# Interband optical properties of $Ni_3Al$

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We present calculations of interband optical conductivity of the compound $Ni_3Al$ using the linear-muffin-tin-orbital method. Calculations are performed using constant matrix elements and calculated matrix elements. We find that matrix elements play a significant role in influencing the magnitude of optical conductivity $\sigma(\omega)$ and the position of peaks. Since $Ni_3Al$ is weakly ferromagnetic (magnetic moment equal to $0.31\mu_B$ per unit cell), we have also calculated $\sigma(\omega)$ using the ferromagnetic band structure but without spin-orbit coupling. A comparison of our results with experimental data is presented.

## I. INTRODUCTION

The intermetallic compound $Ni_3Al$ has been a subject of intensive study both theoretically as well as experimentally. Experimental studies for $Ni_3Al$ include those of magnetic properties,[1–4] low-temperature specific heat,[4] de Haas–van Alphen effect,[5] elastic constants,[6–9] and optical properties.[10] A number of theoretical studies[11–19] have also been undertaken for this compound. In our earlier publication,[20] we reported a calculation of the electronic structure and Fermi surface (FS) of $Ni_3Al$ in the paramagnetic and ferromagnetic states. Calculations were done self-consistently using the linear-muffin-tin-orbital (LMTO) method[21,22] within the atomic-sphere approximation (ASA) and including the combined correction terms. The exchange and correlation potential used was the von Barth–Hedin potential (BH XC potential).[23] The calculations were performed at the experimental value of 6.776 a.u. (3.586 Å) for the lattice constant and the Wigner-Seitz (WS) spheres around Ni and Al sites were taken to be of different radii following Andersen, Jepsen, and Sob.[24] The WS radii at Ni and Al sites were 2.553 and 2.855 a.u., respectively. A good agreement was obtained between the calculated and experimentally measured FS, suggesting that the LMTO method can be used with reasonable confidence to calculate the FS topology for this compound. In fact, we found that a shift in Fermi energy ($E_F$) of 12 mRy was sufficient to bring the calculated FS in agreement with the experimental areas. The value of the corresponding shift for ferromagnetic Ni (Ref. 25) was 9 mRy. Our calculation showed that the electrons at the FS have predominantly Ni-$d$ character. We found a significantly large value for the enhancement factor $[(\gamma_{\text{expt}}/\gamma_{\text{theor}})-1]$, where $\gamma$ is the coefficient of electronic specific heat. Since $Ni_3Al$ is not a superconductor we attribute the large value of enhancement factor to spin fluctuation.

The FS gives information regarding states near $E_F$. Our calculations have already shown that these are well represented within the LMTO method. The optical data, on the other hand, span all the states above as well as below $E_F$. It would be useful to know if the LMTO can give these states equally well. We have therefore decided to calculate the optical conductivity of $Ni_3Al$ in the paramagnetic state as well as in the ferromagnetic state to ascertain how well it compares with the available data. This is a natural sequel to our earlier paper.

van der Heide *et al.*[10] have measured the optical properties of $Ni_3Al$ at room temperature under high vacuum conditions using the method of spectroscopic ellipsometry. Measurements made on single-crystal and polycrystalline samples yielded identical results. Using an earlier self-consistent band calculation of Buiting, Kubler, and Mueller,[15] based on the augmented-spherical-wave (ASW) method of Williams, Kubler, and Gelatt,[26] van der Heide *et al.*[10] have calculated the joint density of states (JDOS) and the optical conductivity $\sigma(\omega)$ as JDOS/$\omega$, to compare with their own experimental data (here $\omega$ represents the energy). Based on this comparison

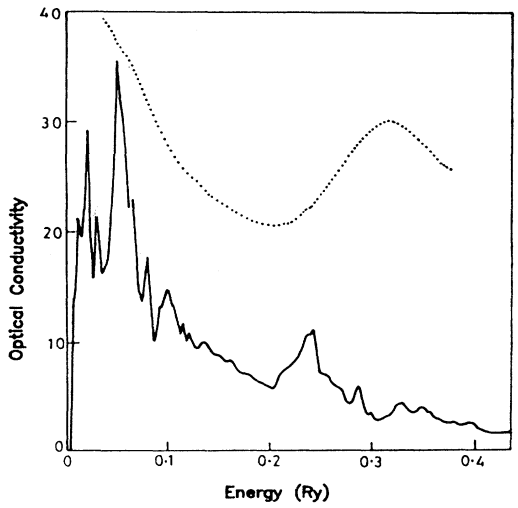


FIG. 1. The optical conductivity of $Ni_3Al$ from the experimental data (dots) and from the calculation (full curve in arbitrary units) of van der Heide *et al.* (Ref. 10).

