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Classification
Physics Abstracts
72.15N — 75.30F

Phason excitations in the SDW state of $(TMTSF)_2PF_6$

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(Received 30 December 1992, accepted 5 February 1993)

Abstract. — We have measured the frequency dependent conductivity at various temperatures below the SDW transition in $(TMTSF)_2PF_6$. We find two distinct resonances which we ascribe to internal deformations and pinned mode oscillations of the SDW condensate. Furthermore, the spectral weight of the pinned mode resonance is found to be nearly temperature independent below the SDW transition. Our findings are contrasted with various models and suggestions on the electrodynamics of the SDW ground state.

Considerable progress has been made recently in exploring the collective mode dynamics of the broken symmetry state called the spin density wave (SDW) [1]. The electrodynamical response $\hat{\sigma}$ of the ground state is determined by both the collective mode $\hat{\sigma}_{SDW}$ and single particle $\hat{\sigma}_{sp}$ excitations. As for a superconductor, single particle excitations are expected for photon energies $\hbar\omega > 2\Delta$ where 2Δ is the SDW gap. The collective mode response arises at zero frequency for a superconductor, while for a SDW impurities shift the mode to a finite frequency ω_0, the so-called pinning frequency. In either case, in the clean limit ($\hbar\tau^{-1} < \Delta$, where τ^{-1} is the scattering rate) at $T = 0$ K all the spectral weight, defined as

$$\int_0^\infty \mathrm{Re}\,\hat{\sigma}(\omega)\,\mathrm{d}\omega = \frac{\omega_p^2}{8} \tag{1}$$

where $\omega_p^2 = \frac{4\pi ne^2}{m_b}$ with n the number density of electrons and m_b the bandmass, is expected to be associated with the collective mode. This is in contrast to that found for charge density wave (CDW) systems where, due to the large effective mass m^* of the condensate, the spectral weight of the collective mode resonance is small [2].

We have recently observed [3] a low lying resonance at frequencies well below the expected single particle gap $\frac{2\Delta}{h} \cong 30\ \mathrm{cm}^{-1}$ (as determined from the temperature dependence of the dc conductivity $\sigma = \sigma_0 \exp\left(-\frac{\Delta}{kT}\right)$) in the model compound $(TMTSF)_2PF_6$ below the SDW transition ($T_{SDW} = 11.5$ K). We argued that this resonance is due to the oscillatory response of the SDW condensate, the $q = 0$ phason excitation ; and, in clear contrast with that expected from equation (1), a strongly reduced spectral weight has been found.

Several models have been proposed to account for our experimental findings. One explanation, that the effective mass is large due to coupling to phonons, can be ruled out as no lattice distortion has been found in the SDW state [4]. It has also been suggested [5] that, as for superconductors, long range Coulomb forces shift the spectral weight away from the pinning frequency ω_0 up to the plasma frequency ω_p. This model, based on the so-called Anderson-Higgs mechanism [6], should apply in a translationally invariant system only to the longitudinal mode ($q \parallel \mathrm{E}$) while leaving the transverse mode ($q \perp \mathrm{E}$), as sampled with optical measurements, unaffected. However, the transverse and longitudinal modes may be mixed due to anisotropy effects and the randomly situated impurities [7]. It has also been suggested [8] that the mode observed may well be due to internal deformations [7] and not to the oscillatory response of the SDW. If this is the case, the pinned mode resonance — which should be at frequencies well above the spectral range where the modes due to internal deformations occur — would most likely be above the gap, i.e. $\hbar\omega_0 > 2\Delta$. Recently, it has been suggested that commensurability effects may play an important role and that these effects lead to bound state resonances below the gap [9]. These bound state resonances may have a large spectral weight and thus (because of the conservation of total spectral weight) may lead to the reduction of the intensity of the pinned mode resonance. Impurities can also lead to bound states [10, 11], and in an analogous way also lead to a reduction of the collective mode spectral weight. Finally, it was also proposed [12] that impurities may lead to a mixing of the $q = 0$ magnon and phason modes, and that the low lying resonance observed is a magnon excitation at the antiferromagnetic resonance frequency.

