Time-dependent Laudau theory of charge-density waves in transition-metal dichalcogenides*

W. L. McMillan
Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801
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A time-dependent Landau theory is proposed for the transition-metal dichalcogenides coupling the motion of the charge-density waves to the electron liquid. The dissipative terms are related to the metallic resistance. The "phason" or "phonon" excitations of the charge-density wave are found to be overdamped. Resistive anomalies are predicted in the normal state.

I. INTRODUCTION

The anomalous properties of transition-metal dichalcogenides such as $TaSe_2$ have recently been interpreted as being due to charge-density-wave (CDW) formation.[1-3] We have proposed a Landau theory[4] to describe the phase transitions and static distortions of the charge-density-wave structure. For the triple incommensurate charge-density wave, the low-energy distortions are analogous to phonons in a crystal.

In this paper, we write down a time-dependent version of the Landau theory and calculate the dynamic behavior of these "phonon" modes. The modes are found to be overdamped. The motion of the CDW order parameter is coupled to the flow of the electron liquid, and this flow is resistive. The electrical resistance provides the dissipative mechanism in the Landau theory. We calculate the resistance anomaly in the normal state due to order-parameter fluctuations and to impurities. This theory should provide the basis for a study of pinning of the CDW to impurities, but we have not yet undertaken such a study.

II. TIME-DEPENDENT LANDAU THEORY

The order parameter for the charge-density wave is the electronic charge density of the d-band conduction electrons. We write for the electronic charge density

$$\rho(\vec{r}) = \rho_0(\vec{r})[1 + \alpha(\vec{r})] \ , \tag{1}$$

where $\rho_0(\vec{r})$ is the charge density in the normal state, and $\alpha(\vec{r})$ is the real order parameter. In order to describe the observed triple CDW state, it is convenient to use three complex order parameters ψ_i, one for each CDW,

$$\alpha(\vec{r}) = \mathrm{Re}[\psi_1(\vec{r}) + \psi_2(\vec{r}) + \psi_3(\vec{r})] \ . \tag{2}$$

The free energy of one layer given previously[4] is then

$$F = \int d^2r \Big[a\alpha^2 - b\alpha^3 + c\alpha^4 + d(|\psi_1\psi_2|^2 + |\psi_2\psi_3|^2 + |\psi_3\psi_1|^2) + U\rho_0\alpha + e\sum_i |(\vec{q}_i\cdot\vec{\nabla} - q_i^2)\psi_i|^2 + f\sum_i |\vec{q}_i\times\vec{\nabla}\psi_i|^2 \Big], \tag{3}$$

where $U(\vec{r})$ is the impurity potential, and $|\vec{q}_1| = |\vec{q}_2| = |\vec{q}_3| = 2\pi/\lambda$, where λ is the wavelength of the incommensurate CDW. The three $\vec{q}_i$ vectors lie in the ΓM directions in the Brillouin zone 120° apart. In order to study the commensurate CDW, one must assume that a–f are periodic with the periodicity of the crystal lattice. In the present paper, we study only the incommensurate wave and replace a–f by their constant parts a_0–f_0. We assume

$$a_0 = a'(T - T^*), \tag{4}$$

and that the other parameters are temperature independent

The CDW order parameter $\alpha(\vec{r})$ describes the short-wavelength components of charge density. The long-wavelength components of the d-band conduction-electron charge density are treated as an incompressible fluid. The macroscopic charge density is ρ_0, and the incompressible fluid motion is described by the velocity field $\vec{v}(\vec{r})$, where

$$\vec{\nabla}\cdot\vec{v} = 0. \tag{5}$$

The kinetic energy density is

$$(m^*\rho_0/2e)\,\vec{v}^2 \ , \tag{6}$$

where m^* is an average d-band effective mass.

We write two terms in the power dissipation

$$D = \int d^2r \left(\eta\vec{v}^2 + \sum_i \gamma \left|\frac{\partial\psi_i}{\partial t} + \vec{v}\cdot\vec{\nabla}\psi_i\right|^2\right), \tag{7}$$

where the first term arises from the electrical resistance when the electron fluid moves relative to the crystal lattice, and the second term is the dissipation arising from the motion of the CDW relative to the electron fluid. In Sec. III, we will relate γ and η.

