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Light-induced polaron magnetization in EuTe at temperatures reaching 150 K

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We demonstrate that light creates a highly magnetized region in a magnetic semiconductor far above its critical temperature. A near-gap photon generates a quasiparticle of nonzero magnetic moment, named magnetic polaron, which is constituted by the photoexcited electron and about 1000 spin-polarized lattice atoms surrounding the photoexcited electron. The photoinduced magnetization follows a Langevin function, whose shape uniquely determines the magnetic moment of an individual polaron. In EuTe at 5 K the magnetic moment reaches a giant value of over 500 Bohr magnetons, thus the photoinduced magnetization saturates with a magnetic field of only 50 mT, which characterizes the magnetic polaron system as superparamagnetic. The polaron has an average lifetime of 15 μs. When temperature is increased its magnetic moment decreases, but at 150 K it still has a large value of about 80 Bohr magnetons. The paramagnet of polarons is fully controlled by light. Because the magnetic polaron affects only spin orientation, but not the charge distribution, in the superparamagnetic state the ideal optical quality of the host semiconductor is preserved.

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I. INTRODUCTION

Light control of magnetic order is a fascinating topic, and understanding its mechanisms is essential for designing and optimizing applications. It has been predicted a few decades ago that in magnetic semiconductors a photogenerated conduction electron polarizes lattice spins within the reach of its wave function, forming a quasiparticle named magnetic polaron [1,2]. In practice, observing magnetic polarons and determining their characteristic parameters (magnetic moment, radius, lifetime, etc.), has been until now a formidable challenge, because interpretation of experiments has had to rely on elaborate theories, as is the case of diluted magnetic semiconductors and their nanostructures [3–6]. In the latter the density of magnetic atoms is a small fraction of the density of native atoms. Intrinsic magnetic semiconductors [7,8], on the other hand, have a density of magnetic atoms many orders of magnitude greater than diluted ones, hence they have a potential for creating a much larger polaron-associated magnetization. However, progress on intrinsic magnetic semiconductors has been slow, because it has often been considered that the quality of samples is insufficient [9], or that their complex many-body scenarios make it impossible to fully understand their electronic and optical properties [9,10]. Nevertheless, intrinsic magnetic semiconductors grown by molecular beam epitaxy (MBE) display superior structural quality and chemical perfection [11], and MBE samples unraveled novel linear [12,13] and nonlinear [14,15] optical properties, which are very well described by standard mean-field solid-state theories [16–18]. Photogeneration of magnetic polarons in MBE material has been inferred from their band-edge photoluminescence (PL) [13,19]; however, because polaron-related PL is present only at very low temperatures (below 25 K) [13] and quenched in magnetic fields [19] as in EuTe, or simply absent, as in EuSe, knowledge of polarons is still very limited.

In this work we demonstrate that magnetic polarons can be readily photoexcited in an intrinsic magnetic semiconductor, using photoinduced Faraday rotation measurements—a technique that can be used at any temperature and field, unlike PL studies reported hitherto. The photoinduced magnetization is described by a Langevin function, whose shape uniquely determines the magnitude of the magnetic moment of an individual polaron, independently of the calibration of the measuring setup. Remarkably, the magnetic moment estimated using the Langevin function does not require any knowledge of the properties of an individual polaron, such as its lifetime, generation efficiency and circular birefringence, which is needed if the polaron magnetic moment is estimated from the absolute value of the Faraday rotation angle. The magnetic moment of an individual photoexcited polaron in EuTe achieves the huge value of more than 500 Bohr magnetons (μ_B) at 5 K, with a lifetime of 15 μs. The huge polaron magnetic moment implies that a small magnetic field of only 50 mT is sufficient to align all polarons. Photoinduced polarons were detected at a record high temperature of 150 K, which is more than ten times the critical temperature, when their magnetic moment is reduced to $80\mu_B$. In distinction from conventional superparamagnets that consist of a suspension of large-moment nanoparticles in a foreign material, and are thus optically diffuse, the polaron paramagnet does not disturb the ideal optical properties of the host semiconductor crystal.

