Interpretation of the optical properties of Nb

Bong-Soo Kim
Iowa State University

Bruce N. Harmon
Iowa State University, harmon@ameslab.gov

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

Follow this and additional works at: http://lib.dr.iastate.edu/physastro_pubs

Part of the Atomic, Molecular and Optical Physics Commons, and the Condensed Matter Physics Commons
Interpretation of the optical properties of Nb

Abstract
The interband optical conductivity of Nb was calculated from a self-consistent relativistic band structure. For the optical conductivity, the major relativistic effect is the lowering of the $s$-like bands relative to the $d$-like bands, not spin-orbit splitting. $k$-space searches identified the regions of the Brillouin zone contributing to the principal structures of the optical conductivity. The regions were found to be large volumes of the zone that did not include symmetry points or lines. Along with previous results for Mo, the good agreement between theory and experiment suggests that the effects of the omitted self-energy are small for these two bcc metals.

Keywords
Ames Laboratory, niobium, Brillouin zone

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

Comments
Interpretation of the optical properties of Nb

Bong-Soo Kim, B. N. Harmon, and David W. Lynch

Department of Physics and Ames Laboratory, Iowa State University, Ames, Iowa 50011

(Received 17 February 1989)

The interband optical conductivity of Nb was calculated from a self-consistent relativistic band structure. For the optical conductivity, the major relativistic effect is the lowering of the $s$-like bands relative to the $d$-like bands, not spin-orbit splitting. $k$-space searches identified the regions of the Brillouin zone contributing to the principal structures of the optical conductivity. The regions were found to be large volumes of the zone that did not include symmetry points or lines. Along with previous results for Mo, the good agreement between theory and experiment suggests that the effects of the omitted self-energy are small for these two bcc metals.

Previously we reported the optical conductivity calculated both with nonrelativistic and relativistic self-consistent energy bands for Mo. It was found that relativistic bands, as well as calculated dipole matrix elements, were needed for good agreement with experiment, and it was suggested that both might be needed in the interpretation of the optical properties for all $4d$ transition metals. For Mo, the omission of the self-energy seemed not to be important. In this paper, we calculated the relativistic self-consistent energy bands and the interband optical conductivity of Nb, and show that relativistic bands produce better agreement with experiment than nonrelativistic bands.

The calculated energy bands are displayed along symmetry directions in Fig. 1. The spin-orbit splitting, added to the bands as a perturbation in the final stage, was very small. It was at most 0.08 eV at $\Gamma$ and had a negligible effect on the calculated optical conductivity. The results from the present calculation are quite similar to the self-consistent, relativistic results of Ref. 3, but differences of up to $\sim 10$ mRy are present. This is because Elyashar and Koelling used a weaker exchange-correlation potential [they used the $\alpha = \frac{8}{3}$, or the so-called Kohm-Sham-Gaspar approximation, while we used that of Hedin and Lundqvist (see Ref. 4)], which results in our $d$ bands being somewhat lower than those of Ref. 3. Elyashar and Koelling also used a general potential, whereas we have used the muffin-tin approximation with 63.5% of the unit cell being inside the muffin-tin sphere. This approximation results in shifts in $E(k)$ as large as 10 mRy. The shifts caused by relativistic effects are larger and act in such a way to lower the energies of the $s$ states relative to the $d$ states (e.g., at $\Gamma$ the $\Gamma_{12}-\Gamma_1$ separation is 60 mRy larger for the relativistic calculation). Except for this difference the bands and Fermi surface are similar to the reported nonrelativistic, non-self-consistent bands, and nonrelativistic self-consistent bands.

The calculated interband optical conductivity is shown in Fig. 2 along with the experimental result and the relativistic self-consistent results of Ref. 3. Dotted line: calculated interband conductivity including matrix elements. Dashed line: calculated interband conductivity including matrix elements, using nonrelativistic bands. Dotted line: calculated interband conductivity from Jani et al. (Ref. 7). Dashed-dotted line: experimental total conductivity (Ref. 8). Drude term is added to all calculated spectra.

