Revival of the spin-Peierls transition in $Cu_{1-x}Zn_xGeO_3$ under pressure

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Pressure- and temperature-dependent susceptibility and Raman scattering experiments on single-crystalline $Cu_{0.986}Zn_{0.014}GeO_3$ have shown an unusually strong increase of the spin-Peierls phase transition temperature upon applying hydrostatic pressure. The large positive pressure coefficient (7.5 K/GPa)—almost twice as large as for the pure compound (4.5 K/GPa)—is interpreted as arising due to an increasing magnetic frustration which decreases the spin-spin correlation length, and thereby weakens the influence of the nonmagnetic Zn substitution. [S0163-1829(98)02013-X]

I. INTRODUCTION

The spin-Peierls (SP) transition[1] in low-dimensional magnetic compounds is the magnetic analog of the well-known Peierls transition[2] in low-dimensional metals. The interest in this magnetoelastic phenomenon, which has been studied widely in organic compounds in the 1980s,[3] has been strongly renewed with the discovery of inorganic materials [$CuGeO_3$ (Ref. 4), α-NaV_2O_5 (Ref. 5)] exhibiting this unusual phase transition. In particular $CuGeO_3$ has attracted strong experimental and theoretical attention over the past few years[6]. There are at least three reasons for this: In the first place $CuGeO_3$ allows for the growth of large single crystals which permits for experiments which were difficult to perform in the past. Second, $CuGeO_3$ allows for well-controlled substitutions (e.g., $Cu_{(1-x)}Zn_xGeO_3$, $CuGe_{(1-y)}Si_yO_3$) which provides a useful method to study its structural and, in particular, its magnetic properties. A surprising observation is that the SP phase transition is extremely sensitive to substitution (5% of Zn substitution for Cu already makes the SP phase disappear). Equally surprising is that substitutions, which inherently increase the disorder in the system, lead to the formation of a long-range-ordered antiferromagnetic phase at low temperatures.[7–10]

In the third place $CuGeO_3$ cannot be considered as a "classical" spin-Peierls system. The susceptibility in the uniform phase ($T > T_{SP}$) strongly deviates from the Bonner-Fisher behavior[11,12] and the spin-Peierls transition temperature strongly increases upon applying hydrostatic pressure despite an apparently decreasing nearest-neighbor (NN) exchange interaction.[13–15] There may be several reasons for the deviations from the classical behavior, notably a frustrating next-nearest-neighbor (NNN) interaction,[11,12] the two dimensionality of the magnetism in the system,[16] and the degen-eracy of the energy scales for phonon and spin excitations in $CuGeO_3$ which calls for a description of $CuGeO_3$ in terms of mixed spin-phonon excitations rather than more or less decoupled spin and phonon excitations. At present, however, it is still unclear which of the above effects plays the dominant role in $CuGeO_3$, although it has been shown that one can certainly not neglect the frustration in the system induced by the next-nearest-neighbor interactions. The values reported for the frustration in $CuGeO_3$, based on one-dimensional (1D) adiabatic approximations, are in the range from $\alpha = J_{nnn}/J_{nn} = 0.24$–$0.36$,[11,12] and may even be higher in $CuGeO_3$ under hydrostatic pressure.[15,17] Within the 1D adiabatic approximations one then expects $CuGeO_3$ to have a spin gap, even without dimerization, which is small for $\alpha_c < \alpha \lesssim 0.5$, but increases strongly upon approaching the Majumdar-Gosh (MG) point ($\alpha = 0.5$).[18]

The influence of nonmagnetic impurities on one-dimensional spin-Peierls systems has been studied theoretically by several authors.[19–22] One of the main conclusions drawn from these studies is that one cannot consider such an impurity as a local site defect. In fact, it is expected that a nonmagnetic impurity induces local antiferromagnetic correlations over a length scale which is determined by the spin-spin correlation length ($\xi \sim 8$–10 sites[19]). This enhancement of the local antiferromagnetic correlations may explain the extreme sensitivity of the SP phase to substitutions and possibly also[23] the low-temperature antiferromagnetic (AF) long-range order in $Cu_{(1-x)}Zn_xGeO_3$ and $CuGe_{(1-y)}Si_yO_3$. Experimentally, evidence for these enhanced correlations may be found in Raman scattering data on substituted compounds.[10] The Raman phonon modes activated in the SP phase of pure $CuGeO_3$ appear in substituted compounds already at much higher temperatures, and are even observed in compounds having a substitution level for which the SP

phase has completely vanished.

