Microscopic coexistence of a two-component incommensurate spin density wave with superconductivity in underdoped $NaFe_{0.983}Co_{0.017}As$

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We have performed ^{75}As and ^{23}Na nuclear magnetic resonance (NMR) measurements on a single crystal of $NaFe_{0.9835}Co_{0.0165}As$ and found microscopic coexistence of superconductivity with a two-component spin density wave (SDW). Using ^{23}Na NMR we measured the spatial distribution of local magnetic fields. The SDW was found to be incommensurate with a major component having magnetic moment ($\sim$0.2 μ_B/Fe) and a smaller component with magnetic moment ($\sim$0.02 μ_B/Fe). Spin lattice relaxation experiments reveal that this coexistence occurs at a microscopic level.

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The relationship between antiferromagnetism (AFM) and superconductivity in unconventional superconductors, such as cuprates and heavy fermion superconductors, is an intriguing problem of substantial current interest.[1,2] In particular, demonstrating coexistence of these two condensations on a microscopic scale is of special importance given the antithetical nature of magnetism and superconductivity.[3–5] Although it has been shown that iron-based superconductivity[6] can coexist with a spin density wave (SDW),[7–17] in order to better understand this phenomenon it would be helpful to have a clear determination of the spatial distribution of local fields using a high-resolution probe. In this paper, we identify an incommensurate SDW that coexists with superconductivity in underdoped $NaFe_{0.983}Co_{0.017}As$ (NaCo17) taking advantage of very narrow NMR spectra that provide a faithful visualization of the local field distribution. We find that the SDW has unusual character appearing with two components, one with an amplitude an order of magnitude larger than the other.

Theory predicts that coexistence of an SDW and superconductivity is possible where phases of the superconducting wave functions on different portions of the Fermi surface with $s_\pm$ gap symmetry are different by a factor of π for either isotropic or anisotropic superconducting gaps.[18] It has been argued that an incommensurate SDW is more likely to coexist with superconductivity than for a commensurate SDW.[19,20] In contrast, for s_{++} gap symmetry where the phase of the superconductor is a constant, coexistence is only possible when the superconducting gap has nodes.[21] Nuclear magnetic resonance (NMR) gives a direct measure of distributions of local magnetic fields, utilized previously to study SDW's in pnictides.[9,12,13] Here we investigate a high-quality pnictide single crystal, free of paramagnetic impurities, using two resonances, ^{75}As and ^{23}Na. The two nuclei are located on opposite sides of the Fe layer, Fig. 1(b), providing complementary views of the local fields as they develop in the SDW state. The As nucleus has a strong hyperfine coupling to the conduction electrons, in contrast to the Na nucleus, which is much more weakly coupled by a factor of $\sim$20.[22] Owing to the unusually narrow linewidth of ^{23}Na NMR in NaCo17, $\sim$4 kHz at $T = 30$ K for $H_0 = 16.36$ T, we have determined the spatial distribution of the incommensurate SDW in the normal and superconducting states shown schematically in the phase diagram in Fig. 1(a).

The ^{75}As and ^{23}Na NMR experiments were performed at Northwestern University and the National High Magnetic Field Laboratory in Tallahassee, Florida. The range of temperature was from 2.2 K to room temperature with external magnetic fields from $H_0 = 6.4$ to 16.36 T parallel to the c axis or ab plane. For the results reported here, the central-transition linewidth of the ^{23}Na NMR has been stable at 2.5 kHz for two months, taken at room temperature with $H_0 = 6.4$ T parallel to the c axis; our criterion that there is no degradation of the sample. During the same period the linewidth of the ^{75}As NMR has remained stable at 8 kHz. The $\sim 3 \times 2 \times 0.3$-mm^3 crystal of $NaFe_{0.983}Co_{0.017}As$ was grown at the University of Tennessee and found to have T_c of 18 K from magnetization measurement in low field. Hahn-echo sequences ($\pi/2$-$\pi/2$) were used to obtain spectra, and spin-lattice relaxation rates, $1/T_1$, at the central transition ($-1/2 \leftrightarrow 1/2$) with a $\pi/2$ pulse of $\approx$6 μs. A frequency sweep method was used to cover the broadened NMR spectra in the SDW state. T_1 was obtained with a saturation recovery method for $T_1 < 2$ s. For longer T_1 the more efficient progressive saturation technique[23] was used.

