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Measurement of T_c suppression in tungsten using magnetic impurities

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We have measured the effects of dilute magnetic-atom doping on the superconducting transition temperature of tungsten thin films. Our ''T_c tuning'' technique is accurate, precise, and simple. Experiments were performed using dc-magnetron-sputtered tungsten films with undoped values of T_c in the range of 70–150 mK. The magnetic-atom doping was achieved using ion implantation. Specific T_c suppressions of between 5% and 65% were targeted and observed in this study. The transition width of each undoped sample was ≈1 mK and the transition widths remained sharp after implantation with $^{56}\mathrm{Fe}^+$ ions. Our data are in good agreement with predictions of a linear dependence of T_c suppression with increasing magnetic-atom concentration, in the small concentration limit. At higher concentrations, antiferromagnetic coupling between the magnetic dopant atoms becomes important and the T_c-suppression effect is diminished. We use our T_c data to calculate the Abrikosov–Gor'kov (AG) and Ruderman–Kittel–Kasuya–Yosida (RKKY) spin-flip relaxation parameters τ_{AG} and τ_{RKKY}. We conclude with a brief discussion of applications of the T_c-tuning technique, and present our plans for future studies in this area. © *1999 American Institute of Physics.* [S0021-8979(99)03624-5]

MOTIVATION AND BACKGROUND

Thin films of superconducting materials have been used as phonon sensors for many years. The exact design of phonon sensors varies according to the intended applications.[1] For example, some of the cryogenic detectors developed by the Cold Dark Matter Search (CDMS) collaboration to search for weakly interacting massive particle (WIMP) dark matter rely on collecting thermal phonons using overlapping thin films of superconducting aluminum and tungsten.[2] These phonon sensors are called ''W/Al quasi-particle-trap-assisted electrothermal-feedback transition-edge sensors (QETs).''[3] The energy sensitivity of a QET phonon sensor depends critically on the T_c of the superconducting W film, and also on the width of the superconducting transition.

Our tungsten and aluminum films are deposited using a Balzers 450 dc-magnetron sputtering system and a 99.999% pure, vacuum-pressed tungsten target purchased from Johnson Matthey. This metalization system provides high-quality films with sufficiently narrow superconducting transitions (width ≤1 mK), however, the ''as-deposited'' W films often have values of T_c above our target value of 65 mK. The exact value of the tungsten film T_c obtained in a given metalization run depends significantly on the details of deposition parameters and specific vacuum chamber conditions. The variability in T_c from one batch of devices to the next is likely due to differing stress characteristics in the deposited films, as well as to minute quantities of various residual impurities present in our vacuum chamber during deposition. Because film deposition parameters cannot be absolutely controlled, and even slight deviations from our prescribed deposition recipe can give rise to nonoptimal QET T_c values, we recently focused on developing a technique to adjust the T_c of our sputtered tungsten films, by postprocessing. We were specifically interested in developing a technique which would suppress T_c by a prescribed amount, yet no broaden the superconducting transition nor degrade device performance.

DEMONSTRATION OF T_c-TUNING TECHNIQUE

Our idea was to use the precision and accuracy of conventional ion-implantation techniques to tune the T_c of individual W films. To study this possibility, we used a collection of 350-Å-thick, sputtered W films with as-deposited T_c's of ≈70–150 mK. The films were deposited in a series of separate depositions; the substrates used were identical, and consisted of a 210-Å-thick layer of amorphous silicon on top of a (100) silicon wafer. Immediately adjacent samples, each of area ≈0.25 cm^2, were taken from our as-deposited W films and separately ion-implanted with $^{56}\mathrm{Fe}^+$ ions at 50 keV. At this energy, a negligible fraction of the incident ^{56}Fe ions fully penetrate a 350-Å-thick tungsten film. Doses ranging from 1.00×10^{12} to 1.00×10^{13} ions/cm^2 (±0.01%) were used in the study presented here. A plot of the depth profile

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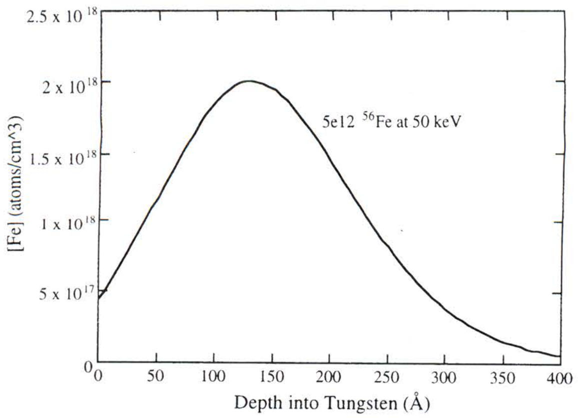

