MA 14 Molekularer Magnetismus

Zeit: Samstag 13:45–16:45 Raum: TU H1012

MA 14.1 Sa 13:45 TU H1012

Quantum corrections for the classical density of states of Heisenberg systems — •Matthias Exler, Jochen Gemmer, and Jürgen Schnack — Universität Osnabrück, Fachbereich Physik

We present a method to add quantum corrections to the classical density of states of Heisenberg spin systems. The classical density of states is either available analytically, or we calculate it numerically using a Wang-Landau algorithm.

We use products of spin-coherent states as the source of the quantum corrections. Their energy expectation values match those of the corresponding classical spin configurations by construction. However, they are no eigenstates of the Heisenberg Hamiltonian and therefore yield energy distributions different from delta functions. We calculate moments of these spectra which can then be used to modify the classical density of states in order to get an estimate for the quantum spectrum.

We perform these calculations for different values of the spin quantum number. Therefore, we can give an impression of how the classical limit is approached with increasing spin quantum number.

MA 14.2 Sa 14:00 TU H1012

Quantum Computation Using Endohedral Fullerenes — •Boris Naydenov[1], Wofgang Harneit[1], Christian Spudat[1], Michael Scheloske[1], Heike Süss[2], and Jürg Hulliger[2] — [1]Institut für Experimentalphysik, Freie Universität Berlin, Arnimalle 14, 14159 Berlin, Germany — [2]Dept. f. Chemie und Biochemie, Universität Bern, Freiestrasse 3, 3012 Bern , Switzerland

The endohedral atom in N@C$_{60}$ and P@C$_{60}$ is in its atomic state, it is well shielded from the surrounding and it possesses a very sharp EPR signal. These molecules are very suitable for quantum computation (QC) purposes, because of their very long relaxation times and the possibility of good scalability [1]. We present here our work and demonstrate the first steps towards the QC using our fullerenes: one qubit operations, decoherence times (T$_2$ up to 150 μs) and the alignment of our molecules in 1D channels [2]. The latter provides a way to selectively address the (±3/2;±1/2) and (1/2;-1/2) transitions, giving access to all internal spin degrees of freedom.

[1] W. Harneit, Phys. Rev. A 65 (2003) 032322

[2] H.I. Süss, M. Lutz, J. Hulliger, Cryst. Eng. Comm. 4 (2002) 610

MA 14.3 Sa 14:15 TU H1012

Spin lattice relaxation rates of heterometallic wheels — •Mohammed Allalen and Jürgen Schnack — Universität Osnabrück, Fachbereich Physik, D-49069 Osnabrück, Germany

In this work we present theoretical estimates of the exchange parameters and the proton nuclear spin-lattice relaxation rate of heterometallic $\{Cr_7M\}$ wheels using exact diagonalization of the underlying Heisenberg Hamiltonian. Heterometallic $\{Cr_7M\}$ wheels are antiferromagnetically coupled molecular magnetic rings where in the original ring of eight Chromium ions ($s = 3/2$) one ion is replaced by M=Fe, Cu, Zn or Ni with spin quantum number $s = 2, 1/2, 0,$ and $s = 1$, respectively. One of our results is that the nearest neighbor coupling is almost not altered by substituting another paramagnetic ion with the exception of iron.

MA 14.4 Sa 14:30 TU H1012

Modifying the molecular magnetism of ferric stars by intermolecular interaction — •H. Rupp[1], A. Scheurer[2], S. Sperner[2], I. Bernt[2], U. Reimann[2], R.W. Saalfrank[2], and P. Müller[1] — [1]Physikalisches Institut III, Universität Erlangen-Nürnberg, E. Rommel Str. 1, 91058 Erlangen — [2]Institut für Organische Chemie, Universität Erlangen-Nürnberg, Henkestr. 42, 91054 Erlangen, Germany