In order to address these questions we have conducted experiments at various temperatures over an extensive spectral range which spans the audio to infrared frequencies. Standard radiofrequency (rf) techniques were utilized up to 1 GHz, and the components of the optical conductivity $\hat{\sigma} = \sigma_1 + i\sigma_2$ were evaluated from the time dependence of the discharge current [13]. In the micro- and millimeter wave spectral range resonant cavities operating at 3, 7, 9, 12, 35, 60, 100 and 150 GHz were employed, and the surface impedance $\hat{Z}_s = R_s + iX_s$ was measured. Our measurement techniques and analysis are described elsewhere [14]. For a good metal R_s is proportional to the absorptivity $A(\omega) = 1 - R(\omega)$, where $R(\omega)$ is the reflectivity, and thus can be combined with optical experiments, conducted between 15 and $10^4\ \mathrm{cm}^{-1}$, to construct $A(\omega)$ over a broad spectral range. The optical measurements described here were made on mosaics and the procedure has been described in more detail in reference [15]. A Kramers-Kronig (KK) analysis was used to evaluate $\sigma_1(\omega)$ at various temperatures.

In figure 1 we show the absorptivity at two temperatures, one above and one well below T_{SDW}. The absorptivity values measured with different techniques are shown together with a Drude and Lorentz oscillator fit. At high frequencies $\left(\frac{\omega}{2\pi} > 100\ \mathrm{cm}^{-1}\right)$ a temperature independent feature is found which is discussed in detail elsewhere [15]. Above T_{SDW}, at 20 K, the frequency dependent conductivity can be well described with a simple Drude expression. The Drude fit gives a relaxation time $\frac{1}{2\pi\tau} = 3\ \mathrm{cm}^{-1}$, placing it well into the clean limit. This leads, with $\sigma_{\mathrm{dc}}(20\ \mathrm{K}) = 3 \times 10^4\ \Omega^{-1}\ \mathrm{cm}^{-1}$ and a carrier concentration $n = 1.4 \times 10^{21}\ \mathrm{cm}^{-3}$ to a bandmass of $m_b = 20\, m_e$, in qualitative agreement with the bandmass obtained from magnetic measurements [16]. Below T_{SDW} the measured absorptivity can be well reproduced using a Drude oscillator to model the uncondensed electrons and an underdamped Lorentz oscillator for the microwave resonance. An extrapolation between the measured points is made by joining the different sets of data together using the oscillator fit as a guide. At each temperature $A(\omega)$ is transformed through a KK analysis to give $\sigma_1(\omega)$, as shown in figure 2.

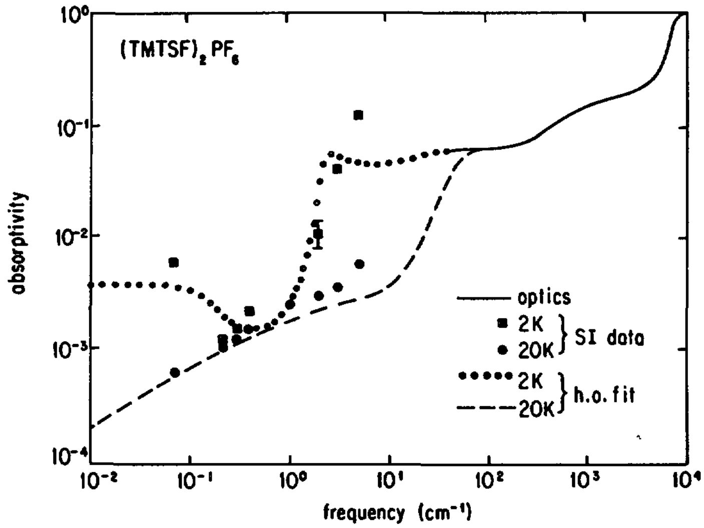

Fig. 1. — The frequency dependence of the absorptivity both above ($T = 20$ K) and below ($T = 2$ K) T_{SDW} in $(TMTSF)_2PF_6$. Optical measurements (solid line) are shown together with both surface impedance (SI) measurements (circles and boxes) and a simple harmonic oscillator (h.o.) fit (dashed and dotted line).

