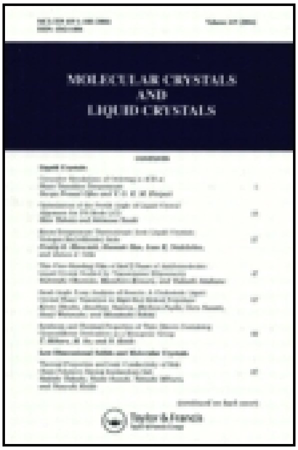

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FAR INFRARED RESPONSE OF THE CHARGE DENSITY WAVE IN $K_{0.3}MoO_3$

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Abstract Optical reflectivity has been measured in the far infrared region on $K_{0.3}MoO_3$ single crystals, using polarized light. At 300 K the reflectivity spectrum is metal-like (p ∥ b axis) and at 5 K a very strong polarization dependent phonon spectrum is observed. At photon energies below 8 meV a giant structure dominates the whole spectrum, polarized along the b-axis. The structure, reaching reflectivity values of 97% is assigned to the oscillations of the phase of the pinned CDW. Optical constants are calculated by means of the Kramers-Kronig relation: the results are compared with the predictions of mean field theory ($m^*_{CDW}, \lambda, T_C^{MF}$).

REFLECTIVITY MEASUREMENTS

The optical reflectivity of a large single crystal of $K_{0.3}MoO_3$ (cluster structure [1]) has been measured in an extended photon energy range from 12 eV down to 1 meV using linearly polarized light, in a temperature region between 5 and 300 K. In the far-infrared (FIR) part of the spectrum we have used a Bruker-Fourier spectrophotometer with TGS detectors down to 25 cm^{-1} and with a liquid Helium cooled germanium bolometer from 100 to 8 cm^{-1}. As in ref. [2] the incident light was polarized parallel to the metallic b-axis and perpendicular in the [102] direction. The whole spectrum is shown in Fig. 1: one notes that for p∥b the 300 K spectrum is metal-like with a plasma edge at about 1.3 eV and a reflection shoulder at 0.15 ≈ 0.2 eV. For $T<T_C$ = 180° K the metal-like reflectivity turns into a semiconductor like spectrum, shown for T = 5 K also in Fig. 1: the shoulder at 0.2 eV has developed into a broad reflexion maximum and typical phonon lines appear for photon energies below 0.12 eV (insert Fig. 1). In the FIR region a very high reflectivity peak, reaching a value of 97% stands out from the other maxima. The temperature dependence of this unusual maximum is shown in Fig. 2. The reflectivity behaviour above T_C is presented in the insert of Fig. 2. Some samples also show a weak maximum at 200 K in the same energy region which disappears at room temperature.

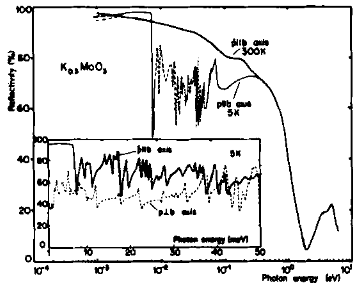

FIGURE 1 Polarized reflectivity of $K_{0.3}MoO_3$ at 5 and 300 K: to be noted the very strong structure in the FIR and the two possible $\omega \to 0$ extrapolations.

Kramers-Kronig transformation

The spectra have been analyed by means of the Kramers-Kronig relation (R,θ) and discussed in terms of the dielectric functions ε_1, ε_2 and of optical conductivity σ_1.

The dielectric functions show a very strong structure corresponding with the giant reflectivity peak: ε_2 shows a large peak at 1.8 meV (ω_{TO}) which reaches a value of about 7000. Consequently ε_1 has a very large dispersion in the same region. It intersects the abscissa with $d\varepsilon_1/d\omega > 0$, yielding ω_{LO}, at 7.4 meV. The ε_1, ε_2 values of the other phonon lines are around 100≈200. The dc values of ε_1 coming from this structure are very sensitive to the $\omega \to 0$ extrapolation. With an extrapolation as shown in Fig. 1 these values are between 2000 and 3000. The derivation of the transversal frequency ω_{TO} depends somewhat on the $\omega \to 0$ extrapolation. Our measurements show for all samples that dR/dω is positive at 5 K in the energy region between 10 and 15 cm^{-1}, yielding a transversal frequency of 1.8 meV. If we extrapolate the reflectivity curve for $h\omega < 15cm^{-1}$ with dR/dω = 0 towards ω = 0 we can reduce ω_{TO} at most by a factor 6 ($\omega_{TO} \approx$ 0.3 meV): ε_{stat} will be enhanced by a factor 10 and consequently, since ω_{LO} is indipendent from the extrapolation, the oscillator strength will also be enhanced. It is also possible that other structures in the reflectivity are present for ω<<THz, i.e. in the MHz or GHz region. Such structures will not influence at all the results in the FIR (δ-functions compared with FIR structures) but they would enhance the ε_{stat} by several orders of magnitude at ω<<THz.

