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Summary

The Colour of Charge Density Wave Order

The beauty of condensed matter physics lies in the emergent orders out of the systems consisting of a huge number of particles. The collective behaviours of the massive particles refuse to be reduced into a single particle's attributes [173]. At long wavelength and low energy scales, the properties of the many-body system are dominated by the global parameters, such as symmetry and topology, and the details of the system's composition or interaction do not matter as shown in Chap. 2 [174, 175]. By introducing symmetry into physics, we can revisit the physical world with a different angle, a better angle in some sense. We can easily understand the connections between seemingly different phenomena, such as the superconductivity and the charge density wave or the Peierls instability and the Stoner instability. However, if we move from this "elegant" domain of low energies and long distances to the more complicated domain of high energies and short distances, the kind and strength of interactions and the physical compositions play more important roles than before [175]. As we discussed in previous chapters, CDW order lies on the delicate edge between the two domains. To briefly explain this, the coherence length of CDW orders is normally in the order of 10 nm [176, 177], which is larger than the symmetry breaking phase in strongly correlated materials ($\sim$1 nm) but much smaller than the conventional BCS superconductors (100 $\sim$1000 nm) [178, 179]. So in TMDCs, the symmetry and interaction both affect the physical properties and give rise to so many different CDW orders.

Chap. 4, therefore, is a retro study and a test on the weakly-coupled picture of the CDW order. For the first time, the FIR spectra of 1T-VSe_2 are brought to light. From the optical functions, such as reflectivity and the optical conductivity $\sigma_1(\omega)$, we observe the very typical optical response of the general symmetry-breaking mechanism. In the CDW state, the reflectivity approaches 1 at low energies and the spectral weight within the same energy range depletes. The Drude response persists in the CDW phase and is suppressed as the temperature decreases. First, we investigate the spectral weight transfer with the finite energy cut-off. With different energy cut-offs, the spectral weight as the functions of the temperature possesses an obvious kink at T_c when part of the spectral weight is transferred away from the selected energy windows. We conclude that the FIR spectral weight transfer has two directions: one to the energy range below our detection limit and the other to the higher energies as the result of the electronic structure's reconstruction. Then we study the asymptotic behaviours of the observed optical functions. Due to the distinct optical responses of the single-particle and the collective excitation, we can split the optical response of the collective excitation in the CDW state and confirm its existence in VSe_2, i.e. the sliding mode. The spectral weight that is transferred below the detecting limit fits well with the weight of the sliding mode obtained from the asymptotic behaviour of the dielectric function $\varepsilon_1(\omega)$. So far it seems to be a nice and elegant example of the Goldstone theorem, where we have the (possibly) gapless excitation protected by the single particle gap after the symmetry of the system is spontaneously broken. However, once we take a closer look, can we interpret the spectral weight depletion in the FIR range $\sigma_1(\omega)$ as opening an optical gap? Our collaborators already show that in VSe_2 the nesting alone is not enough to explain its CDW instability and the nesting region is small on the Fermi surface [14]. The momentum dependence of the electron-phonon coupling strength is necessary to obtain the right wave vector of the incommensurate charge modulation in VSe_2. In the end, only a small portion of the Fermi surface will be gapped. With such a premise, the large and clear spectral weight depletion is not likely to be the optical gap opened on the Fermi surface. To verify this deduction, we calculate the matrix elements of the possible optical transitions in the Brillouin zone. With a limited gapped area on the Fermi surface, the optical gap can still be detected if the matrix elements in the corresponding area are large. The calculation result, however, does not support the gap opening in $\sigma_1(\omega)$ since the maxima of the matrix elements do not overlap with the gapped region. To understand the spectral weight depletion and the suppression of the Drude component, we propose several possible mechanisms including the opening of the optical gap which we just discussed. Combined with our transport data, the most likely scenario we conclude is the temperature dependence of the low energy bands that reduces the number of free electrons at low temperatures. Furthermore, we observe four optical phonons in the FIR range that are only present in the CDW state. We conclude that VSe_2 conforms with a weakly coupled CDW system where symmetry breaking is the dominant factor of the low energy excitations.

