

Pseudogap-Driven Sign Reversal of the Hall Effect

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We present a calculation of the Hall coefficient in $2H$ -TaSe₂ and $2H$ -Cu_{0.2}NbS₂ based on their electronic structure extracted from angle-resolved photoemission spectra. The well-known semiclassical approach, based on the solution of the Boltzmann equation, yields the correct value for the normal-state Hall coefficient. Entering the charge density wave state results in the opening of the pseudogap and redistribution of the spectral weight. Accounting for this allows us to reproduce the temperature dependence of the Hall coefficient, including the prominent sign change, with *no* adjustable parameters.

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While in simple metals the Hall coefficient is essentially temperature independent, in unconventional materials it can exhibit strong variations with temperature. In particular, in the transition metal dichalcogenides, known as charge density wave (CDW) bearing compounds, the Hall coefficient changes sign from positive to negative soon after the transition into a CDW state [1,2]. A similar sign change of the Hall coefficient has recently been discovered in high temperature superconductors (HTSC) [3]. Another common departure of the CDW and HTSC compounds from conventional solid state theory is the presence of a pseudogap in their excitation spectra [4–7]. The pseudogap results in a depletion of electronic states at the Fermi level. Since the charge dynamics in the crystal is restricted to a narrow energy range around the Fermi level, the opening of the pseudogap reduces the effective number of charge carriers that apparently affects transport properties of the solid. Since the pseudogap can be highly anisotropic [6,7], the quantitative investigation of this effect requires a momentum-resolved experimental technique. In this Letter we suggest a procedure to calculate the Hall coefficient from angle-resolved photoemission spectroscopy (ARPES) data [8] and show that the origin of its sign change in $2H$ -TaSe₂ is intimately related to the pseudogap phenomenon.

In our calculations we assume the electric field \mathbf{E} to be parallel to ab plane, and the magnetic field \mathbf{B} to be parallel to the c axis (thus, current \mathbf{j} flows in the ab plane). This experimental geometry is common for investigations of two-dimensional compounds, in particular, the dichalcogenides [1,2,9,10]. In the low-field limit, the current density is related to the electrical field by means of the conductivity tensor:

$$\mathbf{j} = \sigma \mathbf{E}, \quad \sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ -\sigma_{xy} & \sigma_{xx} \end{pmatrix}. \quad (1)$$

The components of the conductivity tensor are derived from the solution of the semiclassical Boltzmann equation. Neglecting k_z dispersion and taking into account the identity of the a and b axes, σ_{xx} and σ_{xy} are expressed through the integrals over the Fermi surface in the first Brillouin zone (formulas are given in SI units):

$$\sigma_{xx} = \frac{e^2}{2\pi L_c h} \int \tau(\mathbf{k}) v_F(\mathbf{k}) dk \quad (2)$$

$$\sigma_{xy} = \frac{e^3 B}{L_c h^2} \int \frac{\tau^2(\mathbf{k}) v_F^2(\mathbf{k})}{\rho(\mathbf{k})} dk, \quad (3)$$

where τ is the quasiparticle lifetime, v_F is the *renormalized* Fermi velocity, ρ is the Fermi surface curvature radius, dk is the element of the Fermi surface length, L_c is the size of the elementary cell along the c axis, h is the Planck's constant, and e is the elementary charge. Mathematically equivalent formulae, but less convenient for our discussion, can be found in the literature [11–13]. By definition, the Hall coefficient is equal to the Hall electrical field over the magnetic field and the current density: $R_H \equiv E_H / (B \cdot j)$. In terms of the conductivity tensor it is expressed in the following way:

$$R_H = \frac{\sigma_{xy}}{B \sigma_{xx}^2}. \quad (4)$$

ARPES gives us a complete knowledge about the band structure. Therefore the only thing missing to calculate the conductivity tensor from Eqs. (2) and (3) is the transport lifetime τ , which should not be mixed with the quantum lifetime τ_q , seen in ARPES [14,15]. If we assume that τ is momentum independent, then it cancels out and the expression for R_H reduces to

$$R_H = \frac{4\pi^2 L_c}{e} \frac{\int v_F^2(\mathbf{k})/\rho(\mathbf{k})dk}{(\int v_F(\mathbf{k})dk)^2}, \quad (5)$$

where R_H is expressed in m^3/C [16]. For $2H\text{-TaSe}_2$, $L_c = 12.7 \text{ \AA}$ [17], and all other quantities that enter Eq. (5) can be extracted straightforwardly from ARPES spectra as v_F is seen in the energy-momentum cuts [15,18], ρ and the integration path are seen in the Fermi surface maps [19].

