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Anomalous current–voltage characteristics of SFIFS Josephson junctions with weak ferromagnetic interlayers

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Abstract

We present a quantitative study of the current–voltage characteristics (CVC) of SFIFS Josephson junctions (S = bulk superconductor, F = metallic ferromagnet, I = insulating barrier) with weak ferromagnetic interlayers in the diffusive limit. The problem is solved in the framework of the nonlinear Usadel equations. We consider the case of a strong tunnel barrier such that the left SF and the right FS bilayers are decoupled. We calculate the density of states (DOS) in SF bilayers using a self-consistent numerical method. Then we obtain the CVC of corresponding SFIFS junctions, and discuss their properties for different set of parameters including the thicknesses of ferromagnetic layers, the exchange field, and the magnetic scattering time. We observe an anomalous nonmonotonic CVC in case of weak ferromagnetic interlayers, which we attribute to DOS energy dependencies in the case of small exchange fields.

Introduction

It is well known that superconductivity and ferromagnetism are two competing antagonistic orders. In superconductors (S) electrons form Cooper pairs with opposite spins and momenta, while in ferromagnetic metals (F) electron spins tend to align in parallel. Nevertheless, it is possible to combine S and F layers in one hybrid structure, which leads to the observation of many striking phenomena. The reason is the superconducting proximity effect, i.e., the superconducting correlations leakage into a
ferromagnetic metal due to Andreev reflection [1-7]. As a consequence, the real part of the pair wave function exhibits damped oscillatory behavior in a ferromagnetic metal. Hence, since the oscillations are spatially dependent, it is possible to realize a transition from “0” to “π” phase states in S/F/S structures upon changing the F layer thickness [1]. The proximity effect is characterized by the two length scales of decay and oscillations of the real part of the pair wave function in a ferromagnetic layer, \( \xi_0 \) and \( \xi_{D2} \), correspondingly [1]. If we consider the exchange field \( h \) as the only important parameter of a ferromagnetic material, both lengths are equal to \( \xi_h = \sqrt{D_f / h} \), where \( D_f \) is the diffusion constant in the ferromagnetic metal.

The existence of such phenomena enables the creation of so-called Josephson π junctions with a negative critical current [1,2]. Oscillations of the pair wave function in the F layer leads to several interesting phenomena in S/F/(S) systems, including nonmonotonic critical temperature dependence [8-12], Josephson critical current oscillations [13-41], and density of states (DOS) oscillations [42-45]. S/F hybrid structures have many promising applications in, e.g., single-flux quantum circuits [46,47], spintronic devices [48], memory elements [49-58] and spin-valves [59-65], magnetoelectronics [66-68], qubits [69], artificial neural networks [70], microrefrigerators [71,72], and low-temperature sensitive electron thermometers [73].

However, junctions with a ferromagnetic interlayer as well as other normal metal junctions (for example, SFNFS), proposed as elements of novel superconducting nanoelectronics, have limited applicability since such junctions have low resistance values [74,75]. This situation is resolved by addition of an insulating barrier (I) yielding a SFIFS layer sequence, which allows one to realize much larger values of the product \( J_c R_n \), where \( J_c \) is the critical current of the junction and \( R_n \) its normal state resistance [36-38]. Recently, SFIFS junctions attracted much attention and have been extensively studied both experimentally [32-41] and theoretically [23,45,76-80]. For instance, the current–voltage characteristics (CVC) of SFIFS Josephson junctions with a strong insulating layer were studied in [45]. They exhibit interesting nonmonotonic behavior for weak ferromagnetic interlayers, i.e., small enough exchange fields. The reason for this behavior is the shape of the density of states in the F layer. At small exchange fields the decay length of superconducting correlations in the ferromagnetic material, \( \xi_f \), is large enough, which leads to profound variations of the superconducting density of states in the F layer as a function of the energy and results in a corresponding CVC behavior. With an increase of the exchange field the \( \xi_h \) decreases, which suppresses the superconducting correlations in the F layer and makes the SFIFS CVC similar to the \( I-V \) curve of the FIS junction.

In this paper we study the current–voltage characteristics of SFIFS Josephson junctions with two ferromagnetic interlayers. SFIFS structures were also proposed for various applications in memory elements [56-58], single-flux quantum circuits [47], and as injectors in superconductor–ferromagnetic transistors [81-84], which can be used as amplifiers for memory, digital, and RF applications. In this work we study the current–voltage characteristics of a SFIFS junction as shown below in Figure 1. We present a quantitative model of the quasiparticle current in SFIFS junctions for different sets of parameters characterizing the ferromagnetic interlayers. In case of weak ferromagnetic metals we find an anomalous nonmonotonic shape of the current–voltage characteristics at subgap voltages and compare the results with the CVC of SFIFS junctions [45]. We ascribe this behavior to DOS energy dependencies in case of small exchange fields in the F layers. The shape is smeared if we include a finite magnetic scattering rate. The anomalous nonmonotonic shape of the current–voltage characteristics of SFIFS junctions with weak ferromagnetic layers looks similar to the fine structures of quasiparticle currents, recently obtained experimentally on similar systems [82-85].

The paper is organized as follows. In the first section (“Model”) we formulate the theoretical model and basic equations and introduce the self-consistent numerical iterative method for calculating the density of states in S/F bilayers. In the next section (“Results and Discussion”) we present and discuss the results for the density of states in S/F bilayers in case of subgap values of the exchange field and the current–voltage characteristics of SFIFS junctions. Finally we summarize the results in the last section (“Conclusion”).

Model

In this section we present the theoretical model we use in our studies. The geometry of the considered system is depicted in Figure 1. It consists of two superconducting electrodes and a

![Figure 1: Schematic representation of the SFIFS hybrid structure](image)
pair of ferromagnetic interlayers, with thicknesses $d_{1}$ and $d_{2}$. The system contains three interfaces: two S/F (superconductor/ferromagnet) boundaries and one tunnel F-I-F interface. Each of these interfaces is described by the dimensionless parameter $\gamma_{n} = R_{n} \sigma_{n}/\xi_{n}$ ($n = 0, 1, 2$), which is proportional to the resistance $R_{n}$ across the interface [86-88]. Here $\sigma_{n}$ is the conductivity of the $n$th layer and $\xi_{n} = \sqrt{\tau_{n}}/2\pi T_{c}$ is the coherence length, where $T_{c}$ is the critical temperature of the superconductor $S$ (here and below we assume $\hbar = k_{B} = 1$). In this paper we consider the diffusive limit, when the elastic scattering length $\ell$ is much smaller than the characteristic decay length of the real part of the pair wave function in the ferromagnet, $\xi_{n}$, which we introduce later in Equation 13 and Equation 14. We assume that the S/F interfaces are not magnetically active. We also neglect the nonequilibrium effects [89-91] and use the Matsubara Green’s functions technique, which has been developed to describe many-body systems in equilibrium at finite temperature [92].

In our model the tunneling barrier is located between two F layers at $x = 0$ (Figure 1), whereas the other interfaces at $x = -d_{1}$ and $x = d_{2}$ are identical and transparent. This case corresponds to $\gamma_{n1} = \gamma_{n2} \ll 1$ and $\gamma_{0} \gg 1$. In case of a sufficiently strong tunnel barrier ($\gamma_{0} \gg 1$), the two S/F bilayers in the SFIFS junction are decoupled, i.e., the amplitudes of two-electron processes between left and right F layers are negligibly small. Hence, the quasiparticle current through the SFIFS junction, biased by the voltage $eV$, can be calculated by using the Werthamer formula [93],

$$I = \frac{1}{eR} \int_{-\infty}^{\infty} dE N_{11}(E - eV) N_{22}(E) \left[ f(E - eV) - f(E) \right],$$

where $N_{11,2}(E)$ are the densities of states (DOS) in the corresponding ferromagnetic layer at $x = 0$, $f(E) = [1 + e^{E/T_{c}}]^{-1}$ is the Fermi–Dirac distribution function, and $R = R_{0}$ is the resistance across the F-I-F interface. Both densities of states $N_{11,2}(E)$ are normalized to their values in the normal state.

