Ultrasonic investigation of the organic spin-liquid compound EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

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Ultrasonic measurements performed on the spin-liquid organic compound EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ reveal an inhomogeneous elastic behavior above 40 K due to the rotational motion in the EtMe$_3$Sb cations. Two velocity softening anomalies and concomitant attenuation peaks at 40 K and 160 K are respectively attributed to the rotation of methyl and ethyl groups. At lower temperatures, the elastic character is homogeneous and an anomaly is observed below 2 K. A magnetic field study suggests that two kinds of excitations contribute to this anomaly. First, a softening of the velocity toward the lowest temperatures which is not dependent on magnetic field implies other degrees of freedom than spins. Second, spin excitations coupled to longitudinal acoustic phonons yield a velocity softening anomaly that completely disappears for field values higher than 7 T. Consistent attenuation anomalies are also observed for both contributions. These data suggest that only one kind of spin excitation exists in the spin-liquid ground state below 1.5 K. A spinon-phonon interaction likely explains the magnetic field dependent elastic anomalies. However, no spin-gap-like excitations related to a pairing instability of the spinons could be detected in this experiment.

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I. INTRODUCTION
A quantum spin-liquid (QSL) ground state that was early proposed to be realized in spin-1/2 frustrated systems [1] has been reported only recently in a few frustrated magnets, especially the quasi-two-dimensional (2D) organic compounds $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [2] and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [3,4]. In both compounds, nuclear magnetic resonance (NMR) measurements have shown no long range order down to a temperature corresponding to $10^{-4}$ the nearest-neighbor exchange constant $J$. Frustration brought on by this exchange interaction is known to be insufficient to destroy long-range magnetic order [5] but quantum fluctuations can even at the zero temperature. The nature of the QSL states has attracted theoretical and experimental attention for decades, but a detailed description of the ground state still remains elusive. These QSL states may possess exotic elementary excitations which obey either fermionic or bosonic statistics and have gapped or gapless energy dispersion [6,7].

To understand the nature of the QSL ground states, the detailed structure of the fluctuations and of the low-lying elementary excitations must be investigated in the zero temperature limit. For example, the presence or absence of an excitation gap is of primary importance for characterizing the spin correlations and the correlation length scale [8]. In the $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ compound the presence of a spin excitation gap is controversial [8,9] and a second order phase transition near $\sim 6$ K complicates further the situation [10]. Although a broad hump structure is observed in heat capacity measurements around 4 K [11], a similar phase transition is likely absent in EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ and a more homogeneous QSL state is attained at low temperatures [3,12]. A sizable linear term is found in the temperature dependence of the thermal conductivity in the zero limit [13], indicating gapless excitations with an extremely long mean free path in analogy with excitations near the Fermi surface in pure metals; its magnetic field dependence suggests a concomitant appearance of spin-gap-like excitations at low temperatures. It was then concluded that two kinds of excitations are found for EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ in the low temperature regime. Torque magnetometry measurements down to low temperatures (30 mK) and up to high fields (32 T) [14] revealed a distinct residual paramagnetic susceptibility comparable to that in a half-filled two-dimensional metal, demonstrating the magnetically gapless nature of the ground state.

The existence of low lying excitations in organic spin-liquid materials $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, as revealed in thermal conductivity measurements appears to support the picture of mobile particles called spinons which form a Fermi surface and which are coupled to $U(1)$ gauge fields [15,16]. It was then suggested that the 6 K transition in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ could result from an instability of the Fermi surface which leaves a finite density of states intrinsically or due to impurities [17]. To test this picture of spinons in the organic salts, a few experiments have been suggested, metalliclike spin transport [18] and spinon-phonon coupling [19], for which specific temperature dependencies of the ultrasonic attenuation were predicted. Indeed, a recent ultrasonic investigation [20] suggested that a spinon-phonon coupling was responsible for a velocity softening below 20 K and that the phase transition near 6 K was due to a pairing instability of the spinons.