FIG. 1. Calculated Fe concentration vs depth into the W film for one Fe-ion implantation dose used in our study.

for one of our implants (5.00×10^{12} cm^{-2} at 50 keV), showing the concentration of implanted $^{56}\mathrm{Fe}^+$ ions versus depth into the tungsten film, is presented in Fig. 1. The plot in Fig. 1 was calculated using PROFILE CODE™, a software simulation package purchased from Implant Sciences, Inc. The sputtering target used to deposit all of our tungsten films contains a residual impurity concentration of 0.9 ppm Fe. In our study, this Fe concentration would correspond to an equivalent 50 keV Fe-atom dose of $\approx 1.3\times 10^{11}$ cm^{-2}.

In Fig. 2, we plot the measured film T_c versus ion dose for 50 keV $^{56}\mathrm{Fe}^+$ implanted into a 350-Å-thick tungsten film with unimplanted superconducting T_c of (135 ± 2) mK. In Fig. 3, we plot the T_c data in an alternate form; we plot T_c (control sample)$-T_c$ (implanted sample) versus peak $^{56}\mathrm{Fe}^+$ concentration in atomic ppm, calculated for each of the measured implant doses, and for the one control sample. Our control sample was not implanted; it contained only the 0.9 ppm Fe contributed from the high-purity W sputtering target. A quadratic fit to the combined data provides an empirical relationship between [Fe], the peak concentration of Fe ions in our W samples, and ΔT_c, the resulting shift in the transition temperature:

$$\Delta T_c = -3.16 + 1.66[\mathrm{Fe}] - 0.00457[\mathrm{Fe}]^2,$$

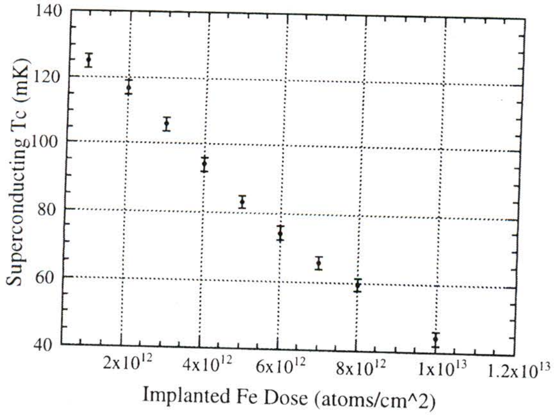

FIG. 2. Measured superconducting transition temperature for 350-Å-thick tungsten films implanted with $^{56}\mathrm{Fe}^+$ at 50 keV kinetic energy.

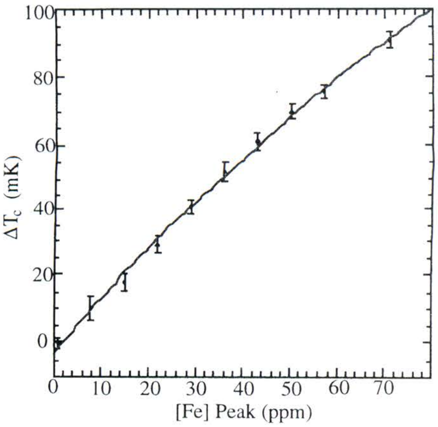

FIG. 3. Measured T_c suppression for tungsten samples as a function of peak Fe concentration in our tungsten films. The control sample with 0.9 ppm [Fe] had a T_c of 135 ± 2 mK. The plotted points include the [Fe]=0.9 ppm from the sputtering target. The fitted curve is a quadratic.

where $\Delta T_c = T_c$ (control sample)$-T_c$ (implanted sample) is measured in mK and [Fe] is in ppm, atomic fraction.

