Effect of Coulomb Interactions on Spin-Density-Wave Dynamics

K. Maki
Department of Physics, University of Southern California, Los Angeles, California 90089-0484

G. Grüner
Department of Physics and Solid State Science Center, University of California, Los Angeles, Los Angeles, California 90024
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The effect of Coulomb and electromagnetic interactions on the dynamics of spin-density waves is discussed. We show that effects analogous to the Anderson-Higgs mechanism for superconductors occur, and discuss the experimental evidences for such a mechanism.

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Coulomb effects play an important role in the electrodynamics of superconducting[1] and charge-density-wave[2] (CDW) ground states. In the absence of Coulomb effects the excitation spectrum is gapless for both types of condensates. The velocity of the excitations is equal to the Fermi velocity V_F for the superconducting ground state, and screening effects are important only in the immediate vicinity of T_c. Also, Coulomb effects lead[3,4] to a shift of the dispersion relation up to the plasma frequency $\omega_p = 4\pi ne^2/m_b$, where n is the number of carriers and m_b the band mass. Typically, $\omega_p \gg 2\Delta$ and there are no excitations below the gap frequency $2\Delta/\hbar$. For a charge-density-wave ground state the phason excitations have a velocity $c = (m_b/m^*)^{1/2}V_F$. Here m^* is the effective mass, which is typically[2] $(10^2\text{–}10^3)m_b$. The consequence of the large m^* is twofold. First, Coulomb effects become important[5,6] only at temperatures well below T_{CDW}; and second, Coulomb effects lead to a massive mode at frequency $\omega_p^* = \omega_p(m/m^*)^{1/2}\varepsilon_\Delta^{-1/2}$, where ε_Δ is the dielectric constant associated with the single-particle excitations across the gap Δ. As $m^* \gg m_b$, $\omega_p^* \ll \omega_p$ and is typically smaller than 2Δ. These features have several important consequences on the frequency-dependent conductivity $\sigma(\omega)$, which due to random impurities, is determined both by the transverse and by the longitudinal mode.[5] There is a contribution from single-particle excitations across the gap 2Δ which exhausts nearly all the spectral weight associated with the ac response. The collective-mode contribution appears at the so-called pinning frequency which occurs well below the gap frequency. The contribution of the collective mode to the spectral weight of the ω-dependent conductivity,

$$A = \int \sigma_{\mathrm{CDW}}(\omega)\,d\omega = \pi ne^2/2m^* , \tag{1}$$

is small, but finite as $T \to 0$ and reflects the large dynamical mass m^*.

The electrodynamics of the spin-density-wave (SDW) ground state is expected to be different from that of the superconducting and charge-density-wave state. Because the dynamical mass m^* is equal to the band mass m_b, and because of pinning effects (which are comparable to pinning effects in charge-density-wave systems), it is expected[7] that the collective-mode contribution (1) appears at finite frequencies $\omega_0 < 2\Delta$, and (2) contains the full spectral weight of the frequency-dependent conductivity with no single-particle contributions to $\sigma(\omega)$ at $\omega = 2\Delta$ and above. (Both statements are appropriate in the so-called clean limit where the inverse relaxation time $1/\tau \ll \Delta$.)

We have recently reported[8] on our frequency-dependent conductivity measurements, performed in the spin-density-wave state of the organic linear chain compounds tetramethyltetraselenofulvalene$_2X$ [$(\mathrm{TMTSF})_2X$] with $X = \mathrm{PF_6}$ and $\mathrm{NO_3}$. Our experimental observations are dramatically different from the expected frequency-dependent response. In the spin-density-wave state we find clear evidence for single-particle excitations across the gap, in agreement with earlier optical studies.[9] The analysis of our experimental results at temperature $T = 0.5T_{\mathrm{SDW}}$ leads to a low-lying resonance at $\omega_0 \ll \Delta$ in the $\mathrm{PF_6}$ salt, with a spectral weight which, if interpreted in terms of Eq. (1) would lead to a large effective mass $m^* \simeq 100m_b$. In the $\mathrm{NO_3}$ salt,[10] experiments at low temperature do not give evidence for a collective mode, and $\sigma(\omega < 2\Delta)$ is dominated by thermally excited carriers across the gap.