In order to obtain the densities of states in ferromagnetic layers, $N_{11,2}(E)$, we use a self-consistent two-step iterative procedure, described below. As far as $\gamma_{0} \gg 1$, we can neglect the influence of the right F layer on the density of states in the left S/F bilayer and vice versa (see Figure 1). Thus we need to obtain the DOS at the outer border of each S/F bilayer. That can be done by solving the Usadel equations in the S/F bilayer system [94].

In the following, we use the $\theta$-parameterizations of normal ($G = \cos \theta$) and anomalous ($F = \sin \theta$) Green’s functions and write the Usadel equations in the F layers in the form [94,95],

$$\frac{D_{F}}{2} \frac{\partial^{2} \theta_{F}^{\pm}(\xi)}{\partial \xi^{2}} = \left( \frac{\omega \pm i\hbar}{\tau_{F}} + \frac{\cos \theta_{F}^{\pm}(\xi)}{\tau_{m}} \right) \sin \theta_{F}^{\pm}(\xi) + \frac{1}{\tau_{m}} \sin \left( \theta_{F}^{-} + \theta_{F}^{+} \right) \pm \frac{1}{\tau_{so}} \sin \left( \theta_{F}^{-} - \theta_{F}^{+} \right),$$

where the positive and negative signs correspond to the spin-up ("↑") and spin-down ("↓") states, respectively. In terms of the electron fermionic operators $\psi_{↑,↓}$ the spin-up state corresponds to the anomalous Green’s function $F_{↑} \sim \langle \psi_{↑} \psi_{↓} \rangle$, while spin-down state corresponds to $F_{↓} \sim \langle \psi_{↑} \psi_{↓} \rangle$. The expressions $\omega = 2\pi T(n + 1/2)$ are the Matsubara frequencies, where $n = 0, \pm 1, \pm 2, \ldots$, and $\hbar$ is the exchange field in the ferromagnet. The scattering times are labeled here as $\tau_{F}$, $\tau_{m}$, and $\tau_{so}$, where $\tau_{so}$ corresponds to the magnetic scattering parallel (perpendicular) to the quantization axis, and $\tau_{so}$ is the spin–orbit scattering time [96-99].

Assuming a strong uniaxial anisotropy in ferromagnetic materials, in which case there is no coupling between spin-up and spin-down electron populations, we neglect $\tau_{m}$ ($\tau_{m}^{-1} \approx 0$). We also assume the ferromagnets to have a weak spin–orbit coupling and thus neglect the spin–orbit scattering time $\tau_{so}$. After taking into account all the assumptions, the Usadel equations in the ferromagnetic layers for different spin states can be written as

$$\frac{D_{F}}{2} \frac{\partial^{2} \theta_{F}^{\pm}(\xi)}{\partial \xi^{2}} = \left( \frac{\omega \pm i\hbar}{\tau_{F}} + \frac{\cos \theta_{F}^{\pm}(\xi)}{\tau_{m}} \right) \sin \theta_{F}^{\pm}(\xi),$$

where $\tau_{m} = \tau_{F}$ is the magnetic scattering time. In the superconducting layer $S$ the Usadel equation reads [94]

$$\frac{D_{S}}{2} \frac{\partial^{2} \theta_{S}^{\pm}(\xi)}{\partial \xi^{2}} = \omega \sin \theta_{S} - \Delta(\xi) \cos \theta_{S},$$

where $D_{S}$ is the diffusion coefficient in the S layer and $\Delta(\xi)$ is the pair potential in the superconductor. We note that $\Delta(\xi)$ vanishes in the F layer.

Equation 3 and Equation 4 must be supplemented with corresponding boundary conditions. At the S/F interfaces we apply the Kupriyanov–Lukichev boundary conditions. For example, at the left S/F interface they are written as [86],

$$\xi_{F} = \frac{\partial \theta_{F}^{\pm}}{\partial \xi} \big|_{x = d_{1}} = \xi_{S} \frac{\partial \theta_{S}^{\pm}}{\partial \xi} \big|_{x = d_{1}},$$
Similar equations can be written at the right S/F interface at \( x = d_f^2 \). Here \( \gamma = \frac{\xi_s}{\sigma_n} \), where \( \sigma_n \) is the conductivity of the S layer and \( \xi_s = \sqrt{D_s / 2\pi T_c} \) is the superconducting coherence length. The parameter \( \gamma \) defines the strength of the inverse proximity effect, i.e., the suppression of superconductivity in the adjacent S layer by the ferromagnetic layer F. We consider the parameter \( \gamma \) to be relatively small \( \gamma \ll 1 \), which corresponds to a rather weak suppression.

To calculate the density of states in the S/F bilayer we should set the boundary conditions at the outer boundary of the ferromagnet \( (x = 0) \),

\[
\left( \frac{\partial \theta_j}{\partial x} \right)_0 = 0.
\]  

(7)

To complete the boundary problem we also set a boundary condition at \( x = \pm \infty \),

\[
\theta_j(\pm \infty) = \arctan \left( \frac{\Delta}{\omega} \right),
\]  

where the Green’s functions acquire the well-known bulk BCS form. We notice that the density of states at \( x = \pm \infty \) is given by standard BCS equation,

\[
N_j(E) = \text{Re} \left[ \cos \theta_j \left( i\omega \rightarrow E + i0 \right) \right] = \frac{|E| \Theta(|E| - \Delta)}{\sqrt{E^2 - \Delta^2}},
\]  

(9)

where \( \Theta(x) \) is the Heaviside step function.

Finally the self-consistency equation for the superconducting order parameter takes the form,

\[
\xi_n \gamma B_1 \left( \frac{\partial \theta_j}{\partial x} \right)_{d_f^1} = \sin \left( \theta_j - \theta_j^f \right),
\]  

(6)

\[
N_{j\uparrow}(E) = \frac{N_{j\uparrow}(E) + N_{j\downarrow}(E)}{2}, \quad j = 1, 2,
\]  

(11)

where \( N_{j\uparrow}(E) \) and \( N_{j\downarrow}(E) \) are the spin-resolved densities of states written in terms of the spectral angle \( \theta \).

\[
N_{j\uparrow\downarrow}(E) = \text{Re} \left[ \cos \theta_j \gamma \left( i\omega \rightarrow E - i0 \right) \right], \quad j = 1, 2.
\]  

(12)

To obtain \( N_{1,2} \), we use a self-consistent two-step iterative procedure [95,100-102]. In the first step we calculate the pair potential coordinate dependence \( \Delta(x) \) using the self-consistency equation in the S layer (Equation 10). Then, by proceeding to the analytical continuation in Equation 3 and Equation 4 over the quasiparticle energy \( i\omega \rightarrow E + i0 \) and using the \( \Delta(x) \) dependence obtained in the previous step, we find the Green’s functions by repeating the iterations until convergence is reached.