We have demonstrated the T_c-tuning process on several additional W films with unimplanted T_c's of 70–150 mK. In the simplest approach, we used the data shown in Fig. 2 to determine the dose of $^{56}\mathrm{Fe}^+$ needed to shift the T_c of a given film the desired amount. We assumed a straight-line fit to the low-dose data plotted in Fig. 2, then shifted the y intercept of that line up or down to match the T_c of the new unimplanted control sample. The slope ($=1.5$ mK/ppm) was not adjusted because the $^{56}\mathrm{Fe}^+$-implanted energy for all samples was held at 50 keV. The ion dose necessary to shift T_c by a prescribed amount was then read from the x axis of the intercept-adjusted graph. Using this simple approach, we have successfully tuned several films to within a few mK of a specified target T_c. Most importantly, for all of our samples, the narrow superconducting transition widths of ≈ 1 mK were maintained even after the T_c-tuning procedure was completed.

In Fig. 4, we show the 135 mK film data, along with a fit to the theoretical prediction of Abrikosov and Gor'kov (AG) for the general dependence of T_c on magnetic-impurity concentration. Based on the AG model,[4] we assume an antisymmetric exchange coupling between the magnetic impurity ions (Fe) and the lattice host atoms in our W films. We assume a one-electron Hamiltonian is sufficient to describe the W system. We also assume the Fe concentration is sufficiently low that a dilute-limit approximation is valid, and that the presence of impurity atoms results only in the scattering of W conduction electrons. That is, we assume no coupling exists between one impurity atom and another. Finally, we assume the pair potential for Cooper pairs in the W film is position independent. For reference, the coherence length in our superconducting W films is $\xi_0\approx 0.3$ μm, which is much greater than the film thickness ≈ 350 Å. In the AG model, the interaction between a W-atom electron and an iron impurity can be represented by a perturbing Hamiltonian term of the form:

$$H_{\mathrm{perturbation}} = K(\mathbf{r}_e - \mathbf{r}_i)\mathbf{S}_e\cdot\mathbf{S}_i,$$

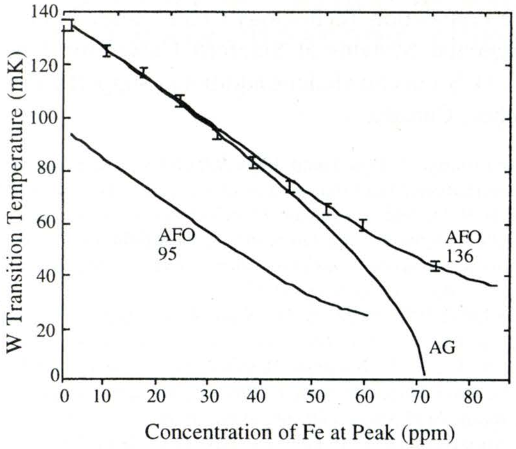

FIG. 4. Superconducting transition temperature of tungsten as a function of iron dopant concentration (at peak of ion-implant distribution). Curve "AG" corresponds to the Abrikosov and Gor'kov model. Curves "AFO" correspond to the AG model modified to include antiferromagnetic ordering between Fe atoms. Fit "AFO 136" corresponds to $T_{co}=136$ mK. For comparison, we include fit "AFO 95," the predicted curve for films with $T_{co}=95$ mK. Our Fe in W data are best fit for the AFO model with $\alpha=-(0.06\pm0.01)$.

where $\mathbf{r}_e$ and $\mathbf{r}_i$ are the position vectors of the W-atom electron and the magnetic impurity atom, respectively. Similarly, $\mathbf{S}_e$ and $\mathbf{S}_i$ are the spin vectors of the W-atom electron and the magnetic-ion impurity. $K(\mathbf{r}_e-\mathbf{r}_i)$ is the exchange coupling constant, which can be positive or negative because it operates differently (asymmetrically) on the two electron (↑↓) in a Cooper pair. The AG model for superconductors with magnetic impurities yields the following relationship between the reduced transition temperature T_c/T_{co} and the reduced magnetic-impurity concentration x/x_c of a sample:[5]

$$\ln(T_c/T_{co})=\psi\{1/2\}-\psi\{(1/2)+(1/4)(e^{-\gamma})(x/x_c)\times(T_{co}/T_c)\},$$

where ψ is the digamma function, i.e., ψ is the derivative of the logarithm of the Γ function:

$$\psi=-\gamma-(1/z)-\sum_{k=1}^{\infty}\{1/(z+k)-(1/k)\},$$

and γ is the Euler–Mascheroni constant ($\gamma=0.577\,215\,66\ldots$). At low-impurity concentrations the AG model predicts a linear decrease in T_c as a function of x/x_c. At substantially higher concentrations, i.e., when $T_c \ll T_{co}$, the model predicts an extremely rapid decrease of T_c such that the superconductivity vanishes altogether at a certain critical concentration x_c. The critical concentration can be evaluated from the slope of the T_c vs x curve in the low concentration limit:

$$\left.\frac{dT_c}{dx}\right|_{x=0}\cong\frac{-\pi^2T_{co}}{8e^{\gamma}\,x_c}.$$

For our W data, and referenced to the peak of each Fe-implant distribution, this calculation yields a critical concentration $x_c=69$ ppm.