It is well known that for conventional metals Eq. (5) yields a result consistent with direct measurements of R_H [20]. We find that it also provides good agreement with the experiment for NbS_2 . In case of TaSe_2 Eq. (5) provides the correct R_H at high temperatures, but becomes inapplicable at lower temperatures, at the Fermi surface reconstruction onset. Formula (2) and (3), and, hence, (5) imply that all energy bands are *equally and uniformly populated* with

electrons. Although this assumption often holds, a *complex picture of the spectral weight distribution* does not appear to be a rare occasion for unconventional materials [4,7,22]. For such a case, Eq. (5) can be modified by introducing a factor $D(\mathbf{k})$ that takes into account the distribution of the spectral weight, i.e., the behavior of the density of states (DOS) near the Fermi level:

$$R_H = \frac{4\pi^2 L_c}{e} \frac{\int D(\mathbf{k})v_F^2(\mathbf{k})/\rho(\mathbf{k})dk}{(\int D(\mathbf{k})v_F(\mathbf{k})dk)^2}, \quad (6)$$

where

$$D(\mathbf{k}) = \int_{-\infty}^{+\infty} \text{DOS}_{\mathbf{k}}(\omega) \left(-\frac{\partial f(\omega)}{\partial \omega} \right) d\omega, \quad (7)$$

which is the temperature weighted DOS at the Fermi level, $f(\omega)$ is the Fermi function, and ω is the binding energy.

