terms of the very large longitudinal displacements of (110) GaAs,[2] which at the film surface show a mixed longitudinal and z-transverse polarization. We have found previously[2] that this structure is present also in the clean GaAs spectrum at the same frequency Ω_L. However, we want to stress that in the case of the clean surface the peak is due to the elasto-optic coupling while the Al cover causes the peak to originate from the ripple mechanism.

In conclusion, by describing the interaction of the light with the metallic film through the ripple mechanism and by using the theory of elasticity for the displacement field, it is possible to reproduce the surface features of both the continuous and the discrete part of the Brillouin spectrum. Our analysis indicates the existence of a strong interaction between the long-wavelength modes in the two media.

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Conductivity of the One-Dimensional Conductor Tetrathiafulvalene-Tetracyanoquinodimethane (TTF-TCNQ) near Commensurability

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We report an important drop in the longitudinal conductivity of TTF-TCNQ occurring at constant temperature in the metallic phase and in a narrow pressure domain located around 19 kbar. The drop of conductivity has been related to a $\times 3$ commensurability of the longitudinal distortion with the lattice. This experiment, together with x-ray diffuse scattering of TTF-TCNQ could suggest that a significant fraction of the metallic conductivity below ambient temperature is due to the fluctuating collective mode.

The organic conductor TTF-TCNQ exhibits several experimental features characteristic of one-dimensional (1D) metallic behavior.[1,2] Large values of electron-electron interactions in TTF-TCNQ are necessary for the interpretation of the enhancements observed in spin susceptibility[3] and nuclear relaxation rate.[4] However, the electron-phonon interaction reveals its presence through the existence of structural phase transitions at low temperature.[5] Understanding of the metallic conductivity remains the main puzzle for TTF-TCNQ and its derivatives. Various theories have been proposed for the resistivity: (a) the single-particle scattering picture: intramolecular phonon scattering,[6] second-order scattering against librations,[7] interchain electron-electron scattering,[8] electron-spin fluctuation scattering[9]; (b) the collective-mode picture: important contribution from fluctuating charge-density waves.[10-12] The study of the TTF-TCNQ phase diagram[13] has shown the existence of a narrow pressure domain in the vicinity of 20 kbar, where, as a result of the increase of the charge transfer, the wavelength of the lattice distortion becomes commensurate with the underlying lattice. The peaking of a single-phase transition temperature around 20 kbar and its first-order character have suggested the occurrence of a ×3 commensurability ($2k_F = \frac{1}{3}b^*$) at this pressure.[13,14] Moreover, high-pressure optical reflectance studies[15] allow us to discard the possibility of important changes in the band structure other than band filling.