 

they were able to identify the origin of the various experimentally observed structures in the optical conductivity. The experimental curve showed a decrease in $\sigma(\omega)$ as $\omega$ increases with a minimum at 0.21 Ry followed by a maximum at 0.32 Ry; thereafter $\sigma(\omega)$ decreases with increasing $\omega$. The theoretical JDOS/$\omega$ did show most of this structure along with many others but was unable to bring out the main peak at 0.32 Ry suggesting the need for inclusion of matrix elements to calculate $\sigma(\omega)$ (see Fig. 1).

In Sec. II we give the methodology, in Sec. III we present a comparison of our results with the available calculations and experimental data, and in Sec. IV we give our conclusion.

## II. METHODOLOGY

The interband contribution to the dielectric function for both the spins is given by[27]

$$\varepsilon_2^b(\omega)=\frac{8}{3\pi\omega^2}\sum_n\sum_{n'}\int_{BZ}|P_{nn'}(\bar{k})|^2\frac{dS_k}{\nabla\omega_{nn'}(\bar{k})} .$$

The above expression is written in atomic units with $e^2=1/m=2$ and $\hbar=1$. Here $\omega$ is the photon energy (Ry) and $P_{nn'}(\bar{k})$ is the dipolar matrix element between the initial $|n\bar{k}\rangle$ and final $|n'\bar{k}\rangle$ states with their eigenvalues $E_n(\bar{k})$ and $E_{n'}(\bar{k})$, respectively,

$$\omega_{nn'}(\bar{k})=E_{n'}(\bar{k})-E_n(\bar{k})=\omega$$

and the constant energy surface is

$$S_k=\{\bar{k};\omega_{nn'}(\bar{k})=\omega\} .$$

Using the LMTO-ASA, $P_{nn'}(\bar{k})$ can be easily calculated. Instead of going into detail, we refer the reader to Refs. 27 and 28.

## III. RESULTS AND DISCUSSIONS

Figure 2 shows the LMTO-ASA (Refs. 21 and 22) band structure of paramagnetic $Ni_3Al$ along some high-symmetry directions. Six bands 14–19 cut the Fermi level $E_F$. On comparison with the band structure given by van der Heide *et al.*, our band structure shows similar structure everywhere. The band structure of ferromagnetic $Ni_3Al$ is almost similar and therefore not presented. For the majority carriers six bands (14–19) intersect $E_F$ while for minority carriers five bands (13–17) do so. These differences will affect the optical conductivity.

Based on the theory discussed in the preceding section, we extended our self-consistent LMTO-ASA–based calculation for $Ni_3Al$ to calculate the optical conductivity. The calculations are done at two levels of sophistication. First the JDOS is calculated and then JDOS/$\omega$ gives the optical conductivity where the matrix elements are constant. Thereafter, optical matrix elements are calculated within the LMTO-ASA and then $\sigma(\omega)$ is calculated.

Results of our calculations of the $\sigma(\omega)$ with constant matrix elements and with matrix elements properly included are given in Figs. 3(a) and 3(b). The results of earlier calculation and experimental data are also shown in Fig. 1. The $\sigma(\omega)$, from JDOS/$\omega$ [Fig. 3(a)], compares well with the calculations of van der Heide *et al.*[10] We obtain peaks at 0.05 and 0.2 Ry with a minimum in between these two peaks at 0.1 Ry and thereafter a regular decrease in $\sigma(\omega)$. These peaks can be identified as transitions from bands 8–12 to band 16 near $\Gamma$ and from bands 15 and 16 to band 17 at $X$. The second peak results from transitions from bands 3–6 to bands 14 and 15 near $\Gamma$.

