From Ising Resonant Fluctuations to Static Uniaxial Order in Antiferromagnetic and Weakly Superconducting $CeCo(In_{1-x}Hg_x)_5(x=0.01)$

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$CeCo(In_{0.990}Hg_{0.010})_5$ is a charge doped variant of the *d*-wave $CoCoIn_5$ superconductor with coexistent antiferromagnetic and superconducting transitions occurring at $T_N = 3.4$ and $T_c = 1.4$ K, respectively. We use neutron diffraction and spectroscopy to show that the magnetic resonant fluctuations present in the parent superconducting phase are replaced by collinear *c*-axis magnetic order with three-dimensional Ising critical fluctuations. No low-energy transverse spin fluctuations are observable in this doping-induced antiferromagnetic phase and the dynamic resonant spectral weight predominately shifts to the elastic channel. Static ($\tau > 0.2$ ns) collinear Ising order is proximate to superconductivity in $CeCoIn_5$ and is stabilized through hole doping with Hg.

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Strong magnetic fluctuations are not compatible with conventional superconductivity [1]; however, they are believed to be consistent with a superconducting gap with nodes such as *d*-wave symmetry [2]. The critical point separating magnetic order and superconductivity is often proximate to new phases [2–7]. For example, the cuprate superconducting dome is bracketed by both a pseudogap phase [8,9] and a Fermi liquid [10]. In pnictides, nematic order [11] occurs in the vicinity of superconductivity. A signature that magnetic fluctuations are important for new superconducting orders is the presence of a magnetic resonance peak observed in many magnetic unconventional superconductors including cuprates [12–15], $CeCu_2Si_2$ [16], UPd_2Al_3 [17–20], pnictides [21,22], and $CeCoIn_5$ [23–25] associated with a gap function that undergoes a change in sign [26,27]. Magnetic resonant excitations also occur when other order parameters are present [28] with the observation of an exciton mode in CeB_6 an example of this [29]. We investigate the magnetic fluctuations in the $CeCoIn_5$ *d*-wave superconductor charge doped to long-range antiferromagnetic (AFM) order. The results illustrate the instability of transverse "spin waves" in unconventional superconductors in favor of Ising-like fluctuations which are condensed via charge doping.

$CeCoIn_5$ displays unconventional superconductivity with a transition temperature of $T_c = 2.3$ K [30] and a *d*-wave superconducting order parameter [31]. The crystallographic structure consists of a tetragonal unit cell with layers of magnetic Ce^{3+}-In planes stacked along *c*. Neutron scattering shows the electronic normal state consists of overdamped magnetic excitations peaked near $\vec{Q} = (1/2, 1/2, 1/2)$ indicative of antiferromagnetic interactions between the Ce^{3+} ions, both within the *a*-*b* plane and along *c*. The commensurate magnetic spin response differs from nonsuperconducting $CeRhIn_5$ (at ambient pressure), which displays a magnetic Bragg peak at the incommensurate $\vec{Q} = (1/2, 1/2, 0.297)$ [32] characterizing a helical magnetic structure [33]. The *a*-*b* magnetic helix in $CeRhIn_5$ contrasts with the commensurate *c*-axis polarized resonant fluctuations that dominate the neutron response in superconducting $CeCoIn_5$ [23].

Doping impurities into superconductors has been used to break superconducting Cooper pairs revealing competing phases [34]. Efforts in the "115" system originally were directed to alloying on the Co site in $CeCoIn_5$ with either Rh or Ir as a means of tuning from superconducting to magnetic order [35,36]. However, this phase diagram is complex with $CeRhIn_5$ displaying both helical magnetic order [33] and a low-temperature superconducting phase under pressure [37–43]. Several commensurate magnetic phases are also believed to compete with helical magnetic order [32,44,45] and superconductivity with Rh-Ir alloying. Also, $CeIrIn_5$ is a superconductor with a reduced T_c of

~0.4 K [46]. Replacing Ce by La has been shown to result in a suppression of T_c [47–49].

Another means of electronic tuning $CeCoIn_5$ with nonmagnetic impurities is through the In site with either electron doping (with Sn) or hole doping (with Cd, Hg, or Ru) [50–54]. Doping magnetic Yb on the Ce site has also been pursued; however, it is unusual as the suppression of the superconductivity order parameter with doping is very mild [55,56] and penetration depth measurements [57] even suggest that nodal *d*-wave superconductivity may be replaced by a fully gapped order parameter [58]. In contrast, hole doping with Cd, Hg, or Zn on the In site strongly suppresses superconductivity [59–62] in favor of a commensurate antiferromagnetic state with a characteristic wave vector of $\vec{Q} = (1/2, 1/2, 1/2)$ [32,63,64]. We note that this commensurate order contrasts with the incommensurate spin-density wave reported at large magnetic fields in the superconducting state [65–70].

