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Weak-localization effects in $V_{1-x}Si_x$ amorphous alloys with high Si content

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We present high-field magnetoresistance measurements on a series of $V_{1-x}Si_x$ amorphous alloys with high Si content ($0.50 \le x \le 0.86$). For alloys with $x \le 0.75$ the magnetoresistance-versus-field curves are characteristic of weak localization effects in the presence of strong spin-orbit coupling despite rather low values of the disorder parameter. But a tentative fit of experimental curves at high field from model equations is made difficult on the one hand by the electron-electron interaction term becoming important at high field and on the other hand by the possible inadequacy of model formulations of weak localization effects in the high H/T limit. However, reasonable values for the inelastic and spin-orbit relaxation times are tentatively proposed. Due to phase separation occurring for $x \ge 0.82$, the metal-nonmetal transition could not be observed.

I. INTRODUCTION

In recent years theoretical and experimental works on localization phenomena in disordered systems have become widely developed. Concerning the three-dimensional materials, the amorphous metallic alloys are an excellent tool for studying these effects. The analysis of measurements of resistivity versus temperature[1] and of magnetoresistance[2,3] in the light of theoretical models has allowed in various systems a quantitative determination of the inelastic and spin-orbit relaxation times. Up to now, most of the available data on magnetoresistance were obtained under weak or moderate magnetic field ($H \le 10$ T) and for alloys away in composition from a metal-nonmetal transition. Over this field range theoretical predictions were rather well verified. However, some authors have already shown the limits of the models in presence of high magnetic field.[4,5]

In this paper, we present the results of magnetoresistivity measurements on the amorphous vanadium-silicon system ($V_{1-x}Si_x$) with high Si content ($0.50 \le x \le 0.86$) and under strong pulsed magnetic field ($H \le 32$ T). Results on superconducting amorphous $V_{1-x}Si_x$ alloys ($x < 0.30$) were already discussed in a previous paper.[6] For silicon concentrations between 50–75 at. % the alloys do not become superconductors above 1.4 K. We show that magnetoresistance measurements can be interpreted in terms of weak localization and interelectronic interactions. In addition, the use of a high magnetic field allows us to determine with a reasonable degree of confidence the spin-orbit scattering frequency even when the spin-orbit coupling is strong. When the silicon concentration reaches 85 at. %, one expects a metal-semiconductor transition to occur. A detailed study of the transition is precluded in this system from the fact that the V-Si alloys become phase separated in the amorphous state for Si concentrations higher than 82 at. %. Consequently, due to a remanent metallic phase the resistivity-versus-temperature measurement remains basically metallic up to $x = 0.86$. However the magnetoresistance quickly increases when reaching the concentration of 83 at. % of silicon.

II. BRIEF THEORETICAL BACKGROUND

A. Weak localization

The expression for the magnetoresistance due to weak localization has been established by various authors.[7,8] In the presence of strong spin-orbit coupling, the equation takes the following form:

$$\frac{\Delta\rho}{\rho} = \alpha\rho \frac{e^2}{2\pi^2\hbar} \left[\frac{eH}{\hbar}\right]^{1/2} \left[\frac{1}{2} f_3 \left[\frac{H}{H_i}\right] - \frac{3}{2} f_3 \left[\frac{H}{H_{so}}\right]\right], \tag{1}$$

with

$$H_i = \frac{\hbar}{4eD} \tau_i^{-1} \tag{2}$$

and

$$H_{so} = \frac{\hbar}{4eD} (\tau_i^{-1} + 2\tau_{so}^{-1}) = H_i + H'_{so} \tag{3}$$

where τ_i and τ_{so} are the electronic relaxation times for inelastic and spin-orbit scattering respectively. D is the diffusion constant. The function $f_3(h)$ was given by Kawabata.[7] The prefactor α is one in the theoretical results for free electrons. The contribution of impurities with a localized moment can be discarded in the V-Si alloys, so spin-flip scattering is unlikely to occur.