Chap. 5 deals with the system where the energy scale is higher and the characteristic length is shorter. We study the Se doped 1T-TaS_2 at two different doping levels. This group of materials features non-negligible electronic correlations and strong interlayer coupling. It possesses various CDW orders in the phase diagram. Samples with the doping level $x = 0.8$ have the bifurcation of the low-temperature states that depends on the cooling history and the cooling rate, while samples with the doping level $x = 1.0$ do not. One of the possible low-temperature states is meta-stable, which we think is related to the hidden/mosaic state observed in pristine TaS_2. The other is the CCDW ground state. Based on our optical data, we plot the phase diagram that contains the island of the meta-stable state. We obtain the optical functions of each CDW phase for the two doping levels and observe several optical gaps opened in different CDW orders. As the CDW gap of the ICCDW phase is not obvious in the spectra, we turn our focus to the gap related to the meta-stable state. We observe a strong spectral weight depletion below 100 meV during the phase transition from the NCCDW state to the meta-stable state during cooling, and another optical gap with a different shape during the transition from the meta-stable state to the CCDW state during heating. By studying the temperature evolution of the normalised optical conductivity, we attribute the formations of the two gaps to different physical mechanisms. The gap that is associated with the formation of the meta-stable state during cooling conforms with the scenario where the low energy spectral weight is gradually depleted. Its size is approximately fixed during cooling while the spectral weight continuously decreases like the pseudogap observed in the cuprate superconductors. The other gap formed in the CCDW state, however, shows a similar temperature evolution with the opening of the spontaneous symmetry-breaking gap, which has a certain degree of temperature dependence on its gap size. Considering the mosaic pattern of domains and the finite density of states observed in the hidden state, we conclude that the meta-stable state is obtained through fast cooling when the stacking faults lead to the formation of dense domain walls. In each domain, the commensurate CDW order is with a very short coherence length. No long-range order is formed in this stage, and the depleted density of states near the Fermi surface accounts for the formation of the pseudogap. By increasing the temperature, the stacking faults can be corrected by the thermal fluctuation and the system relaxes into the true ground state. The long-range order is established then, which explains the gap-opening scenario we observe in the heating process.

Samenvatting

De schoonheid van de natuurkunde van gecondenseerde materie ligt in de emergentie van de systemen die bestaan uit een enorm aantal deeltjes. Het collectieve gedrag van de massieve deeltjes weigert te worden gereduceerd tot de eigenschappen van één enkel deeltje. Op lange golflengte- en lage energieschalen worden de eigenschappen van het veel-deeltjes-systeem gedomineerd door globale parameters, zoals symmetrie en topologie, en doen de details van de samenstelling van het systeem of van de interactie er niet toe, zoals beschreven in hoofdstuk 2 [174, 175]. Door symmetrie in de natuurkunde te introduceren, kunnen we de natuurkundige wereld vanuit een andere hoek bekijken, een betere hoek in zekere zin. We kunnen gemakkelijk de verbanden begrijpen tussen schijnbaar verschillende verschijnselen, zoals supergeleiding en ladingsdichtheidsgolven (CDW) of de Peierls-instabiliteit en de Stoner-instabiliteit. Als we echter van dit “elegante” domein van lage energieën en lange afstanden overgaan naar het meer gecompliceerde domein van hoge energieën en korte afstanden, spelen het soort en de sterkte van de interacties en de fysische samenstellingen een belangrijkere rol dan voorheen. Zoals wij in vorige hoofdstukken hebben besproken, ligt de CDW-orde op het dunne rand tussen beide domeinen. Om dit kort uit te leggen: de coherentielengte van CDW-ordes ligt normaal gesproken in de orde van 10 nm [176, 177], wat groter is dan de symmetriebrekende fase in sterk gecorreleerde materialen ($\sim$1 nm) maar veel kleiner dan de conventionele BCS-supergeleiders (100 $\sim$1000 nm) [178, 179]. Dus in TMDC’s beïnvloeden de symmetrie en de interactie beide de fysische eigenschappen en geven aanleiding tot zoveel verschillende CDW-ordes.

