Inelastic Neutron-Scattering Measurements of Incommensurate Magnetic Excitations on Superconducting LiFeAs Single Crystals

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Magnetic correlations in superconducting LiFeAs were studied by elastic and by inelastic neutron-scattering experiments. There is no indication for static magnetic ordering, but inelastic correlations appear at the incommensurate wave vector $(0.5 \pm \delta, 0.5 \mp \delta, 0)$ with $\delta \sim 0.07$ slightly shifted from the commensurate ordering observed in other FeAs-based compounds. The incommensurate magnetic excitations respond to the opening of the superconducting gap by a transfer of spectral weight.

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Superconductivity in the FeAs-based materials [1] appears to be closely related to magnetism as the superconducting (SC) state emerges out of an antiferromagnetic (AFM) phase by doping [1–4] or by application of pressure [5]. The only FeAs-based exception to this behavior has been found in LiFeAs, which is an ambient-pressure superconductor with a high T_C of $\sim$17 K without any doping [6–8]. LiFeAs exhibits the same FeAs layers as the other materials but $FeAs_4$ tetrahedrons are quite distorted [8], suggesting a different occupation of orbital bands. Indeed, angle-resolved photoemission spectroscopy (ARPES) studies on LiFeAs find an electronic band structure different from that in LaOFeAs or $BaFe_2As_2$-type compounds [9]. The Fermi-surface nesting, which is proposed to drive the spin-density wave (SDW) order in the other FeAs parent compounds, is absent in LiFeAs [9], suggesting that this magnetic instability is less relevant. The main cause for the suppression of the nesting consists in the hole pocket around the zone center which is shallow in LiFeAs [10]. Consequently, there is more density of states near the Fermi level which might favor a ferromagnetic (FM) instability. Using a three-band model Brydon *et al.* [10] find this FM instability to dominate and discuss the implication for the SC order parameter proposing LiFeAs to be a spin-triplet superconductor with odd symmetry. However, other theoretical analyses of the electronic band structure still find an AFM instability which more closely resembles those observed in the other FeAs-based materials [11].

Inelastic neutron scattering (INS) experiments revealed magnetic order and magnetic excitations in many FeAs-based families [2,12–14]. Strong magnetic correlations persist far beyond the ordered state, and, most importantly, the opening of the SC gap results in a pronounced redistribution of spectral weight [13–15], which is frequently interpreted in terms of a resonance mode. Recently a powder INS experiment on SC LiFeAs reported magnetic excitations to be rather similar to those observed in the previously studied materials [16] but with a spin gap even in the normal-conducting phase. Magnetic excitations observed in a recent single-crystal INS study on non-SC Li deficient $Li_{1-x}FeAs$ ($x \sim 0.06$) were described by spin waves associated with commensurate antiferromagnetism, again with a large temperature independent spin gap of 13 meV [17]. We have performed INS experiments on SC single-crystalline LiFeAs finding incommensurate magnetic correlations which still can be associated with the SDW order in the other FeAs-based compounds. These incommensurate excitations show a clear response to the SC phase.

Single crystals of LiFeAs were grown similarly as described in Ref. [18]. Further information documenting the good chemical, crystalline, and SC properties of our sample crystals is given in the Supplemental Material [19]. First neutron experiments were performed with small samples containing natural Li (about $12 \times 12 \times 0.3\,\mathrm{mm}^3$) focusing on elastic analyses. We searched for magnetic superstructure peaks, in particular, near the propagation vectors of the known SDW order in FeAs-based compounds [2]. With the high sensitivity of single-crystal neutron diffraction we may rule out this SDW ordering with a magnetic moment larger than $0.07\mu_B$, which is significantly below the ordered moment, for example, in LaOFeAs [20]. None of the scans along main-symmetry directions indicate magnetic ordering. For most of the INS experiments we used two large coaligned crystals of a total weight of 1.4 g grown with the ^{7}Li isotope to reduce the neutron absorption. INS experiments were performed at the thermal spectrometers IN20 [Institut Laue Langevin (ILL)], 1T and 2T [both Laboratoire Léon

Brillouin (LLB)], as well as on the cold spectrometers 4F (LLB) and IN14 (ILL). On IN20 we used the flat-cone detector with silicon (111) analyzers fixing the final energy to 18.7 meV.

