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Observation of Insulating Nanoislands in Ferromagnetic GaMnAs

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Resonant Raman data on ferromagnetic GaMnAs reveal the existence of a new kind of defect: insulating nanoislands consisting of substitutional MnGa acceptors surrounded by interstitial MnI donors. As indicated by the observation of a sharp 1S3/2 → 2S1/2 Raman transition at ~703 cm−1, the acceptor-bound holes inside the islands are isolated from the metallic surroundings. Instead, Mn-bound excitons do couple to the ferromagnetic environment, as shown by the presence of associated Raman magnon side bands. This leads to an estimate of 5–10 nm for the nanoisland radius. The islands disappear after annealing due to the removal of the MnI ions.

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The III-Mn-V diluted magnetic semiconductor (DMS) and, in particular, GaMnAs have attracted much attention recently due to their potential applications in spintronics [1]. It is now widely accepted that the ferromagnetism in these systems arises from the hole-mediated interaction between local magnetic moments. While the majority of the Mn ions in GaMnAs exists in the form of substitutional MnGa acceptors, a small amount also occurs as interstitial MnI donors. In addition to reducing the hole density, the interstitials form antiferromagnetic MnGa0-MnI pairs and MnGa-MnI-MnGa complexes, which reduce the number of magnetically active MnGa ions [2,3]. A recent hot-electron photoluminescence (PL) study indicates that the holes from MnGa form a detached impurity band for a wide range of manganese concentrations [4]. The MnI ions can be removed by low-temperature annealing [2,3,5].

In this Letter, we report on a resonant Raman scattering (RRS) study of GaMnAs. Consistent with recent theoretical analyses [2] as well as with the hot-PL results [4], which show evidence of the coexistence of paramagnetic (PM) and ferromagnetic (FM) regions below the Curie temperature, TC, and infrared absorption work [6], which suggests the existence of Mn acceptors isolated from the FM environment, our results conclusively show the occurrence of a new type of defect: paramagnetic nanoislands containing magnetically-inactive MnGa acceptors surrounded by MnI donors. The RRS technique has been used successfully to probe the 1S3/2 levels of Mn acceptors in insulating GaAs [7]. Our study focuses on the 1S3/2 → 2S1/2 transition of isolated neutral MnGa acceptors in DMS GaMnAs, which reveals itself as a Raman line at ~703 cm−1. In addition, we identified several Raman sidebands as due to the interaction between magnons from the FM environment and MnGa-bound excitons. These observations indicate that the distance between the MnGa acceptor and the FM surroundings is ~5–10 nm. We find that the hole-bound transitions disappear after the sample is annealed, and we interpret this as due to the outdiffusion of MnI ions [2].

We studied several FM GaMnAs films and a lightly-doped PM GaAs:Mn sample. The FM samples were grown on semi-insulating (001) GaAs substrates by low-temperature molecular-beam epitaxy (MBE). Two as-grown FM films are 300-nm thick with nominal Mn concentrations 1.4% and 2.6%, and the same Curie temperature of 44 K. The third as-grown FM film is 120-nm thick with 9% Mn and TC = 60 K. A small portion of this sample was annealed for one hour in a nitrogen gas atmosphere at 270°C. The PM sample, grown on (111) GaAs, has a concentration of 0.04% [7]. Samples were placed inside a liquid-helium flow magnetic cryostat for low temperature measurements. We used a cw Ti:sapphire laser whose wavelength γL could be tuned from 750 nm to 835 nm. RRS spectra were obtained with a Dilor XE system. The laser power density on the samples was ~8 mW/cm2. The experiments in a magnetic field were carried out in a near-backscattering Faraday geometry with fields oriented along z, the direction perpendicular to the films’ surface. The Raman geometry is described using standard notation: z(σ+, σ−)z denotes the backscattering configuration where the incident and scattered beams are represented, respectively, by z(σ+) and σ−z. Here σ+ and σ− stand for right and left circular polarizations, whereas z and z indicate the directions of the corresponding wave vectors.