The ^{23}Na NMR spectra are shown in Fig. 2. In the paramagnetic state, $T \gtrsim 30$ K, the central and satellite transitions have very narrow linewidths $\approx$2.5 (4) kHz in 6.4 (16.36) T, even in high fields, demonstrating high crystal quality. On cooling, NaCo17 enters into the SDW state at $T_{SDW} \approx 27$ K. Since Na or As atoms are centered symmetrically above or below four Fe atoms, there is a net magnetic field, H_{SDW}, at the As or Na sites along the c axis due to in-plane magnetic moments, m, at the Fe site.[25] Thus, a peak in the NMR spectrum at H_0 at high temperatures splits into $H_0 - H_{SDW}$ and $H_0 + H_{SDW}$ in the SDW state. H_{SDW} can be expressed as 4 $A_{HF}\,m$ where A_{HF} is the hyperfine coupling constant, 0.023 T/μ_B for Na.[22,26] This splitting is clearly demonstrated at $T = 18$ and 20 K, Fig. 2. We could not sweep the complete spectrum owing to available time so the spectra in Fig. 2(b) above 72 (184.52) MHz at

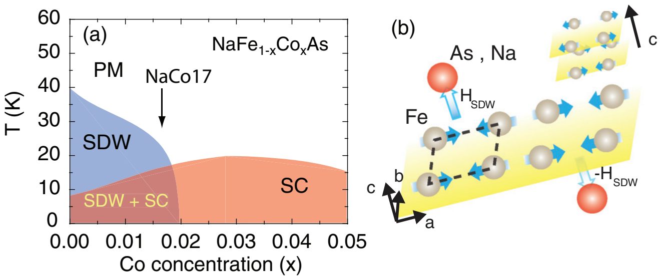

FIG. 1. (Color online) (a) The phase diagram of $NaFe_{1-x}Co_xAs$ as a function of Co concentration.[24] The downward arrow indicates the doping concentration of our Co-underdoped NaCo17 crystal. (b) Alignment of magnetic moments in the SDW state. There is a net alternating magnetic field, H_{SDW}, at both As and Na sites along the c axis.

T = 20 (18) K were limited at high frequency and are not shown. From the distribution of the local fields as two broad humps, associated with either the central or the satellite transitions, we can identify that the SDW is incommensurate. Otherwise, each NMR transition would be split into discrete spectra.

Additionally, there is an unexpected peak in the middle of the ^{23}Na spectrum in the SDW state. We attribute this peak to a small amplitude SDW (S-SDW). The absence of the S-SDW at the satellite position indicates that the electric field gradient (EFG) is significantly different or disordered at the location of those Na atoms that contribute to the narrow middle peak of the central transition, which is only sensitive to second order in the EFG. However, the satellites are broadened by the EFG at first order. Existence of an SDW below T_c provides evidence for its coexistence with superconductivity, where the transition temperature was determined from coil detuning and spin lattice relaxation. From the known hyperfine field[22] we determined that the moment m for the larger SDW (L-SDW) was $0.2\,\mu_B$ at $T = 4.2$ K and $H_0 = 16.36$ T. If we treat the additional contribution to the linewidth of the narrower middle

