

**Magnetic and transport properties of amorphous Tb-Si alloys near the metal-insulator transition**

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The magnetic and transport properties of amorphous  $\text{Tb}_x\text{Si}_{1-x}$  alloys for  $x$  near the metal-insulator transition ( $\approx 0.14$ ) have been studied as a function of temperature, magnetic field, and composition. Local Tb magnetic moments act to localize extended-state carriers at low temperatures, similarly to Gd, causing a sharp drop in conductivity as a function of temperature. The spin-glass freezing seen in amorphous Gd-Si alloys is drastically affected by the randomly oriented local anisotropy of Tb; amorphous Tb-Si alloys show a broad poorly defined susceptibility peak and a strong frequency dependence which is not seen in amorphous Gd-Si alloys. The magnetic exchange interactions are strong but balanced ferromagnetic and antiferromagnetic, as in amorphous Gd-Si alloys, with little dependence on  $x$  and no effect of the metal-insulator transition. The Tb effective moment in the paramagnetic state shows similar effects to Gd: a peak at the metal-insulator transition due to polarization of carriers and suppression below the expected value far from it.

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**I. INTRODUCTION**

The behavior of amorphous rare-earth-Si alloys ( $a$ -RE-Si) at the metal-insulator (M-I) transition is of significance as an example of the dramatic effects of introducing magnetic moments into a semiconducting material. Transport measurements on  $a$ -Gd-Si show strong localization of extended-state carriers caused by the presence of the Gd local moments and many orders of magnitude negative magnetoresistance.<sup>1,2</sup> This extremely large magnetoresistance (MR) has allowed tunneling spectroscopy studies of the electron density of states through the three-dimensional M-I transition on a single sample for the first time.<sup>3</sup> The localizing effects of the Gd local moments and the negative magnetoresistance occur below a characteristic temperature  $T^*$  which is of order 50–100 K. Magnetic studies show strong balanced ferromagnetic and antiferromagnetic exchange interactions which suppress the magnetization well below the noninteracting Brillouin function and lead to a spin-glass freezing at temperatures below 10 K (dependent on composition).<sup>4</sup> The effective moment  $p_{eff}$  in the paramagnetic state above the spin-glass freezing shows a surprising composition dependence, with a large peak at the M-I transition and a significant suppression away from this composition.<sup>4</sup> Optical properties show strong effects of the correlated electron behavior near the M-I transition, with nonspectral weight conserving shifts out to surprisingly high energy (1 eV).<sup>5</sup> All of these properties indicate the nontrivial effects of local magnetic moments on the correlated electron state found near the M-I transition. Theoretical studies of these materials to date indicate the importance of lattice polarons and of the strong effect of the spin disorder on the carrier wave functions via a local moment-carrier exchange interaction.<sup>6,7</sup>

To date, nearly all data have been taken on  $a$ -Gd-Si alloys. Gd is a  $4f^75d^16s^2$  atom, virtually always trivalent. The  $\text{Gd}^{3+}$  ion has a large moment  $J=S=7/2$  and  $L=0$  due to the half-filled  $f$  shell; hence single-ion anisotropy is negligible. Tb is a  $4f^85d^16s^2$  atom, also nearly always trivalent and with nearly identical ionic radius to Gd. The  $\text{Tb}^{3+}$  ion has

$S=3$ ,  $L=3$ ,  $J=6$ , hence also a large local moment but with strong local anisotropy (due to  $L\neq 0$ ) which is randomly oriented in the amorphous structure.

The enormous MR as well as the spin-glass freezing and strong inter-rare-earth magnetic interactions are believed to be related to an exchange interaction  $J_{sf}$  between the local rare earth magnetic moments and the conduction electrons. In metals such as crystalline Gd and  $\text{GdSi}_2$ ,  $J_{sf}$  results in an indirect (RKKY) exchange interaction  $J$  between the Gd ions with a magnitude proportional to  $J_{sf}^2$  and a sign which oscillates with distance between the ions.  $J$  is ferromagnetic for Gd (Curie temperature  $T_c=293$  K) and antiferromagnetic for  $\text{GdSi}_2$  (Néel temperature  $T_N=27$  K). In crystalline  $\text{TbSi}_2$ , the exchange interaction  $J$  is also antiferromagnetic with  $T_N=16$  K. These Néel temperatures scale with the ratio of the de Gennes factor  $(g-1)^2J(J+1)$  (Gd:Tb 15.75:10.5=1.5); one piece of evidence that RKKY is the exchange mechanism. Disorder alone does not change the nature of the interactions:  $a$ - $\text{Gd}_x\text{Ge}_{1-x}$  is ferromagnetic with  $T_c>150$  K for  $x>0.5$ .<sup>8</sup> Evidence from measurements on ternary  $a$ - $(\text{Gd}_y\text{Y}_{1-y})_x\text{Si}_{1-x}$  (Ref. 9) suggests that an indirect RKKY-like exchange mechanism is also the source of the strong magnetic interactions in  $a$ - $\text{Gd}_x\text{Si}_{1-x}$  for  $x<0.2$ , despite the localizing of the conduction electrons, as long as the localization length exceeds the inter-Gd distance. An indirect RKKY-like interaction in  $a$ - $\text{Tb}_x\text{Si}_{1-x}$  alloys is likely to be similar to that of  $a$ - $\text{Gd}_x\text{Si}_{1-x}$  alloys due to their similar atomic structure and ionic radius, with the strength expected to scale as the de Gennes factor. However, due to the nonzero  $L=3$ ,  $a$ - $\text{Tb}_x\text{Si}_{1-x}$  is expected to show the influence of random anisotropy on magnetic properties.

