

High Field Magneto-Optic Measurements of Amorphous Gd-Si Alloys

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Abstract

The temperature and magnetic field dependence of the magneto-optic Kerr effect was used to determine the temperature and field dependence of the magnetization of giant negative magnetoresistance amorphous $\text{Gd}_x\text{Si}_{1-x}$ films at fields up to 25T. Amorphous $\text{Gd}_x\text{Si}_{1-x}$ has been previously shown to exhibit spin glass freezing and suppression of the magnetization well below the non-interacting Brillouin function for Gd^{3+} at fields to 6T at 4K, indicative of strong mixed ferromagnetic and antiferromagnetic interactions. The Kerr rotation was found to be proportional to the directly measured magnetization at low fields. The high field Kerr data show that the magnetization is not saturated even at 25T and 4K, reaching a value of approximately 0.6 of the expected saturation magnetization value, indicating interactions of strength greater than 25T. Kerr ellipticity was small.

Introduction of magnetic moments into semiconducting materials produces large and not yet fully understood effects. Amorphous $\text{Gd}_x\text{Si}_{1-x}$ alloys possess enormous (many orders of magnitude) negative magnetoresistance for $x \sim 0.1-0.15$, near the metal-insulator (M-I) transition.[1,2] Transport and tunneling spectroscopy (proportional to the electronic density of states near the Fermi energy) data has been taken to 20T (33T in some cases), including data through a magnetic field driven M-I transition for the critical concentration $x = 0.14$. [3,4] These properties show extremely strong magnetic field dependence, with no indication of saturation up to 20T. By contrast, preliminary data on the anomalous Hall effect show saturation near 10T.[5] The magnetoresistance is nearly certainly due to a strong s - f exchange interaction between the carriers and local Gd moments which localizes the carriers when Gd moments are randomly oriented in zero field. With increasing field, the Gd moments align, which we have suggested causes a shift downward of the mobility edge relative to the Fermi energy and a consequent delocalization of carriers, due to the reduced exchange disorder. Gd moment alignment is expected to be proportional to the magnetization normalized to the saturation value $M(H)/M_s$; characterization of the temperature and field dependent magnetization $M(T, H)$ and knowledge of the saturation value of magnetization M_s are thus essential to even a qualitative explanation of the magnetoresistance and magnetically-driven shifts in the electronic density of states. Quantitative theories of the magnetoresistance involving magnetic polarons,[6] lattice polarons,[7] or shifts in the mobility edge [8] have been proposed; tests of these also require knowledge of $M(H, T)$.

Previous magnetization measurements in a SQUID magnetometer between 0 and 6T have shown spin glass freezing at a temperature T_f approximately 5-10K (dependent on composition x) and suppression of M well below the Brillouin function for non-interacting moments at 2K in 6T, with a large high field susceptibility.[9] Curie-Weiss fits to the low field susceptibility χ (both AC and DC) above T_f show a Curie-Weiss constant θ near 0K ($< 2.5\text{K}$ for all x) and an effective moment p_{eff} with an unexpectedly low value for Gd and a non-trivial dependence on composition x . The suppression of magnetization is indicative of strong antiferromagnetic interactions; the near-zero value of Curie-Weiss constant θ indicates that these antiferromagnetic interactions are balanced by equally strong ferromagnetic interactions. Gd is usually an extremely simple local moment, virtually always trivalent, with $p_{\text{eff}} = g\sqrt{J(J+1)} = 7.9 \mu_B$. In these amorphous Gd-Si alloys with x near the M-I transition, however, the moment is suppressed far from the M-I transition to values as low as $5.5 \mu_B$ and shows a large peak at the M-I transition. The peak at the M-I transition has been suggested to be related to polarization of the carriers at the M-I transition, as seen in P:Si [10]; we suggest here that it is possible that far from the M-I transition, a reduced moment may be the result of polarization of Si dangling bonds surrounding the Gd atoms. The effect of the M-I transition on p_{eff} has recently been confirmed by

experiments on amorphous Tb-Si alloys and by adding non-magnetic Y to Gd-Si alloys, which adds conduction electrons without changing the Gd moment concentration.[11,12]

Thus the magnetization data taken to date show remarkable magnetic phenomena related to the M-I transition. They indicate strong mixed ferromagnetic/antiferromagnetic Gd-Gd interactions which are likely to be an indirect exchange interaction, mediated by the conduction electrons, despite their localization at the metal-insulator transition (note that the localization length can enormously exceed the Gd-Gd distance).[9] They also indicate a non-trivial dependence of the effective magnetic moment in the paramagnetic state on composition around the M-I transition, related to effects seen in crystalline P:Si.