In this paper, we report an ultrasonic experiment on the other organic spin-liquid candidate, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, that confirms a slightly different character of the QSL ground state in comparison with $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. The high temperature profile of the longitudinal sound velocity along a direction perpendicular to the organic layers presents a frequency dependent inhomogeneous character originating
from the rotation of the cations EtMe3Sb. At much lower temperatures the inhomogeneous character has practically disappeared and a well defined softening anomaly independent of frequency is observed below 1.5 K down to the lowest attained temperature of 40 mK. A magnetic field study reveals that only one kind of magnetic excitations could likely explain the temperature profile of this elastic anomaly. The results suggest the appearance of a spinon Fermi surface below 1.5 K without any pairing instability.

II. EXPERIMENT

High-quality single crystals of EtMe3Sb[Pd(dmit)2]2 were grown as thin platelets using an aerial oxidation method. Two crystals with typical maximum sizes 3 × 2.5 × 0.1 mm3 were used for the pulsed echo ultrasonic experiment which requires samples with parallel shiny surfaces on which piezoelectric transducers can be glued. The ultrasonic technique is used in the transmission mode: the transducers are installed on the parallel surfaces of the platelet, the ab plane, so the ultrasonic waves propagate along the ε∗ axis. Given the reduced thickness of the crystal along the ε∗ axis (≈ 100 μm), the sample is glued with silicone seal onto a CaF2 delay line in order to separate the first transmitted acoustic echo (through the crystal-CaF2 assembly) from the electric pulse. Only the longitudinal acoustic mode can be reliably studied with this setup because it has a high velocity that allows its time separation from other parasitic echoes. Because of the monoclinic crystal structure, mode conversion occurs at the different interfaces and the transverse modes are difficult to isolate from other echoes. The longitudinal acoustic pulses are generated with LiNbO3 piezoelectric transducers resonating at 30 MHz and odd overtones bonded to the crystal and the CaF2 delay line with silicone seal. We measure the variation of longitudinal acoustic velocity relative to its value at a fixed temperature \( T_0 \), \( \Delta V/V = (V(T) - V(T_0))/V(T_0) \), and the corresponding variation of the attenuation \( \Delta \alpha \). Since the crystal structure is monoclinic, the acoustic mode is not pure but it is dominated by the compression modulus \( C_{33} \); after subtraction of the contribution of the CaF2 crystal, the velocity variations are related to the variations of the compressibility along a direction perpendicular to the two-dimensional \( \text{Pd(dmit)}_2 \) planes, \( \Delta C_{33}/C_{33} = 2\Delta V/V \) since \( C_{33} = \rho V^2 \), where \( \rho \) is the mass density. The velocity variations were not corrected to take account of the thermal expansion since these effects are usually negligible. We could also measure the variation of the attenuation \( \Delta \alpha \) by monitoring the amplitude of the first transmitted echo. A magnetic field up to 16 T could be applied along the ε∗ axis. The experiment is performed in a variable temperature insert (VTI) fridge for temperatures between 2 and 300 K, and in a helium dilution fridge for lower temperatures.

III. RESULTS AND DISCUSSION

The silicone seal used to bond the piezoelectric transducers and the crystal shows a glass transition near 200 K that affects importantly the acoustic signal and this is why the data are only analyzed below this temperature. Ultrasonic data obtained at two frequencies of 106 and 177 MHz are shown in Fig. 1 for two thermal cycles indicated by arrows. When decreasing the temperature from room value, the velocity generally increases smoothly in agreement with the stiffening of the interplane cohesion forces and saturates at low temperatures. This is the behavior observed below 190 K for the other spin-liquid candidate κ-(BEDT-TTF)2Cu2(CN)3 as shown in Fig. 1(a) for comparison (dashed line). The temperature profile of the velocity for EtMe3Sb[Pd(dmit)2]2 shows important deviations from this behavior. For the decreasing temperature cycle, two frequency dependent elastic anomalies are observed on \( \Delta V/V \) [Fig. 1(a)]; the first one appears as a decrease of the velocity below \( \sim 160 \) K and the second one looks rather like a very large softening dip centered around 40 K, the amplitude of the dip increasing rapidly with frequency. For the increasing temperature cycle, the same anomalies are observed but the data clearly separate from the decreasing cycle ones above 40 K, revealing then an inhomogeneous elastic behavior up to 190 K. The attenuation data shown in Fig. 1(b) confirm this inhomogenous character, the thermal cycles being markedly different only above 40 K. Two attenuation peaks are observed near 160 and 40 K consistently with the velocity data. Below 40 K, the elastic behavior is practically homogeneous: the increase of \( \Delta V/V \) is not found frequency dependent and there is an apparent absence of thermal cycling effects.