Figure 1 shows the maps recorded with the flat-cone detector on IN20. Figure 1(a) represents the results for Co-doped $BaFe_2As_2$ in the SC phase. This pattern demonstrates how easily this instrument may detect the magnetic signal which is found to be similarly strong to that of the phonon scattering around the nuclear Bragg peaks. The same experiment on LiFeAs shown in Fig. 1(b) immediately reveals the differences of the magnetic scattering. Although the phonons in both samples yield signals of similar strength, there is no comparably strong magnetic signal visible in LiFeAs. However, there clearly is magnetic scattering near $Q = (0.5, 0.5, 0)$ which is displaced in the transverse direction to $Q_{\mathrm{inc}} = (0.5 \pm \delta, 0.5 \mp \delta, 0)$ with $\delta \sim 0.07$. Throughout this Letter we label all reciprocal-space vectors in reduced lattice units referring to the lattice with a 3.8 Å parameter. The incommensurate excitation is also visible in the pattern obtained with $l = 0.5$ at 5 meV energy transfer and in the $l = 0$ pattern with 10 meV energy transfer; see Figs. 1(c) and 1(d). The observation of a comparably strong signal for finite l suggests an essentially two-dimensional magnetic correlation; therefore, we neglect a possible out-of-plane modulation in the following discussion. By normalizing with the phonon signals, we may compare the magnetic scattering in LiFeAs and in Co-doped $BaFe_2As_2$. Taking into account the scattering lengths and the reciprocal-energy factor and assessing the phonon dispersion of frequency and of dynamical structure factors with the aid of phenomenological lattice-dynamics models, we may roughly estimate the incommensurate magnetic signal per Fe in LiFeAs at 5 meV and 2 K to be a factor of 8 less intense than the commensurate scattering in Co-doped $BaFe_2As_2$ appearing at 8.5 meV and 10 K [15], but note that integration over the Brillouin zone will recover a factor of 2. The constant-energy maps do not give any indication of a FM scattering in LiFeAs.

Scans across the incommensurate magnetic signal were obtained from the IN20 scattering maps and by additional experiments on cold and thermal triple-axis spectrometers. A few examples of transverse scans, $(h\ 1 - h\ 0)$, are shown in Fig. 2. The profiles were fitted with two symmetric Gaussians appearing at $Q_{\mathrm{inc}} = (0.5 \pm \delta, 0.5 \mp \delta, 0)$ with $\delta \sim 0.07$, to extract the incommensurability and the amplitude of the magnetic signal. In the normal-conducting state we were able to follow the incommensurate signal between 1.5 and 10 meV, finding almost no energy dependence of the incommensurability δ; see Fig. 3(a). Combining the data obtained on the thermal

