Generalized susceptibility and magnetic ordering in rare-earth nickel boride carbides

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(Received 23 January 1995)

The generalized susceptibility for rare-earth nickel boride carbides has been calculated using the normal-state electronic structure. Peaks in the susceptibility occur near wave vectors corresponding to those observed for the incommensurate spin density waves in $HoNi_2B_2C$ between approximately 4.7 and 6.0 K [A. I. Goldman *et al.*, Phys. Rev. B **50**, 9668 (1994)].

The recently discovered[2–5] family of rare-earth nickel boride carbides RNi_2B_2C, where R stands for a rare-earth element, has attracted considerable interest both theoretically and experimentally.[1,3,6–9] Theoretical studies[8,9] have so far been focused on understanding the mechanism for the relatively high superconducting transition temperature observed in nonmagnetic $LuNi_2B_2C$ (16.6 K), and unlike the high-T_c materials, electron-phonon coupling has been strongly suggested as the mechanism responsible for the superconducting phase transition. Superconductivity has been observed not only for the compounds containing nonmagnetic rare-earth elements, but also for those containing magnetic rare-earth elements (Tm, Er, Ho).[3,6] The interplay between magnetism and superconductivity of these rare-earth nickel boride carbides, which is reminiscent of the magnetic superconductors RRh_4B_4 and RMo_6S_8,[10–13] has already been the focus of many experimental studies. Among these materials, $HoNi_2B_2C$ (T_c=8 K) is of particular interest. Earlier resistivity and upper critical field measurements on powder samples by Eisaki *et al.*[6] demonstrated that $HoNi_2B_2C$ exhibits reentrant behavior with zero field in a small temperature range around 5 K. Further investigations on single-crystal samples clarified that the resistivity anomaly is only present with a small applied field (20–200 G).[1,7] Susceptibility measurements by Eisaki *et al.*[6] and those by Canfield *et al.*[7] show that below 5 K, the compound is in an antiferromagnetic state that coexists with superconductivity. The recent neutron scattering experiment carried out by Goldman *et al.*[1] on single crystals further confirmed that below approximately 4.7 K, this compound is in a simple antiferromagnetic state coexisting with superconductivity. This experiment also detected two incommensurate magnetic structures at $\mathbf{q} = (0, 0, 0.915c^*)$ and at $\mathbf{q} = (0.585a^*, 0, 0)$ appearing for temperatures between approximately 4.7 and 6.0 K. This finding strongly suggests that pair breaking due to these incommensurate spin-density-wave states is responsible for the near reentrant behavior reported earlier in this compound at around 5 K.

Although considerable progress has been achieved towards a theoretical understanding of antiferromagnetic superconductors,[14] no complete microscopic theory including incommensurate magnetic structures is presently available to assess in detail how the magnetic structure and superconductivity influence each other. If we regard the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) type interactions between the rare-earth $4f$ moments as the driving force for the magnetic orderings that occur, then the origin of the magnetic structures must be attributed to susceptibility maxima arising from the conduction band electronic structure. Such maxima may be caused by Fermi surface nesting, or possibly by effects associated with the superconducting state. The latter mechanism has been discussed by Machida and Matsubara.[15] Since the experiments revealed three magnetic structures below T_c for the $HoNi_2B_2C$ crystal,[1] it is of particular interest to identify the role, if any, of the normal-state electronic structure in producing these structures.

Here we present our results for the calculated generalized susceptibility function which exhibits maxima suggesting that the two incommensurate structures are caused by features of the normal-state electronic structure. On the other hand, no susceptibility maximum was found at the commensurate $\mathbf{q}$=(0,0,1) wave vector corresponding to the lowest-temperature antiferromagnetic ordering. This suggests that the superconducting state may be partially responsible for the commensurate antiferromagnetic phase below 4.7 K.

