Charge-Density-Wave-Induced Modifications to the Quasiparticle Self-Energy in 2H-$TaSe_2$

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The self-energy of the photohole in 2H-$TaSe_2$ is measured by angle-resolved photoemission spectroscopy as a function of binding energy and temperature. In the charge-density wave (CDW) state, a structure in the self-energy is detected at ~65 meV that cannot be explained by electron-phonon scattering. A reduction in the scattering rates below this energy indicates the collapse of a major scattering channel with the formation of the CDW state accompanying the appearance of a bosonic "mode" in the excitation spectrum of the system.

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In general, the quasi-2D electronic systems have a weaker tendency towards the formation of charge-density wave (CDW) and spin-density wave (SDW) instabilities than their 1D counterparts. This is because the Fermi surfaces in 2D (generalized cylinders) can be only partially nested. However, under favorable nesting conditions, or driven by saddle-point singularities, the electronic susceptibility may be enhanced enough for a CDW to develop. Since the Fermi surface is only partially gapped, the system may retain metallic character even in the CDW state. The 2D character and the existence of an anisotropic gap make these systems similar to the high T_C superconductors (HTSCs). In particular, the low-energy one-electron-like excitations ("quasiparticles") may show certain similarities. Angle-resolved photoelectron spectroscopy (ARPES) represents a powerful technique for studying the one-electron properties in low-dimensional materials. It measures the occupied component of the single-particle spectral function $A(\mathbf{k}, \omega)$, thus providing direct insight into the fundamental many-body interactions of the system. The technique has the important advantage that it momentum resolves. Studies of momentum-resolved gaps and self-energies, as well as their temperature dependencies, can help in understanding the fundamental mechanisms responsible for producing a variety of phases in strongly correlated low-dimensional systems. As such, not only are the more exotic systems currently under investigation, but many "simple" systems are also being reinvestigated. Indeed, in recent studies, the effects of the electron-phonon coupling on single-particle states have been directly observed [1,2]. The change in a state's width and dispersion near the Fermi level provide a signature of the electron-phonon coupling. However, similar effects have been detected in some systems where such a correspondence cannot be established. For example, in the cuprate superconductor, $Bi_2Sr_2CaCu_2O_{8+\delta}$, a "kink" in the dispersion and a narrowing of the state have also been detected [3]. However, in that system, the doping and temperature dependencies rule out the electron-phonon coupling as the mechanism and point towards a different type of coupling resulting in modifications of the single-particle spectrum [4].

In the present paper, we report a detailed ARPES study of low-energy electronic excitations in 2H-$TaSe_2$. 2H-$TaSe_2$ undergoes a second-order transition to an incommensurate CDW at 122 K, followed by a first-order lock-in transition to a 3×3 commensurate CDW phase at 90 K [5]. The driving mechanism for the CDW transition is still under debate. The two most recent ARPES studies have provided alternative explanations: Liu *et al.* [6] found the opening of a large gap in the extended saddle band region when the system entered the CDW state; Straub *et al.* [7], in a study of 2H-$NbSe_2$, concentrated on the nesting properties of the Fermi surface, and found large portions that, although not gapped, could be nested. In both studies, the corresponding "nesting vectors" cannot be easily related to the CDW wave vector.

In the present study we concentrate on the properties of the one-particle excitation spectrum and provide evidence of a possible collective mode that exists in the CDW state. This mode causes significant renormalization of the one-particle spectrum at low energies. Simultaneously, the one-particle scattering rates at low energies are greatly reduced in the CDW state, indicating a collapse of the phase space available for scattering. The mode may be attributed to the electron-hole pair creation in those regions that are gapped, or more precisely, to the scattering from fluctuations associated with the CDW order parameter. The observation of a mass or velocity renormalization in the ungapped region of the Fermi surface accompanying the formation of a gap elsewhere in the zone is very reminiscent of the behavior observed for the high T_C superconducting materials, as discussed above [3,4].

The experiment reported here was carried out on a high resolution photoemission facility based on undulator beam line U13UB at the National Synchrotron Light Source. This facility employs a Scienta SES-200 electron spectrometer which simultaneously collects a large energy (0.5 to 1 eV) and angular window (~12°) of the photoelectrons. This reduces the time needed for data acquisition

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and ensures that the whole region of interest in k space is recorded under identical conditions of temperature and surface cleanliness. The combined instrumental energy resolution was set to $\sim$6 meV, small enough to make no significant contribution to the photoemission peak widths measured here. The angular resolution was better than $\sim$0.2°, translating into a momentum resolution of $\sim$0.005 $Å^{-1}$ at the 15.2 eV photon energy used in the study. Samples, grown by a chemical reaction with iodine as a transport agent [8], were mounted on a liquid He cryostat and cleaved *in situ* in the UHV chamber with base pressure 3×10^{-9} Pa. The temperature was measured using a silicon sensor mounted near the sample and was rechecked by fitting the Fermi edge, taking account of the full experimental energy resolution.

