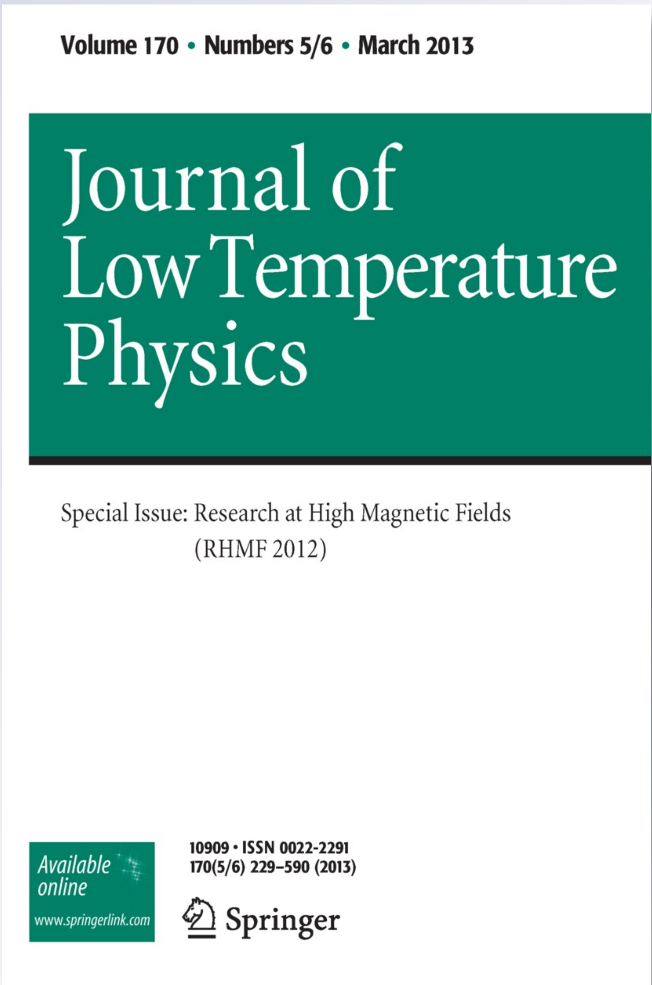
High-field ESR Studies of the Quantum Spin Dimer System $Ba_3Cr_2O_8$

D. Kamenskyi, J. Wosnitza, J. Krzystek, A. A. Aczel, H. A. Dabkowska, A. B. Dabkowski, G. M. Luke & S. A. Zvyagin

Journal of Low Temperature Physics

ISSN 0022-2291
Volume 170
Combined 5-6

J Low Temp Phys (2013) 170:231-235
DOI 10.1007/s10909-012-0722-4



Springer

J Low Temp Phys (2013) 170:231–235
DOI 10.1007/s10909-012-0722-4

High-field ESR Studies of the Quantum Spin Dimer System $Ba_3Cr_2O_8$

D. Kamenskyi · J. Wosnitza · J. Krzystek · A.A. Aczel · H.A. Dabkowska · A.B. Dabkowski · G.M. Luke · S.A. Zvyagin

Received: 9 June 2012 / Accepted: 6 September 2012 / Published online: 20 September 2012

Abstract Results of systematic high-frequency electron spin resonance (ESR) studies of $Ba_3Cr_2O_8$, a weakly coupled $S = \frac{1}{2}$ dimer system, in magnetic fields up to 25 T are reported. Two pairs of ESR gapped modes corresponding to transitions from a spin-singlet ground state to the first excited triplet states with gaps, $\Delta_{AB} = 563$ GHz and $\Delta_{CD} = 399$ GHz, are revealed below $H_{c1} = 12.5$ T. The detection of the ground-state excitations by means of ESR clearly indicates the presence of a non-secular term allowing these transitions. A complex structure of the microwave absorption spectrum in magnetic fields above H_{c1} is observed, those peculiarities are discussed.

Keywords Electron spin resonance · Field-induced phase transition · Dimers

1 Introduction

Field-induced phase transitions in $S = \frac{1}{2}$ quantum magnets have recently received a considerable amount of attention, particularly in the context of the quantum criticality problem. The corresponding evolution of the ground state and the magnetic

D. Kamenskyi · J. Wosnitza · S.A. Zvyagin (✉)
Dresden High Magnetic Field Laboratory (HLD), Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany
e-mail: s.zvyagin@hzdr.de

J. Krzystek
National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32310, USA

A.A. Aczel · H.A. Dabkowska · A.B. Dabkowski · G.M. Luke
Department of Physics and Astronomy, McMaster University, Hamilton, Ontario L8S 4M1, Canada

