

# Experimental report

10/04/2016

**Proposal:** 4-03-1712

**Council:** 10/2014

**Title:** Localized high-energy magnetic excitations in a new dilute magnetic semiconductor

**Research area:** Physics

**This proposal is a new proposal**

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**Samples:**  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2\text{As}_2$  ( $y=0.15$ ,  $x=0;0.2$ )

Instrument	Requested days	Allocated days	From	To
IN4	0	4	13/07/2015	17/07/2015
IN8	7	0		

## Abstract:

A recently discovered dilute magnetic semiconductor  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2\text{As}_2$  represents a model system with decoupled spin and charge doping. This material can be accurately described by the density functional theory, resulting in specific theoretical predictions as to the doping-dependent energies of exchange interactions. Here we propose to verify these predictions experimentally on powder samples of  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2\text{As}_2$  using the IN8 spectrometer.

# EXPERIMENTAL REPORT

Proposal number: 4-03-1712

Instrument: IN4

Dates of the experiment: 13 – 17 July, 2015

In the present experiment, we studied polycrystalline samples of diluted magnetic semiconductors (DMS) [2?] by mapping out the energy-momentum space:

- To verify the theoretical predictions [4] of Mn-Mn antiferromagnetic exchange parameters by checking the energy scale of localized magnetic excitations in the INS spectrum
- To verify the theoretically predicted [4] dependence of the excitation energies on the K doping concentration

Our experiment was performed at the disk chopper time-of-light spectrometer IN4. We studied 4 DMS samples with the general composition  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2$ : K-doped sample with  $x = 0.2, y = 0.15$  [3], undoped  $\text{Ba}(\text{Zn}_{1-y}\text{Mn}_y)_2$  with  $y = 0.15$  and  $0.2$  and the reference sample  $\text{BaZn}_2\text{As}_2$ . All samples are characterized by the  $I4/mmm$  space group, lattice parameters are  $a = b = 4.121 \text{ \AA}$  and  $c = 13.575 \text{ \AA}$ . All polycrystalline samples were wrapped in aluminum foil, fixed in the holder, and mounted in an orange-type cryostat. The incident neutron wavelength was fixed at  $1.2 \text{ \AA}$  ( $56.81 \text{ meV}$ ). All measurements were performed at  $1.6 \text{ K}$ .

The measured INS intensity from each sample is shown as momentum-energy color maps in Fig. 1 (panels a–d). The shown INS signal is normalized for the mass of each sample. One can see non-dispersive magnetic excitations around  $1 - 2 \text{ \AA}^{-1}$ , at the same time the high- $|\mathbf{Q}|$  part of the figure is dominated by phonon scattering. In order to better separate the magnetic signal from the nonmagnetic background, we sequentially subtracted the measured intensity of the reference compound  $\text{BaZn}_2\text{As}_2$  from the signal of each DMS sample. The results of the subtraction are shown in Fig. 2 (panels a–c), where one can clearly see the same low- $|\mathbf{Q}|$  magnetic excitations from Fig. 1. The energy scale of these excitation agrees with the theoretical predictions for the non-doped samples. To reveal the detailed shape and energy evolution of magnetic signal with doping, we plotted in Fig. 3 a momentum-integrated INS intensity at low  $|\mathbf{Q}|$ , estimated from the data in Fig. 1 (a–d) and Fig. 2 (a–c) [panels (a) and (b), respectively]. We can see that in both non-doped  $\text{Ba}(\text{Zn}_{1-y}\text{Mn}_y)_2$   $y = 0.15, 0.2$  samples, the peak is centered around  $13 \text{ meV}$  independently of the Mn concentration. K-doping leads to a significant broadening of the magnetic excitation and a slight shift of the center of the peak towards lower energies, as one would expect for a damped oscillator under the assumption of coupling to the continuum of particle-hole excitations. This behavior differs from the theoretical prediction, where a much more considerable change in the energy of the magnetic peak was anticipated. The main findings of our experiment are:

- We revealed magnetic excitations in the undoped Mn- $\text{BaZn}_2\text{As}_2$  at the theoretically predicted energy.
- The energy of the localized magnetic excitation is independent of Mn concentration for the  $y = 0.15, 0.2$  samples, indicating that they can be still treated in the dilute limit.
- Electronic doping by potassium introduces charge carriers and, as a consequence, broadens and softens the magnetic excitation, which appears as a damped harmonic oscillator with approximately the same energy as in the undoped compound. In contrast to the theoretical prediction that anticipated a nearly twofold decrease of the peak energy, we only observe a minor softening from  $13 \text{ meV}$  to  $11 - 12 \text{ meV}$ .

These results are being prepared for publication.