Pressure-dependent experiments on CuGeO$_3$ (Refs. 17 and 24) have shown that T_{SP} strongly increases upon increasing pressure ($\partial T_{SP}/\partial p = 4.5$ K/GPa), in good agreement with earlier predictions derived from zero-pressure specific heat and thermal expansion experiments[13] using the Ehrenfest relation ($\partial T_{SP}/\partial p = 5$ K/GPa). It has been argued[15,17] that the pressure dependence of T_{SP} may be due to a strong positive pressure coefficient of the NNN frustration in CuGeO$_3$ ($\partial \alpha/\partial p > 0$). Recently, thermodynamic experiments on substituted compounds by Lorenz *et al.*[25] have led to the surprising prediction that $\partial T_{SP}/\partial p$ of substituted compounds is enhanced by a factor of 2–3 over that of the pure compound. Motivated by these predictions, we report here on the pressure dependence of T_{SP} for a Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ single crystal as determined from pressure-dependent susceptibility and Raman scattering experiments. At low pressure, where the above-mentioned prediction should be valid, the susceptibility experiments indeed show a strongly enhanced $\partial T_{SP}/\partial p$, quantitatively in agreement with the predicted $\partial T_{SP}/\partial p \approx 8.1$ K/GPa.[25] In contrast to the pure compound, however, Raman scattering experiments show that the pressure dependence of T_{SP} strongly deviates from a linear behavior, and approaches the pressure dependence of the pure compound for higher pressures. The remainder of this paper is organized as follows: Sections II and III describe the experimental results of the susceptibility and Raman scattering experiments, respectively. The subsequent section discusses these results in terms of a frustration- (and thus pressure-) dependent spin-spin correlation length. Finally, the last section presents the main conclusions of this paper.

II. SUSCEPTIBILITY

The Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ single crystal was grown from the melt by a floating zone method using an image furnace.[26] For our measurements of the susceptibility under pressure we use a piece ($5 \times 3 \times 2$ mm^3) of the crystal studied in Ref. 25. On the one hand, the specific heat and thermal expansion data presented there prove the high quality of this crystal. On the other hand, using the same sample enables an unambiguous comparison between the measured pressure derivatives of T_{SP} and the predictions from thermodynamic properties.[25]

The ac susceptibility was measured in a pressure range $0 \leq p \leq 0.43$ GPa with a mutual inductance bridge at a frequency of 6.1 kHz in a magnetic field of about 1 Oe. Both the primary coil and the astatic secondary coils are located inside the Teflon cup of a piston cylinder pressure cell which is similar to that described by Thompson.[27] A methanol:ethanol (4:1) mixture was used as pressure transmission medium in order to guarantee hydrostatic pressure conditions. The pressure was controlled at low temperatures by simultaneously measuring the superconducting transition temperature of a small piece of lead.

In Fig. 1 the raw data of the ac susceptibility measurements are presented for several pressures. The overall temperature dependence of the ac signal is mainly determined by a smooth background, which does not change strongly as a function of pressure. This background signal is nearly temperature independent above about 12 K and increases at
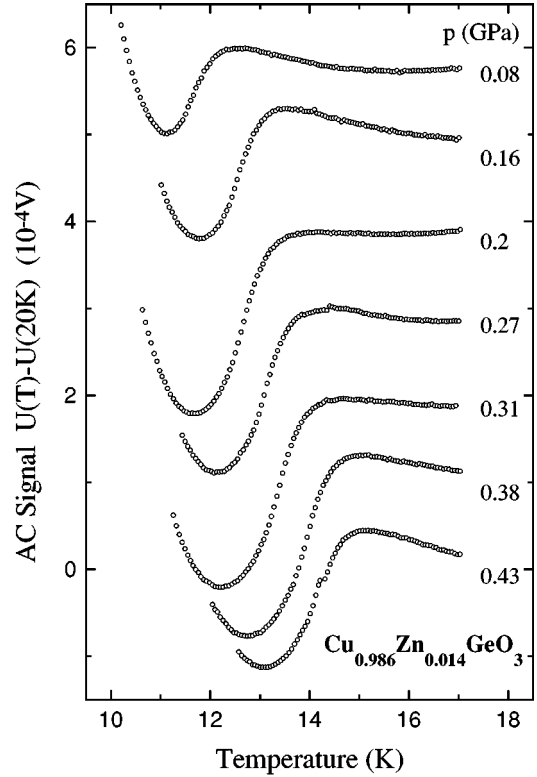

FIG. 1. Raw data of the ac susceptibility measurements (output signals of the lock-in amplifier) for several pressures. The data are subsequently shifted by 10^{-4} V for clarity.

lower temperatures.[28] Due to this background and its pressure dependence, a precise quantitative extraction of the susceptibility of Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ is not possible. In particular, one can not separate the Curie-like increase of χ present in doped CuGeO$_3$ at low temperatures[29] from the background signal. However, the pronounced anomalies at the spin-Peierls transition, which is associated with a rapid decrease of the susceptibility in a small temperature range, are clearly visible at all pressures. Besides the apparent increase of T_{SP} we do not find significant pressure dependences of the susceptibility anomaly at the corresponding T_{SP}.

