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MEASUREMENTS OF MAGNETIC RELAXATION PROCESSES IN QUASICRYSTALS

Z. Jagličić[*,] — J. Prizmič[***] — J. Dolinšek[***,****] — Z. Trontelj[*,***]**

The nature of spin-glass-like transition in icosahedral quasicrystals Tb-Mg-Zn and Tb-Mg-Cd was studied by the thermoremanent magnetization (TRM) decay as a function of magnetic field. The observed linear dependence of the TRM on the magnetic field in the low-field regime demonstrates difference to canonical spin glasses.

Keywords: quasicrystal, magnetic properties, spin glass

1 INTRODUCTION

In classical crystallography a crystal is defined as a three dimensional periodic arrangement of atoms with translational periodicity along the three principal axes. Thus it is possible to obtain an infinitely extended crystal structure by aligning unit-cells until the space is filled up. Among the consequences of periodicity is the fact that the only rotational symmetries that are possible are 2-, 3-, 4-, and 6-fold rotations.

However, in 1982 Dan Shectman observed electron diffraction patterns of a rapidly cooled Al-Mn alloy, which showed six, so called forbidden, 5-fold symmetry axes. Along with the 3- and 2- fold symmetry axes he concluded that the alloy had the symmetry of an icosahedron [1]. The symmetry of an icosahedron does not permit to build up a crystal by periodically aligning unit-cells. Soon, many other structures (*eg* Al-Pd-Mn, Al-Cu-Fe, RE-Mg-Zn(Cd), RE= rare earth) with the same or other in classical crystallography forbidden symmetry properties (8-, 10-, 12-fold symmetry axes) were discovered and they are called quasicrystals. In spite of the aperiodicity, the positions of the atoms in quasicrystal structure is exactly defined - the quasicrystals posses a long-range order.

Quasicrystals have characteristic physical properties. Some properties resemble those of periodic crystals, for instance the morphology of quasicrystals; some are rather similar to the properties of amorphous alloys. The latter, for example, applies to the transport anomalies observed in quasicrystals. Most quasicrystals are alloys of elements that are good metals and one would expect high conductivity. But contrary, quasicrystals are poor conductors, and their electric conductivity increases with increasing temperature. Quasicrystals are very poor thermal conductors. At room temperature they are hard and brittle.

Quasicrystals can be used as materials for surface coatings. As hard and thermal isolating thin protective layers they are already exploited in mechanical engineering.

2 MAGNETIC PROPERTIES OF QUASICRYSTALS AND SPIN GLASSES

Although many Al-based quasicrystals contain transition metals, which carry localized magnetic moments, they usually do not seem to exhibit magnetic ordering, and mostly show weak paramagnetic or diamagnetic behaviour. For quasicrystals of a high structural quality, magnetic moments appear to be effectively screened, and only a small fraction of all transition metal atoms in the structure actually demonstrates a non-zero magnetic moment [2]. In Al-based quasicrystals *d* electrons of the transition metal elements represent the basic reorientable magnetic dipoles, which are difficult to classify as localized or itinerant in an unambiguous way.

The magnetic structure is much more defined in the RE-containing icosahedral families RE-Mg-Zn and RE-Mg-Cd. The magnetic moments of the *f* electrons of the rare-earth elements are sizable and well localized in these quasicrystals.

Many of the RE-containing quasicrystals exhibit spin-glass-like freezing: *eg* a large difference between field-cooled and zero-field-cooled magnetic susceptibilities below a freezing temperature T_f, a frequency-dependent cusp in the ac susceptibility and slow thermoremanent magnetization time decay [3,4,5,6].

In order to be classified as a spin glass, a spin system must posses two fundamental properties[7]: a) frustration (the interaction between spins is such that no configuration can simultaneously satisfy all the bonds and minimize the energy at the same time) and b) randomness (the spins are positioned randomly in the sample). In canonical spin glasses (dilute magnetic alloys formed from a noble metal host and a magnetic impurity, such as eg Cu-Mn) the interaction between spins is the indirect, conduction-electrons mediated Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction. This interaction oscillates in space and can be either ferromagnetic or antiferromagnetic, depending on the distance between spins. Combined with randomness, the RKKY interaction results in frustration.

[*] Institute of Mathematics, Physics and Mechanics, Jadranska 19, 1000 Ljubljana, Slovenia, E-mail: zvonko.jaglicic@uni-lj.si, [**] Faculty of Civil and Geodetic Engineering, University of Ljubljana, Jamova 2, Ljubljana, Slovenia, [***] Faculty of Mathematics and Physics, University of Ljubljana, Jadranska 19, 1000 Ljubljana, Slovenia, [****] Jožef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia.

