


**Absence of Kondo effect in CeNiGe<sub>3</sub> revealed by coherent phonon dynamics**Longyu Wang,<sup>1,2,\*</sup> Yadong Han,<sup>1,2,\*</sup> Junhong Yu,<sup>1</sup> Fang Xu<sup>①,2</sup>, Hang Zhang,<sup>1,2</sup> Chunsheng Gong,<sup>3</sup> Hechang Lei,<sup>3,†</sup> and Jianbo Hu<sup>①,2,‡</sup><sup>1</sup>Laboratory for Shock Wave and Detonation Physics, Institute of Fluid Physics, China Academy of Engineering Physics, Mianyang 621900, China<sup>2</sup>State Key Laboratory for Environment-Friendly Energy Materials, Southwest University of Science and Technology, Mianyang 621010, China<sup>3</sup>Department of Physics and Beijing Key Laboratory of Opto-electronic Functional Materials & Micro-nano Devices, Renmin University of China, Beijing 100872, China (Received 10 July 2021; revised 14 November 2021; accepted 15 November 2021; published 29 November 2021)

Cerium-based intermetallic CeNiGe<sub>3</sub> has been generally believed to be a heavy-fermion material with Kondo behavior at low temperatures. Using femtosecond-resolved coherent phonon spectroscopy, we present a temperature-dependent dynamic investigation of the bosonic quasiparticles in CeNiGe<sub>3</sub>. Our data do not agree with the heavy-fermion expectation and instead show that the phonon stiffening from room temperature down to 5 K can be well explained by the anharmonic effect in the absence of a Kondo mechanism. Furthermore, the coherent lattice vibration located at  $\sim 7.9$  THz exhibits a mode splitting at  $T^+ \approx 105$  K, which is close to the energy scale of the first excited crystal field splitting of the Ce  $4f$  level. We argue that an orbital crossover may account for this intriguing observation.

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Investigation of cerium-based materials has been the research frontier of condensed matter physics since the strong electronic correlation in their  $4f$  electrons can give rise to various exotic ground states [1–3]. Among these correlation-driven ground states, Kondo effect-caused insulators are well documented and can be characterized by the formation of a narrow band gap (i.e., the energy scale of several tens of meV) at low temperatures with the Fermi energy level in the gap [4–6]. Such a Kondo insulator is distinct from the typical band insulators (e.g., SiO<sub>2</sub> or diamond) and the energy gap opening is the result of a collective Kondo hybridization between localized  $f$  electrons and conduction electrons [7,8]. Recently, a cerium-based ternary intermetallic CeNiGe<sub>3</sub>, has attracted much interest due to the predicted coexistence of the magnetic ordering and the Kondo effect at low temperature [9–11], a phenomenon by far only experimentally observed in some uranium compounds [12]. To confirm this expectation, on one hand, static transport characterizations (including magnetic susceptibility, electrical resistivity, and Hall effect measurements) have identified the existence of an antiferromagnetic ordering at  $T_N = 5.5$  K [9], consistent with the predicted well-organized  $4f$  magnetic moments [11]. On the other hand, the existence of Kondo behavior in CeNiGe<sub>3</sub>, which is thought to be naturally guaranteed in materials with an unstable electronic  $4f$  shell, still lacks experimental verifications.

As aforementioned, the Kondo effect in electronic correlation systems manifests as the emergence of collective hybridization and the formation of an energy gap at the coherent temperature  $T^*$ . In this respect, femtosecond-resolved coherent phonon spectroscopy is eminently suitable to verify the existence of the Kondo effect [13–18] since it provides a nonequilibrium way to study the dynamics of collective bosonic excitations in CeNiGe<sub>3</sub> whose response is directly associated with the emergence of an energy gap in the vicinity of the Fermi level [7,19]. Therefore, in this work, we report a comprehensive study of the temperature-dependent dynamics of bosonic quasiparticles in CeNiGe<sub>3</sub> utilizing coherent phonon spectroscopy. Our spectroscopic data show that the photoexcited bosonic quasiparticle dynamics is akin to the observation in typical semimetals [20,21], which can be well explained by an anharmonic phonon-phonon scattering process [22,23], and the temperature evolution of coherent phonon modes does not reveal any Kondo-like behavior. Furthermore, we have observed a peculiar coherent phonon mode splitting at  $T^+ \approx 105$  K, which is in great agreement with the energy scale of the first excited level of the crystalline electric field (CEF) [9,10], suggesting an orbital crossover around this temperature [24,25].

