

Structural transformation of near-equiatomic V-Ru compounds at high pressure

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The structural transformation of near-equiatomic V-Ru compounds has been studied resistively under hydrostatic pressure up to ~ 21 kbar. It is found to be suppressed by the application of pressure. The rate of suppression increases as the Ru content approaches the critical concentration for the occurrence of the transformation. A critical pressure of 14 ± 2 kbar is obtained for the stabilization of the cubic phase down to 0 K in a sample containing 46-at.% Ru. The results are interpreted as support for the suggestion that the enhanced superconductivity in this compound system is associated predominantly with the singular feature of its electron energy spectrum.

I. INTRODUCTION

The crystal lattice of near-equiatomic V-Ru compounds with Ru concentration C larger than a critical value $C_k \approx 45.5$ at.% undergoes a cubic $B2$ (CsCl) to tetragonal $B2$ transformation upon cooling.¹ The structural transformation temperature T_L increases rapidly with increase of C . C_k is also associated with the dramatic appearance of superconductivity, with the superconducting transition temperature T_c peaking sharply about it.¹⁻³ The structural transformation is accompanied by an anomalous behavior of the electrical resistivity,¹ magnetic susceptibility,¹ and Knight shift⁴ of ^{51}V . From the form of these anomalies, it was proposed^{1,4} that the structural transformation is electronic in nature and results in a large drop in d -electron density of states in going from the cubic to the tetragonal phase. Alternatively, on the basis of his sound-velocity measurements, Testardi⁵ has suggested that the T_c peak at C_k is caused by the softness of the lattice.

The concurrence of structural transformation and high-temperature superconductivity has stimulated extensive investigations both experimental and theoretical on these two phenomena, particularly for the A15 compounds, where attempts have been made to account for the high T_c of these compounds.^{6,7} Some approaches have emphasized the fine structure of the d -electron subbands, characteristic of the peculiar linear-chain arrangement of the transition-metal atoms in these compounds. Others have treated the softness of lattice, which ultimately leads to the structural transformation, as the dominant factor. While the former, which is specific to the A15 compounds, is capable of accounting for the signs of the pressure dependences for T_c for V_3Si ($\partial T_c / \partial P > 0$)^{8,9} and Nb_3Sn

($\partial T_c / \partial P < 0$),^{8,10,11} provided the appropriate assumptions are made regarding the d -subband occupation and its variation with pressure,¹¹ the latter¹² predicts a general quadratic pressure dependence for T_c , with a maximum T_c at a pressure where $T_c = T_L$. While any deviation from linearity of the measured variation^{8,9} of T_c with pressure for V_3Si is at least an order of magnitude less than that predicted,¹³ the measurements do not extend to high enough pressure to test the possibility of T_c passing through a maximum at T_L .

Measurements¹⁴ of the variation of T_c and T_L under pressure for a range of $\text{Hf}_{1-x}\text{Zr}_x\text{V}_2$ compounds are also consistent with $\partial T_c / \partial P > 0$ for $\partial |T_L - T_c| / \partial P < 0$, but again the pressure does not reach that required for $T_c = T_L$.

In order to examine further the interrelationship between T_L and T_c we have undertaken a high-pressure study of the near-equiatomic composition alloys in the V-Ru system. Although these compounds can hardly be regarded as high- T_c superconductors, their maximum $T_c \sim 5.5$ K is high relative to their structure class. Other than the V-Ru system, only a very limited number of superconductors with $B2$ structure has been found and none of these has a T_c greater than 3 K.¹⁵

A detailed account of the influence of pressure upon T_c for the V-Ru compounds has already been presented.³ The present paper extends a preliminary report¹⁶ on the pressure dependence of T_L , as determined from measurements of the electrical resistivity as a function of temperature at fixed pressure up to ~ 21 kbar for samples with $C = 46, 47$, and 48 . These results are related to the previous conclusions, which were based upon the T_c measurements^{1,3,4} regarding the nature of the influence of the structural distortion upon T_c .