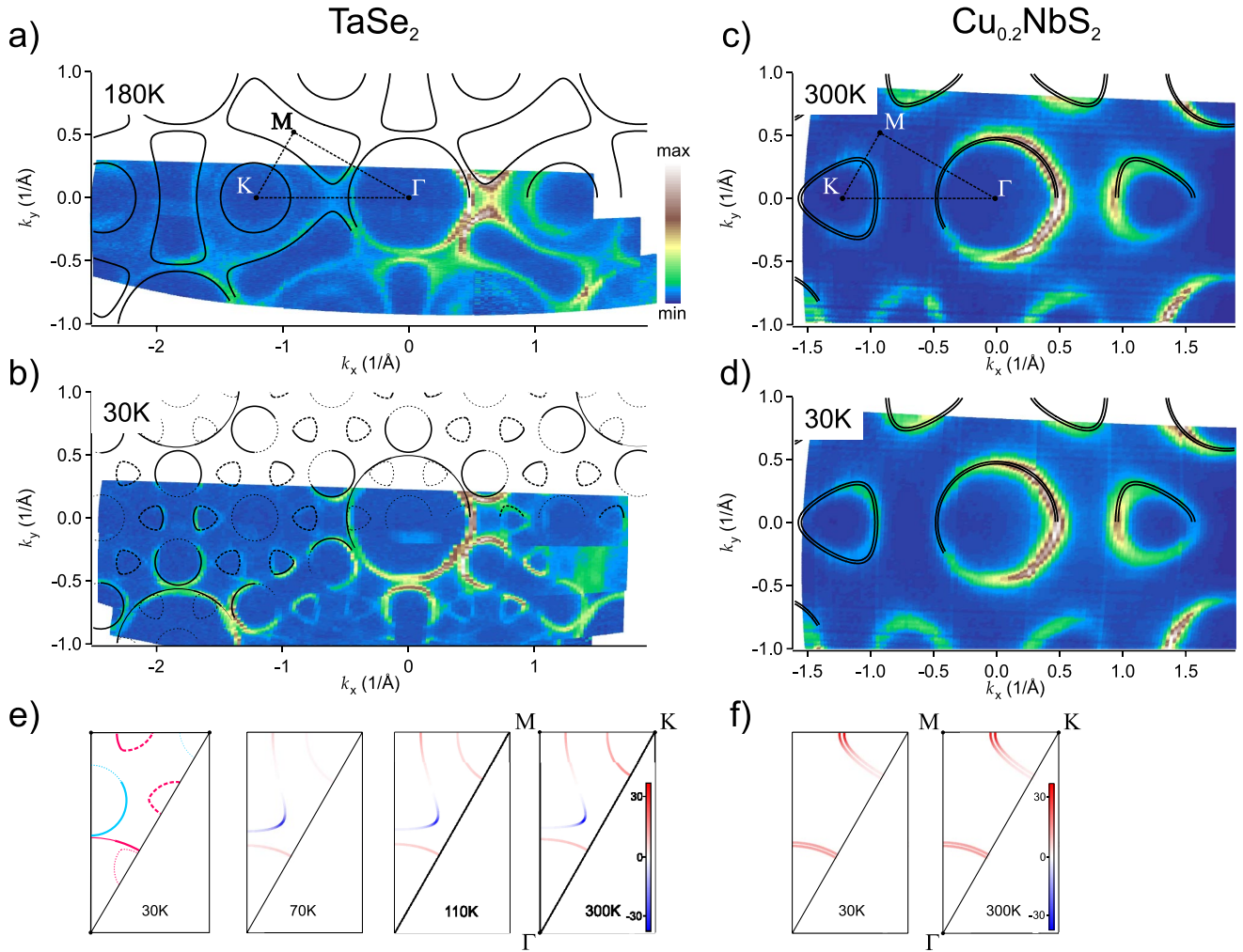


FIG. 1 (color online). Evolution of the Fermi surface of TaSe_2 and $\text{Cu}_{0.2}\text{NbS}_2$ with temperature. The Fermi surface of TaSe_2 changes topology with cooling (a, b). Absence of changes and uniform spectral weight distribution in the spectra of $\text{Cu}_{0.2}\text{NbS}_2$ (c),(d). Solid black lines in (a),(c),(d) are the tight-binding fit to the data. For the CDW-reconstructed Fermi surface (b) different types of guidelines represent the spectral weight distribution: the stronger line corresponds to the higher spectral weight. The relative contribution to σ_{xy} from different parts of the Fermi surface at different temperatures is shown in the irreducible part of the Brillouin zone for TaSe_2 (e) and $\text{Cu}_{0.2}\text{NbS}_2$ (f).