Hoofdstuk 4 is daarom een retrostudie en een test van het zwakgekoppelde beeld van de CDW-orde. Voor het eerst worden de FIR-spectra van 1T-VSe$_2$ aan het licht gebracht. Uit de optische functies, zoals de reflectiviteit en de optische geleidbaarheid $\sigma_1(\omega)$, blijkt de zeer typische optische respons van het algemene symmetrie-brekende mechanisme. In de CDW-toestand benadert de reflectiviteit 1 bij lage energieën en vermindert het spectraal gewicht binnen hetzelfde energiegebied. De Drude respons blijft bestaan in de CDW-fase en wordt onderdrukt naarmate de temperatuur afneemt. Eerst onderzoeken we de overdracht van het spectrale gewicht met de eindige energieafsnijding. Bij verschillende energieafsnijdingen vertoont het spectrale gewicht als functie van de temperatuur een duidelijke knik bij T$_c$ wanneer een deel van het spectrale gewicht wordt overgebracht uit het geselecteerde energie gebied. Wij concluderen dat de overdracht van het FIR spectrale gewicht in twee richtingen verloopt: een naar het energiegebied onder onze detectielimiet en een naar de hogere energieën als gevolg van de reconstructie van de elektronische structuur. Vervolgens bestuderen we het asymptotische gedrag van de waargenomen optische functies. Door de verschillende optische reacties van de enkel-deeltje en de collectieve excitatie kunnen wij de optische reactie van de collectieve excitatie in de CDW-toestand splitsen en het bestaan ervan in VSe_2, d.w.z. de glijdende modus, bevestigen. Het spectrale gewicht dat onder de detectielimiet wordt overgedragen past goed bij het gewicht van de glijdende modus dat wordt verkregen uit het asymptotische gedrag van de diëlektrische functie $\varepsilon_1(\omega)$. Tot dusver lijkt het een mooi en elegant voorbeeld van het Goldstone theorema, waarbij we de (mogelijk) kloofloze excitatie beschermd zien door de kloof van één enkel deeltje nadat de symmetrie van het systeem spontaan is gebroken. Kunnen we echter, als we beter kijken, de afname van het spectrale gewicht in het FIR-gebied van $\sigma_1(\omega)$ interpreteren als het openen van een optische kloof? Onze partners hebben al laten zien dat in VSe_2 de Fermi-oppervlak nesteling alleen niet voldoende is om de CDW-instabiliteit te verklaren en dat het nestelingsgebied klein is op het Fermi-oppervlak [14]. De impulsafhankelijkheid van de elektron-fononkoppelingssterkte is noodzakelijk om de juiste golfvector van de onevenredige ladingsmodulatie in VSe_2 te verkrijgen. Uiteindelijk zal slechts een klein deel van het Fermi-oppervlak ontsloten zijn. Met een dergelijke vooronderstelling is het niet waarschijnlijk dat de grote en duidelijke spectrale gewichtsafname door het openen van de optische kloof op het Fermi-oppervlak komt. Om deze conclusie te verifiëren, berekenen wij de matrixelementen van de mogelijke optische overgangen in de Brillouin-zone. Bij een beperkte opening op het Fermi-oppervlak kan de optische kloof nog steeds worden opgespoord als de matrixelementen in het overeenkomstige gebied groot zijn. Het resultaat van de berekening ondersteunt echter niet de opening van de kloof in $\sigma_1(\omega)$, aangezien de maxima van de matrixelementen niet overlappen met het gebied waar de kloof zich bevindt. Om de afname van het spectrale gewicht en de onderdrukking van de Drude-component te begrijpen stellen wij verschillende mogelijke mechanismen voor, waaronder de opening van de zojuist besproken optische kloof. In combinatie met onze transportgegevens concluderen wij dat het meest waarschijnlijke scenario de temperatuurafhankelijkheid van de lage energiebanden is, waardoor het aantal vrije elektronen bij lage temperaturen afneemt. Verder observeren wij vier optische fononen in het FIR-bereik die alleen aanwezig zijn in de CDW-toestand. Wij concluderen dat VSe_2 voldoet aan een zwak gekoppeld CDW-systeem waarin symmetriebreking de dominante factor is van de lage-energie-excitaties.

