The evolution of electron dispersion in the series of rare-earth tritelluride compounds obtained from their charge-density-wave properties and susceptibility calculations

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We calculated electron susceptibility of rare-earth tritelluride compounds \( \text{RTe}_3 \) as a function of temperature, wave vector and electron-dispersion parameters. Comparison of results obtained with the available experimental data on the transition temperature and on the wave vector of a charge-density wave in these compounds allowed us to predict values and the evolution of electron-dispersion parameters with the variation of atomic number of rare-earth element R.

INTRODUCTION

In the last two decades the rare-earth tritelluride compounds \( \text{RTe}_3 \) (\( \text{R} = \text{rare-earth elements} \)) were actively studied, both theoretically\cite{1,2} and experimentally by various techniques\cite{3–18}. A very rich electronic phase diagram and the interplay between different types of electron ordering\cite{7–9} as well as amazing physical effects in electron transport even at room temperature\cite{15–17} stimulate this interest. These compounds undergo a transition to a unidirectional density wave\cite{3} in the Te plane as developed in\cite{4}, where the square net of Te atoms in each conducting layer forms two orthogonal \( p_{xy} \) orbitals. The resulting in-plane electron dispersion can be written down as

\[
\varepsilon_1(k_x, k_z) = -2t_\parallel \cos \left( \frac{(k_x + k_z) a}{2} \right) - 2t_\perp \cos \left( \frac{(k_x - k_z) a}{2} \right) - E_F, \\
\varepsilon_2(k_x, k_z) = -2t_\parallel \cos \left( \frac{(k_x - k_z) a}{2} \right) - 2t_\perp \cos \left( \frac{(k_x + k_z) a}{2} \right) - E_F, \tag{1}
\]

In the optical conductivity measurements, but not yet confirmed by the X-ray studies. At lower temperature the \( \text{RTe}_3 \) compounds become magnetically ordered\cite{8}. In addition to all this, at high pressure the \( \text{RTe}_3 \) compounds become superconducting\cite{8}.

To understand the richness of this phase diagram and the physical properties in each phase it is very helpful to have information about the evolution of electronic structure of \( \text{RTe}_3 \) compounds with the change of the atomic number of rare-earth element R. Unfortunately, the ARPES data are available only for very few compounds of this family and, in spite of a notable progress in instrumentation, still have a large errorbar. The electron transport measurements are much more sensitive but they only give indirect information about the electronic structure, because of a large number of electron scattering mechanisms.\cite{13,17} In this paper we use the extensive experimental data on the evolution of the CDW\( \text{D}_1 \) wave vector \( Q_{CDW1} \) and transition temperature \( T_{CDW1} \) to study the evolution of the electronic structure of \( \text{RTe}_3 \) compounds. We calculate the electron susceptibility, responsible for CDW\( \text{D}_1 \) instability, as a function of the wave vector and temperature at various parameters, which determine the electron dispersion. The comparison of the results obtained with available experimental data allows us making predictions about the evolution of these electron-structure parameters with the atomic number of rare-earth element R.

CALCULATION

At temperatures \( T > T_{CDW1} \) the in-plane electron dispersion in \( \text{RTe}_3 \) is described by a 2D tight binding model of the Te plane as developed in\cite{4}, where the square net of Te atoms in each conducting layer forms two orthogonal chains created by the in-plane \( p_x \) and \( p_z \) orbitals. Correspondingly, \( x \) and \( z \) are the in-plane directions. In this model \( t_\parallel \) and \( t_\perp \) are the hopping amplitudes (transfer integrals) parallel and perpendicular to the direction of the considered \( p \) orbital. The resulting in-plane electron dispersion can be written down as
where the calculated parameters for DyTe₃ are \( t∥ = 1.85 \) eV, \( t⊥ = 0.35 \) eV \cite{4} and the in-plane lattice constant \( a \approx 4.305\text{Å} \). The Fermi energy \( E_F \) is determined from the electron density, namely, from the condition of 1.25 electrons for each \( p_x \) and \( p_z \) orbitals \cite{4}. This condition gives us \( E_F = -2t∥ \cos(\pi(1 - \sqrt{3/8})) \). It is slightly (by 10%) less than the originally used Fermi-energy value \( E_F = -2t∥ \sin(\pi/8) \), inaccurately determined \cite{4} from the same condition (see Appendix). The resulting expression shows the relation between these two parameters \( t∥ \) and \( E_F \), which is important because they both affect the electron susceptibility.