II. RESULTS AND INTERPRETATIONS

The light-induced magnetic polarons were investigated by measuring the photoinduced Faraday rotation (PFR) using an optical time-resolved two-color pump-probe technique [see Fig. 1(a)]. The energy of the pump photons was tuned between 1.8 and 3.1 eV, which crosses the EuTe band gap at 2.2 eV, whereas the probe photon energy was fixed to a value below the EuTe band gap. A magnetic field was applied normal to the surface of the sample, which is parallel to the [111] crystalline direction. The EuTe sample was grown by molecular beam epitaxy on a (111)-oriented BaF_2 substrate. The thickness of the EuTe epitaxial layer was 1.5 μm, and the surface of the

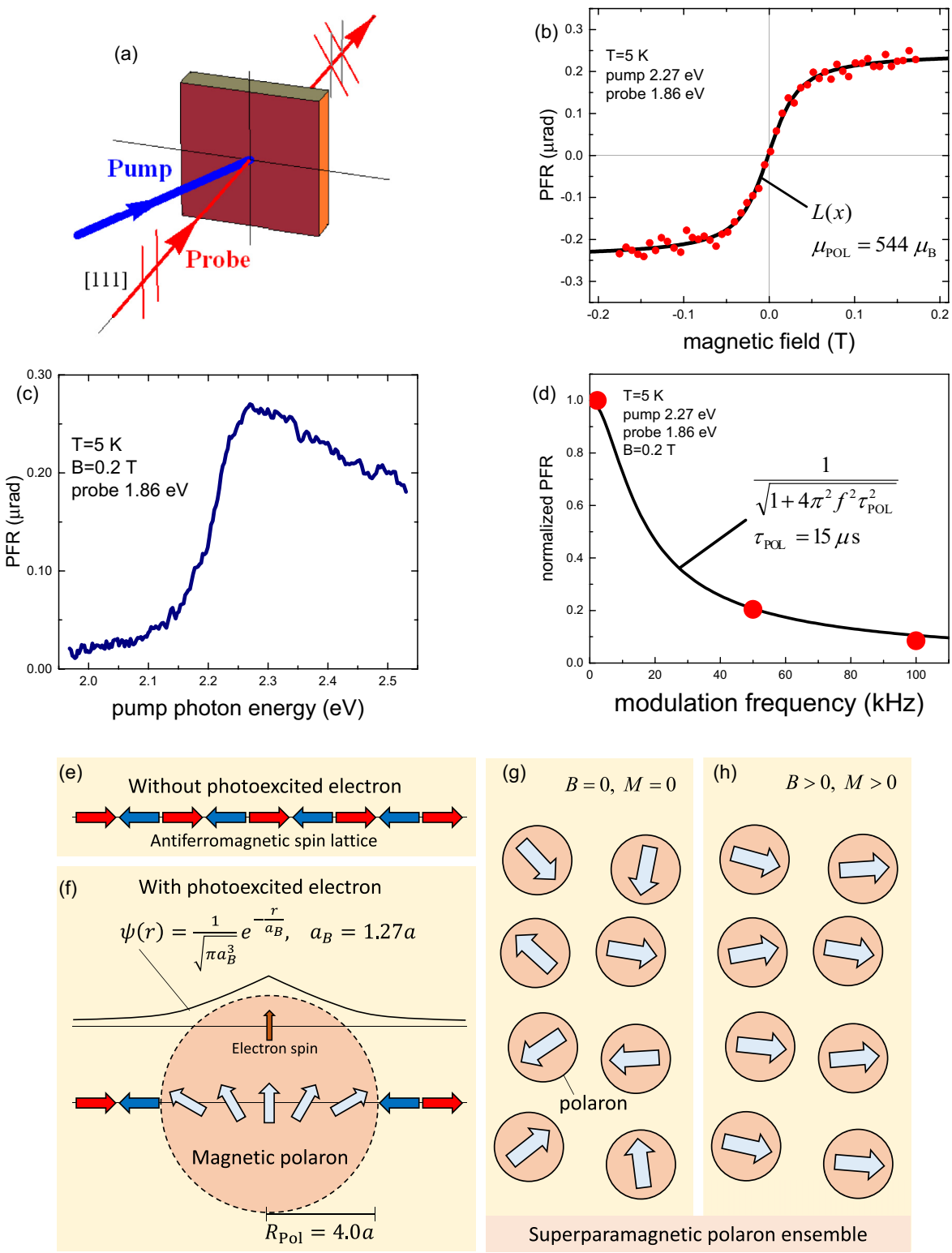

FIG. 1. (a) Geometry of experiment. Magnetic polarons in a longitudinal magnetic field are excited and probed by light. (b) Typical photoinduced Faraday rotation (PFR) signal as a function of the applied magnetic field at $T = 5$ K. (c) PFR as a function of the pump energy at $T = 5$ K and $B = 0.2$ T. (d) PFR as a function of the pump modulation frequency at $T = 5$ K and $B = 0.2$ T. (e) In equilibrium, the lattice is antiferromagnetic. (f) The photoexcited electron is described by a Bohr envelope wave function, and polarizes the lattice spins into a preferential direction in a radius of $R_{\mathrm{Pol}} = 4a$, forming a magnetic polaron. (g) The photoinduced polarons form a paramagnetic ensemble: at zero external field each polaron has an arbitrary orientation, and the net magnetic moment is zero. (h) Because the magnetic moment of the polaron is so large, a small magnetic field of 50 mT tesla is sufficient to align all polarons at 5 K.

epitaxial layer was capped with 200 nm of BaF_2. The experiments were performed using a variable temperature optical cryostat containing a superconducting coil to generate the magnetic field applied in the Faraday geometry. For optical excitation we used a variety of monochromatic sources: a xenon lamp passed through a monochromator, a frequency doubled Nd:YAG laser, or the frequency-doubled mode-locked Ti-sapphire tunable laser emitting pulses with a duration of 1.5 ps at a frequency of 76 MHz. The excitation light was modulated at 2.33 kHz using a chopper or at 50 and 100 kHz using a photoelastic modulator. The photoinduced Faraday rotation signal was independent from the polarization of the optical excitation. The probe light was a semiconductor