Except for small effects in lattice constants caused by the lanthanide contraction, the electronic structures for RNi_2B_2C should be similar for all the trivalent R elements. In our calculation of the generalized susceptibility function

$$\chi(\mathbf{q}) = \sum_{n,m,\mathbf{k}} \frac{f[\epsilon_m(\mathbf{k})]\{1 - f[\epsilon_n(\mathbf{k}+\mathbf{q})]\}}{\epsilon_n(\mathbf{k}+\mathbf{q}) - \epsilon_m(\mathbf{k})}, \qquad (1)$$

where $f(\epsilon)$ is the Fermi-Dirac distribution function, we have used the energy band structure calculated for $LuNi_2B_2C$. We expect that if the $4f$ states are treated as core states in the other rare-earth compounds of the family the resulting energy band structures should be similar to that of $LuNi_2B_2C$.

In generating the self-consistent one-electron potential for $LuNi_2B_2C$ within the local density-functional ap-

proximation (LDA), 349 irreducible **k** points were used in the scalar relativistic, tight-binding, atomic-sphere-approximation, linear muffin-tin orbital (ASA-LMTO) program. Our energy band results agree well with those already in the literature.[8,9] To calculate the generalized susceptibility function, the whole reciprocal unit cell was divided into 40 × 40 × 40 parallelepipeds, and energies were calculated at 4531 irreducible **k** points. Each parallelepiped was further cut into six tetrahedra and the linear tetrahedron method was employed in the **k** space integration. Since bands that are away from the Fermi level only contribute to a structureless background, we only include five bands in our calculation, the middle three of which are those crossing the Fermi level.

The results of $\chi(\mathbf{q})$ for **q** along the a axis and c axis are shown in Fig. 1 and Fig. 2, respectively. Within the resolution of our calculation, a sharp peak at around **q**= (0.6, 0, 0) is evident (see Fig. 1) and a broad peak at **q**= (0, 0, 0.9) is also found (see Fig. 2). These are within the theoretical resolution of the wave vectors corresponding to the incommensurate ordered states. We note that along the c^* axis there is another peak at **q** = (0, 0, 0.3) which is higher than the **q** = (0, 0, 0.9) peak. Without including matrix elements or a proper treatment of the effects caused by the superconducting state in our calculations, we are unable, for now, to say much about the significance of this peak. A full treatment of the appropriate matrix elements, accurate to a few degrees Kelvin, is difficult and, to our knowledge, has never been accomplished. Besides the possible suppression of the peak by small matrix elements, it is also possible that the susceptibility in the presence of a superconducting gap will be reduced in the small **q** region.

We found that the sharp peak along the a axis at **q**=(0.6, 0, 0) is due to strong Fermi surface nesting. To illustrate this, we show in Fig. 3 four slices of Fermi surface intersected by planes perpendicular to the c axis. The c-direction coordinates of these planes in units of c^* are 0.0, 0.5, 0.8, 1.0. The nesting at **q**=(0.6, 0, 0) is highlighted by an arrow shown in Fig. 3 and it is quite apparent that the nesting is strong. However, we did not find a similar strong nesting for **q**=(0, 0, 0.9) which is consistent with the broad feature shown in Fig. 2.

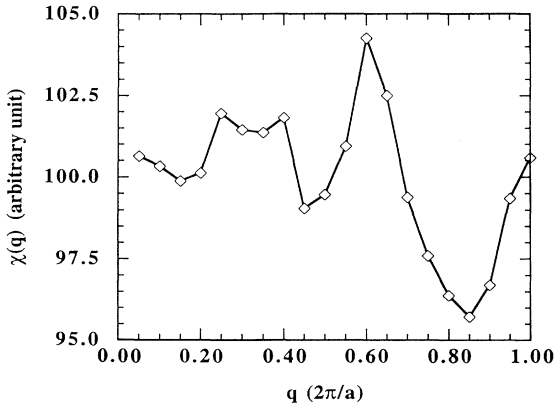

FIG. 1. Generalized susceptibility along the a^* axis.

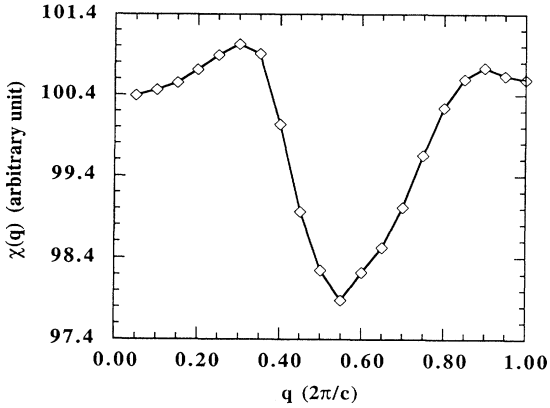

FIG. 2. Generalized susceptibility along the c^* axis.

