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Photoemission near the Fermi energy in one dimension

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Angle-resolved photoemission spectra taken with high energy resolution on the one-dimensional compound $(TaSe_4)_2I$ confirm the suppression of the spectral intensity at the Fermi edge for one-dimensional systems and lend support to correlation effects as its most probable cause.

The photoemission spectrum of nearly-one-dimensional materials like $(TaSe_4)_2I$ is a very interesting problem in condensed-matter physics. Several experiments have suggested a very low spectral intensity near the Fermi energy E_F.[1] Dardel *et al.*[2] examined this striking result with very high energy resolution, concluding that the effect is most likely not related to a low quasiparticle density of states near E_F, but rather to a significant difference between the excitation spectrum and the density of states.

Several mechanisms were proposed to explain the suppression of spectral intensity near E_F, but no final evidence in favor of one of them could be extracted from angle-integrated photoemission data.[2] We did perform high-energy-resolution, *angle-resolved* studies of $(TaSe_4)_2I$. The results strongly favor one of the hypotheses formulated in Ref. 2: the suppression of the photoemission intensity near E_F is due to correlation effects that drive an infrared catastrophe, as suggested by Schulz[3] in the Luttinger model framework.

The samples studied in our experiments were similar to those of Ref. 2; the most significant differences in the procedure were the use in our study of monochromatized synchrotron radiation, and a high angular resolution of $\pm 0.5°$. The energy resolution in the different runs of our experiments ranged from 25 to 50 meV.

Typical experimental results are shown in Figs. 1 and 2. In Fig. 1 we see a series of high energy and angular resolution spectra, taken at a constant photon energy of 22 eV, and at different azimuthal angles. The spectra exhibit two important features: first, a dispersive peak that does not cross the Fermi energy, but reaches its maximum energy, approximately 0.5 eV below E_F, at a k vector corresponding to the border of the first Brillouin zone ($k_{\parallel}=0.245$ Å^{-1} in the direction of the crystal).

Second, we also see that the photoemission spectral intensity is very weak near E_F, as was observed in the angle-integrated spectra of Ref. 2. Note that the intensity is weak, but not zero. This is clear from Fig. 2, which shows the spectral region near E_F. For all directions in Fig. 2, we do see detectable signal up to the Fermi level, but never an indication of a band crossing a Fermi edge.

Several photon energies were used in the experiments, in the range 15–45 eV. The results of these tests indicate that the suppression of the near-edge spectral signal is not a trivial final-state effect. Similarly, experiments performed with different photon polarizations rule out polarization effects as a possible cause of the low intensity near

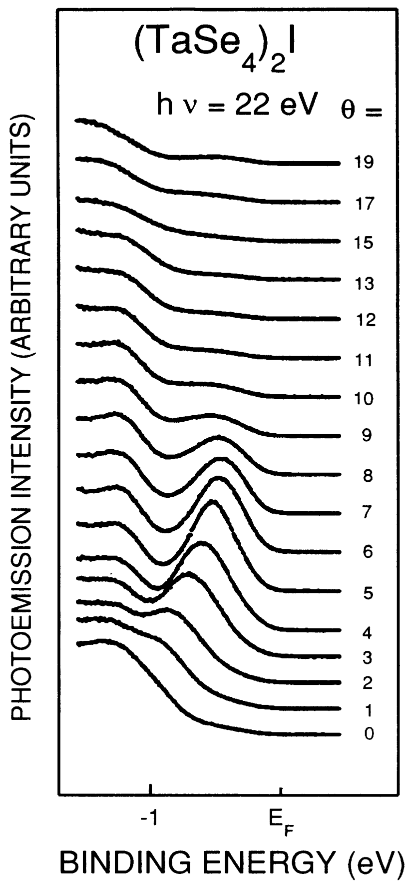

FIG. 1. A series of high-energy-resolution angle-resolved photoemission spectra, taken along the direction of the sample and for different values of the azimuthal angle.

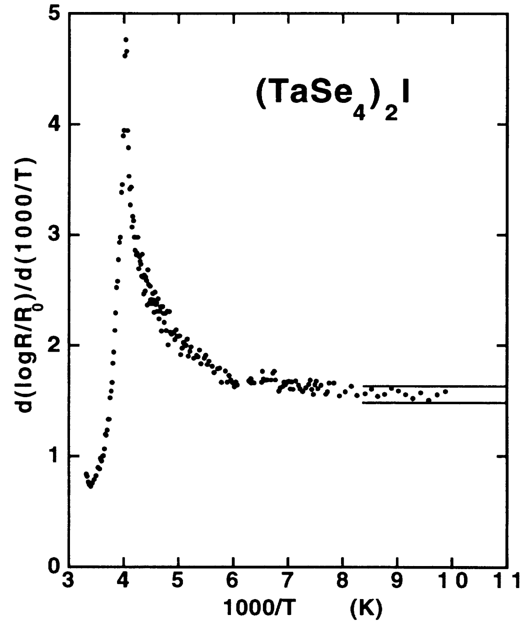

FIG. 3. Transport measurements of the half-width of the pseudogap (see Ref. 4). The asymptotic extrapolated value of the plot of $d[\ln(R/R_0)]/d(1000/T)$ (where R is the resistance, R_0 its high-temperature value, and T is the temperature) gives the half-width of the pseudogap in units of 10^3 K. The horizontal lines give a half-width of 120–135 meV.