laser operating at 665 nm or the fundamental pulses of the mode-locked tunable Ti-sapphire laser. For measuring the Faraday rotation angle of the linearly polarized probe beam, a homodyne technique based on phase-sensitive balanced detection was used. To minimize heating of the sample by the excitation light, the lowest possible excitation power was used, whereby a circular area of radius $\sim$100 μm was illuminated by a light power of 1 μW. By studying the shape of the PFR signal as a function of the magnetic field, which is described by a Langevin function and is therefore sensitive to the temperature of the sample, we certified that for the pump power we used the sample temperature differed from the thermal bath by at most 0.3 K. It was also observed that high excitation powers caused a detectable modulation of the bulk magnetization, generating an unwanted signal with opposite phase to the photoinduced polaron signal, which was avoided at the low excitation powers used here.

Figure 1(b) shows the PFR signal at $T = 5$ K, for a pump and probe photon energy of 2.27 and 1.86 eV, respectively, as a function of the magnetic field. The PFR signal is a direct measure of the photoinduced magnetization component along the probe wave vector [3,20]. Notice that the photoinduced magnetization quickly saturates with field, which is in sharp contrast with the linear low-field magnetization of EuTe (see, for instance, Ref. [21]). As seen in Fig. 1(c), the PFR shows a steplike increase when the energy of the pump photons resonates with the EuTe band gap, demonstrating that the PFR is provoked by photogenerated free conduction band carriers. To interpret the B dependence we assume that the photogenerated electrons polarize the lattice spins through the band-lattice exchange interaction and form magnetic polarons, as outlined in Figs. 1(e) and 1(f) (see also Ref. [18] for a complete theoretical model). Notice that the photogenerated hole is strongly localized in an Eu site, therefore it should not generate an effective exchange field, in contrast to the photoexcited electron. If we assume that the magnetic moment of a polaron is free to point in any direction except for the Zeeman interaction as sketched in Figs. 1(g) and 1(h), the photoinduced magnetization will be described by the Langevin formula (see, for instance, Ref. [20])

$$L(x) = \coth(x) - \frac{1}{x}, \qquad x = \frac{\mu_{\mathrm{Pol}} B}{k_B T}, \tag{1}$$