Having shown our results for the susceptibility function, we want to make some remarks about the relevance of this calculation to the magnetic structures observed by Goldman *et al.*[1] First, we believe that it is the indirect (RKKY) coupling among the magnetic rare-earth ions mediated by the conduction electrons that drives the system toward an ordered structure. The exchange integral between the $4f$ and the conduction electrons is the relevant parameter for the matrix elements which should go into a complete evaluation of the spin ordering and excitation spectrum.[16] This means that there should be sufficient amplitude of the conduction electron wave functions at the rare-earth site to sustain any peak feature in our generalized susceptibility function (which was calculated assuming constant matrix elements). Although we have not carried out a full calculation with the proper matrix elements included, we did check that there is indeed conduction electron density at the rare-earth site for states near the Fermi level. Our observation is also consistent with the existing literature on the electronic structure of $LuNi_2B_2C$.[9]

Second, since these compounds with different rare earths have slightly different lattice constants[4] even though they all have the same crystal structure, we do

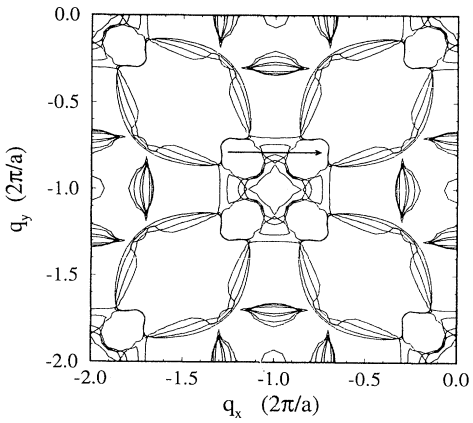

FIG. 3. Fermi surface cross section on the planes perpendicular to the c axis.

not expect that our calculation will apply *precisely* for the whole family of compounds.[17] Neither do we expect our calculation to give peaks at *exactly* those wave vectors as found experimentally. Our results do provide strong evidence, however, that the two incommensurate magnetic structures observed in $HoNi_2B_2C$ are driven by the normal-state electronic structure. We did not find a separate peak at $\mathbf{q}=(0,0,1)$ corresponding to the antiferromagnetic ordering along the c axis as observed experimentally below 4.7 K. To explain this low-temperature ordering it may be necessary to consider changes of the susceptibility function caused by the electronic structure entering the superconducting phase.

As for the interplay of superconductivity and magnetism, one may argue that the effective coupling arising from the nesting of the Fermi surface is reduced as the superconducting gap develops. If peaks in $\chi(\mathbf{q})$ broaden and flatten out, the spin-wave spectrum (for the corresponding incommensurate state) will become soft for $\mathbf{q}$ near the ordering wave vector. This in turn will lead to an increase in spin-flip scatterings and pair breaking. In this short paper we do not address the details of these interactions, as we believe they require a careful, self-consistent treatment. A model with such a treatment should be able to account in detail for the rich phenomena observed, namely the appearance and disappearance of the incommensurate magnetic structures between approximately 4.7 and 6 K,[1] the near-reentrant behavior[6,7] related to the pair breaking in the incommensurate structures, and the antiferromagnetic ordering below 4.7 K.[1,6,7]

To conclude, we have presented evidence for the normal-state electronic structure being responsible for the incommensurate magnetic structures observed in $HoNi_2B_2C$. For $\mathbf{q}$ around $(0.6,0,0)$, a strong Fermi surface nesting was found. A complete self-consistent theory to incorporate all the experimental findings is anticipated.

We wish to thank P. Canfield, C. Stassis, and A. I. Goldman for keeping us informed of their experimental results which motivated our calculations. Ames Laboratory is operated by the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This work was supported by the Director for Energy Research, Office of Basic Energy Sciences.

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