Separation of the magnetic phases at the Néel point in the diluted spin-Peierls magnet CuGeO₃

V. N. Glazkov, A. I. Smirnov
P. L. Kapitza Institute for Physical Problems RAS, 117334 Moscow, Russia

K. Uchinokura, T. Masuda
Department of Advanced Materials Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
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The impurity induced antiferromagnetic ordering of the doped spin-Peierls magnet Cu₁₋ₓMgₓGeO₃ was studied by electron spin resonance (ESR) technique. Crystals with the Mg concentration x < 4% demonstrate a coexistence of paramagnetic and antiferromagnetic ESR modes. This coexistence indicates the separation of a macroscopically uniform sample in the paramagnetic and antiferromagnetic phases. In the presence of the long-range spin-Peierls order (in a sample with x = 1.71%) the volume of the antiferromagnetic phase immediately below the Néel point Tₐ is much smaller than the volume of the paramagnetic phase. In the presence of the short-range spin-Peierls order (in samples with x = 2.88%, x = 3.2%) there are comparable volumes of paramagnetic and antiferromagnetic phases at T = Tₐ. The fraction of the antiferromagnetic phase increases with lowering temperature. In the absence of the spin-Peierls dimerization (at x = 4.57%) the whole sample exhibits the transition into the antiferromagnetic state and there is no phase separation. The phase separation is explained by the consideration of clusters of staggered magnetization located near impurity atoms. In this model the areas occupied by coherently correlated spins expand with decreasing temperature and the percolation of the ordered area through a macroscopic distance occurs.

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I. INTRODUCTION

Quasi-one-dimensional magnet CuGeO₃ is a unique inorganic compound demonstrating a spin-Peierls phase transition. The spin-Peierls transition may occur in a crystal containing spin S = 1/2 antiferromagnetic chains due to the spin-lattice instability with respect to the dimerization of magnetic ions. Below the transition temperature Tₛₚ = 14.5 K the lattice period along the chain direction becomes doubled and the exchange integral alternates taking in turn two values J ± δJ. Due to this alternation the ground state is a singlet separated from the excited triplet states by an energy gap Δ = 2 meV. Thus at low temperatures pure crystals appear to be almost nonmagnetic and a small residual magnetic susceptibility is provided only by defects. The amplitude of the atomic displacements resulting in the dimerization can be chosen as the order parameter of the spin-Peierls phase. Impurities substituting magnetic or nonmagnetic ions disturb the homogeneity of the spin-Peierls phase in CuGeO₃. The doping diminishes the transition temperature and results in an antiferromagnetic long-range ordering.

The spin-Peierls dimerization and the impurity-induced magnetic order were found to coexist at low impurity concentration x. Stimulation of the long-range antiferromagnetic order by impurities was explained in Refs. 8–11. The violation of the dimerization around an impurity results in the formation of a soliton-like spin cluster with an antiferromagnetic correlation of neighboring spins and staggered magnetization. Overlapping of clusters and the weak interchain exchange result in the long-range three-dimensional antiferromagnetic order.

The phase transition to the antiferromagnetic state and T − x phase diagram were studied for different types of doping atoms.14,16 For antiferromagnetic phases stimulated by the impurities of Zn, Mg, Si the easy axis of the ordered state is c axis, and for Ni-doping - the a axis. The phase diagram contains areas of a uniform (i.e., without dimerization) paramagnet, of a dimerized paramagnet (i.e., of spin-Peierls state), and of the antiferromagnetic state. It was shown experimentally that there is no threshold concentration of impurities for the occurrence of the magnetic order.17 Further detailed investigations revealed subtle features of the phase diagram shown on the Fig. 1 for Mg-doped crystals of CuGeO₃ following Ref. 20. The first order phase transition between dimerized and uniform antiferromagnetic phases at a critical value of concentration was found.14 The transformation from the dimerized to the uniform phase in Mg-doped samples occurs in a concentration range between 2.37% and 2.71%. The uniform phase demonstrates a higher value of the Néel temperature. Further, the state with a short-range spin-Peierls order was detected for Mg-doped samples and the transition between short range and long-range ordered spin-Peierls phases was found.16 Finally, the reentrant phase transition into the undimerized state from the dimerized antiferromagnetic phase was observed.16 A theoretical consideration of the transition between dimerized and uniform antiferromagnetic phases and of the reentrant transition was given in Ref. 11.