Figure 1 shows the photoemission intensity, recorded in the CDW state at $T = 34$ K, as a function of binding energy and momentum along the line through the two-dimensional Brillouin zone indicated in the inset of the figure. The figure shows a band crossing the Fermi level at a point on the holelike Fermi surface S_C, centered at Γ [9]. This particular Fermi surface is preserved in the CDW transition with no gap forming, independent of $\mathbf{k}_F$ [6,7]. The most remarkable feature in Fig. 1 is the kink in the band's dispersion, accompanied by a sharpening in the vicinity of the Fermi level. Also shown in the figure are cuts through the intensity map at constant momenta or energy distribution curves (EDCs). In this energy range, the EDCs show a two-peaked structure, behavior that is characteristic of the interaction of the photohole with some excitation of the system with energy range limited approximately to the energy scale of the kink (see [1,2] and references therein). Such an interaction renormalizes the mass and lifetime of the photohole but conserves the total charge. The self-energy, $\Sigma(\mathbf{k}, \omega)$, describes this interaction, the real part corresponding to the shift in energy and the imaginary part the scattering rate or inverse lifetime. Both components of the self-energy may be extracted directly from an ARPES spectrum since the spectral intensity $I(\mathbf{k}, \omega)$ is given by $I^0(\mathbf{k})A(\mathbf{k}, \omega)f(\omega)$ where $A(\mathbf{k}, \omega)$ represents the spectral function, $I^0(\mathbf{k})$ incorporates the dipole matrix elements, and $f(\omega)$ is the Fermi distribution function. As recently noted by LaShell *et al.* [2], in the limit of a momentum independent self-energy and matrix elements, the spectral intensity takes the simple form

$$I(\mathbf{k}, \omega) \propto \frac{\mathrm{Im}\Sigma(\omega)}{[\omega - \epsilon_{\mathbf{k}} - \mathrm{Re}\Sigma(\omega)]^2 + [\mathrm{Im}\Sigma(\omega)]^2} f(\omega), \tag{1}$$

where ϵ_k is the noninteracting dispersion. The real and imaginary components of the self-energy, $\mathrm{Re}\Sigma(\omega)$ and $\mathrm{Im}\Sigma(\omega)$, may then be extracted directly from a momentum-distribution curve (MDC), the intensity as a function of momentum at constant binding energy. With this method, the fitting is possible without imposing any particular model for the interaction. We approximate the

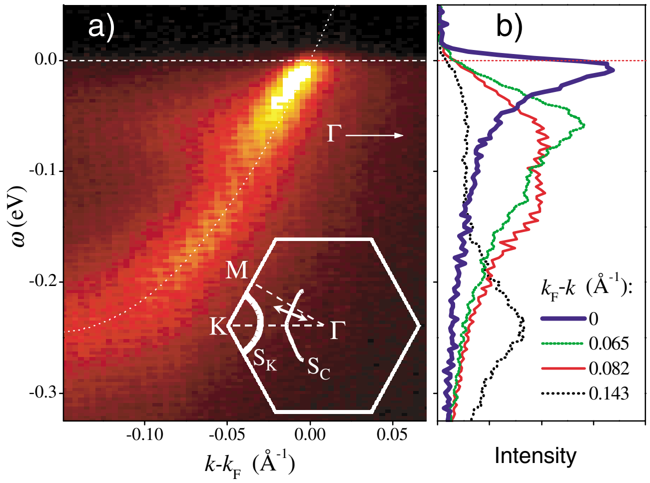

FIG. 1 (color). (a) The photoemission intensity in the CDW state ($T = 34$ K) as a function of binding energy and momentum along the line indicated in the inset by the double-headed arrow. The intensity is represented by a false color map, with yellow and white representing the highest intensity. The dispersing state is a part of the holelike Fermi surface S_C, centered at Γ. This Fermi surface is not gapped in the CDW state. (b) EDCs, measured for several momenta as discussed in the text.

noninteracting dispersion with a second-order polynomial [10] that coincides with the measured dispersion at $k = k_F$ and at higher binding energies, close to the bottom of the band: $\mathrm{Re}\Sigma = 0$ for $\omega = 0$ and $\omega < -200$ meV. Figure 2 shows several MDCs with corresponding fits. In contrast to the line shapes in Fig. 1(b) for EDCs, the line shapes in Fig. 2 are approximately Lorentzian at low binding energies developing an asymmetry at higher binding energies. The latter asymmetry mostly reflects the quadratic term in the noninteracting dispersion. The advantage of using MDCs in the analysis is obvious in that the self-energies are more dependent on energy than on momentum.