B. Interelectronic interactions

The magnetoresistance of a weakly disordered electron gas, arising from spin splitting of conduction electron energies, has been calculated by Lee and Ramakrishnan[9] in the case of a three-dimensional system:

$$\left[\frac{\Delta\rho}{\rho}\right]_{\text{int spin}}=\rho\frac{e^2}{4\pi^2\hbar}F\left[\frac{k_BT}{2\hbar D}\right]^{1/2}g_3\left[\frac{g\mu_BH}{k_BT}\right] . \tag{4}$$

F is the screening parameter for the Coulomb interaction.

The orbital contribution given by Isawa and Fukuyama[10] has the form

$$\left[\frac{\Delta\rho}{\rho}\right]_{\text{int orb}}=\rho g(T,H)\frac{3}{8}\frac{e^2}{\hbar}\left[\frac{1}{D\hbar}\right]^{1/2}\times\frac{(k_BT)^2}{(4DeH)^{3/2}}\Phi(T,H) . \tag{5}$$

The expression of the coupling constant $g(T,H)$ was given by McLean and Tsuzuki.[11] Another expression of the orbital contribution to the magnetoresistance was derived by Al'tshuler *et al.*[8] But it is less accurate than the upper one in the presence of high magnetic field.

III. EXPERIMENT

The samples were prepared in an ultrahigh vacuum chamber by simultaneous condensation on cooled substrates (77 K) of vanadium and silicon evaporated using two electron-beam guns. The pressure was better than 2×10^{-8} Torr during the evaporation process, the ultimate obtainable vacuum being 1×10^{-9} Torr. The evaporation rates were controlled and measured using two quartz monitoring systems (QMS) whose vibration period is a linear function of the deposited mass. Thus the alloy composition can be determined after calibration of the QMS. Alloys with silicon content higher than 82 at. % were found to be phase separated from our electron microscope observations. The substrates used were glass plates with preevaporated chromium-gold electrodes for resistivity measurements.

The magnetoresistance experiments were performed at the Service CNRS des Champs Magnétiques Intenses in Toulouse. Our measurements were performed at temperatures between 4.2 and 25 K. We have done also measurements at higher temperatures ($T\sim100$ K) to make sure that the classical normal magnetoresistance is negligible in these high resistive systems. The magnetic field is quasistatic with the following characteristics: maximum field, 32 T; increasing time, 40 ms; decreasing time, 500 ms.

The magnetoresistance is measured by a classical four-probe technique using alternating current at high frequency (100 KHz) to avoid any spurious signal induced by the field at low frequency. In order to check that the magnetic sweep does not introduce a temperature rise, the data were computer recorded during both the increasing and decreasing phases of the pulse. In all cases, data obtained for the two phases were identical.

IV. EXPERIMENTAL DATA AND ANALYSIS

The field dependence of the magnetoresistance at several temperatures ranging between 4.2 and 20 K is shown in Fig. 1(a) for the amorphous $V_{0.48}Si_{0.52}$ alloy. Similar behavior is observed for other amorphous $V_{1-x}Si_x$ alloys with $x\leq0.74$. The magnetoresistance first increases at low field, then goes through a maximum before decreasing at high field. Such a behavior can be accounted for only if Eq. (1) plays a predominant role and, therefore, if there is a significant contribution arising from weak localization in the presence of strong spin-orbit coupling. This is obviously true in the low H/T limit, while the electron-electron interactions are expected to be more significant in the high H/T limit as can be seen from analytical expressions (4) and (5).