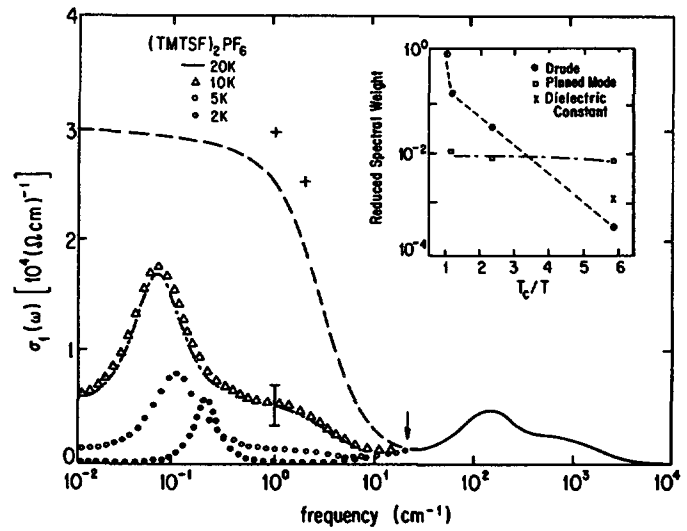

Fig. 2. — The frequency dependent conductivity both above and below T_{SDW} for $(TMTSF)_2PF_6$ determined from a Kramers Kronig analysis of figure 1. The single particle gap, determined from dc conductivity measurements, is indicated in the figure with an arrow. The pluses (+) are the values of σ_1 as determined directly from a measurement of both R_s and X_s in the microwave region. In the inset, the temperature dependence of the normalized spectral weight of the various contributions to $\sigma_{SDW}(\omega)$ below T_{SDW} are displayed, where each has been normalized by the spectral weight of the Drude response at 20 K.

A resonance at temperatures below T_{SDW} is clearly evident near 0.1 cm^{-1} This resonance was previously associated [3] with the pinned mode resonance. The single particle gap, as established from the temperature dependence of the dc conductivity, is indicated by the arrow in figure 2. Earlier [3] we have associated the increase of $\sigma_1(\omega)$ around this frequency with the gap in the SDW state, however, the fact that this structure is also observed well above T_{SDW} [15] suggests that it has a different origin and is most probably due to an interband transition. The reason for the absence of the gap structure in the optical conductivity is the same as that in a superconductor : in the clean limit $\sigma_1(\omega)$ is small in the spectral range around 2Δ, and consequently the effect of the removal of this spectral weight leads to only minor changes in the reflectivity.

The detailed form of the low lying resonance was explored by combining the optical data with experiments in the rf spectral range. Below T_{SDW}, the conductivity $\hat{\sigma}(\omega)$ has contributions from both the thermally excited electrons $\hat{\sigma}_{sp}(\omega)$ and the condensed electrons $\hat{\sigma}_{SDW}(\omega)(\hat{\sigma}(\omega) = \hat{\sigma}_{sp}(\omega) + \hat{\sigma}_{SDW}(\omega))$. $\hat{\sigma}_{SDW}$ for $(TMTSF)_2PF_6$, together with the conductivity observed [17] in the material $K_{0.3}MoO_3$ in the CDW state are displayed in figure 3. In both cases we find a broad structure at low frequencies together with a well defined resonance in the high frequency end of the spectrum. In the CDW system the low frequency behavior has been analyzed in detail and is now well understood [7] to be due to the internal deformations of the collective mode. Furthermore, the resonance which appears at 100 GHz for $K_{0.3}MoO_3$ is due to the oscillations of the entire collective mode. This resonance is usually referred to as the pinned mode resonance at the pinning frequency ω_0. Extensive experiments [2, 18] on several materials with a CDW ground state clearly establish that ω_0 is indeed determined by pinning to the impurities, and that it represents the oscillations of the entire collective mode subject to an average restoring force whose spring constant is given by $k = m^* \omega_0^2$. Much less is known