The results of the fitting procedure, which produces pairs of $\mathrm{Re}\Sigma$ and $\mathrm{Im}\Sigma$ for every MDC are shown in Fig. 3 for several temperatures. We have also included $\mathrm{Im}\Sigma$ obtained by fitting EDCs when the latter have a Lorentzian line shape. The real part of the self-energy is concentrated in the region of binding energies less than 150 meV. At the lowest temperature, it has a maximum at a binding energy of $\sim$65 meV, approximately coincident with the value corresponding to the sharp drop in $\mathrm{Im}\Sigma$. Such behavior is indicative of the scattering of the photohole from some collective excitation or "mode" of the system. The striking similarity with the behavior recently observed in an ARPES study of the photohole interacting with phonons [1,2] would point to the electron-phonon coupling as the source of this behavior. This would imply the presence of $\sim$70 meV phonons in the CDW state. However, the highest calculated and measured phonon frequency is $\sim$40 meV [11]. The measured temperature dependence of the self-energy also argues against phonons.

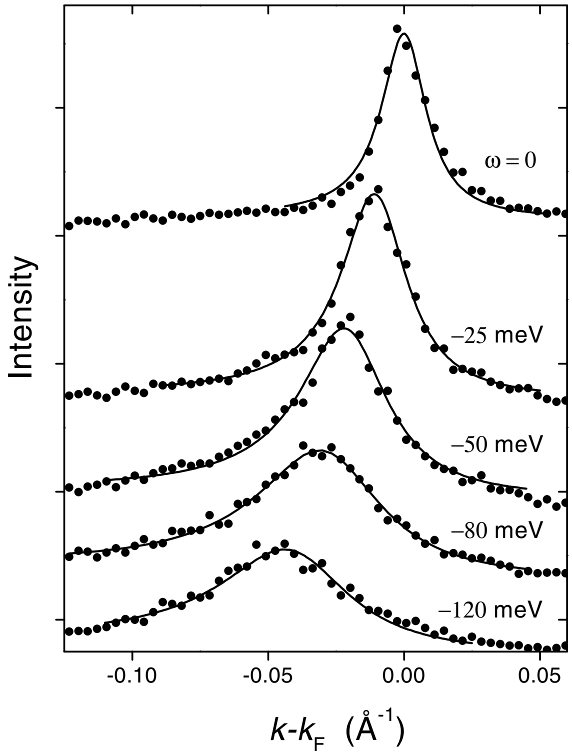

FIG. 2. MDCs, measured at different binding energies (symbols), fitted with a momentum-independent spectral function (solid lines) as discussed in the text.

With increasing temperature, the peak in ReΣ loses its magnitude and the structure shifts to lower energies. At a temperature of 111 K, only a small peak is left at a binding energy of ∼30 meV, and this survives in the normal state to at least 160 K. This peak may be of the same CDW origin, but may also be caused by conventional electron-phonon coupling, since it is within the range of the phonon spectrum.

At low temperatures the imaginary part of the self-energy or scattering rate shows a sharp reduction for binding energies lower than 70 meV. As the temperature increases, this reduction becomes less pronounced. A more detailed temperature dependence for ImΣ(0) is shown in Fig. 4. The in-plane resistivity and the Drude scattering rate measured by Vescoli *et al.* [12] are also shown. In a simple Drude-type model, the conductivity (the inverse of resistivity) in a 2D system is proportional to the integral of $k_F l(\mathbf{k}_F)$ over the Fermi surface, weighted by geometrical factors defined by the field direction. Here k_F is the Fermi wave vector and $l(\mathbf{k}_F) = 1/\Delta k(\mathbf{k}_F) = v_F^0(\mathbf{k}_F)/\mathrm{Im}\Sigma(\omega = 0, \mathbf{k}_F)$ is the mean-free path, $v_F^0(\mathbf{k}_F)$ being the bare Fermi velocity. The striking similarity between the scattering rates measured here and the resistivity indicates that this component of the Fermi surface plays an essential role in the transport. Indeed, similar behavior is found over the whole central Fermi surface, S_C, while the sections centered at K points and flat saddle regions show much higher scattering rates and/or are gapped in the CDW state [6]. In addition to a significant zero-temperature offset [13], the single-particle scattering rate measured here has an approximately 6 times larger change over the same temperature interval than the Drude one. This is because transport currents are insensitive to small-angle scattering, whereas ARPES is sensitive to all scattering events.