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One of the consequences of frustration and randomness in canonical spin-glasses is a highly degenerate free-energy landscape. The most direct evidence of such a free-energy landscape is the observation of slow relaxation effects in the thermoremanent magnetization (TRM) time decay and characteristic dependence of the amplitude of the TRM on the temperature, time and magnetic field.[8,9]

In the TRM experiment (see also Fig. 1) one cools down the sample in a small magnetic field H_{fc} (up to 100 Oe) from above transition temperature T_f to a measuring temperature $T_m < T_f$, where one lets the system age for time t_w. The spin system reaches an equilibrium magnetization M_0 which is proportional to the H_{fc}. At the end of the waiting time t_w, the field H_{fc} is cut to zero and the TRM decay is measured as a function of time. The TRM is of the order of 10% of the M_0 immediately after the field is cut off and decays very slowly.

The amplitude of the TRM M_{TRM} recorded at a fixed time after the H_{fc} is cut off increases with waiting time t_w in spin glasses. However, the dependence of a relative amplitude of the TRM M_{TRM}/M_0 on the field H_{fc} in canonical spin glasses such as CuMn is just opposite: the M_{TRM}/M_0 increases with decreasing the H_{fc}. Such behaviour is a direct consequence of a highly degenerate free-energy landscape. It is theoretically explained in terms of the ultra-slow spin dynamics in an ultrametric free energy and experimentally verified. [9]

The basic open question here is whether the RE-containing quasicrystals fall into the same universality class as canonical spin glasses or whether the spin-glass-like phase in the RE-containing quasicrystals is fundamentally different. The interaction between spins in quasicrystals is still the spatially oscillating RKKY interaction. So one of the two basic requirements for canonical spin glasses - namely frustration - is fulfilled. But the second requirement - randomness - is not fulfilled in quasicrystals as magnetic moments of the RE atoms are at well defined positions in the quasicrystalline lattice, which is long-range ordered.

It has been discovered that spin-glass-like phases develop also in systems where randomness is absent. [10] These are geometrically frustrated antiferromagnets where triangular or tetrahedral distribution of nearest-neighbor antiferromagnetically coupled spins frustrates a nonrandom, perfectly ordered periodic system. Many of their magnetic properties resemble the situation in canonical spin glasses. However, the important difference is a formation of magnetic clusters in the geometrically frustrated systems. In the case of non-interacting clusters such system may be viewed as a superparamagnet. Though superparamagnetic clusters below the blocking temperature exhibits several similar features as spin-glasses, they show difference in the most basic physical property - the free energy landscape. For spin-glasses the free energy landscape is highly degenerate while in superparamagnets it exhibits a single global minimum. The aim of our study is to throw some additional light on the nature of spin-glass-like transition in RE-containing quasicrystals that belong to the frustrated but geometrically ordered systems.

3 MEASUREMENTS OF TRM

Two members of the RE-containing icosahedral families RE-Mg-Zn(Cd) were used in our study: $Tb_9Mg_{34}Zn_{57}$ (Tb-Mg-Zn) and $Tb_{11.6}Mg_{35.2}Cd_{53.2}$ (Tb-Mg-Cd). The Tb-Mg-Zn quasicrystal was grown by the self-flux technique[3] while the Tb-Mg-Cd was prepared by a high-frequency induction melting method [5].

Magnetization as a function of magnetic field and time, and susceptibility as a function of temperature were measured with a Quantum Design MPMS-XL-5 SQUID magnetometer and a home made SQUID susceptometer.

Both quasicrystals exhibit a spin-glass-like transition at T_f = 5.8 K and 12.5 K in Tb-Mg-Zn and Tb-Mg-Cd, respectively. We have done many TRM time decay measurements changing measuring temperature T_m, waiting time t_w and magnetic field H_{fc} in both quasicrystals. A typical TRM time-decay experiment is shown in Fig. 1.

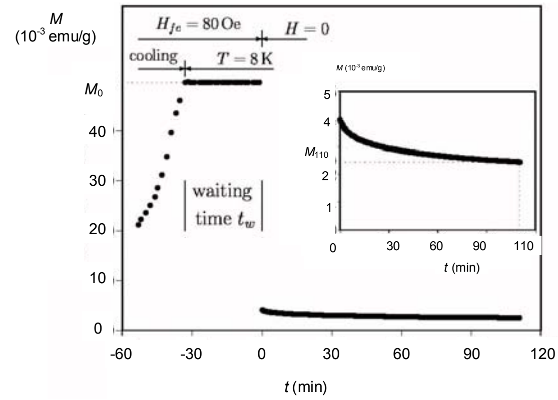

Fig. 1. A typical TRM time-decay experiment. The Tb-Mg-Cd quasicrystal was cooled from above T_f to a measuring temperature T_m = 8 K which is below T_f, where one lets the system age for a time t_w = 30 min. At t = 0 the field was cut to zero and the magnetization decay was measured as a function of time (enlarged in the inset).