Figure 1(a) shows RRS spectra excited at different laser wavelengths. The feature indicated by the dashed line, which appears on the high-energy side of the PL peak, is ascribed to the 1S3/2 → 2S3/2 Mn acceptor transition (we note that, due to exchange coupling between the j = 3/2 hole and the S = 5/2 Mn2+ core, the acceptor ground state is actually a F = 1 multiplet [7,8]). Our assignment is supported by the facts that this peak is only observed in samples containing Mn and that its position, 703 cm−1...
acceptors, denoted by Mn-bound holes in GaAs \[8\]. Furthermore, the PL in GaAs \[10\] is due to the recombination of electrons with Mn-bound holes. Here, the broad peak, PL, is due to the different between the 1S\(_{3/2}\) and 2S\(_{3/2}\) binding energies of Mn-bound holes in GaAs \[8\].

The Raman process is dominated by transitions involving the 1S\(_{3/2}\) and 2S\(_{3/2}\) impurity bands \[4,11,12\], which should be significantly broader than those of the low concentration PM specimen. We believe that the presence of neutral Mn acceptors is closely related to the existence of Mn\(_i\) in the FM samples. The interstitial ions, acting as double donors, can interact with neighboring Mn\(_{Ga}\) ions to form Mn complexes \[2,3\], in the neighborhood of which, the density of free holes is reduced facilitating the existence of solitary Mn\(_{Ga}\) acceptors. On the other hand, the Mn\(_i\) ions can be partly removed by annealing, which causes their outdiffusion to the surface \[2,3\].

The effect of low-temperature annealing on the 1S\(_{3/2}\) \(\rightarrow\) 2S\(_{3/2}\) transition of the 9% sample is shown in Fig. 1\(c\). Clearly, the Raman peak disappears after annealing. This can be explained by the increase in the hole density, which opens the initially isolated Mn\(_{Ga}\) ions to become interactive with the FM environment. Previous measurements have shown that annealing at 250 °C for 30 minutes can increase the hole concentration by ~10%–30% and lead to an enhancement of \(T_C\) by a factor of ~1.7 \[13\]. Also, isolated Mn ions located outside the ferromagnetic environment have been reported to exist in the DMS InMnSb \[14\].

Results in an applied magnetic field, of magnitude \(B\), reveal a strong dependence of the scattering on the polarization of the incident light; see Fig. 2\(a\). As shown in Fig. 2\(b\) at 2.6 K, the 1S\(_{3/2}\) \(\rightarrow\) 2S\(_{3/2}\) intensity increases (decreases) with increasing field if the exciting light is \(\sigma^-\) (\(\sigma^+\)) polarized. Referring to the simplified, single-particle RRS process depicted in Fig. 2\(c\), such a behavior can be accounted for by the Zeeman splitting of the ground \(F = 1\) multiplet, for which the gyromagnetic factor is \(g \approx 2.75\) \[7,8\], and thermal population effects. At sufficiently low temperatures, the bound holes reside for the most part in the lowest-lying state, \(m_F = -1\), involving mostly \(m_j = 1/2\), 3/2 states from the GaAs valence band \[15\]. To minimize the Coulomb repulsion between the two holes in \(A^0\)\(X\), the Raman process is dominated by transitions associated with states that have the same magnetic quantum number. These excitations are allowed mainly in one of the two circular polarizations because of the predominance of positive values of \(m_j\). Conversely, when the thermal energy is much larger than the Zeeman splitting, the \(m_F\) populations become comparable and, as illustrated by the 7 K data in Fig. 2\(b\), the difference in peak heights becomes negligible. These considerations also explain why
the scattering does not depend on the incident light polarization at \( B = 0 \); see Fig. 2(a). The theoretical curves in Fig. 2(b) were obtained using the optical selection rules for zinc blende semiconductors and the acceptor wave functions listed in [15].

Figure 3 shows the temperature dependence and selection rules of the acceptor transition in the 2.6% sample at \( B = 6.9 \) T. At large fields, the Raman feature splits into several peaks. In addition to the central band, labeled “0”, there are three sidebands marked “−1”, “+1” and “+2” (these sidebands were not seen in the PM sample and are not resolved in the spectra of Fig. 2(a) at \( B = 3 \) T). The separation between neighboring peaks is nearly the same as that of Mn-bound holes [16]. Therefore, we assign the peak “−1” to the \( 1S_{3/2} \rightarrow 2S_{3/2} \) transition with one magnon emitted, and peaks “+1” and “+2” to \( 1S_{3/2} \rightarrow 2S_{3/2} \) excitation followed by the absorption of one and two magnons, respectively. These assignments are consistent with the temperature-dependent results and the properties of GaMnAs magnons [16].