The elastic anomalies likely originate from rotations in the EtMe3Sb cations as recently observed in the nuclear spin-lattice relaxation rates \( (1/T_1) \) of the proton sites [21].
The NMR results revealed that a broad maximum observed at \( T_{\text{Me}} \approx 40 \text{ K} \) is ascribed to the rotation of the methyls (Me), and a maximum at \( T_{\text{Et}} \approx 180 \text{ K} \) is originating from the rotation of ethyl (Et) of the EtMe3Sb cation. These rotations of the methyls appears as a universal phenomenon in \([\text{Pd(dmit)}]_2\) salts irrespective of their magnetic ground states. Since the longitudinal elastic waves are propagating perpendicularly to the cation layers, it is thus not surprising to reveal over this temperature range an inhomogeneous character and softening anomalies due to these rotations. Unfortunately, this could hinder the identification of any physical phenomenon related to the spin-liquid ground state within the Pd(dmit)\(_2\) layers, especially at low temperatures where quantum rotational motion of the methyl and ethyl groups can survive even near absolute zero \([13,22]\). However, the data presented in Fig. 1 suggest that, well below 40 K, the crystal may present a nearly homogeneous elastic character that can favor the investigation of the spin-liquid ground state.

The homogeneity of the elastic character below \( T_{\text{Me}} \) is further examined in Fig. 2, where velocity and attenuation data at 106 MHz are presented for different thermal cycles. The first increase to 60 K (curves 1) was performed at a rate of 1.2 K/min after keeping the sample at 2 K for 24 h. Then, after decreasing the temperature to 2 K at the same rate, the second thermal increase (curves 2) revealed an upward shift of the data only for \( T < 50 \text{ K} \). Following the same procedure, the third thermal cycle (curves 3) produces a similar shift but with a smaller amplitude and the well defined anomaly on \( \Delta \alpha \) at \( T_{\text{Me}} \) has been washed out. No further significant changes in the data were obtained for repeated thermal cycles. These results reveal several characteristics of the low temperature elastic behavior. The rotational motion related to the EtMe3Sb cations persists down to 2 K and long relaxation times are associated to it. These relaxation effects produce a smaller attenuation as expected, but it decreases the velocity which is rather counterintuitive. It is clear that repeated thermal cycles over \( T_{\text{Me}} \) induce disorder at low temperatures but, as long as the temperature is kept below \( T_{\text{Me}} \) for a long period (curves 1), the disorder appears stabilized and temperature cycling at the lowest temperatures are highly reproducible. This last point is very important since we will now investigate the temperature range below 2 K in search of features related to the spin-liquid ground state.

We know that spin degrees of freedom couple easily with the lattice in various systems to yield softening anomalies on compression elastic moduli \([23,24]\). We may thus wonder if such a spin-lattice coupling could be responsible for the anomaly centered at \( T_{\text{Me}} \), since the spin susceptibility of EtMe3Sb[Pt(dmit)]\(_2\) displays a maximum around the same temperature of 40 K. This is unlikely because the resulting softening anomaly is expected to be frequency independent in clear contradiction with our observations. Moreover, such an anomaly is completely absent for the other organic spin-liquid candidate \( \kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \) [Fig. 1(a)] that presents a very similar spin susceptibility \([2]\). Nevertheless, spin degrees of freedom are present over the low temperature range as revealed by a magnetic field study presented later in this paper.