As shown, e.g., in Ref. 30 the spin-Peierls transition temperature can be derived precisely from a sharp maximum of the temperature derivative of the magnetic susceptibility. The T_{SP} obtained in this way are shown as a function of pressure in Fig. 2. Together with the data we show a line, which represents a pressure derivative of $\partial T_{SP}/\partial p = 8.1(10)$ K/GPa as predicted from the Ehrenfest relation for $p \to 0$.[25] The measured, nearly linear, pressure dependence of T_{SP} up to 0.43 GPa is in excellent agreement with the predicted behavior. Fitting the data obtained from the susceptibility measurements by a linear increase of T_{SP} reveals an only slightly smaller slope of $\partial T_{SP}/\partial p = 7.3(3)$ K/GPa. We mention, however, that this slope systematically increases with decreasing pressure range considered for the linear fit. For example, fitting the data up to 0.27 GPa yields 8.2(5) K/GPa, whereas the slope $\partial T_{SP}/\partial p$ above 0.2 GPa only amounts to 6.3(7) K/GPa. Thus the susceptibility data are also consistent with a slight decrease of $\partial T_{SP}/\partial p$ with increasing pressure. The susceptibility experiments are particularly suited for the low-pressure region ($p < 1$ GPa), where
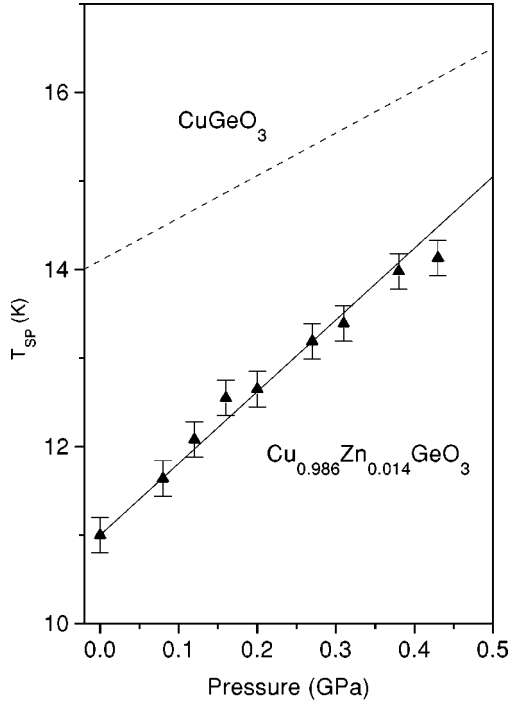
FIG. 2. Pressure dependence of T_{SP} as determined from the susceptibility data of $Cu_{0.986}Zn_{0.014}GeO_3$ (solid symbols). The lines give the predicted pressure dependences from the Ehrenfest relation [solid line, $Cu_{0.986}Zn_{0.014}GeO_3$, $\partial T_{SP}/\partial p = 8.1$ K/GPa (Ref. 25); dashed line, $CuGeO_3$, $\partial T_{SP}/\partial p = 5$ K/GPa (Ref. 13)].

FIG. 3. (ZZ) polarized Raman spectra of $Cu_{0.986}Zn_{0.014}GeO_3$ recorded at $T = 7$ K for $p = 0, 1.2, 1.6$, and 2.9 GPa. The curves have been given an offset for clarity. The positions of observable Ar^+-laser plasma lines are denoted by an asterisk.

also the prediction made in Ref. 25 should hold. These low-pressure data are shown in Fig. 2 (solid symbols) which also displays a linear fit to the data (solid line) and the prediction made in Ref. 25 (dashed line). The measured, nearly linear, pressure dependence of T_{SP} clearly agrees well with the predicted behavior in this pressure range.

