Spatially Resolved Inhomogeneous Ferromagnetism in (Ga, Mn)As Diluted Magnetic Semiconductors: A Microscopic Study by Muon Spin Relaxation

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(Received 12 May 2008; published 7 July 2008)

Thin epitaxial films of the diluted magnetic semiconductor (DMS) GaMnAs have been studied by low energy muon spin rotation and relaxation (LE-μSR) as well as by transport and magnetization measurement techniques. LE-μSR allows measurements of the distribution of magnetic field on the nanometer scale inaccessible to traditional macroscopic techniques. The spatial inhomogeneity of the magnetic field is resolved: although homogeneous above T_c, below T_c the DMS consists of ferromagnetic and paramagnetic regions of comparable volumes. In the ferromagnetic regions the local field inhomogeneity amounts to 0.03 T.

DOI: 10.1103/PhysRevLett.101.027202 PACS numbers: 75.50.Pp, 75.70.−i, 76.75.+i

The spin of the carriers has thus far played a minor role in semiconductor devices mainly because the semiconductors used for industrial applications, such as Si and GaAs, are nonmagnetic. On the other hand, the enhanced spin-related phenomena realized in diluted magnetic semiconductors (DMS) [1,2] open the way to applications in the field of spintronics [3,4]. Discovery of ferromagnetism (FM) in InMnAs [5] and later in the canonical DMS—GaMnAs films [6], which have the benefit of being compatible with GaAs technology—has led to intense studies of the FM in these materials for spintronics applications. Although there is an emerging consensus that FM in DMS originates from the interaction between itinerant carriers (holes in the case of GaMnAs) and localized magnetic moments, the microscopic theory of FM in these materials is still the subject of considerable debate. Theoretical models encounter major difficulties caused by the absence of experimental data on the distribution of the magnetic field in DMS on the nanometer scale. In particular, neither the strong-coupling nor the weak-coupling models (double exchange [7] and kinetic exchange [8]) can fully describe the experimental observations (for a recent review see [9]).

Note that these limiting cases correspond to two different length scales of hole localization relative to the separation between Mn ions, thus also defining different length scales of homogeneity of the FM state. If holes remain strongly localized by the Mn acceptor, the FM transition is expected to follow the percolation threshold dynamics of bound magnetic polarons [10–12]. Alternatively, if hole states are extended over lengths longer than the average distance between Mn acceptors, the Zener model becomes valid [13–15]. In both cases microscopic modeling of the FM exchange requires knowledge of the magnetic field distribution on the level of nanometers.

So far, however, the magnetic properties of GaMnAs have been studied mainly by macroscopic techniques (e.g., SQUID magnetometry, ferromagnetic resonance, magneto-optical measurements or magnetotransport), which do not probe the local nanometer-scale distribution of magnetic fields. Here the unique sensitivity of polarized positive muons as a local magnetic probe makes muon spin relaxation (μSR) ideally suited for mapping the magnetic state on the atomic scale [16,17]. This approach has already contributed greatly to the understanding of magnetic materials [16,17] and semiconductors [18]. In particular, the recent development of μSR with a polarized low energy muon beam (LE-μSR) [19] tunable in the kilo-electron-volt range (which allows one to vary the implantation depth between a few nanometers and a few hundred nanometers) provides a powerful tool for mapping magnetic field distributions in thin magnetic films by following the μ^+ decay at different depths, that reveals the effect of local magnetic fields on the time-dependent muon spin polarization (asymmetry) [16,17,20].

In this Letter we present investigations of GaMnAs by LE-μSR, and we demonstrate that the ferromagnetism of this material is highly inhomogeneous, consisting of ferromagnetic and paramagnetic (PM) regions whose sizes exceed the nanometer range.

DMS samples of $Ga_{1-x}Mn_xAs$ with layer thicknesses from 46 to 100 nm and with x between 0.02 to 0.06 were grown by low temperature molecular-beam epitaxy on semi-insulating (001) GaAs substrates. Several samples were cleaved into two sections, one of which was annealed at 280 °C for 1 h in nitrogen. All samples exhibit FM with T_c between 60 and 130 K as determined by magnetization (SQUID) measurements, and metallic resistivity below T_c. The annealing significantly increases T_c, reduces resistivity, and increases saturation magnetization, indicating removal of interstitial Mn and a corresponding increase of hole concentration [21] [see Figs. 2(a) and 2(b)].