In this Letter we use the possibility of achieving commensurability under pressure as a new experimental tool to investigate the resistivity of TTF-TCNQ up to 28 kbar. The b-axis resistivity was measured on single crystals ($2\times0.3\times0.06$ mm^3) using the standard four-probe, ac, phase-sensitive techniques at 70 Hz. High pressure was generated by He gas up to 12 kbar and by isopentane in a Teflon capsule above. Readings of sample voltages and a Cu-Constantan thermocouple inside the pressure cell were digitized and automatically recorded during slow cooling and warming cycles at constant pressure. Samples used for this study showed conductivity peak ratios (CPR), at atmospheric pressure, ranging from 10 to 25. For all samples the "unnested voltage" ratios were better than 10^2 at 300 K and exceeded 20 at T_{peak}. No phase shifting or data irreproducibility after T and P cycles has been noticed within 2%. In the present study we have observed a single and sharp transition in the commensurability domain, peaking at 74 K under 19 kbar. The transition into the commensurate charge-density wave (CDW) state is of first order as clearly shown by the hysteresis of about 1 K observed at the transition, Fig. 1.[16] The constant-temperature pressure dependence of the conductivity of three samples is displayed in Fig. 2. The conductivity has been normalized at 400 $(\Omega\ \text{cm})^{-1}$ under ambient conditions for all samples. The minimum temperature of 85 K, at which the pressure dependence of the conductivity has been studied for all samples, is already well into the metallic regime ($d\rho/dT>0$), whatever the pressure is. The striking feature emerging from Fig. 2 is the loss of conductivity occurring around 19 kbar (when CDW and lattice become commensurate). The amplitude of the conductivity loss weakens in the case of different samples at a fixed temperature [say 85 K, as the CPR drops CPR =25, 16, 11 for samples (a), (b), and (c), respectively]. The overall pressure dependence of the conductivity becomes smaller at low temperature [$d\ln\sigma/dP\approx 28\%$ kbar^{-1} and 7% kbar^{-1} at 290 and 80 K, respectively, for sample (a) around atmospheric pressure].[17] To discuss the properties of the system at a pressure corresponding to a phase transition into the commensurate state ×3, it is essential to note that the effects of commensurability are described by a cubic term in the Landau free-energy expansion[13,18,19] similar to the Landau theory of the layered compounds.[20] Because of the cubic term, the phase transition into the commensurate CDW state is first order and the transition temperature T_{c1} is *higher* than the temperature T_{c2} of the second-order transition occurring in the absence of the cubic term (i.e., in the incommensurate case). According to the TTF-TCNQ phase diagram[13] a value $T_{c2}\approx 60$ K at 19 kbar provides an estimate of the commensurability-induced transition-temperature enhancement, $T_{c1}-T_{c2}\approx 15$ K.

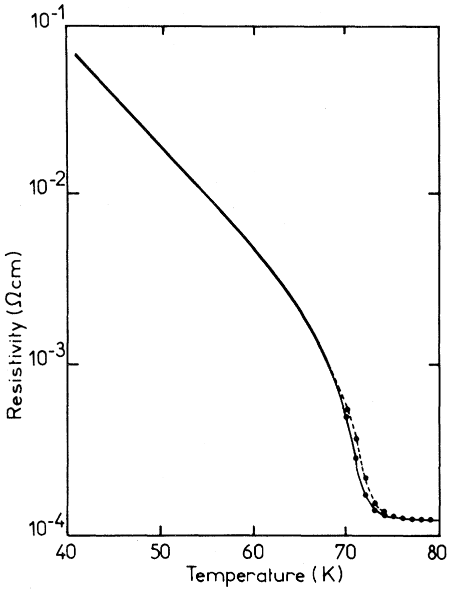

FIG. 1. Resistivity vs temperature at a pressure situated in the commensurability range. Experimental points have been displayed in the transition region. The lines drawn through these points (unbroken means cooling, broken means warming) indicate a 1-K hysteresis at T_{c1}.

Data shown in Fig. 2 can be discussed following two ways: (i) In the incommensurate case rigid translations of fluctuations into the CDW state (sliding-mode conductivity) are possible since the energy of a fluctuation is independent of its phase.[21] In the commensurate case, how-