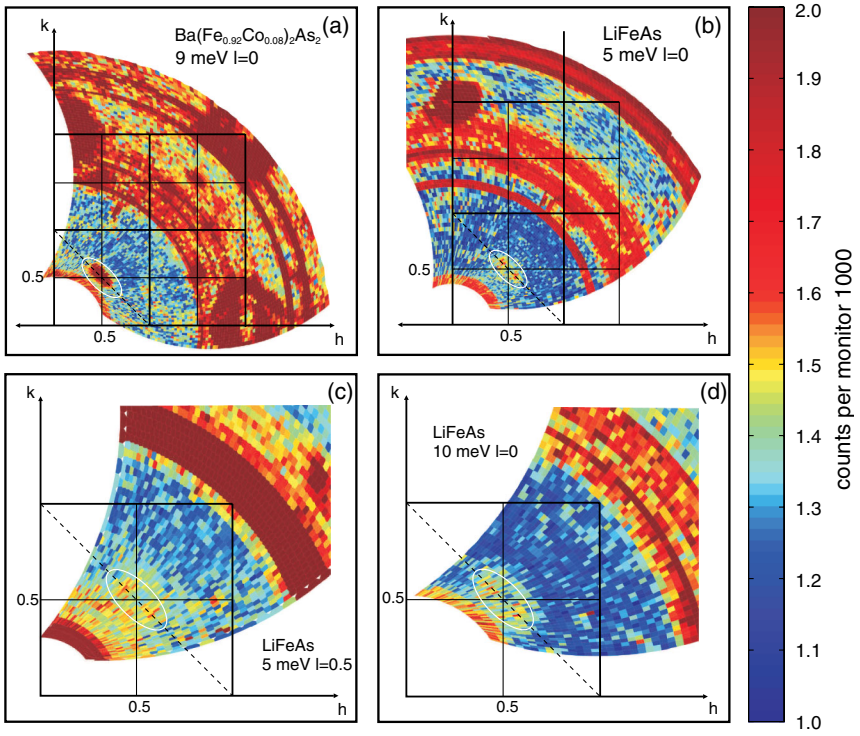

FIG. 1 (color online). Distribution of neutron-scattering intensity measured with the flat-cone detector on IN20. The energy transfer is constant in all maps. (hkl) planes with fixed but finite l component are studied by tilting the detector and the sample. Rings of scattering arise from polycrystalline construction material. (a) The scattering of optimally Co-doped $BaFe_2As_2$ crystal in the SC phase ($T = 10$ K). One easily identifies the magnetic mode near $Q = (0.5, 0.5, 0)$ which is of comparable strength to the phonon scattering around the strong nuclear Bragg peaks. (b)–(d) Intensity maps measured on LiFeAs at an energy transfer of 5 meV and $l = 0$ ($T = 2$ K), of 5 meV and $l = 0.5$ ($T = 2$ K), and of 10 meV and $l = 0$ ($T = 2$ K) for (b), (c), and (d), respectively.

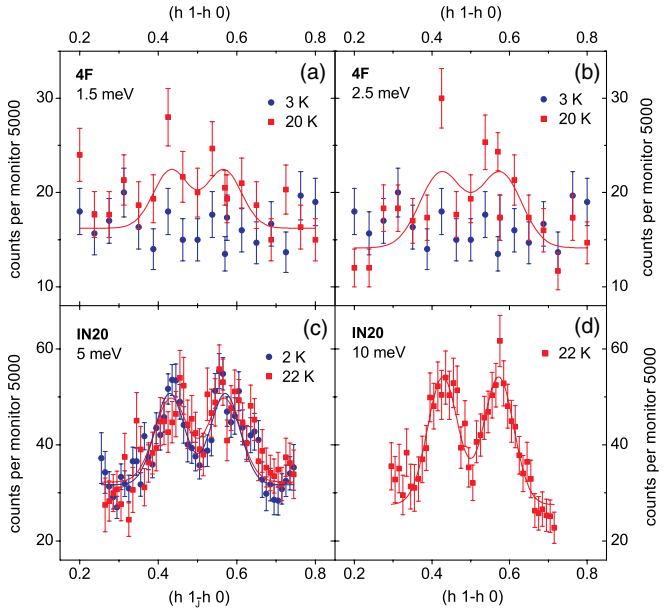

FIG. 2 (color online). Constant-energy scans across the incommensurate positions of magnetic scattering $Q_{\rm inc} = (0.5 + \delta, 0.5 - \delta, 0)$ with $\delta \sim \pm 0.07$. The scans in (a) and (b) were recorded on the cold 4F spectrometer with $k_f = 1.55$ Å^{-1} with an energy transfer of 1.5 and 2.5 meV, respectively. One clearly recognizes the incommensurate signal even at this low energy in the normal state, but the signal disappears in the SC phase. (c), (d) Scans taken from the flat-cone data at 5 and 10 meV, respectively. Lines are fits with two symmetric Gaussians.

and on the cold triple-axis spectrometers we also obtain the energy dependence of the strength of this signal. After correcting for the monitor and for the Bose factor we may deduce the imaginary part of the generalized susceptibility, $\chi''(Q_{\rm inc}, E)$, which is shown in Fig. 3(b) for the temperature of 22 K. $\chi''(Q_{\rm inc}, E)$ can be well described by a single relaxor function relating χ'' with the real part of the susceptibility at zero energy and a characteristic energy Γ: $\chi''(Q_{\rm inc}, E) = \chi'(Q_{\rm inc}, 0)\frac{\Gamma E}{\Gamma^2 + E^2}$ yielding $\Gamma = 6.0 \pm 0.6$ meV. This rather low value of the characteristic energy signals that LiFeAs is quite close to the corresponding SDW instability associated with full softening of the characteristic energy. The incommensurate scattering in LiFeAs and its spectrum closely resemble the incommensurate magnetic excitations arising from nesting in Sr_2RuO_4 where the corresponding SDW instability can be induced by a small substitution [21].

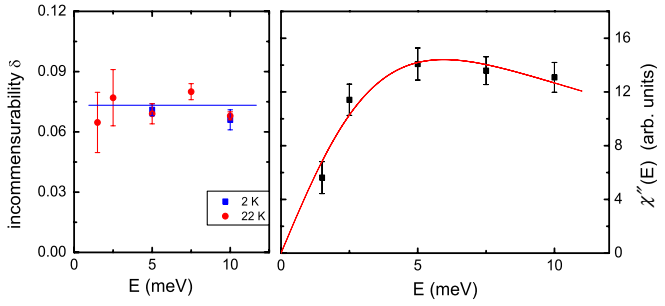

FIG. 3 (color online). (a) Energy dependence of the incommensurability of magnetic scattering in LiFeAs. (b) The energy dependence of the amplitude of the signal can be well described by a single relaxor function (line), see text, with a characteristic energy of $\Gamma = 6.0 \pm 0.6$ meV signaling a near SDW instability.