Fully polarized low energy positive muons (with typical energy of 4 keV) from the LE muon beam line at the Paul Scherrer Institute (Switzerland) were implanted into the GaMnAs layer. Muon spin relaxation spectra of the ensemble of about 2×10^6 muons were recorded at various temperatures, magnetic fields, and implantations depths.

So far most of the research in condensed matter physics employed highly energetic 4.2 MeV muons with stopping range on the scale of millimeters in a solid. Low energy μSR makes use of 100% polarized positive muons of tunable energy (between 0.5 and 30 keV) to study local (nanoscale) magnetic properties of thin films as a function of the muon penetration depth [19].

In the μSR experiment one follows the time evolution of the polarization of the muon ensemble $P(t)$ via detection of the decay positron, which is emitted preferentially in the direction of the muon spin at the moment of the decay. The quantity $P(t)$ contains the information about the interaction of the muon magnetic moment with its magnetic environment. Each muon stopped in the lattice (typically at an interstitial site) experiences the net effect of external and local magnetic fields and therefore precesses at the characteristic frequency $\omega = \gamma_\mu B$ (where $\gamma_\mu = 2\pi \times 135.5$ MHz/T is the muon gyromagnetic ratio and B is the magnetic field at the muon site). The μSR technique offers its extreme sensitivity to the detection of small internal magnetic fields of electronic and/or nuclear origin: routine measurements of the internal magnetic fields below 0.1 G are performed in systems with either very small and/or dilute and random magnetic moments.

In a translationally invariant ferromagnet or antiferromagnet a single precession frequency may be observed. If the muons experience different magnetic fields, $P(t)$ shows a distribution of precession frequencies (a "line shape") with the correspondent width. Thus the local magnetic field distributions in the material can be measured. If the distribution is broad when averaged over the sample only a fast relaxation of the muon polarization is observed. By contrast, in a paramagnetic environment the polarization signal exhibits only slow exponential relaxation. In case of different magnetic environments, $P(t)$ is the superposition of the corresponding signals and it is therefore possible to detect and quantify different magnetic fractions in the material. Note that the μSR technique requires no application of external magnetic fields (which could alter the ground state of the material significantly). Details of the technique and data analysis are described in [17,19].

Figure 1 shows typical μSR spectra in zero field (ZF) in GaMnAs (2.2% Mn) below and above the FM transition. In zero field we find no precession in any of the FM samples studied. This is in contrast to coherent muon precession observed in systems displaying long-range FM order (e.g., elemental Fe, Co, Ni, Gd, and Dy, or the concentrated FM semiconductor EuS [22]). Instead, below T_c the muon polarization (proportional to the positron decay asymmetry) clearly consists of fast- and slow-relaxing components.

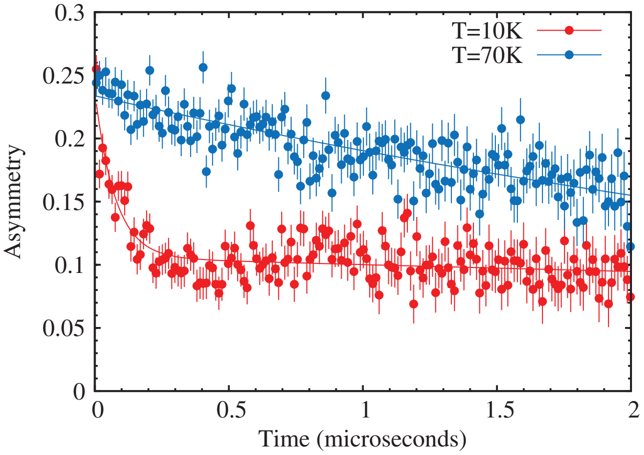

FIG. 1 (color). Muon spin relaxation spectra in GaAs:2.2%Mn in zero external magnetic field below the FM transition at $T_c = 60$ K (red dots) and above it (blue dots).

Figure 2(c) presents the temperature dependence of the relaxation rates for both components, which differ by 2 orders of magnitude. The fast-relaxing component is a clear signature of the FM state: it disappears above T_c and when muons stop in the nonmagnetic substrate.

The signal is typical of highly inhomogeneous magnets with local magnetic field $B_{\text{loc}} \approx \Delta B$. Its relaxation rate λ_f provides a measure of the magnetic field inhomogeneity at the muon site (expected to be an interstitial site in the GaAs matrix). Specifically, $\lambda_f = \gamma_\mu \Delta B$ gives the width of the magnetic field distribution experienced by the muon ensemble to be $\Delta B = 0.03$ T. This average magnetic field value is significantly less than the dipolar field of about 0.3 T produced by the moment of a single Mn (d^5 + hole) complex ($4\mu_B$) on the nearest interstitial site, probably reflecting some suppression of the magnetic moment of the Mn (d^5 + hole) complex [9].