The AG curve shown in Fig. 4 fits our data well at low-impurity concentrations, where T_c decreases linearly with increasing impurity concentration. For higher concentrations, however, i.e., for doping levels exceeding ≈30–40 ppm Fe, our data deviate substantially from the AG curve. In particular, our data show that the T_c suppression due to antisymmetric electron–ion exchange coupling in the film becomes less significant at the higher Fe concentrations studied. At these higher concentrations it is probable that the weak coupling approximation for the W–Fe system is not valid and Fe–Fe interactions can be no longer ignored. Indeed, we believe that the observed deviation from the AG model at high Fe concentrations (i.e., $T_c \ll T_{co}$) is due to antiferromagnetic interactions between the Fe impurities in our samples. The antiferromagnetic ordering increases the superconducting order parameter, which allows the system to superconduct when it would otherwise be normal ($x \geqslant x_c$). In contrast, *ferromagnetic* (rather than antiferromagnetic) coupling between impurities would inhibit the persistence of the superconducting state. Including the effect of nearest-neighbor magnetic interactions between the dopant atoms, we get

$$\ln(T_c/T_{co})=\psi\{1/2\}-\psi\{(1/2)+(1/4)(e^{-\gamma})(x/x_c)\times(T_{co}/T_c)[1+\alpha(x/x_c)(T_{co}/T_c)]\},$$

where α is a magnetic coupling parameter[5]

$$\alpha=\tau_{\mathrm{AG}}(x_c)/\tau_{\mathrm{RKKY}}(x_c,T_{co}),$$

$\alpha>0$ for ferromagnetic ordering (parallel spins) and $\alpha<0$ for antiferromagnetic ordering (antiparallel spins). The best fit to our data shown in Fig. 4 has $\alpha=-0.06\pm0.01$. The parameters α_{AG} and τ_{RKKY} (Ruderman–Kittel–Kasuya–Yosida) give the relative strengths of the impurity–host atom (Fe–W) interactions and the impurity–impurity (Fe–Fe) interactions, respectively. Using our data, we can readily calculate τ_{AG} and τ_{RKKY} for the Fe–W system. In the dilute dopant limit:

$$\left.\frac{dT_c}{dx}\right|_{x=0}=\frac{\pi}{4\tau_{\mathrm{AG}}x}.$$

Thus

$$\tau_{\mathrm{AG}}(x_c)=-\frac{\pi}{4(-1.5\ \mathrm{mK/ppm})(69\ \mathrm{ppm})}=7.6\ \mathrm{K}^{-1},$$

and

$$\tau_{\mathrm{RKKY}}(x_c,T_{co})=(7.6\ \mathrm{K}^{-1})/(-0.06)=-130\ \mathrm{K}^{-1}.$$

Magnetic ordering in superconductors is discussed in depth by Roshen and Ruvalds,[5] Maple,[6] Fulde and Keller,[7] and Dupont, Ziemniak, and Usadel.[8]

CONCLUSION AND FUTURE DIRECTIONS

The T_c-tuning technique we have developed is reliable and reproducible, and offers flexibility for fabricating cryogenic devices with specific superconducting transition temperatures. The fact that the measured transition widths of our T_c-suppressed films are not affected by the doping process is encouraging, since we expect that a broadening of the transition would accompany a decrease in the critical current of our superconducting films. For our primary applications to cryogenic particle and photon detectors, such a decrease in critical current would adversely affect the overall performance of our devices. Soon, we will measure directly the effects of dilute Fe-atom doping on the critical current and heat capacity of the W films.

We are in the process of extending our T_c-suppression work to W–Co and W–Ni systems, to further study the effects on T_c of magnetic ordering in superconducting films. Those results will be presented in a future publication.

ACKNOWLEDGMENTS

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