Measurement of $M(H)$ to high field ($>6T$) is therefore necessary to permit an analysis of the high field transport and tunneling data, and for better characterization of the strength of the interactions and the value of the saturation magnetization M_s . Characterization of M_s versus x may also be of help in understanding p_{eff} versus x .

Samples are made by vapor deposition techniques, limiting the sample volume to approximately 10^{-4} cm^3 , ruling out high field VSM measurements due to small signals. Instead, we used the Magneto-Optic Kerr effect (MOKE) to measure $M(H, T)$. The polar Kerr rotation in many materials is proportional to the alignment of the magnetic moments along the film normal, with magnetic field applied normal to the sample. For the rare earth elements such as Gd, the Kerr rotation peaks in the ultraviolet, but still has a significant rotation at the wavelength of a HeNe laser (2.5-3 minutes of arc at 1.96 eV at 105K) [13]. MOKE has for example been used to study the magnetic properties of thin epitaxial Gd films.[14] In the present case, measurements were performed at the National High Magnetic Field Lab in Tallahassee in fields to 25T using a polarization modulated ellipsometer built on a high field probe (Mok3). This ellipsometer is based on a HeNe laser and a Hinds photo-elastic modulator [15]. 4000Å-thick samples of a -Gd-Si and the non-magnetic analog a -Y-Si on amorphous SiN-covered Si substrates were mounted with blade springs and thermal vacuum grease on the sample stage. The probe was cooled down in a He atmosphere to 4.5K, after which the gas was evacuated. The laser signal impinges at perpendicular incidence on the sample and is reflected back up through the probe, through the photoelastic modulator and a polarization analyzer. The intensity is detected by a photodetector; the amplitude of the signal is input to two lock-in amplifiers. In this technique, the signal at the modulator frequency ω is proportional to the polar (perpendicular) Kerr ellipticity and the signal at two ω is proportional to the polar Kerr rotation. The DC component of the signal is proportional to the laser intensity. Prior to the measurements, the angle of the polarization analyzer was adjusted to minimize the two- ω signal. This guarantees that the angle between the polarizer and analyzer is the exactly the same for each measurement ($=45$ degrees) and thus the calibration factor can be considered constant [16]. In order to compensate for possible drift of the laser intensity, all data were divided by the measured intensity of the laser, a DC-signal. In order to remove the residual background signal originating from the substrate, the amorphous

Si background, and the optics, data measured on the non-magnetic $a\text{-Y}_{0.12}\text{Si}_{0.88}$ sample were subtracted from the $a\text{-Gd}_x\text{Si}_{1-x}$ data at each temperature. The residual zero field offset in the two-omega signal was subtracted from each set of data. A known Kerr rotation material (CoSm) was measured as a test of the equipment.

All samples were prepared by electron beam co-evaporation in a UHV deposition system. Compositions and total number of atoms were determined by Rutherford Backscattering analysis. Further details of sample preparation and characterization may be found in refs 1-4. Data were taken as a function of magnetic field between + and - 25T at fixed temperatures (4.5K, 10K, 20K, 30K, and 60K) for $x=0.12$, 0.14, and 0.19. The temperature varied by approximately $\pm 0.5\text{K}$ throughout each measurement, due to heating as the magnetic field is ramped from $\pm 25\text{T}$ to 0 and back. The Kerr ellipticity signal of all samples was found to be much smaller than the Kerr rotation signal. Kerr rotation for $a\text{-Y-Si}$ ($x=0.12$) was subtracted from the measured values, which are then normalized at $H = 6\text{T}$ and $T=4\text{K}$ to the known Gd-Si sample magnetization M (in emu/cm^3) for $x=0.14$ at 6T, 4K from previous SQUID magnetometer measurements. Data at all other temperatures and fields and for the other two compositions measured are then scaled with this single normalization factor. Figure 1 shows this normalized Kerr rotation data for the $a\text{-Gd}_x\text{Si}_{1-x}$ sample with $x=0.14$. The agreement shown between SQUID $M(H)$ and MOKE rotation data at fields below 6T for temperatures from 4K to 60K is very good and shows that the Kerr rotation is indeed proportional to the alignment of Gd moments $M(H)/M_s$.