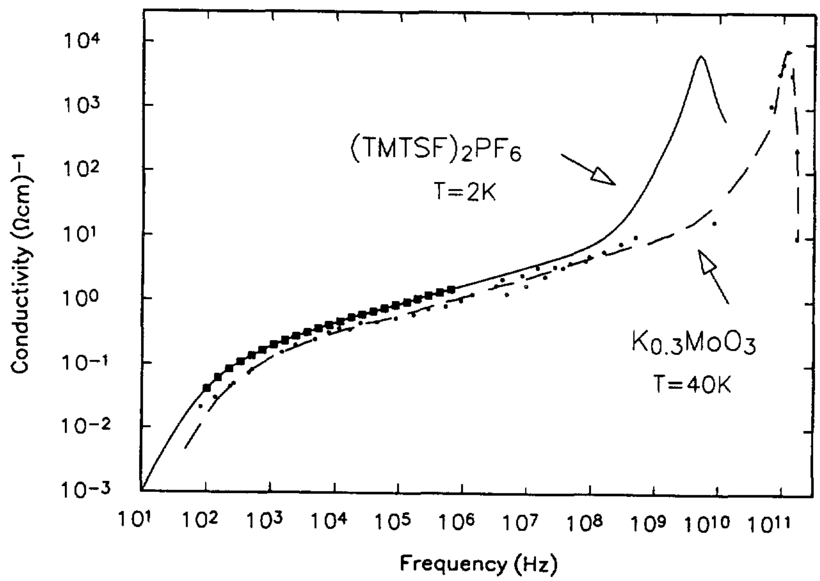

Fig. 3. — The frequency dependence of the real part of the SDW conductivity $\hat{\sigma}_{SDW}(\omega)$ for $(TMTSF)_2PF_6$ and $\hat{\sigma}_{CDW}(\omega)$ for the CDW compound $K_{0.3}MoO_3$. The measured conductivity is shown (boxes and circles) together with the output of the KK analysis (solid and broken lines). The measurements shown were made at different temperatures, but in each case the measurement was made well below the transition temperature.

about the electrodynamical response of the SDW materials. Experiments on SDW samples with varying impurity concentrations are currently underway in order to determine the dependence of $\sigma_1(\omega)$ on impurity concentration. However, the striking similarity of the frequency dependent response of these two systems seen in figure 3 strongly suggests that the low frequency mode in $(TMTSF)_2PF_6$ is due to internal oscillations of the SDW and the mode around $0.1\ cm^{-1}$ is the response of the pinned SDW. Therefore, we conclude that for $K_{0.3}MoO_3$ and $(TMTSF)_2PF_6$ both the internal mode deformations and the pinned mode resonance appear well within the single particle gap, $\hbar\omega_0 < 2\,\Delta$, in contrast to the suggestion of reference [7].

The temperature dependence of the spectral weight of the pinned SDW mode was evaluated by fitting the observed conductivity with a function of the form

$$\hat{\sigma} = \hat{\sigma}_{sp} + \hat{\sigma}_{SDW} = \frac{n(T)\,e^2\,\tau}{m_b(1 - i\,\omega\tau)} - \frac{i\,\Omega_p^2\,\omega}{4\,\pi[(\omega_0^2 - \omega^2) - i\,\omega\Gamma]} \tag{2}$$

where the first term $\hat{\sigma}_{sp}$ is the Drude response of the thermally excited electrons, taken from the temperature dependent dc conductivity $\sigma_{dc} = \frac{ne^2\,\tau}{m_b}$ below the transition, and the second Lorentzian term $\hat{\sigma}_{SDW}$ describes the pinned mode resonance, with Ω_p the SDW plasma frequency and Γ the SDW damping. A temperature independent τ has been used for $T < T_{SDW}$. The spectral weight associated with the two contributions in equation (2) is displayed in the inset of figure 2 as a function of temperature. It is clear from the figure that in contrast to the temperature dependent Drude contribution, the spectral weight of the pinned mode resonance is only weakly temperature dependent and is significantly reduced from the spectral weight associated with the Drude conductivity above the SDW transition. Our observations therefore rule out the Anderson-Higgs mechanism [5] as it would lead to an exponential freeze out of the spectral weight of the pinned mode resonance with decreasing temperature. The strong reduction in the spectral weight of the pinned mode has been independently confirmed with the measured low frequency dielectric constant ε [13]. Just below T_{SDW}, ε is dominated by low frequency excitations, but at $T = 2$ K these low frequency modes freeze out and ε is determined by the pinned mode resonance. The spectral weight obtained by this method is also displayed in the inset of figure 2 and it is more than two orders of magnitude below the spectral weight associated with the Drude response at 20 K.