The characteristic lengths of the decay and oscillations of the real part of the pair wave function in the ferromagnetic layer at the Fermi energy, \( \xi_{1,2} \), are given in our model by [45],

\[
\frac{1}{\xi_{1}} = \frac{1}{D_f} \sqrt{\frac{1}{\tau_m} + \frac{1}{\tau_m}}
\]  

(13)

\[
\frac{1}{\xi_{2}} = \frac{1}{D_f} \sqrt{\frac{1}{\tau_m} + \frac{1}{\tau_m} - \frac{1}{\tau_m}}
\]  

(14)

We see from these equations that with an increase of the magnetic scattering rate \( \alpha_m = 1/\tau_m \Delta \) the length of decay \( \xi_{1} \) decreases, while the length of oscillations \( \xi_{2} \) increases. In the absence of magnetic scattering \( \xi_{f1} = \xi_{f2} = \xi_0 = \sqrt{D_f / \hbar} \).

Results and Discussion

In this section we present the results of the DOS energy dependencies in SF bilayers at the free boundary of the F layer for \( h \leq \Delta \). The densities of states for \( h \geq \Delta \) were thoroughly discussed in [45]. Then we calculate the corresponding CVC of the SFIFS junction using the Werthamer formula (Equation 1). In the case of \( h \leq \Delta \) we obtain an interesting nonmonotonic behavior of the quasiparticle current, presented in a subsection below (“Current–voltage characteristics of SFIFS junctions”). At large exchange fields the decay length \( \xi_{1} \) of the real part of the pair wave function in the F layer becomes small (see Equation 13 and Equation 14), and the amplitude of DOS variations tends to zero. In this case the CVC of SFIFS junction tends to follow Ohm’s law for \( h \gg \Delta \). The ferromagnetic materials with small
exchange fields can be fabricated as discussed in [103]. We also note that the DOS at the end of an SF bilayer in case of a domain wall in the ferromagnetic layer was studied in [104].

**Density of states in SF bilayers for** \( h \leq \Delta \)

Figure 2 and Figure 3 show the DOS energy dependencies for different values of \( h \leq \Delta \) and for relatively thick F layers. In our calculations we fix the temperature at \( T = 0.1T_c \), where \( T_c \) is the critical temperature of the superconductor S. In Figure 2 the characteristic “finger-like” shape of DOS is observed along with a minigap for \( d_f = 2\xi_f \) (Figure 2a,c). At larger \( d_f \) and/or at larger \( h \) the minigap closes (Figure 2c and Figure 3a,c). In the absence of magnetic scattering (\( \alpha_m = 1/\tau_m\Delta = 0 \)) we can roughly estimate the critical value \( h_c \) of the exchange field at which the minigap closes as [45]

\[
    h_c \sim E_{1\text{Th}}, \quad E_{1\text{Th}} = \frac{D_f}{d_f^2}, \quad (15)
\]

where \( E_{1\text{Th}} \) is the Thouless energy and \( d_f \) is the thickness of the F layer in the SF bilayer (\( d_{f1} \) or \( d_{f2} \) for the left or right SF bilayer, respectively, in Figure 1). Since we consider subgap values of \( h \), the minigap closes at rather large values of \( d_f \) in the absence of magnetic scattering.

After the minigap closes the DOS at the Fermi energy \( N_f(0) \) rapidly increases to values larger than unity with further increase of \( d_f \) and then it oscillates around unity while its absolute value exponentially approaches unity [45]. This is the well-known damped oscillatory behavior with the lengths of decay and oscillations given by Equation 13 and Equation 14, respectively. Figure 2b,d and Figure 3b,d show that stronger magnetic scattering leads to the minigap closing at smaller values of \( d_f \). With the increase of \( \alpha_m = 1/\tau_m\Delta \) the period of oscillations increases (\( \xi_{f1} \) in Equation 14 increases). At the same time the DOS variation amplitude becomes smaller and DOS features smear, since for larger \( \alpha_m \) the dumped exponential decay of oscillations occurs faster (\( \xi_{f1} \) in Equation 13 decreases).

Finally, in Figure 4 we present plots for spin-resolved densities of states given by Equation 12 for both zero and finite magnetic scattering.

**Current–voltage characteristics of SFIFS junctions**

Using the densities of states \( N_{1,2}(E) \) obtained in the subsection above, we calculate a set of quasiparticle current curves using Equation 1 for various values of parameters describing properties of ferromagnetic material, which include the thicknesses of the F layers, \( d_{f1} \) and \( d_{f2} \), the exchange field \( h \), and the magnetic scattering rate \( \alpha_m \). In our calculations we fix the temperature at \( T = 0.1T_c \), where \( T_c \) is the critical temperature of the superconducting lead.

Figure 5 shows the CVC of a symmetric SFIFS junction, where \( d_{f1} = d_{f2} = d_f \) in the absence of magnetic scattering. For thin
enough ferromagnetic interlayers, $d_f/\xi_n = 0.5$, and a small enough value of the exchange field, $h = 0.5\Delta$, we observe CVC that resemble the $I$-$V$ characteristic of a SNINS Josephson junction with a characteristic peak at $eV \approx 2\Delta$ (see Figure 5a, solid black line) [101]. With an increase of the exchange field $h$ this peak becomes smeared (see Figure 5b–d, solid black line). Increasing $d_f$ and/or $h$ produces a set of $I$-$V$ curves among which the red dashed line in Figure 5d is the most interesting because it exhibits a nonmonotonic behavior. The reason of this atypical nonmonotonic behavior will be explained later.

Figure 6 shows the current–voltage characteristics of SFIFS junctions at subgap values of the exchange field. We observe a nonmonotonic behavior for thick enough ferromagnetic layers at $h \leq \Delta$. Let us consider the CVC in Figure 6b, red dashed line. We can explain its behavior as well as any other nonmonotonic CVC behavior as the signature of the DOS energy dependence. The anomalous nonmonotonic $I(V)$ dependence arises from the shape features of the densities of states, see Figure 7. In symmetric SFIFS junctions, $N_{f1}(E) = N_{f2}(E) \equiv N_f(E)$ in Equation 1, which can be well approximated by taking $T = 0$ for small temperatures $T \ll T_c$. In this case the Fermi–Dirac distribution function $f(E)$ can be represented with the Heaviside step function $\Theta(-E)$ [and $f(E - eV)$ with $\Theta(eV - E)$]. As a result, the limits of integration in Equation 1 shrink to the interval $[0, eV]$. 

![Figure 4](image-url)

Figure 4: Spin-resolved DOS $N_{f\uparrow}(E)$ on the free boundary of the F layer in the FS bilayer obtained numerically in the absence of magnetic scattering, $\alpha_m = 0$ (plots a and c) and in the case of finite magnetic scattering, $\alpha_m = 0.5$. Plots a, b: $h = 0.5\Delta$, $d_f = 2\xi_n$; plots c, d: $h = 0.3\Delta$, $d_f = 3\xi_n$ (c) and $d_f = 2\xi_n$ (d). The black solid line corresponds to $N_f(E)$, the red dashed line corresponds to $N_{f\uparrow}(E)$, and the blue dash-dotted line corresponds to $N_{f\downarrow}(E)$.

![Figure 5](image-url)

Figure 5: Current–voltage characteristics of the symmetric ($d_{f1} = d_{f2} = d_f$) SFIFS junction in the absence of magnetic scattering for different values of exchange field $h$. The temperature $T = 0.1T_c$. In each graph the curves were calculated for different values of F layer thickness $d_f$, $d_f = 0.5\xi_n$ (black solid line), $d_f = 1.0\xi_n$ (red dashed line), $d_f = 1.5\xi_n$ (blue dash-dotted line). The plots correspond to specific values of the exchange field $h$: plot (a) to $h = 0.5\Delta$, (b) to $h = 1.0\Delta$, (c) to $h = 2.0\Delta$ and (d) to $h = 3.0\Delta$.