Hoofdstuk 5 behandelt het systeem met een hogere energieschaal en een kortere karakteristieke lengte. Wij bestuderen de Se-gedoteerde 1T-TaS_2 op twee verschillende doteringsniveaus. Deze groep materialen heeft niet-verwaarloosbare elektronische correlaties en een sterke koppeling tussen de lagen. Zij bezit verschillende CDW-ordes in het fasediagram. Monsters met het doteringsniveau $x = 0{,}8$ hebben een bifurcatie van de lage-temperatuurtoestanden die afhangt van de koelgeschiedenis en de koelsnelheid, terwijl monsters met het doteringsniveau $x = 1{,}0$ dat niet hebben. Een van de mogelijke lage-temperatuurtoestanden is metastabiel, wat volgens ons verband houdt met de verborgen/mosaïsche toestand die in ongerept TaS_2 wordt waargenomen. De andere is de CCDW-grondtoestand. Op basis van onze optische gegevens tekenen we het fasediagram dat het eiland van de metastabiele toestand bevat. Wij verkrijgen de optische functies van elke CDW-fase voor de twee doteringsniveaus en observeren verschillende optische kloven die in verschillende CDW-ordes worden geopend. Aangezien de CDW-kloof van de ICCDW-fase niet duidelijk is in de spectra, richten wij onze aandacht op de kloof die gerelateerd is aan de metastabiele toestand. Wij nemen een sterke spectrale gewichtsvermindering onder 100 meV waar tijdens de faseovergang van de NCCDW-toestand naar de metastabiele toestand tijdens afkoelen, en een andere optische kloof met een andere vorm tijdens de overgang van de metastabiele toestand naar de CCDW-toestand tijdens opwarmen. Door de temperatuurevolutie van de genormaliseerde optische geleiding te bestuderen, schrijven wij de vorming van de twee kloven toe aan verschillende fysische mechanismen. De kloof die samenhangt met de vorming van de metastabiele toestand tijdens afkoelen is in overeenstemming met het scenario waarin het laag energetische spectrale gewicht geleidelijk wordt uitgeput. De grootte ervan ligt tijdens het afkoelen ongeveer vast, terwijl het spectrale gewicht voortdurend afneemt, zoals de pseudo-kloof die in de cupraten supergeleiders wordt waargenomen. De andere in de CCDW-toestand gevormde kloof vertoont echter een vergelijkbaar temperatuurverloop met de opening van de spontane symmetrie-brekende kloof, die een zekere mate van temperatuurafhankelijkheid aan de kloofgrootte heeft. Gezien het mozaïekpatroon van domeinen en de eindige toestandsdichtheid in de verborgen toestand, concluderen wij dat de metastabiele toestand wordt verkregen door snelle afkoeling wanneer de stapelfouten leiden tot de vorming van dichte domeinwanden. In elk domein heeft de overeenkomstige CDW-orde een zeer korte coherentielengte. In dit stadium wordt geen lange-afstandsorde gevormd, en de verarmde toestandsdichtheid nabij het Fermi-oppervlak verklaart de vorming van de pseudo-kloof. Door de temperatuur te verhogen kunnen de stapelfouten worden gecorrigeerd door thermische fluctuatie en ontspant het systeem zich naar de ware grondtoestand. De lange-afstandsorde wordt dan gevestigd, wat het kloof-opening scenario verklaart dat we waarnemen tijdens het verhittingsproces.

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