All the experimental features reported in relation to the QSL ground state in EtMe3Sb[Pt(dmit)]\(_2\) were observed below 2 K. We have thus performed our ultrasonic experiment in a dilution refrigerator. In order to get rid of any residual disorder due to rotational motion previously discussed, we kept the crystal at the lowest attained temperature of 40 mK for a few days before initiating the experiment. If cation rotational motion is still active over this temperature range, it does not produce supplementary entropy so that repeated thermal cycles from 40 mK to 2 K were always highly reproducible. However, there is a possibility that quantum rotational motion may affect the measurement at the lowest temperatures.

The velocity data \( \Delta V/V \) at 177 MHz are presented in Fig. 3(a) for \( T \lesssim 1.5 \text{ K} \). The velocity increase, already observed going to low temperatures (Fig. 2), is maintained down to \( \sim 1 \text{ K} \) where a maximum is reached; then \( \Delta V/V \) decreases rapidly for lower temperatures and nearly saturates around 100 mK. In our ultrasonic experiment applied to thin organic compounds, the crystal is glued on a CaF\(_2\) delay line which is a poor thermal conductor. Thermalization of the crystal is mainly assured by the gold wires installed on the emitting piezoelectric transducer. Thus it is important to verify that the sample’s temperature is not affected by the level of acoustic power. We show in Fig. 3(a) three \( \Delta V/V \) curves obtained at different power levels that were sufficiently low so that our RuO\(_2\) temperature sensor is not heated up over its base temperature of 38 mK. We observed that the temperature dependence of \( \Delta V/V \) is not affected by the selected range of levels above approximately 300 mK. For the lowpower data, the decrease of the velocity toward low temperatures is quasilinear without any saturation that is induced by increasing the level by 3 and 6 dB. Decreasing the power level even more greatly deteriorates the signal to noise ratio but it does not modify further the temperature profile in the low temperature range. This is why all the subsequent ultrasonic data were performed with lowpower level. We have also investigated the frequency dependence of this softening anomaly. We compare in Fig. 3(b) the temperature dependence of \( \Delta V/V \) obtained at 177 MHz and 248 MHz. In spite of a deterioration of the signal to noise ratio due to the reduced acoustic power generated by higher harmonics of the piezoelectric transducers, we may consider that no frequency effects are
We thus consider the low power curve of Fig. 3(a) as our reference elastic behavior in the QSL state.

It is known that generic magnetic fluctuations couple with acoustic longitudinal modes to yield a softening of the elastic moduli \[23,24\]. The observation of an elastic softening anomaly on the velocity at these low temperatures could be related to this kind of coupling in the QSL ground state of EtMe₃Sb[Pd(dmit)₂]₂. To verify this, we have investigated the effects of a magnetic field applied perpendicular to the Pd(dmit)₂ layers. We compare in Fig. 4 the temperature dependence of the ultrasonic data below 2 K in zero magnetic field with the ones obtained at the maximum field value of 16 T. For the velocity \[\Delta V/V\] data [Fig. 4(a)], we can distinguish two temperature ranges with opposite field effects, negative (softening) for \(T > 1.5\) K and positive (stiffening) for \(T < 1.5\) K; moreover, the velocity at the lowest temperature of 40 mK is apparently not modified by the magnetic field. Because of the very small propagation length and the reduced acoustic power level, the attenuation is difficult to measure at low temperatures with a reasonable signal to noise ratio. However, at 177 MHz, the attenuation variation \[\Delta \alpha\] could be measured with very good reproducibility. These data are shown in Fig. 4(b) at the same magnetic field values. In zero field, \(\Delta \alpha\) decreases quasilinearly with temperature down to \(-0.8\) K where the decreasing rate is enhanced; at the lowest temperatures, it saturates with a tendency to weakly increase below 300 mK. In a 16 T field, no effects are observed until the temperature has decreased below \(-1.5\) K; then the attenuation decreases more rapidly relative to the zero field curve and shows a clear minimum near 300 mK.