![Figure 6](image-url)

Figure 6: Current–voltage characteristics of a symmetric SFIFS junction for different values of the subgap exchange field $h$ in the absence of magnetic scattering at a temperature $T = 0.1T_c$. In each graph the curves were calculated for different values of F layer thickness, $d_f$, $d_f = 2\xi_n$ (black solid line) and $d_f = 3\xi_n$ (red dashed line). The plots correspond to specific values of the subgap exchange field $h$: plot (a) to $h = 0.1\Delta$, (b) to $h = 0.3\Delta$, (c) to $h = 0.5\Delta$ and (d) to $h = 0.7\Delta$. 

![Image](image-url)
Hence, the current through the junction can be written as,

$$ I = \frac{1}{eR} \int_0^E dE \ N_f(E - eV) N_f(E). $$

Using this expression, the origin of the nonmonotonic behavior of the CVC can be explained. At $eV = 0$ the upper limit of the integral in Equation 16 is zero and the current is zero. With the increase of the voltage, the current first increases linearly due to the broader region of integration as in Ohm’s law. The first feature that is shown in Figure 7a is a significant change in the slope of the current. Figure 7b shows the relative positions of the densities of states $N_f(E - eV)$ and $N_f(E)$ in this case, where almost no peak overlap can be seen, resulting in relatively small values of the integral in Equation 16. As we proceed to larger values of $eV$, we reach the first local maximum of the CVC, which corresponds to a maximum overlap of the densities of states $N_f(E - eV)$ and $N_f(E)$ at $eV/\Delta \approx 1$ (see Figure 7c). The second maximum of the quasiparticle current occurs at $eV/\Delta \approx 1.68$, which corresponds to a perfect DOS peak overlap at $E/\Delta \approx 1$ (Figure 7d). For large enough values of the voltage $eV$, a product of the DOS $N_f(E - eV)$ $N_f(E)$ $\approx 1$ and its integration does not produce any features. Thus, the CVC eventually coincides with Ohm’s law in this case. In fact any shape of a SFIFS $I$–$V$ curve can be explained and understood in this way. We note that in this paper we present the densities of states in SF bilayers only for subgap values of the exchange field. For $h \geq \Delta$ the DOS energy dependencies in SF bilayers can be found in [45].

Based on the properties of the density of states in FS bilayers we can see that even the tiny exchange field $h$ can dramatically modify the current introducing anomalous nonmonotonic behavior in case of thick enough F layers (see Figure 5 and Figure 6). It is important to understand how the CVC of a SFIFS junction transforms as the exchange field $h$ increases. In Figure 8 we demonstrate the plot of current–voltage characteristics calculated for a wide range of exchange field values $h$ in the absence of magnetic scattering. From this plot it can be clearly seen that while for relatively small (subgap) values of the exchange field many interesting features appear in the structure of the current, at larger values of $h$ these features are smeared and the CVC approaches Ohm’s law. Figure 9 shows the current–voltage characteristics in the case of an asymmetric SFIFS junction, i.e., when $d_{f1} \neq d_{f2}$ in the case of zero magnetic scattering.

In this section we also present the current–voltage characteristics of a SFIFS junction in the presence of magnetic scattering for $d_{1} = 3\xi_{n}$. The temperature is $T = 0.1T_{c}$. The curves correspond to different values of $h$, from $h = 0$ to $h = 1.2\Delta$ with increments equal to $0.1\Delta$. The exchange field $h = 0$ corresponds to the case of a SNINS junction [101].
metric SFIFS junctions. The insets show the CVC in case of zero magnetic scattering. For very small \( h \) nonzero magnetic scattering leads to smearing of characteristic features of the current as shown in Figure 10. At larger subgap values of the exchange field \( h \) we see a “triple kink” structure (Figure 10c). For large enough values of \( \alpha_m \) the nonmonotonic behavior of the quasiparticle current will be smeared and the current approaches Ohm’s law. This is due to the fact that increasing \( \alpha_m \) the length of the superconducting correlations decay in the ferromagnetic layers decreases, see Equation 13, and the suppression of superconducting correlations in the F layers occurs faster.

We can compare these results with the \( I-V \) characteristics of SIFS Josephson junctions [45]. In this case at zero magnetic scattering we may also observe the nonmonotonic behavior, but with only one peak [see Ref. [45], Figure 6 (c)]. In case of finite magnetic scattering the CVC has a “double kink” structure [see Ref. [45], Figure 7 (a, c)]. In SIFS junctions the overlap of subgap DOS structures \( N_f(E - eV) N_g(E) \) in the integrand of the current equation, Equation 16, produce more complex behavior of the \( I-V \) characteristics.

We also notice that in recent experiments on SFIFS junctions as injectors of superconductor-ferromagnetic transistors some fine structures of the subgap quasiparticle current was observed [82-85], which looks similar to our theoretical results.

**Conclusion**

In this work we have presented the results of CVC calculations of a SFIFS junction for different set of parameters including the thicknesses of the ferromagnetic layers, \( d_{\perp 1} \) and \( d_{\perp 2} \), the exchange field, and the magnetic scattering time \( \alpha_m = 1/\tau_m \Delta \).

We considered the case of a strong insulating barrier such that the left SF and the right FS bilayers are decoupled. In order to obtain the current–voltage characteristics we first calculated the densities of states on the free boundary of the F layer in each SF bilayer utilizing an iterative self-consistent approach. Using the numerically obtained DOS we have derived the quasiparticle current of a SFIFS junction in the case of symmetric \( d_{\perp 1} = d_{\perp 2} \) and asymmetric \( d_{\perp 1} \neq d_{\perp 2} \) structures. We have paid much attention to the case of a SFIFS junction with weak ferromagnetic interlayers with exchange fields \( h \leq \Delta \). It was demonstrated that the CVC exhibits interesting and unusual features in this case, which can be ascribed to typical DOS behavior. We have provided a simple physical explanation for such anomalous CVC behavior. We have also illustrated how the CVC shape evolves as one increases the exchange field \( h \). It should be emphasized that taking into account finite magnetic scattering leads to the smearing of characteristic features and, in particular cases, to a “triple kink” shape of the current.
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High dynamic resistance elements based on a Josephson junction array

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Full Research Paper

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Abstract

A chain of superconductor–insulator–superconductor junctions based on Al–AlOx–Al nanostructures and fabricated using conventional lift-off lithography techniques was measured at ultra-low temperatures. At zero magnetic field, the low current bias dynamic resistance can reach values of \( \approx 10^{11} \Omega \). It was demonstrated that the system can provide a decent quality current biasing circuit, enabling the observation of Coulomb blockade and Bloch oscillations in ultra-narrow Ti nanowires associated with the quantum phase-slip effect.

Introduction

The field of modern nanoelectronics is facing stagnation with respect to further miniaturization, deviating from Moore’s law [1]. Typically, two main reason are quoted: severe heat dissipation per unit volume (surface), and various quantum phenomena that drive the operation of ultra-small devices and make them different from devices in the conventional (classical) regime. The radical solution to the first problem is to build the critical elements using superconductors. The basics of this approach were developed in the late 1980s, resulting in rapid single flux quantum (RSFQ) logic [2]. Since that time the concept has continued to develop. However, the corresponding systems so far have not developed into mass market commercial products, being limited solely to particular “cost-no-object” applications. Currently, the field of superconducting electronics is developing much faster mainly due to the understanding that (even taking into consideration the necessity of refrigeration) the energy consumption of next generation supercomputers can be as low as \( \approx 10 \) MW, which is compared to values of \( \approx 100 \) MW for conventional semiconductor complementary metal–oxide–semiconductor (CMOS) technology. In addition to heat dissipation, another issue is the speed of processing. It has been shown that the operational frequency of superconducting logic can be at least 100 times higher than for CMOS-based devices. It is universally accepted
that the limiting factor for the speed of operation of various superconducting devices is the high-frequency impedance, e.g., originating from kinetic inductance. The effect should be taken into consideration for various cryoelectronic applications.