In order to compare the relative contributions to magnetoresistance arising from localization and from electronic interactions, one needs an estimation of the diffusion constant (D) and of the screening parameter (F). These quantities can be readily calculated from values of residual resistivity and by assuming one s electron per vanadium atom and four s electrons per silicon atom in a free electron model. The validity of a free-electron model for such alloys with high silicon content is certainly questionable. However, the values for the coefficient α in Eq. (1) as obtained from the initial slope

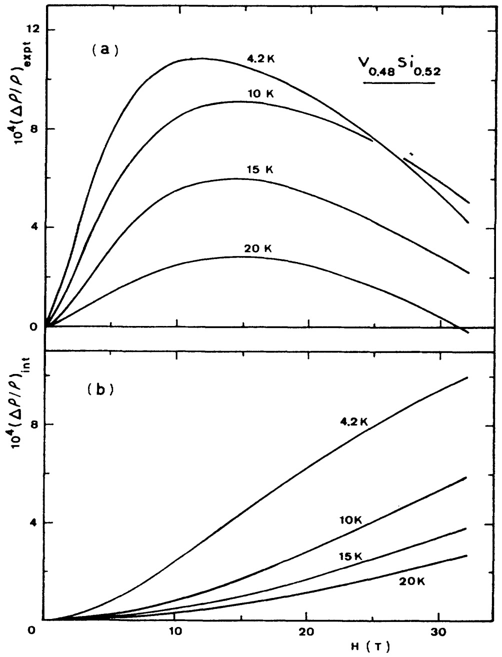

FIG. 1. (a) Experimental magnetoresistance measured at various temperatures for the amorphous $V_{0.48}Si_{0.52}$ alloy. (b) Contribution to the magnetoresistance arising from the spin component of interelectronic interactions. Calculations are made using the values of D and F given in Table I.

TABLE I. Experimental values of resistivity and calculated values for the diffusion constant D and the screening parameter F for three $V_{1-x}Si_x$ amorphous alloys. (α was obtained from the initial slope of $\Delta\rho/\rho$ vs H at 4.2 K.)

α	F	D ($m^2 s^{-1}$)	ρ ($\mu\Omega$ cm) T=4.2 K	x
1.5	0.43	0.81×10^{-4}	345	0.52
1.5	0.43	0.64×10^{-4}	433	0.60
1.6	0.43	0.42×10^{-4}	646	0.75

of $\Delta\rho/\rho$ versus H are of the order 1.5 (Table I) and therefore they do not depart too strongly from unity expected in a free-electron model. Values calculated for D and F within this approximation together with experimental values of residual resistivity for the alloys with $x=0.52$, 0.60, 0.74 are listed in Table I. Using these values for D and F, the contribution of electron-electron interactions to magnetoresistance were calculated according to analytical expressions[12] for Eqs. (4) and (5). The orbital contribution [Eq. (5)] is practically negligible as compared with the contribution from Eq. (4). As shown in Fig. 1(b) for the $V_{0.48}Si_{0.52}$ alloy, the contribution from electron-electron interactions is weak under low magnetic field but it increases strongly in the high field regime becoming then of the same order of magnitude as the experimental magnetoresistance. Taking into account the approximation made for the estimation of the diffusion constant and the screening parameter, it is clear that the accurate determination of the respective contributions of localization and interaction is very hazardous in the high H/T limit, while the localization effects are obviously predominant in the low H/T region.

This is further evidenced by fitting the magnetoresistance data after correcting for the calculated contributions from electron-electron interactions. Values for α are readily obtained from the low field region at 4.2 K. Values for H_i and H_{so} can be safely determined from the overall curve at 20 K, since for this temperature the value of H/T is moderate. Values for H_i at other temperatures can be easily determined from the low field curve, since $H'_{so}=H_{so}-H_i$ (spin-orbit scattering) is temperature independent.