Phase excitations for spin-density waves result in charge fluctuations, and in the absence of screening are subject to long-range Coulomb interactions. In contrast to CDW's, the phase velocity is V_F, significantly larger than the velocity of charge-density-wave phason excitations c. The large phason velocity of the SDW state is expected to lead (in analogy to what is observed in superconductors) to two effects. First, screening is important only in the intermediate vicinity of T_{SDW}. Second, Coulomb effects raise the phason spectrum to the plasma frequency $\omega_p \gg 2\Delta$ in an analogy to the Anderson-Higgs mechanism[3,4] for superconductors, and the transverse mode is also fundamentally modified. In this Letter we calculate optical properties of the SDW ground state and evaluate explicitly the effects of Coulomb and electromagnetic interactions. We show that the Anderson-Higgs mechanism applies for spin-density-wave condensates, and that the consequence of Coulomb interactions is a spectral weight of collective-mode excitations which is strongly reduced, and ideally, disappears as $T \to 0$. We discuss various experimental results obtained on $(TMTSF)_2X$ salts which support our conclusions on the importance of Coulomb and electromagnetic interactions.

The electrodynamical response of the spin-density-wave state was discussed earlier[11] in the limit where long-range Coulomb interactions are neglected.

In terms of the phason propagator D_ϕ, the collective-mode contribution to the conductivity σ_{coll} is given by

$$\sigma_{\text{coll}}(\omega) = (ne^2/m_b) i\omega f(T,\omega) D_\phi \,, \tag{2}$$

where[5,11]

$$D_\phi^{-1} = \frac{8\Delta^2}{U f(T,\omega)} (1 - \tfrac{1}{2} U \langle [\delta\Delta, \delta\Delta] \rangle) \,. \tag{3}$$

In Eq. (3) U is the on-site Coulomb Hubbard interaction, $\delta\Delta$ is the $q = 2k_F$ component of the spin density n, $f(T,\omega)$ is the dynamical condensate density, and ω_0 is the pinned mode frequency. $f(T,\omega)$ has a nonanalytical temperature dependence, determined by the equation,[11]

$$f(T,\omega) = [2\Delta(T)]^2 \int_{\Delta(T)}^{E_F} \frac{dE}{[E^2 - \Delta^2(T)]^{1/2}} \frac{\tanh(\beta E/2)}{4E^2 - \omega^2} \,, \tag{4}$$

where $\beta = 1/kT$. For $\omega/\Delta \ll 1$, $f(T, \omega \ll \Delta) \to 0$ as $T \to T_{\text{SDW}}$ and $f(T, \omega \ll \Delta) \to 1$ for $T \to 0$; at $T \ll T_{\text{SDW}}$, $1 - f(T, \omega \ll \Delta) \sim \exp(-\beta\Delta)$. In the absence of Coulomb interactions[5,11] $D_\phi = \omega_0^2 - \omega^2$, and

$$\sigma_{\text{coll}}(\omega) = (ne^2/m_b) i\omega f(T,\omega) (\omega^2 - \omega_0^2)^{-1} \,. \tag{5}$$

ω_0 is determined by the interaction of the collective mode with impurities,[12] and here we treat it as a free parameter. It is expected that for nominally pure specimens, where ω_0 is due to the residual impurity concentration, $\hbar\omega_0 \ll \Delta$. Relaxation time effects are not included in Eq. (5); they lead to the broadening of the collective-mode resonance, but do not influence the parameters such as the spectral weight. Equation (5) gives[7,11] a collective mode at $\omega = \omega_0$, which contains all the oscillator strength with no contributions to $\sigma(\omega)$ from excitations across the single-particle gap, in clear disagreement with the experimental findings. The effect of Coulomb interactions is incorporated[13] by replacing the correlation function $\langle [\delta\Delta, \delta\Delta] \rangle$ with

$$\langle [\delta\Delta, \delta\Delta] \rangle + \frac{4\pi e^2}{q^2} \frac{\langle [\delta\Delta, n] \rangle \langle [n, \delta\Delta] \rangle}{1 - (4\pi/q)(e^2/2) \langle [n,n] \rangle} \,, \quad \text{longitudinal} \,, \tag{6a}$$