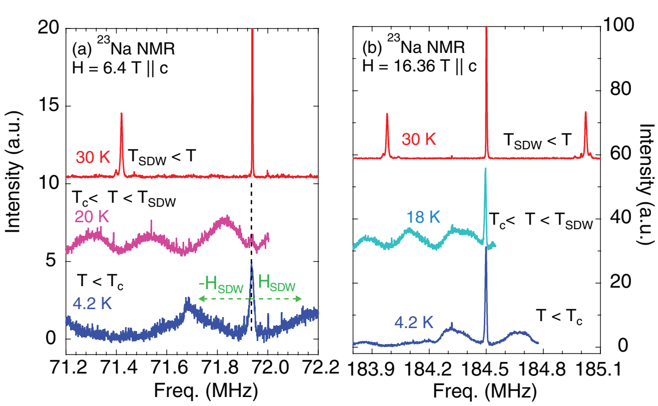

FIG. 2. (Color online) (a) ^{23}Na NMR spectra from NaCo17 in $H_0 = 6.4$ T parallel to the c axis. (b) Same as described in (a) but for $H_0 = 16.36$ T. T_{SDW} and T_c are 27 and 14 K (12 K), respectively, in H = 6.4 T (16.36 T). Two types of SDW were found in the SDW state: a large amplitude SDW (L-SDW) with a splitting in the spectrum ~0.25 MHz (0.02 T), and a small SDW (S-SDW), broadening the spectrum ~0.02 MHz (0.0018 T).

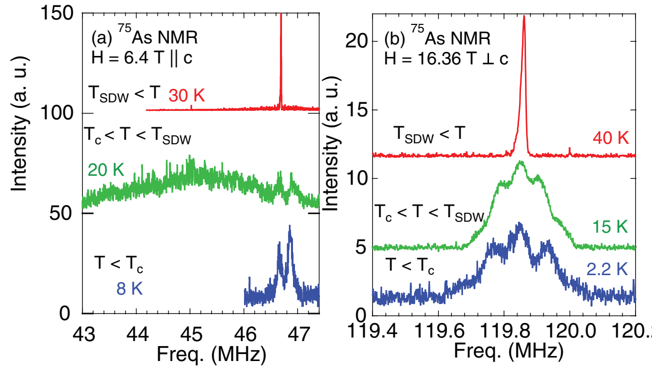

FIG. 3. (Color online) (a)^{75}As NMR spectra from NaCo17 in $H_0 = 6.4$ T parallel to the c axis. The splitting near 46.65 MHz in the SDW and superconducting states indicates that the central peak in the ^{23}Na NMR is due to a S-SDW not inhomogeneity. (b) ^{75}As NMR spectra in $H_0 = 16.36$ T || ab plane.

peak as an unresolved splitting, we find the moment for the S-SDW to be $0.02\,\mu_B$, under the same conditions.

In Fig. 3(a) we show ^{75}As spectra for the paramagnetic phase at $T = 30$ K, in the SDW state $T = 20$ K, and in the superconducting state at $T = 8$ K for $H_0 = 6.4$ T along the c axis. In the SDW state, the wide hump at low frequency for the NMR central transition indicates formation of a large moment SDW. The frequency interval from the unshifted central transition to the peak of the hump is ~0.2 T (1.5 MHz) at $T = 20$ K, as compared with ^{23}Na NMR for which the corresponding interval is ~0.009 T (0.1 MHz). If we allow for the hyperfine field ratio, $A_{HF}^{Na}/A_{HF}^{As} \approx 1/20$,[22] the ^{75}As and ^{23}Na NMR consistently depict the same amplitude for the L-SDW at $T = 20$ K. We also observe an appearance of two ^{75}As peaks at the unshifted position with a splitting of 0.034 T (0.25 MHz) that indicates a small moment SDW with (~$0.018\,\mu_B$) at the Fe site, also in-plane, consistent with the broadening of the ^{23}Na spectra discussed above. It is interesting that the center of the split spectrum in Fig. 3(a) does not quite match the peak of the spectrum at $T = 30$ K. On further cooling, the splitting persists within the superconducting state. We did not perform a frequency sweep to cover the whole range of the L-SDW at T = 8 K due to a drastic decrease in the signal-to-noise ratio in the superconducting state and the rather wide spread in H_{SDW} at the As site. We note that the small magnetic moment S-SDW is consistent with reports of the amplitude of the SDW observed by neutron scattering on the same material, ~$0.03\,\mu_B$.[15]

