Enhancement of the upper critical field in the cubic Laves-phase superconductor HfV$_2$ by Nb doping

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Abstract

We report the effect of Nb doping on the upper critical field of the cubic Laves-phase superconductor HfV$_2$ studied in a series of HfV$_{2-x}$Nb$_x$ samples with $0 \leq x \leq 0.3$ under pulsed fields up to 30 T. The undoped HfV$_2$ undergoes a martensitic structural transition around 110 K, and becomes superconducting below $T_c = 9.4$ K. Upon Nb doping, while the structural transition is suppressed for $x \geq 0.1$, a maximum in $T_c$ of 10.1 K and zero-temperature upper critical field $B_{c2}(0)$ of 22.4 T is found at $x = 0.2$, which is ascribed to an increase of the density of states at the Fermi level. For all samples, the temperature dependence of $B_{c2}$ can be well described by the Werthamer–Helfand–Hohenberg theory that takes into account both the spin paramagnetic effect and spin orbit scattering. In addition, a comparison is made between the $B_{c2}$ behavior of HfV$_{2-x}$Nb$_x$ and those of Nb–Ti and Nb$_3$Sn.

Keywords: Cubic Laves-phase superconductors, Upper critical field, Density of states at the Fermi level

(Some figures may appear in colour only in the online journal)

1. Introduction

HfV$_2$ based superconductors with the C15 cubic Laves-phase structure have received considerable attention because of their potential as high-field superconducting magnets [1, 2]. Similar to A15-type Nb$_3$Sn, HfV$_2$ undergoes a martensitic transition involving two successive structural modifications from cubic to tetragonal then to orthorhombic in the temperature range between 100 and 115 K [3], followed by a superconducting transition below $T_c \sim 9$ K [4]. Despite its relatively low $T_c$, the upper critical field $B_{c2}$ of HfV$_2$ has been found to be $\sim 20$ T at 4.2 K, and can be improved to $\sim 23$ T by alloying with ZrV$_2$ [5], which is the highest for superconductors composed only of transition metal elements. Furthermore, compared with Nb$_3$Sn, the HfV$_2$-based C15 compounds are more resistant to neutron radiation [6] and less brittle [7], which make them promising for application in nuclear fusion reactor.

For superconductors with spin-singlet Cooper pairs, superconductivity (SC) can be destroyed by the application of magnetic field via two effects. The first one is the orbit effect, which is a manifestation of the Lorentz force. The second one is the spin paramagnetic effect, which tends to align the spin of Cooper pairs and can be partially cancelled out by introducing the spin–orbit scattering. The investigation of the $B_{c2}$ has been shown to provide valuable information on the
contributions from these effects [8–10]. In the case of Hf1-xZrxV2, the Bc2 value was measured across 0 \leq x \leq 1, by only at 4.2 K [5]. In addition to Zr, various other dopants, such as Ta [11], Nb [12], Fe [13], Co [13], Ni [13], have been introduced in the HfV2 system to investigate the impurity effect on superconductivity and structural transition. For example, dilute Nb doping (<5%) is found to suppress the structural transition and increase Tc [12]. However, no systematic study on the temperature and doping dependences of Bc2 has been reported for any of these dopants.

In this paper, we present a study on the superconducting properties of HfV2−xNb x for x in the range of 0–0.3, focusing on the Bc2(T) behavior. Bulk SC with indication of strong coupling nature is observed in this x range, and a maximum Tc of 10.1 K is found at x = 0.2. The Bc2(T) data for all x values are well fitted by the WHH model including the effects of spin paramagnetism and spin orbit scattering. The extrapolated Bc2(0) exhibits a maximum of 22.4 T also at x = 0.2, which is 4.1 T higher than that of undoped HfV2. The enhancement of both Tc and Bc2(0) is attributed to an enhanced density of states of the Fermi level [N(EF)], as revealed by the specific heat results. We also compare the Bc2(T) curve at the optimal doping with those of Nb0.44Ti0.56 and Nb5Sn, and discuss its implication on the Bc2 improvement.