**II. RESULTS AND DISCUSSION**

CeNiGe<sub>3</sub> has recently been predicted as a Kondo system with the antiferromagnetic ordering at low temperature [9,10]. As shown in Fig. 1(a), CeNiGe<sub>3</sub> crystallizes in the orthorhombic SmNiGe<sub>3</sub>-type structure (*Cmmm*, No. 65) with a sequence of Ge<sub>2</sub>Ni-CeGe<sub>2</sub>Ce-NiGe<sub>2</sub> layers along the  $b$  axis [26]. In this lattice structure, Ce<sup>3+</sup> occupies a single  $4j$  site and the well-localized  $4f$  magnetic moments

\*These authors contributed equally to this work.

†hlel@ruc.edu.cn

‡jianbo.hu@caep.cn

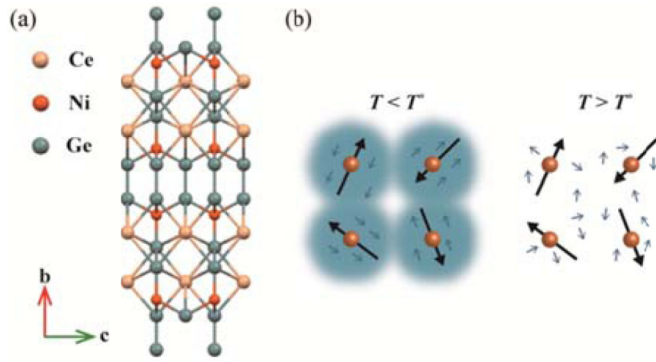


FIG. 1. Crystal structure of CeNiGe<sub>3</sub> and the Kondo effect concepts. (a) The ball-and-stick model oriented to the *bc* plane. Ce: yellow; Ni: red; Ge: green. (b) A schematic illustration of 4*f* electrons below and above the coherence temperature  $T^*$ .

permit the emergence of antiferromagnetism below  $T_N = 5.5$  K, which has been revealed by the transport measurements of the thermal, magnetic, and electrical properties [10,27,28]. While the Kondo behavior in CeNiGe<sub>3</sub>, which manifests as the collective hybridization between the localized *f* and free conduction electrons below the coherence temperature  $T^*$  [see the schematic in Fig. 1(b)] still lacks experimental verifications [7,8,29]. To confirm the existence of the Kondo effect, we have performed an ultrafast time-resolved coherent phonon spectroscopy measurements on high-quality single-crystal CeNiGe<sub>3</sub> at a center wavelength of 789 nm ( $\sim 1.57$  eV) using a Ti: sapphire femtosecond laser with a pulse width of  $\sim 20$  fs, taken from room temperature down to 5 K. More details about the experimental setup can be found in previous publications [15,30–35].

Figure 2(a) shows the measured  $\Delta R/R_0$  signals at three representative temperatures with pump polarization perpendicular to the *b* axis. Upon photoexcitation with a pump fluence of  $4.1 \mu\text{J}/\text{cm}^2$ , the transient reflectivity trace can be decomposed into an electronic part (i.e., a non-oscillatory response due to the excitation and relaxation of nonequilibrium carriers) which has been subtracted by exponential fittings [14,15,34], and a coherent oscillating part which is normally attributed to the generation of optical phonons at the center of the Brillouin zone via the displacive excitation mechanism [36] or a photoexcitation induced Raman process [37]. Please note that with such a low excitation fluence, we have estimated the temperature increase induced by laser heating is less than 2 K. The frequencies of coherent phonon modes in CeNiGe<sub>3</sub> can be clarified from the Fourier transform (FT) of the time domain signal. As shown in Figs. 2(b) and 2(c), four coherent phonon modes with the frequencies of  $\sim 3.6$ ,  $\sim 5.2$ ,  $\sim 7.9$ , and  $\sim 8.8$  THz can be identified from the temperature dependence of the corresponding FT spectrum. It is noteworthy that all these four modes can be clearly resolved in our measured temperature range, indicating that photoinduced phase transition or point-group symmetry change can be ruled out in the following observations [38,39].