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[1] H. Ohno, Science **281**, 281 (1998).

[2] T. Dietl, Science **281**, 281 (2010).

[3] K. Zhao *et al.*, Nature Commun. **4**, 1442 (1442).

[4] J. K. Glasbrenner *et al.*, Phys. Rev. B **90**, 140403(R) (2014).

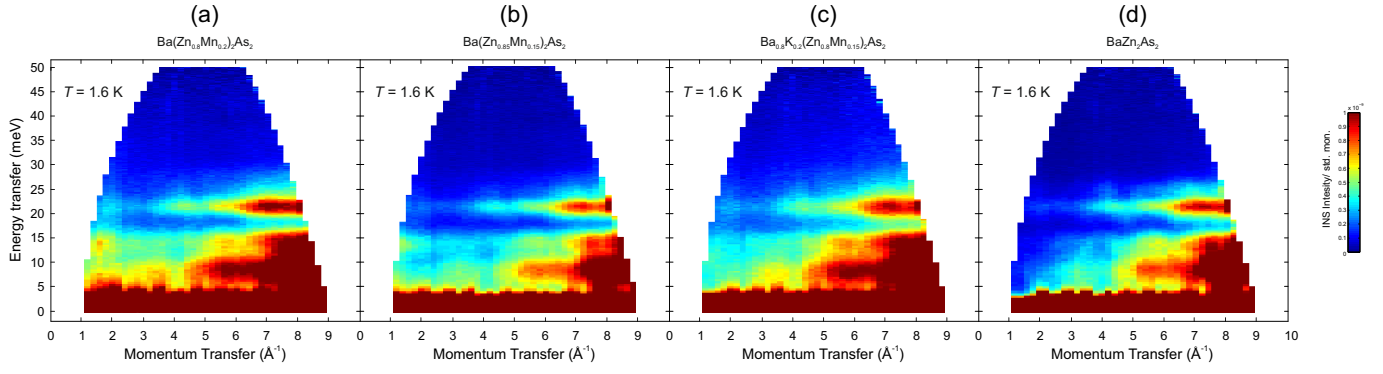


Fig. 1. Raw momentum-energy maps of DMS:  $\text{Ba}(\text{Zn}_{1-y}\text{Mn}_y)_2$   $y = 0.15, 0.2$  [panel (a,b)],  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2$   $x = 0.2, y = 0.15$  [panel (c)], the reference sample  $\text{BaZn}_2\text{As}_2$  [panel (d)]. The intensity in each panel is normalized for the mass of each sample. Localized magnetic excitations can be seen at low  $|\mathbf{Q}|$  around  $1\text{--}2\text{ \AA}^{-1}$ .