In  $a$ -Gd-Si, the conductivity depends on the presence of the magnetic Gd ion below a characteristic temperature  $T^*$ . Above this temperature, the conductivities of  $a$ -Gd-Si and its nonmagnetic analog  $a$ -Y-Si are nearly identical. Below it, the conductivity of  $a$ -Gd-Si plummets below that of  $a$ -Y-Si of similar composition and we find large effects of a magnetic field. One of the crucial unanswered questions is what physical property and what interaction determines  $T^*$ . In the crys-

talline dilute magnetic semiconductors, the energy scale is set by the polaronic binding energy, which scales with both the strength of the  $sd$  exchange in those materials and the Coulomb binding of an electron to the impurity donor ion. While this model cannot be directly applied to the present materials, due to their much higher concentration of electrons and consequently higher electron/local moment ratio, it might nonetheless be anticipated that  $T^*$  would similarly scale with  $J_{sf}$  and hence should not depend strongly on the rare earth; this would be consistent with the models presented in both Refs. 6 and 7.

In this paper, we present work on the transport and magnetic properties of  $a$ -Tb $_x$ Si $_{1-x}$  alloys. The purpose of the work is to determine (1) the dependence of the characteristic temperature  $T^*$  on the rare earth (RE) ion, (2) the composition dependence of  $p_{eff}$  in another rare-earth-Si system, (3) the strength of the Tb-Tb exchange interactions and if they scale as the de Gennes factor, as in simple RE metals, and (4) the effects of the random anisotropy of Tb on the magnetic properties such as the spin-glass state. We have therefore measured the temperature and field dependence of magnetization  $M(T,H)$ , low-field susceptibility  $\chi(T)$ , and the temperature dependence of the conductivity  $\sigma(T)$  for  $a$ -Tb $_x$ Si $_{1-x}$  alloys as a function of Tb concentration  $x$ .

## II. EXPERIMENTAL RESULTS

Samples are made by electron beam coevaporation under UHV conditions onto amorphous Si-N-coated Si substrates held near room temperature. Compositions and Tb and Gd concentrations (atoms/cm<sup>2</sup>) were determined by Rutherford backscattering (RBS). The thickness is 4000–5000 Å, measured by Dektak and confirmed by RBS. ac and dc susceptibility ( $\chi_{ac}$  and  $\chi_{dc}$ ) and magnetization  $M(H,T)$  were measured using a superconducting quantum interference device (SQUID) magnetometer.

Figure 1 shows  $\chi$  versus temperature  $T$  (1.8–300 K) for  $a$ -Tb $_x$ Si $_{1-x}$  for  $x=0.133$ .  $\chi_{dc}$  is defined as  $M/H$  and was measured at 100 Oe; data are shown for zero-field-cooled (ZFC) and field-cooled (FC) states.  $\chi_{ac}$  was measured in an ac field of 5 Oe (in zero dc field). A temperature-independent constant  $b$  due to core, substrate, and SQUID magnetometer background contributions has been subtracted from the raw data. Comparing these data with the data of  $a$ -Gd $_x$ Si $_{1-x}$ ,<sup>4</sup> the difference is striking. The  $a$ -Gd $_x$ Si $_{1-x}$  data indicate a classic spin glass, with a sharp peak in  $\chi_{ac}$  at a freezing temperature  $T_f$  and a small but significant shift in  $\chi_{dc}(T)$  below  $T_f$ . The  $a$ -Tb-Si data show a broad peak in  $\chi_{ac}$  and a large difference between  $\chi_{ac}$  and  $\chi_{dc}$  below this broad peak.