A.A. Aczel
Quantum Condensed Matter Division, Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

excitation spectrum was studied in a number of magnetic systems with a large diversity of magnetic structures, ranging from simple weakly-isolated isotropic spin dimer systems ($TlCuCl_3$ [1] and $BaCuSi_2O_6$ [2]) to quasi-two-dimensional networks of strongly interacting dimers with frustrated interactions ($SrCu_2(BO_3)_2$ [3]). In some cases such field-induced phase transitions can effectively be described in terms of the field-induced magnon Bose-Einstein condensation (BEC) [2, 4]. An important property of such systems is that they must posses $U(1)$ rotational symmetry (as required by the BEC theory), or at least the anisotropy must be small enough not to alter the BEC universality class of the phase transition.

As a powerful technique to probe the low-energy excitations spectrum in such spin systems, electron spin resonance (ESR) plays a particular important role [5–8]. In this work, we report on ESR studies of the $S=\frac{1}{2}$ weakly coupled dimer system $Ba_3Cr_2O_8$. This material belongs to a new class of systems with the general formula $A_3M_2O_8$, where $A = Ba^{2+}$ or Sr^{2+}, and $M = Cr^{5+}$ or Mn^{5+}. At room temperature, these compounds crystallize in the $R\overline{3}m$ space group with the crystal structure consisting of MO^{4-} tetrahedra and isolated A^{2+} ions. $Ba_3Cr_2O_8$ has magnetic Cr^{5+} ions ($3d^1$, $S=\frac{1}{2}$) and lattice parameters $a = 5.739$ Å, $c = 21.375$ Å determined at room temperature. Cr^{5+} ions, coordinated tetrahedrally, form dimers along the crystallographic c axis arranged in the triangular lattice in the ab plane. The distance between the Cr^{5+} ions in the dimers is $d = 3.934$ Å, while the minimal distance between Cr^{5+} ions from neighboring dimers is $d = 5.739$ Å [9]. This makes $Ba_3Cr_2O_8$ an excellent model system for three-dimensional weakly coupled dimers with dominant intradimer coupling $J_0 = 2.38(2)$ meV and weak interdimer couplings of less than 0.52(2) meV [10].

Recently, magnetic properties of this compound have been studied by means of a variety of experimental techniques [10, 11]. Two critical fields, $H_{c1} = 12.5$ T and $H_{c2} = 23.6$ T (determined at $T = 0$) have been extracted from the phase diagram with a maximal transition temperature $T_c \approx 2.7$ K at about 18 T. In this region, the system is in a long-range antiferromagnetically ordered state. The first transition was described in terms of the magnon BEC [11]. Inelastic neutron-scattering measurements [10] showed a complex excitation spectrum consisting of three excitation modes with pronounced dispersions, which can be described assuming spatially anisotropic inter-dimer interactions in three crystal domains in $Ba_3Cr_2O_8$ formed as result of a cooperative Jahn-Teller-like structural phase transition at $T \simeq 70$ K. This transition is accompanied by the lowering of the symmetry from rhombohedral R-$3m$ to monoclinic $C2/c$ [10].

Previously, ESR studies of $Ba_3Cr_2O_8$ have been performed in the frequency range from 70 to 380 GHz in magnetic fields up to 13 T [12]. Three ESR modes were observed, two gapped modes and one gapless, with $g = 1.94$ ($H \perp c$). The gap values, 546(9) GHz and 406(4) GHz, were determined by extrapolating the frequency-field dependences of these modes to zero field. Noticeably, the observation of these modes suggests the presence of intra-dimer Dzyaloshinsky-Moriya (DM) interaction in this compound.

In this paper, we report systematic studies of the magnetic excitation spectrum of this compound by means of tunable-frequency ESR spectroscopy in the extended frequency range, 50–700 GHz, and in magnetic fields up to 25 T. Two gaps, $\Delta_{AB} =$

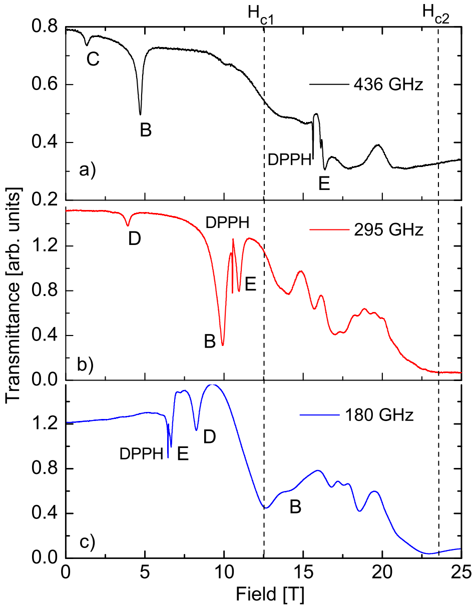


Fig. 1 ESR transmission spectra of $Ba_3Cr_2O_8$ at frequencies 436 GHz (**a**), 295 GHz (**b**), 180 GHz (**c**) taken in magnetic fields up to 25 T at $T = 1.4$ K. The first, $H_{c1} = 12.5$ T, and the second, $H_{c2} = 23.6$ T, critical fields (determined at $T = 0$) are shown by *vertical dashed lines* (Color figure online)