We apply neutron diffraction and spectroscopy to study the static and dynamic magnetism in Hg doped $CeCoIn_5$. $CeCo(In_{1-x}Hg_x)_5$ samples were grown from In/Hg flux. Nominal Hg substitution for In of 7% and 9% resulted in $x = 1.0\%$ and 1.3%, respectively [Fig. 1(d)]. Elastic scattering measurements used the D23 diffractometer (ILL, France) and spectroscopic measurements were done on the IN12 (ILL, France) and the MACS (NIST, USA) cold triple-axis spectrometers using a coalignment of ~150 crystals (total mass of 4 g). Experimental details are provided in the Supplemental Material [71].

The static magnetic properties of antiferromagnetic and superconducting $CeCo(In_{0.990}Hg_{0.010})_5$ and $CeCo(In_{0.987}Hg_{0.013})_5$ are shown in Fig. 1 and compared to helically magnetically ordered $CeRhIn_5$ [33]. The intensity of the magnetic Bragg peak is a measure of the magnetic order parameter and is fit to a power law near T_N with $I(T) \propto |M(T)|^2 \propto (T_N - T)^{2\beta}$. Hg doped $CeCoIn_5$ samples in Figs. 1(a) and 1(b) are shown with a best fit of $\beta = 0.33 \pm 0.02$ and 0.31 ± 0.02 with magnetic transitions of $T_N = 3.41$ and 3.51 K, respectively. $CeRhIn_5$ is illustrated in Fig. 1(c) for reference with a similar analysis giving $\beta = 0.19$. The Hg doped samples are within error of the three-dimensional Ising universality class where $\beta = 0.326$ [72]. In contrast, $CeRhIn_5$ is consistent with a two-dimensional order parameter, a property reflected in the low-energy magnetic excitations [33,73], and the anisotropy of the correlation lengths [74]. Therefore, three-dimensional critical dynamics coexist with superconductivity in $CeCo(In_{1-x}Hg_x)_5$. A similar result has been noted for pnictide superconductors near the boundary between antiferromagnetism and superconductivity [75–77].

The antiferromagnetic order parameter in $CeCo(In_{0.990}Hg_{0.010})_5$ [Fig. 1(a)] is suggestive of a saturation at the superconducting transition T_c. Confirming this is the comparison in Fig. 1(b) that illustrates the magnetic order parameter for a Hg doping of 1.3% with a superconducting transition $T_c = 1.4$ K. NMR measurements indicate that magnetism forms via localized droplets, which are effectively decoupled from superconducting components of the sample [78]. Despite this apparent decoupling of magnetic and superconducting orders, the magnetic order parameter for Figs. 1(a) $CeCo(In_{0.990}Hg_{0.010})_5$ and 1(b) $CeCo(In_{0.987}Hg_{0.013})_5$ shows a saturation at the superconducting T_c, therefore indicating superconductivity interrupts the continuous formation of magnetic order. We note that a similar low temperature saturation of the magnetic order parameter has been reported in coexistent antiferromagnetic order and superconductivity $CeCo(In_{1-x}Cd_x)_5$ [63].