### 2. Experimental details

Polycrystalline HfV2−xNb x samples with x = 0, 0.1, 0.2, and 0.3 were prepared by the arc-melting method. High purity Hf, V (>99.99%, 200 mesh) and Nb (>99.99%, 200 mesh) powders were weighed according to the stoichiometric ratio and melted in an arc furnace under high-purity argon atmosphere. The melts were turned over and remelted by the same procedure several times to ensure homogeneity, following by rapid cooling on a water-chilled copper plate. The samples were used as cast without further annealing treatment, which was reported to result in Tc degradation [14]. The phase purity of the samples was checked by powder x-ray diffraction patterns at room temperature using the PowderX software. The chemical composition of theses samples was examined with an energy-dispersive x-ray (EDX) spectrometer (Model Octane Plus) affiliated to a Zeiss Supratm 55 schottky field emission scanning electron microscope. The spectra were collected on at least 3 different locations of each sample for averaging. The electrical resistivity was measured using a standard four-probe method. Resistivity and specific heat measurements down to 1.8 K and up to 9 T were carried out on regular-shaped samples in a Quantum Design PPMS-9 Dynacool. The typical dimension is 2 mm × 0.8 mm × 0.45 mm for resistivity measurements. The magnetoresistance under pulsed fields up to 30 T was measured at the National High Magnetic Field Center in Wuhan. The dc magnetization measurements down to 1.8 K were performed using a commercial SQUID magnetometer (Quantum Desgin MPMS3).

### 3. Results and discussion

Figure 1(a) shows the room temperature XRD patterns for the HfV2−xNb x samples. All the major peaks can be indexed on the cubic C15 structure with the space group Fd3m [3], and a schematic structure is shown in figure 1(b). The lattice parameter is plotted as a function of the nominal Nb content x in figure 1(c). For undoped HfV2 (x = 0), the refined a = 7.382(1) Å is in good agreement with the previous reports [3, 4]. In this compound, the V atoms form a network of corner sharing tetrahedron, and the nearest V–V distance dV−V = 2.616 Å is even smaller than that (2.620 Å) in body-center-cubic V. With increasing x, the a-axis expands monotonically to 7.399(1) Å. This is in line with the expectation that Nb substitutes V rather than Hf since r(V) < r(Nb) < r(Hf), where r is the atomic radius. In addition to the main C15 phase, there exist small extra diffraction peaks, which is ascribed to the Hf-based solid solution impurity. Base on the peak intensity at ~36.7° (marked by the asterisks), the impurity fraction is estimated to vary from ~8% to ~12%, depending on the Nb content. On the other hand, the chemical compositions of these samples were examined by an EDX spectrometer (Model Octane Plus) affiliated to a Zeiss Supratm 55 schottky field emission scanning electron microscope. The spectra were collected on at least 3 different locations of each sample for averaging. The electrical resistivity was measured using a standard four-probe method. Resistivity and specific heat measurements down to 1.8 K and up to 9 T were carried out on regular-shaped samples in a Quantum Design PPMS-9 Dynacool. The typical dimension is 2 mm × 0.8 mm × 0.45 mm for resistivity measurements. The magnetoresistance under pulsed fields up to 30 T was measured at the National High Magnetic Field Center in Wuhan. The dc magnetization measurements down to 1.8 K were performed using a commercial SQUID magnetometer (Quantum Desgin MPMS3).
temperatures of the resistive transitions, are 9.4 K, 9.88 K, 10.1 K, and 9.84 K for \( x = 0, 0.1, 0.2, \) and 0.3, respectively. The occurrence of SC in these HfV\(_{2-x}\)Nb\(_x\) samples is corroborated by the magnetic susceptibility results shown in figure 2(b). For each \( x \), a strong diamagnetic signal is detected, and its onset temperature corresponds well to the midpoint of the resistive transition except for \( x = 0 \), where the onset temperature coincides with the completion of the resistive transition. When cooling below 9 K, all the zero-field cooling susceptibility curves become flat, and the shielding fractions \(-4\pi\chi_{ZFC}\), where \( \chi_{ZFC} \) is the zero-field cooling susceptibility in the unit of emu cm\(^{-3}\), are calculated to vary between 110\% and 150\% without correction for the demagnetization effect.