E_F.

What is, then, the cause of this low intensity? The angle-integrated photoemission data of Ref. 2 could not, strictly speaking, rule out a very low density of states near the Fermi level. This hypothesis, however, did not seem reasonable because a very low density of states would be difficult to reconcile with the observed formation of a Peierls distortion.[2] Our present results support

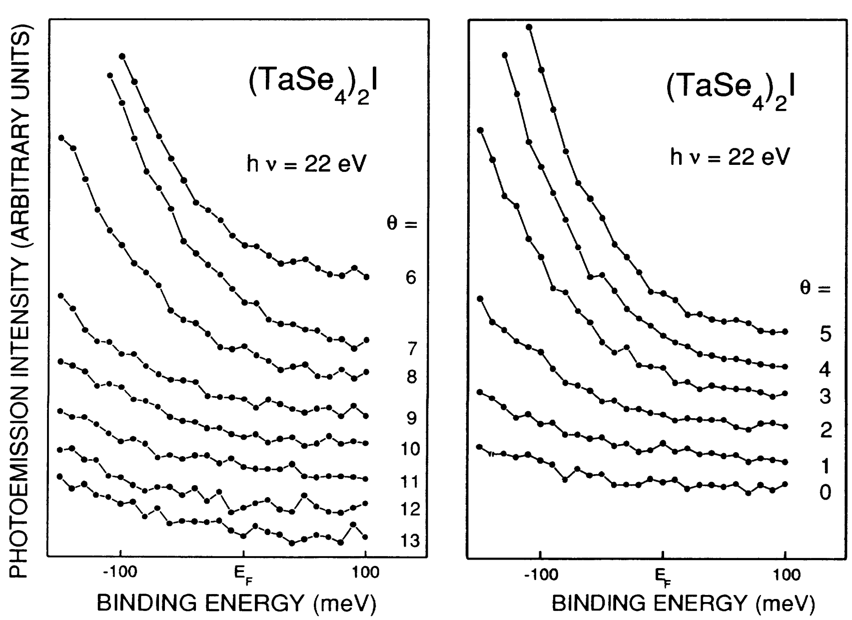

FIG. 2. Magnified plots of the spectral region immediately below E_F, showing the vanishing photoemission signal for all directions and no band crossing the Fermi edge.

this point of view: if we assume that the spectra are a faithful representation of the quasiparticle states, then the topmost band would not get closer than 0.5 eV to E_F, and this does not seem plausible even in the presence of the fluctuation effects discussed below.

We agree, therefore with Ref. 2 in believing that there is an attenuation of the photoemission signal near E_F, which causes a discrepancy between the angle-integrated spectral function and the density of states. Of the three possible mechanisms discussed there, the first one (fluctuation effects) was not supported by the temperature dependence of the spectra, in agreement with our measurements performed at different temperatures.

Note that the presence of a pseudogap was derived from optical and transport experiments for this material;[4] therefore, it is important to accurately measure its half-width and test the possibility that the dispersive band of Fig. 1 is simply at its lower edge of the pseudogap. We measured the gap half-width following the transport procedure of Wang *et al.* (see Ref. 4), and the results are shown in Fig. 3; the asymptotic value of this plot gives the half-width of the pseudogap. We obtain a half-width of 120–135 meV, in full agreement with all types of measurements in Ref. 4. This value, even considering the worst possible experimental uncertainty (horizontal lines in Fig. 3) cannot be reconciled with the minimum distance of the dispersive band from the Fermi level.

Second, there is the possibility of excitation of phonons and shift of the spectral weight towards higher binding energies.[5] This explanation seems unlikely, however, based on our angle-resolved data. We do not see evidence of phonon satellites, and a phonon shift of the dispersive band of Fig. 1 would require very strong electron-lattice coupling plus large energy shifts of the order of 250–500 meV. All this seems quite unreasonable, whereas the data suggest a simple attenuation of the signal in the region immediately below E_F.

The attenuation could be explained by correlation effects leading to an infrared catastrophe.[3,6] In three dimensions, the spectral function would be given by the density of states multiplied by the renormalization factor, which leads to attenuation. For one-dimensional systems, the attenuation is expected[2,7] to be more drastic; instability with respect to the emission of electron-hole pairs leads to a breakdown of the quasiparticle picture, and to a vanishing spectral intensity at E_F. This picture agrees quite well with what we observe.

In summary, the added capability of angular resolution to high energy resolution produced data that lend support to the hypothesis of spectral intensity suppression by correlation effects. Although other, more complicated and therefore less likely, mechanisms cannot be ruled out, this conclusion would also agree with the apparent universality of the low photoemission intensity from one-dimensional systems. The results open interesting questions on other low-dimensional crystals like the high-temperature superconductors, specifically on the interpretation of photoemission data for these highly correlated systems.[8]

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