In addition to RSFQ computers which exploit classical 2-bit logic, during the last decades, there has been an increasing interest in quantum computing utilizing nonclassical approaches. There have been multiple suggestions regarding how to build quantum logic elements, such as quantum bits (qubits), including superconducting systems based on the Josephson effect. It has been shown that physics behind a Josephson junction (JJ) is dual to a quantum phase-slip junction (QPSJ) [3], whereby the corresponding QPSJ-based qbit operation has also been demonstrated [4]. At the same time, the quantum dynamics of a JJ (or a QPSJ) is strongly determined by the environment [5,6]. In particular, the utilization of devices based on quantum fluctuations of the macroscopic phase, \( \phi \), requires stabilization of the quantum conjugated quantity – charge \( q \). The most straightforward approach is to use high-Ohmic on-chip current-biasing elements [7-10]. However, it was later understood that resistive dissipative elements inevitably act as a source of Johnson noise, leading to degradation of system performance [11].

Here we present an experimental study of a quasi-1D chain of JJs. A sufficient high-frequency impedance was demonstrated to study the QPS phenomena without the undesired impact of Johnson noise typically associated with dissipative elements [12]. The \( I-V \) dependence studied in [12] demonstrated clear and expected characteristics at low current, \( I \rightarrow 0 \): the so-called “Bloch nose” (back-bending of \( I-V \)), while at finite current values, the corresponding singularities were not so pronounced. The purpose of this paper is to provide an in-depth analysis of the \( I-V \) dependence of the same JJ chains used in the current-biasing elements in [12].

**Experimental**

Conventional lift-off electron-beam lithography followed by ultrahigh vacuum deposition of materials was used for the fabrication of the nanostructures. Hybrid QPSJ samples were made of Ti, Al and aluminum oxide [12]. The high-impedance JJs studied in this paper, similar to those from [12], were fabricated from superconducting thin film Al oxidized in situ to form tunnel barriers. Each sample consisted of 25 pairs of JJs connected in parallel where the area of each superconductor–insulator–superconductor (SIS) contact was about 100 \( \times \) 100 nm (Figure 1). The samples were analyzed by scanning electron microscopy (SEM) (Figure 1) and atomic force microscopy (AFM).

Transport measurements were made inside a \(^3\)He/\(^4\)He dilution refrigerator at temperatures below 400 mK, corresponding to the superconducting transition of Ti QPSJs [10,12]. All input/output lines were carefully filtered [13] to reduce the impact of the noisy electromagnetic environment. When necessary, a small magnetic field, up to 0.05 T, was applied using small superconducting coils wound directly on the sample holder cap.

**Results and Discussion**

The ultimate goal of this work is to study the quantum dynamics of the QPSJ, a system dual to JJ [3], including the observation of Coulomb blockade and Bloch oscillations [14]. Given that the macroscopic phase, \( \phi \), and the charge, \( q \), are quantum conjugated values, \( [\phi, q] = i\hbar \), in order to enable the high rate of phase fluctuations, one should define the charge. The phase–charge duality in conventional JJ systems is well established [15-17]. Hence, to enable the phase fluctuation regime, the electric current, \( I \), through a QPSJ, which is just the time derivative of charge, \( I = dq/dt \), should be stabilized. The focus of this manuscript is to study the transport properties of JJ chains to be used as current-biasing elements of a QPSJ. Note that here the finite electric current is maintained by correlated Cooper pair tunneling at a voltage bias \( V \) across the QPSJ exceeding the particular Coulomb blockade threshold, \( V_C \) [14]. The tunneling happens at the Bloch oscillation rate, \( f_B \). The synchronization of this “internal” periodic process with the external drive, \( f_{RF} \), should result in quantized singularities (Bloch steps) at current values \( I(n) = n(2e/\hbar f_{RF}) \), where \( 2e \) is the charge of the Cooper pair and \( n = 1,2,3... \) are integers. Furthermore, the study of QPSJ \( I-V \) characteristics demonstrating Coulomb blockade at zero current, \( I = 0 \), and voltages of \( V < V_C \) requires only the high-Ohmic environment with resistance \( R_{env} \) exceeding the quantum value \( R_Q = h/e^2 \approx 26 \text{ k}\Omega \). While at finite current values, \( I > 0 \), one needs current stabilization at high frequency \( f_{RF} \) which further requires high values of the high-frequency impedance, \( Z_{env}(f_{RF}) \). The observation of a pronounced
Coulomb blockade has been observed in JJs using both a high-resistive dissipative environment [7,8] and nonlinear Josephson elements with high dynamic resistance and/or kinetic inductance [6,18]. However, extended attempts to observe Bloch oscillation phenomena at finite currents in JJs provided rather modest results [7,8,19]. The recent progress in understanding the QPS phenomena [20] in ultra-narrow superconducting channels has revived interest in this topic, resulting in the observation of a decent Coulomb blockade [9,10], while quite blurred Bloch steps at finite current values have been detected so far [10]. Later it was understood that the straightforward approach of using a high-Ohmic dissipative environment, \( R_{\text{env}} > R_Q \), is far from optimal, as it introduces Johnson noise, washing out the desired current singularities [11]. Various JJ-based systems were suggested which take advantage of the high kinetic inductance of superconducting quantum interference devices (SQUIDs) [21,22] (\( J_k = \cos^{-1}(\Phi/\Phi_0) \)) at a degeneracy point when \( \Phi/\Phi_0 \to \pi/2 \), where \( \Phi \) is the magnetic flux through the SQUID area and \( \Phi_0 \) is the magnetic flux quantum, \( \Phi_0 = h/2e = 2 \times 10^{-15} \text{ Wb} \). Hence the SQUID-based approach requires application of a finite magnetic field. Given that the electromagnetic horizon of our QPSJ is of the order of \( \approx 100 \mu \text{m} \) [23-25], the corresponding high-impedance current biasing circuit should be of appropriate (small) dimensions. Thus the area of the SQUID is small, and hence a magnetic field corresponding to \( \Phi/\Phi_0 \to \pi/2 \) can easily reach the \( \approx 10 \text{ mT} \) range. At such a magnetic field, two undesirable effects might happen both with the biasing superconducting leads and with the QPSJ. Namely, the formation of Abrikosov vortices and a noticeable suppression of the energy gap. Consequently, in our approach, we opted for a non-dissipative (superconducting) high-impedance environment under zero magnetic field.

Our quasi-one-dimensional arrays of SIS junctions contain loops forming SQUIDs (Figure 1). The Josephson current is very small (Figure 2a), \( I_c < 10 \text{ pA} \), and application of the magnetic field only monotonically suppresses the superconducting gap. The corresponding \( I-V \) dependence can be understood as a tunnel characteristic of multiple SIS junctions connected in series. The \( I-V \) characteristics (Figure 2a) with a gap of \( \approx 10 \text{ mV} \) corresponds well with 25 SIS junctions connected in series, each being a Al–AlO\(_x\)–Al junction with a gap of about 400 \( \mu \text{V} \). The charging energy, \( E_c = e^2/2C \), of each SIS contact (considering it to be a plate capacitor with dielectric constant \( \varepsilon = 10 \), area \( 100 \times 100 \text{ nm} \) and distance between plates \( \approx 2 \text{ nm} \)) is about two orders of magnitude higher than the Josephson energy, \( E_J = I_c/2e \). As \( E_J < < E_c \), the physics of the system is dominated solely by charging phenomena. At zero magnetic field and small current bias, the dynamic resistance \( R_{\text{dy}} = dV/dI \) of the JJ chain can reach \( \approx 10^{11} \Omega \) (Figure 2b), while at a higher bias, \( R_{\text{dy}} (I \gg 0) \) approaches 100 k\( \Omega \).