Figure 2 shows  $\chi_{ac}$  of  $a$ -Tb $_{0.103}$ Si $_{0.897}$  on an expanded scale for different measurement frequencies (15–950 Hz). The peak height increases and shifts to lower temperatures with decreasing frequency, consistent with previous results on  $a$ -Gd-Si alloys and with classical spin glasses. However, in  $a$ -Tb-Si, the peaks in  $\chi_{ac}$  are very broad, with a poorly defined freezing temperature  $T_f$ , and the height is strongly frequency dependent. By contrast, in earlier work on  $a$ -Gd-Si films,  $\chi_{ac}$  was shown to be quite sharp with a small but characteristic dependence of peak height and position with

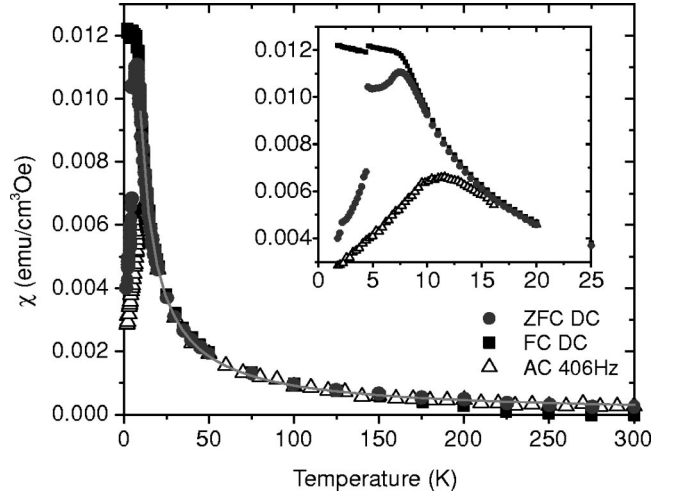


FIG. 1.  $\chi_{dc}$  and  $\chi_{ac}$  for Tb $_{0.133}$ Si $_{0.867}$ .  $\chi_{dc}$  was measured in 100 Oe, applied either after cooling (zero field cooled) or at 300 K (field cooled).  $\chi_{ac}$  was measured in a 406 Hz, 5 Oe ac magnetic field. The line is a Curie-Weiss law fit above  $T_f$ . Inset: same data on an expanded scale.

frequency. The shift in freezing temperature  $\Delta T_f/T_f$  per decade of frequency for  $a$ -Gd-Si was shown to be 0.04, similar to the classic spin glasses, and the increase in peak height with decreasing frequency is of order 1%.<sup>4</sup> For  $a$ -Tb-Si,  $\Delta T_f/T_f$  is only slightly larger (0.067), but more importantly,  $T_f$  is poorly defined and the shift in peak height with decreasing frequency is enormous ( $\sim 33\%$ ). This frequency dependence explains the large difference between  $\chi_{dc}$  and  $\chi_{ac}$  for  $a$ -Tb-Si (Fig. 1), which is not seen in  $a$ -Gd-Si.<sup>4</sup> This strong frequency dependence below the nominal freezing temperature continues into time scales normally considered dc, as evidenced by the discontinuity seen in  $\chi_{dc}$  at 4.6 K; at this temperature the SQUID magnetometer takes nearly 15 min to stabilize temperature, compared to 2–3 min at other temperatures. During that 15 min, the  $a$ -Tb-Si ZFC magnetization in 100 Oe relaxes appreciably to the FC value; this relatively rapid (minutes) relaxation below the freezing tem-

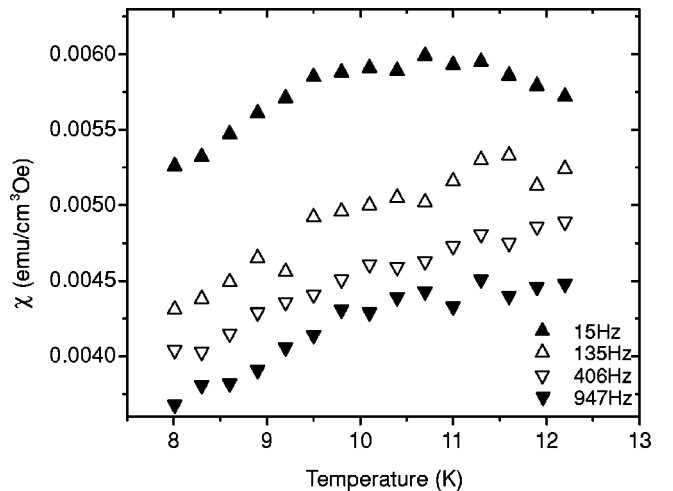


FIG. 2.  $\chi_{ac}$  vs  $T$  for  $a$ -Tb $_{0.103}$ Si $_{0.897}$  at different frequencies in a 5 Oe field.

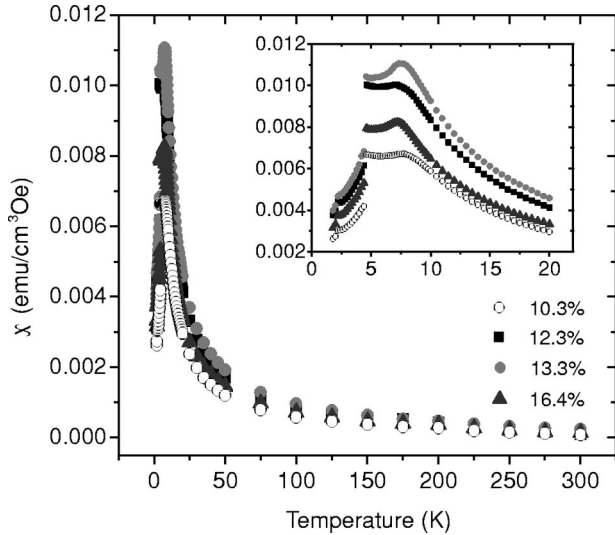


FIG. 3.  $\chi_{dc}$  vs  $T$  for  $x=0.103$ – $0.164$  measured at 100 Oe after cooling to 2 K in zero field. The data below the freezing temperature are strongly time dependent. Inset: same data on an expanded scale.

perature is not seen in  $a$ -Gd-Si.