Comparing the data on $Cu_{0.986}Zn_{0.014}GeO_3$ presented so far to the results for pure $CuGeO_3$ it is apparent that the pressure derivative in the doped compound is significantly larger. In other words, the reduction of T_{SP} due to doping with 1.4% Zn decreases from $\simeq 3$ K at $p = 0$ to $\simeq 2$ K at $p = 0.5$ GPa. Since an extrapolation of our data of doped $CuGeO_3$ would yield the unreasonable result that T_{SP} increases above the value of pure $CuGeO_3$ at high pressures ($p \gtrsim 2$ GPa), we have studied the behavior in this pressure range using Raman scattering.

III. RAMAN SCATTERING

Pressure- and temperature-dependent Raman scattering experiments have been performed on thin platelets ($100 \times 100 \times 40 \ \mu m^3$) of the same $Cu_{0.986}Zn_{0.014}GeO_3$ single crystal as used for the susceptibility measurements. These platelets were mounted in a clamp-type diamond anvil cell, using a methanol:ethanol mixture as the pressure medium. The cell has subsequently been mounted in a He flow cryostat with a temperature regulation better than 1 K. Polarized Raman spectra have been recorded in a quasibackscattering geometry using the 514-nm line of an Ar^+ laser for excitation and a charge-coupled-device- (CCD-) equipped DILOR-XY spectrometer for detection. The polarizations of incident
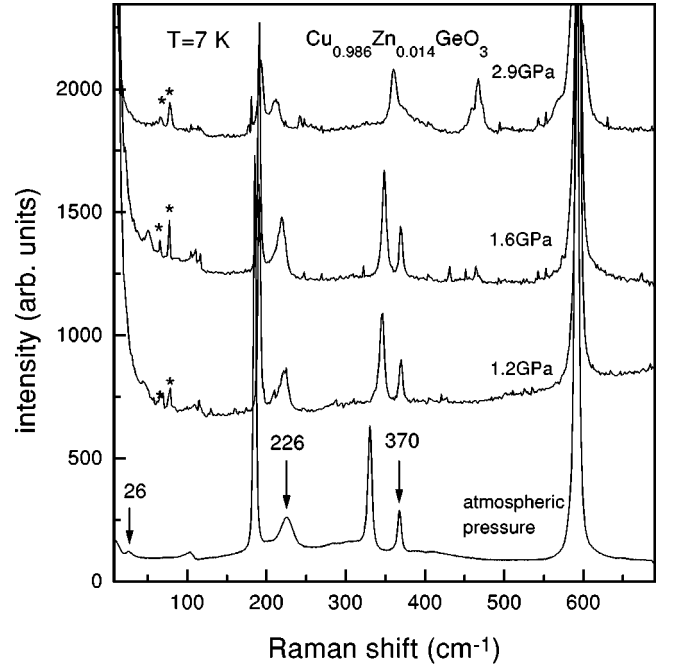
and scattered light have both been chosen parallel to the c axis of the orthorhombic crystal structure (i.e., along the Cu-O chains).

Typical spectra observed for different pressures at $T = 7$ K are presented in Fig. 3. For atmospheric pressure one observes three strong peaks at 184, 330, and 592 cm^{-1} which have been assigned to A_g modes of the orthorhombic crystal structure.[31] The four additional modes observed at 26, 105, 226, and 370 cm^{-1} are characteristic of the SP phase.[32,33] The two modes at 105 and 370 cm^{-1} can be assigned to Brillouin zone boundary phonons, and are activated in the SP phase through the magnetic and/or structural cell doubling. The low-frequency mode at 26 cm^{-1} arises due to transitions from the singlet ground state to the lowest magnetic singlet state.[34] The energy of this mode is somewhat smaller than that of the corresponding mode in the pure compound (30 cm^{-1}), indicating that Zn-substituted crystals have a smaller spin gap. This is in good agreement with the observed reduction of the transition temperature in $Cu_{0.986}Zn_{0.014}GeO_3$ (T_{SP} [$Cu_{0.986}Zn_{0.014}GeO_3$]$\approx 11$ K, T_{SP} [$CuGeO_3$]≈ 14 K). Finally, the broad, asymmetric response, peaking at 226 cm^{-1} is generally thought to arise from two-magnon scattering due to the Fleury-Loudon[35] mechanism, and should reflect a weighted two-magnon density of states.

