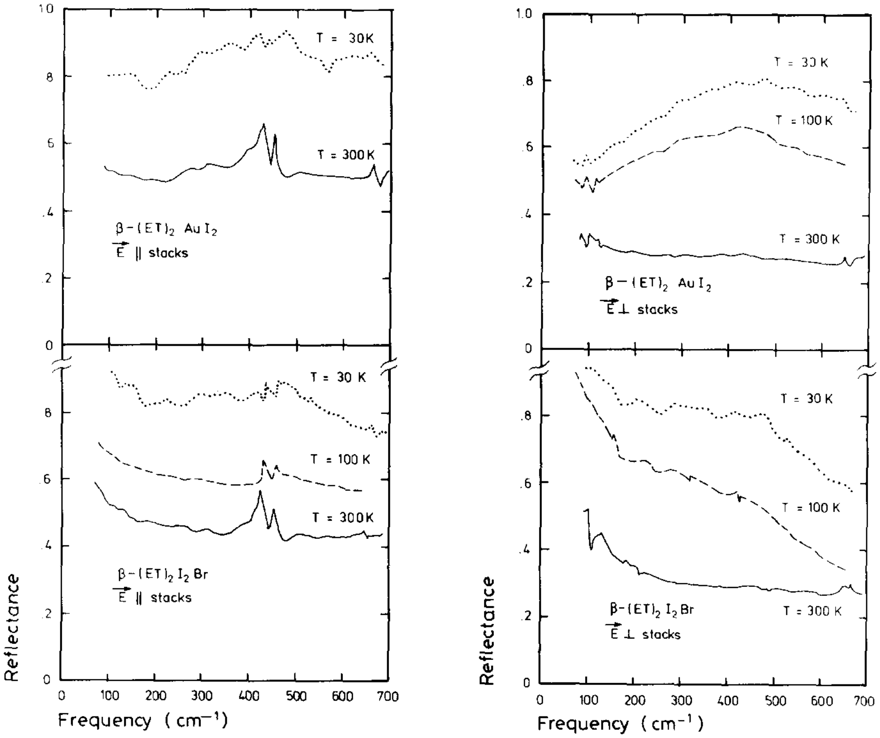
Fig. 5. Dielectric function of β-$(ET)_2AuI_2$ at two temperatures: T = 300K and 30K. E||stacks.

TABLE 1

Unscreened plasma frequencies and average transfer integrals. Data for $(ET)_2I_3$ are from Ref. [9]. ε_{core} = 3.6-4.0

X	$\omega_{p\parallel}$ cm^{-1}	$\omega_{p\perp}$ cm^{-1}	$\langle t_\parallel \rangle$ eV	$\langle t_\perp \rangle$ eV
I_3^-	9600	5700	.19	.08
I_2Br^-	9300	5300	.18	.07
AuI_2^-	10300	5800	.22	.09

Finally, we show in Fig. 6 details in the far infrared reflectance spectra. Along the stacks the temperature dependence of the strong doublet at 430-460 cm^{-1} is noteworthy. The line shape is inverted as the conductivity peak moves down at low temperature. For both polarizations it is of some interest to observe that the AuI_2^--material (with a superconducting ground state) is less Drude-like than the non-superconducting material. Whether there is a connection is not clear at present.



(a) (b)

Fig. 6. Far infrared reflectance of $(ET)_2X$, $X = AuI_2^-$, I_2Br^- at two temperature: T = 300K and 30K. (a) $\vec{E}\parallel$ stacks. (b) $\vec{E}\perp$ stacks.

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