Figure 3 shows  $\chi_{dc}$  versus  $T$  (ZFC) for four samples of  $a$ -Tb-Si of different composition. All samples show a peak in the susceptibility indicative of some type of magnetic freezing, but below the peak show a strong time dependence, including the discontinuity at 4.6 K discussed above. The freezing temperature, defined as the peak in the susceptibility, depends very little on composition. The data above the magnetic freezing temperature are independent of measurement frequency for all samples.

Most significantly, as found in  $a$ -Gd-Si, the susceptibility in the paramagnetic state above the freezing does not increase monotonically with composition as would be expected. In particular,  $\chi$  for  $x=0.133$  (near the M-I transition) has higher susceptibility than  $\chi$  for  $x=0.164$ . Data for the paramagnetic state for all samples were fit to a Curie-Weiss law  $\chi=A/(T-\theta)$ . In this expression, the Curie-Weiss constant  $A=np_{eff}^2\mu_B^2/3k_B$ , where  $p_{eff}$  is the effective moment,  $\theta$  is their average net interaction, and  $n$  is the number of Tb atoms/cm<sup>3</sup>, determined by RBS. (Note that RBS determines atoms/cm<sup>2</sup>, which is divided by thickness to obtain atoms/cm<sup>3</sup>; however, this same thickness is then used to calculate the sample volume. Thickness is therefore canceled out of this calculation of  $p_{eff}$  from  $\chi$  and only sample area plus atoms/cm<sup>2</sup> from RBS is required.) The composition and  $n$  are shown in Table I.  $\theta$  for Tb-Si sample is less than 0.4 K; it was therefore simply taken as zero in the fits. For  $a$ -Gd-Si,  $\theta$  ranged from 0 to a maximum of 2.5 K, small but significantly larger than  $a$ -Tb-Si.

The effective moment in the paramagnetic state  $p_{eff}$  shows a strong dependence on composition, as was seen in  $a$ -Gd-Si, and is less than the expected value for all  $x$ . For Tb<sup>3+</sup>,  $L=S=3$ ,  $J=6$  moments,  $p_{eff}=g[J(J+1)]^{1/2}=9.74$ . The magnitude of  $A$  and the effective moment  $p_{eff}$  for  $a$ -Gd <sub>$x$</sub> Si<sub>1- $x$</sub>  and  $a$ -Tb <sub>$x$</sub> Si<sub>1- $x$</sub>  are plotted in Fig. 4.  $p_{eff}$  is largest for samples near the M-I transition ( $\sim 12$

TABLE I. The composition  $x$  in  $a$ -Tb <sub>$x$</sub> Si<sub>1- $x$</sub>  and the number of Tb atoms/cm<sup>3</sup> determined by RBS ( $\theta$  was less than 0.4 K and was therefore taken to be zero for all compositions).

Composition	$n_{Tb}$ (Tb atoms/cm <sup>3</sup> )
0.103	$4.856 \times 10^{21}$
0.123	$5.732 \times 10^{21}$
0.133	$6.164 \times 10^{21}$
0.164	$7.471 \times 10^{21}$

– 14 at% RE) and drops significantly when samples are strongly insulating or metallic. The values of  $p_{eff}$  for the expected Tb<sup>3+</sup> and Gd<sup>3+</sup> are shown with arrows in Fig. 4(b).

Figure 5 shows the freezing temperature  $T_f$  for  $a$ -Gd <sub>$x$</sub> Si<sub>1- $x$</sub>  and  $a$ -Tb <sub>$x$</sub> Si<sub>1- $x$</sub>  (defined as the peak in  $\chi_{ac}$ , as measured at either 135 Hz or 406 Hz; the shifts with measurement frequency are small on this scale). Although  $T_f$  for