$$\langle [\delta\Delta, \delta\Delta] \rangle + \frac{4\pi e^2}{q^2} \frac{\langle [\delta\Delta, j] \rangle \langle [j, \delta\Delta] \rangle}{1 - (4\pi e^2/q^2) \langle [j,j] \rangle} \,, \quad \text{transverse} \,, \tag{6b}$$

for the two modes. Substituting these expressions into Eq. (3), and then into Eq. (2) and assuming that f is independent of frequency, we obtain the collective-mode contribution to the conductivity for both polarizations,

$$\begin{aligned} \sigma_{\text{coll}}(\omega) &= \frac{ne^2}{m_b} i\omega f \left[\omega^2 + i\omega\Gamma_p - \omega_0^2 - \omega_p^2 f \left(1 - \frac{\omega_p^2(1-f)}{\omega^2 + i\omega\Gamma_n} \right)^{-1} \right] \\ &\approx \frac{ne^2}{m} i\omega f \left\{ f(\omega^2 - \omega_p^2 - \omega_0^2 f)^{-1} + (1-f) \frac{1}{\omega^2 + i\omega\Gamma_p^* - \omega_0^2(1-f)} \right\} \,, \end{aligned} \tag{7}$$

where $\omega_p = (4\pi e^2/m_b)^{1/2}$ is the plasma frequency. Γ_n and Γ_p are the phason and quasiparticle damping constants and $\Gamma_p^* = \Gamma_p(1-f) + \Gamma_n f$. For the $(TMTSF)_2X$ salts, $\omega_p = 10\,000$ cm^{-1}, and $\Delta = 15$ cm^{-1}, and it is expected that $\omega_0 < \Delta/\hbar, \omega_p$ We have made use of the fact that ω_0 is smaller than the other frequencies when deriving the final form of $\sigma_{\text{coll}}(\omega)$ in Eq. (7). Note that the first term in Eq. (7) is negligible as $f(\omega_p) = O(2\Delta/\omega_p)^2 \ll 1$. The real part of the complex conductivity is given by

$$\sigma_1(\omega) = \frac{ne^2}{m_b} \left\{ \frac{1}{\omega} \text{Im} f(\omega) + (1-f) f \frac{\omega\Gamma_p^*}{[\omega - \omega_p(1-f)^{1/2}]^2 + (\omega\Gamma_p^*)^2} \right\} \,, \tag{8}$$

where

$$\text{Im} f(\omega) = \frac{\pi}{2} \frac{1}{Z(Z^2-1)^{1/2}} \tanh\left(\frac{\beta\omega}{2} \right) \theta(Z-1) \tag{9}$$

with $Z = \omega/2\Delta$ for both the longitudinal and transverse modes.

In Eq. (8) the first term appearing at frequency $[\omega_p^2 + \omega_0^2 f]^{1/2}$ represents the plasma frequency and is analogous to the plasma frequency in superconductors. As $\omega_p > 2\Delta$, the spectral weight of this high-frequency mode is transferred to the spectral weight of the single-particle excitation spectrum, which has the usual onset at the gap frequency. The second term which appears at a renormalized pinning frequency $\omega_0^* = \omega_0(1-f)^{1/2}$ is due to the pinned collective mode. The temperature dependence of the spectral weight of this mode is given by

$$A_{\text{coll}} = \tfrac{1}{2}\pi(ne^2/m_b)f(1-f) \tag{10}$$

for both modes. According to Eq. (4) A is strongly temperature dependent and for $T \ll T_{\text{SDW}}$, $A_{\text{coll}} \sim \exp(-\beta\Delta)$, giving a small contribution to the total spectral weight. The latter which includes contributions from both the collective mode and single-particle excitations together with the high-frequency mode is constant, given by $A_{\text{total}} = \frac{1}{2}\pi(ne^2/m_b)$. From Eq. (8) we also find that the low-frequency dielectric constant is given by

$$\varepsilon(\omega \to 0) = 1 + f(T,0)\left(\frac{\omega_p}{2\Delta}\right)^2 + f(T,0)\left(\frac{\omega_p}{\omega_0}\right)^2 . \tag{11}$$

The first term is due to single-particle excitations; the second term is due to the collective mode. The latter gives no contribution to ε at frequencies $\Delta > \omega \gg \omega_0^*$, and in this limit

$$\varepsilon(\omega \gg \omega_0^*) = 1 + f(T,0)(\omega_p/2\Delta)^2 , \tag{12}$$

independent of temperature.