In the ferric star Fe(III)(Fe(III)L$_2$)$_3$, the outer Fe ions are linked via two ligands $L = H_3CN(CH_2CH_2O)_2$ to the central one in a star-like topology. SQUID and EPR measurements revealed a strong antiferromagnetic intramolecular Heisenberg coupling $J = -40\,K$ and zero field splitting parameter $D = -0.57\,K$ resulting in a $S = 10/2$ ground state. Using 2DEG ballistic Hall probes, magnetization measurements have been performed on the two complexes $FeFe_3 * 4\,CHCl_3$ and $FeFe_3 * 3\,CH_2Cl_2$ which differ only in the solvent used for the crystallization process. In both types of crystals, chains of stars are coupled via hydrogen bonds. The strength of this intermolecular coupling depends on the solvent used. $FeFe_3 * 4\,CHCl_3$ shows step-like hysteresis at temperatures below 0.8 K but $FeFe_3 * 3\,CH_2Cl_2$ even up to 1.2 K. In addition, the two compounds differ significantly in the shape of their magnetization hysteresis. We discuss the observed features in terms of intermolecular interactions between the stars.

MA 14.5 Sa 14:45 TU H1012

^{1}H-NMR on Fe$_6$(tea)$_6$ single crystals — •Roland Leppin, Lars Schnelzer, and Bernd Pilawa — Physikalisches Institut, Universität Karlsruhe (TH)

The Fe$_6$(tea)$_6$ cluster consists of 6 Fe^{III} ions, which are linked by the (tea)-ligand (tri ethanol aminato) to a ringlike structure. The symmetry axis of the molecule is parallel to the hexagonal axis c of the crystal structure so that the molecules can be easily oriented with respect to an external magnetic field. The temperature dependence of the proton T_1 rate has been measured for various orientations of the magnetic field. These measurements reveal a strong dependence of the T_1 rate on the direction of the magnetic field. The temperature dependence of the T1 rate for perpendicular orientation ($B \perp c$) shows the characteristic variation known for powder samples *i.e.* an exponential increase of the T_1 rate at low temperature, a maximum of the T_1 rate at $\sim$ 40 K and a linear increase for $T > 120$ K. In general, the T_1 rate increases, when the field is turned towards the c direction and the T_1 peak disappears for parallel field orientation. The origin of the T_1 anisotropy will be discussed in this contribution.

MA 14.6 Sa 15:00 TU H1012

Hysteresis and Metastability of the Antiferromagnetic Heisenberg Spin-Icosahedron — •Christian Schröder[1,2], Heinz-Jürgen Schmidt[3], Jürgen Schnack[3] und Marshall Luban[2] — [1]University of Applied Sciences Bielefeld, Dept. of Electrical Engineering and Computer Engineering, Bielefeld, Germany — [2]Ames Laboratory & Iowa State University, Dept. of Physics, Ames, Iowa, USA — [3]University of Osnabrück, Dept. of Physics, Osnabrück, Germany

Mounting on the vertices of an icosahedron classical or quantum spins that interact via nearest-neighbor isotropic antiferromagnetic Heisenberg exchange one generates a system that exhibits surprising physical properties when it is placed in an external magnetic field. At $T = 0$ K the classical system undergoes an abrupt magnetic transition at a critical field B_{crit} reminiscent of a first order phase transition. Moreover, this system shows hysteresis effects and metastabilty with an infinite lifetime at $T = 0$ K and a characteristic lifetime distribution for finite T. These properties of the spin-icosahedron may also be of interest for potential applications in the area of nanomagnetic switches. Here, we present a full analytical description of the classical behavior for $T = 0$ K and discuss finite temperature results from numerical quantum and classical calculations.

MA 14.7 Sa 15:15 TU H1012

Quantendynamik des Néel-Vektors in antiferromagnetischen molekularen Rädern und Gittern — •O. Waldmann, C. Dobe und H.-U. Güdel — Departement für Chemie und Biochemie, Universität Bern, CH-3012 Bern, Schweiz.