For calculation we use the Kubo formula for the susceptibility of quantity \( A \) with respect to quantity \( B \) (see §126 of \cite{20}): 

\[
\chi(\omega) = \frac{i}{\hbar} \int_{0}^{\infty} \left\langle \left[ \hat{A}(t), \hat{B}(0) \right] \right\rangle e^{i\omega t} dt. \tag{2}
\]

For the free electron gas in the terms of matrix elements it becomes

\[
\chi(\omega) = \sum_{ml} A_{ml} B_{lm} \frac{n_F(E_m) - n_F(E_l)}{E_l - E_m - \omega - i\delta}, \tag{3}
\]

where \( m \) and \( l \) denote the quantum numbers \( \{k,s,\alpha\} \), which are the electron momentum \( k \), spin \( s \), and the electron band index \( \alpha \). In the CDW response function the quantities \( A \) and \( B \) are the electron density, so that Eq. (2) is a density-density correlator. To study the CDW onset one needs the static susceptibility at \( \omega = 0 \) but at a finite wave vector \( \mathbf{Q} \). Electron spin only leads to a factor 4 in susceptibility, but the summation over band index \( \alpha \) must be retained if there are more than one band crossing the Fermi level. As a result we have for the real part of electron susceptibility

\[
\chi(\mathbf{Q}) = \sum_{\alpha,\alpha′} \int \frac{4\pi d\mathbf{k}}{(2\pi)^{d}} \frac{n_F(E_{k,\alpha}) - n_F(E_{k+\mathbf{Q},\alpha′})}{E_{k+\mathbf{Q},\alpha′} - E_{k,\alpha}}, \tag{4}
\]

where \( n_F(\varepsilon) = 1/(1 + \exp((\varepsilon - E_F)/T)) \) is the Fermi-Dirac distribution function, \( d \) is the dimension of space. Since the dispersion in the interlayer \( y \)-direction is weak in RTe₃ compounds, we can take \( d = 2 \). And each of the band indices \( \alpha \) and \( \alpha′ \) may take any of two values 1, 2, because in RTe₃ two electron bands cross the Fermi level. Here we assume that the matrix elements \( A_{ml} \) and \( B_{lm} \) do not depend on the band index. This means that due to the e-e interaction the electrons may scatter to any of the two bands with equal amplitudes. This assumption has virtually no effect on both temperature and \( \mathbf{Q} \)-vector dependence of the electron susceptibility, because the latter is determined mainly by the diagonal (in the band index) terms, which are enhanced in RTe₃ by a good nesting.

Using Eq. (4) we calculate the electron susceptibility \( \chi \) as a function of CDW wave vector \( \mathbf{Q} \) and temperature for various parameters \( t∥ \) and \( t⊥ \) of the bare electron dispersion \cite{11}. The CDW phase transition happens when \( \chi(\mathbf{Q},T)U = 1 \), where the interaction constant \( U \) only weakly depends on the rare-earth atom \( R \) in RTe₃ family. The position of susceptibility maximum \( \chi(\mathbf{Q}) \) gives the wave vector \( Q_{CDW} \) of CDW instability as a function of the band-structure parameters \( t∥ \) and \( t⊥ \). The value of susceptibility in its maximum as a function of temperature \( \chi_{max}(T) \) gives the evolution of CDW transition temperature \( T_{CDW} \) as a function of \( t∥ \) and \( t⊥ \).