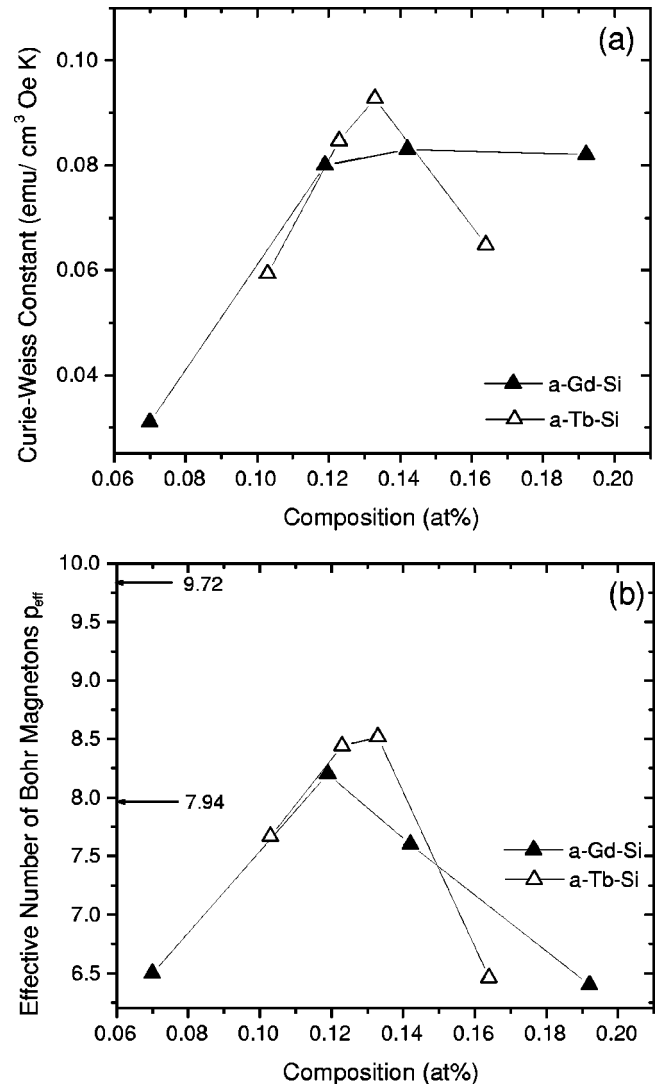


FIG. 4. The Curie-Weiss constant  $A$  and effective number of Bohr magnetons  $p_{eff}$  for  $a$ -Tb-Si and  $a$ -Gd-Si of different composition.

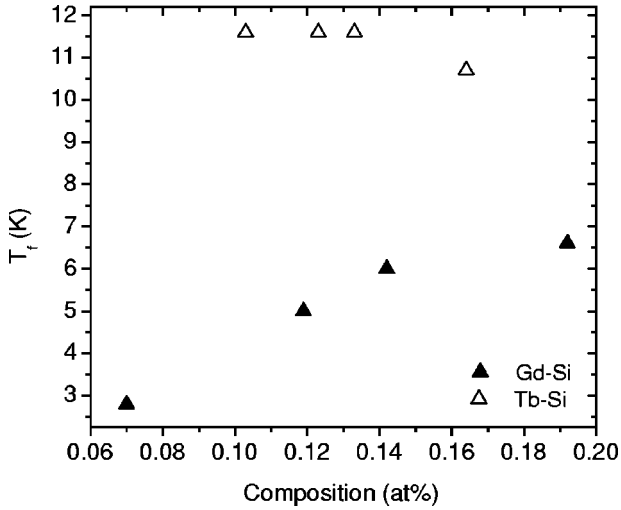


FIG. 5. Freezing temperature  $T_f$  vs composition for  $a$ -Tb-Si and  $a$ -Gd-Si.  $T_f$  is defined from the peak in  $\chi_{ac}$  at either 406 or 135 Hz.

$a$ -Tb-Si alloys is poorly defined (due to the breadth of the peaks), it is clear that the magnetic freezing occurs at much higher temperatures than for  $a$ -Gd-Si alloys; it is also not significantly dependent on composition. These are indications of the crucial role played by the local random magnetic anisotropy  $L \neq 0$  in the freezing as will be further discussed below.

Figure 6 shows the high-field data  $M(H)$  for  $Tb_{0.123}Si_{0.877}$ , at several temperatures. The small background magnetization  $M(H)$  measured at 300 K is subtracted from all data.  $M(H)$  does not saturate in fields up to 50 kOe even at 2 K and values are significantly below the expected saturation magnetization. These results are identical to what was seen in  $a$ -Gd-Si and indicate the presence of strong antiferromagnetic interactions.

Figure 7 shows  $M(H)$  at 4 K for several  $a$ -Tb-Si samples normalized to the expected saturation value ( $M_s = ngJ\mu_B$ ), along with a  $Gd_{0.14}Si_{0.86}$  sample. For the  $a$ -Tb-Si alloys, all the data coincide, showing an independence of composition