On including the matrix elements we see a significant change in the $\sigma(\omega)$. The positions of the maximum and minimum are shifted towards the higher-energy side. Now the maximum is at 0.12 Ry, a minimum at 0.3 Ry, and a next maximum at 0.4 Ry, and thereafter $\sigma(\omega)$ decreases with increase in $\omega$. On comparison with data, we see that neither the $\sigma(\omega)$ from JDOS/$\omega$ nor the $\sigma(\omega)$ using the calculated matrix elements gives an agreement with the experimental $\sigma(\omega)$ as far as peak positions are concerned. However, the general trend of the $\sigma(\omega)$ from the matrix element calculations agrees better with the experimental $\sigma(\omega)$. We note that our $\sigma(\omega)$ gives two peaks arising due to transitions from bands 5 and 6 to 14 and 15 near $\Gamma$ and from 3 and 4 to 14 and 15 near the same

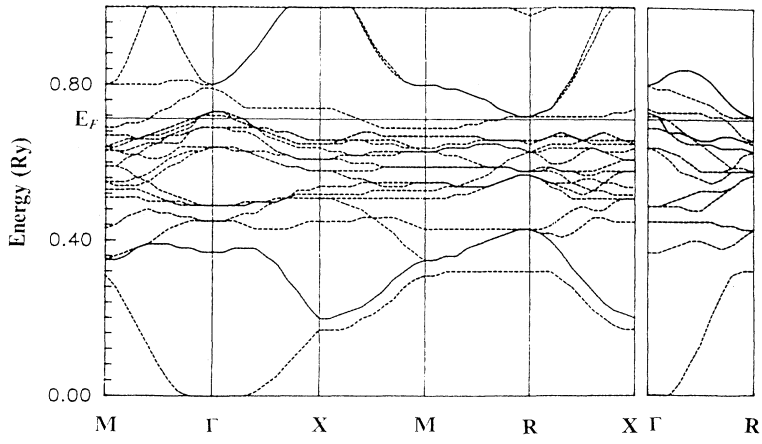


FIG. 2. The band structure of $Ni_3Al$ (paramagnetic) along high-symmetry lines.

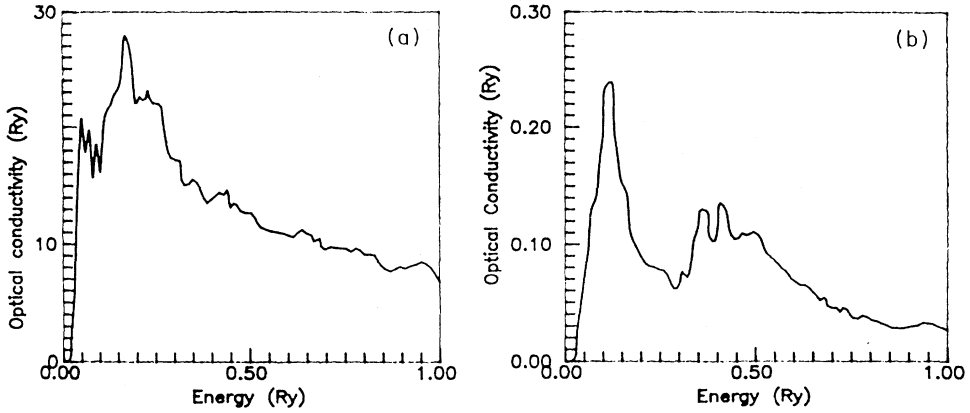


FIG. 3. The interband optical conductivity for $Ni_3Al$ using 95 $k$ points in the IBZ (a) with constant matrix elements and (b) with calculated matrix elements.

high-symmetry point. Inclusion of the matrix elements brings out these two peaks more clearly than the constant matrix element calculation where they are hardly discernible. Inclusion of broadening could again merge the peaks. To compare with experiment one must include this broadening. The experimental analysis of van der Heide *et al.* is unable to determine the magnitude of this broadening giving values of $\tau$ as $1\times10^{-15}$ or $3.4\times10^{-16}$ sec (about 0.6 and 2.0 eV).