To glean insights into whether there exists an energy gap induced by the Kondo effect below a critical temperature ( $T^*$ ) in CeNiGe<sub>3</sub>, we have extracted the  $T$ -dependent amplitudes and frequencies of all coherent phonon modes. As

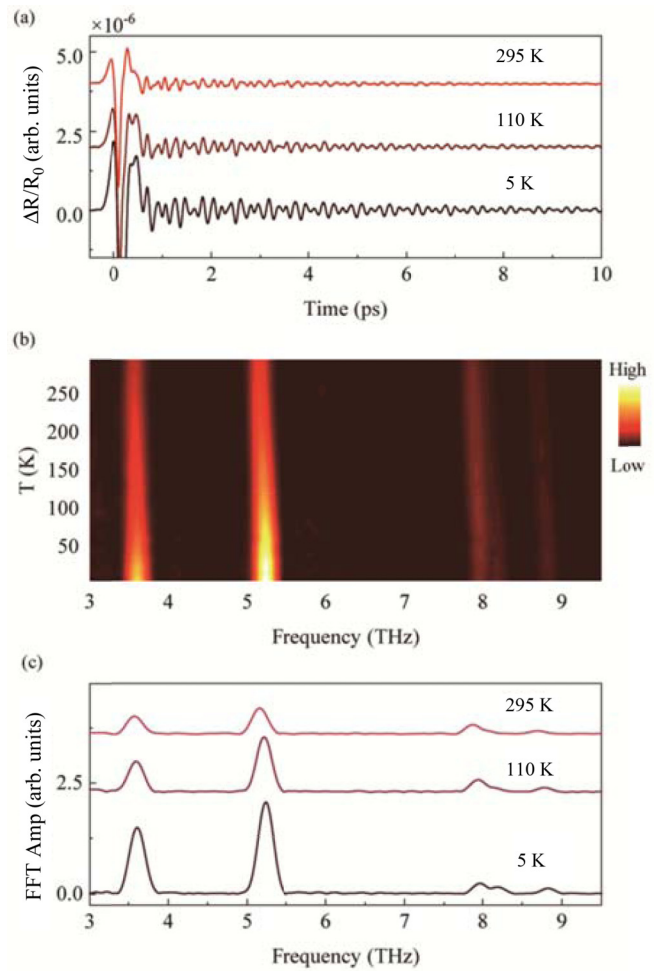


FIG. 2. Temperature-dependent coherent phonon spectra in CeNiGe<sub>3</sub> with the pump fluence of  $\sim 4.1 \mu\text{J}/\text{cm}^2$ . (a) Three representative time traces of coherent lattice oscillations at the temperature of 295, 110, and 5 K. (b) The pseudocolor photograph of frequency- and temperature-dependent photoinduced coherent phonon modes at all the measured temperatures. (c) The FT spectra at 295, 110, and 5 K.

demonstrated in various reports [7,19,40], if the collective hybridization and the consequent gap opening in the density of states (DOS) appears, a sharp downturn of mode frequency should be expected at low temperatures since the gap in the DOS constrains the electron-lattice scattering in the vicinity of the Fermi energy and as a consequence reduces the energy of coherent phonons. Such a frequency change can be related to the density of Kondo singlets ( $b_i$ ), which is proportional to the population of quasiparticles ( $N$ ) excited across the narrow gap in the form of  $[1-N(T)/N(T^*)]$  [40]. However, in our spectroscopic data, frequencies of all coherent phonon modes [see Fig. 3(a)] display a hardening trend with decreasing temperature and become saturated in the even lower temperature range, which is highly inconsistent with the Kondo effect. Indeed, this temperature-dependent phonon frequency change in CeNiGe<sub>3</sub> agrees well with the anharmonic effect observed in numerous other materials [41–44], in which the optical phonon can decay into acoustic modes of lower energy and cause a renormalization of its self-energy. To test this