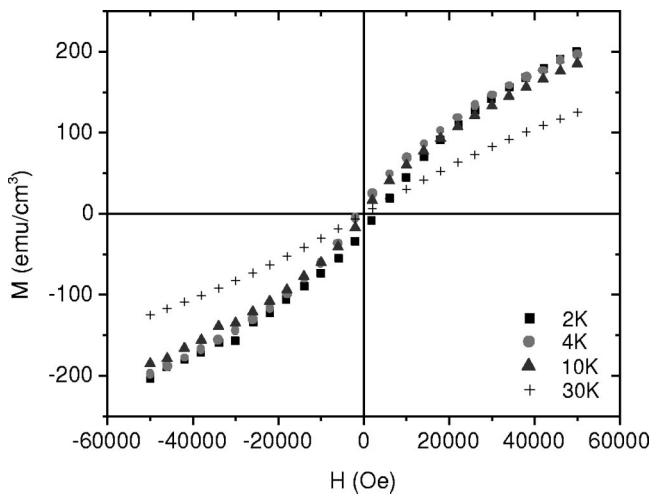


FIG. 6.  $M(H, T)$  for  $a$ - $Tb_{0.123}Si_{0.877}$  for various temperatures from 2 to 30 K.

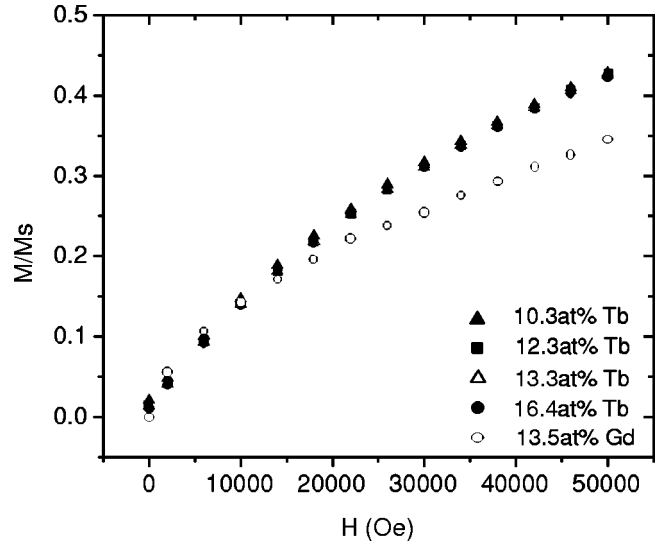


FIG. 7. Normalized magnetization vs field for  $a$ -Tb-Si of different composition and for  $a$ - $Gd_{0.135}Si_{0.865}$  at 4 K.  $M_s$  for the 4  $a$ -Tb-Si samples shown is 405, 478, 514, and 623  $emu/cm^3$  and 403 for the Gd sample, assuming trivalent  $Tb^{3+}$  and  $Gd^{3+}$  and RBS-determined concentrations shown in Table I.

at these relatively high fields.  $M(H)/M_s$  is larger for the  $a$ -Tb-Si samples than for  $a$ -Gd-Si samples, which initially suggests weaker anti-ferromagnetic interactions [ $M(H)$  closer to the noninteracting Brillouin function]. However, an examination of the Brillouin functions for  $Tb^{3+}$  and  $Gd^{3+}$  shows that these observed  $M(H)$  data are in fact nearly exactly equivalently suppressed below the noninteracting  $M(H)$  values. To make a crude estimation of the strength of interaction, the observed  $M(H)/M_s$  at 50 kOe and 4 K requires an equivalent field of 2780 Oe and 2825 Oe for  $a$ -Tb-Si and  $a$ - $Gd_{0.135}Si_{0.865}$  which means an antiferromagnetic interaction strength of order 47 220 Oe and 47 175 Oe, respectively, virtually identical numbers. The exchange interaction was expected to be proportional to the de Gennes factor  $(g - 1)^2 J(J + 1)$  where  $g$  is the Lande factor and  $J$  the total angular momentum. The difference of the de Gennes factor for  $a$ -Tb-Si and  $a$ -Gd-Si is more than 33%, while the difference of the interaction strength estimated from the  $M(H)$  data is negligible. We conclude therefore that the interaction strength is not scaling with the de Gennes factor.

Figure 8 shows the conductivity  $\sigma$  versus temperature for  $a$ -Tb-Si and  $a$ -Gd-Si samples of various compositions. Conductivities were measured by Van der Pauw four-point-probe measurement from 4 K to room temperature. For  $a$ -Gd-Si, the M-I transition occurs between 14 and 15 at. % Gd.<sup>3</sup> From the present data, the M-I transition for  $a$ -Tb-Si occurs between 13.3 and 16.4 at. % Tb, and (based on the high value of  $\sigma$  at 2 K for the 16.4 at. % sample) is likely to be close to 14 at. % Tb, as for  $a$ -Gd-Si samples. Again we see a characteristic temperature  $T^*$  below which the conductivity drops sharply; qualitatively, this temperature does not appear different for  $a$ -Gd-Si compared to  $a$ -Tb-Si.