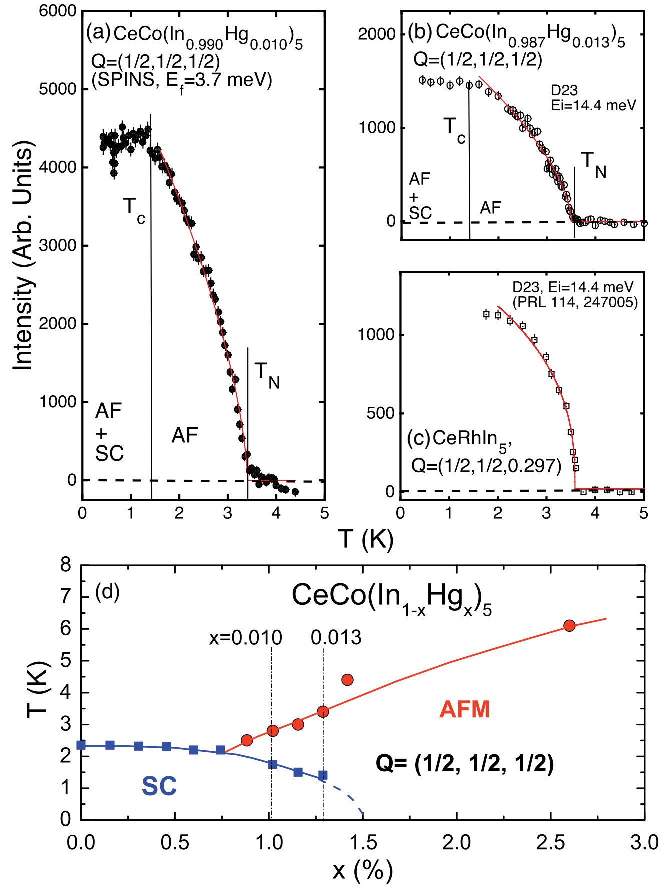

FIG. 1. The magnetic Bragg peak intensity measured as a function of temperature in superconducting and antiferromagnetic (a) $CeCo(In_{0.990}Hg_{0.010})_5$, (b) $CeCo(In_{0.987}Hg_{0.013})_5$, and (c) magnetic helically ordered and nonsuperconducting $CeRhIn_5$ (from Ref. [33]). The SC transition T_c and Néel T_N temperatures are indicated by vertical lines. (d) Temperature composition $T - x$ phase diagram of $CeCo(In_{1-x}Hg_x)_5$ determined from specific heat measurements.

The low temperature Ce^{3+} ordered moment in $CeCo(In_{0.987}Hg_{0.013})_5$ was measured by calibrating against 7 nuclear Bragg peaks to be $0.98 \pm 0.2\mu_B$, as outlined in the Supplemental Material [71], while the ordered moment of $CeRhIn_5$ and Cd doped $CeCoIn_5$ is $\sim 0.3\mu_B$ and $0.7\mu_B$, respectively [45,63,78]. Hg doped magnetic order is characterized by a magnetic moment pointing along the c axis evidenced by a large suppression of intensity at the (1/2, 1/2, 3/2) and (1/2, 1/2, 5/2) magnetic Bragg peaks (see Supplemental Material [71]). This contrasts with the in-plane helical order of $CeRhIn_5$ and also Rh doped $CeCoIn_5$ [44].

We now discuss the dynamics in superconducting and antiferromagnetic $CeCo(In_{0.990}Hg_{0.010})_5$ summarized in Fig. 2 and compared to superconducting $CeCoIn_5$ [23]. Figures 2(a) and 2(b) illustrate low temperature constant $\vec{Q} = (1/2, 1/2, 1/2)$ scans taken in the superconducting state of both $CeCoIn_5$ and $CeCo(In_{0.990}Hg_{0.010})_5$. The scans have been normalized to sample mass and confirmed through a comparison of the elastic incoherent scattering. The superconducting resonance peak in $CeCoIn_5$ at $\sim$0.5 meV is not observed in antiferromagnetic and superconducting $CeCo(In_{0.990}Hg_{0.010})_5$ within experimental resolution at $T = 0.5$ K with the solid line in Fig. 2(b) denoting the measured high temperature background. Instead, at