This variety of phases is caused by the competition
between the gapped dimerized state and the antiferromagnetic state which is gapless in the exchange approximation. The spin-Peierls state does not allow the three dimensional antiferromagnetic ordering in the pure compound and, on the other hand, impurities restore the antiferromagnetic correlations and suppress the spin-Peierls dimerization.

The goal of this work is an ESR study of magnetic properties of different antiferromagnetic phases and of the phase transitions at various phase boundaries of the phase diagram. The previous ESR investigations have revealed the multispin nature of clusters formed near impurity ions. The gap in the zero-field ESR frequency in the antiferromagnetic phase. Thus the evolution from isolated clusters with local staggered magnetization to the long-range antiferromagnetic order may be followed using the ESR technique. We used single crystal samples of Cu$_{1-x}$Mg$_x$GeO$_3$ from the measurements of the phase diagram shown on Fig. 1, or the samples grown by the same method on the same installation. The phase transitions studied in the present work are marked on Fig. 1.

As a result of this study we found that the formation of the antiferromagnetic order at small impurity concentrations is accompanied by the microscopic phase separation into the paramagnetic and antiferromagnetic phases and this phase separation differs in temperature evolution for antiferromagnetic phases coexisting with a long-range and a short-range ordered spin-Peierls background.

II. SAMPLES AND EXPERIMENTAL DETAILS

The single crystals of the Cu$_{1-x}$Mg$_x$GeO$_3$ with $x = 0$, 1.71%, 2.88%, 3.20%, 4.57% were grown by the floating zone method. The impurity distribution was checked by inductively coupled plasma atomic emission spectroscopy technique and was found to be uniform within 0.1% (see Ref. 2). We used single crystals with the dimensions of about 1×2×2 mm.

The concentration of the residual magnetic defects (both of the structure and impurity type) may be estimated from measurements of the ESR intensity of a nominally pure ($x = 0$) sample (see the inset of Fig. 7). This intensity rapidly decreases below the transition temperature due to the freezing out of the gapped triplet excitations. The minimum values of the ESR intensity and of the static susceptibility observed at 4.5 K equal 5% of these values at $T_{SP}$. This intensity and susceptibility correspond to the concentration of the residual defects $x_{def} \sim 0.07\%$ per Cu-ion.

The ESR spectra were taken by means of a spectrometer with a set of transmission-type resonators. Measurements were carried out in the frequency range 9 – 75 GHz at temperatures 1.5 – 15 K. The magnetic resonance absorption line was recorded as a dependence of the transmitted microwave power on the applied magnetic field. The reduction of the transmitted signal is proportional to the microwave power absorbed by the sample. For the paramagnetic state the integrated intensity of absorption is proportional to the static susceptibility.

III. EXPERIMENTAL RESULTS

The temperature evolution of the ESR line for the sample with the impurity concentration $x = 4.57\%$ is typical of an antiferromagnet: at the decrease of the temperature starting from the Néel point ($T = T_N$) the single resonance line shifts to lower fields when the magnetic field is perpendicular to the easy axis of spin ordering (see Fig. 2).

The ESR lines of the samples with $x = 1.71\%$ and $x = 2.88\%$ are shown on the Fig. 3 and Fig. 4. At $T = T_N$ the resonance line splits into two spectral components. Similar splitting was observed for the sample with $x = 3.20\%$. Below the Néel temperature the ESR line is well described as a sum of the two Lorentzian components (see inset of Fig. 3). The component shifting to lower fields with decreasing temperature has a nonlinear field-dependence of the resonance frequency with strong anisotropy, shown in Fig. 5 for the sample with $x = 3.20\%$. We note this spectral component as an antiferromagnetic resonance line since this frequency-field dependence with two gaps is typical for two-axes antiferromagnets. The other spectral component has a linear frequency-field dependence with a temperature independent $g$-factor. We note this absorption mode as a paramagnetic resonance corresponding to the $g$-factor values $g_a = 2.14$ and $g_b = 2.21$ measured for the field orientations along $a$ and $b$ axes correspondingly. These values coincide within the experimental errors with $g$-factors obtained for pure compound.

The temperature dependence of the resonance fields at a fixed frequency is shown in Fig. 6 for the sample with $x = 2.88\%$. The temperature when the low-field line starts to shift from the paramagnetic resonance position corresponds well to the value of $T_N$ obtained by susceptibility measurements.