![FIG. 1. Calculated electronic structure of Nb. Solid line: self-consistent relativistic bands. Dashed line: nonrelativistic bands calculated with potential of Ref. 4. The two sets of bands were aligned at the Fermi energies.](image1)

![FIG. 2. Optical conductivity for Nb. Solid line: calculated interband conductivity including matrix elements. Dashed line: calculated interband conductivity including matrix elements, using nonrelativistic bands. Dotted line: calculated interband conductivity from Jani et al. (Ref. 7). Dashed-dotted line: experimental total conductivity (Ref. 8). Drude term is added to all calculated spectra.](image2)
cent result from Jani et al.\textsuperscript{7} The latter made a nonrelativistic, self-consistent calculation with the linear combination of Gaussian orbitals method. Their result, which included matrix elements, was in better agreement after broadening the spectrum than the results from previous work.\textsuperscript{9,10} We used the analytic tetrahedron linear energy method\textsuperscript{11} for the calculation of the energy denominator in the expression for the joint density of states. There were 440 tetrahedra included in the irreducible $\frac{1}{16}$ Brillouin zone. The calculated spectrum without matrix elements, but with $k$ conservation, is similar to that calculated with matrix elements (which were assumed constant within each tetrahedron). Without matrix elements, there are two spurious peaks around 5.5 and 6.7 eV, and the peak at 3 eV is bigger than in the spectrum with matrix elements. The matrix elements deemphasize the structure around 3 eV and remove the two spurious structures, as shown in Fig. 3. The Drude contribution with $\sigma_0 = 140 \times 10^{15}$ sec\(^{-1}\) and $\tau = 13.9 \times 10^{-15}$ sec (Ref. 12) is added to all calculated spectra (it has a small effect of raising the conductivity by about 4\% at 2 eV and about 1\% at 3 eV). The absolute magnitude of the calculated spectrum agreed with the experimental values very well, so the calculated dipole matrix elements are probably quite accurate. The above results are in agreement with our previous results for Mo,\textsuperscript{1} where it was shown that relativistic effects play an important role in the determination of the optical conductivity. The downward relative shift of the s-like parts of the band structure with respect to the d-like bands is significant. These shifts account for the better agreement in the relativistic calculation.

The origin of the structures in the optical conductivity were found using an energy and a wave-vector window. Figure 4 shows the distribution of the strength of the contributions in the Brillouin zone for the peak at 2.8 eV. Each tetrahedron is represented by a point if it makes a contribution to the interband conductivity in this spectral region, with the size of dots denoting the strength of the contribution. The largest contributions do not come from the symmetry lines, but from general points in the Brillouin zone. The main contributions to structure come from transitions from band 3 to band 4. Transitions from band 1 to band 3 and from band 2 to band 3 gave smaller contributions near the $F$ edge. The biggest structure in the conductivity spectrum is at 4.7 eV, and the contributions to this structure are shown in Fig. 5.
These transitions occurred throughout all of the irreducible wedge. The largest contribution is near the center of the irreducible wedge, arising from transitions from band 2 to band 4. Transitions from band 1 to band 3 and from band 1 to band 4 occurred near the $\Gamma$ corner and the $\Delta$ edge, respectively. Around the $P$ corner, transitions from band 3 to band 4 occurred. The structure at 7.8 eV in the spectrum arises mainly from transitions from band 1 to band 6 near $\Delta$, as shown in Fig. 6. Generally, transitions occurred at general points of the Brillouin zone, where the weights can vary greatly between points, but all transitions are allowed. The overall effect of the dipole matrix elements is not dramatic, although their use results in better agreement between theory and experiment and also determine the absolute magnitude for the conductivity.

We have obtained good quantitative agreement between theory and experiment for the optical interband conductivity, with both the magnitude of the spectrum as well as the peak positions being well represented, the exception being the main peak which is about 0.3 eV off from the experimental result. It is found that almost all transitions were between $d$ states strongly hybridized with $s$ and $p$ states. Transition between the $s$-like first band and $d$ states above $E_F$ are responsible for the structure near 8 eV. Relativistic self-consistent bands and dipole matrix elements were needed to obtain the best peak positions and magnitudes. Along with the previous results for Mo, we believe that relativistic calculations are required to give the best results for the optical properties for the all 4$d$, and presumably 5$d$, transition metals. For Nb we have found the contributions to the spectra come generally from large regions of the Brillouin zone, not from regions near any special points or symmetry lines. It appears that self-energy corrections make small net effects in the optical properties. This is also suggested by an explicit calculation of the self-energy corrections which found the shift of bands caused by the self-energy is negative for all states within $\pm 4$ eV of the Fermi energy for Cr, so the net effects between pairs of bands tend to cancel each other for the optical conductivity. This suggests that for improvement between theory and experiment, it is probably best to concentrate on improvements in the potential, before attempting the much more difficult self-energy correction.

This work was carried out at the Ames Laboratory, which 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 for Energy Research, Office of Basic Energy Science.