Similar to the situation in the pure compound,[24,17] the application of hydrostatic pressure leads to drastic changes in the observed Raman spectra. With increasing pressure the two-magnon peak shifts to lower frequencies (208 cm^{-1} at 2.9 GPa), reflecting an increasing magnetic frustration in the system. In contrast to this, but for the same reasons, the gap-related mode at 26 cm^{-1} rapidly shifts to higher frequencies upon applying pressure. Simultaneously this mode also broadens, and at 2.9 GPa the singlet response is hardly observed anymore. Most likely, this broadening is due to
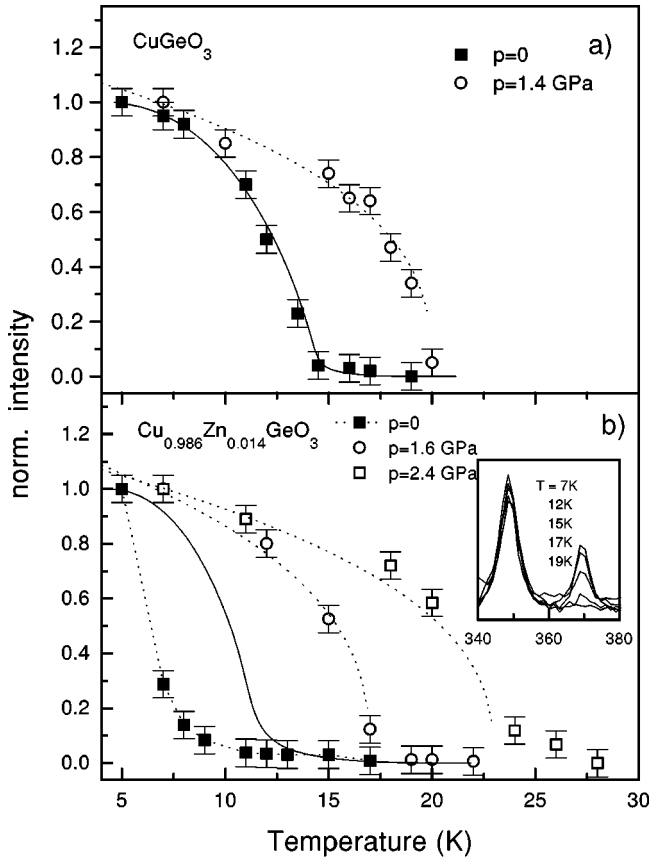
FIG. 4. (a) Intensity 370-cm^{-1} mode in CuGeO$_3$ as a function of temperature for $p=0$ (solid symbols), and 1.4 GPa (open symbols). The solid line gives the experimental temperature dependence of the spontaneous strain at $p=0$ GPa (Ref. 25). Dashed line is a guide to the eye. (b) Intensity of the 370-cm^{-1} mode in Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ as a function of temperature for $p=0$ (solid symbols), 1.4, and 2.4 GPa (open symbols). The solid line gives the experimental temperature dependence of the spontaneous strain at $p=0$ GPa (Ref. 25). Dashed lines are a guide to the eye. The inset shows the temperature dependence of the (ZZ) polarized spectrum of Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ for $p=1.6$ GPa.

nonhydrostatic effects at higher pressure, as is also reflected in the width of the phonon modes.

A good method to determine T_{SP} for the pure compound is to monitor the temperature dependence of the singlet mode.[17] As is the case for the triplet spin-Peierls gap,[36] the energy of the singlet mode strongly decreases upon increasing temperature until it vanishes at T_{SP}. Alternatively, it has been shown that also the intensity of the spin-Peierls active 370-cm^{-1} phonon may be used to determine T_{SP}.[24] The intensity of this mode shows a temperature behavior characteristic for a second-order phase transition, i.e., an intensity which varies as the square of the order parameter. This is demonstrated for CuGeO$_3$ in Fig. 4(a), which shows a comparison between the temperature dependence of the intensity of the 370-cm^{-1} phonon (solid symbols) with that of the experimentally determined spontaneous strain (solid line),[25] which should vary as the square of the structural order parameter. For the pure compound the above-mentioned methods yield essentially the same results and are in good agreement with the transition temperature obtained using other methods (e.g., thermal expansion, susceptibility, heat capac-

ity), as shown in Refs. 13, 25, and 37.