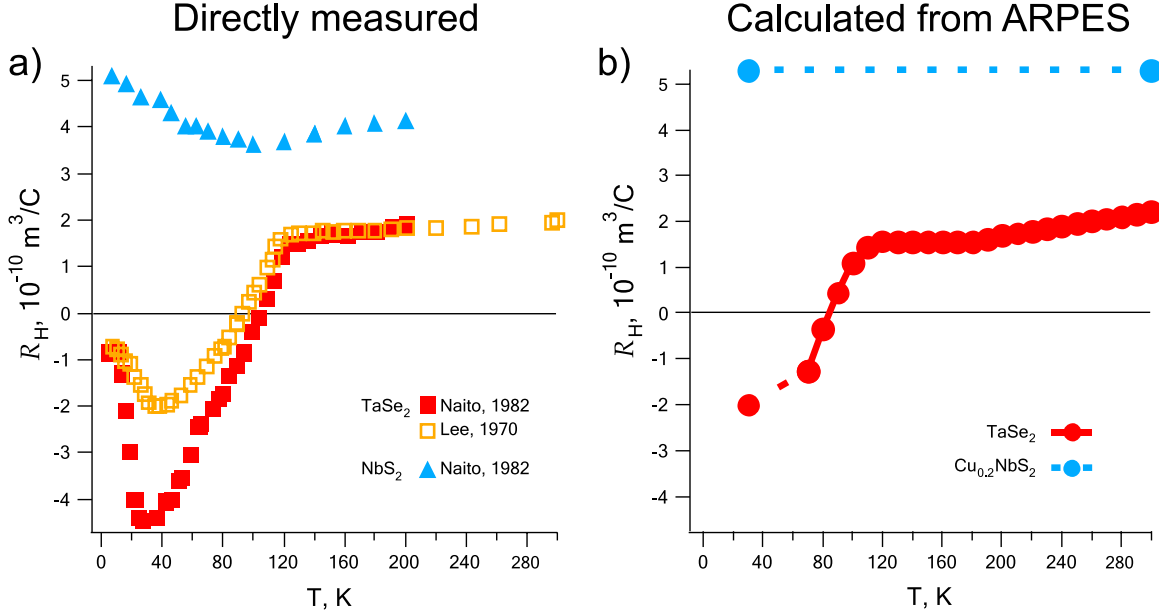


FIG. 2 (color online). The temperature dependence of the Hall coefficient. The Hall coefficient in NbS_2 has a weak temperature dependence, while in TaSe_2 it changes sign (a). The Hall coefficient of $\text{Cu}_{0.2}\text{NbS}_2$, calculated in approximation of the equally “populated” bands, agrees well with the directly measured one [23], while in the case of TaSe_2 one should take into account the spectral weight redistribution and the opening of the pseudogap (b). The discrepancy between two experimental curves for TaSe_2 is due to the charge density wave suppression by impurities.

Note that in the simplest case $\text{DOS}_{\mathbf{k}}(\omega) = 1$, and $D(\mathbf{k}) = f(-\infty) - f(+\infty) = 1$, so we arrive back at formula (5). In case of the pseudogap-modified spectra, based on the experimental data, we modeled the DOS using the following function:

$$\text{DOS}_{\mathbf{k}}(\omega) = \begin{cases} 1, & |\omega| \geq 2\Delta; \\ |\omega|/2\Delta, & |\omega| < 2\Delta. \end{cases} \quad (8)$$

To obtain the experimental input on the electronic structure of TaSe_2 and $\text{Cu}_{0.2}\text{NbS}_2$ we carried out a series of ARPES measurements for temperatures ranging from 300 down to 30 K [see Figs. 1(a) and 1(b) and, for further details, Ref. [7]]. TaSe_2 undergoes a transition to the incommensurate CDW state at 122 K and to the commensurate one at 90 K. In the spectra of TaSe_2 the pseudogap is already present at room temperature, and begins to increase sharply upon the transition into the incommensurate CDW state, evolving to the band gap in the commensurate CDW state [7]. The magnitude of the pseudogap depends on the position in the Brillouin zone. In the case of TaSe_2 the K barrel is affected by the pseudogap most of all, so its contribution to the Hall coefficient has the strongest variation with temperature, and is the primary reason for the Hall coefficient to change sign. Fermi surface reconstruction also implies opening of the pseudogap on the parts of the bone-shaped sheet around the M point and a fading of the Γ barrel ($\text{DOS}_{\mathbf{k}}(\omega) = \text{const} < 1$) near the point where it approaches the M bone; see Figs. 1(a) and 1(b) and Ref. [7]. In Fig. 1(e) the contribution to σ_{xy} from different

parts of the Fermi surface is shown for several temperatures. The above described procedure was also used in conjunction with $\text{Cu}_{0.2}\text{NbS}_2$ that exhibits no CDW. As follows from the spectra, the electronic structure exhibits no considerable temperature dependence [Figs. 1(c), 1(d), and 1(f)], and is characterized by a uniform distribution of the spectral weight. The calculated Hall coefficient of $\text{Cu}_{0.2}\text{NbS}_2$ [23] shows a weak temperature dependence. Comparing the result of the calculations with the experimental measurements we find good agreement for both the studied compounds [Fig. 2], which implies the implementation of the pseudogap effect into the semiclassical formula is correct.

In conclusion, we have shown that the suppression of the spectral weight at the Fermi level and its nonuniform distribution over the Fermi surface contours, which are related to the pseudogap formation and consequent Fermi surface folding upon entering the CDW state, are indispensable for attaining a quantitative understanding of the temperature dependence of the Hall coefficient in TaSe_2 . Our findings hint that accounting for the pseudogap and the Fermi surface reconstruction phenomenon may also be fruitful for understanding other physical properties of CDW systems and unconventional superconductors [3,24].

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$$R_H = -1/ne, \quad \sigma_{xx} = ne^2\tau/m$$
 where $n = (2\pi^2L_c)^{-1} \times \int f(\epsilon(\mathbf{k}))d^2\mathbf{k}$ and $m = \hbar k_F/v_F$.
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- | $R_H(10^{-10} \text{ m}^3/\text{C})$ | Calculation | Experiment |
|--------------------------------------|-------------|------------|
| Na | -2.38 | -2.50 |
| K | -4.49 | -4.20 |
| Rb | -5.4 | -5.0 |
| Cu | -0.530 | -0.517 |
| Nb | +0.752 | +0.875 |
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