The main panel of figure 3(a) shows the temperature dependence of the specific heat \( C_p/T \) for the HfV\(_{2-x}\)Nb\(_x\) samples. A \( C_p \) anomaly can be seen at \( \sim110 \) K for HfV\(_2\), in accordance with the martensitic structural transformation. However, the small magnitude and relatively broad width of this anomaly signify that the structural transition is of second order rather than first order \([12]\). This is probably because that our sample is slightly off stoichiometric, and, indeed, its \( T_m \approx 110 \) K is slightly lower than that the usual value of 116 K \([12]\). For \( x > 0.1 \), no such anomaly is discernible, in line with the previous study \([12]\). This indicates that the structural transformation is suppressed completely by this Nb doping level. According to \([12]\), the structural transition in HfV\(_2\) is due to the Fermi surface nesting. The doping of Nb introduces additional electrons, which fill unoccupied \( d-d \) hybrid states and result in a change in the Fermi energy. This could deteriorate the nesting condition and hence suppress the structural transition. On the other hand, a sharp \( C_p \) jump around \( T_c \) is found for all samples, which can be seen more clearly from the \( C_p/T \) versus \( T^2 \) plot shown in the inset of figure 3(a). This confirms the bulk nature of SC in these HfV\(_{2-x}\)Nb\(_x\) samples. Following \([12]\), the normal-state data are analyzed by the Debye model \( C_p/T = \gamma_e + \beta_3 T^2 + \beta_4 T^4 \), where \( \gamma_e \) and \( \beta_i \) (\( i = 3, 5, 7 \)) are the electronic and phonon specific heat coefficients, respectively. Here the high order terms are included since there is a downward curvature of the data. The best fits yield \( \gamma_e = 50 \) mJ mol\(^{-1}\) K\(^{-1}\), 65 mJ mol\(^{-1}\) K\(^{-2}\), 68 mJ mol\(^{-1}\) K\(^{-2}\), and 62 mJ mol\(^{-1}\) K\(^{-2}\) for \( x = 0, 0.1, 0.2, \) and 0.3, respectively, which are comparable to the previous study \([12]\). Since \( \gamma_e \) is proportional to \( N(E_F) \), these results not only indicate a large \( N(E_F) \) in HfV\(_{2-x}\)Nb\(_x\)\([15]\), but also point to an increase of \( N(E_F) \) by Nb doping. Figure 3(b) shows the temperature dependence of the normalized electronic specific heat \( C_{el}/\gamma_e T \) after subtraction of the phonon contribution. The normalized specific heat jumps \( \Delta C_{el}/\gamma_e T_c \) for these samples are about 1.7, which are significantly larger than the value 1.43 of the Bardeen–Cooper–Schrieffer (BCS) theory \([16]\) and point to a strong coupling superconducting state. In such case, the \( C_{el}/\gamma_e T \) jump can be analyzed by a modified BCS model, or the so-called \( \alpha \) model, where \( \alpha = \Delta_0/T_c \) and \( \Delta_0 \) is the size of fully isotropic gap at 0 K \([17]\). It turn out that the data of \( x = 0 \) can be well reproduced with \( \Delta_0/T_c = 2 \), which is very

Figure 2. Temperature dependence of (a) resistivity and (b) magnetic susceptibility near the superconducting transition for the HfV\(_{2-x}\)Nb\(_x\) samples. The resistivity data are normalized for better comparison. The inset in (a) shows a magnification of the normal-state resistivity data, and the anomaly due to the structural transition is marked by the arrow.