The SIS junction chain has been used to current bias narrow Ti nanowires [12], with cross sections demonstrating various phenomena attributed to the QPS effect [10,26-33]. The observation of Coulomb blockade and Bloch steps [12] confirms the usefulness of the suggested concept, that is, the utilization of SIS junction chains.

Summarizing, we can conclude that chains of series-connected tunnel SIS junctions can provide high dynamic resistance at low current, which is necessary to stabilize the charge of a quantum circuit and hence enable the high level of phase fluctuations. The absence of dissipation makes such elements very useful for experimental studies of mesoscopic scale objects at ultralow temperature applications, where even a very small amount of Johnson noise may overheat electrons above the phonon bath. However, the non-linearity of a SIS junction \( I-V \) characteristic makes them less useful at finite current biases, which dramatically reduces the dynamic resistance. A promising solution might be the utilization of superconducting circuits with a high...
level of kinetic inductance capable to provide a sufficient impedance at high frequencies.

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Epitaxial growth and superconducting properties of thin-film PdFe/VN and VN/PdFe bilayers on MgO(001) substrates

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Full Research Paper

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Abstract

Single-layer vanadium nitride (VN) and bilayer Pd₀.₉₆Fe₀.₀₄/VN and VN/Pd₀.₉₂Fe₀.₀₈ thin-film heterostructures for possible spintronics applications were synthesized on (001)-oriented single-crystalline magnesium oxide (MgO) substrates utilizing a four-chamber ultrahigh vacuum deposition and analysis system. The VN layers were reactively magnetron sputtered from a metallic vanadium target in Ar/N₂ plasma, while the Pd₁₋ₓFeₓ layers were deposited by co-evaporation of metallic Pd and Fe pellets from calibrated effusion cells in a molecular beam epitaxy chamber. The VN stoichiometry and Pd₁₋ₓFeₓ composition were controlled by X-ray photoelectron spectroscopy. In situ low-energy electron diffraction and ex situ X-ray diffraction show that the 30 nm thick single-layer VN as well as the double-layer VN(30 nm)/Pd₀.₉₂Fe₀.₀₈(12 nm) and Pd₀.₉₂Fe₀.₀₄(20 nm)/VN(30 nm) structures have grown cube-on-cube epitaxially. Electric resistance measurements demonstrate a metallic-type temperature dependence for the VN film with a small residual resistivity of 9 μΩ·cm at 10 K, indicating high purity and structural quality of the film. The transition to the superconducting state was observed at 7.7 K for the VN film, at 7.2 K for the Pd₀.₉₆Fe₀.₀₄/VN structure and at 6.1 K for the VN/Pd₀.₉₂Fe₀.₀₈ structure with the critical temperature decreasing due to the proximity effect. Contrary to expectations, all transitions were very sharp with the width ranging from 25 mK for the VN film to 50 mK for the VN/Pd₀.₉₂Fe₀.₀₈ structure. We propose epitaxial single-crystalline thin films of VN and heteroepitaxial Pd₁₋ₓFeₓ/VN and VN/Pd₁₋ₓFeₓ (x ≤ 0.08) structures grown on MgO(001) as the materials of a choice for the improvement of superconducting magnetic random access memory characteristics.
Introduction

Since its invention, rapid single-flux quantum (RSFQ) logic [1,2] based on superconducting digital electronics has been seriously considered as an alternative to semiconductor electronics for supercomputing applications [3-5]. Merging it with magnetism [6-8] has given a birth to superconducting spintronics [9,10]. The latter concept was implemented in the US Cryogenic Computing Complexity (C3) Program [11-13] with the goal “to demonstrate a small-scale computer based on superconducting logic and cryogenic memory that is energy-efficient, scalable and able to solve interesting problems”, opening prospects of reaching 100 PFLOPS/s with about 200 kW of electric power consumption including the cryogenic cooling. Niobium-based Josephson junction technology is currently implied to be used for the logics fabrication, however, hybrid Josephson junctions incorporating magnetic components are also considered for the mainframe computation components [9,14-19], and cache and main memories [8,20-25]. It is argued that the use of magnetic Josephson junctions in single-flux quantum electronics significantly reduces the number of junctions and interconnects in the circuits [26] and also has other important advantages such as wide operation margins and low bit-error rate [27]. The magnetic material has to be magnetically soft, tunable and weak in the sense of small spin-polarization of the conduction band [10,28]. The latter provides a large superconducting coherence length and hence bypasses a necessity to deposit flat, nanometer-thick continuous layers expected for strong elemental ferromagnets. A combination of niobium as a superconductor with a Pd$_{1-x}$Fe$_x$ alloy as a soft and weak ferromagnet was considered as material of choice for superconducting magnetic random access memories (MRAM) [8,29,30]. However, no further developments towards a prototype using a Pd$_{1-x}$Fe$_x$ alloy have been demonstrated. There are indications of non-homogeneous, nanoclustered magnetism in Pd$_{0.96}$Fe$_{0.04}$ films grown on niobium [31], which may cause a shortening of the spin-memory length [32] and a reduction of the Josephson critical current.

In general, the metallic Nb lattice (body-centered cubic with $a_{Nb} = 329.4$ pm) poorly matches that of the palladium-rich Pd$_{1-x}$Fe$_x$ alloys (face-centered cubic with $a_0 = 389$ pm). Therefore, a good crystallinity of the layer stack can hardly be expected. In the resulting polycrystalline films, crystallite boundaries and crystal lattice imperfections can lead to the segregation of iron impurities and to nanoclustering of the alloy. Following the development of a way to grow single-crystalline, magnetically homogeneous epitaxial Pd$_{1-x}$Fe$_x$ films on MgO(001) single-crystalline substrates [33], we propose fully epitaxial Pd$_{1-x}$Fe$_x$/VN and VN/Pd$_{1-x}$Fe$_x$ ($x \leq 0.08$) building blocks as an alternative choice for superconducting MRAM materials, in which vanadium nitride (VN) serves as the superconductor. The magnetic anisotropies of a 20 nm thick Pd$_{0.96}$Fe$_{0.04}$ film of the first-generation epitaxial sample of VN/Pd$_{0.96}$Fe$_{0.04}$ on MgO(001) were studied by using a ferromagnetic resonance technique in [34].

Results and Discussion

Sample preparation

Single-crystalline MgO(001) (henceforth designated MgO) epitaxial substrates (CRYSTAL GmbH, Germany) with a size of $10 \times 5 \times 0.5$ mm$^3$ were annealed at 800 °C for 5 min in the ultrahigh vacuum (UHV) molecular beam epitaxy (MBE) chamber with a residual pressure below $10^{-10}$ mbar (SPECS, Germany). Then, depending on the desired structure, either the Pd$_{1-x}$Fe$_x$ alloy layer or the VN layer was deposited. The Pd$_{1-x}$Fe$_x$ layers were grown by means of UHV MBE following a three-step procedure described in detail in [33]. Metallic Pd (99.95% purity, EVOCHEM GmbH, Germany) and Fe (99.97% purity, ChemPur GmbH, Germany) were co-evaporated from the pre-calibrated high-temperature effusion cells to obtain the desired Pd$_{1-x}$Fe$_x$ composition.