Die Dynamik der Untergittermagnetisierungen in Antiferromagneten kann bei tiefen Temperaturen durch den Néel-Vektor beschrieben werden. Wir untersuchten die Néel-Vektor-Dynamik in den molekularen Rädern Cr8 und CsFe8 und in dem Gitter Mn-[3 × 3] mittels inelastischer Neutronenstreuung (INS) und/oder Hochfeld-Drehmomentmagnetometrie. Im Cr8-Rad, welches eine nur schwache magnetische Anisotropie aufweist, konnten wir die quantisierte Rotation des Néel-Vektors mittels INS zweifelsfrei beobachten (PRL **91**, 237202, 2003) - 50 Jahre nach der Vorhersage dieser Anregungsmode von Antiferromagneten durch Anderson. Im CsFe8-Rad führt eine ausgeprägte easy-axis-Anisotropie zu einer neuen Situation. Hier konnten wir Tunneln des Néel-Vektors nachweisen; die Analyse der Tunnelresonanz im INS-Spektrum demonstriert dessen Kohärenz. Damit ist uns die erstmalige Beobachtung von mesoskopischer Quantenkohärenz in einem antiferromagnetischen Cluster gelungen (cond-mat/0410447). Im Mn-[3 × 3]-Gitter beobachteten wir spektakuläre Oszillationen in der Feldabhängigkeit des Drehmoments (PRL **92**, 096403, 2004). Diese neuartigen Quanten-Magnetooszillationen resultieren aus dem Wechselspiel zwischen Rotation des Néel-Vektors, magnetischer Anisotropie und angelegtem Magnetfeld. Diese Arbeiten zeigen die Vielfalt von neuartigen Quantenphänomenen in antiferromagnetischen Nanomolekülen auf.

MA 14.8 Sa 15:30 TU H1012

Spin transition and superexchange coupling in metallo-organic assemblies — •Yves Bodenthin[1], Guntram Schwarz[2], Dirk G. Kurth[2,3], Helmuth Möhwald[2], and Ullrich Pietsch[1] — [1]Institute of Physics, University of Potsdam, P.O. Box 601553, D-14415 Potsdam, Germany — [2]Max-Planck-Institute of Colloids and Interfaces, D-14424 Golm, Germany — [3]National Institute for Materials Science 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

We report on a novel approach to cause spin crossover by introducing mechanical strain through an amphiphilic phase transition in a metallo-supramolecular architecture. Using SQUID and temperature resolved XMCD (x-ray magnetic circular dichroism) we observed a diamagnetic to paramagnetic phase transition which is connected to a phase transition of the amphiphilic layers within the superstructure. It was investigated by EXAFS, x-ray reflectivity and in-plane diffraction that this structural phase transition induces sufficient mechanical strain to distort the octahedral coordination sphere of the tightly coupled Fe^{2+} ions. Consequently, the energetic separation of the subsets of the iron d-orbitals changes and gives rise to spin transition from S=0 to S=2. Ferromagnetic behavior of PAC was observed above the spin-crossover temperature by at a system with a reduced number of amphiphilic molecules. The appearance of molecular ferromagnetism in these supra-molecular structures can be explained by a super-exchange mechanism. Using element specific XMCD we observed a positive spin density at the iron ion and a negative spin density located at the ligand. This model is confirmed by first ab-initio calculations in term of density-functional approach.

MA 14.9 Sa 15:45 TU H1012

Pressure dependence of superexchange mediated antiferromagnetism in $X\mathrm{Cl}_2(\mathrm{PM})_2$ ($X = $ Fe, Co, Ni; PM = pyrimidine) — •S. Süllow[1], J. Kreitlow[1], A.U.B. Wolter[1], D. Menzel[1], J. Schoenes[1], K. Doll[2] und R. Feyerherm[3] — [1]Institut für Festkörperphysik, TU Braunschweig, 38106 Braunschweig — [2]Institut für Mathematische Physik, TU Braunschweig, 38106 Braunschweig — [3]Hahn-Meitner-Institut, 14109 Berlin

We present measurements of the magnetic susceptibility $\chi(T)$ on the pyrimidine bridged transitions metal complexes $X\mathrm{Cl}_2(\mathrm{PM})_2$ ($X = $Fe,Co,Ni; PM = pyrimidine = $C_4N_2H_4$) under externally applied pressure. From our data we derive the pressure response of the antiferromagnetic phase transition temperatures (ambient pressure values of T_N: 6.4 K for $X = $Fe, 4.7 K for Co and 15.9 K for Ni). For the three materials the transition temperatures increases by about 15% at a highest pressure of $\sim$8kbar. We compare our data with electronic structure calculations and find very good agreement between the theoretically predicted pressure response of T_N and the bulk module of $\mathrm{FeCl}_2(\mathrm{PM})_2$, $B_0 = 15\,\mathrm{GPa}$, with our experimental observations.