The amplitude of the TRM recorded at a fixed time after the H_{fc} is cut off increases with t_w. This is in agreement with spin-glass-like behaviour.

However the dependence of the amplitude of the TRM M_{TRM} on the magnetic field H_{fc} does not follow the spin-glass prediction. Fig. 2 shows several TRM time-decay experiments measured in Tb-Mg-Zn quasicrystals in different magnetic field H_{fc}, while t_w and T_m were not changed. One can observe increase of the M_{TRM} with increasing field H_{fc}.

We measured the M_{TRM} at fixed time after the magnetic field was cut off and plot it against the field H_{fc}. Two sets of measurements, one for each of the two studied quasicrystals, are shown in Fig. 3. Two essential information can be obtained from this plot. The first is obvious linear M_{TRM} vs. H_{fc} dependence. The second is the fact that linear $M_{TRM}(H_{fc})$ fit does not cross the y-axes through the zero magnetization value. The reason is a remanent magnetic field of the order of few tenths of Oe in a superconducting coil of the MPMS magnet. This field causes small additional, independent on H_{fc}, magnetic signal measured when the field should be zero . Despite the obvious linear $M_{TRM}(H_{fc})$ dependence, the plot of a relative amplitude M_{TRM}/M_0 vs. H_{fc} could demonstrates an increase for smaller H_{fc} (this misleading plot is not shown here). That happens because the constant magnetic signal is relatively more effective for low H_{fc}.

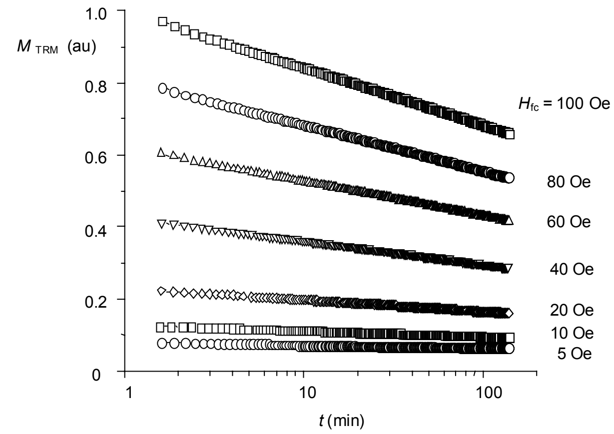

Fig. 2. The TRM time-decays in Tb-Mg-Zn quasicrystal in several magnetic fields H_{fc}. Only the magnetization after the magnetic field was cut off is shown here. t_w = 30 min and T_m = 4.0 K for all measurements.

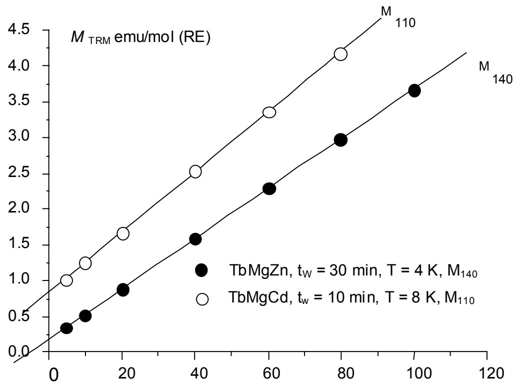

Fig. 3. Thermoremanent magnetization M_{140} 140 min (Tb-Mg-Zn) and M_{110} 110 min (Tb-Mg-Cd) after the magnetic field H_{fc} was turned off.

To confirm the linear $M_{TRM}(H_{fc})$ dependence we have performed a new set of measurements with a home made SQUID susceptometer where the magnetic field is generated with a non-superconducting cooper coil. In this way we completely eliminated the effect of the remanent magnetic field of superconducting coil. For the measurements in small magnetic fields we used the Quantum Design MPMS system again. The field was generated with a non-superconducting cooper coil that is built in the MPMS system for ac measurements. We wrote a programming sequence to control a dc current through this coil. The maximal field we can reach in this way is ±8 Oe.