Figure 2 shows similar normalized Kerr rotation data for the three $a\text{-Gd}_x\text{Si}_{1-x}$ samples ($x=0.12$, 0.14, 0.19) at 4.5K, after subtracting the $a\text{-Y-Si}$ data. A single normalization factor (the same as used for Fig. 1) is used to convert Kerr rotation to M for all three samples. The data taken on $x=0.12$ and 0.19 samples were taken earlier in the experiment and are not as clean as the $x=0.14$ data (there are for example drifts in the measurement particularly for the $x=0.19$ sample, leading to offsets at $H=0$ on cycling the field and causing increased uncertainty in the high field susceptibility), which were avoided later in the experiment), but the qualitative trend is very clear and also indicates a lack of magnetic saturation at 25T, indicative of extremely strong antiferromagnetic interactions. The value found at 6T from the MOKE for the $x=0.12$ sample agrees well (within 10%) with that measured directly by SQUID magnetometer.

The data shown in Figs. 1 and 2 indicate that magnetization is not saturated even at 25T. Assuming a saturation magnetization corresponding to Gd^{3+} with $J=7/2$, the reduced magnetization M/M_s at 25T is approximately 0.6, where the non-interacting Brillouin function would give a value of 0.9999 for 25T and 5K. Based on this data, the strength of antiferromagnetic Gd-Gd interactions suppressing M must exceed 25T (equivalent to 120K assuming $J=7/2$ for Gd^{3+}). There must also be equally strong ferromagnetic interactions in order to give the near zero net interaction strength measured by the low field susceptibility (Curie-Weiss θ near zero). The lack of saturation makes it impossible to determine the saturation magnetization M_s , which will require yet higher fields to determine.

The large high field susceptibility and suppression below saturation magnetization at 25T are consistent with the lack of saturation seen previously in resistivity and tunneling data, but not consistent with preliminary analysis of the anomalous Hall effect data which suggested a saturation of M by 10T. Further analysis of this transport and tunneling spectroscopy data will be made.

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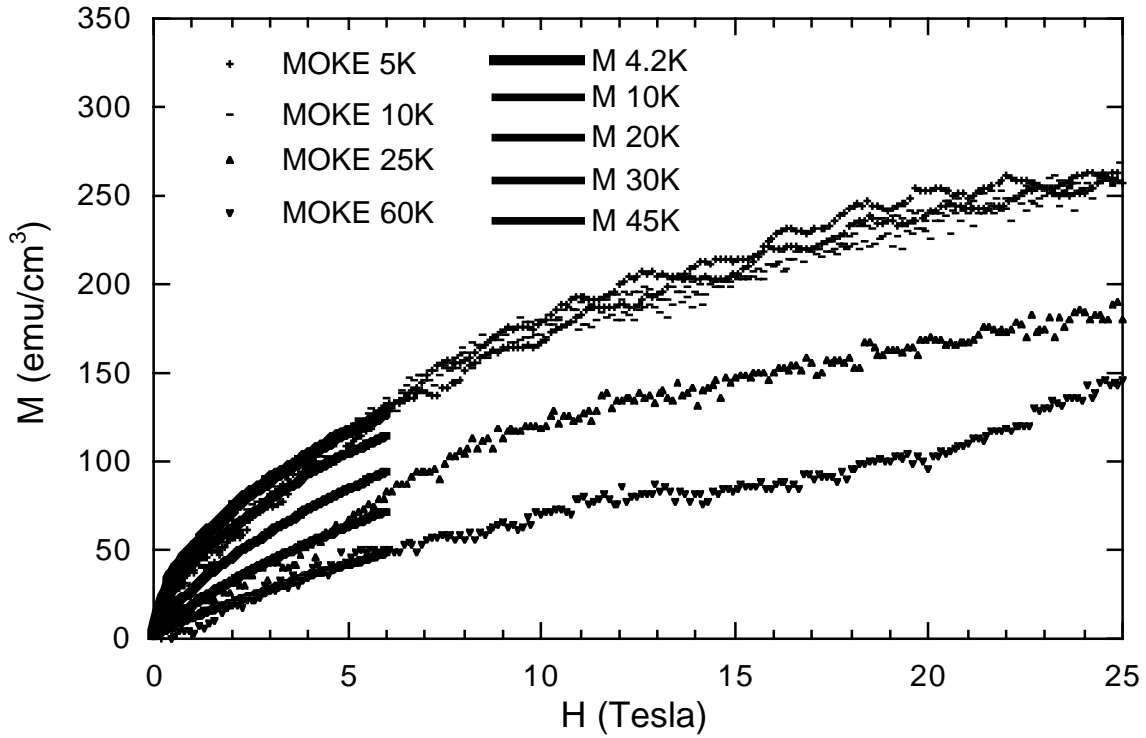


