Magnetic and Superconducting Properties of Vanadium Nanoconstrictions

Hiroki Takata, Yuji Inagaki, Tatsuya Kawae
Department of Applied Quantum Physics, Kyushu University, Fukuoka. 819-0385, Japan
E-mail: 2te13283s0s.kyushu-u.ac.jp

Koichiro Ienaga
Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan

Hiroyuki Tsujii
Department of Physics, Faculty of Education, Kanazawa University, Kanazawa 920-1192, Japan

Abstract. We study the magnetic and superconducting properties in a paramagnetic vanadium (V) nanoconstriction with changing its size using a mechanically controllable break junction technique. In the normal state the magnetoresistance effect is observed below the diameter \( d \approx 8 \text{ nm} \). Moreover, a Fano resonance appears around zero-bias voltage in the differential conductance for the atomic-size contacts and changes shape as the size of the constriction changes. On the other hand, below the superconducting critical temperature the superconducting gap features in V contacts are largely different from those in Pb contacts which exhibit typical features expected in superconducting point contacts. Only a single Andreev anomaly at \( 2\Delta \), where \( \Delta \) is the superconducting energy gap, is observed in the spectra of the V contacts, while two anomalies at \( \Delta \) and \( 2\Delta \) appear in the case of the Pb contacts. In the tunnel conductance regime, the structure of the superconducting quasiparticle tunneling density of states is not seen in the V spectra in contrast to the Pb spectra. The origin of these features is discussed.

1. Introduction

When the system size is decreased below the characteristic length such as the correlation length, qualitatively different behavior associated with the reduced dimensionality can be observed. For example, the low dimensionality brings about an enhancement in the magnetization [1] or an emergence of magnetic anisotropy [2, 3]. One of the important issues for the low dimensional magnetic system is the appearance of ferromagnetism, which occurs in nonmagnetic materials. A spontaneous long-range ferromagnetic ordering is observed only in several 3\( d \) transition (Fe, Co, and Ni) and 4\( f \) rare-earth (Gd, Tb, Dy, Ho, Er, and Tm) metals, while several theoretical calculations predict the appearance of the ferromagnetism in transition metals for systems of reduced dimensionality, e.g., clusters or ultrathin films [4, 5]. Actually, the ferromagnetic ordering is found in various transition metals, e.g., Ru monolayers on a C(0001) substrate [6] and Pd nanometer-sized particles [7].
We have investigated ferromagnetic transitions of paramagnetic Pd caused by downsizing via transport measurements using a mechanically controllable break junction (MCBJ) technique [8]. A clear magnetoresistance (MR) effect with hysteresis is observed in Pd nanoconstrictions. These results suggest that the increase of the lattice parameter or of the surface to volume ratio gives rise to a ferromagnetic transition by stretching the sample wire using the MCBJ technique.

Here, we focus on metallic vanadium (V). Bulk V exhibits a superconducting transition at $T_C = 5.3$ K, while the emergence of a magnetic moment is reported for V fine particles [9]. In addition, a theoretical study predicts the magnetic behavior of V metal when the lattice parameter is increased [10]. From these results, we consider that the magnetic behavior may be detected in V nanoconstrictions by stretching a V wire using the MCBJ technique as in the case of Pd. This implies that the V nanocontact is a good candidate to study the competition between superconductivity and magnetism.

In this study, we have examined magnetic and superconducting properties of V nanoconstrictions using the MCBJ technique. From the MR and differential conductance measurements for $T > T_C$, we suggest that magnetic moments are induced in the V constriction when the size is decreased to the nanometer range. For $T < T_C$, only a single Andreev anomaly at $2\Delta$ is observed in the V nanoconstrictions, while two anomalies at $\Delta$ and $2\Delta$ are seen in the Pb nanoconstrictions. In the tunnel conductance regime, the structure of the superconducting quasiparticle tunneling density of states is not seen in the spectra of the V system in contrast to the case of the Pb system. However, for the V nanoconstrictions, we cannot obtain evidence for impurity effects by the induced moments in the superconducting tunnel spectra of the junctions.