To obtain the ESR spectra with two resolvable components one should take the microwave frequency close to the antiferromagnetic resonance gap. In this case the difference between the paramagnetic resonance field and that of a gapped antiferromagnetic resonance mode will be more significant, helping to resolve two spectral components. As shown on Fig. 5 the spectrum of antiferromagnetic resonance of the doped CuGeO$_3$ has two branches with different gaps. The observation of the first or the second branch by the field-sweep technique depends on the field orientation. Therefore to meet the condition mentioned we selected not only the microwave frequency from the set of the resonant frequencies of the microwave resonator, but also the orientation of the magnetic field with respect to crystal axes.
ues of gaps depend also on Mg-concentration, the data for different samples are obtained at different microwave frequencies and orientations of the external field.

The temperature dependences of the integrated intensities of both components are shown on Fig. 7 for \( x = 1.71\% \), and on Fig. 8 for \( x = 2.88\% \) and 3.20\%. The remarkable feature of the two-component ESR spectrum is the large intensity of the paramagnetic line in a temperature range below the Néel point. For the low concentration \( x = 1.71\% \) the intensity of the paramagnetic line below the Néel point is close to the integral intensity above \( T_N \), and the intensity of the antiferromagnetic resonance mode is much smaller than that of the paramagnetic mode. For concentrations of \( x = 2.88\% \) and 3.20\% on the contrary, the intensity of the antiferromagnetic resonance is larger than the intensity of paramagnetic mode. For the concentration \( x = 4.57\% \) there is no distinguishable paramagnetic mode below Néel temperature. We ascribe the whole intensity to the antiferromagnetic mode and take the intensity of the paramagnetic resonance as zero. The \( x \)-dependences of the relative intensities of the paramagnetic and antiferromagnetic modes extrapolated to \( T_N \) from low temperatures are plotted in Fig. 9. Because of significant errors occurring near \( T_N \) in the determination of the intensity of a weak and wide antiferromagnetic component near the narrow paramagnetic line the intensity of the antiferromagnetic component tends to be underestimated.

IV. DISCUSSION

A. Possible reasons for the paramagnetic mode in the antiferromagnetic state

The identical values of the \( g \)-factor of the paramagnetic spectral component below and above \( T_N \) indicate that this ESR signal is due to Cu\( ^{2+} \) ions. The intensity of this component decreases with decreasing temperature, therefore it can not be ascribed to isolated Cu ions located at the surface or at structure defects. The isolated spins would show an increase in intensity according to the Curie-law.

The paramagnetic resonance signal might be due to the triplet excitations of the spin-Peierls magnet, which are present both in pure and doped crystals. However, due to the gap in the triplet excitations spectrum the intensity of this signal should rapidly drop with cooling below \( T_{SP} \). To estimate the ESR intensity provided by the triplet excitations we take the whole intensity in the temperature range \( T_N < T < T_{SP} \) to be due to the additive contributions of the impurity Curie-like part and the triplet excitation part:

\[
I(T) = \frac{C}{T-\theta} + \frac{D}{T} \exp\left(-\frac{\Delta}{kT}\right) \tag{1}
\]

Here \( C, \theta \) are the Curie constant and Curie-Weiss temperature, \( D \) is a constant depending on dimerization.

The results of the fitting of the ESR intensity by this formula for the pure sample and for the doped sample with \( x = 1.71\% \) in the temperature range between \( T_N \) and \( T_{SP} \) are shown on the inset of Fig. 7. We find that for \( x = 1.71\% \) the triplet part of the ESR intensity at the Néel point should be of about of \( 10^{-3} \) of the observed value and thus could not provide the paramagnetic resonance signal observed.

