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Editors:

Dieter Ziessow

Wolfgang Lubitz

Friedhelm Lendzian

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EPR of Incommensurate Phases in Ferroelastic $MgSiF_6 \cdot 6H_2O : Mn^{2+}$: New Approaches

P.G. Skrylnik and A.M. Ziatdinov
Institute of Chemistry, Far-East Branch of RAS, Vladivostok 690022, Russia

The single crystals of improper ferroelastic $MgSiF_6 \bullet 6H_2O$ have been investigated by means of X - band EPR on the admixture ions Mn^{2+}. At $T_{i1} \cong 370K$ and $T_C \cong 299K$ the crystals undergo phase transitions from paraelastic phase to incommensurate phase and from incommensurate phase to ferroelastic phase, respectively [1]. The temperature decrease below T_{i1} is accompanied by smooth inhomogeneous broadening followed by splitting of the Mn^{2+} admixture ions lines of EPR spectra hyperfine structure. Moreover, the EPR lineshape is typical for incommensurate one-dimensional modulated systems with quadratic modulation of an order parameter. The inhomogeneous phase similar to that in magnesium fluorosilicate have been observed in mixed single crystals $Mg_xZn_{1-x}SiF_6 \bullet 6H_2O$. It is worth mentioning that the phase forms at $x \approx 20\%$, indicating on a special role of Mg^{2+} ions in the incommensurability emersion.

The calculations of the temperature dependence of modulation parameter h_2 ($h_2 \sim (T_{i1} - T)^{2\beta}$) allow to determine the critical index $\beta = 0{,}36 \pm 0{,}02$ of a transition to incommensurate phase. The lineshape in inhomogeneous phase is formed mainly by the modulation ΔD of the fine structure parameter D. However, the angular dependence of the lineshape shows that the angle φ of complex ions orientation around crystal axis C_3 must be a primary order parameter. This statement seems to be physically reasonable and accounts for the principal experimental data. We suppose that parameter D is connected with φ. For the reasons of symmetry, we should conclude that $\Delta D \sim \varphi^2$ (quadratic case).

A smooth evolution of incommensurate phase follows to the predictions of a classical theory [2] between T_{i1} and T_{i2} ($T_{i2} \cong 343K$) : the modulation follows from a plane-wave regime to multisoliton regime with decreasing soliton density n_S at $T \to T_{i2}$. Below T_{i2} the spectra lineshape changes differ essentially from magnetic resonance spectra evolution in conventional incommensurate systems with one-dimensional modulation. The calculations performed in particular show that n_S undergoes a step-wise decrease at T_{i2} to a small value about $\approx 0{,}1$ (Fig. 1). A successful description of the lineshape may be in principle obtained between T_{i2} and $T_C + 10K$ taking into account the variation of spin - lattice relaxation rate T_1^{-1} over the incommensurate spectral distribution.

This phenomenon was predicted theoretically and observed experimentally by direct

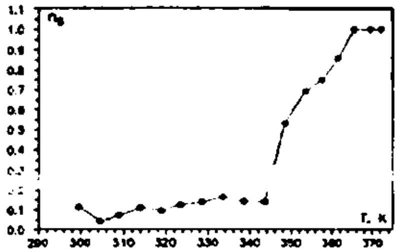

Fig. 1. The temperature dependence of soliton density n_S for the pattern taking into account variation of T_1^{-1} over the spectrum.

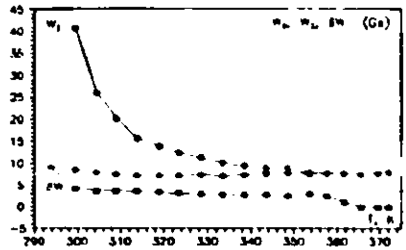

Fig. 2. The temperature dependence of the parameters W_0, W_2 and δW of linewidth variation induced with T_1^{-1} variation.

T_1^{-1} measurements in some compounds [2, 3]. The reason for such a variation may be different contributions of amplitudon and phason fluctuations to the T_1^{-1} at different parts of the inhomogeneous magnetic resonance lines [2, 3]. However, below $T \approx T_C + 10K$ the value of variation indicated becomes too large (Fig. 2. Parameters W_0 and W_2 indicate the value of Lorentzian linewidth at the points of incommensurate spectral distribution singularities, δW is a deviation from linear function in a proposed parabolic dependence $W(H)$).

Therefore, we have proposed and examined another pattern of reversible crystal division into subsystems of domains and soliton-like areas too. The model allows to describe rather satisfactorily the experimental spectra with two distinct components, from T_{i2} to T_C. It is notable that soliton-like component lineshape is typical for multidimensional 2q (or 3q) incommensurate modulation rather than one-dimensional case at $T > T_{i2}$. The step-wise changes in parameters describing EPR spectrum lineshape at T_{i2} may be attributed to the presence of nearly independent aforementioned subsystems below T_{i2}. The soliton density n_S is almost invariable within some temperature range. However, the interactions between these subsystems probably restores at $T \rightarrow T_C$ resulting in soliton density n_S decrease (close by T_C) followed by a transition to commensurate ferroelastic phase.

[1] A.M. Ziatdinov and V.G. Kuryavyi, Ferroelectrics, 143, 99 - 107 (1993).

[2] Incommensurate Phases in Dielectrics, edited by R. Blinc and A.P. Levanyuk, North - Holland, Amsterdam, (1986).

[3] R. Blinc, T. Apih, J. Dolinsek, U. Mikac, D.C. Ailion and P.-H. Chan, Physical Review, B51, 1354 - 1357 (1995).