2. Experimental

V nanoconstrictions were prepared by the following procedure. A commercial V polycrystalline wire of 0.2 mm in diameter with a purity of 99.8 %, which contains 200 ppm of Fe and 10 ppm of Mn magnetic impurities and other nonmagnetic impurities such as 1,000 ppm of Cu and 500 ppm of Si, was used as the sample. The V wire, which is notched at its center, was rigidly fixed on top of a phosphor-bronze bending beam and mounted in a three-point bending configuration in a vacuum chamber. After cooling the chamber down to liquid He temperature, the V wire was stretched by bending the beam to an appropriate size with a micro screw to prevent contamination at the contact due to outgassing. A piezo element was then used for precise control of the constriction size. By combining these methods, the size of the V constrictions can be varied from a few micrometers down to an atomic scale. Note that the absence of temperature gradients and/or variations, in the MCBJ experiments at liquid He temperatures, favors a stable contact geometry. By applying this method, we study the size dependence of the magnetic properties for $T > T_C$ and the superconducting properties for $T < T_C$.

The resistance was measured with an LR700 AC resistance bridge (Linear Research Inc.) [8]. The magnetic field of $|H| \leq 600$ Oe with a typical sweep speed of 400 Oe/min generated by a superconducting coil is applied parallel to the current flow in the constriction. We emphasize that the V constrictions are retained without a break for a sufficiently long time to perform the measurements because of reduced thermal fluctuations at low temperatures. The differential conductance $dI/dV$ is measured as a function of the bias voltage using a lock-in technique with $f = 1$ kHz [11].

3. Results and Discussion

Figures 1(a), 1(b) and 1(c) show the MR in the V nanoconstrictions at zero-field resistances of $\sim 4.9 \Omega$, $\sim 13 \Omega$ and $\sim 31 \Omega$ at $T = 6$ K, where the diameter is estimated to be $d \sim 13$ nm, 8 nm and 5 nm from the Sharvin relation [12], respectively. In the MR measurements, the magnetic field decreases from +600 Oe to −600 Oe and then increases to +600 Oe. The MR effect appears when
Figure 1. (a) MR in the V constriction at a zero-field resistance of $\sim 4.9 \, \Omega$ and for the sweep field range of $|H| \leq 600 \, \text{Oe}$. The diameter is estimated be $d \sim 13 \, \text{nm}$ from the Sharvin relation [12]. (b) MR at a zero-field resistance of $\sim 13 \, \Omega$, where $d$ is estimated be $\sim 8 \, \text{nm}$. (c) MR at a zero-field resistance of $\sim 31 \, \Omega$, where $d$ is estimated be $\sim 5 \, \text{nm}$.

Figure 2. The conductance (left panel) and the differential conductance $dI/dV$ (right panel) in the final stage of the breaking, which are measured simultaneously. The three right panels show the evolution of $dI/dV$ for the regions I, II and III in the conductance. The dashed lines are fits by Eq.(1), where $T_K$ is given in the figure.

the resistance of the constriction is larger than $\sim 10 \, \Omega$ by stretching the V wire. The resistance at $\pm 600 \, \text{Oe}$ is almost the same, indicating that the MR effect is not related to the direction of the magnetic field. Note that MR is not observed in Al nanoconstrictions as shown in Fig. 1 of ref. 8. From these facts, we conclude that magnetic moments are induced by downsizing of V. In Ni and Pd nanoconstrictions, a large hysteresis is confirmed in the ferromagnetic region [8], while no clear hysteresis is seen in the present measurements. These results suggest that the
induced moments are paramagnetic and the origin of the MR is considered to be caused by the alignment of the induced moments by magnetic fields.

To further study the emergence of magnetic moments by downsizing, we measure the continuous evolution of the conductance and differential conductance $dI/dV$ spectra in the final stage of the breaking with stretching the wire at very low speed. The typical data below $G(0) < 5G_0$, where $G(0)$ is the zero-bias conductance, i.e., $dI/dV$ at $V = 0$, are shown in Figs. 2(a) and 2(b). Here, the curves are plotted in the atomic conductance unit $G_0 (= 2e^2/h)$. All the $dI/dV$ spectra represent a Fano-like resonance with a peak or dip shape anomaly at around zero bias, which changes its shape through the stretching of the constriction as shown in Fig. 2(b). From these features, we conclude that the origin of the Fano-like resonance is not caused by the presence of 200 ppm of Fe magnetic impurities in the V wires.