In Fig. 5, we analyze more thoroughly these field effects on the velocity data by performing field scans at fixed temperatures. Below 1.5 K, \(\Delta V/V\) increases rapidly with field up to approximately 7 T where saturation is observed. For higher temperatures (3.1 K), the velocity weakly decreases after going through a small maximum near 3 T; this last field behavior has been observed with practically the same amplitude up to 25 K, indicating that a magnetoelastic coupling is present over a wide temperature range. The inset of Fig. 5 shows a scan at 100 mK up to 2 T that confirms the absence of field effects at the lowest temperatures [Fig. 4(a)].

The magnetic field investigation strongly suggests that two types of excitations contribute to the elastic anomaly observed in the QSL ground state in a temperature range below 1.5 K. Since the field effects saturate around \(-7\) T, one type of excitation that is insensitive to field gives rise to the softening anomaly represented by the 16 T \(\Delta V/V\) data of Fig. 4(a) and the concomitant attenuation \(\Delta \alpha\) of Fig. 4(b). The other types of excitations are magnetic in nature since they are strongly sensitive to field: their effects on the velocity and attenuation (not shown because of poor signal/noise ratio) disappear completely for \(H > 7\) T. We can thus express the elastic anomalies on the velocity and attenuation as a superposition of two contributions below 1.5 K,

\[
\Delta V/V = (\Delta V/V)_0 + (\Delta V/V)_H, \\
\Delta \alpha = (\Delta \alpha)_0 + (\Delta \alpha)_H.
\]
where \((\Delta V/V)_0, (\Delta \alpha)_0\) and \((\Delta V/V)_H, (\Delta \alpha)_H\) are respectively the field insensitive and sensitive contributions. The field sensitive contributions are evaluated by subtracting the 16 T data from the zero field ones (Fig. 4) after performing a smoothing process. These contributions are shown in Fig. 6 as a function of temperature. We observe that magnetic excitations below \(\sim 1.5\) K induce in zero magnetic field a softening of the velocity that presents a maximum in the range as the NMR relaxation experiment \([25]\). The anomaly appears as a superposition of two contributions, an observation that could be in apparent agreement with thermal conductivity measurements \([13]\). However, their relation with the QSL ground state remains to be clearly established.

The first contribution to the velocity anomaly \((\Delta V/V)_0\) represented by the 16 T data of Fig. 4(a) is not dependent on magnetic field; it produces a continuous softening below \(\sim 1\) K down to the lowest temperatures. The existence of low energy magnetic excitations called spinons \([15,16]\) may justify the appearance of an anomaly due to a spinon-phonon interaction as suggested for the QSL candidate \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\) \([20]\). However, a continuous softening of the velocity down to 40 mK is not realistic since the spinon-phonon interaction should disappear as \(T \to 0\) K because of a vanishingly small number of acoustic phonons. Moreover, it is difficult to understand why such an anomaly should not be affected by a 16 T magnetic field whose energy is rather large compared to the energy scale of the excitations. Thus we must conclude that this contribution to the elastic anomaly is not related to the QSL ground state and that it originates from degrees of freedom other than spins, for example, the quantum rotational motion of the methyl groups which is expected to survive even at the lowest temperatures. A more thorough frequency dependence of this anomaly, not essential to our study, is needed to identify these degrees of freedom and the coupling mechanism.

The second contribution \((\Delta V/V)_H\) (Fig. 6) has clearly a magnetic origin and is possibly related to the spinon-phonon interaction that progressively sets in for \(T < 1.5\) K when a spinon Fermi surface appears. This was the kind of scenario suggested for \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\) below 20 K \([20]\). At lower temperatures, the rapid and complete disappearance of the softening for \(T < 300\) mK can be possibly attributed to two processes. It can be due to a pairing instability of the spinons that decreases the number of excitations as reported below 6 K for \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\) \([20]\). More likely the loss of softening is due to a progressive disappearance of the spinon-phonon interaction as the number of acoustic phonons goes to zero. Although these two processes may occur simultaneously, the last one is privileged because of the absence of field effects on \(\Delta V/V\) below 100 mK (inset Fig. 5).