563 GHz and $\Delta_{CD} = 399$ GHz, have been observed. A complex picture of radiation absorption was revealed above H_{c1}.

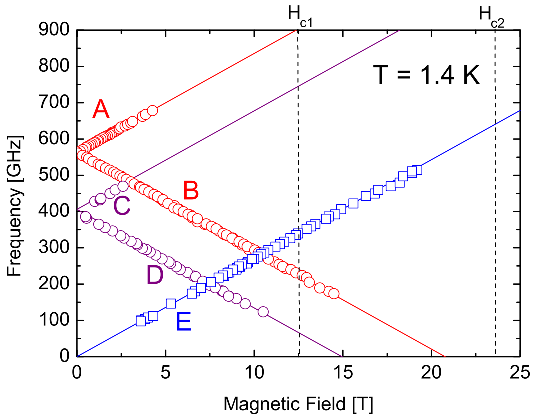
2 Experimental Details

Single crystalline $Ba_3Cr_2O_8$ samples with typical sizes of $4 \times 2 \times 0.2$ mm^3 were used [13]. Experiments have been performed using two tunable-frequency ESR spectrometers operated in combination with a 16 T superconducting magnet at the Dresden High Magnetic Field Laboratory, and a 25 T resistive magnet at the National High Magnetic Field Laboratory, Tallahassee, USA [14]. The magnetic field was applied along the c axis. Backward Wave Oscillators (BWO) and Virginia Diodes Inc. microwave sources were employed as tunable sources of mm- and submm-wavelength radiation. 2,2-Diphenyl-1-picrylhydrazyl (DPPH) was used as a standard marker.

3 Results and Discussion

ESR spectra obtained at frequencies 180, 295 and 436 GHz in magnetic fields up to 25 T are shown in Fig. 1. Five modes have been observed in magnetic fields below the first critical field, $H_{c1} = 12.5$ T. The frequency-field dependencies of the observed modes are shown in Fig. 2. Two sets of ESR modes (A, B and C, D, respectively) with gaps, $\Delta_{AB} = 563 \pm 1$ GHz (27 K) and $\Delta_{CD} = 399 \pm 1$ GHz (19.1 K), have been observed. These excitations correspond to ESR transitions between the spin-singlet ground state and the first excited spin-triplet states. The obtained data are consistent with results of inelastic neutron scattering [10]. In accordance to that, the modes A

Fig. 2 Frequency-field dependence of ESR excitations in $Ba_3Cr_2O_8$ taken at $T = 1.4$ K. The first, $H_{c1} = 12.5$ T, and the second, $H_{c2} = 23.6$ T, critical fields (determined at $T = 0$) are shown by *vertical dashed lines*. *Solid lines* are guides for the eye (Color figure online)



and B correspond to excitations in the center of the Brillouin zone, at $k = 0$. The modes C and D correspond to transitions from the ground state to the first excited state at $k_c = \pi/c$. Sunglet-triplet ESR transitions are normally forbidden in a simple dimer system. However, a non-secular term such as DM interaction or alternating g-tensors can mix the wave functions of the spin-singlet ground and triplet exited states, allowing these transitions to occur in $Ba_3Cr_2O_8$. Below H_{c1}, mode E corresponds to $\Delta S_z = \pm 1$ transitions inside the excited triplet. The slope of frequency-field dependencies of modes A, B, C, D, and E corresponds to $g = 1.94$, which coincides with earlier observations [10].

The extrapolation of the frequency-field dependencies of modes D and B to zero frequency gives $H^*_{c1} = 14.7$ T and $H^*_{c2} = 20.7$ T (for $T = 1.4$ K). Interestingly, these values are different from that extracted from the temperature-field phase diagram $H_{c1} = 13.7$ T and $H_{c2} = 22.8$ T (determined at $T = 1.4$ K) and $H_{c1} = 12.5$ T and $H_{c2} = 23.6$ T (determined at $T = 0$) [11]. This observation seems natural assuming a complex dispersion of magnetic excitations in $Ba_3Cr_2O_8$ [12], with extreme values of excitation energies different from Δ_{AB} and Δ_{CD}.