Referring to the previous results [13], we fit the $dI/dV$ spectra by the sum of a Fano resonance and a background component $g_0$ given by the following equation:

$$\frac{1}{G_0} \frac{dI}{dV} \propto g_0 + A \frac{(q + \epsilon)^2}{1 + q^2 + \epsilon^2}. \quad (1)$$

Here, $\epsilon = (eV - \epsilon_0)/k_BT_K$ is the bias shifted with respect to the center of the resonance $\epsilon_0$, and normalized by the natural width of the resonance, $k_BT_K$; $k_B$ is Boltzmann’s constant and $T_K$ is a fitting parameter; $q$ is the asymmetry parameter of the Fano theory and $A$ is the amplitude of the signal.

The $dI/dV$ spectra are well reproduced by the Fano resonance as shown in Fig. 2(b). It is well known that the Fano resonance appears due to the interference of scattering events between the continuum and discrete energy states. Hence, the present results imply the emergence of discrete energy state in the V nanoconstriction. The parameter $T_K$ is almost the same in the stretching process despite changing the contact shape and size, which strongly suggests that the Fano resonance does not come from structural origins such as dislocations induced by stretching. Recalling the results on MR effect, the discrete energy level is likely originating from the induced magnetic moments by downsizing. Therefore, it would be reasonable to consider that the induced moment is screened by the conduction electrons due to the Kondo effect, as in the case of the single magnetic adatom systems. Note that $T_K \sim 200$ K in the present experiments is in reasonable agreement with that in single magnetic adatoms on nonmagnetic metallic surfaces [13].

Next, we present the results in the superconducting temperature region. We show the size dependence of $dI/dV$ spectra in Pb and V nanoconstrictions in Figs. 3(a) and 3(b), respectively, where the diameter is varied from about 1 to 10 nm. The coherence lengths of Pb and V are 83 nm and 44 nm, respectively, implying that superconductor-normal-metal-superconductor (SNS) junctions are formed at the constrictions. In the Pb nanoconstriction, which is a reference to the V nanoconstriction, the clear anomalies due to the Andreev reflection are seen at $\Delta$ and $2\Delta$ together with the sharp peak caused by the Josephson current at around $V = 0$. It is seen that the energy gap is consistent with the energy gap for bulk Pb of $2\Delta = 2.3$ meV. On the other hand, the anomaly is only observed at $2\Delta$ in the V nanoconstrictions. The experimental value seems to be a little smaller than the gap for bulk V of $2\Delta = 1.4$ meV. In the diameter smaller than 3 nm, the anomalies due to the Andreev reflection disappear for both systems. Since the constriction is prepared by the MCBJ technique, the disorder and dislocations likely increase in the constrictions with decreasing the size. It seems that the Andreev reflection is suppressed due to appearance of a nonuniform structure in the smaller constrictions for both systems.

The difference of $dI/dV$ spectra between the two systems is also observed in the region of an atomic-size contact. The $dI/dV$ spectra below 10$G_0$ for Pb and V are shown in Figs. 4(a) and 4(b), respectively. The conductance of a one-atom sized contact is estimated to be at $G(0) \sim$
Figure 3. (a) The size dependence of $dI/dV$ spectra at $T = 1.5$ K for the bias voltage range of $|V| \leq 6$ meV in the Pb nanoconstrictions. Note that the superconducting transition temperature of bulk Pb is $T_C = 7.2$ K. The arrows correspond to $\Delta$ and $2\Delta$, where $\Delta$ is the superconducting energy gap of bulk Pb. (b) $dI/dV$ spectra at $T = 2$ K in the V nanoconstrictions. Note that $T_C$ of bulk V is 5.3 K. The arrows correspond to $2\Delta$ of bulk V.

Figure 4. (a) $dI/dV$ spectra for the atomic-size region of the Pb nanoconstriction below $10G_0$. (b) $dI/dV$ spectra for the atomic-size region of the V nanoconstriction.
2\(G_0\) for both systems from the measured histogram of the distribution of conductance plateaus in the breaking process, indicating that the contacts with conductance below \(2G_0\) are in the tunnel regime. Figure 4(a) shows that the broad peak due to the Josephson current changes to a dip-shape spectra in the tunnel regime for \(G(0) \leq 2G_0\) in Pb contact. The spectra can be fitted by a quasiparticle tunneling theory as shown by the red line in Fig. 4(a) with the energy gap \(2\Delta\) of 2.53 meV and the width \(\Gamma\) of 1.1 meV [14], which are consistent with those obtained in STM experiments. In contrast, for the V contact, the broad conductance peak at \(V = 0\) due to the superconducting anomaly survives down to the atomic-size of the contact without changing shape. Moreover, the dip-shape spectra is not seen in the tunnel regime, i.e., the conductance in the region of sub-atomic V contact size does not reveal quasi-particle tunneling of the superconducting state.