**RESULTS AND DISCUSSION**

First we analyze the evolution of CDW₁ wave vector. The experimentally observed dependence of \( Q_{CDW} \) on the atomic number of rare-earth atom \( R \) can be taken, e.g., from Ref. \cite{14}: \( Q_{CDW} \) monotonically increases by \( \approx 10\% \) with the increase of \( R \)-atom number from \( Q_{CDW_{LaTe₃}} ≈ 0.275 \) r.l.u. in LaTe₃ to \( Q_{CDW_{TmTe₃}} ≈ 0.303 \) r.l.u. in TmTe₃. The dependence of CDW wave vector \( c \)-component, \( Q_{CDW_{1}} = (0,0,Q_{CDW_{1}}) \), on the perpendicular hopping term \( t⊥ \), calculated using Eq. (4), is shown in Fig. 1. As we can see from this graph, \( Q_{CDW_{1}}(t⊥) \) demonstrates approximately linear dependence. The value \( t⊥ = 0.35 \) eV, proposed in Ref. \cite{2} from the band structure calculations, is located in the middle of this plot. The obtained \( Q_{CDW_{1}}(t⊥) \) dependence is rather weak: while \( t⊥ \) increase dramatically, from 0.2 to 0.5eV, \( Q_{CDW_{1}} \) changes by only \( \sim 8\% \) in \( \text{Å}^{-1} \). In the reciprocal lattice units (r.l.u.) this variation is slightly stronger, as the lattice constant \( c \) decreases with the atomic number from \( c ≈ 4.407\text{Å} \) in LaTe₃ to \( c ≈ 4.28\text{Å} \) in ErTe₃ and TmTe₃, and the r.l.u. correspondingly increases in \( \text{Å}^{-1} \). However, just the \( Q_{CDW_{1}}(t⊥) \) dependence cannot explain the observed evolution of the CDW₁ wave vector with the \( R \)-atom number, because it is too weak.

The dependence \( \chi(t⊥) \) is shown in Fig. 2. The electron susceptibility varies within one percent of its’ maximum value and thus remains almost constant. The \( \chi_{CDW_{1}} \) values are calculated on the wave vectors \( Q_{CDW_{1}} \), obtained
for each value of $t_\perp$ as a position of the susceptibility maximum. From this plot we conclude that the parameter $t_\perp$ has almost no effect on the CDW$_1$ transition temperature. Hence, to interpret the evolution of CDW$_1$ transition temperature $T_{CDW1}$ and of its wave vector $Q_{CDW1}$ with the rare-earth atomic number, one needs to consider their $t_\parallel$-dependence.

The dependence $Q_{CDW1}(t_\parallel)$ is shown in Fig. 3. The interval of this plot comprises the values $t_\parallel = 1.7\,\text{eV}$ and $t_\parallel = 1.9\,\text{eV}$, obtained in Ref. [4] from the band structure calculations for the lightest and heaviest rare-earth elements. $Q_{CDW1}(t_\parallel)$ demonstrates sublinear monotonic dependence, but $Q_{CDW1}$ there increases with the increasing of parameter $t_\parallel$. It is opposite to the dependence $Q_{CDW1}(t_\perp)$. Comparing Fig. 3 with the experimental data on $Q_{CDW1}$, summarized in Ref. [14], we may conclude that the parameter $t_\parallel$ increases with the atomic number of the rare-earth element. According to the band structure calculations in Ref. [4] this transfer integral indeed increases from $t_\parallel = 1.7\,\text{eV}$ in LaTe$_3$ to $t_\parallel = 1.9\,\text{eV}$ in LuTe$_3$. Thus, our conclusion qualitatively agrees with the band-structure
calculations in Ref. [4]. However, according to our calculation the variation of $t_\parallel$ with the atomic number of the rare-earth element must be stronger in order to account for the observed $Q_{\text{CDW}1}$ dependence.

In Fig. 4 we plot the calculated $\chi(t_\parallel)$ dependence, which is approximately linear. Similarly to our calculations of $\chi(t_\perp)$, the susceptibility value is taken in its maximum as a function of the wave vector $Q_{\text{CDW}1}$. $\chi$ changes significantly—about 35% of its maximum value in the full range of parameter $t_\parallel$ change. The $Q_{\text{CDW}1}$ transition temperature $T_c$ is given by the equation $|U\chi(Q_{\text{CDW}1}, T_c)| = 1$. Since the susceptibility increases with the decrease of temperature, the largest value of $\chi$ corresponds to the highest value of CDW transition temperature. We assume that the electron-electron interaction constant $U$ remains almost the same for considered series of RTe$_3$ compounds, because they have very close electronic structure. The result obtained (see Fig. 4) is comparable to the change of transition temperature $T_{\text{CDW}1}$ observed in the RTe$_3$ series [8]. The value $t_\parallel = 1.85\text{eV}$ in DyTe$_3$ is the reference point. The experimentally observed transition temperature to $\text{CDW}_1$ state in TmTe$_3$ is $T_{\text{CDW}1} = 245\text{K}$, while for GdTe$_3$ it is $T_{\text{CDW}1} = 380\text{K}$ and for DyTe$_3$ it is $T_{\text{CDW}1} = 302\text{K}$. [8] This transition temperature is reduced by 35% of its maximum value from GdTe$_3$ to TmTe$_3$. Thus, we may assume that this range of $t_\parallel$ describes the whole series of compounds from TmTe$_3$ to GdTe$_3$. Moreover, basing on our calculations, we predict the values $t_\parallel \approx 1.37\text{eV}$ in GdTe$_3$, $t_\parallel \approx 1.96\text{eV}$ in HoTe$_3$, $t_\parallel \approx 2.06\text{eV}$ in ErTe$_3$, and $t_\parallel \approx 2.20\text{eV}$ in TmTe$_3$.