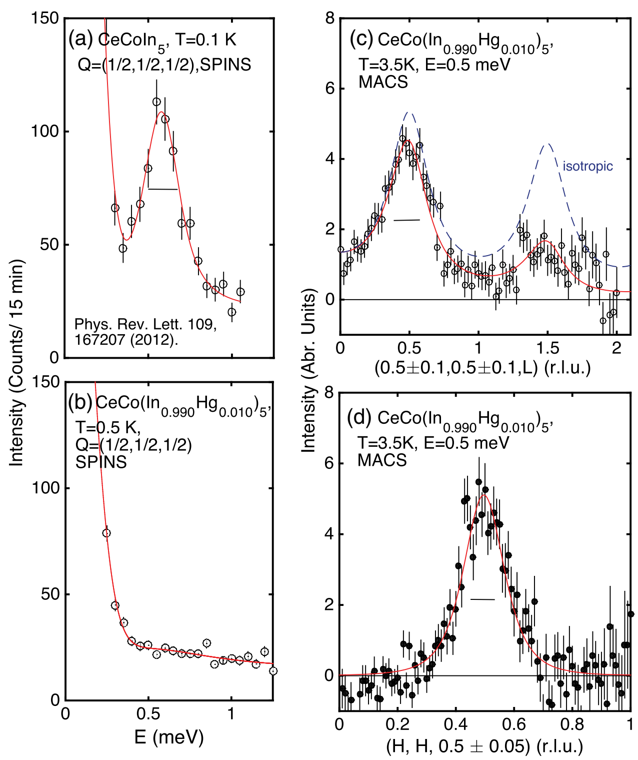

FIG. 2. Constant $\vec{Q} = (1/2, 1/2, 1/2)$ scans for superconducting (a) $CeCoIn_5$ and superconducting and antiferromagnetic (b) $CeCo(In_{0.990}Hg_{0.010})_5$ with both plots normalized to the same absolute scale. Panel (c) illustrates the critical magnetic fluctuations showing polarization along c and (d) antiferromagnetic correlations within the a-b plane of $CeCo(In_{0.990}Hg_{0.010})_5$. The solid lines in (c) and (d) are fits to Eq. (1) corresponding to Ce^{3+} moments polarized along the [001] direction. The dashed line is the same fit to a model with the isotropic magnetic moments having no preferential direction.

temperatures near T_N [Fig. 1(a)] of 3.5 K, magnetic critical scattering associated with the development of long-range static AFM order in $CeCo(In_{0.990}Hg_{0.010})_5$ is observed with the momentum dependence illustrated in Figs. 2(c) and 2(d) from background corrected scans taken using MACS.

Figures 2(c) and 2(d) show that the magnetic critical dynamics in $CeCo(In_{0.990}Hg_{0.010})_5$ is highly anisotropic, mimicking the Ising-like critical scattering discussed above. The solid line in Figs. 2(c) and 2(d) is a fit to

$$I(\vec{Q}) \propto f(Q)^2[1 - (\hat{Q}\cdot\hat{c})^2] \times \frac{\sinh(c/\xi_c)}{\cosh(c/\xi_c) + \cos(\vec{Q}\cdot\hat{c})} \frac{\xi_{ab}^2}{[1 + (\xi_{ab}|\mathbf{Q}_{ab} - \mathbf{Q}|)^2]^2}, \quad (1)$$

which represents the momentum dependence of short-range antiferromagnetic Ce^{3+} moments polarized along the [001] direction with dynamic correlation lengths of $\xi_c = 6.8 \pm 0.7$ and $\xi_{ab} = 6.3 \pm 0.5$ Å at $E = 0.5$ meV. $f(Q)^2$ is the magnetic form factor [79]. The dashed line in Fig. 2(c) is the momentum dependence expected for no preferential Ce^{3+} moment direction. A Lorentzian squared function was chosen to describe the in-plane momentum dependence as it is normalizable in two dimensions. The ratio of the dynamic correlation lengths is $\xi_{ab}/\xi_c \sim 1$, illustrating a strong three-dimensional character. The dynamic correlation length along [001] of $CeCo(In_{0.990}Hg_{0.010})_5$ is comparable to the value (6.5 ± 0.9 Å) for the low temperature resonance peak in superconducting $CeCoIn_5$ [80] illustrating that both the polarization and dynamic correlation lengths have similarities to the parent compound resonant fluctuations. The uniaxial and three-dimensional nature of the fluctuations is consistent with the Ising universality class extracted from the magnetic order parameter discussed above.

The energy dependence of the critical magnetic fluctuations of $CeCo(In_{0.990}Hg_{0.010})_5$ is shown in Fig. 3. Figure 3(a) displays a constant momentum slice at 3.5 K, illustrating that these critical fluctuations show little momentum dependence with energy transfer. Confirming this are constant energy slices at $E = 0.5$ [Fig. 3(b)] and 1.0 meV [Fig. 3(c)], which show little change in the line shape and also the ratio of the dynamic correlation lengths ξ_c/ξ_{ab} with energy transfer. The three-dimensional character of the critical correlations is robust with energy transfer and there is no observable evidence of fluctuations perpendicular to the [001] direction or transverse to the ordered low-temperature magnetic moment direction.

