The electronic structure of $R\text{NiC}_2$ intermetallic compounds

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First-principles calculations of the electronic structure of members of the $R\text{NiC}_2$ series are presented, and their Fermi surfaces investigated for nesting propensities which might be linked to the charge-density waves exhibited by certain members of the series ($R = \text{Sm}, \text{Gd} \text{ and } \text{Nd}$). Calculations of the generalized susceptibility, $\chi_0(q, \omega)$, show strong peaks at the same $q$-vector in both the real and imaginary parts for these compounds. Moreover, this peak occurs at a wavevector which is very close to that experimentally observed in SmNiC$_2$. In contrast, for LaNiC$_2$ (which is a superconductor below 2.7K) as well as for ferromagnetic SmNiC$_2$, there is no such sharp peak. This could explain the absence of a charge-density wave transition in the former, and the destruction of the charge-density wave that has been observed to accompany the onset of ferromagnetic order in the latter.

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I. INTRODUCTION

The idea that the Fermi surface (FS), through instabilities in the electronic structure (such as nesting features or van Hove saddle points), can drive low-dimensional systems into new ground states such as spin-density or charge-density waves (CDW), is well-known (for example, the transition metal dichalcogenides [1] and Cr [2]). More recent theoretical studies of the role of the FS in CDW formation [3, 4] have contributed greatly to our understanding of the phenomenon, and have clarified a number of misconceptions. Specifically, those authors emphasize that simply inspecting the FS for possible nesting features without actually calculating the real part of the electronic susceptibility does not help in predicting CDW instabilities [4]. They further urge caution about attributing such nesting as the only (or even main) driving force for such instabilities. In this paper we calculate the electronic structure, FS and susceptibilities of several members of the $R\text{NiC}_2$ family which have been shown to exhibit a fascinating array of electronic instabilities [5, 6]. SmNiC$_2$ has recently been found to host an interesting interplay between CDW and ferromagnetic order [7]. On cooling below a temperature of 148K, a resistivity anomaly and the appearance of satellite peaks in x-ray scattering indicate the formation of a CDW. The critical phonon softening, inferred from the thermal diffuse scattering above 148K (and which also disappears at the ferromagnetic transition) occurs at two specific wavevectors, namely $q_1 = (0.5, 0.52, 0)$ and $q_R = (0.5, 0.5, 0.5)$ [7]. Of these two wavevectors, it is the incommensurate $q_1$ modulation which develops into a CDW. Below 17.7K the satellite peaks suddenly disappear and there is a sharp decrease in the resistivity, and both phenomena are coincident with the appearance of ferromagnetic order [7]. Shimomura et al. tentatively suggest that the diffuse scattering associated with the phonon softening, the formation of the CDW state and their disappearance at the (first order) ferromagnetic transition could be collectively understood from a knowledge of the FS of SmNiC$_2$ [7]. In this paper we address this issue through ab initio calculations of the electronic structure of the $R\text{NiC}_2$ system.

II. ELECTRONIC STRUCTURE CALCULATIONS

The $R\text{NiC}_2$ family possess the orthorhombic CeNiC$_2$ structure (Space Group 38, Amm2) [8], which is shown in Fig. 1a. The electronic band structure of $R\text{NiC}_2$ was calculated for several $R$ (La, Nd, Sm and Gd) using the scalar-relativistic linear muffin-tin orbital (LMTO) method within the atomic sphere approximation (ASA) including combined-correction terms [9]. Here, the potential is described within the local density approximation for non-magnetic calculations [10] and the lo-

FIG. 1: (color online) The (a) crystal structure, (b) electronic band structure, and (c) density of states of SmNiC$_2$. 

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TABLE I: Lattice parameters used in the electronic structure calculations, accompanied by the references from which they were taken. The density of states at the Fermi level, $N(E_F)$, obtained from the calculations, is also given.

| R  | a (Å) | b (Å) | c (Å) | Ref. | $N(E_F)$ [states per (eV · f.u.)] |
|----|-------|-------|-------|------|-----------------------------|
| La | 3.956 | 4.563 | 6.204 | [12] | 4.34                        |
| Nd | 3.765 | 4.544 | 6.146 | [12] | 2.64                        |
| Sm | 3.703 | 4.529 | 6.098 | [13] | 3.84                        |
| Gd | 3.649 | 4.518 | 6.077 | [12] | 4.28                        |

FIG. 2: (color online) The FS of SmNiC$_2$, (a) in 3D with the first Brillouin zone marked, and (b) several contours in planes perpendicular to $c^*$. The arrow in (b) denotes the experimentally observed CDW wavevector $q = (0.5, 0.52, 0)$. All units are in terms of $2\pi/(a, b, c)$.

FIG. 3: (color online) $\chi_0(q)$ for SmNiC$_2$. The arrow denotes the peak in both real and imaginary parts of $\chi_0(q)$ at $q = (0.5, 0.56, 0)$.

where $f(\epsilon)$ is the Fermi function and the sum runs over bands $n$ and $n'$ through the Brillouin zone. The Brillouin zone integration is performed using the tetrahedron method of Rath and Freeman [14]. As recently emphasized by Johannes et al., although the imaginary part of $\chi_0(q)$ depends only on details of the electronic structure near the Fermi energy, it is the real part of $\chi_0(q)$ (which has contributions coming from a bandwidth-sized window of energies) that could indicate a CDW instability [4]. Both the real and imaginary parts of the non-interacting susceptibility were calculated for SmNiC$_2$, and are shown in Fig. 3. It is noteworthy that there is a strong peak at the same $q$ vector in both the real and imaginary parts of $\chi_0(q)$. Closer inspection reveals that the peak is at a wavevector of $(0.5, 0.56, 0)$, which is very close to the CDW vector $q_1$ identified by Shimomura et al. [7]. It should be borne in mind when comparing the experimental $q_1$, and theoretical peak in $\chi_0(q)$ that our calculations have been performed at the experimental lattice constant, rather than at the energy minimum of the LMTO calculation. Nevertheless, excellent qualitative, and good quantitative agreements are found between experiment and these band structure calculations.
calculated along the [1,1,1] direction (shown in Fig. 4) and again a peak was found at a vector of (0.5, 0.5, 0.5), which matches the \( q \) vector associated with the thermal diffuse scattering by Shimomura et al. [7].