The dependence $\chi(t_\perp)$ calculated at temperature above the transition is shown in Fig. 5. It is important to note there that with the decrease of temperature the wave vector does not shift and thus does not change its value, as it shown in Fig. 3 the position of maximum of susceptibility is almost the same for two different temperatures. Thus, the electronic susceptibility in Fig. 5 is calculated on the same $Q_{\text{max}}$ wave vectors in Fig. 4 but has lower value with the increase of temperature from 240 K to 400 K.

The transition temperatures and conducting band parameters for varies RTe$_3$ compounds are summarized in Table I. The $t_\parallel$ increases with the increase of the atomic number of rare-earth element R; the $t_\perp$ decreases with the increase of the atomic number of rare-earth element R according to $Q_{\text{max}}(t_\perp)$, but since the electronic susceptibility does not depend on $t_\perp$ we cannot calculate the values.

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|c|c|}
\hline
Compound & $T_{\text{CDW}1}$ & $T_{\text{CDW}2}$ & $t_\parallel$ & $t_\perp$ & $E_F$ & \\
& K & K & eV & eV & eV & \\
\hline
GdTe$_3$ & 377 & 4.320 & 1.37 & 0.35 & 0.95 & \\
DyTe$_3$ & 306 & 4.302 & 1.85[4] & 0.35[4] & 1.28 & \\
HoTe$_3$ & 284 & 4.290 & 1.96 & 0.35 & 1.35 & \\
ErTe$_3$ & 267 & 4.285 & 2.06 & 0.35 & 1.42 & \\
TmTe$_3$ & 244 & 4.275 & 2.20 & 0.35 & 1.52 & \\
\hline
\end{tabular}
\caption{List of parameters describing the dispersion and the CDW transition temperatures $T_{\text{CDW}1}$ and $T_{\text{CDW}2}$ for R = Gd, Dy, Ho, Er, Tm.}
\end{table}
Our suggested values of the transfer integrals $t_{\parallel}$ and $t_{\perp}$ assume that the effective electron-electron interaction at CDW wave vector does not depend considerably on the rare-earth element R.

**CONCLUSIONS**

To summarize, we have calculated the electron susceptibility on CDW$_1$ wave vector in the rare-earth tritelluride compounds as a function of temperature, wave vector, and two tight-binding parameters ($t_{\parallel}$ and $t_{\perp}$) of the electron dispersion. From these calculations we have shown that the parameter $t_{\perp}$ has almost no effect on the CDW$_1$ transition temperature $T_{CDW1}$ and weakly affects the CDW$_1$ wave vector $Q_{CDW1}$. On contrary, the variation of parameter $t_{\parallel}$ with the atomic number $n$ of rare-earth element drives the variation of both $T_{CDW1}$ and $Q_{CDW1}$. Note that the
FIG. 6: The total susceptibility as a function of wave vector $Q_{\text{max}}$ near its maximum calculated at $T=240$ K (solid blue line) and at $T=400$ K (dashed red line).

increase of $t_\parallel$ and of $t_\perp$ has opposite effects on $Q_{\text{CDW1}}$. Using the experimentally measured transition temperatures $T_{\text{CDW1}}$, we estimated the values of $t_\parallel$ from our calculations for the whole series of RTe$_3$ compounds from TmTe$_3$ to GdTTe$_3$.

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