where μ_{Pol} is the magnetic moment of an individual polaron. It must be stressed that the S-like shape of the Langevin function dependence on field is determined by the one and only parameter—the magnetic moment of the individual particles forming a paramagnet—therefore it defines uniquely the magnitude of the polaron magnetic moment, without the necessity of any additional modeling or assumptions, and independently of the calibration of the Faraday rotation setup. Figure 1(b) shows that Eq. (1) provides an excellent fit of the experimental data, where the polaron magnetic moment best fit value is $\mu_{\mathrm{Pol}} = 544\mu_B$. A photoinduced magnetization that is resonant with the EuTe band gap [Fig. 1(c)], and caused by an ensemble of identical independent particles of magnetic moment of several hundred Bohr magnetons, as obtained from the Langevin fit, is exactly what is expected from photoinduced magnetic polarons [13,18]. The Langevin estimate is in excellent agreement with the polaron magnetic moment estimated from low temperature PL [13], and therefore validates the self-consistent magnetic polaron theory of Ref. [18], which for EuTe predicts $\mu_{\mathrm{Pol}} = 610\mu_B$ at $T = 0$ K. From Ref. [18] the radius of the polaron sphere is expected to be $R_{\mathrm{Pol}} = 4a$, independent of temperature and magnetic field, where $a = 6.6$ Å is the EuTe *fcc* lattice parameter. This sphere contains $\frac{4}{a^3}\frac{4}{3}\pi R_{\mathrm{Pol}}^3 \sim 1100$ Eu atoms, therefore on average each Eu atom contributes with $\sim\frac{1}{20}$ of its spin $S = 7/2$ to the total polaron magnetic moment. The giant μ_{Pol} explains why a small magnetic field of only 50 mT is sufficient to fully saturate the polaron magnetization, as seen in Fig. 1(b). The light-induced magnetization is generated in a surface layer of width equal to the light penetration depth $\frac{1}{\alpha} \sim 0.1$ μm, where $\alpha \sim 10$ $\mu\mathrm{m}^{-1}$ is the optical absorption coefficient at the band edge [22].

To investigate the photoexcited polaron lifetime τ_{Pol}, the PFR signal was measured as a function of the time delay between the arrival at the sample of the probe and pump pulses, using an optical delay line of 1 m length. It was found that PFR remains constant when the delay is varied between 0 and 4 ns, indicating that τ_{Pol} is much longer than a few nanoseconds, and a much larger delay line would be required. The polaron lifetime was therefore investigated by examining the dependence of the intensity of the PFR signal on the modulation frequency of the pump light, which for a single exponential recombination kinetics is given by

$$\mathrm{PFR}(f) = \frac{\mathrm{PFR}(0)}{\sqrt{1 + 4\pi^2 f^2 \tau_{\mathrm{Pol}}^2}}. \tag{2}$$

Figure 1(d) shows that Eq. (2) fits the experimental data very well and yields the polaron lifetime $\tau_{\mathrm{Pol}} = 15$ μs. This result is in perfect agreement with the lifetime estimated using the absolute value of the PFR angle [see Fig. 1(b)] and the low-field Verdet constant for EuTe [22].

By measuring the PFR as a function of magnetic field at different temperatures and fitting PFR with a Langevin function at each temperature, the corresponding polaron magnetic moment was obtained, and the result is plotted in Fig. 2(a). A temperature increase reduces the polaron magnetic moment, because the photoexcited electron loses the power to polarize lattice spins when they are more thermally agitated. To demonstrate this interpretation, the dashed line in Fig. 2(a) represents the calculated temperature dependence of the magnetic moment of a EuTe sphere of radius $R_{\mathrm{Pol}} = 4a$, under the sole effect of the exchange field, B_{Xf}, generated by