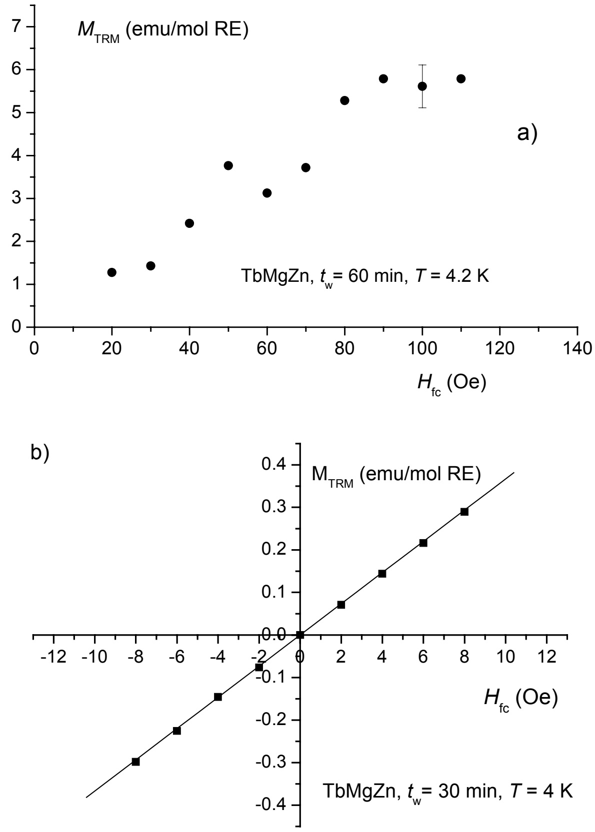

Fig. 4. TRM measured with a home made SQUID susceptometer (a) and with a MPMS system (b). In both cases the magnetic field was generated with a non-superconducting cooper coil. A magnetic shield from an environmental noise is not so sophisticated in the home made SQUID susceptometer as in the MPMS system. This is a reason for larger deviation of the data points obtained with a home made susceptometer from the liner fit (a).

The results of the TRM time decay measurements with a home made SQUID susceptometer and with a MPMS system using the cooper coil are shown in Fig. 4. The linear fits of $M_{TRM}(H_{fc})$ now show no offset at $H_{fc} = 0$ and confirm the linear $M_{TRM}(H_{fc})$ dependence with no increase of M_{TRM}/M_0 for small H_{cf}.

4 CONCLUSIONS

Magnetic properties of two icosahedral Tb-Mg-Zn(Cd) quasicrystals were studied. These quasicrystals belong to the class of geometrically-frustrated magnetic systems. Some of their magnetic properties can be attributed to the canonical spin-glass behaviour and to the superparamagnets. However, the linear relation between M_{TRM} and H_{fc} is essentially different from the canonical spin-glass-like behaviour. It is compatible with a single global minimum in the free energy landscape of superparamagnets where M_{TRM} is proportional to H_{fc}.

References

[1] D. SHECHTMAN, I. BLECH, D. GRATIAS, Phys. Rev. Lett. **53**, 1984, p. 1951.

[2] J. DOLINŠEK, M. KLANJŠEK, Z. JAGLIČIĆ, A. BILUŠIĆ, A. SMONTARA, J. Phys.: Condens. Matter **14**, 2002, p 6975.

[3] I.R.FISHER, K.O. CHEON, A.F. PANCHULA, P. CANFIELD,M. CHERNIKOV, H.R. OTT, K. DENNIS, Phys. Rev. B **59**, 1999, p 308.

[4] T.J. SATO, H. TAKAKURA, A.P. TSAI, K. SHIBATA, K. OHOYAMA, K.H. ANDERSEN, Phys. Rev. B **61**, 2000, p 476.

[5] T.J.SATO, J. GUO, A.P.TSAI, J. Phys: Condens. Matter **13**, 2001, L105.

[6] J. DOLINŠEK, Z. JAGLIČIĆ, M. A. CHERNIKOV, I. R. FISHER, P. C. CANFIELD, Phys. Rev. B **64**, 2001, p 224209.

[7] K. BINDER, A. P. YOUNG, Rev. Mod. Phys. **58**, 1986, p 801.

[8] M. LEDERMAN, R. ORBACH, J.M. HAMMANN, M. OCIO, E. VINCENT, PHYS. REV. B **44**, 1991, P 7403.

[9] D. CHU, G.G. KENNING, R. ORBACH, Philos. Mag. B **71**, 1995, p 479.

[10] A.S. WILLS, V. DUPOIS, E. VINCENT, J. HAMMANN, R. CALEMCZUK, Phys. Rev. B, 2000, p R9264.

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Zvonko Jagličić, born in 1966, in Murska Sobota, Slovenia. Graduated from the Faculty of Mathematics and Physics of the University of Ljubljana in 1990. PhD studies completed in 1996. He works in solids state physics (magnetism, SQUID sensors) and teaches physics at the Faculty of Civil and Geodetic Engineering in Ljubljana.