The interpretation of high-field ESR spectra (Fig. 1) above H_{c1} is more challenging. The transmissions exhibit a very complex structure, although being well reproducible for up and down magnetic field sweeps. The presence of hysteresis found in the vicinity of H_{c2} by means of magnetization and magneto-caloric effect [11] might suggest more sophisticated picture of magnetic interactions in $Ba_3Cr_2O_8$, possibly involving a complex magnetic domain structure.

4 Conclusions

In conclusion, we present results of high-frequency ESR studies of the weakly-coupled dimer system $Ba_3Cr_2O_8$ in magnetic fields up to 25 T. Two pairs of gapped modes corresponding to transitions from a spin-singlet ground state to the first excited triplet with gaps, $\Delta_{AB} = 563$ GHz and $\Delta_{CD} = 399$ GHz, were observed below $H_{c1} = 12.5$ T. The observation of the ground-state ESR excitations clearly indicates the presence of a non-secular term allowing such transitions in this compound with monoclinic structure, which breaks the $U(1)$ rotational symmetry. We hope that our results (in particular, the observation of complex transmittance spectra above H_{c1}) will stimulate further theoretical and experimental investigations of this material, which would allow to accurately estimate the anisotropy in $Ba_3Cr_2O_8$ and to determine the magnon BEC applicability criteria for the field-induced antiferromagnetically-ordered phase.

Acknowledgements We thank O. Cepas and J. Deisenhofer for helpful discussions. This work has partly been supported by the DFG and EuroMagNET II (EU contract No. 228043). A portion of this work was performed at the National High Magnetic Field Laboratory, Tallahassee, FL, which is supported by NSF Cooperative Agreement No. DMR-0654118, by the State of Florida, and by the DOE.

References

1. W. Shiramura, K. Takatsu, H. Tanaka, K. Kamishima, M. Takahashi, H. Mitamura, T. Goto, J. Phys. Soc. Jpn. **66**, 1900 (1997)
2. M. Jaime, V.F. Correa, N. Harrison, C.D. Batista, N. Kawashima, Y. Kazuma, G.A. Jorge, R. Stein, I. Heinmaa, S.A. Zvyagin, Y. Sasago, K. Uchinokura, Phys. Rev. Lett. **93**, 087203 (2004)
3. H. Kageyama, K. Yoshimura, R. Stern, N. Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C.P. Slichter, T. Goto, Y. Ueda, Phys. Rev. Lett. **82**, 3168 (1999)
4. T. Nikuni, M. Oshikawa, A. Oosawa, H. Tanaka, Phys. Rev. Lett. **84**, 5868 (2000)
5. H. Nojiri, H. Kageyama, Y. Ueda, M. Motokawa, J. Phys. Soc. Jpn. **72**, 3243 (2003)
6. V.N. Glazkov, A.I. Smirnov, H. Tanaka, A. Oosawa, Phys. Rev. B **69**, 184410 (2004)
7. S.A. Zvyagin, J. Wosnitza, J. Krzystek, R. Stern, M. Jaime, Y. Sasago, K. Uchinokura, Phys. Rev. B **73**, 094446 (2006)
8. S.E. Sebastian, P. Tanedo, P.A. Goddard, S.-C. Lee, A. Wilson, S. Kim, S. Cox, R.D. McDonald, S. Hill, N. Harrison, C.D. Batista, I.R. Fisher, Phys. Rev. B **74**, 180401(R) (2006)
9. T. Nakajima, H. Mitamura, Y. Ueda, J. Phys. Soc. Jpn. **75**, 054706 (2006)
10. M. Kofu, J.-H. Kim, S. Li, S.-H. Lee, H. Ueda, Y. Qiu, H.-J. Kang, M.A. Green, Y. Ueda, Phys. Rev. Lett. **102**, 037206 (2009)
11. A.A. Aczel, Y. Kohama, M. Jaime, K. Ninios, H.B. Chan, L. Balicas, H.A. Dabkowska, G.M. Luke, Phys. Rev. B **79**, 100409 (2009)
12. M. Kofu, H. Ueda, H. Nojiri, Y. Oshima, T. Zenmoto, K.C. Rule, S. Gerischer, B. Lake, C.D. Batista, Y. Ueda, S.-H. Lee, Phys. Rev. Lett. **102**, 177204 (2009)
13. A.A. Aczel, H.A. Dabkowska, P.R. Provencher, G.M. Luke, J. Cryst. Growth **310**, 870 (2008)
14. S.A. Zvyagin, J. Krzystek, P.H.M. van Loosdrecht, G. Dhalenne, A. Revcolevschi, Physica B **346–347**, 1 (2004)