The Magnetic Susceptibility of Transition-Metal A15-Type Phases*

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The temperature dependence of the paramagnetic susceptibility was measured for several binary A15-type compounds of transition elements. Measurements were made between room temperature and either 3°K or the superconducting transition temperature, whichever was higher. The susceptibilities were found to have, in most cases, a small linear temperature dependence. Molar susceptibilities $\chi_M$, superconducting transition temperatures $T_c$, and electronic specific heat coefficients $\gamma$ for these compounds, when plotted as a function of the valence-electron concentration $e/a$, reveal peaks at an $e/a$ of about 6.5 for all three properties. Compounds containing the 4d-series transition-elements molybdenum or niobium have higher $T_c$ and lower $\chi_M$ values than compounds containing the 3d transition-element chromium or vanadium. It appears that an increased mixing of atoms on either of the two crystallographic sites in the A15-type structure can be associated with a significant decrease in the paramagnetic susceptibility of the compound.

1. INTRODUCTION

Intermetallic compounds with the A15-type structure often possess unusually good superconducting properties. A satisfactory theoretical model for this behavior has not been developed. The existence of mutually orthogonal one-dimensional "chains" of atoms in the A15-type structure has been the basis of

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band-structure calculations by Weger\textsuperscript{1} and by Labbé, Barisic, and Friedel.\textsuperscript{2} The calculations, however, were not in good agreement with the more general augmented plane-wave (APW) calculations published by Mattheiss.\textsuperscript{3} The band-structure calculations in the three investigations suggest that $d$-electron wave functions interact strongly at the Fermi level.

Experimental NMR and susceptibility measurements\textsuperscript{4--7} for the A\textsubscript{15}-type compounds V\textsubscript{3}X have been discussed in a previous paper by Clogston and Jaccarino.\textsuperscript{6} In that study X represented, for the most part, nontransition elements. They observed that the Knight shift for vanadium atoms in these compounds is invariably positive, but the Knight shift for the X atoms is negative except for cobalt. Often the magnetic susceptibility and Knight shift were temperature dependent, particularly for compounds with high superconducting transition temperatures. The model proposed by Clogston and Jaccarino accounted for the observations on this limited class of A\textsubscript{15}-type compounds. To further examine the characteristics of this interesting structure type, measurements have been extended to several binary A\textsubscript{15}-type compounds in which both atomic species are transition elements.

The present investigation was motivated by the availability of well-characterized compounds, which, in most cases, were the identical specimens employed in previous studies of long-range atomic ordering\textsuperscript{8} and superconducting transition temperatures.\textsuperscript{9,10} Consequently, the present study overcomes the deficiency of inadequate sample characterization that has limited the usefulness of many previous investigations.

### 2. EXPERIMENTAL PROCEDURE

All compounds used in this investigation were prepared by arc melting elemental materials (> 99.9\% pure) in an inert gas atmosphere. Further information on the annealing treatments and x-ray diffraction procedures is given in publications by van Reuth and Waterstrat.\textsuperscript{8,11} All stoichiometric (A\textsubscript{A}X) and most of the other compounds were submitted to spectrographic analysis to establish that the purity of the starting materials had not been adversely affected during sample preparation. Irregularly shaped samples of 200–500 mg were used after etching in a 10\% HNO\textsubscript{3} solution containing a trace of HF to remove possible surface contamination. No surface discoloration occurred during etching.

Magnetic susceptibilities were measured by the Faraday force method. The samples were placed in a commercially pure (2S) aluminium bucket that was suspended from one arm of an Ainsworth microbalance by means of a fine, high-purity gold-wire chain. A section of the chain was replaced by a thin quartz rod, which served as a thermal barrier. A 15-in. electromagnet, fitted with tapered pole caps, provided constant field gradients (up to 100 Oe/cm) in the vertical direction and magnetizing fields (up to 14 kOe) in the horizontal direction. Isothermal measurements, between either 3\textdegree{}K or the superconducting transition temperature (whichever was higher) and 300\textdegree{}K, were made in a variable-temperature cryostat. A copper resistance thermometer and a germanium thermistor, located in the tail of the cryostat, permitted temperature determinations to ±0.1\textdegree{}K. The calibra-