### III. DISCUSSION

A comparison of magnetization and conductivity properties shows that  $a$ -Tb-Si has very similar behavior to  $a$ -Gd-Si,



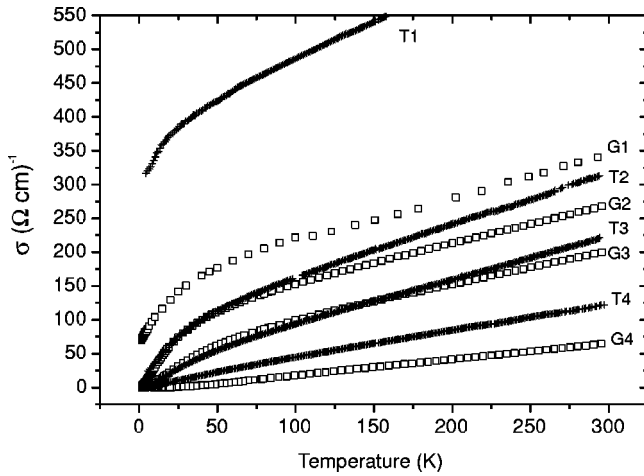


FIG. 8. The conductivity  $\sigma$  vs temperature for  $a\text{-Tb}_x\text{Si}_{1-x}$  ( $x = 0.164, 0.133, 0.123, \text{ and } 0.102$  for T1, T2, T3, and T4, respectively) and  $a\text{-Gd}_x\text{Si}_{1-x}$  ( $x = 0.15, 0.14, 0.13, \text{ and } 0.11$  for G1, G2, G3, and G4, respectively).

with similarly large effects of local moments on low-temperature transport (below  $T^*$ ) and qualitatively similar magnetic properties. High-field  $M(H, T)$  show a strong suppression of magnetization below the noninteracting Brillouin function and a large high-field susceptibility, indicative of strong magnetic interactions. Local random anisotropy due to  $L \neq 0$  RE ions such as Tb suppresses the magnetization compared to alloys containing  $L = 0$  Gd ions in many RE alloys.<sup>15</sup> The fact that the normalized magnetization of  $a\text{-Tb-Si}$  is comparable at a given  $T$  and  $H$  to that of  $a\text{-Gd-Si}$  (both are similarly reduced compared to the noninteracting Brillouin function) suggests that the Tb-Tb interactions are comparable to the Gd-Gd interactions in  $a\text{-Gd-Si}$ , inconsistent with the expected scaling with the de Gennes factor described in the Introduction. The susceptibility in the paramagnetic state shows a very low net magnetic interaction (Curie-Weiss constant  $\theta < 0.3$  K), slightly smaller than in  $a\text{-Gd-Si}$ .  $\theta$  is a rough measure of the average strength of the interactions, suggesting that in both  $a\text{-Tb-Si}$  and  $a\text{-Gd-Si}$ , antiferromagnetic interactions are nearly perfectly balanced by ferromagnetic interactions leading to  $\theta \ll T_f$  and much less than the absolute value of the interactions which are greater than 29 K ( $> 47$  kOe for  $J = 6$ ).

The Tb effective moment (from the susceptibility in the paramagnetic state) shows a strong composition dependence with a peak at the M-I transition, exactly as seen in  $a\text{-Gd-Si}$ . The magnitude of the susceptibility in the paramagnetic state of all other materials with rare earth moments we are aware of increases monotonically as a function of magnetic moment concentration.<sup>15</sup> Nonmonotonic dependence occurs only when nontrivial effects such as many body correlations become important, such as at the metal-insulator transition in crystalline P:Si and similarly doped semiconductors. In these systems, the magnetic moment of the material develops out of the magnetic polarization of conduction electrons in singly occupied Coulomb states near the Fermi energy.<sup>10</sup> In P:Si, the formation and interaction of these magnetic moments results in a susceptibility which has a peak at the M-I

transition, much as we see here, and a characteristic temperature dependence  $\chi$  proportional to  $T^\alpha$  where  $\alpha$  ranges from 1 in the insulating low-doping limit (Curie law) to 0 in the metallic high-doping limit (Pauli susceptibility).<sup>14</sup> In P:Si and related materials, the susceptibility is comparatively small; the only moments are related to electron states near the Fermi energy with  $S = 1/2$ , and the overall electron concentration is of order  $10^{18} \text{ cm}^{-3}$ , much smaller than in the present materials. In the present magnetically doped semiconductors, there are both local rare earth moments and localizing conduction electron moments which interact with the local rare earth moments. The susceptibility is large, the change in effective moment is large, due in part to the increased moment per ion, and the temperature dependence is Curie like ( $\alpha = 1$ ) for all concentrations. The interacting rare earth magnetic moments are close together and hence do not form the singlet states which are the essence of the Bhatt-Lee model for P:Si, instead interacting via an RKKY-like interaction. The Bhatt-Lee model should therefore not be expected to apply in a quantitative way. Nonetheless, we suggest that part of the model applies: specifically that singly occupied states occur near the Fermi energy and are polarized by proximity to the rare earth ions, analogous to the interactions which give rise to the susceptibility peak in P:Si at the M-I transition.