van der Heide *et al.*[10] calculated JDOS/$\omega$ using eight-million pseudorandom points in the irreducible Brillouin zone (IBZ). To perform these calculations they fitted 22 bands to the Fourier-series expansion containing 40 stars of reciprocal vectors. The fit was made to 89 $k$ points in the IBZ with rms error of 1 mRy. For the $k$ integration, we have used the usual tetrahedron method.[29,30] In this method the matrix elements $P_{nn'}(\bar{k})$ and $\omega_{nn'}(\bar{k})$ at four corners of each tetrahedron are calculated with the help of the eigenvectors and eigenvalues obtained through the LMTO, whereas for any other $k$ point a linear interpolation scheme is used. For this a high number of $k$ points is required so that the difference between two successive $k$ points is so small that a linear interpolation scheme becomes valid. A test of the convergence of $\varepsilon_2(\omega)$ as a function of the number of $k$ points has been made by Knab and Koenig[31] for copper with 161, 946, and 2856 $k$ points in the IBZ. They reached the conclusion that the use of a finer mesh of 946 and 2856 $k$ points, as compared to 161 $k$ points, modifies only slightly the intensities of the peaks but not their positions. In the present work, we do not wish to do a comprehensive test of convergence, but we simply want to check the stability of the calculated results. With this objective in mind, we have performed calculations with 95 and 161 $k$ points. These results with 161 $k$ points are given in Figs. 4(a) and 4(b) (for constant and calculated matrix elements, respectively). The corresponding results for 95 $k$ points are already given in Fig. 3. When we compare Figs. 3 and 4, we do not see any dramatic difference. Henceforth, all calculations are done with 95 $k$ points.

Since $Ni_3Al$ is a weak ferromagnet with a magnetic moment of $0.31\mu_B$ per unit cell,[3] we have also performed calculations of $\sigma(\omega)$ for the majority carriers and for the minority carriers. Since the transition between up and

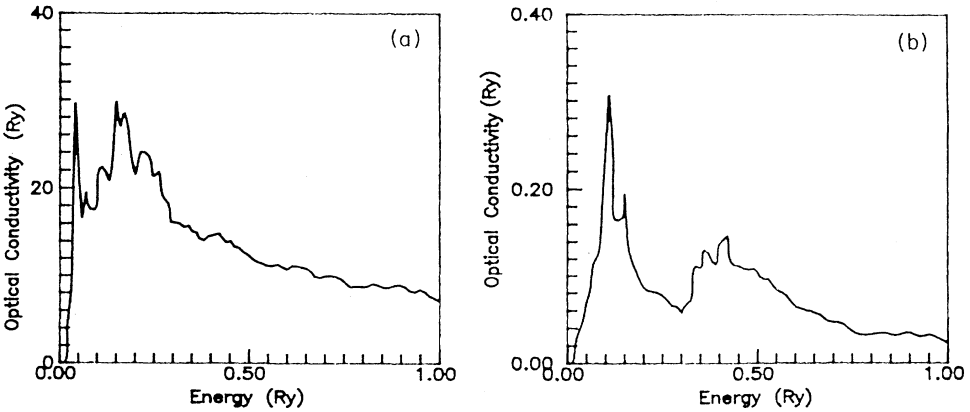


FIG. 4. The interband optical conductivity for $Ni_3Al$ using 161 $k$ points in the IBZ (a) with constant matrix elements and (b) with calculated matrix elements.