II. EXPERIMENTAL

Three samples ($C=46, 47$, and 48) of length ~ 3 mm and cross section ~ 0.6 mm² were cut from the V-Ru buttons prepared for the experiments in Ref. 1. Pt leads were spot-welded onto the samples. A standard four-lead technique using a PAR HR-8 lock-in amplifier was employed to measure the electrical resistivity ρ at 28 Hz. No difference between this low-frequency ρ and the dc ρ was found. The temperature was determined by a Au + 0.07-at. %-iron-Chromel thermocouple for the sample with $C=46$, and by a Chromel-Alumel thermocouple for the samples with $C=47$ and 48 . The Chromel-Alumel thermocouple was calibrated against the Au + 0.07-at. %-iron-Chromel thermocouple¹⁷ at low temperature and atmospheric pressure. To determine the T_c , a Ge resistance thermometer was used. The hydrostatic pressure environment was provided by a fluid mixture of 1:1 *n*-pentane and isoamyl alcohol (for $C=46$ up to 21 kbar) or 1:1 kerosene and silicone oil (for $C=47$ and 48 up to 12 kbar). The clamp technique was employed to contain the pressure which was generated in a press at room temperature. The high-pressure clamp was then removed from the press and cooled slowly inside the cryostat. A superconducting Pb-manometer¹⁸ situated next to the sample was used to determine the pressure at low temperature for $C=46$. For $C=47$ and 48 , the pressure was determined at room temperature by a gauge which had been calibrated against the 25.4-kbar Bi transition. No correction to these values was made to allow for the small pressure loss¹⁹ which occurs on cooling due to the freezing of the medium. The electrical leads were brought out from the high-pressure cell through a Stycast 2850 FT Epoxy seal.²⁰

III. RESULTS AND DISCUSSION

We have measured and show in Fig. 1 the temperature dependence of ρ at atmospheric pressure normalized to its value at 273 K for samples with $C=45, 46, 47$, and 48 . The normalized resistivity ρ_n initially follows the normal metallic behavior and decreases upon cooling, but then rises rapidly at a temperature which depends strongly on C and finally levels off. It has been demonstrated¹ that this anomalous rise in ρ_n is associated with the cubic to tetragonal structural transformation. Since the general temperature dependence of ρ_n prior to the transformation is similar for all samples, the form of ρ_n for the sample with $C=45$, which behaves normally, is considered to be representative of that expected for samples with $C > 45$, had there been no structural change. Hence one may estimate the fractional change in ρ_n due to the transformation for different values of C . It is evident from Fig. 1 that

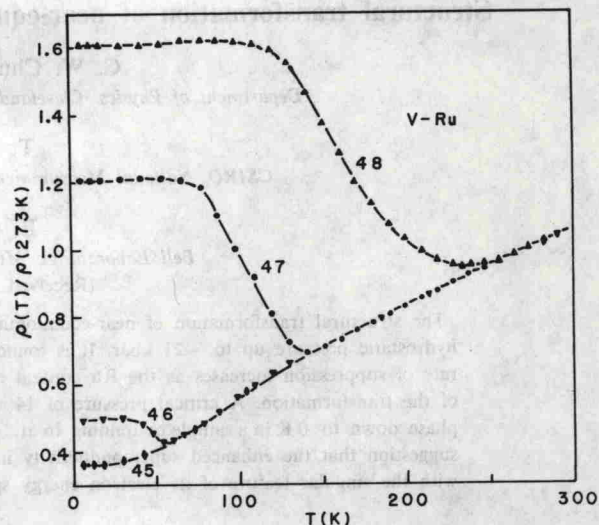


FIG. 1. Temperature dependence of the normalized resistivity. The numbers represent the atomic percent of Ru.

the fractional change of ρ_n increases with C in accord with the changes observed for the other electronic properties.

The three samples with $C=46, 47$, and 48 were studied under hydrostatic pressure. At temperatures above the structural transformation ρ was found to be independent of compression to within $\pm 3\%$ up to ~ 21 kbar. Typical results are represented by those for the sample with $C=46$ shown in Fig. 2. Here we have plotted ρ , normalized to that at 72 K, as a function of temperature at the pressure, in kbar, denoted by the number associated with each curve. All curves vary in a similar way before the anomalous rise in ρ begins to appear. For $P \geq 15$ kbar, no ρ anomaly was detected above T_c . Between ~ 15 and ~ 21 kbar, ρ_n at 10 K decreases slightly with pressure, but by no more than 4%, and all ρ_n -vs- T curves fall into the shaded region in Fig. 2.

In Ref. 1 the lattice distortion for the V-Ru compounds was characterized by a transformation temperature T_L , defined to be the temperature at which the minimum in ρ occurred. While it is clear from Fig. 2 that T_L , defined in this manner, is displaced towards lower temperature under pressure, it is difficult to determine the exact P dependence since the anomaly in ρ is suppressed under pressure, particularly for the sample with $C=46$. To better describe the P effect on the structural transformation, we have considered a quantity $\Delta\rho_n$ as a measure of the fractional change in electronic properties due to the transformation. Because of the similar behavior of ρ_n prior to the appearance of the transformation both with and without pressure,