The equation of motion for the electron fluid is

$$\frac{m^*\rho_0}{e}\frac{d\vec{v}}{dt} = \rho_0\vec{E} - \frac{1}{2}\frac{\partial D}{\partial\vec{v}} \ , \tag{8}$$

where $\vec{E}$ is the electric field.

Neglecting inertial effects, the equation of motion for the order parameter is

$$\frac{\partial F}{\partial \psi_i^*} = -\frac{1}{2}\frac{\partial D}{\partial \dot{\psi}_i^*}, \tag{9}$$

where

$$\dot{\psi}_i = \frac{\partial \psi_i}{\partial t}.$$

III. VISCOSITY COEFFICIENTS

We now relate the two viscosity coefficients to the resistivity. Applying a uniform electric field to the pure metal, the equations of motion are

$$\frac{m^*\rho_0}{e}\frac{d\vec{v}}{dt} = \rho_0\vec{E} - \eta\vec{v} - \frac{1}{2}\sum_i \vec{\nabla}\psi_i\left(\frac{\partial \psi_i^*}{\partial t} + \vec{v}\cdot\vec{\nabla}\psi_i^*\right) + \text{c.c.}, \tag{10}$$

$$\frac{\partial \psi_i}{\partial t} + \vec{v}\cdot\vec{\nabla}\psi_i = 0 \tag{11}$$

In the steady state, the CDW moves with the electron liquid, and the current density is

$$\vec{j} = \rho_0\vec{v} = \rho_0^2\vec{E}/\eta = \sigma\vec{E}, \tag{12}$$

where σ is the electrical conductivity. Thus,

$$\eta = \rho_0^2/\sigma \tag{13}$$

for the pure metal. If the CDW is pinned to the lattice by impurities, dislocations, or grain boundaries, we have

$$\frac{\partial \psi_i}{\partial t} = 0, \tag{14}$$

instead of Eq. (11), and we find

$$\sigma = \rho_0^2/(\eta + \tfrac{3}{2}\gamma\psi_0^2 q_1^2). \tag{15}$$

Up to this point, we have considered the charge in the CDW and the uniform charge of the electron fluid as two separate variables obeying macroscopic equations of motion. In order to relate the two viscosity coefficients, we temporarily adopt a more microscopic picture and treat the charge in the CDW as part of the macroscopic charge.

Consider a single CDW moving with respect to the electron fluid:

$$\psi_1(\vec{r}) = \psi_0 e^{i(\vec{q}_1\cdot\vec{r}-\omega t)}, \tag{16}$$

$$\rho(\vec{r}) = \rho_0[1 + \psi_0\cos(\vec{q}_1\cdot\vec{r} - \omega t)]. \tag{17}$$

From the continuity equation, we have

$$\vec{v}(\vec{r}) = (\rho_0\psi_0\omega\vec{q}_1/q_1^2)\cos(\vec{q}_1\cdot\vec{r} - \omega t). \tag{18}$$

The dissipation per unit volume is

$$\int d^3r\ \eta\vec{v}^2 = \frac{\eta\rho_0^2\psi_0^2\omega^2}{2q_1^2}, \tag{19}$$

which is represented in the original physical picture by the second term of Eq. (7).

$$\int d^3r\ \gamma\left|\frac{\partial \psi_1}{\partial t}\right|^2 = \gamma\psi_0^2\omega^2. \tag{20}$$

Equating (19) and (20), we find

$$\gamma = \eta\rho_0^2/2q_1^2. \tag{21}$$

This microscopic picture would be quantitative if the wavelength of the CDW were much longer than an atomic length. This is not the case for CDW's since the wavelength is directly related to the Fermi wave vector $\lambda = \pi/k_F$. Thus, we expect (21) to provide only an order of magnitude estimate for γ. We now return to the original physical picture treating the CDW and the electron fluid as separate variables.