The comparison of the constant-energy scans above and below the SC transition in Figs. 2(a) and 2(b) reveals a pronounced shift of spectral weight associated with the SC transition in LiFeAs. The incommensurate signals at 1.5 and 2.5 meV become almost fully suppressed in the SC phase. In contrast there is evidence for an increase in intensity at higher energies. In order to elucidate this transfer of spectral weight we performed constant-Q scans at the position of the incommensurate signal, $Q_{\rm inc}$, which are shown in Fig. 4. At low energy in the SC phase the scattering at $Q_{\rm inc}$ is suppressed to the background, while the signal is enhanced in the energy range 6–10 meV. With the present statistics there is no indication for magnetic scattering persisting in the SC state for $E < 4$ meV, suggesting a clean gap in the magnetic excitations in the SC state. ARPES and specific heat measurements on LiFeAs indicate two gaps opening in the bands in LiFeAs which amount to $2\Delta_1 = 2.4$ meV and $2\Delta_2 = 5.2$ meV [22]. The higher gap value agrees with our observation that transfer of spectral weight from below 4.5 meV to above 4.5 meV occurs upon entering the SC phase. Because of the limited statistics of the temperature-dependent data shown in Figs. 5(a)–5(c) we may not yet fully determine the relation between the spectral-weight shift and the SC transition. But the data, in particular that in Fig. 5(b), strongly suggest that the transfer of spectral weight represents the response of the system to the opening of the SC gaps. By measuring the depolarization of the polarized neutron beam due to the shielding of the guide fields we may ascertain the good homogeneity of the SC phase in the large SC single crystals ($T_c = 16.4$ K); see Fig. 5(d).

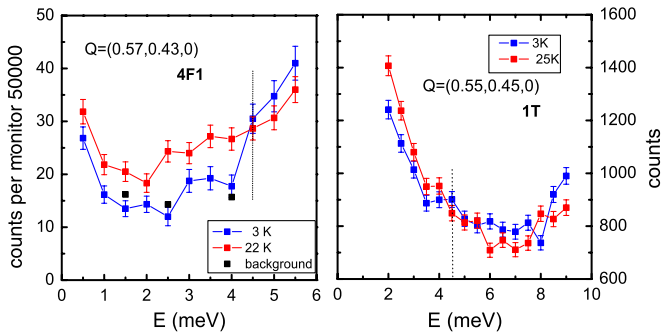

FIG. 4 (color online). Energy dependence of the INS intensity at $Q_{\rm inc}$ at temperatures above and below the SC transition. Panel (a) shows the raw intensity measured on the cold spectrometer (with high-energy resolution, $k_f = 1.55$ Å $- 1$), while panel (b) shows data measured on the thermal spectrometer with lower resolution. Vertical bars indicate the energy of the crossover of the shift of spectral weight.

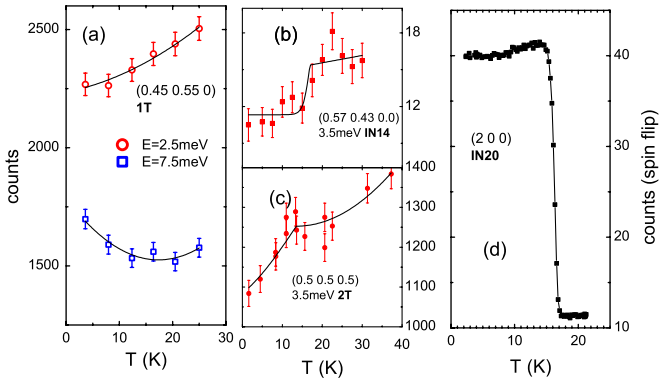

FIG. 5 (color online). Temperature dependence of scattering intensity at Q_{inc} for fixed energy transfers, data are measured on the 1T spectrometer (a) and on IN14 (b) with the two large crystals; data in (c) were taken with a smaller crystal on 2T in a (110)/(001) scattering geometry where the incommensurate signal is integrated with the relaxed vertical resolution. Lines are guides to the eye. (d) Temperature dependence of spin-flip scattering at the nuclear Bragg peak (200) reflecting the neutron depolarization due to SC shielding.