Figure 1 Dependence of normalized polar Kerr rotation on temperature and magnetic field for a - $\text{Gd}_x\text{Si}_{1-x}$ for $x=0.14$ after subtracting offset at $H=0$ and a -Y-Si rotation. MOKE data is normalized at $H=6\text{T}$ and 5K to match SQUID magnetometer-determined $M(H=6\text{T}, 4\text{K})$ in emu/cm^3 . SQUID magnetization data M shown as solid lines (to 6T); MOKE normalized data shown with symbols (to 25T). There is no hysteresis in $M(H,T)$ above 0.1T .

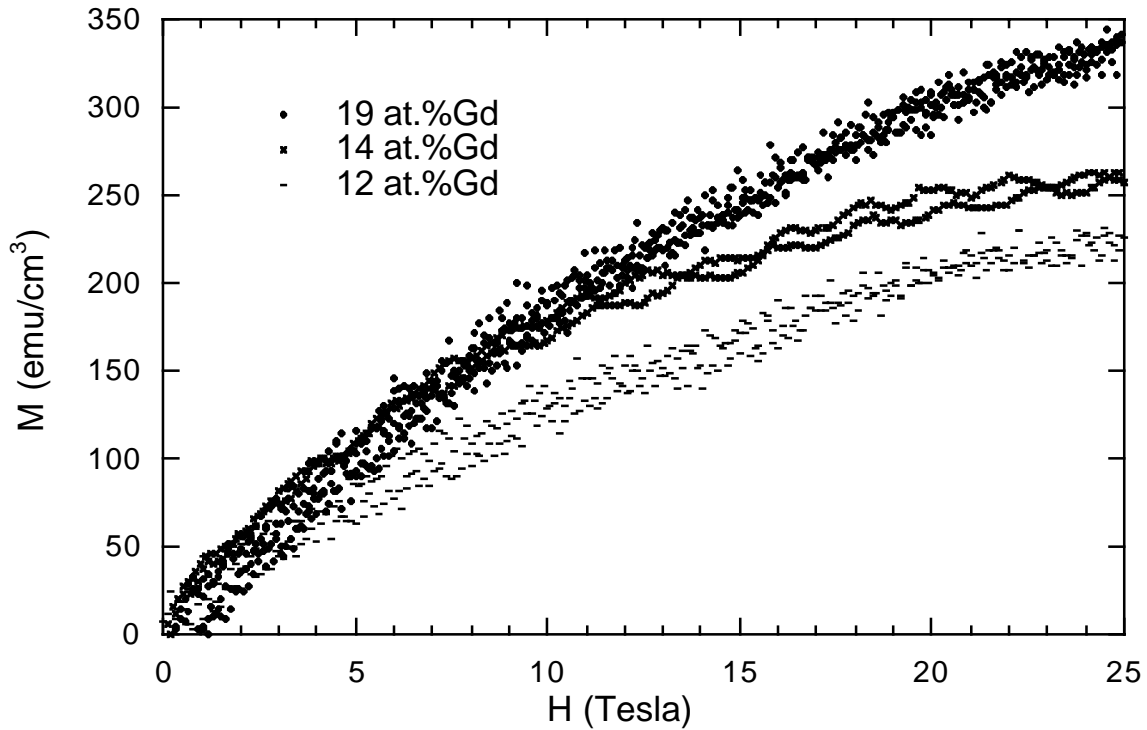


Figure 2 Normalized polar Kerr rotation for $x=0.12$, 0.14 , and 0.19 at 4.5K after subtracting offset at $H=0$ and $a\text{-Y-Si}$ rotation and normalizing to give M in emu/cm^3 using the same constant determined from $x = 0.14$ at 6T and 4.5K . The uncertainty in the $x=0.19$ data is larger than the others due to drifts in background. The expected saturation magnetizations are 370 , 443 , and $618 \text{ emu}/\text{cm}^3$ for $x=0.12$, 0.14 , and 0.19 respectively, based on Gd^{3+} and the RBS measured density of Gd atoms (see ref. 9).