If the external magnetic field, H_0, is aligned perpendicular to the c axis, then the net magnetic field at either As or Na sites in the SDW state becomes $H_{net} = \sqrt{H_0^2 + H_{SDW}^2}$, where H_{SDW} is parallel to the c axis and its shift in NMR frequency is quadratically suppressed. Thus, there is no splitting expected in the SDW state when H_0 is strictly parallel to the ab plane. If there is misalignment of H_0 from the ab plane, H_{net} changes to $\approx H_0 \pm H_{SDW}\sin\theta$, where θ is the misalignment angle. Then we expect two peaks at $H_0 \pm H_{SDW}\sin\theta$. Figure 3(b) shows ^{75}As NMR spectra in the normal state ($T = 40$ K), SDW ($T = 15$ K), and superconducting ($T = 2.2$ K) states where $H_0 = 16.36$ T parallel to the ab plane. In the SDW state and below T_c there are three peaks, two on each side of the unshifted

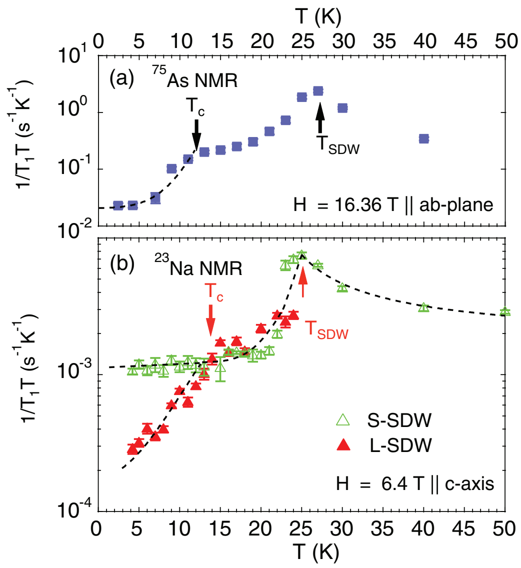

FIG. 4. (Color online) (a) $1/^{75}T_1T$ measured for the S-SDW spectra at $H_0 = 16.36$ T $||ab$ plane. (b) $1/^{23}T_1T$ at the peak in the spectrum corresponding to each of L-SDW and S-SDW, $H_0 = 6.4$ T $||c$ axis. For both nuclei a decrease of the rate on cooling through T_c shows coexistence of the L-SDW with superconductivity on a microscopic scale. Note that there is minimal change of the transition temperatures with magnetic field in the case of S-SDW from $1/^{23}T_1T$.

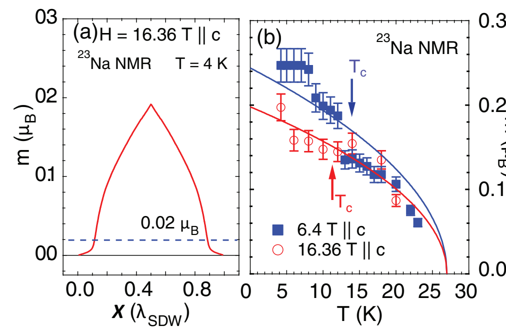

FIG. 5. (Color online) (a) Spatial distribution of magnetic moment, m, with period λ_{SDW} in the superconducting state calculated from ^{23}Na NMR.[26] The behavior of the S-SDW is separated from the L-SDW by the dashed blue line. (b) The temperature dependence of the most probable magnetic moment for the L-SDW from the ^{23}Na NMR spectrum is shown for two magnetic fields.

position stemming from the L-SDW and misalignment ($\sim 1^\circ$), and the central peak appears to be from the S-SDW. However, in order to distinguish which portion of the spectra corresponds to nuclei affected by superconductivity, we have measured spin-lattice relaxation rates, $1/T_1T$.

Figure 4(a) shows $1/^{75}T_1T$ at the middle peak of the three-peak spectrum in Fig. 3(b) (S-SDW) with $H_0 = 16.36$ T $\sim$parallel to the ab plane. On cooling, $1/^{75}T_1T$ increases due to spin fluctuations[22,27,28] until $T_{\mathrm{SDW}} = 27$ K. Below this temperature, $1/^{75}T_1T$ suddenly drops Arrhenius-like.[29] Angle-resolved photoemission (ARPES) measurements on a similar NaCo17 sample[15] have shown that there is a remnant density of states at the Fermi surface in the SDW state, and consequently $1/^{75}T_1T$ saturates at low temperatures. On further cooling, below T_c, the rate decreases more rapidly due to opening of superconducting gaps at the Fermi surface.[12,13] Thus, the existence of superconductivity at the spatial location of the S-SDW is clear. Additionally, $1/^{75}T_1T$ at either of the two side peaks corresponding to the L-SDW was found to be the same as the middle peak within 10% at $T = 2.4$ K and consequently we infer that this coexistence with superconductivity exists at a microscopic level uniformly throughout the sample.