IV. FLUCTUATION-MODE RELAXATION TIMES

In the normal state, we write

$$\psi_i(\vec{r}) = \sum_q \psi_q e^{i(\vec{q}+\vec{q}_1)\cdot\vec{r}} \tag{22}$$

$$\vec{v}(\vec{r}) = \sum_q \vec{v}_q e^{i\vec{q}\cdot\vec{r}}. \tag{23}$$

The fluctuation modes are uncoupled with the fluid relaxation time

$$\tau_q^v = m^*\rho_0/e\eta, \tag{24}$$

and the CDW relaxation time

$$\tau_q^\psi = \gamma/2\epsilon_q, \tag{25}$$

$$\epsilon_q = \tfrac{1}{2}a_0 + e_0(\vec{q}_1\cdot\vec{q})^2 + f_0(\vec{q}_1\times\vec{q})^2. \tag{26}$$

In the incommensurate phase, the modes are uncoupled at long wavelength with τ^v and τ^ψ given by (24) and (25) with

$$\epsilon_q^t = (\tfrac{1}{4}e_0 + \tfrac{3}{4}f_0)\,q_1^2q^2, \tag{27}$$

$$\epsilon_q^l = (\tfrac{3}{4}e_0 + \tfrac{1}{4}f_0)\,q_1^2q^2 \tag{28}$$

for the transverse and longitudinal "phonon" modes of the CDW "lattice." These modes are overdamped with long relaxation times at long wavelength.

V. RESISTIVE ANOMALIES

A. Pure metal

A charge-density wave opens up an energy gap in the band structure at the Fermi surface, and this affects the resistivity in two ways. First of all, carriers freeze out across the gap and the number of charge carriers is reduced provided the CDW is pinned to the lattice; if the CDW moves with the electron liquid, this does not affect the conductivity. Secondly, the density of states available for electron-phonon scattering is reduced and the conductivity increased. For the pure metal, the second effect can be represented in the context of the Landau theory as a reduction in η and we *assume* that

the following expression is valid:

$$\eta = \eta_0 - \eta_1 \sum_i |\psi_i|^2 . \tag{29}$$

In the normal state, we take the thermal expectation value of $|\psi_i|^2$ and find

$$\langle |\psi_i|^2 \rangle = [kT/4\pi(e_0 f_0 q_1^4)^{1/2}] \ln(\xi/\bar{a}) , \tag{30}$$

where ξ is the longitudinal correlation length

$$\xi = [2 e_0 q_1^2 / a'(T - T^*)]^{1/2} \tag{31}$$

and $\bar{a}$ is the lattice spacing. The conductivity of the pure metal increases logarithmically with $T - T^*$.

$$\sigma = \rho_0^2 \Big/ \left[\eta_0 - \frac{3\eta_1 kT}{4\pi(e_0 f_0 q_1^4)^{1/2}} \ln\left(\frac{\xi}{\bar{a}}\right) \right] . \tag{32}$$

B. Impurity resistance

According to Ref. (4), an impurity is dressed with a charge-density wave cloud even in the normal state. For an impurity at the origin, we have

$$\psi_i(\vec{r}) = -\frac{U_0}{2} \sum_{\vec{q}} \frac{e^{i(\vec{q}+\vec{q}_i)\cdot\vec{r}}}{\frac{1}{2} a_0 + e_0(\vec{q}\cdot\vec{q}_i)^2 + f_0(\vec{q}\times\vec{q}_i)^2} , \tag{33}$$

where U_0 is the space integral of the impurity potential. The electron fluid moving past the impurity with velocity $\vec{v}$ experiences an additional dissipation

$$\int d^2r \sum_i \gamma |\vec{v}\cdot\vec{\nabla}\psi_i|^2 = \frac{3\gamma U_0^2 v^2}{32\pi(e_0 f_0)^{1/2}} \ln(\xi/\bar{a}) , \tag{34}$$

which leads to an impurity resistivity of

$$\frac{1}{\sigma_I} = \frac{3\gamma U_0^2 N}{32\pi\rho_0^2(e_0 f_0)^{1/2}} \ln\left(\frac{\xi}{\bar{a}}\right) \tag{35}$$

in addition to the electron-phonon resistivity given by (32), here N is the impurity density. For the dirty material, the correlation length is finite at the phase transition.

Thus, we expect a pretransition increase in conductivity for the pure metal and a decrease for the dirty metal.

VI. CONCLUSIONS

We have proposed a dynamical form of the Landau theory of charge-density waves in transition-metal dichalcogenides. We have found that the "phonon" modes of the charge-density wave lattice are overdamped with long lifetimes at long wavelength. We have also predicted resistive anomalies in the normal state.

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