Figure 3. (a) Temperature dependence of specific heat \( (C_p) \) for the HfV\(_{2-x}\)Nb\(_x\) samples. The anomaly around 110 K for \( x = 0 \) is marked by the arrow. The inset shows the low temperature data plotted as \( C_p/T \) versus \( T^2 \). The solid lines are Debye fits to the normal-state data (see text for details). (b) Normalized electronic specific heat plotted as a function of \( T/T_c \) for these samples. The solid, dashed, and dashed dot lines denote the calculated \( C_{el}/T \) curves from the BCS theory and \( \alpha \) model.
close to that reported previously [18] and indeed larger than the BCS value of 1.764. For Nb-doped samples, the data almost overlap with each other and can be better fitted with \(\Delta_0/T_c = 2.25\) for \(T/T_c < 0.6\). Nevertheless, at higher \(T/T_c\), a discrepancy exists between the experimental and theoretical results, which is probably due to either the error in the estimation of phonon specific heat contribution or the existence of gap anisotropy.

To obtain \(B_{c2}(T)\) for the HfV\(_2\)–Nb\(_x\) samples, temperature dependent resistivity measurements at constant fields up to 9 T (in PPMS) are combined with isothermal magnetoresistivity measurements under pulsed fields up to 30 T. Examples for \(x = 0.2\) are shown in figures 4(a) and (b), respectively, and the suppression of SC with increasing field is evident in both cases. For consistency, the value of \(T_c (B_{c2})\) in each curve is determined as the onset temperature (field) of the resistive transition, and the resulting \(B_{c2} - T\) phase diagrams are displayed in figure 4(c). The initial slopes of \(B_{c2}(T)\), \(\frac{dB_{c2}}{dT} \bigg|_{T=T_c}\), are found to be \(-5.1\) T/K, \(-6.0\) T/K, \(-6.0\) T/K, and \(-5.4\) T/K for \(x = 0, 0.1, 0.2,\) and 0.3, respectively, which are typical for HfV\(_2\) based superconductors [18].

According to the WHH theory [8], the zero-temperature orbital upper critical field \(B_{c2}^{orb}(0)\) in the dirty limit is given by

\[
B_{c2}^{orb}(0) = -0.693 \left( \frac{d\rho}{dB} \right)_{T=T_c}.
\]

Using this formula, the calculated \(B_{c2}^{orb}(0)\) for the HfV\(_2\)–Nb\(_x\) samples falls between 33.2 and 41.9 T, which is much higher than the estimated \(B_{c2}(0) \sim 20\) T. This indicates that the spin paramagnetic effect is at work, and hence the Cooper pairs are in the spin-singlet state. Note that the paramagnetically limiting field \(B_{c2}^p(0)\) can be expressed as

\[
B_{c2}^p(0) = B_{c2}^{orb}(0) / \sqrt{1 + \alpha_M^2},
\]

where \(\alpha_M = \sqrt{2} B_{c2}^{orb}(0)/B_p(0)\) is the Maki parameter and \(B_p(0)\) is the Pauli limiting field at 0 K [9]. Since \(B_p(0) = \Delta_0/\sqrt{2}\mu_B\), where \(\mu_B\) is the Bohr magneton, the \(\alpha_M\) values are

![Figure 4](image-url)

**Figure 4.** (a) Temperature dependence of resistivity for \(x = 0.2\) under various magnetic fields up to 9 T. (b) Isothermal field dependence of resistivity up to 30 T for the same sample at different temperatures. The solid lines and arrows are a guide to the eyes. (c) Upper critical field \(B_{c2}\) versus temperature phase diagram for the HfV\(_2\)–Nb\(_x\) samples.

![Figure 5](image-url)

**Figure 5.** Normalized upper critical field \(h^*\) plotted as a function of the reduced temperature \(t = T/T_c\) for the HfV\(_2\)–Nb\(_x\) samples. (a) \(x = 0\); (b) \(x = 0.1\); (c) \(x = 0.2\); (d) \(x = 0.3\). The solid lines in each panel denote the theoretical WHH curves in the absence of spin paramagnetic effect and spin–orbit scattering \((\alpha_M = 0, \lambda_{SO} = 0)\), in the absence of spin–orbit scattering \((\alpha_M = 0)\), and in the presence of both spin paramagnetic effect and spin–orbit scattering \((\alpha_M = 0, \lambda_{SO} = 0)\). See text for details.
Table 1. Structural, chemical and physical parameters of the HfV$_{2-x}$Nb$_x$ samples.

