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Novel muonium centers—magnetic polarons—in magnetic semiconductors

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ABSTRACT

Muonium centers are readily formed in a wide variety of insulators and semiconductors due to enormous capture cross sections of the unscreened Coulomb potential produced by positive muons. Following the initial capture, different processes of electron localization produce two basically different muonium atoms—deep centers with the characteristic length scale of about the Bohr radius and shallow centers where electron is delocalized over many dozens of lattice spacings. In magnetic semiconductors, additional localization mechanism—exchange interaction–causes electron localization around the muon within about one lattice spacing. This novel muonium center—bound magnetic polaron—is found in Eu and Sm chalcogenides and ferromagnetic spinels.

1. Introduction

Recent progress in the field of spintronics is mainly connected to diluted magnetic semiconductors (DMS) which become ferromagnetic (FM) by addition of several percent of magnetic impurities. Successful demonstration of the injection of polarized spins into light-emitting diode structures and the electric field control of FM ordering [1,2] in DMS resulted in the emergence of a new field in electronics—*semiconductor spintronics*—involving the use of spin states in semiconductor materials. However, addition of magnetic impurities into DMS shifts a semiconductor to the metallic side of a metal–insulator transition. Our recent μSR studies of DMS [3,4] show that this circumstance strongly affects muonium atom formation: we found no muonium formed via transport and delayed capture of electrons by the positive muon—the mechanism which governs much of Mu formation in pure semiconductors [5–9].

As persistence of ferromagnetism in intrinsic (concentrated) magnetic semiconductors (MS) does not require addition of magnetic ions, one may expect Mu formation in MS via delayed electron capture. This may open up a possibility to study Mu formation in different magnetic states of MS (also using electric field techniques) and thus monitor electron transport and capture on the FM background.

In many μSR experiments the various neutral and ionic states of Mu may be successfully used as spectroscopic probes, without knowledge of exactly how these states are reached following muon implantation, to obtain information on systems traditionally interesting from the condensed state of physics point of view: superconductors and superfluids, metals and magnets, semiconductors and insulators, etc. It is in the cases where Mu is used as a model for hydrogen or a model for Coulomb defect center that knowledge of any peculiarities in its formation mechanism(s) is indispensable. Thus the widely accepted concept of Mu as a light hydrogen isotope has initiated extensive studies of different phenomena which complement our knowledge of states and dynamics of simple atoms in matter. In particular, studies of Mu dynamics in insulators and semiconductors have revealed different mechanisms of quantum tunneling phenomena [10] which are dramatically less pronounced for the heavier H atom. An investigation of semiconductors [11] has led to discovery of a variety of different muonium states, for which the analogous hydrogen-like defect centers where not all previously known. In such cases, an understanding of the different mechanisms by which muonium and hydrogen defect centers are created and detected is extremely important.

An electron in the conduction band is normally a free carrier. Localization raises the electron's kinetic energy and is thus unlikely unless some local interaction lowers its energy at least as much. A familiar example is the attractive Coulomb potential of a positive donor ion or a positive muon in semiconductors or

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insulators. A less obvious example is the *exchange* energy decrease when a number of magnetic ions with weak direct coupling experience a strong FM coupling mediated by their exchange interactions with the aforementioned electron. Since this interaction increases with the local probability density of the electron, it favors localization and can be sufficient to *autolocalize* the electron into a FM "droplet" on the scale of the lattice spacing in an antiferromagnetic or paramagnetic (PM) "sea". This quasiparticle is called a *magnetic polaron* (MP) [12,13] and is of fundamental interest.

The concept of MP is by now ubiquitous in any discussion concerning the physical properties of both MS and DMS. Nevertheless MP has eluded direct observation until now. Here we present the results of our studies of Eu and Sm chalcogenides EuO, EuS, EuSe, SmS and magnetic spinels $CdCr_2Se_4$ and $HgCr_2Se_4$ were we found novel Mu states—MP bound to positive muons.

In order to form the MP bound to μ^+ one has to ensure that the change in the free energy in the process of the electron capture

$$\Delta F = \frac{\hbar^2}{2m_* R^2} - J\frac{a^3}{R^3} - \frac{e^2}{\varepsilon R} \qquad (1)$$

has a minimum as a function of R—the radius of the electron localization around the muon. The increase in the electron kinetic energy due to localization [first term in Eq. (1)] should be compensated by the combine efforts of the Coulomb interaction [third term in Eq. (1)] and exchange interaction with coupling J [second term in Eq. (1)] (a is lattice spacing, m^* is the electron effective mass and ε is dielectric constant of the host MS). The exchange contribution to the localization amounts to a difference between the PM order of the host and the enhanced FM order in the MP.

Note that the muon acts both as positive defect center and the source for electrons created during its thermalization process [5–9]. In semiconductors, this phenomenon produces a model system with which to study electron capture by and release from the donor center (positive muon) [8].

