International Journal of Scientific Research in Science and Technology
© 2018 IJSRST | Volume 4 | Issue 2 | Print ISSN: 2395-6011 | Online ISSN: 2395-602X

A Critical Study of Lattice Dynamics and Elastic Properties of the 4 Electron System

Dr. Anil Kumar Singh
M.Sc., Ph.D.(Physics)
B.R.A. Bihar University, Muzaffarpur (Bihar)

INTRODUCTION

CeN is the first member of the series of rare-earthmononitrides,1,2which all crystallize in the rock-salt crystalstructure. The rare-earth mononitrides have received renewedattention due to their peculiar electronic and magnetic proper-ties and their potential application in spintronics.3–6CeN is dif-ferent from the other members of the series in several respects.The lattice constant of CeN is significantly smaller1,4than forthe other rare-earth mononitrides, while the susceptibility ischaracteristic of a temperature-independent paramagnet,1,7incontrast to the local moment magnetism encountered in thelater nitrides.4Furthermore, the thermal expansion shows anunusual increase in lattice parameters at elevated temperaturesaccompanied by a dip in the susceptibility7–9and an increasein resistivity and Seebeck coefficients.10 Also, a changein reflectivity with temperature has been observed,11 andanomalous lattice expansion upon alloying with O occurs.12All these observations suggest that the rigid trivalent pictureof the Ce ions in CeN with localized f1electrons is inadequate,and instead felectrons contribute to the chemical bonding.In the mixed-valent picture,13–15 the localized felectronshybridize weakly with the conduction electrons, leading toa ground state composed of a mixture of f0and f1ions. Thisleads to an effective Ce valence in CeN between 3 and 4, whichbecomes very sensitive to external parameters like pressure andtemperature. Core-level photoemission spectroscopy on CeNshow clear signals of both valencies,16 while the interpretationof absorption spectra is less clear.8,17 Valence photoemissionexperiments16,18 as well as inverse photoemission spectra19 areoften interpreted in the Gunnarsson-Sch¨onhammer model20(GS), which considers a single Ce impurity in a conductionsea. The Ce ion fluctuates between the f0and f1configuration.

Models the mixed-valent state. However, for CeN theenergy of the f1state is much lower than that of the f0state,and the GS picture of the ground state becomes that of Kondoscreened local moments.18 The derived Kondo temperature forCeN is in between those of the αand γphases of elementalCe (Ref. 18).

The effective Ce valence is near to 3 (seeRef. 21). In contrast, optical conductivity data have beeninterpreted in terms of an effective valence of 3.48,22 while theeffective valence extracted from the lattice constant is muchcloser to 4, around 3.85 at room temperature but approaching3 at temperatures around 1100 K.7This latter result is inaccordance with the Kondo model, where an effective valencearound 3 at high temperatures is reached as a result of thedestruction of the screening and the formation of disorderedlocal fmoments.

V. KANCHANA et al. PHYSICAL REVIEW B 84, 205135 (2011)state is not trivalent in the traditional sense with localizedcorelike felectrons. The inverse photoemission spectrumof CeN (Ref. 19)showsfcharacter just above the Fermilevel, which also concurs with the occurrence of a Ce fbandpinned to the Fermi level. However, not all measured spectralproperties of the unoccupied 4fstates are reproduced by thispicture. In particular, the distinct feature observed19 at 6 eVabove the Fermi energy cannot be explained but rather reflectsthe large on-site Coulomb correlation energy associatedwith the formation of f2ions in the inverse photoemissionprocess. The optical conductivity calculated in the bandpicture is in excellent agreement with experimental data, incontrast to the result based on a localized (corelike) pictureof felectrons.25 Also, the magnetic susceptibility and linearspecific heat coefficient calculated assuming f-band formationcompares favorably with the experimental data.25 In particular,the latter comes out too high in the Kondo

