Optical properties of RNi2B2C (R=Y,Tb,Er,Dy)

S. J. Lee  
*Iowa State University*

B. K. Cho  
*K-JIST*

Paul C. Canfield  
*Iowa State University, canfield@ameslab.gov*

David W. Lynch  
*Iowa State University, dlynch@iastate.edu*

Follow this and additional works at: [http://lib.dr.iastate.edu/physastro_pubs](http://lib.dr.iastate.edu/physastro_pubs)  
Part of the [Atomic, Molecular and Optical Physics Commons](http://lib.dr.iastate.edu/physastro_pubs/atomic) and the [Condensed Matter Physics Commons](http://lib.dr.iastate.edu/physastro_pubs/condensed)

The complete bibliographic information for this item can be found at [http://lib.dr.iastate.edu/physastro_pubs/113](http://lib.dr.iastate.edu/physastro_pubs/113). For information on how to cite this item, please visit [http://lib.dr.iastate.edu/howtocite.html](http://lib.dr.iastate.edu/howtocite.html).

This Article is brought to you for free and open access by the Physics and Astronomy at Iowa State University Digital Repository. It has been accepted for inclusion in Physics and Astronomy Publications by an authorized administrator of Iowa State University Digital Repository. For more information, please contact [digirep@iastate.edu](mailto:digirep@iastate.edu).
Optical properties of RNi2B2C (R=Y,Tb,Er,Dy)

Abstract
The optical properties of single crystals of RNi2B2C (R=Y,Tb,Er, and Dy) were measured between 1.7 and 5.2 eV at room temperature using a spectroscopic ellipsometer. The spectra for all compounds are similar and the peak positions appear in a similar energy region. The similarity of the spectra of RNi2B2C (R=Tb,Er,Dy) to that of YNi2B2C indicates that the rare-earth 4f states are not actively involved in the optical transitions.

Keywords
Ames Laboratory, YNi2B2C, spectroscopic ellipsometer

Disciplines
Atomic, Molecular and Optical Physics | Condensed Matter Physics | Physics

Comments
Optical properties of $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Y, Tb, Er, Dy}$)

S. J. Lee  
*Ames Laboratory, Iowa State University, Ames, Iowa 50011*

B. K. Cho  
*Department of Materials Science and Technology, K-JIST, Kwang-Joo, 500-712, Korea*

P. C. Canfield and D. W. Lynch  
*Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011*

(Received 2 October 2000; revised manuscript received 9 March 2001; published 18 May 2001)

The optical properties of single crystals of $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Y, Tb, Er, and Dy}$) were measured between 1.7 and 5.2 eV at room temperature using a spectroscopic ellipsometer. The spectra for all compounds are similar and the peak positions appear in a similar energy region. The similarity of the spectra of $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Y, Tb, Er, Dy}$) to that of $\text{YNi}_2\text{B}_2\text{C}$ indicates that the rare-earth 4$f$ states are not actively involved in the optical transitions.

DOI: 10.1103/PhysRevB.63.233103  
PACS number(s): 68.35.Ja, 71.20.—b, 71.20.Eh, 74.25.Gz

The magnetic, transport, and electrical properties of the magnetic superconductors $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{rare earth, Y}$) have been investigated widely because they offer a system for studying the interaction of superconductivity with magnetism. The crystal structure of $\text{RNi}_2\text{B}_2\text{C}$ is the ThCr$_2$Si$_2$ type with additional carbon atoms in each $R$ layer. Adding C atoms between boron-boron bonds results in the expansion of the lattice along the $c$ axis and contraction along the $a$ and $b$ axes. As a result, the structure can be viewed as a layered system in geometrical structure, reminiscent of the high-$T_c$ oxide superconductors. A series of papers on band structure calculations on LuNi$_2$B$_2$C and YNi$_2$B$_2$C have been published, concluding that the electronic properties are three dimensional rather than two dimensional. However, there have been no band structure calculations reported on samples with $R$ other than Y and Lu, in which the 4$f$ shell is not completely closed, because the local density approximation (LDA) does not treat well the strong correlations of the localized 4$f$ electrons in the unfilled 4$f$ shell. Therefore, the optical experimental data presented in this paper provide useful information on the electronic structure for compounds with an unfilled 4$f$ shell.