Although initial electron capture solely due to Coulomb potential is viewed as a common phenomenon, subsequent electron localization leads to formation of two quite different Mu centers: compact (atomic) Mu states found in Si, Ge, GaAs, etc. [11] and dilated over many lattice spacings shallow Mu states detected in II–VI semiconductors CdS, CdTe, ZnO, etc. [14].

In magnetic chalcogenides and spinels, the second term in Eq. (1) causes further electron localization as compared to a typical Mu shallow donor state which is readily detected by transverse magnetic field μ^+SR techniques.

2. Experimental

Prior to μ^+SR experiments all the samples were characterized by XRD and SQUID measurements. XRD data have not revealed any traces of alien phases. Examples of SQUID data on one of Eu chalcogenides are shown in Figs. 1 and 2.

Similar measurements on one of FM spinels are presented in Figs. 3 and 4.

Time-differential μ^+SR experiments were performed on the M15 surface muon channel at TRIUMF using the *HiTime* apparatus. In order to get rid of demagnetization effects in applied magnetic fields [15] we used a ball-shaped samples from 6.5 to 7 mm in diameter.

At room temperature, in high transverse magnetic field we detected three-frequency precession in all the samples studied. Fourier transforms of the corresponding spectra are shown in Figs. 5–8.

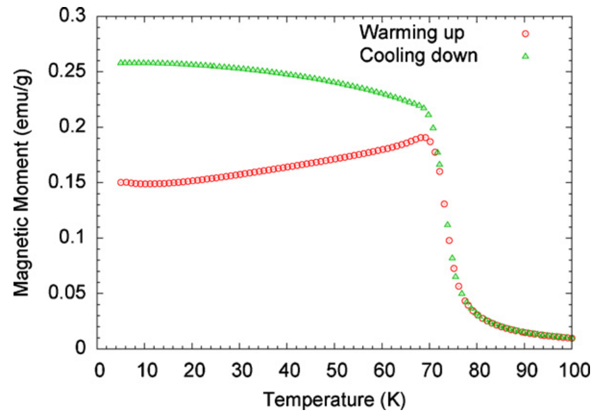

Fig. 1. Temperature dependence of the magnetization of EuO sample in magnetic field 0.001 T. Data show both warming and cooling behavior. FM transition temperature $T_c = 69$ K is found in good agreement with literature data.

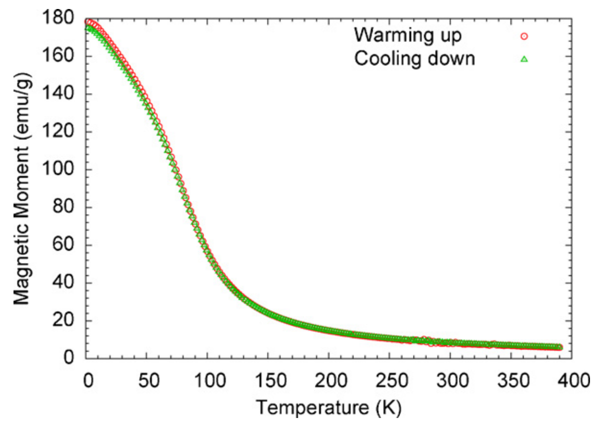

Fig. 2. Temperature dependence of the magnetization of EuO sample in magnetic field 5 T. Data show both warming and cooling behavior.

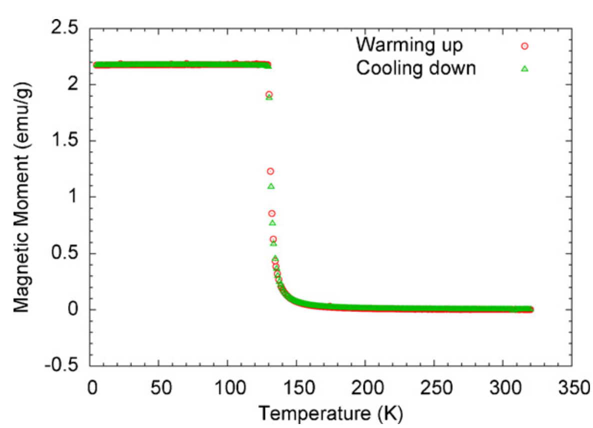

Fig. 3. Temperature dependence of the magnetization of $CdCr_2Se_4$ sample in magnetic field 0.005 T. Data show both warming and cooling behavior. FM transition temperature $T_c = 130$ K is in good agreement with literature data.

At lower temperature, muon precession spectra consist of just one line broadened due to interaction with magnetic moments of the corresponding atoms (Eu, Sm or Cr). We claim that the central line is the signal from bare muons which avoid electron capture, and that the two satellite lines represent characteristic muonium spectra from those muons that managed to capture and localize electrons.