In opposition with reported thermal conductivity measurements \([13]\), we are thus left with only one contribution to the elastic anomaly originating from magnetic excitations. Nevertheless, our measurements can be reconciled with the thermal transport ones by examining closely the magnetic field dependence. At low temperatures, a substantial contribution to heat transport comes from a spin-mediated process, an observation that is reinforced by an important magnetic field

![FIG. 5. (Color online) Ultrasonic velocity data \((V(H) - V(0))/V(0)\) as a function of magnetic field at fixed temperatures, \(T = 0.25\) K (black), 0.75 K (red), 1.10 K (blue), and 3.10 K (gray); the curves have been shifted vertically from the 0.25 K one to help the comparison. The vertical dashed line indicates the field where the data likely saturate. Inset: field scan at 100 mK up to 2 T.](image)

![FIG. 6. (Color online) Magnetic field sensitive components of the velocity data \((\Delta V/V)_H\) (blue) and of the attenuation \((\Delta \alpha)_H\) (red) as a function of temperature.](image)
dependence below 1 K. Indeed, at the lowest temperatures ($T < 1$ K), a magnetic field oriented perpendicular to the Pd(dmit)$_2$ plane produces a steep increase of the thermal conductivity $\kappa_{xx}$ for low field values and a change of rate near the range 6–8 T. This behavior is less pronounced for higher temperatures near and above 1 K. The existence of excitations, magnetic in nature, over this temperature range is confirmed by the magnetoelastic coupling anomaly (Fig. 6). In zero magnetic field, such a coupling between these excitations and phonons possibly reduces the spin-mediated heat transport. For increasing magnetic field, the magneto-elastic coupling rapidly vanishes and disappears completely near 7 T and the heat transport due to spin excitations is enhanced accordingly.

The presence of anomalies in $^{13}$C NMR relaxation time experiments [25] near 1 K suggests a continuous phase transition that appears to be confirmed below 1.5 K in our ultrasonic experiment. The phase transition could be related to the growth of a spinon Fermi surface as in the other QSL candidate $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. Although the NMR data strongly suggest, together with the thermal conductivity ones, that the ground state does not have fully gapless fermionic magnetic interactions, the presence of spin-gap-like excitations were not detected in our experiment and they are moreover, not necessary to explain the magnetic field dependence. However, the presence of spin-gap-like excitations could be short-circuited by a vanishing magnetoelastic coupling.

IV. CONCLUSION

We have performed a pulsed ultrasonic experiment on a 100 $\mu$m thick EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ organic crystal which is considered as a spin-liquid candidate. At high temperatures, the elastic character is found highly inhomogeneous due to the rotational motion of the methyl and ethyl groups of the EtMe$_3$Sb cations separating the Pd(dmit)$_2$ layers. These motions give rise to frequency dependent softening peaks near 40 K (methyl) and 160 K (ethyl) on the velocity of longitudinal waves propagating perpendicular to the organic planes. Although long relaxation times are associated to these rotations, the elastic character is found practically homogeneous at low temperatures as long as $T$ is kept well below 40 K. Then, the QSL ground state could be properly investigated below 2 K. A velocity softening anomaly was indeed observed over this temperature range and magnetic field effects reveal that two kinds of excitations contribute to it. First, a field independent contribution originates from the quantum rotational motion of the methyl groups that persist at very low temperatures when the acoustic phonons have practically disappeared. Second, a strongly field dependent contribution is due to a coupling between spin excitations and acoustic phonons. These spin excitations or spinons appear below 1.5 K consistently with a progressive phase transition as reported in NMR measurements. However, in opposition with the other QSL organic compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, no pairing instability of these spinons could be observed at the lowest temperatures due to a vanishing magnetoelastic coupling following the loss of acoustic phonons. If spin-gap-like excitations are present in this system, they could not be identified undoubtedly in our experiment.

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