Since the present data do not allow one to use the singlet response as an indicator for the presence of the SP phase over the whole pressure range, we will use the intensity of the 370-cm^{-1} mode to determine T_{SP}. As an example, the inset of Fig. 4(b) shows the temperature dependence of this mode for $P=1.6$ GPa. Relative to the $A_g(2)$ mode at 347 cm^{-1}, this mode, which is observed as a relatively strong Lorentzian-shaped mode at $T=7$ K, rapidly loses intensity as the temperature increases, and finally disappears around $T_{SP}=18$ K. Figure 4(b) shows the temperature dependence of the intensity of the 370-cm^{-1} phonon as a function of temperature (normalized to the intensity at the lowest temperature) for $p=0,1.6$, and 2.4 GPa. For higher pressure ($p>1$ GPa) the intensity indeed shows the expected behavior; i.e., it qualitatively seems to reflect the squared order parameter of a second-order phase transition. In contrast to this and to the situation in the pure compound, the temperature dependence of the intensity of the 370-cm^{-1} mode at $p=0$ does not seem to reflect the square of the structural order parameter. This becomes more clear from a comparison of the temperature dependence of the intensity of the 370-cm^{-1} mode with the spontaneous strain measured on the same sample [Fig. 4(b), solid line]. Although the spontaneous strain still shows the behavior of a relatively well-defined second-order phase transition with $T_{SP}=11.5$ K, the intensity of the 370-cm^{-1} mode drops much more rapidly with temperature and, moreover, remains nonzero up to temperatures well above the phase transition.[38] These observations put into question the interpretation of a structurally induced intensity for the 370-cm^{-1} mode in the SP phase. Indeed, the intensity of this mode in the pure compound also shows a good correlation with the square of the singlet energy (the square of the "magnetic order parameter"). This is not surprising since the singlet energy directly correlates with the SP gap Δ, which itself is related to the structural order parameter δ ($\Delta=\delta^{2/3}$ within Cross-Fisher theory[39]). This indicates that the intensity of the 370-cm^{-1} mode may not be induced by the structural distortion in the SP phase, but rather by the doubling of the magnetic unit cell.[40] It furthermore strongly suggests that the relation between the structural distortion and the size of the SP gap may drastically change upon substitutions, strongly deviating from the Cross-Fisher prediction.[39]

IV. DISCUSSION

At low pressures the transition temperature in the substituted compound indeed increases much faster with pressure ($\partial T_{SP}/\partial p \approx 7.5$ K/GPa) as compared to the increase for the pure compound ($\partial T_{SP}/\partial p \approx 4.5$ K/GPa) (see Fig. 2). The steep linear behavior observed for low pressures clearly cannot persist indefinitely. Indeed, the transition temperatures obtained for higher pressure from the Raman data show a pronounced nonlinear behavior at higher pressures (see Fig. 5, solid squares). In this regime, the pressure dependence of the transition temperature for Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ approaches that of the pure CuGeO$_3$ compound.

In order to get some understanding of the strong pressure dependence of T_{SP} in Cu$_{(1-x)}$Zn$_x$GeO$_3$, consider first the strong dependence of T_{SP} on substitutions. As has been
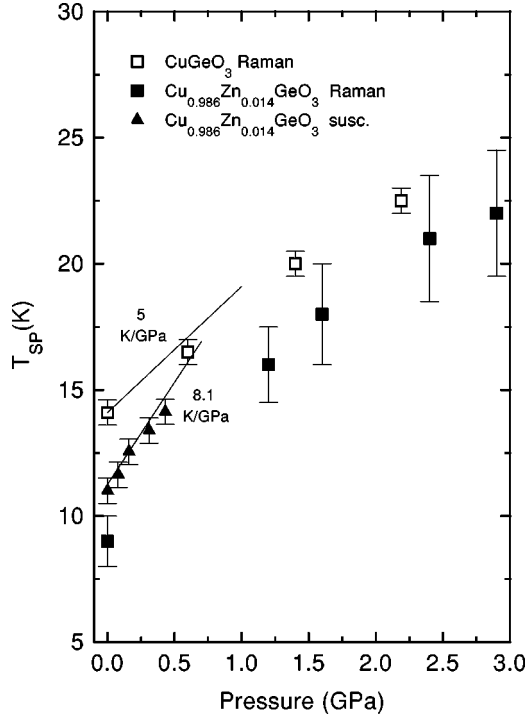
FIG. 5. Dependence of T_{SP} as a function of pressure for CuGeO$_3$ (open symbols) and Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ (solid symbols: triangles, obtained from susceptibility data; solid squares, obtained from Raman data). The solid lines give the predictions for the zero-pressure derivatives.

shown by, e.g., Fukuyama et al.,[19] a substitution at a single site does not only perturb this site, but leads to the formation of local AF ordering and suppression of the dimerization over several lattice sites. The spatial extent of the impurity is typically of the order of the spin-spin correlation length $\xi \sim (J/\Delta)a$. This immediately explains the strong reduction of T_{SP} upon substitution, and may possibly also explain the occurrence of a long-range-ordered AF ground state in the substituted crystals.[19,21,22] As has been pointed out by Mostovoy and Khomskii,[22] one expects the SP transition to be fully suppressed when the average distance between the impurities becomes of the order of the spin-spin correlation length, i.e., for concentrations of about 10% ($\xi \sim 8a-10a$).