COMPUTATIONAL DETAILS

Series of theoretical tools have been employed to studythe electronic structure of CeN. The all-electron full potential(FP) linear muffin-tin orbital method30 (LMTO) is usedto calculate the total energies as well as the ground-stateproperties.31 Here the crystal is divided into two regions:nonoverlapping muffin-tin spheres surrounding each atomand the interstitial region between the spheres. We used adouble-κspdfLMTO basis (each radial function within thespheres is matched to a Hankel function in the interstitialregion of decay constant κ) for describing the valence bands.Basis functions of Ce(5s,6s,5p,5d,4f) and N(2s,2p) characterhave been used in the calculation. Within the spheres, thepotential is expanded in terms of spherical harmonics, whilein the interstitial region it is expanded in terms of planewaves.

CONCLUSIONS

The ground-state properties and electronic structure of CeNhave been investigated by several theoretical tools. The resultshave been discussed in relation to available experimentalinformation and with emphasis on the nature of the Ce fstatesin this compound. The lattice constant and bulk modulus arecalculated with the generalized gradient approximation for thetotal energy, which includes a

cohesive contribution due tothe felectrons, and agree well with available experimentalinformation. The elastic constants and phonon dispersionrelations obtained using the GGA, as well as the Fermisurface, are other fundamental properties by which futureexperimental determinations may assist in clarifying the roleof the fstates in this compound. The unusual experimentalresults for the specific heat and lattice expansion at elevatedtemperatures suggest that a localization transition occursin CeN with temperature, similar to the α→γtransitionobserved for elemental cerium. This is consistent with theKondo picture of CeN, where the screening of Ce fmomentsis destroyed by increased temperatures, leading to fluctuatinglocal moments. In the density-functional framework, thismay be the state represented in the SIC-LSD approximationwith one localized (corelike) fstate per Ce atom, in whichcase virtually no cohesive energy is associated with the felectrons.

ACKNOWLEDGMENT

SO.E. acknowledges support from the Swedish Research Council, KAW Foundation, STEM, and ERC (Grant No.247062-ASD). Part of the calculations were carried out atthe Center for Scientific Computing in Aarhus (CSCAA)supported by the Danish Center for Scientific Computing.Y.M. acknowledges support from the National Natural Sci-ence Foundation of China

REFRENCES:

1. Malterre, C. Brouder, G. Krill, E. Beaurepaire, B. Carri´ere, andD. Chandesris, Europhys. Lett. 15, 687 (1991).18F. Patthey, S. Cattarinussi, W.-D. Schneider, and Y. Baer, Europhys.Lett. 2, 883 (1986);
2. F. Patthey, J.-M. Imer, W.-D. Schneider,H. Beck, Y. Baer, and B. Delley, Phys. Rev. B 42, 8864 (1990).19E. Wuilloud, B. Delley, W.-D. Schneider, and Y. Baer, J. Magn.Magn. Mater. 47-48, 197 (1985).20O.
3. Gunnarsson and K. Sch ¨onhammer, Phys.Rev.Lett.50, 604(1983); Phys. Rev. B 28, 4315 (1983).21Reference 18 quotes both values of nf=0.82 (text) and nf=0.92(table and figure) for the fcount in their Kondo model calculationsfor CeN, corresponding to effective valencies of 3.18 or 3.08.19 gives a value of nf=0.83 (figure).22J. Schoenes, in Handbook on the Physics and Chemistry of theActinides, edited by A. J. Freeman and G. H. Lander (North-Holland, Amsterdam, 1984), Vol. 1, Chap. 5;
4. J. Schoenes, inMoment Formation in Solids. Proceedings of the NATO AdvancedStudy Institute, edited by W. J. L. Buyers , p. 237.23
5. W. E. Pickett and B. M. Klein, J. Less-Common Met. 93, 219(1983).24M. S. S. Brooks, J. Magn. Magn. Mater. 47-48, 260 (1985).25A.