The optical conductivity and dielectric function for LuNi$_2$B$_2$C and YNi$_2$B$_2$C have been studied to understand the mechanism of superconductivity in these materials. Bommeli et al. identified a superconducting gap of 5.6 and 6.9 meV for LuNi$_2$B$_2$C and YNi$_2$B$_2$C, respectively, by measuring the far-infrared optical reflectivity at 6 K. They concluded that the superconducting mechanism for Lu and Y compounds can be understood within the framework of conventional BCS theory. From the density of states at the Fermi energy, the experimental value of the electronic heat coefficient ($\gamma$) and coupling constant ($\lambda$) can be obtained. These can be used for studying the mechanism of superconductivity in the compound. The large value of the mass enhancement factor for LuNi$_2$B$_2$C and YNi$_2$B$_2$C indicates a conventional BCS mechanism for the superconductivity. The mass enhancement factor arises due to the electron-electron and electron-phonon interactions. The electron-phonon interaction increases the transition temperature while the electron-electron interaction reduces the transition temperature, that is, the electron-electron interaction acts against superconductivity. The electron-electron interaction can be obtained from experimental data on the isotope shift coefficient of a superconductor and is usually taken to be between 0.1 and 0.15. Using these values and the renormalized Bardeen-Cooper-Schrieffer formula or McMillan’s formula, the estimated electron-phonon coupling constants for LuNi$_2$B$_2$C and YNi$_2$B$_2$C are between 0.8 and 1.1, which indicates that the superconductivity in LuNi$_2$B$_2$C and YNi$_2$B$_2$C can be described by the conventional phonon mechanism.

Bommeli et al. also found that the structural anisotropy does not affect the optical data because polycrystalline and single crystalline samples produced equivalent experimental results. This is consistent with the band structure calculation, suggesting that these are electronically three-dimensional even though they have layered crystal structures. Widder et al. obtained the dielectric function of YNi$_2$B$_2$C by linking optical reflectance and electron energy loss measurements. They applied a sum rule to the free-carrier contribution to the optical conductivity and obtained an unscreened plasmon energy of $h\omega = 4.25$ eV.

YNi$_2$B$_2$C is a good reference system for studying the physical properties of the $\text{RNi}_2\text{B}_2\text{C}$ series because Y 4$f$ states are empty and located far above the Fermi level $E_F$ and do not affect the physical properties of YNi$_2$B$_2$C. We measured optical data on a single crystal of YNi$_2$B$_2$C and analyzed the optical response in the visible and UV region using the electronic structure of YNi$_2$B$_2$C. The optical spectra of $\text{RNi}_2\text{B}_2\text{C}$ ($R = \text{Tb, Er, Dy}$) were also measured and compared to those of YNi$_2$B$_2$C.

Polycrystalline stoichiometric $\text{RNi}_2\text{B}_2\text{C}$ compounds were synthesized by arc-melting together high-purity Y, Tb, Er, Dy (Ames Laboratory, 99.99%), Ni (99.95%), B (99.5%), and C (99.99%) under argon gas on a water-cooled copper hearth. An excess of
5 wt % of B over that required for stoichiometry was added to the stoichiometric mixture to compensate the loss of mass during arc melting. Single crystals were then grown from the polycrystalline bulk sample by a high-temperature flux method using Ni$_2$B as a solvent. Powder x-ray diffraction patterns of pulverized single crystals could be indexed well with the known structure type, indicating a single phase. The most intense (211) line of Ni$_2$B was seen and is due to small amounts of flux remaining on the surface of the grown crystals. The crystals separated from the flux are platelike with the crystallographic $c$ axis perpendicular to the plate surface. The crystal surface was polished to remove the remaining flux and to make the surface mirrorlike for optical measurements.