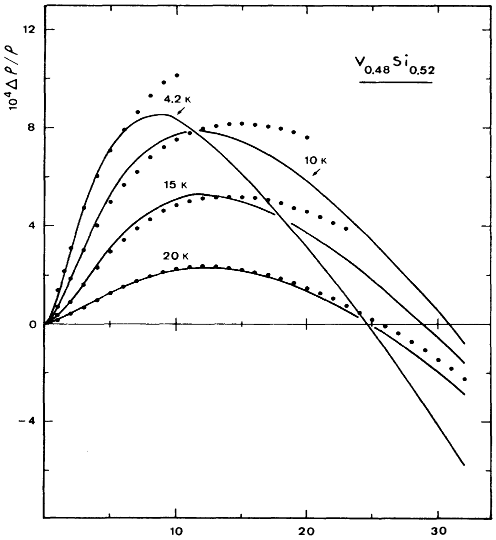

FIG. 2. Experimental magnetoresistance corrected for interelectronic contribution (solid lines) and best fit obtained from Eq. (1) (dots) for the amorphous $V_{0.48}Si_{0.52}$ alloy.

As an example, the best fit for the corrected magnetoresistance of the $V_{0.48}Si_{0.52}$ alloy at different temperatures is shown in Fig. 2. As expected the fit is fair for moderate values of H/T but it becomes poor at low temperature and high field. Such departures arise both from the uncertainties on the interelectronic contribution in high field and from the inadequacy of the theoretical expressions in the high H/T limit. However, we believe that the values for the characteristic frequencies for inelastic and spin-orbit scattering, τ_i^{-1} and τ_{so}^{-1} (as deduced from H_i and H'_{so}, respectively, see Table II) have at least a semiquantitative meaning, since they were obtained from the low and moderate H/T regime.

V. DISCUSSION

A few points deserve to be discussed at this stage, namely the pertinence of the weak localization models for these alloys with high silicon content, the temperature dependence of the inelastic scattering frequency and the concentration dependence of the spin-orbit scattering frequency. One could be surprised that weak localization models for magnetoresistance allow a quantitative analysis of the experimental results in our highly resistive alloys in which the disorder parameter $k_F l = 3Dm/\hbar$ is lying between 1.1 for $x=0.74$, and 2.1 for $x=0.52$. In fact, it was recently shown[13] that theoretical models based on perturbation calculations are valid not only for $k_F l \gg 1$ but also for $k_F l \simeq 1$. In the case of the $V_{1-x}Si_x$ system, we obtained a reasonable agreement between experimental and theoretical results except in the high H/T limit for $0.52 \le x \le 0.74$. When increasing x, $k_F l$ still decreases and the experimental data cannot be described through weak localization

TABLE II. Fit parameters for our amorphous alloys $V_{1-x}Si_x$.

		τ_i^{-1} (s^{-1})					H_i (T)					
τ_{so}^{-1} (s^{-1})	H'_{so} (T)	25 K	20 K	15 K	10 K	4.2 K	25 K	20 K	15 K	10 K	4.2 K	x
1.43×10^{12}	5.85		7.60×10^{11}	3.83×10^{11}	1.96×10^{11}	0.98×10^{11}		1.55	0.78	0.4	0.2	0.52
1.02×10^{12}	5.25	6.78×10^{11}				0.77×10^{11}	1.75				0.2	0.60
6.74×10^{11}	5.33		3.21×10^{11}	1.26×10^{11}		0.28×10^{11}		1.27	0.5		0.11	0.74

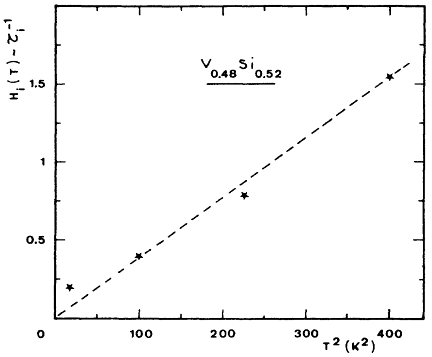

FIG. 3. Inelastic scattering frequency as a function of the square of the temperature for the amorphous $V_{0.48}Si_{0.52}$ alloy.

models.