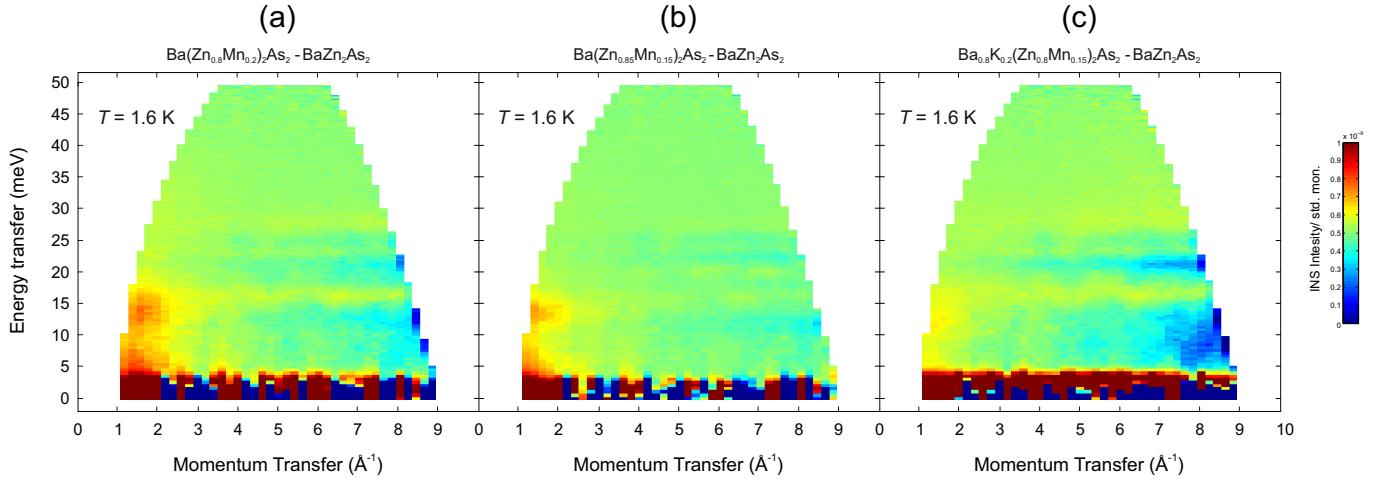


Fig. 2. Momentum-energy maps of DMS:  $\text{Ba}(\text{Zn}_{1-y}\text{Mn}_y)_2$   $y = 0.15, 0.2$  [panel (a, b)] and  $\text{Ba}_{1-x}\text{K}_x(\text{Zn}_{1-y}\text{Mn}_y)_2$   $x = 0.2, y = 0.15$  [panel (c)] after subtraction of the signal from the reference sample  $\text{BaZn}_2\text{As}_2$ . One can clearly see magnetic excitations in low- $|\mathbf{Q}|$  region.

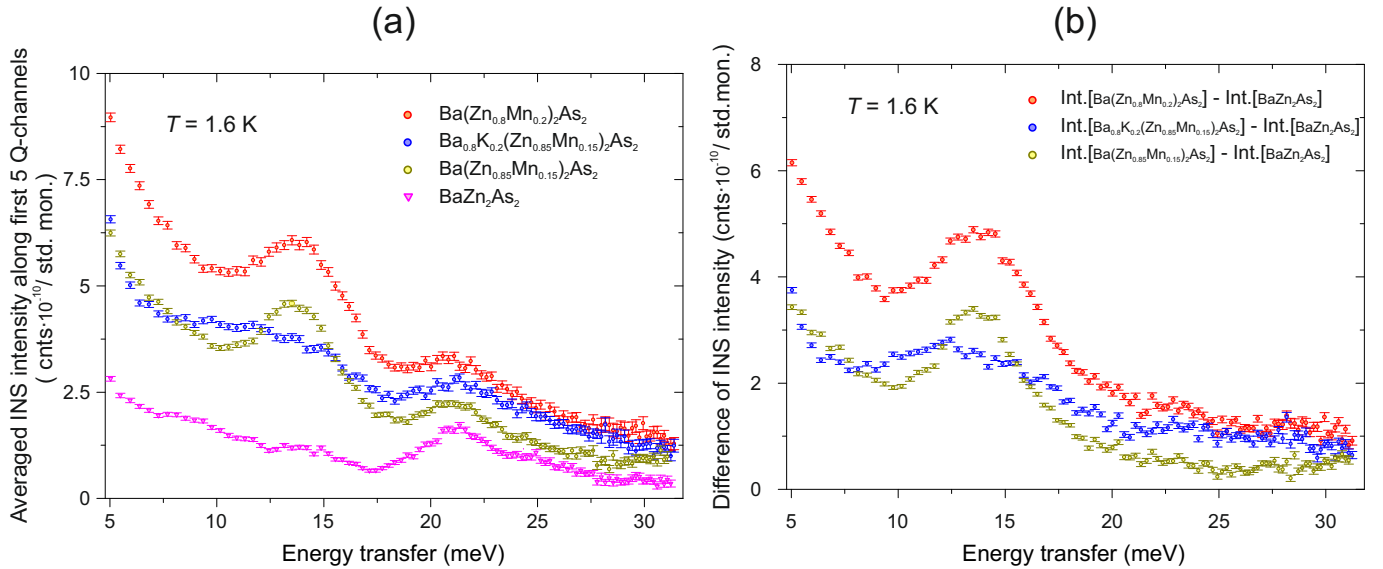


Fig. 3. (a) Energy cuts through the INS data, integrated in the low- $\mathbf{Q}$  region (first 5  $|\mathbf{Q}|$  channels) form the INS signal in Fig. 1 [here panel (a)]. (b) Magnetic signal obtained by subtraction of the nonmagnetic reference signal from  $\text{BaZn}_2\text{As}_2$ . Red (blue, yellow) circles correspond to  $\text{Ba}(\text{Zn}_{0.8}\text{Mn}_{0.2})_2$ ,  $\text{Ba}_{0.8}\text{K}_{0.2}(\text{Zn}_{0.85}\text{Mn}_{0.15})_2$ , and  $\text{Ba}(\text{Zn}_{0.85}\text{Mn}_{0.15})_2$ , while pink triangles show nonmagnetic background from the reference sample  $\text{BaZn}_2\text{As}_2$ . One can see a clear peak around 13 meV for  $\text{Ba}(\text{Zn}_{1-y}\text{Mn}_y)_2$   $y = 0.15, 0.2$  and a somewhat broader and slightly softened peak for  $\text{Ba}_{0.8}\text{K}_{0.2}(\text{Zn}_{0.85}\text{Mn}_{0.15})_2$ .