| $x$     | $T_c$ (K) | $\gamma_e$ (mJ mol$^{-1}$ K$^{-2}$) | $T_b$ (K) | $B_{c2}^0$ (T) | $B_{c1}^0$ (T) | $\rho_c$ (m$\Omega$ m) | $\lambda_{so}$ |
|---------|-----------|-------------------------------|----------|----------------|----------------|------------------------|---------------|
| 0       | 32.0      | 5.1                           | 12.9     | 18.3           | 18.3           | 2.38                   | 0.5           |
| 0.1     | 31.5      | 4.8                           | 12.4     | 17.8           | 17.8           | 2.47                   | 0.4           |
| 0.2     | 31.0      | 4.5                           | 12.0     | 17.3           | 17.3           | 2.47                   | 0.5           |
| 0.3     | 29.5      | 4.0                           | 11.6     | 16.8           | 16.8           | 2.51                   | 0.3           |

(a) $T_c$ (K); (b) $\gamma_e$ (mJ mol$^{-1}$ K$^{-2}$); (c) $T_b$ (K); (d) $B_{c2}^0$ (T); (e) $B_{c1}^0$ (T); (f) $\rho_c$ (m$\Omega$ m); (g) $\lambda_{so}$.

Figure 6. Nb content $x$ dependence of (a) $T_c$, (b) $B_{c2}(0)$, (c) $\gamma_e$, and (d) $\alpha_M$ and $\lambda_{so}$. The vertical line is a guide to the eyes.

Table 2. Space group and superconducting parameters of HfV$_{1.8}$Nb$_{0.2}$, Nb$_{0.44}$Ti$_{0.56}$, and Nb$_3$Sn. The data for Nb$_{0.44}$Ti$_{0.56}$ and Nb$_3$Sn are taken from [21] and [22], respectively.

| $x$     | Space group | $T_c$ (K) | $B_{c2}(0)$ (T) | $B_{c1}(0)$ (T) | $\rho_c$ (m$\Omega$ m) | $\lambda_{so}$ |
|---------|-------------|-----------|----------------|----------------|------------------------|---------------|
| 0.0     | Im3m        | 8.9       | 14.2           | 15.5           | 2.9                    | 4.5           |
| 0.1     | Pm3n        | 17.6      | 28.9           | 30.2           | 2.4                    | 4.5           |
| 0.2     | Fd3m        | 10.1      | 22.4           | 24.1           | 2.3                    | 4.5           |

Figure 7. Comparison between the $B_{c2}$-$T$ phase diagrams of HfV$_{1.8}$Nb$_{0.2}$, Nb$_{0.44}$Ti$_{0.56}$ [21], and Nb$_3$Sn [22]. The dashed line represents the theoretical curve for HfV$_{1.8}$Nb$_{0.2}$ assuming the same spin–orbit $\lambda_{so}$ as Nb$_{0.44}$Ti$_{0.56}$.

To gain more insight, the values of $T_c$, $B_{c2}(0)$, $\gamma_e$, $\alpha_M$ and $\lambda_{so}$, from the above results are plotted as a function of the Nb content in figure 6. It turns out that all of $T_c$, $B_{c2}(0)$, and $\gamma_e$ follow a nonmonotonic $x$ dependence with a maximum at $x = 0.2$, hinting at a close relation between them, while no obvious feature is seen in the data of $\alpha_M$ and $\lambda_{so}$. In particular, compared with undoped HfV$_2$, the $B_{c2}(0)$ value is 4.1 T ($\sim$22%) larger at $x = 0.2$, which is remarkable since $T_c$ is increased only by 0.7 K ($\sim$7%). For strong coupling superconductors, their $T_c$ is given by the MaMillan formula [19]

$$T_c = \frac{\Theta_D}{1.45} \exp \left[ -\frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right],$$