The reflectivity spectrum polarized along the b-axis at 5 K presents in terms of optical conductivity σ_1 three strong peaks (Fig. 3): one located at 1.8 meV, the second centered at 0.07 eV and the third at 0.2 eV: the last is due to electronic transitions across the Peierls-gap. The strong line at 1.8 meV has a weight about 10 to 20 times larger than the other phonon contributions in the energy region between 8 and 50 meV. This line does not exist in the p$\perp$b spectrum. The intensity of this mode is very

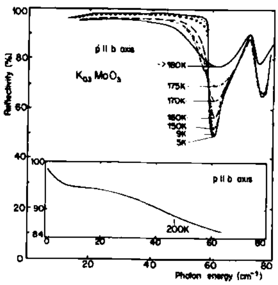

FIGURE 2 Temperature dependence of the reflectivity observed in the FIR for light polarized parallel to the coducting axis: the structure is assigned to the pinned Fröhlich phase mode.

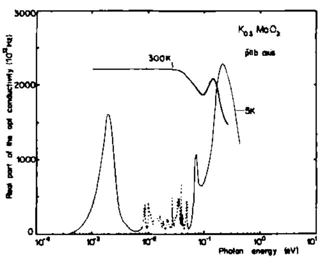

FIGURE 3 Optical conductivity of the blue bronze at 5 and 300 K: the FIR structure is the resonance of the oscillating CDW.

strong temperature dependent; the line becomes overdamped for $T \approx T_C$ and disappears for T > 200 K.

Such a large oscillator strength in the FIR region is quite unusual: the interpretation of this structure with the model of a normal phonon excitation is quite inappropriate: a cluster-cluster oscillation ($K_3Mo_{10}O_{30}$ per cluster) is to be excluded since the red bronze $K_{0.33}MoO_3$, with a similar crystallographic structure, [3] does not show a strong activity in the FIR region: the red bronze reaches reflectivity values of about 30% in the FIR region. The large structure at 1.8 meV cannot be only due to a simple phonon excitation. The line with ω_{TO} = 1.8 meV is therefore assigned to a pinned Fröhlich $2k_F$ phase mode.

If we assume that all the conduction electrons are condensed in the CDW, it turns out from

$$\omega_p^{CDW} = \sqrt{\frac{4\pi e^2 N}{m^*_{CDW}\varepsilon_{opt}}} = 7.4 \text{ meV}$$

that $m^*_{CDW} \approx 900\ m_e$ taking 6 electrons per unit cell and an estimated ε_{opt} of about 150 in the plasma frequency formula (if ε_{opt} = 100 or 250, m^*_{CDW} will be ≈ 1200 or ≈ 600 m_e). When the Fermi energy E_F is known, mean field theory allows one to determine the dimensionless electron-phonon coupling parameters λ. We have calculated the dispersion relation $E(k)[\Gamma\text{-}X]$ for one chain of $K_{0.3}MoO_3$ [4] using an LCAO method. The calculation yields a conduction band width of 1 eV, a Fermi energy E_F of 0.7 eV, a 3/4 full band, a K_F = 3/4 b*/2 and a density of states D(E) at E_F of 1.8 states per eV per spin for a double-degenerate $t_{2g}\text{-}p_\pi$ band.

From this calculation we then obtain the mean field parameters: λ = 0.3, $T_c^{MF} \approx$ 600-700 K and m^*_{CDW} = 800 m_e = 800 m_e: the effective mass agrees quite well with the experimental results.

With knowledge of the transversal frequency ω_{TO} it is possible to estimate the treshold field E_c [5] for the depinning of the CDW: for a sinussoidal pinning potential we found $E_c \approx$ 170 KV/cm which is in contrast to the value obtained from the non Ohmic behaviour of the electrical conductivity: E_c = 0.1 V/cm [6]. It is possible that the latter value is related to a motion of only a part of the CDW through the presence of dislocations in the CDW lattice. In this case the small electrical field E_c could be connected directly to the very strong mid-gap structure (0.07 eV) present in the optical conductivity.

In Summary, we want to mention the similarity between the blue bronze CDW properties and those of KCP [7-10].

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