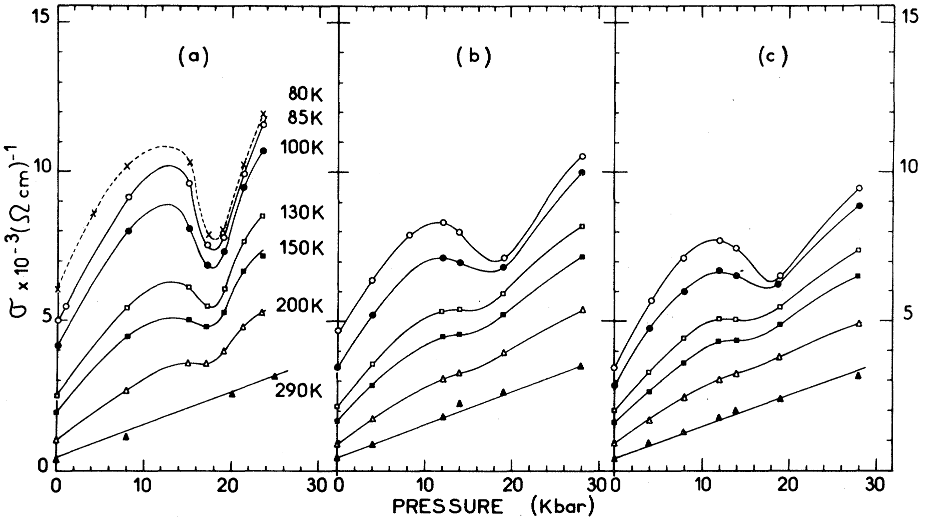

FIG. 2. Pressure dependence of the conductivity of TTF-TCNQ at constant temperature. Results for three samples have been displayed, corresponding to CPR = 25, 16, and 11 for (a), (b), and (c), respectively. The sequence of temperatures is similar for the three samples. A $T = 80$ K curve has been drawn only for sample (a) since for this sample T_{peak} at 19 kbar was situated below 80 K.

ever, the energy of a fluctuation does depend on its phase through the phase-dependent cubic term.[19] Therefore, a finite restoring force prevents translations of the fluctuating CDW so that its phase is pinned to some equilibrium position thereby suppressing the sliding-mode contribution to the conductivity.[22] Thus, a commensurability of low order is a drastic pinning mechanism for the fluctuating CDW and it can well explain the lowering of the conductivity in the commensurate case, as observed in the present study. (ii) If the resistivity is dominated by a single-particle-like scattering mechanism, the behavior around 19 kbar suggests an increase in the scattering rate and/or a decrease in the density of states at the Fermi level associated with the commensurability.[23] In any of the above-mentioned single-particle theories the resistivity is independent of the phase of the CDW fluctuations but eventually depends on the amplitude.

On the other hand, the main effect of the commensurability-induced cubic term is to pin the phase of the fluctuations, whereas the amplitude is only slightly affected.[24] Moreover, it should be noticed that the dip in the conductivity at 19 kbar sets in at about 200 K, which is in fair agreement with the temperature at which $2k_F$ fluctuations are first seen in x-ray experiments under atmospheric pressure.[25,26] Consequently, we tend to believe that the fluctuating CDW picture is likely to explain the behavior of the conductivity around commensurability. In this picture, at 19 kbar, the conductivity is mainly due to the single-particle scattering and is consequently weakly sample dependent as shown from Figs. 2(a)-2(c), whereas on either side of commensurability the sample dependence might be explained by the pinning of fluctuations arising from impurities or crystal defects.[27] However, the important question of the fluctuating CDW conductivity would be definitely settled by the measurement around commensruability of properties which are sensitive to the single-particle electron-scattering time only.[28] Finally, preliminary experiments performed on the selenium analog compound, TSeF-TCNQ, have shown a similar loss of conductivity around 7.5 kbar. According to the existence of maxima in both the low-temperature gap and the ratio of the low-temperature gap to transition temperature, a ×3 commensurability is very likely to occur in TSeF-TCNQ around 7.5 kbar.[29] No such behavior has been observed in hexamethylene TTF and hexamethylene-tetraselenafulvalene tetracyanoquinodimethane in the 30-kbar range since the charge transfer is already above $\frac{2}{3}$ at atmospheric pressure.[30]

In conclusion, this Letter has reported an experiment which brings the evidence that commensurability interferes significantly with the conductivity mechanism in the metallic phase of TTF-TCNQ. The loss of conductivity brought about by the commensurability is, however, restricted to the temperature domain in which 1D $2k_{\mathrm{F}}$ reflections are observed by x rays, under ambient pressure. A tentative explanation of the data has been given suggesting a significant contribution of CDW fluctuations to the conductivity in the incommensurate situation.

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