Presently we can only speculate on the origin of the missing spectral weight in the SDW state. As the total spectral weight must be the same both below and above the transition

$$\int_0^\infty \mathrm{Re}\ \hat{\sigma}_{SDW}(\omega)\,d\omega = \int_0^\infty \mathrm{Re}\ \hat{\sigma}_{Drude}(\omega,\,T > T_{SDW})\,d\omega = \frac{\pi ne^2}{2\,m_b} \tag{3}$$

there must be excitations associated with the SDW response in addition to those found in the microwave region and below. One likely explanation is that the missing spectral weight is associated with bound states accompanying the SDW condensate. In contrast to CDW's, it is expected that bound states due to impurities can readily occur, and the reasoning is as follows [10]. In a CDW the periodically modulated charge density can adjust to the electrostatic impurity potentials by adjusting its local phase to minimize the total energy (electrostatic + elastic) gain. On the other hand, the SDW ground state can be viewed as two CDW's, one for each spin direction, with a CDW modulation on the sublattices which differs by π. As the pinning in an SDW is also due to electrostatic forces [10, 19], a full adjustment of the phase cannot occur simultaneously for both spin sublattices, leaving a net energy gain which is much larger than for a CDW. However, the electrostatic energy can be lowered by removing electrons from the condensate to form a bound state about the impurity. In addition, the spectral weight of these bound states is expected to be significantly larger than in CDW systems. The spatial extension of a bound state is given by the coherence length $\xi = \frac{\hbar v_F}{\pi \Delta}$, and with a Fermi velocity $v_F \cong 10^7$ cm/s, and single particle gap $\Delta/h \cong 15\ \mathrm{cm}^{-1}$, $\xi \cong 10^3$ Å, which is significantly larger than the coherence length for typical CDW systems. As the matrix element for optical transitions of the bound state is proportional to its dipole moment and hence ξ, a large coherence length leads to a large optical spectral weight associated with that transition.

Several microscopic models also suggest a broad spectrum of impurity induced bound states [10, 11] below the single particle gap. An alternative model [9] suggests that as these materials are near to commensurability, soliton excitations could occur below the gap. These excitations would also have a spatial extension of $\xi = \frac{\hbar v_F}{\pi \Delta}$ and therefore would have a large spectral weight. At present we do not have clear spectroscopic evidence for states below the single particle gap. Furthermore, since the missing spectral weight is much smaller ($\cong 5\ \%$) than that associated with the temperature independent feature above the gap, we cannot experimentally determine whether the missing weight is above or below the continuum of single particle excitations.

Acknowledgments.

This research was supported by the National Science Foundation grant DMR 89-13236. Useful discussions with H. Fukayama, K. Maki, G. Mihály, and A. Zawadowski are gratefully acknowledged.

References

[1] GRUNER G. and MAKI K., *Comments Condensed Matter Phys.* **15** (1991) 145.
[2] GRUNER G., *Rev. Mod. Phys.* **60** (1988) 1129.
[3] QUINLIVAN D., KIM Y., HOLCZER K., GRÜNER G. and WUDL F., *Phys. Rev. Lett.* **65** (1990) 1816.
[4] POUGET J. P. (private communication).
[5] MAKI K. and GRÜNER G., *Phys. Rev. Lett.* **66** (1991) 782.
[6] ANDERSON P. W., Basic Notions of Condensed Matter Physics (Benjamin/Cummings, CA, 1984).
[7] LITTLEWOOD P., *Phys. Rev. B* **36** (1987) 3108 ;
WONNEBERGER W., *Synthetic Metals* **43** (1991) 3793.
[8] LITTLEWOOD P. (private communication).
[9] WONNEBERGER W., *Solid State Commun.* **80** (1991) 953.
[10] TÜTTO I. and ZAWADOWSKI A., *Phys. Rev. Lett.* **60** (1988) 1442.
[11] ZAWADOWSKI A. and TÜTTO I., *Synthetic Metals* **29** (1989) F 469.
[12] FUKAYAMA H. (private communication).
[13] MIHALY G., KIM Y. and GRÜNER G., *Phys. Rev. Lett.* **66** (1991) 2806.
[14] DONOVAN S., KLEIN O., HOLCZER K. and GRÜNER G., to be published.
[15] DONOVAN S., DEGIORGI L., KIM Y. and GRÜNER G., submitted to *Europhys. Lett.* **19** (1992) 433.
[16] MORTENSEN K., TOMKIEWICZ Y. and BECHGAARD K., *Phys. Rev. B* **25** (1982) 3319. Using the measured value of χ for a one dimensional free electron gas gives $m_b/m_e = 3$.
[17] MIHALY G., KIM T. W. and GRÜNER G., *Phys. Rev. B* **39** (1989) 13009.
[18] KIM T. W., DONOVAN S., GRÜNER G. and PHILIPP A., *Phys. Rev. B* **43** (1991) 6315.
[19] TUA P. F. and RUVALDS J., *Phys. Rev. B* **32** (1985) 4660.