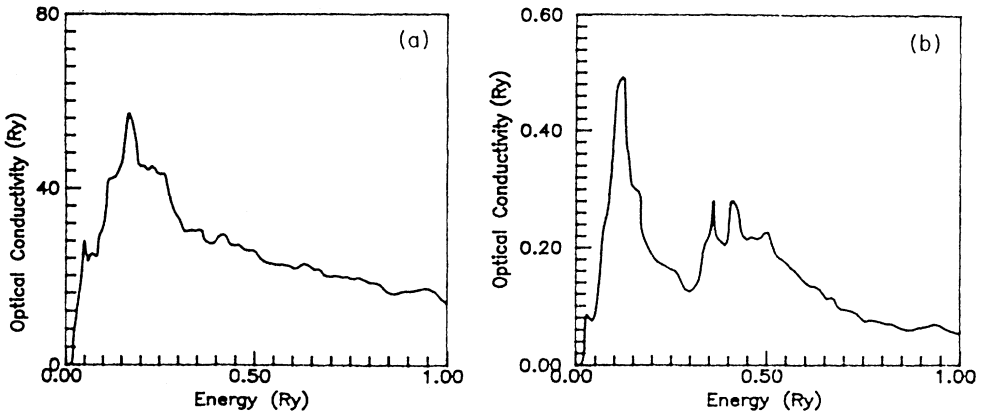


FIG. 5. The total interband optical conductivity for $Ni_3Al$ in the ferromagnetic case (a) with constant matrix elements and (b) with calculated matrix elements.

down spins will occur at low energies where the intraband transitions are dominant, we do not expect any significant change in $\sigma(\omega)$ if transitions between the majority and minority carriers are included. Thus $\sigma_{\text{tot}}(\omega)=\sigma_{\text{maj}}(\omega)+\sigma_{\text{min}}(\omega)$ is plotted in Figs. 5(a) and 5(b). Once again, in comparison with earlier paramagnetic calculations, we do not see any significant change in the structure of $\sigma(\omega)$ and in the position of maxima and minima. Thus, we may say that in a weakly ferromagnetic compound like $Ni_3Al$ the exchange interaction is not expected to lead to any major changes in $\sigma(\omega)$.

## IV. CONCLUSIONS

We have performed calculations of $\sigma(\omega)$ for $Ni_3Al$ with the LMTO-ASA with constant matrix elements and with calculated matrix elements. Our constant matrix element calculated JDOS/$\omega$ yields a maximum at 0.05 Ry, a minimum at 0.1 Ry, and another peak at 0.2 Ry. This is to be compared with the experimental data[10] where there is a shoulder at 0.06 Ry, a minimum at 0.2 Ry, and a maximum at 0.32 Ry. Inclusion of the matrix elements yields a $\sigma(\omega)$ which is in better overall agreement with the experiment but the positions of the maximum and minimum are changed. We obtain a maximum at 0.12 Ry, a minimum at 0.3 Ry, and a maximum at 0.4 Ry. While the constant matrix element calculations underestimate the position of the maximum and minimum, the $\sigma(\omega)$ with the optical matrix elements overestimates these. Such disagreement between theory and experiment is not uncommon. In order to improve the agreement between theory and experiment, one needs to include final-state effects for the transitions involving $d$ states. These have to be calculated as has been done by Kulikov *et al.*[32] for antiferromagnetic Cr. Since these calculations are tedious to perform, we adopt an alternate approximate method suggested by Janak, Williams, and Moruzzi.[33] Inclusion of the self-energy correction modifies the one-electron eigenvalues $E_n(\bar{k})$ to $E_n'(\bar{k})$ given by

$$E_n'(\bar{k})=E_n(\bar{k})+\Lambda_{nk}[E_n(\bar{k})-E_F]$$

for states near the Fermi level $E_F$. For simplicity one takes $\Lambda_{nk}$ to be a constant. Since the electron wave functions do not change, we write the optical conductivity as

$$\sigma(\omega)=\frac{1}{1+\Lambda}\sigma\left[\frac{\omega}{1+\Lambda}\right].$$

For copper one finds $\Lambda=0.05$ for good agreement with the data, whereas for Ni $\Lambda=-0.12$ and for Fe $\Lambda=-0.1$.[34] Although we have not adjusted $\Lambda$ to fit the experimental data, our calculations suggest that $\Lambda$ of about $-0.25$ would result in good agreement with the data.