The main consequences of the Coulomb effects on the electrodynamics of spin-density waves are as follows. In contrast to what is predicted for $\sigma(\omega)$ in the absence of Coulomb forces, $\sigma_1(\omega)$ has two important contributions, one coming from single-particle excitations at frequencies $\omega > 2\Delta/\hbar$ and one reflecting the collective mode at the frequency ω_0^*. The spectral weight of the collective-mode contribution is strongly temperature dependent, and vanishes in the $T \to 0$ limit, with all the spectral weight associated with the single-particle excitations at zero temperature. As both the longitudinal and transverse mode display the same temperature dependence of the spectral weight, the same conclusion applies for a $\mathbf{q} \to 0$ mode which contains both polarizations due to the symmetry breaking brought about by the impurities.[5]

Next, we discuss the various experimental results on the frequency- and electric-field-dependent response which, we believe give strong support for the importance of Coulomb effects suggested by us. As discussed earlier, the frequency-dependent conductivity observed[8-10] in $(TMTSF)_2PF_6$ and $(TMTSF)_2NO_3$ gives clear evidence for single-particle excitations across the gap, and this contribution contains nearly all the spectral weight associated with $\sigma(\omega)$. The collective-mode contribution observed in the PF_6 salt at $T \simeq 0.5T_{\text{SDW}}$ has a small spectral weight in full agreement with the theory. At $T = 0.5T_{\text{SDW}}$ where the experiments were conducted, $1-f \approx 10^{-2}$ and consequently, the spectral weight A_{coll} is 2 orders of magnitude smaller than the total oscillator strength including both collective-mode and single-particle contributions. The small spectral weight was interpreted earlier[8] as evidence for a large dynamical mass, and the present theory suggests that it arises as the consequence of small spin-density-wave mass and significant Coulomb effects. The dc conductivity is significantly higher in the NO_3 salt than in the PF_6 salt in the SDW state, and $\sigma(\omega < 2\Delta/\hbar)$ is dominated by carrier excitations across the gap, except well below T_{SDW}. Our experiments[10] at $T = 0.2T_{\text{SDW}}$ do not indicate any low-lying mode, and we suggest that this is due to the freezing-out of the collective-mode spectral weight at low temperatures. Low-frequency dielectric-constant measurements conducted on the PF_6 salt[14] also clearly establish a reduced spectral weight associated with the low-frequency excitations. Various experiments conducted at radio frequencies well below T_{SDW} give an upper limit of $\varepsilon < 10^7$ at $T < 0.5T_{\text{SDW}}$. In the absence of Coulomb effects, Eq. (9) would lead, with $\omega_0 = 5$ GHz, to $\varepsilon = 10^9$, 2 orders of magnitude higher than the measured value. In contrast, Eq. (10) leads with $\omega_p = 10000$ cm^{-1} and $\Delta = 15$ cm^{-1} to $\varepsilon = 10^{+7}$ at frequencies $\omega \gg \omega_0^*$ in broad agreement with the upper limit of ε established by experiment.

The threshold field for nonlinear conduction is related to the pinning frequency, and

$$E_T = (2eV_F)^{-1}(1-f)^{-1}\omega_0^{*2} . \tag{13}$$

Consequently, the simple relation[2] between E_T and ω_0^*, involving only the temperature-independent constant, is not valid for spin-density waves. However, from Eqs. (11) and (13) we find that the universal relation[2] between the low-frequency dielectric constant and threshold field $\varepsilon E_T = Ae$ (with A a constant of the order of 1) is not modified by the Coulomb interactions, but a significantly reduced εE_T value could result from experiments where ε is measured at frequencies $\omega \gg \omega_0^*$.

In TMTSF salts, the pinning potential, and consequently ω_0, and also ε and E_T are strongly sample dependent, being determined by residual impurities.[15,16] Consequently, experiments where the ω- and E-dependent conductivity are measured on the same specimens will be required to clarify the relation between the field- and frequency-dependent response.

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