Figure 2(d) strongly suggests that FM and paramagnetic regions coexisting in the same sample lead to distinct μSR signals whose amplitudes are proportional to volume fractions occupied by the two phases [16,17], a distinct long-range inhomogeneity being superimposed upon a local short-range magnetic field inhomogeneity in the FM state. The observation of the slow-relaxing component below T_c indicates that about 50% of muons do not feel any magnetic field of electronic origin, their relaxation rate (0.1 MHz, which corresponds to ΔB less than 0.1 mT) being consistent with fields produced by nuclear moments. Above T_c the fast component disappears, consistent with the onset of PM phase, where fast fluctuations of Mn magnetic moments average out local magnetic field to zero [16,17].

The two fractions in the FM phase are found in all samples independent of Mn content, film thickness, and annealing history. We found the same behavior at various depths within the sample (near the surface, near the GaMnAs/substrate interface, or near the center of the magnetic layer) with a depth resolution of 10 nm. The two-component picture (FM and PM regions) remains the same below T_c for zero-field and field-cooling procedures.

Muon spin relaxation spectra observed in external magnetic fields $B_{\rm ext}$ transverse to the incoming muon spin and parallel to the surface of the film confirm this finding, as seen in Fig. 3. Fast- and slow-relaxing fractions appear below T_c, while only the slow component is present above T_c. Strong confirmation that two phases are present comes from the fact that the precession frequency of the slow-relaxing component corresponds exactly to external magnetic field. This is inconsistent with the FM state: in a FM medium muon precession frequency should exhibit a significant shift with respect to the frequency corresponding to the applied magnetic field [16,17]. The ferromagnetic state in a DMS is not an exception: recent experiments in bulk DMS CdGeAs$_2$:Mn clearly showed a magnetic field shift on the muon site dependent on the magnetization [23].

The absence of a magnetic field shift, along with negligible relaxation, provides insight into the sizes of the two phases. A simple estimate of the dipole field produced by Mn moments shows that even muons stopped furthest from the magnetic ions—halfway between them—should experience fields an order of magnitude higher than the measured value. This necessarily means that for a reasonably uniform distribution of Mn in GaMnAs the observed long-range inhomogeneity has a length scale significantly greater than the mean distance between magnetic ions (about 0.9 and 1.3 nm for 6% Mn and 2% Mn, respectively). Thus a length scale of several nanometers is needed to explain the present muon spin relaxation results.

This observation is relevant to whether a spatially uniform FM spin order is a real ground state of the FM DMS on a scale greater than the mean distance between magnetic ions [24]. Nanoscale phase separation into paramagnetic and FM regions has been suggested to occur near the metal-insulator phase transition in p-type FM DMSs below