We have also performed $1/^{23}T_1T$ measurements at different spectral positions: the central peak (S-SDW) and the hump corresponding to the L-SDW of the ^{23}Na NMR spectra in different fields along the c axis. The results are shown in Fig. 4(b).

The spin-lattice relaxation rate $1/^{23}T_1T$ behaves similar to $1/^{75}T_1T$ increasing on cooling in the normal state and dropping abruptly below $T_{\mathrm{SDW}} = 27$ K and then saturating in the SDW state. However, at the S-SDW position in the ^{23}Na spectra below T_c, $1/^{23}T_1T$ saturates similar to what was noted for optimally doped $\mathrm{NaFe_{0.975}Co_{0.025}As}$(NaCo25)[22] in contrast to $1/^{23}T_1T$ at the L-SDW position which drops below T_c similar to $1/^{75}T_1T$.

We can calculate[26,30] the real space distribution of local fields with periodicity λ_{SDW} attributed to an incommensurate SDW with magnetic moment $m = H_{\mathrm{SDW}}/4A_{\mathrm{HF}}$ at the Fe sites using our ^{23}Na NMR spectra in $H_0 = 16.36$ T. Figure 5(a) shows this spatial distribution as a function of distance, x, in units of the SDW period, where there are two components to m. One is the dominant L-SDW with a large magnetic moment, $m = 0.2\,\mu_B$ and the other is the small amplitude SDW (S-SDW) with moment, $m = 0.02\,\mu_B$, evident in the foot of the distribution below the dashed line in Fig. 5(a). The kink at $x = 0.5$ is an artifact from setting the maximum range of the H_{SDW}. Allowing for effects of doping, this m corresponds well with the magnetic moments from NMR measurements in undoped NaFeAs, where m was found to be $\approx 0.3\,\mu_B$.[26] We also note that the small moment SDW amplitude is comparable to that found in neutron scattering measurements, $\sim 0.03\,\mu_B$[15] and we suggest that this might be the dominant contribution in these experiments. Temperature and magnetic field dependence of the most probable moment from the ^{23}Na NMR spectrum is shown in Fig. 5(b). Below T_c, the moment slowly increases on cooling in contrast to the decrease reported from neutron scattering measurements, which in that case corresponds to the order of magnitude smaller moment.[7,10,15] As the ARPES measurements in Co underdoped NaFeAs indicate that significant energy band modification due to the SDW happens at the energy levels that are far from the Fermi energy, more than -50 meV.[15] Thus, the discrepancy between the NMR measurement and the neutron scattering can be explained by the different energy excitation scale between the two probes.

The most natural explanation for the existence of a small moment S-SDW is the disruption of the L-SDW that takes place in the vicinity of a Co atom substituted on an Fe site. This will suppress the magnetic moment and affect the local H_{SDW} sensed by ^{23}Na and ^{75}As NMR. If this were to correspond to near-neighbor and next-near-neighbor Na positions relative to Co, it would affect ∼ 20% of the spectrum in good agreement with our assessment of the S-SDW fraction of the NMR spectra. Furthermore, the S-SDW should be transverse to the L-SDW to coexist with it. Measurements with well-known and controlled concentrations of dopant will be necessary to confirm this hypothesis.

In summary, we have performed ^{23}Na and ^{75}As NMR experiments to show microscopic coexistence of an incommensurate spin density wave and superconductivity, compatible with $s_{\pm}$-wave pairing symmetry for $NaFe_{0.983}Co_{0.017}As$. The SDW has two components differing by an order of magnitude in amplitude, the smaller of which might be associated with the Co substituting on an Fe site.

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