The properties of SNS junction in the V contacts are largely different from those in the Pb contacts, which are understood as typical features in SNS junction. However, we cannot determine whether the induced moments by downsizing affect the superconducting properties in the V contacts, because the influence of dislocations or strain in the contact cannot be excluded. To clarify this problem, further experiments are needed. For instance, since the temperature at 2 K is not sufficient to study the tunnel spectra in detail, we prepare the \(dI/dV\) measurements below \(T = 1\) K.

4. Conclusion
We have studied the transport properties of V nanoconstrictions with changing its size using the MCBJ technique. In the normal region for \(T > T_C\), MR is observed below the diameter \(d \sim 8\) nm. In constrictions of atomic size a Fano resonance appears around zero bias in the differential conductance which varies with constriction size. These results suggest the emergence of magnetic moments in the V nanoconstriction by downsizing. In the superconducting region for \(T < T_C\), we study the differential conductance spectra of the V nanoconstrictions by comparing with those of the Pb nanoconstrictions to investigate the induced magnetic impurity effect by downsizing in the superconducting tunnel junction. Only a single Andreev anomaly at \(2\Delta\) is observed in the conductance spectra of the V nanoconstrictions, while two anomalies at \(\Delta\) and \(2\Delta\) are seen in the Pb nanoconstrictions. In the tunnel regime of the V contacts, the structure of the quasiparticle tunneling density of states is not seen in the spectra. However for the V constrictions, there is no evidence for impurity effects by the induced moments in the superconducting tunnel spectra of the junctions.

References
[1] Tischer M, Hjortstam O, Arvanitis D, Hunter Dunn J, May F, Baberschke K, Trygg J, Wills J. M, Johansson B, and Eriksson O, 1995, \textit{Phys. Rev. Lett.} \textbf{75} 1602.
[2] Daalderop G. H. O, Kelly P. J, and Schuurmans M. F. H, 1994, \textit{Phys. Rev.} \textbf{B 50} 2874.
[3] Koide T, Miyachi H, Okamoto J, Shidara T, Fujimori A, Fukutani H, Amemiya K, Takeshita H, Yuasa S, Katayama T, and Yoda Y, 2001, \textit{Phys. Rev. Lett.} \textbf{87} 257201.
[4] Zhu M. J, Bylander D. M, and Kleinman L, 1990 \textit{Phys. Rev.} \textbf{B 42} 2874.
[5] Blugel S, 2005, \textit{Phys. Rev. Lett.} \textbf{68} 851.
[6] Pfandzelter R, Steierl G, and Rau C, 1995 \textit{Phys. Rev. Lett.} \textbf{74} 3467.
[7] Shinohara T, Sato T, and Taniyama T, 2003 \textit{Phys. Rev. Lett.} \textbf{91} 197201.
[8] Jenaga K, Nakashima N, Inagaki Y, Tsuji H, Kimura T, and Kawae T, 2012 \textit{Appl. Phys. Lett} \textbf{101} 123114.
[9] Akoh H, and Tasaki A., 1977, \textit{J. Phys. Soc. Jpn.} \textbf{42} 791.
[10] Liu F, Khanna S. N, and Jena P, 1991, \textit{Phys. Rev} \textbf{B 43} 8179.
[11] Jenaga K, Nakashima N, Inagaki Y, Tsuji H, Honda S, Kimura T, and Kawae T, 2012 \textit{Phys. Rev. B} \textbf{86} 064404.
[12] Sharvin Y. V, 1965, \textit{Sov. Phys. JETP} \textbf{21} 655.
[13] Ternes M, Heinrich A. J, and Schneider W-D, 2009 \textit{J. Phys.: Condens. Matter} \textbf{21} 053001.
[14] Dynes R. C, Narayanamurti V, and Garo J. P, 1978, \textit{Phys. Rev. Lett.} \textbf{41} 1509.