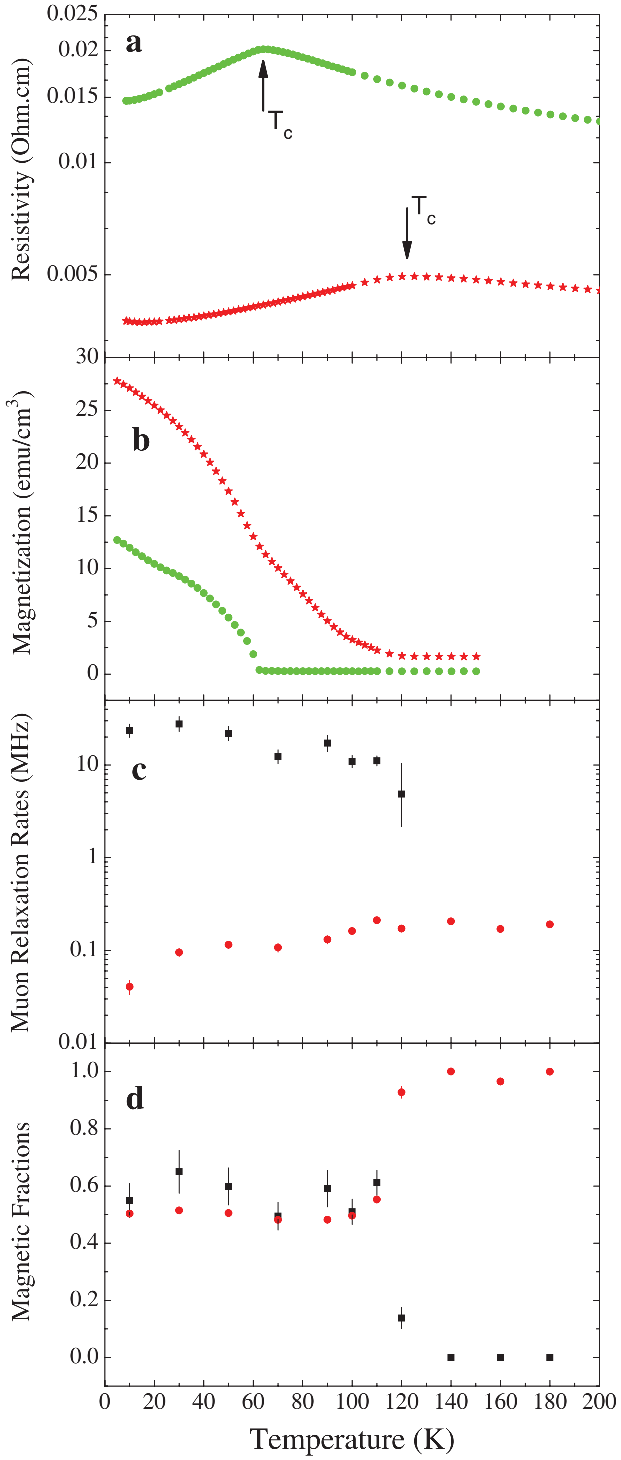

FIG. 2 (color). (a) Resistivity and (b) magnetization before (green dots) and after (red stars) annealing of GaAs:6%Mn. (c) Temperature dependences of the fast (black squares) and slow (red dots) muon relaxation rates in the annealed sample. (d) Corresponding magnetic fractions.

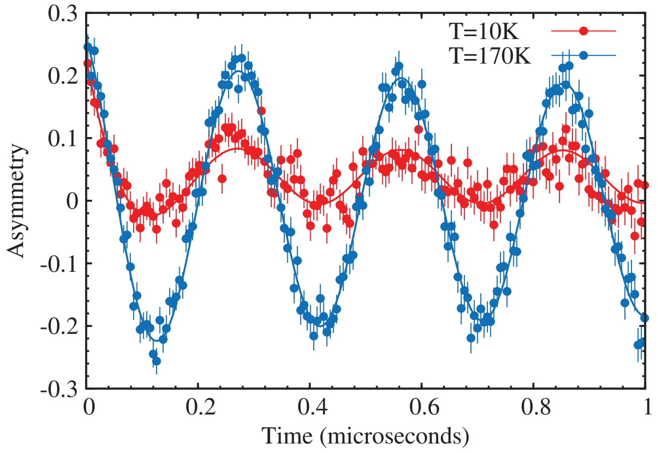

FIG. 3 (color). Muon spin relaxation spectra in GaAs:6%Mn below the FM transition at $T = 10$ K (red dots) and above it at $T = 170$ K (blue dots) in transverse magnetic field $B_{\rm ext} = 26$ mT. For both spectra the precession frequencies correspond to external magnetic field.

T_c when the hole localization length is much greater than the average distance between acceptors [13,14,24], with paramagnetic regions persisting down to the lowest temperatures at locations that are not visited by the delocalized holes [24].

In the alternative case of localized holes the FM transition is viewed as a magnetic percolation [11,12,25,26], which can also result in the coexistence of FM and PM regions. This model, however, assumes effects of clustering [12,26], that should lead to significant influence of annealing on the coexisting phases. However, such FM clusters (predicted to form above T_c and to coalesce at $T = T_c$, leading to a magnetic percolation transition) [12] are not found in our experiment.

Although the model of magnetic phase separation due to hole delocalization over extended nanometer region [13–15] describes our findings more adequately than the case of strongly localized carriers, further theoretical studies are needed to reconcile the short- and long-range inhomogeneities revealed by the present μSR experiments. In particular, the nature of the decomposition of FM GaMnAs into ferromagnetic and paramagnetic regions on the metallic side of metal-insulator phase transition still remains unresolved.

This work was supported by Kurchatov Institute, the (U.S.) National Science Foundation Grant No. DMR06-03752, and was partially performed at the Swiss Muon Source of the Paul Scherrer Institute, Villigen, Switzerland.

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