Calculations for both NdNiC\(_2\) and GdNiC\(_2\) reveal similar electronic structure and Fermi surfaces to the Sm compound, and the nesting properties already identified for SmNiC\(_2\) are still present in a visual inspection of the FS. Furthermore, calculations of both the real and imaginary parts of \( \chi_0(q) \) show comparable structure, the strong peak previously identified for the Sm compound developing in \( Re [\chi(q)] \) for \( q = (0.5, 0.5, 0) \) and \( q = (0.5, 0.5, 0) \) for NdNiC\(_2\) and GdNiC\(_2\), respectively (see Fig. 5). Both the Nd and Gd compounds exhibit anomalies in the resistivity that have been identified with the emergence of a CDW [8]. Although the wavevectors that define these CDW states have not yet been investigated, these results suggest the CDW structure is expected to be similar in nature to that already observed for SmNiC\(_2\), with a weak dependence on the particular rare-earth compound (presumably associated with the lanthanide contraction). The coincidence of the peak structure in both the real and imaginary parts of the susceptility for several different rare-earths, demonstrated in Fig. 5, underlines the importance of the FS (through nesting) in these systems.

Conversely, LaNiC\(_2\) shows no equivalent kink in its resistivity [8] for temperatures down to 12K, raising the question of whether this compound shares the same FS topology, and more specifically, whether the same nesting feature is present. We have addressed this by calculating the electronic structure and \( \chi_0(q) \) for LaNiC\(_2\). The density of states at the Fermi level is shown in Table 4 alongside comparative values for the other rare-earth compounds addressed in this study. The FS, shown in Fig. 6, is found to be topologically quite different, and the features of the FS that give rise to the nesting in SmNiC\(_2\) appear strongly distorted, as demonstrated by the constant \( c^* \) contours shown in Fig. 6. The \( Im [\chi_0(q)] \) shows a broad series of ripples along the \( q = (0.5, \eta, 0) \) direction (see Fig. 7), rather than the strong peak observed for SmNiC\(_2\), compounding the evidence of poor nesting. In the real part of the susceptibility, also shown in Fig. 7, the peak becomes spread out over some \( \sim 0.4b^* \) of the Brillouin zone, which is unlikely to drive a CDW. Calculations for SmNiC\(_2\) in the LaNiC\(_2\) structure yield similar results, emphasizing that the transition of the FS from strong to weaker nesting is a consequence of the structure, rather than the particular rare-earth substitution.

Spin-polarized calculations of the electronic structure have also been performed for SmNiC\(_2\) in order to investigate whether a similar transition in the nesting qualities of the FS (as a consequence of the spin-splitting of the FS) can explain the disappearance of the CDW state that accompanies the onset of ferromagnetism. These calculations have been performed at the same lattice constant used for non-magnetic SmNiC\(_2\), since the structural transition that has been reported at the Curie temperature...
The electronic band structure (a) and FS of LaNiC₄, (b) in 3D with the first Brillouin zone marked, and (c) several contours as for Fig. 2.

FIG. 6: (color online) The electronic band structure (a) and FS of LaNiC₂, (b) in 3D with the first Brillouin zone marked, and (c) several contours as for Fig. 2.

III. CONCLUSION

We have calculated the FS of several (La, Nd, Sm and Gd) members of the RNiC₂ series, accompanied by calculations of both real and imaginary parts of the non-interacting susceptibility χ₀(q). For the Nd, Sm and Gd members, peaks in the imaginary part of χ₀, that can be directly identified with corresponding nesting properties of the FS, still persist in the real part of χ₀, which is the relevant quantity for a CDW instability. The wavevector [0.5, η, 0] corresponding to these peaks in χ₀(q) is close (η = 0.56) to the experimental wavevector (η = 0.52) obtained from x-ray scattering measurements of SmNiC₂ [7]. On the other hand, the FS of LaNiC₂ is found to be markedly different, and the χ₀(q) does not exhibit such strong structure, providing a possible route towards explaining the apparent absence of a CDW state for this particular compound. Spin-polarized calculations for SmNiC₂ at two different magnetic moments (0.17 and 0.32μB) demonstrate the gradual destruction of the nesting properties of the FS with increasing moment. The strong peak in χ₀(q) that is present at zero moment becomes washed out as the spin-polarized bands separate, in agreement with the suggestion of Shimomura et al. that the ferromagnetic transition in this compound adversely affects the nesting properties of the FS and leads to a destruction of the CDW state [7].

In summary, our findings suggest that for the Sm, Gd
and Nd compounds, the FS does indeed nest at the CDW wavevector recently observed for SmNiC$_2$, and more importantly, this strong divergence survives in the real part of the susceptibility. Furthermore, calculations of two isostructural compounds for which CDW formation has not been observed show substantially weaker divergences in their respective susceptibility, lending support to the idea that FS nesting plays an important role in deciding the fate of these systems.

**Note added during revision**

During the preparation of this manuscript, a recent study of the electronic structure of LaNiC$_2$ has been reported [16], in which the authors have relaxed the internal crystal structure. They go on to comment that the FS they obtain is in excellent agreement with our results. In response to their investigation, we have repeated the calculation of LaNiC$_2$ for these parameters, and find no significant change in the FS.

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