The temperature dependence of the critical magnetic fluctuations in $CeCo(In_{0.990}Hg_{0.010})_5$ near $\vec{Q} = (1/2, 1/2, 1/2)$ is displayed in Fig. 4, measured on IN12 and MACS. At low temperatures in the superconducting state there is no evidence of any observable magnetic fluctuations

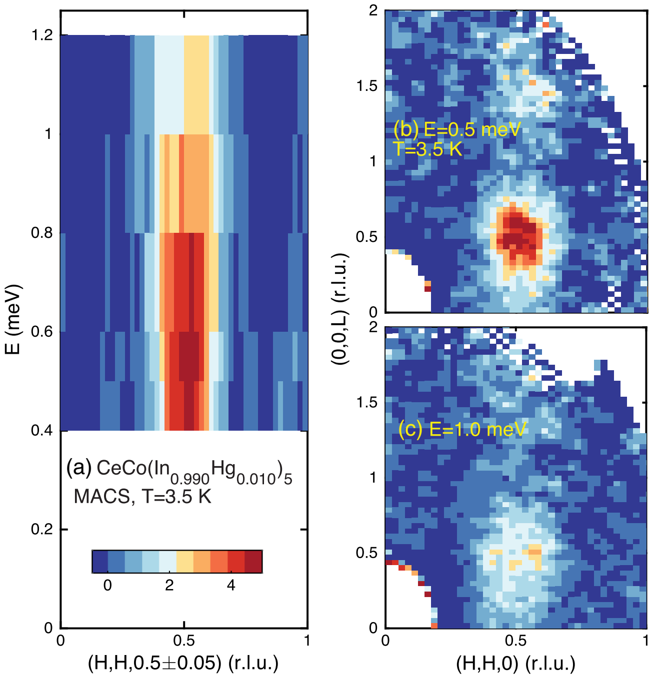

FIG. 3. (a) Constant $\vec{Q} = (H, H, 1/2)$ slice and constant energy slices at (b) 0.5 and (c) 1.0 meV of $\mathrm{CeCo(In_{0.990}Hg_{0.010})_5}$ at 3.5 K.

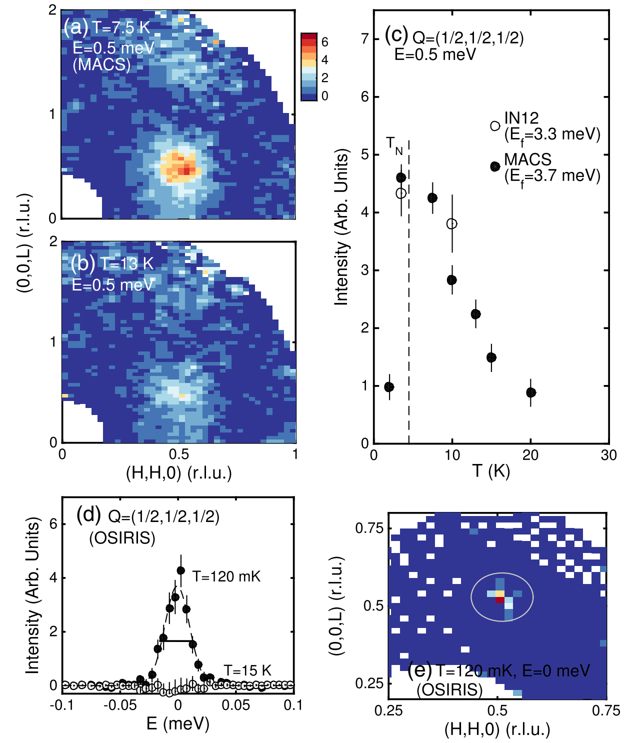

FIG. 4. The temperature dependence of the magnetic critical scattering in $\mathrm{CeCo(In_{0.990}Hg_{0.010})_5}$. (a)–(b) illustrate constant $E = 0.5$ meV slices in the (HHL) scattering plane taken at 7.5 and 13 K, respectively. (c) Intensity of the $\vec{Q} = (1/2, 1/2, 1/2)$ magnetic position as a function of temperature. (d)–(e) illustrate high resolution backscattering measurements probing the temporal nature of the magnetic order. The horizontal bar is the energy resolution showing that the magnetic order is static to within an energy resolution of 25 μeV.

confirmed by measurements on IN12 and also momentum space maps on MACS. Figures 4(a) and 4(b) show further momentum maps illustrating the presence of dynamic magnetic fluctuations above T_N. The background corrected intensity as a function of temperature is shown in Fig. 4(c) displaying a precipitous decrease in magnetic spectral weight below T_N and into the superconducting state. This transfer of spectral weight occurs while static magnetic order is formed (shown in Fig. 1) and results in temporally static order [on order of the experimental resolution of 25 μeV shown in Figs. 4(d) and 4(e)]. Any resonant excitation in $\mathrm{CeCo(In_{0.990}Hg_{0.010})_5}$ with comparable spectral weight to that of parent $\mathrm{CeCoIn_5}$ is either considerably broadened in momentum and energy, or residing within the elastic energy resolution of our measurements.