First, we note that the measured \( g \) is consistent with our interpretation because the magnon \( g \) factor is nearly the same as that of Mn 3d electrons [16]. Second, the fact that the intensity of peak “−1” is significantly smaller at 1.9 K supports our assignment since magnons are thermally excited and, thus, fewer magnons can be emitted at low temperatures. Finally, the observed dominance of one of the two circular polarizations in the scattering with magnon emission (sideband −1) and absorption (sideband 1) is also consistent with our interpretation since the angular momentum of the scattered photon must compensate for the change in the total spin of the magnetic system.

The occurrence of magnon sidebands indicates that some of the states involved in the RRS process interact with the FM environment. For an isolated acceptor, the

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**FIG. 2** (color online). (a) RRS of GaMnAs (1.4% Mn) at 2.6 K showing the dependence of the acceptor scattering on the incident photon polarization; \( \lambda_L = 826.5 \) nm. (b) Magnetic field dependence of the Raman intensity, in arbitrary units. Filled and open circles (squares) indicate \( \sigma^- (\sigma^+ -) \) polarized incident light at, respectively, 7 K and 2.6 K. The curves are from theoretical expressions described in the text. (c) Diagram showing the RRS process at zero field and in an applied magnetic field. The dashed line indicates states in the vicinity of the bottom of the GaAs conduction band. The dominant single-particle transitions are indicated by arrows.

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**FIG. 3**. RRS spectra of the sample with 2.6% Mn showing (a) the temperature dependence and (b) the scattering selection rules. The PL signal has been subtracted. Peak labeled 0 is the \( 1S_{3/2} \rightarrow 2S_{3/2} \) hole transition whereas −1, 1, and 2 are magnon sidebands.
Bohr radius of the $1S_{3/2}$ and $2S_{3/2}$ states are, respectively, $\sim 1$ nm and $\sim 5$ nm [7]. Since the linewidths of the $1S_{3/2} \rightarrow 2S_{3/2}$ transition in the FM and PM samples are the same within experimental uncertainty, as shown in Fig. 1(b), it is apparent that there is little interaction between the $1S_{3/2}$ and $2S_{3/2}$ levels of the MnGa acceptors in the islands and the FM surroundings. Therefore, the sidebands can only be the result of interactions between the FM magnons and the intermediate resonant state in the Raman process, i.e., MnGa-bound excitons. It follows that the radius of the PM islands must be somewhere between the Bohr radius of the $2S_{3/2}$ holes (5 nm) and that of the MnGa bound excitons (10 nm) [7]. We note that inhomogeneous structures of sizes similar to those of the nanoislands have been observed in GaMnAs using cross-sectional scanning tunneling microscopy [17].

An idealized picture of the PM nanoislands is shown in Fig. 5. The islands are insulating regions with a small concentration of holes immersed in the ferromagnetic host containing a much larger hole concentration. Their existence reflects indirectly the presence of MnI ions which, in itself, results from fluctuations favored by the nonequilibrium nature of low-temperature MBE [18]. The MnGa concentration inside the islands is lower than that in the film, while the MnI concentration is much higher. Acting as donors, the MnI ions significantly reduce the density of holes inside the island where the remaining neutral MnGa acceptors cannot interact with each other or with the ferromagnetic metallic surroundings [19]. The PM regions disappear after annealing, which removes MnI ions from the islands to the surface, as indicated by the disappearance of the acceptor transition in the annealed sample in Fig. 1(c).

In summary, the $1S_{3/2} \rightarrow 2S_{3/2}$ transition of holes bound to neutral Mn acceptors and associated magnon sidebands were observed in FM GaMnAs using RRS. The results show that insulating nanoislands of 5–10 nm radius exist in as-grown samples, but disappear after low-temperature annealing due to the removal of interstitial MnI ions.

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**References**