The strong increase of T_{SP} with increasing pressure in pure CuGeO$_3$ has been discussed in terms of a strongly increasing frustration.[15,17] This may, of course, also explain part of the observed increase of T_{SP} in the substituted compound. There is, however, a second effect which arises from the increasing frustration and is of importance in the substituted compounds. As the frustration increases, one expects the spin-spin correlation length to decrease. Indeed, at $\alpha = 0.5$ (the MG point) one expects the ground state to be formed by singlets pairs and to have a correlation length of exactly two spins. Numerical density matrix renormalization group calculations of the correlation length have indeed confirmed this picture.[41] The consequence of the decreasing correlation length is that the influence of impurities is strongly

suppressed upon applying pressure. This actually may explain the observed behavior of $T_{SP}(p)$ in Cu$_{0.986}$Zn$_{0.014}$GeO$_3$. Initially, the increase of the transition temperature is driven by a combination of an increasing frustration and a decreasing influence of the impurities with frustration. At higher pressure the effect of the impurities is strongly suppressed, and the increase of T_{SP} is fully driven by the increasing frustration alone. At these pressures one therefore expects only small differences between the pure and the substituted compounds, in good agreement with the observed behavior.

A further indication of the suppression of the effects of the impurities may be found from the temperature dependence of the intensity of the 370-cm^{-1} mode [see Fig. 4(b)]. As mentioned in the previous section, at low pressures the behavior of the intensity of this mode does not agree well with the temperature dependence of the structural order parameter. In addition, the mode remains observable at temperatures well above the phase transition. This latter observation is consistent with the idea that the mode is induced by magnetoelastic interactions and the magnetic cell doubling. If indeed the presence of impurities leads to enhanced AF correlations around the impurities, then this also leads to a local magnetic unit cell doubling and, hence, to an activation of the 370-cm^{-1} mode in the Raman spectra. Of course, at high enough temperatures the intensity should still disappear due to the decreasing correlation length, i.e., the decreasing spatial ''size'' of the impurities. At high enough pressure ($p > 1$ GPa) the temperature dependence of the intensity of the 370-cm^{-1} mode is again in qualitative agreement with the dependence observed for the pure compound, in good agreement with the idea that the influence of the impurities is strongly reduced for high pressures.

V. CONCLUSIONS

In this paper we have shown, by two different methods, that the pressure dependence of the spin-Peierls transition temperature in Zn-substituted CuGeO$_3$ is strongly enhanced over that of the pure compound. The initial-slope data are found to be in excellent agreement with the predictions using the Ehrenfest relation.[25] At higher pressure the pressure coefficient $\partial T_{SP}/\partial p$ of Cu$_{0.986}$Zn$_{0.014}$GeO$_3$ approaches that of the pure compound. The strong enhancement at low pressure and the approach to the behavior of the pure compound at high pressure are in good agreement with an increasing frustration in these compounds, which leads to a decreasing correlation length and thus to a decrease of the influence of the impurities.

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[1] E. Pytte, Phys. Rev. B **10**, 4637 (1974).

[2] R.E. Peierls, *Quantum Theory of Solids* (Clarendon, Oxford, 1995).

[3] J.W. Bray, L.V. Iterrante, I.S. Jacobs, and J.C. Bonner, in *Extended Linear Chain Compounds*, edited by J. S. Miller (Plenum Press, New York, 1983), Vol. 3.

[4] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. **70**, 3651 (1993).

[5] M. Isobe and Y. Ueda, J. Phys. Soc. Jpn. **65**, 1178 (1996).

[6] J.P. Boucher and L.P. Regnault, J. Phys. I **6**, 1939 (1996).

[7] J.P. Renard, K. Le Dang, P. Veillet, G. Dhalenne, A. Revcolevschi, and L.P. Regnault, Europhys. Lett. **30**, 475 (1995).

[8] L.P. Regnault, J.P. Renard, G. Dhalenne, and A. Revcolevschi, Europhys. Lett. **32**, 579 (1995).

[9] M. Hase, N. Koide, K. Manabe, Y. Sasago, K. Uchinokura, and A. Sawa, Physica B **215**, 164 (1995).

[10] P. Lemmens, M. Fischer, G. Güntherodt, C. Gros, P.G.J. van Dongen, M. Weiden, W. Richter, C. Geibel, and F. Steglich, Phys. Rev. B **55**, 15 076 (1997).