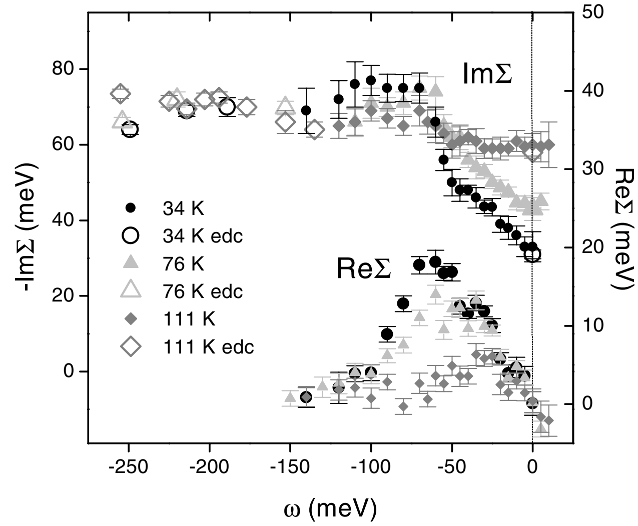

FIG. 3. Self-energies extracted from MDCs for several temperatures. Results for ImΣ obtained from EDCs are shown as open symbols.

The unusually large zero-temperature offset in the normal state resistivity measurements may be caused by conventional impurity/defect scattering, which usually adds a constant temperature-independent term to the normal state resistivity. However, for 2H-$TaSe_2$ all published resistivity

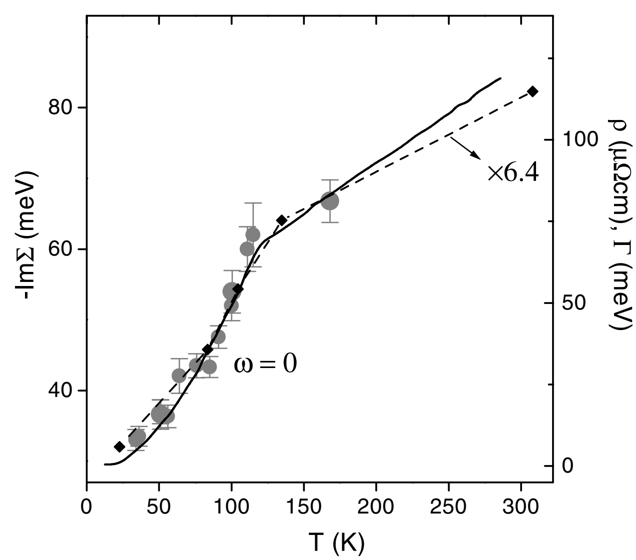

FIG. 4. Temperature dependence of the scattering rate at the Fermi level ($\omega = 0$). Resistivity $\rho(T)$ (solid line) and Drude scattering rate $\Gamma(T)$ (dashed line) from Ref. [12] are given for comparison on the right scale.

curves [12,14] appear to scale in a similar fashion over the whole temperature range, indicating some form of intrinsic disorder in the normal state. As suggested by Vescoli *et al.* [12], this may reflect the presence of fluctuating incoherent CDW segments. To resolve this, it would be instructive to study in more detail the effect of intentionally introduced defects on the resistivity.

We have discussed the experimental observations in terms of scattering from some form of collective mode. It is also possible that the opening of the gap in the "saddle regions" with an associated reduction in the phase space available for scattering would be sufficient to explain the binding energy and temperature dependence of the self-energy. However, the opening of the gap in the single-particle spectrum implies a modification to the response function with the possible simultaneous appearance of a new collective mode. The self-energy behaves then as if the hole was scattered from this mode. The two pictures are equivalent if electronic correlations dominate.

As pointed out by Vescoli *et al.* [12], the in-plane optical response in 2H-$TaSe_2$ is very similar to that measured in high T_C superconductors. In both systems one can resolve a Drude component and a mid-IR component in the gapped low temperature phases. As the Fermi surfaces in both systems are only partially gapped, it is tempting to connect the Drude component in the optical response to portions that are not gapped, while the mid-IR component seems to reflect the gapped regions. Whether the mid-IR structure itself points to the presence of the new mode is not clear. The presence of the well-known resonance in the spin response function of HTSCs shows that a mode with well-defined quantum numbers may indeed be formed. Recently, the strong connection between the commensurate resonance magnetic peak in neutron scattering [15] and the mid-IR optical structure in HTSCs has been noted [16]. Similarities in the optical response would then imply the possibility of a similar mode in the 2H-$TaSe_2$. More detailed studies of the charge and spin responses of the system may help to resolve the nature of this mode.

In summary, we have shown that the single-particle self-energy in 2H-$TaSe_2$ shows significant changes with the opening of the CDW gap implying the existence of a new type of collective excitation, associated with the CDW. Indeed, the observations suggest that the photohole is scattering from fluctuations in the CDW state. We believe that the observed behavior warrants further theoretical investigation in view of the strong similarities with the behavior observed in HTSCs.

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[10] Note that an MDC is a Lorentzian centered at $k = k_F + [\omega - \mathrm{Re}\Sigma(\omega)]/v^0$ with a width $\Gamma = 2|\mathrm{Im}\Sigma(\omega)|/v^0$ only in the case of a linear bare dispersion, $\epsilon_k = v^0(k - k_F)$.

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