with

$$\lambda = \frac{N(E_F)\langle I^2 \rangle}{M(\omega^2)}.$$

where $\Theta_D$ is the Debye temperature, $\mu^*$ is the Coulomb pseudopotential, $\langle I^2 \rangle$ is the average square electron-ion matrix element, $M$ is the atomic mass, and $\omega^2$ is the average square phonon frequency. Hence one could see that $T_c$ increases with increasing $N(E_F)$, which explains the enhancement of $T_c$ by Nb doping. On the other hand, it is noted from figure 5 that the $h^*$ values at 0 K are very similar for the HfV$_{2-x}$Nb$_x$ samples. It thus appears that $B_{c2}(0)$ is nearly proportional to the product of $T_c$ and $(dB_{c2}/dT)_{T=T_c}$. In the dirty limit, we have $(dB_{c2}/dT)_{T=T_c} \approx \gamma_e \rho_c$, where $\rho_c$ is the resistivity value just above $T_c$ [20]. Given that $\gamma_e$ increases by more than 30% with increasing $x$ from 0 to 0.2, it can be speculated that the concomitant enhancement of $B_{c2}(0)$ is also mainly due to an enhanced $N(E_F)$.

Finally, we present in figure 7 a comparison between the $B_{c2}$-$T$ phase diagrams of HfV$_{1.8}$Nb$_{0.2}$, Nb$_{0.44}$Ti$_{0.56}$ [21], and Nb$_3$Sn [22], all of which adopt the cubic structure but with different space groups (table 2). It is noted that the $B_{c2}(T)$

the analysis of $B_{c2}(T)$ behavior for HfV$_2$ based superconductors [18].
curve of HfV1.8Nb0.2 lies between those of Nb0.44Ti0.56 and Nb3Sn. In particular, the \( B_{c2}(0) \) value of HfV1.8Nb0.2 is much higher than that of Nb0.44Ti0.56, despite their similar \( T_c \). This illustrates the importance of C15 structure type for achieving high value of \( B_{c2}(0) \). Furthermore, as can be seen from table 2, \( B_{c2}(0) \) already approaches its orbital-limited value for Nb0.44Ti0.56 and Nb3Sn. By contrast, given its relatively small \( \lambda_{so} \), there is still plenty room for the \( B_{c2}(0) \) enhancement of HfV1.8Nb0.2. Indeed, assuming the same \( \lambda_{so} = 4.5 \) as Nb0.44Ti0.56, \( B_{c2}(0) \) of HfV1.8Nb0.2 would reach \( \sim 33 \) T, which is even higher than that of Nb3Sn. In this respect, it is worthy noting the following two relations: \( \lambda_{so} = 2\hbar/3\pi k_B T_c \tau_{so} \) \[10\] and \( \tau_{so}/\tau_{so} = (Z/137)^{4/5} \) \[23\], where \( \tau_{so} \) is the spin orbit scattering time, \( \tau_{so} \) is the transport lifetime and \( Z \) is the atomic number. Hence, doping with high \( Z \) elements, such as \( 4d \) or \( 5d \) elements, may increase \( \lambda_{so} \) and will be of interest for future studies.

4. Conclusion

In summary, we have studied the superconducting properties, with focus on the upper critical field, of polycrystalline C15-type Laves-phase compound HfV2-xNbx, with \( x \) up to 0.3. While the martensitic structural transition is suppressed for \( x \geq 0.1 \), a maximum in \( T_c \) (\( \sim 10.1 \) K) as well as \( B_{c2}(0) \) (\( \sim 22.4 \) T) is found at \( x = 0.2 \). Furthermore, the specific heat results indicate a strong coupling SC in the whole \( x \) range studied, and an increase of \( N(E_F) \) induced by Nb doping, which is suggested to be responsible for the enhancement of both \( T_c \) and \( B_{c2}(0) \). The \( B_{c2}(T) \) data of all samples are found to be well described by the WHH model considering the spin paramagnetic effect and spin orbit scattering. In addition, a comparison between the \( B_{c2}(T) \) behavior of HfV2-xNbx and those of Nb–Ti and Nb3Sn signifies the importance of spin orbit scattering in the \( B_{c2}(0) \) improvement for the former case. Our results call for further studies to increase the density of states at the Fermi level and spin orbit scattering in the HfV2 based Laves-phase superconductors, which may facilitate the high-field application of these materials.

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