Ellipsometry is widely used to characterize surfaces, interfaces, and thin films. The principle of ellipsometry is that the state of polarization of light is changed on reflection. This change is directly related to the dielectric function of the reflecting material. With rotating analyzer ellipsometry, one measures the complex reflectivity ratio

$$\rho = \frac{r_p}{r_s} = \left| \frac{r_p}{r_s} \right| e^{i\Delta} = \tan \Psi e^{i\Delta},$$

where $r_p$ and $r_s$ are the complex amplitude reflection coefficients for $p$ and $s$ polarized light, and $\Psi$ and $\Delta$ express the change in amplitude and phase between $p$ and $s$ components of the polarized light reflected from a surface. $\Psi$ and $\Delta$ are quantities directly measurable by ellipsometry. The complex dielectric function $\tilde{\varepsilon}$ is related to the complex reflectivity ratio $\rho$ by

$$\tilde{\varepsilon} = \sin^2 \phi_s + \sin^2 \phi_p \tan^2 \phi_p \left( \frac{1 - \rho}{1 + \rho} \right)^2,$$

where $\phi_s$ is the angle of incidence, which is 68° in this experiment. From the complex dielectric function $\tilde{\varepsilon}$, the complex optical conductivity $\tilde{\sigma}$ can be obtained from

$$\tilde{\varepsilon} = 1 - \frac{4 \pi \tilde{\sigma} i}{\omega}.$$

The absorptive part of the optical conductivity $\sigma_1$ is related to the imaginary part of the dielectric function $\varepsilon_2$ by

$$\sigma_1 = \frac{\omega \varepsilon_2}{4 \pi}.$$

The spectrum of the optical conductivity for YNi$_2$B$_2$C is plotted as a solid line together with those for RNi$_2$B$_2$C ($R = \{\text{Tb}, \text{Dy}, \text{Er}\}$) in Fig. 1. There are two peaks in the optical spectra for all compounds. The first peak is between 2.3 and 2.5 eV and the second, which is weaker and broader than the
The first peak, appears between 4.5 and 5.0 eV. For the first peak of YNi$_2$B$_2$C at 2.4 eV, the occupied Ni 3d states and the unoccupied Ni 4p states are expected to be involved in the transitions from analysis of the band structure calculation.\textsuperscript{10,11} The second peak at 4.6 eV may involve contributions from the occupied Y 5p states to the empty Y 4d states located broadly between 4 and 6 eV. The second peak in YNi$_2$B$_2$C was not clearly observed in the measurement of Bommeli \textit{et al.}\textsuperscript{13–15} and of Widder \textit{et al.}\textsuperscript{19,20} who used a Kramers-Kronig transformation of the reflectivity to obtain the optical conductivity. The first peak was observed in the measurement of Widder \textit{et al.}\textsuperscript{19,20} and weakly observed by Bommeli \textit{et al.}\textsuperscript{13–15}

The origin of the first peak for RNi$_2$B$_2$C ($R$= Tb, Dy, Er) is expected to be similar to that of YNi$_2$B$_2$C because the peak positions and shapes are similar. The second peak for Tb, Dy, and Er compounds is mainly caused by the contribution of the Y 5d states of the compounds. The real part of the complex dielectric function is shown in Fig. 2. The large negative curve in the figure is due to the large free-electron gas contribution to $\varepsilon_1$. The first peak in the $\varepsilon_1$ spectra is due to interband transitions. The zero crossing of $\varepsilon_1$ for YNi$_2$B$_2$C occurs at 4.0 eV. This is lower than the unscreened plasma energy obtained by Widder \textit{et al.}\textsuperscript{19} by 0.25 eV, because it is shifted by the interband contribution. The imaginary part of the dielectric function, which can be derived from the real part of the optical conductivity by using Eq. (4), is shown in Fig. 3. Interband contributions to the dielectric function appear between 2.3 and 2.5 eV and weak peaks appear between 4.5 and 5.0 eV.

In summary, we have measured the optical conductivity spectra of RNi$_2$B$_2$C ($R$= Y, Tb, Er, Dy) at room temperature using spectroscopic ellipsometry. The spectra for all compounds are similar and the peak positions appear in a similar energy region. The similarity of the spectra of RNi$_2$B$_2$C ($R$= Tb, Er, Dy) to that of YNi$_2$B$_2$C indicates that the rare-earth 4f states are not actively involved in the optical transitions in the measured photon energy range.

Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This work was supported by the Director of Energy Research, Office of Basic Energy Science. The work at K-JIST was supported by the Brain Korea 21 project.