Far from the M-I transition, the reduced value of  $p_{eff}$  we suggest could be due to polarization of Si local dangling bond states surrounding the Gd or Tb and antiferromagnetically coupled to the RE. To the best of our knowledge, there is no direct evidence for this suggestion, but the presence of dangling bond states and their magnetization in an applied field has been seen in  $a\text{-Si}$ .<sup>11</sup> Local density functional simulations of  $a\text{-Y-Si}$  suggests that Y atoms are surrounded by cages of dangling bond states.<sup>12</sup> X-ray absorption fine structure (XAFS) analysis on  $a\text{-Gd-Si}$  supports the structural model of rare earth ions surrounded by a Si cage, but show no dependence of Si coordination number on rare earth content, which argues against low coordination dangling bond states surrounding the rare earth ions.<sup>13</sup> Whether there are magnetic states associated with the Si bonds surrounding each rare earth ion is a question which could be addressed directly with electron spin resonance (ESR) measurements.

The most significant difference between  $a\text{-Gd-Si}$  and  $a\text{-Tb-Si}$  is seen in the spin freezing. Amorphous Gd-Si shows nearly classic spin-glass properties in its temperature and field dependence (and in its near-perfect balancing of interactions  $\theta \approx 0$ ).<sup>4</sup> By contrast, for  $a\text{-Tb-Si}$ , the peak in the magnetic susceptibility is much broader, strongly frequency dependent, and not dependent on composition. The freezing temperature  $T_f$  is also significantly higher for  $a\text{-Tb-Si}$  alloys than for  $a\text{-Gd-Si}$  alloys. It seems likely that the strong random local anisotropy of Tb due to the nonzero  $L$  is the cause of these differences. Random anisotropy could lead to the observed strong frequency dependence and relaxation in the nominally frozen state below the freezing of  $a\text{-Tb-Si}$ ; the freezing in this material, unlike  $a\text{-Gd-Si}$ , is associated with single-ion anisotropy effects rather than a cooperative exchange as in a spin glass. In this model, the true freezing associated with exchange does not occur until a lower tem-

perature, but this effect is obscured by a single-ion anisotropy-driven blocking of the paramagnetic response. Broadened susceptibility peaks in  $L \neq 0$  rare earth systems have been seen before (e.g., Dy-Cu), but to the best of our knowledge, there is no discussion of an increased frequency dependence.<sup>15</sup> Measurements of the time dependence of magnetization at low temperatures have been used extensively to develop models for spin glasses; such measurements on  $L \neq 0$  rare earth systems would allow testing of the effects of random anisotropy.

The zero-field conductivity  $\sigma(T)$  shows little difference between Tb and Gd. Previous work on low-temperature conductivity (below 4 K) has been used to quantitatively analyze the M-I transition and the effect of a magnetic field in  $a$ -Gd-Si alloys.<sup>2,3,16</sup> The purpose of the present work is to look at the higher-temperature conductivity and in particular, to attempt to answer the crucial question of what physical property causes the temperature  $T^*$  below which the magnetic moment of the rare earth ion causes an increased localization of carriers and enormous negative magnetoresistance. We have found it difficult in the absence of a quantitative model for the conductivity at higher temperatures to define precisely a value for  $T^*$ . We were, for example, unable to find a consistent definition of  $T^*$  for  $a$ -Gd-Si alloys which did not vary with composition  $x$ , despite the fact that by eye there is a sharp break in  $\sigma(T)$ . Nevertheless, by comparing samples of similar conductivity (e.g., G1, T2, and T1 in Fig. 8), a qualitative definition of  $T^*$  was chosen as the temperature where the conductivity of the Tb-Si or Gd-Si drops sharply below that of the comparable nonmagnetic Y-Si

sample. Using this definition of  $T^*$ , which depends on concentration  $x$ ,  $T^*$  does not vary significantly between Tb-Si and Gd-Si, contrary to our own suggestion in the earliest work on this subject.<sup>1</sup> By contrast, preliminary measurements on amorphous Gd-Ge alloys suggest a strong dependence of  $T^*$  on the semiconductor.<sup>17</sup>

To summarize, we have shown that (1) the characteristic temperature below which the conductivity is strongly affected by the local moments does not appear to depend strongly on magnetic ion (Tb vs Gd), (2) the effective moment  $p_{eff}$  in the amorphous Tb-Si system shows the same dependence on composition as it did in the amorphous Gd-Si system, with a strong peak at the M-I transition and significant suppression below the expected  $Tb^{3+}$  value for both metallic and insulating samples, (3) the strengths of exchange interactions are comparable for amorphous Tb-Si and amorphous Gd-Si, in contrast to the different de Gennes factor, but are still nearly perfectly balanced ferromagnetic and antiferromagnetic, and (4) the random anisotropy of the  $L = 0$  Tb ion has a strong effect on the magnetic freezing, causing a poorly defined and strongly frequency-dependent transition more reminiscent of a blocking temperature than the spin-glass freezing seen in amorphous Gd-Si.

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