For larger concentrations the triplet part of the intensity and susceptibility is not well pronounced (there is no evident drop of the magnetic susceptibility at lowering temperature). Nevertheless we can exclude the triplet excitations for the following reasons. At \( x = 3\% \) the triplet excitations are overdamped, hence they should strongly interact with the spins near impurities and the coupling of the antiferromagnetic resonance and of the triplet mode should occur. Depending on the coupling parameter, the resulting ESR spectrum of two exchange coupled spin systems (see, e.g. Ref. 31) should consist of a single line at the intermediate frequency or of two separate lines with frequencies varying simultaneously at the change of external parameters or of the coupling coefficient. Examples of the ESR spectra of the exchange coupled systems including a spin-Peierls magnet, an antiferromagnet with impurities and thermally activated spins in organic molecules may be found in Refs. 29,32,33. In the present study we observe two-component spectrum with constant field of the paramagnetic mode and with the shift of the antiferromagnetic line. Consequently there are no traces of the mutual influence of two modes which should occur if the paramagnetic mode would be due to the triplet excitations. Thus for \( x > 2\% \) the contribution of triplet excitations to ESR intensity at the Néel temperature is also negligible.

Further we prove that the observed two-component ESR signals couldn’t be ascribed to a trivial inhomogeneous distribution of the Mg-concentration, and, hence to different values of \( T_N \) in different parts of the sample. The distribution of the Néel temperature would result in the wide band of absorption, while the observed antiferromagnetic resonance absorption is well described by a single Lorentzian. Besides that the range of the distribution of \( x \) which is necessary to account for the paramagnetic phase is much wider than obtained in control measurements. For example, to have the Néel points in the range 1.5–2.25 K where both signals are present for the \( x = 1.71\% \) sample, we should imagine the concentration distribution in the range 1.3–2.2%. The width of this range is much larger than 0.1% obtained in concentration measurements. The well defined singularities on temperature dependences of the susceptibility and of the resonance field (see Fig. 6) prove that the samples are macroscopically uniform. The width of the transition to the Néel state may be estimated from the susceptibility and ESR data, and we see that it is not larger than 0.1 K.

Summarizing the above analysis of possible reasons for the paramagnetic resonance mode below the Néel point
we state that the observed paramagnetic resonance signal cannot be ascribed to isolated uncontrolled magnetic defects, to triplet excitations or to an inhomogeneous distribution of impurities.

The uniform antiferromagnet is characterized by a single order parameter and should exhibit only antiferromagnetic resonance modes corresponding to a certain type of ordering. The experiments on numerous antiferromagnets show that at the Néel point the paramagnetic resonance converts in the antiferromagnetic resonance, if the frequency is larger than the gap of the antiferromagnetic resonance, as we observe for the sample with \( x = 4.57\% \). If the frequency is much smaller than the gap value, the ESR signal in a usual antiferromagnet disappears at the Néel point. In both cases there is no ESR signal on the paramagnetic resonance frequency below \( T_N \). For the existence of an additional paramagnetic resonance mode in the uniform antiferromagnet an additional spin degree of freedom is necessary. A hypothetical additional mode related to Cu\(^{2+}\) spins in our case should be coupled with the antiferromagnetic resonance mode. As described above, we excluded the spin degrees of freedom of the isolated defects and of the triplet excitations, and we detected the absence of any coupling of two modes. Therefore we come to the conclusion that our system is not microscopically uniform and the coexisting paramagnetic and antiferromagnetic resonance signals should originate from different areas of the sample. Thus a microscopic separation in the paramagnetic and antiferromagnetic phases takes place in macroscopically uniform samples with low impurity concentrations.

**B. Geometrical model**

We explain the microscopic phase separation at low impurity concentrations, when long-range dimerization order occurs, considering the regions of antiferromagnetically correlated spins (spin clusters) appearing near all impurity atoms. The spins within these spin clusters have nonzero average spin projections in form of staggered magnetization, thus the local Néel order parameter can be introduced. Besides that a cluster has a net magnetic moment equal to \( \mu_B \). The drops are placed in space at random with the density of drops corresponding to the value of the concentration of impurities. At high temperatures when the drops are small and do not overlap, the antiferromagnetic order parameter is nonzero within ellipsoids and zero outside them. The phases of the local order parameters of different drops are not coherent and the model shows no long-range antiferromagnetic order. At lowering temperature the drops grow and begin to overlap. The order parameter in the overlapped drops (a conglomerate of drops) is coherent, thus large areas with coherent antiferromagnetic ordering appear due to the formation of conglomerates. This model is illustrated in Fig. 10 by drawing the expanding drops in two dimensional space. One can see on Fig. 10 how the area of the coherent antiferromagnetic order penetrates through a macroscopic distance along both coordinates.