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MA 14.10 Sa 16:00 TU H1012

Simulation of Frequency Domain Magnetic Resonance Spectra on Molecular Magnets — •Nadeschda Kirchner, Joris van Slageren, and Martin Dressel — 1. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart

The FDMRS-technique is an advanced method that allows direct determination of the Zero Field Splittings (ZFS) in magnetic molecules in absence of an external magnetic field. Here we report a FDMRS study on $(\mathrm{PPh}_4)[\mathrm{Mn}_{12}\mathrm{O}_{12}\ (\mathrm{O}_2\mathrm{CEt})\ _{16}(\mathrm{H}_2\mathrm{O})_4]$, $[\mathrm{Mn}_9\mathrm{O}_7(\mathrm{OAc})\ _{11}\ (\mathrm{thme})(\mathrm{py})\ _3\ (\mathrm{H}_2\mathrm{O})\ _2]$, $[\mathrm{Ni}(\mathrm{Himpy})\ _2\mathrm{NO}_3](\mathrm{NO}_3)$ and $[\mathrm{Ni}_4\ (\mathrm{MeOH})\ _4\ (\mathrm{sae})\ _4]$ complexes, in powder form. The FDMRS-spectra of these molecules were simulated in the strong-exchange limit within the ground spin multiplet for all complexes mentioned above. The ZFS-parameters were fitted to the absorption line positions observed in the FDMRS-experiment; they correlate well with those obtained by INS measurements.

We are currently exploring the simulation of FDMRS spectra beyond the strong exchange/spin only approximation. On one hand we are interested in how the simulated magnetic resonance spectrum changes when considering the isotropic exchange interaction and magnetic anisotropy. On the other hand we are interested in orbitally degenerate systems. As an example the results obtained for the $[\mathrm{Ni}_4\ (\mathrm{MeOH})\ _4\ (\mathrm{sae})\ _4]$ cluster will be shown. The comparison of the FDMRS-measurements with their simulation resulting from irreducible tensor operator calculations and the discussion of the origin of magnetic anisotropy in the cluster will be presented.

MA 14.11 Sa 16:15 TU H1012

First-principles DFT calculations of the magnetic anisotropy in transition metal compounds — •Jens Kortus[1], Jordi Cirera[2], and Eliseo Ruiz[2] — [1]Institut de Physique et Chimie des Materiaux de Strasbourg, 23 rue du Loess, F-67034 Strasbourg Cedex 2, France — [2]University of Barcelona, Department of Inorganic Chemistry, Diagonal 647 08028 Barcelona, Spain

We performed first-principles density-functional based calculations on the nature of electronic states and the magnetic coupling of single molecule magnets (SMM) using the massively parallel cluster code NRLMOL [1], which has been used successfully to study several SMM in the past [2], in order to calculate the second order magnetic anisotropy parameters for several mono- and polynuclear transition metal systems. The theoretical results will be compared with available experimental data. Further, we investigate the effect of different basis sets and DFT functionals on the numerical values. Using simple model systems we explore structural dependencies of the magnetic anisotropy on different distortion pathways.

The ability to predict computationally the magnetic anisotropy energy should allow for a microscopic understanding of the magnetic anisotropy parameters based on the electronic structure of SMM, which is crucial for a rational design of molecular magnets.

[1] M. R. Pederson, D. V. Porezag, J. Kortus and D. C. Patton, phys. stat. sol. (b) 217, 197 (2000)

[2] J. Kortus, M. R. Pederson, T. Baruah, N. Bernstein, and C. S. Hellberg, Polyhedron 22, 1871-1876 (2003)

MA 14.12 Sa 16:30 TU H1012

Many-body Landau-Zener effect at fast sweep — •Dmitry Garanin and Rolf Schilling — Institut für Physik, Johannes Gutenberg-Universität, D-55099 Mainz, Germany

We investigate the influence of interaction between tunneling particles and disorder on their avoided-level-crossing transitions in the fast-sweep limit. Whereas the results confirm expectations based on the mean-field arguments that ferromagnetic/antiferromagnetic couplings suppress/enhance transitions, we found large deviations from the mean-field behavior for dipole-dipole interactions (DDI) in molecular magnets Mn-12 and Fe-8. For ideal crystals of the needle, spherical, and disc shapes DDI tends to enhance transitions. This tendency is inverted for the needle shape in the presence of even small disorder in the resonance fields of individual particles, however.