Now we want to discuss the temperature dependence of the inelastic scattering frequency in the case of the alloy $V_{0.48}Si_{0.52}$. In Fig. 3 we have plotted τ_i^{-1} as a function of T^2. The data follow rather well a quadratic behavior: $\tau_i^{-1}=aT^2$, with $a=2\times 10^9$ s^{-1} K^{-2}. Surprisingly the same value for a was obtained in such different amorphous systems as $Cu_{57}Zr_{43}$ ($a=2.5\times 10^9$ s^{-1} K^{-2}) (Ref. 14) or $Cu_{50}Ti_{50}$ ($a=2.43\times 10^9$ s^{-1} K^{-2}).[1] This T^2 dependence of the inelastic scattering frequency has been already derived for many amorphous alloys. According to Bergmann,[15] it has to be ascribed to electron-phonon scattering. However, recent calculations predict a contribution from electron-phonon interaction proportional to T^3.[16] In fact a T^2 term is expected to arise from electron-electron interactions[16] but it is too small to describe the experimental effects. We also plotted our values for τ_i^{-1} as a function of T^3. One obtains a fairly good straight line but with an extrapolation at a finite value of τ_i^{-1} at $T=0$, which is not physical for inelastic scattering. At low temperatures departures from the quadratic temperature dependence of τ_i^{-1} are experimentally observed. This phenomenon has been shown in various metallic glasses by Bieri *et al.*[14] There is also a marked change of the inelastic scattering frequency with the vanadium concentration. In a recent paper Hickey *et al.*[1] also mentioned a Cu dependence of τ_i^{-1} in Cu-Ti alloys. At this time we do not have a clear explanation for this effect.

Now let us consider the spin-orbit scattering frequency. As shown in Table II, it decreases quite linearly with vanadium concentration. Such a behavior was expected since the spin-orbit coupling is mainly related to the vanadium sites. The dependence of τ_{so}^{-1} on x may also be due to changes in microstructure which reduce the spin-orbit rate as x is increased.

However, in an earlier paper concerning the superconducting amorphous alloys $V_{1-x}Si_x$ ($x<0.30$), we have noticed an anomalous behavior of τ_{so}^{-1}. In this low silicon-concentration region the spin-orbit scattering frequency (H'_{so}) linearly increases with x. We thought that the term H'_{so} as calculated from theory contains another contribution having the same symmetry-breaking effect as the spin-orbit coupling. It might be the tendency to form more covalent bonds in the alloys when the Si content increases. But this change in the concentration dependence of τ_{so}^{-1} is not clear at this time. Finally, we can remark that the spin-orbit coupling in our alloys is moderate. It has quite the same strength as in the amorphous $Cu_{50}Y_{50}$ or $Cu_{60}Zr_{40}$.[13] It would be interesting to compare our results to those obtained on $Y_{1-x}Si_x$. The magnetoresistance has been measured by Bieri *et al.*[17] at low field, but only qualitative analysis was done.

VI. CONCLUSION

In this paper we present magnetoresistance measurements on amorphous alloys $V_{1-x}Si_x$ with high silicon content. The results show that weak localization effects are well evidenced. The data are tentatively analyzed by following the current model for non-Boltzmann behavior, although the disorder parameter is very close to unity. We obtained a fair agreement between experimental data and theoretical models for magnetoresistance in the presence of weak localization in the low H/T limit. With increasing H/T discrepancies appear. They are not solely attributable to the inadequacy of models for localization but can arise also from systematic errors in estimating the magnetoresistance due to electron-electron interaction. Indeed in this latter expression one finds the diffusion constant D and the screening parameter F that we have estimated in a free-electron model.

Despite these difficulties we proposed a method for the analysis of the magnetoresistance. By using it we were able to determine in a semiquantitative way the inelastic and spin-orbit scattering frequencies. The main interest of high magnetic fields is the possibility to deduce the spin-orbit scattering time, even when the spin-orbit coupling is strong, from experimental data in the moderate H/T regime.

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