Vanadium nitride layers were synthesized by using reactive DC magnetron sputtering (MS) in the UHV chamber with a base pressure of $p \leq 5 \times 10^{-10}$ mbar (BESTEC, Germany). During this process, the substrate had a temperature of 500 °C. A mixture of high-purity (99.9999%) argon (Ar) from a gas chromatography purification system and high-purity (99.9999%) nitrogen (N$_2$) at a composition of Ar/N$_2 = 60:40$ was used as plasma gas for the reactive synthesis of VN. During the deposition process, the pressure of the Ar/N$_2$ gas mixture in the chamber was automatically kept at $6 \times 10^{-3}$ mbar. A metallic vanadium disk of 99.95% purity (GIRMET Ltd, Russia) was used as a target. The magnetron power was 50 W, the distance between the target and the substrate was 20 cm, and the deposition rate was 0.2 nm/min.

To grow heterostructures, the samples on the molybdenum holder were moved without breaking vacuum via the UHV transfer line between the MBE and MS deposition chambers as well as the analysis chamber (SPECS, Germany).

To perform a comparative study allowing to see only the proximity effect of the ferromagnetic layer on the properties of the superconducting VN layer, the latter was deposited in one run for all studied samples. To do this, we mounted two $10 \times 5 \times 0.5$ mm$^3$ MgO substrates close and parallel to each other on the sample holder and used a system of two orthogonal shutters in the MBE chamber. After depositing a 20 nm thick Pd$_{0.96}$Fe$_{0.04}$ layer onto one substrate (with the second being blocked by the shutter), the sample holder was moved to...
the magnetron chamber, and a 30 nm layer of VN was grown on both substrates. Then, the holder was moved back to the MBE chamber, and a 12 nm thick Pd$_{0.92}$Fe$_{0.08}$ layer was deposited to a half of both samples using the second shutter. The thicknesses of the Pd$_{1-x}$Fe$_x$ layers were adjusted to possess identical magnetic moments. In situ tests of crystallinity, VN stoichiometry and resulting composition of Pd$_{1-x}$Fe$_x$ were taken at each deposition step using low-energy electron diffraction (LEED) and X-ray photoelectron spectroscopy (XPS). Finally, all structures were capped with 10 nm layer of undoped Si by magnetron sputtering to prevent sample deterioration. Thus, a VN film and stacks of Pd$_{0.96}$Fe$_{0.04}$/VN and VN/Pd$_{0.92}$Fe$_{0.08}$ (the first component in a stack being directly deposited to MgO) have been obtained with the identical properties of the VN layer in each sample.

**Crystallinity and epitaxial growth**

The crystallinity and the epitaxial growth of the thin films were examined in situ by using LEED (SPECS, Germany). LEED images were taken of the pristine MgO(001) substrate after annealing (Figure 1a), after the deposition of VN (30 nm) on MgO (Figure 1b), after the deposition of Pd$_{0.92}$Fe$_{0.08}$ on VN (Figure 1c) and after the deposition of VN on Pd$_{0.96}$Fe$_{0.04}$ (Figure 1d). Figure 1b indicates that the individual VN thin film has grown cube-on-cube epitaxially (for an individual Pd$_{1-x}$Fe$_x$ film see the full crystallinity analysis in [33]). Figure 1c,d shows that the Pd$_{0.96}$Fe$_{0.04}$/VN and VN/Pd$_{0.92}$Fe$_{0.08}$ heterostructures are pass-through epitaxial. This is, first of all, due to the good lattice match between MgO, VN and Pd: $a_{\text{MgO}} = 421.2$ pm, $a_{\text{VN}} = 413.7$ pm [35] and $a_{\text{Pd}} = 389.1$ pm. Thus, the lattice mismatch between MgO and VN is only about 1.7%, and between Pd and VN it is as small as 5.95%.

The in situ LEED analysis was corroborated with ex situ X-ray diffraction (XRD, BRUKER D8, Germany) measurements using Cu Kα ($\lambda = 1.5418$ Å) radiation in the Bragg–Brentano geometry with a scanning rate of 0.002°/s in the 2θ range from 17° to 82° and a step width of 0.0153°. Room-temperature XRD patterns of the pristine MgO(001) substrate, the VN thin film on MgO, Pd$_{0.96}$Fe$_{0.04}$ on MgO and the Pd$_{0.96}$Fe$_{0.04}$/VN heterostructure are shown in Figure 2. The 0–2θ scans clearly indicate the single-crystalline structure of the VN and Pd$_{0.96}$Fe$_{0.04}$ thin films and of the Pd$_{0.96}$Fe$_{0.04}$/VN heterostructures. The (002) reflex of the MgO substrate, the (002) reflex of the VN film (30 nm), and the (002) reflex of the Pd$_{0.96}$Fe$_{0.04}$ (20 nm) film can be easily identified.

![Figure 1: LEED patterns of (a) pristine MgO annealed at 800 °C, (b) the VN film, (c) the VN/Pd$_{0.92}$Fe$_{0.08}$ and (d) Pd$_{0.96}$Fe$_{0.04}$/VN structures on the MgO(001) substrate. All patterns were taken at an electron energy of 140 eV.](image)

![Figure 2: XRD patterns of pristine MgO substrate, VN, Pd$_{0.96}$Fe$_{0.04}$ (prepared in a separate deposition experiment) and Pd$_{0.96}$Fe$_{0.04}$/VN.](image)

The significant peak broadening of the diffraction maxima of VN and Pd$_{0.96}$Fe$_{0.04}$ is primarily due to small coherent scattering range $\tau$ along the normal to the film plane (Scherrer broadening); XRD data with accounting for the instrument function [33] yields estimates of $\tau \approx 22.0$ nm for Pd$_{0.96}$Fe$_{0.04}$, $\tau \approx 12.6$ nm for Pd$_{0.92}$Fe$_{0.08}$ and $\tau \approx 30.4$ nm for VN, which agree quantitatively with the film thickness values $d(\text{Pd}_{0.96}\text{Fe}_{0.04}) \approx 21.5$ nm, $d(\text{Pd}_{0.92}\text{Fe}_{0.08}) \approx 12.5$ nm and $d(\text{VN}) \approx 29.8$ nm, respectively, measured ex situ with a BRUKER DektakXT stylus profiler by using the shadow mask method. Thus, LEED and XRD measurements confirm that the VN thin film and the Pd$_{0.96}$Fe$_{0.04}$/VN and VN/Pd$_{0.92}$Fe$_{0.08}$ heterostructures have grown cube-on-cube epitaxially and that all samples are single crystalline.
Stoichiometry and chemical composition

The stoichiometry and chemical composition of the VN and the Pd$_{1-x}$Fe$_x$ layers were analyzed in situ using XPS. The measurements were carried out in the UHV analysis chamber (base pressure $p < 3 \times 10^{-10}$ mbar) equipped with a Mg Kα X-ray source operated at 12.5 kV and 250 W, and a Phoibos-150 hemispherical energy analyzer (all from SPECS, Germany). Figure 3a,b shows the XPS spectra of the as-deposited VN/Pd$_{0.92}$Fe$_{0.08}$ thin film heterostructure. The binding energies of the Fe 2p$_{1/2}$, Fe 2p$_{3/2}$, and Pd 3d$_{3/2}$ and Pd 3d$_{5/2}$ states are 721.0, 707.7, and 340.2 and 335.0 eV, respectively, which agrees well with literature data [33,36].