## ACKNOWLEDGMENTS

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[1]F. R. de Boer, C. J. Schinkel, J. Biesterbos, and S. Proost, J. Appl. Phys. **40**, 1049 (1969).
[2]N. Buis, Ph.D. thesis, University of Amsterdam, 1979.
[3]N. Buis, J. J. M. Franse, and P. E. Brommer, Physica B+C **106B**, 1 (1981).
[4]W. de Wood and P. F. Chatel, J. Phys. F **3**, 1038 (1973).
[5]T. I. Sigfusson, N. R. Bernhoeft, and G. G. Lonzarich, J. Phys. F **14**, 2141 (1984).
[6]R. W. Dickson, H. B. Wachtman, and S. M. Copely, J. Appl. Phys. **40**, 2276 (1969).
[7]J. J. M. Franse, M. Rosen, and E. P. Wohlfarth, Physica B+C **86-88B**, 317 (1981).
[8]F. X. Kayser and C. Stassis, Phys. Status Solidi A **64**, 335 (1981).
[9]F. Wallow, G. Neite, W. Schröer, and E. Nembach, Phys. Status Solidi A **99**, 483 (1888).

[10]P. A. M. van der Heide, J. J. M. Buiting, L. M. tenDam, L. W. M. Schreurs, R. A. de Groot, and A. R. Vrooman, J. Phys. F **15**, 1195 (1985).
[11]S. M. Foiles and M. S. Daw, J. Mater. Res. **2**, 5 (1987).
[12]J. E. Osburn, M. J. Mehl, and B. M. Klein, Phys. Rev. B **43**, 1805 (1991).
[13]G. G. Fletcher, Physics **62**, 41 (1972); **56**, 172 (1971).
[14]D. Hackenbracht and J. Kubler, J. Phys. F **10**, 427 (1980).
[15]J. J. M. Buiting, J. Kubler, and F. M. Mueller, J. Phys. F **13**, L178 (1983).
[16]A. P. Maclin, G. M. Stocks, and W. M. Temmerman, in *High Temperature Alloys, Bethesda,* edited by J. O. Stigler (Metallurgical Society of AIME, Bethesda, MD, 1984).
[17]B. I. Min, T. Oguchi, H. J. F. Jansen, and A. J. Freeman, J. Magn. Magn. Mater. **54-57**, 1091 (1986).
[18]B. I. Min, A. J. Freeman, and H. J. F. Jansen, Phys. Rev. B **37**, 6757 (1988).
[19]X. Jian-hua, B. I. Min, A. J. Freeman, and T. Oguchi, Phys. Rev. B **41**, 5010 (1990).
[20]T. Nautiyal and S. Auluck, Phys. Rev. B **45**, 13 930 (1992).
[21]O. K. Andersen, Phys. Rev. B **12**, 3060 (1975).
[22]H. L. Skriver, *The LMTO Method* (Springer, New York, 1984).
[23]U. von Barth and L. Hedin, J. Phys. C **5**, 1629 (1972).
[24]O. K. Andersen, O. Jepsen, and M. Sob, in *Electronic Band Structure and Its Applications,* edited by M. Yussouff, Lecture Notes in Physics Vol. 283 (Springer, Berlin, 1987).
[25]R. Ahuja, A. K. Solanki, and S. Auluck, Phys. Scr. **45**, 621 (1992).
[26]A. R. Williams, J. Kubler, and C. D. Gelatt, Jr., Phys. Rev. B **19**, 6094 (1979).
[27]C. Koenig and M. A. Khan, Phys. Rev. B **17**, 6129 (1983).
[28]M. Alouani, J. M. Koch, and M. A. Khan, J. Phys. F **16**, 473 (1986).
[29]O. Jepsen and O. K. Andersen, Solid State Commun. **9**, 1763 (1971).
[30]G. Lehman and M. Taut, Phys. Status Solidi **54B**, 469 (1972).
[31]D. Knab and C. Koenig, J. Phys. Condens. Matter **2**, 465 (1990).
[32]N. I. Kulikov, M. Alouani, M. A. Khan, and M. V. Magnitskaya, Phys. Rev. B **36**, 929 (1987).
[33]J. F. Janak, A. R. Williams, and V. L. Moruzzi, Phys. Rev. B **11**, 1522 (1975).
[34]D. G. Laurant, J. Callaway, and C. S. Wang, Phys. Rev. B **20**, 1134 (1979).