The results presented here illustrate that collinear c-axis polarized magnetic order is parent to superconductivity in the $\mathrm{CeCoIn_5}$ system. The magnetic order in $\mathrm{CeCo(In_{1-x}Hg_x)_5}$ replaces the temporally well-defined "Ising-like" [80] and longitudinally polarized [81] resonant fluctuations reported in Nd doped $\mathrm{CeCoIn_5}$ which displays both superconductivity and commensurate magnetic order. The static magnetic structure mimics the predominately [001] polarized resonant magnetic fluctuations present in strongly superconducting samples. The spectral weight of the static magnetic Bragg peak in $\mathrm{CeCo(In_{1-x}Hg_x)_5}$ is similar to $\langle\mu_{\mathrm{eff}}^2\rangle \sim 0.4\mu_B^2$ reported to reside in the resonance peak in parent $\mathrm{CeCoIn_5}$ [23]. Changing the chemical potential through hole doping (Hg) therefore shifts this dynamic spectral weight to $E = 0$ (within the resolution of the measurements discussed here) allowing little spectral weight to form a resonant excitation at finite energy, measured in parent $\mathrm{CeCoIn_5}$. This result is in agreement with the significantly reduced superconducting gap, reflected in the specific heat jump $\Delta C/\gamma T_c \sim 0.8$ in $\mathrm{CeCo(In_{0.990}Hg_{0.015})_5}$ compared to 4.5 in $\mathrm{CeCoIn_5}$ [82].

Our work on $\mathrm{CeCo(In_{1-x}Hg_x)_5}$ shows that a three-dimensional Ising phase is parent to superconductivity in $\mathrm{CeCoIn_5}$. The lack of evidence of any low energy transverse fluctuations and the presence of a commensurate low temperature structure with [001] ordered moments that mimic the polarization and momentum dependence of the resonance peak in $\mathrm{CeCoIn_5}$ supports this description of the magnetic order parameter. This indicates that transverse excitations are likely heavily damped in energy and, hence, are unstable. While transverse magnetic fluctuations have been reported in $\mathrm{CeRhIn_5}$ [73], we note these fluctuations are also unstable with a momentum and energy broadened continuum of magnetic excitations resulting from spontaneous decay, or multiparticle states [33].

Both the resonant peak in $\mathrm{CeCoIn_5}$ and the c-axis polarized three-dimensional fluctuations in $\mathrm{CeCo(In_{1-x}Hg_x)_5}$ differ from the magnetic response in other materials with coexistent magnetism and superconductivity. Isotropic short-range magnetic order is proximate to unconventional superconductivity in the cuprates [83–86], while three-dimensional order is nearby in pnictides [75,76,87]. The anisotropy is also reflected in the magnetic dynamics with a predominately c-axis polarized resonance present in $CeCoIn_5$ compared with the isotropic "spin-1" response measured in the cuprates [88,89]. The underlying anisotropy has been used to explain the magnetic field dependence of the $CeCoIn_5$ resonance peak [28,90] compared to explanations for the cuprates [91]. However, similar to the case discussed here, transverse spin waves are indeed unstable in other unconventional superconductors including iron- and copper-based high-T_c superconductors [88,92–95].

The work here on $CeCo(In_{1-x}Hg_x)_5$ contrasts with the inelastic scattering measurements on Yb doped $CeCoIn_5$ that report a robust resonance peak against doping with a dispersing momentum dependence with increasing energy transfer [96]. This observation was used to argue that the resonance peak is associated with spin fluctuations seen in the superconducting state due to a reduction in dampening, similar to magnon quasiparticles in insulating quantum magnets [97]. In our current work, we report the presence of three-dimensional c-axis magnetic order and the lack of low-energy transverse magnetic dynamics that reflects the Ising-like order. The c-axis polarized magnetic order in $CeCo(In_{1-x}Hg_x)_5$ is similar to the magnetic resonance peak in the superconducting phase of pure $CeCoIn_5$. We do not observe a strong resonance peak at $\sim$0.5 meV despite superconductivity being present, albeit subdued. This indicates that such a gapped spin excitation is not present when a superconducting order parameter is suppressed through hole doping in favor of Ising order. The resonance peak reflects the underlying itinerant response and the superconducting gap symmetry with spectral weight being drawn from competing magnetic orders.

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