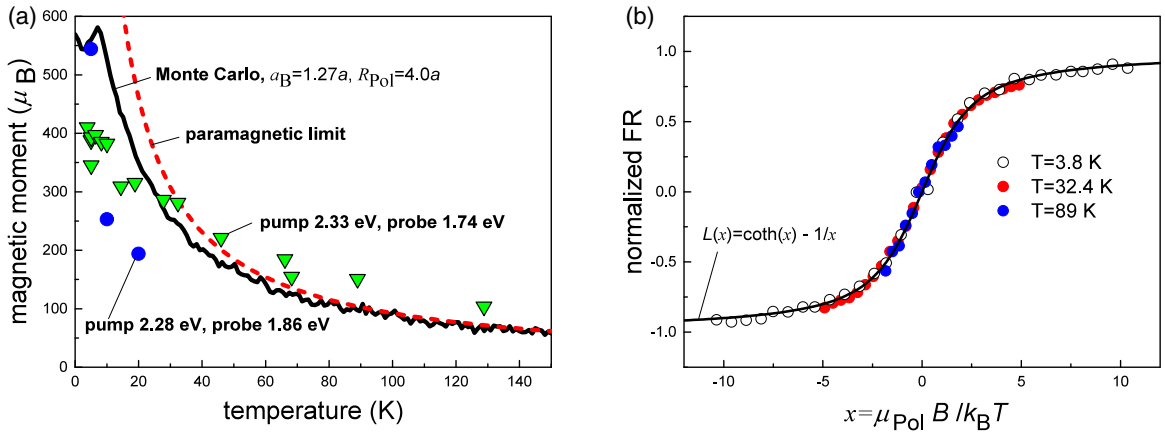

FIG. 2. (a) Magnetic moment of the polaron as a function of temperature. Circles and triangles are the experimental results. The full curve represents the result of the Monte Carlo calculation. The dashed curve shows the magnetic moment of a paramagnetic material under the influence of the average exchange magnetic field generated by the photoexcited electron. (b) Using the magnetic moment values shown in (a), the photoinduced Faraday rotation as a function of the applied field at any temperature collapses into a single Langevin function, shown by the solid line.

the photoexcited electron [18]

$$B_{Xf}(r) = \frac{J_{Xf}}{g_S \mu_B} \frac{a^3}{4} |\psi(r)|^2. \quad (3)$$

The polaron magnetic moment calculated in this way corresponds to the paramagnetic limit, when lattice spins are noninteracting. In Eq. (3) $J_{Xf} = 83$ meV is the band-lattice exchange interaction constant, and $\psi(r) = \frac{e^{-r/a_B}}{\sqrt{\pi a_B^3}}$ is the Bohr envelope wave function of the photoexcited electron, where $a_B = 1.27a$ [18]. Figure 2(a) shows that the paramagnetic approximation indeed agrees with the experimental data at high temperatures. However, at low temperatures the paramagnetic approximation fails, because the antiferromagnetic exchange interaction between Eu lattice atoms becomes effective, and prevents full alignment of the Eu spins with the magnetic field. To demonstrate this point, the solid curve in Fig. 2(a) represents the polaron magnetic moment obtained from a Monte Carlo simulation, where in addition to the exchange field of the photoexcited electron, the exchange interaction between Eu spins [18] was included, using $J_1 = 0.043$ K and $J_2 = -0.150$ K for first and second neighbor exchange interaction constants, respectively [8]. The Monte Carlo calculations are in good agreement with the experimental data, demonstrating that thermal fluctuations of lattice spins fully explain the temperature dependence of the polaron magnetic moment.

The paramagnetic behavior plus the large value of the magnetic moment of a polaron ($540\mu_B$ at 5 K) suggests that the photoexcited magnetic polaron ensemble can be classified as *superparamagnetic*, if the concept defined by Bean [23] is used. To test this further, the measured PFR was plotted as a function of the dimensionless variable $x = \frac{\mu_{Pol} B}{k_B T}$, in which case all curves collapse into a single Langevin function as depicted in Fig. 2(b), confirming the legitimacy of the superparamagnetic polaron model [23].

In conclusion, we have demonstrated the optical generation and monitoring of magnetic polarons, which have a huge magnetic moment and could be observed at temperatures as high as 150 K, confirming theoretical predictions based on photoluminescence measurements done at 4 K. Within the polaron, the magnetization reaches more than a one-twentieth of the saturation value in EuTe. We believe that photogenerated polarons remain in the light-penetration depth layer, and do not diffuse into the interior of the crystal, because PL associated with polarons is observed at ~1.9 eV in zero magnetic field [13], which is about 300 meV below the gap. Such a large binding energy suggests that the polarons are probably anchored by a lattice distortion. The results shown here for EuTe should remain valid for all europium chalcogenides as well as for other magnetic semiconductors with a large band-lattice exchange interaction. The magnetic polaron ensemble forms a paramagnet that is fully controlled by light within a host that has ideal optical quality. The optically controlled magnetism we have discovered demonstrates the strength of magnetic semiconductors, which combine the virtues of a semiconductor with ferromagnetism, and may find applications in light-controlled spintronic devices.

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