At finite temperature the coherence of the antiferromagnetic order parameter which is spatially variable on the wing of the cluster will be destroyed by thermal fluctuations. The distance \( L \) in the chain direction of the region of the coherent antiferromagnetic order parameter may be estimated from the relation:

\[
k_B T = J S^2 \exp\{-2L / \xi_c\}.
\]

The distances of the coherence along transverse directions are taken to be equal to \( \xi_c / v_i \). Therefore at finite temperature the area of the spatially coherent antiferromagnetic order may be considered as an antiferromagnetic drop of the ellipsoidal form with fixed boundaries. This drop is elongated along the spin chain direction and the ratio of the drop dimensions along and transverse to the chain is of about the ratio of corresponding exchange integrals. For CuGe\(\text{O}_3\) we have this ratio according to Refs. 3,8: \( J_b / J = 0.11 \), \( J_a / J = 0.011 \).

The size of an ellipsoidal drop enlarges when temperature is lowering according to the relation \( \xi_c / v_i \). A drop has a net magnetic moment of \( \mu_B \). The drops are placed in space at random with the density of drops corresponding to the value of the concentration of impurities. At high temperatures when the drops are small and do not overlap, the antiferromagnetic order parameter is nonzero within ellipsoids and zero outside them. The phases of the local order parameters of different drops are not coherent and the model shows no long-range antiferromagnetic order. At lowering temperature the drops grow and begin to overlap. The order parameter in the overlapped drops (a conglomerate of drops) is coherent, thus large areas with coherent antiferromagnetic ordering appear due to the formation of conglomerates. This model is illustrated in Fig. 10 by drawing the expanding drops in two dimensional space. One can see on Fig. 10 how the area of the coherent antiferromagnetic order penetrates through a macroscopic distance along both coordinates.

We see that in the process of the formation of large ordered regions there are also islands of zero order parameter and ordered drops within these islands. The ordered parameter propagates in the process of percolation of the ordered phase through a macroscopic distance. The percolation occurs here as it takes place in the phenomenon of the percolation through the randomly placed interpenetrating spheres at a critical value of the volume fraction occupied by spheres 8.

**C. Correspondence between the geometric model and experimental results**

The observed results for \( x = 1.71\% \) sample may be explained on the basis of the described model as follows. At a temperature above the Néel point the drops are small and hence isolated (Fig. 10a). All drops contribute to the Curie-type susceptibility due to their magnetic moments equal to \( \mu_B \). The dimerized spin-Peierls matrix (white
of the antiferromagnetic phase at $T = T_N$ is small when the ordering takes place at the long-range spin-Peierls order; ii) at the short-range spin-Peierls order there are comparable volumes of two phases at the $T = T_N$; iii) in the absence of spin-Peierls dimerization the whole sample becomes ordered at the transition temperature. The first conclusion is in agreement with geometrical model: the volume of a percolating thread is smaller than the sample volume (see Fig. 10c). This observation is to be compared with the result of the percolation theory for interpenetrating spheres placed at random, giving the volume fraction at the percolation point $v_c = 0.286$ for three dimensions and 0.675 for two dimensions.

We can not extrapolate the constructed model to the situation with short range dimerization order. Nevertheless it is natural to propose that in the corresponding concentration range, the behavior of the system should be intermediate between the behaviors of dimerized and uniform crystals with defects. This proposition is shown in Fig. 9 with straight lines interpolating the fraction of the antiferromagnetic phase from a small value at long range spin-Peierls order to unity at undimerized phase. This hypothesis is in a qualitative agreement with our observations. Further detailed investigations of the amount of the ordered phase just below the Néel point for different concentration are of great interest.

It is worth to note that in several previous investigations of the antiferromagnetic phase in doped CuGeO$_3$ the two-component signal was not observed. The single line may be explained here either by frequencies far from the antiferromagnetic resonance gaps or by large impurity concentrations which suppress the dimerization.

The two-component line with the spectra and temperature dependences analogous to the reported in the present work was observed in experiments with Cu$_{0.98}$Zn$_{0.02}$GeO$_3$. 

V. CONCLUSION

ESR measurements reveal the microscopic phase separation at the impurity induced antiferromagnetic ordering in the spin-Peierls magnet CuGeO$_3$. The temperature evolution of the ordered phase volume with the small volume fraction at the Néel point indicates the percolating character of the antiferromagnetic phase transition at low doping level when the antiferromagnetic and spin-Peierls order coexist.