Figure 3c,d shows the XPS spectra of the VN thin film on MgO. The binding energies of the V 2p$_{1/2}$, V 2p$_{3/2}$ and N 1s states are 521.1, 513.6 and 397.4 eV, respectively, which are very close to that given in the literature for crystalline VN [37,38]. The presence of a characteristic satellite at a binding energy of ca. 515 eV is a fingerprint of V in a nitride compound [37]. The chemical composition of the as-grown VN and Pd$_{1-x}$Fe$_x$ layers was analyzed with the CasaXPS software [39]. According to the XPS data, the stoichiometry of synthesized layers was Pd/Fe = 96:4, V/N = 52.5:47.5 and Pd/Fe = 92:8, respectively, with an accuracy of ±0.5%. Neither impurities nor surface contaminations were detected (compare with [40]). All recorded high-resolution XPS spectra of VN and Pd$_{1-x}$Fe$_x$ films were calibrated to the binding energies of crystalline VN at 513.6 eV and of metallic Pd at 335.0 eV [33,37], respectively.

Magnetic moment measurements shown in Figure 4 confirm the composition of Pd$_{0.96}$Fe$_{0.04}$ and Pd$_{0.92}$Fe$_{0.08}$ through the ferromagnetic transition temperature $T_C \approx 125$ K and $T_C \approx 240$ K, respectively [41].

![Figure 4: Saturation magnetization $M_s(T)$ as a function of the temperature of the Pd$_{0.96}$Fe$_{0.04}$/VN (green symbols) and VN/Pd$_{0.92}$Fe$_{0.08}$ (red symbols) heterostructures measured in a magnetic field of 200 Oe.](image)
Temperature dependence of resistance and superconducting transition

A physical property measurement system (QUANTUM DESIGN PPMS-9, USA) was used for studying the temperature dependence of the electrical resistance of the VN thin films and Pd₀.₉₆Fe₀.₀₄/VN and VN/Pd₀.₉₂Fe₀.₀₈ heterostructures in the temperature range of 4.2–300 K. A four-probe resistance measurement scheme was used. Figure 5 shows the measurement results as a function of the temperature for the epitaxial VN film and the heteroepitaxial Pd₀.₉₆Fe₀.₀₄/VN and VN/Pd₀.₉₂Fe₀.₀₈ samples. Table 1 contains the data on the residual resistance ratio RRR (i.e., the ratio of room temperature resistance, \(R_{300K}\), to the resistance at 10 K, \(R_{10K}\)), the superconducting transition temperature (mid-transition criterion) and the width of the superconducting transition (10–90% criterion) for the VN thin film and the heterostructures with Pd₁₋ₓFeₓ.

| structure                        | RRR | \(T_c\) (K) | \(\Delta T_c\) (mK) |
|----------------------------------|-----|-------------|---------------------|
| VN(30 nm)                        | 5.2 | 7.7         | 25                  |
| Pd₀.₉₆Fe₀.₀₄(20 nm)/VN(30 nm)    | 3.5 | 7.2         | 37                  |
| VN(30 nm)/Pd₀.₉₂Fe₀.₀₈(12 nm)    | 2.6 | 6.1         | 50                  |

The temperature dependence of the resistance of the VN thin film is of metallic type and exhibits two temperature intervals, one above 250 K and another one in the range of 80–180 K, of quasi-linear temperature dependence with different temperature coefficients of resistivity (TCR), i.e., \(9.7 \times 10^{-3} \Omega/K\) and \(2.1 \times 10^{-2} \Omega/K\), respectively, marked by red straight lines over the green line in Figure 5a. It is similar to the \(R(T)\) behavior of VN/MgO(011) samples in [42], which was explained by a change in the electron/phonon scattering amplitude upon the structural phase transition from cubic to tetragonal at \(T_s = 250\) K. Below 50 K the \(R(T)\) dependence saturates approaching the residual resistance originating, in general, from impurities and imperfections. Further cooling results in the phase transition to the superconducting state as it is shown in Figure 5b. The RRR value of 5.2 and the room-temperature resistivity of 42.5 \(\mu\Omega\cdot\text{cm}\) for the 30 nm thick VN film are among the best values obtained to date [42-45], indicating the high purity and structural quality of our VN film.

The superconducting transition temperature \(T_c\) of the VN film is 7.7 K (see Table 1), which is well above the temperature of liquid helium, \(LHeT = 4.2\) K. Figure 5b shows a very sharp resistive transition at \(T_c = 7.7\) K with a small width of 25 mK, which is quite remarkable compared to an elemental niobium film of the same (30 nm) thickness deposited in the same chamber and under vacuum conditions (\(\Delta T_c [\text{Nb}(30\text{ nm})] = 10–23\) mK).

Combining the VN film into a heterostructure with a palladium-rich Pd₁₋ₓFeₓ alloy leads to a lowering of \(T_c\) because of the proximity effect [28]. This may shift the material operation temperature close to or even below the \(LHeT\). With the iron content \(x\) in Pd₁₋ₓFeₓ alloy below 0.08 its magnetic properties meet all the requirements for the F-layer in superconducting spintronic S/F/S-type structures, i.e., it is a weak ferromagnet with a low coercive field [41]. It is important that magnetic properties of epitaxial Pd₁₋ₓFeₓ films are precisely controlled with the iron content \(x\) [41], and a perfect cube-on-cube epitaxy is realized with either the MgO(001) substrate or with the supercon-
ducting VN layers in any sequence. Figure 5b shows that 12 nm thick layer of Pd_0.92Fe_0.08 alloy adjacent to the 30 nm VN film lowers $T_c$ from 7.7 K to 6.1 K, which is well above the LHeT. Moreover, Figure 5b demonstrates that the transition stays sharp: the maximum $\Delta T_c$ increases only to 50 mK, and there is no tail towards lower temperatures. Also, there is a room to optimize the superconducting parameters of the VN film towards an increase in $T_c$ by about 1 K [43,44]. In our opinion, the results hint at a possible use of heteroepitaxial combinations of nitrides as superconductors and palladium-rich Pd$_{1-x}$Fe$_x$ alloys as weak tunable ferromagnets to improve the operation characteristics of superconductor–ferromagnet–insulator heterojunctions for superconducting spintronics applications. For example, cubic superconducting MoN$_x$, which is a Josephson junction technology material [4,5,46], exhibits a good epitaxial match with Pd$_{1-x}$Fe$_x$ alloys, $a_0$(MoN) = 416.3 pm.

Conclusion

Fully epitaxial single-crystalline thin films of VN and heteroepitaxial structures of Pd$_{1-x}$Fe$_x$/VN and VN/Pd$_{1-x}$Fe$_x$ ($x = 0.04, 0.08$, respectively) were grown on single-crystalline MgO(001) substrates using a combination of UHV molecular beam epitaxy and magnetron sputtering. The obtained 30 nm thick VN films exhibit a sharp superconducting transition with $T_c = 7.7$ K and $\Delta T_c = 25$ mK. The heteroepitaxial Pd$_{0.96}$Fe$_{0.04}$/VN and VN/Pd$_{0.92}$Fe$_{0.08}$ structures reveal a superconductor–ferromagnet proximity suppression of the transition temperature to $T_c = 6.1$ K. This is, however, well above the liquid helium temperature of 4.2 K and, therefore, suitable for superconducting spintronics. The superconducting transition stays sharp with a somewhat larger width of $\Delta T_c = 50$ mK. Moreover, there is no resistive tail towards lower temperatures. These results, in our opinion, indicate that fully epitaxial Pd$_{1-x}$Fe$_x$/VN and VN/Pd$_{1-x}$Fe$_x$ thin film stacks can be considered as building blocks for superconducting spintronics elements.

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