The experimental data obtained by powder INS and the given interpretation [16] only qualitatively agree with our experiment. The incommensurate character of magnetic excitations in LiFeAs is not easily accessible in a powder experiment, but our results fully disagree with the claim of a spin gap in the normal state that is formulated in Ref. [16]. We speculate that the background and phonon contributions underlying the magnetic signal could not be properly assessed, thereby underestimating the magnetic response of LiFeAs.

Magnetic excitations in LiFeAs clearly differ from those reported for many other FeAs-based superconductors in at least two aspects. The amplitude of the signal is weaker than that in Co-doped $BaFe_2As_2$. More importantly the signal clearly appears at an incommensurate position, whereas those in the FeAs superconductors with a high T_C are all commensurate. Incommensurate scattering has recently been observed in the end member of the $Ba_{1-x}K_xFe_2As_2$ series KFe_2As_2 [23] which, however, exhibits a very low T_C. The less closely related Fe(Te/Se) compounds also exhibit incommensurate magnetic excitations in the SC concentration range [24], but there the incommensurability varies more strongly with energy and the magnetism of the parent compound is fully different.

The incommensurability in the magnetic scattering seems to be the consequence of the suppressed nesting condition in LiFeAs. In the plots relating the SC transition temperature with either the As-Fe-As bond angles [25] or with the anion height from the Fe layer [26], LiFeAs clearly lies on the wing of the distribution away from the regular tetrahedron observed in samples with maximum T_C. The sizable tetrahedron elongation in LiFeAs should cause a different orbital occupation. It appears interesting to note that concerning these distortions LiFeAs resembles $FeTe_{\sim 0.5}Se_{\sim 0.5}$, which exhibits similar incommensurate excitations [24]. It appears likely that doping and structural deformation change the occupation of orbital levels and thereby the character of the magnetic instability. Density-functional theory calculations indicate that doping electrons or holes into the FeAs layers modifies the nesting with the magnetic response shifting from the commensurate position to an incommensurate one [27,28]. Hole doping implies a longitudinal modulation peaking at $Q = (0.5 \pm \delta, 0.5 \pm \delta, 0)$ which indeed is observed in hole-overdoped KFe_2As_2 [23], whereas electron doping results in a transverse modulation peaking at $Q = (0.5 \pm \delta, 0.5 \mp \delta, 0)$ [27,28]. Experimental evidence for such transverse inelastic incommensurability in the FeAs-based materials can so far only be found in the high-energy magnetic excitations in Co-doped $BaFe_2As_2$ [29], which, however, are associated with commensurate scattering at lower energy. Very recently, a static transverse modulation was reported for underdoped $Ba(Fe_{1-x}Co_x)_2As_2$ [30]. Our observation of transversally modulated incommensurate excitations in LiFeAs thus suggests to compare LiFeAs with an electron doped compound. The similarity can arise from the role of the central hole pocket which is shallow in LiFeAs somehow similar to the expected effect of electron doping. The transversally modulated incommensurate response in LiFeAs, however, still seems to be closely related with the commensurate or longitudinally modulated response in the other FeAs-based materials.

Inspection of the Fermi surfaces calculated in Ref. [11] or those fitted to the ARPES data [10] allows one to understand that the commensurate wave vector $(0.5, 0.5, 0)$ is not associated with the strongest magnetic signal in LiFeAs as such nesting is absent in this material. However, an additional shift may partially recover nesting. Taking the orbital character of the Fermi-surface sheets into account, the experimentally determined transversal incommensurability of $\delta \sim 0.07$ agrees with the Fermi-surface maps presented in Refs. [10,11], but a detailed calculation is desirable. Note that deciding between dominating FM or AFM instability depends sensitively on the choice of the interaction.

In conclusion, INS experiments on single-crystalline SC LiFeAs reveal incommensurate magnetic correlations, which appear close to the wave vector of the stronger magnetic signal observed in previously studied FeAs-based superconductors. The loss of commensurate nesting for $q = (0.5, 0.5, 0)$ apparently needs to be compensated by a small shift explaining the incommensurate propagation vector $Q_{\mathrm{inc}} = (0.5 \pm \delta, 0.5 \mp \delta, 0)$ with $\delta \sim 0.07$. These magnetic fluctuations clearly respond to the opening of the SC gap. In the SC phase the magnetic weight at Q_{inc} seems to be fully suppressed below $\sim$5 meV and there is an enhancement of spectral weight compared to the normal state in the energy range 6–10 meV. The magnetic instability in LiFeAs indicates that magnetic correlations in FeAs-based superconductors are more variable than a simple commensurate response.

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