VI. ACKNOWLEDGEMENTS

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VII. FIGURE CAPTIONS

Fig. 1. Phase diagram of the diluted Cu$_{1-x}$Mg$_x$GeO$_3$ following Ref. 21. The phase transitions studied in the present work are marked by signs •.

Fig. 2. Evolution of the ESR line for the sample containing 4.57% Mg. $\mathbf{H} \parallel a$, $f = 31$ GHz, $T_N = 4.20$K.

Fig. 3. Evolution of the ESR line for the sample containing 1.71% Mg. $\mathbf{H} \parallel b$, $f = 36$ GHz, $T_N = 2.25$K. Inset: ESR line at 1.5K and the Lorentzian components.

Fig. 4. Evolution of the ESR line for the sample containing 2.88% Mg. $\mathbf{H} \parallel a$, $f = 26.3$ GHz, $T_N = 4.14$K.

Fig. 5. The spectrum of the antiferromagnetic resonance of the 3.2% Mg doped sample at $T = 1.8$ K for three principal directions of the magnetic field with respect to crystal axes. Dashed lines represent the theoretical calculations following Ref. 27.

Fig. 6. The temperature dependences of the 26.34 GHz ESR fields for the sample containing 2.88% Mg at $\mathbf{H} \parallel a$. The signs $\nabla$ correspond to the antiferromagnetic resonance and $\bigcirc$ to the paramagnetic resonance, $\square$ - magnetic resonance above $T_N$.

Fig. 7. Temperature dependences of the intensities of 36GHz ESR spectral components at $\mathbf{H} \parallel b$. for the samples with impurity concentration $x=1.71\%$. The signs: $\nabla$-above $T_N$, $\bigcirc$ correspond to the antiferromagnetic resonance and $\square$ to the paramagnetic resonance below $T_N$. Solid lines are guide-to-eyes. Inset: 36GHz ESR intensities at $\mathbf{H} \parallel b$ for pure $\bigcirc$ and 1.71% Mg-doped $\bigcirc$ samples at $\mathbf{H} \parallel b$. Solid line 1 represents the fit by formula (1) for $x=1.71\%$, dashed and dotted lines are the first and second terms in (1), solid line 2 is the second term obtained from the fitting by formula (1) for the pure sample.

Fig. 8. Temperature dependences of the intensities of spectral components for the samples with impurity concentrations 2.88% (a), 3.20% (b). The signs: $\nabla$-above $T_N$, $\bigcirc$ correspond to the antiferromagnetic resonance and $\square$ to the paramagnetic resonance below $T_N$. Solid lines are guide-to-eyes. The Néel temperatures are marked by arrows.

Fig. 9. Relative intensities of antiferromagnetic and paramagnetic ESR components just below $T_N$ for different concentrations $x$. The lines are linear interpolations between the uniform and long range spin-Peierls orderings. The arrow indicates the volume fraction of interpenetrating spheres at the percolation point according to percolation theory.

Fig. 10. Illustration of the two dimensional modeling of the formation of the long-range antiferromagnetic order. Spin chains are directed horizontally, drops of the correlated spins are shown by grey filling, spin-Peierls matrix is white, the macroscopic group of drops is marked by black filling. The scale is given in interspin distances. The modeling is performed for $x=0.1\%$ and the following values of $L$ (in interspin distances): $a) L=16$, $b) L=34$; $c) L=50$.

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Fig. 1
Fig. 2

Magnetic Field (kOe) vs. Transmitted Power for different temperatures: 1.65K, 2.69K, 3.17K, 3.72K, 4.2K.
Fig. 3
Fig. 4

Magnetic Field (kOe) vs. Transmitted Power for various temperatures:
- 1.65K
- 2.41K
- 3.31K
- 3.70K
- 4.08K
- 4.2K

Fig. 4
Fig. 5
Fig. 7

Temperature (K) vs. Intensity (arb. units)

Inset: 

- Curve 1
- Curve 2

Temperature scale: $T/T_{SP}$
Fig. 8

Temperature (K)

Intensity (arb. units)
Fig. 9

Components' Intensities at $T_N$

Concentration of Impurities, $x$ (%)
Fig. 10

(a) $T \gg T_N$
$L = 16$

(b) $T > T_N$
$L = 34$

(c) $T = T_N$
$L = 50$