[11] G. Castilla, S. Chakravarty, and V.J. Emery, Phys. Rev. Lett. **75**, 1823 (1995).

[12] J. Riera and A. Dobry, Phys. Rev. B **51**, 16 098 (1995).

[13] H. Winkelmann, E. Gamper, B. Büchner, M. Braden, A. Revcolevschi, and G. Dhalenne, Phys. Rev. B **51**, 12 884 (1995).

[14] H. Takahashi, N. Mori, O. Fujita, J. Akimitsu, and T. Matsumoto, Solid State Commun. **95**, 817 (1995).

[15] B. Büchner, U. Ammerahl, T. Lorenz, W. Brenig, G. Dhalenne, and A. Revcolevschi, Phys. Rev. Lett. **77**, 1624 (1996).

[16] G. Uhrig, Phys. Rev. Lett. **79**, 163 (1997).

[17] P.H.M. van Loosdrecht, J. Zeman, G. Martinez, G. Dhalenne, and A. Revcolevschi, Phys. Rev. Lett. **78**, 487 (1997).

[18] C.K. Majumdar and D.K. Ghosh, J. Math. Phys. **10**, 1388 (1969).

[19] H. Fukuyama, T. Tanimoto, and M. Saito, J. Phys. Soc. Jpn. **65**, 1182 (1996).

[20] G. Martins, M. Laukamp, J. Riera, and E. Dagotto, Phys. Rev. Lett. **78**, 3563 (1997).

[21] M. Fabrizio and R. Mélin, Phys. Rev. Lett. **78**, 3382 (1997).

[22] M. Mostovoy and D. Khomskii, Z. Phys. B **103**, 209 (1997).

[23] M. Mostovoy (private communication).

[24] A.R. Goñi, T. Zhou, U. Schwarz, R.K. Kremer, and K. Syassen, Phys. Rev. Lett. **77**, 1079 (1996).

[25] T. Lorenz, H. Kierspel, S. Kleefisch, B. Büchner, E. Gamper, A. Revcolevschi, and G. Dhalenne, Phys. Rev. B **56**, 501 (1997).

[26] A. Revcolevschi and R. Collongues, C.R. Acad. Sci. **266**, 1767 (1969); A. Revcolevschi and G. Dhalenne, Adv. Mater. **5**, 9657 (1993).

[27] J.D. Thompson, Rev. Sci. Instrum. **55**, 231 (1984).

[28] The low-temperature increase of the signal is strongly frequency dependent and does not represent a magnetic susceptibility.

[29] M. Hase, I. Terasaki, Y. Sasago, K. Uchinokura, and H. Obara, Phys. Rev. Lett. **71**, 4059 (1993).

[30] J.P. Pouget, L.P. Regnault, M. Ain, B. Hennion, J.P. Renard, P. Veillet, G. Dhalenne, and A. Revcolevschi, Phys. Rev. Lett. **72**, 4037 (1994).

[31] S.D. Dević, M.J. Konstantinović, Z.V. Popović, G. Dhalenne, and A. Revcolevschi, J. Phys.: Condens. Matter **6**, L745 (1994).

[32] H. Kuroe, T. Sekine, M. Hase, Y. Sasago, K. Uchinokura, H. Kojima, I. Tanaka, and Y. Shibuya, Phys. Rev. B **50**, 16 468 (1994).

[33] P.H.M. van Loosdrecht, J.P. Boucher, G. Martinez, G. Dhalenne, and A. Revcolevschi, Phys. Rev. Lett. **76**, 311 (1996).

[34] G. Bouzerar, A.P. Kampf, and F. Schönfeld, cond-mat/9701176 (unpublished).

[35] P. A. Fleury, R. Loudon, Phys. Rev. **166**, 514 (1968).

[36] M. Nishi, O. Fujita, and J. Akimitsu, Phys. Rev. B **50**, 6508 (1994).

[37] T. Lorenz, U. Ammerahl, R. Ziemes, B. Büchner, A. Revcolevschi, and G. Dhalenne, Phys. Rev. B **54**, 15 610 (1996).

[38] Similar observations for Zn- and Si-substituted $CuGeO_3$ have been reported in Ref. 10.

[39] M.C. Cross and D.S. Fisher, Phys. Rev. B **19**, 402 (1979).

[40] P.H.M. van Loosdrecht, J.P. Boucher, S. Huant, G. Martinez, G. Dhalenne, and A. Revcolevschi, Physica B **230-232**, 1017 (1997).

[41] G. Bouzerar (private communication).