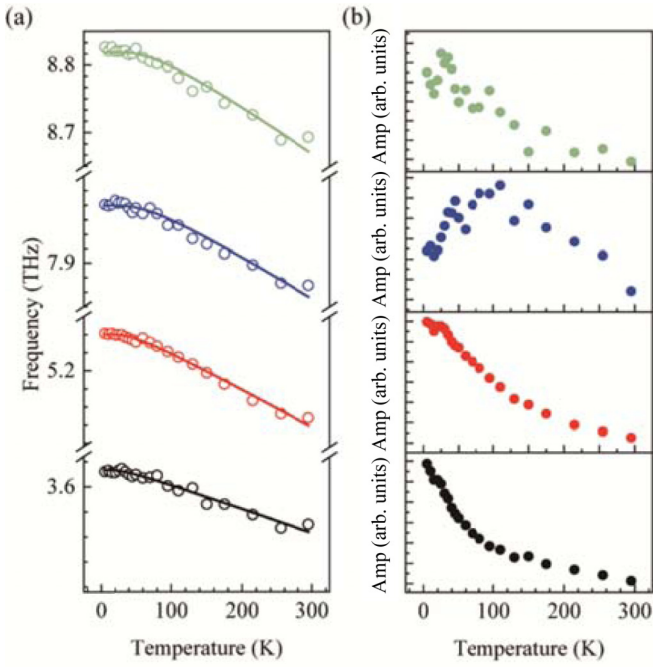


FIG. 3. Temperature-dependent mode analysis of coherent phonons in CeNiGe<sub>3</sub>. (a) The mode frequency as a function of temperature. The solid lines represent the fit using the anharmonic phonon model. (b) The temperature-dependent mode amplitude. A peculiar kind at  $\sim 105$  K for the mode located around  $\sim 7.8$  THz is resolved.

argument, we fit the frequency change using the anharmonic expression, which assumes the coherent phonon decays into two modes of frequency  $\omega_0/2$  or three modes of frequency  $\omega_0/3$  [22,23]:

$$\omega(T) = \omega_{\text{int}} + m \left[ 1 + 2n \left( \frac{\omega_{\text{int}}}{2}, T \right) \right] + n \left[ 1 + 3n \left( \frac{\omega_{\text{int}}}{3}, T \right) + 3n^2 \left( \frac{\omega_{\text{int}}}{3}, T \right) \right], \quad (1)$$

where  $n(\omega, T) = \frac{1}{\exp(\hbar\omega/k_B T) - 1}$  is the Bose-Einstein distribution,  $\omega_{\text{int}}$  is the intrinsic harmonic angular frequency, and  $m$  and  $n$  are fitting parameters.

As shown in Fig. 3(a), the quality of the fit (the solid lines) is quite satisfactory, suggesting that the anharmonic effect should account for the observed phonon energy change in CeNiGe<sub>3</sub>. It is worth mentioning that we have also estimated the size of the phonon softening caused by the Kondo effect using the density of Kondo singlets [7,19]. The calculated results reveal that fitting root mean square deviation (RMSE) for the Kondo singlet density model is as large as  $(3-9) \times 10^{-3}$  (RMSE for the anharmonic phonon model is smaller than  $4 \times 10^{-5}$ ) which rules out the possibility that phonon frequency down-turning caused by the Kondo effect may embed in the fitting uncertainties. Moreover, the monotonical increase of phonon amplitudes (except the mode at  $\sim 7.9$  THz, which will be discussed later) with decreasing temperature in Fig. 3(b) also confirms the absence of energy gap formation

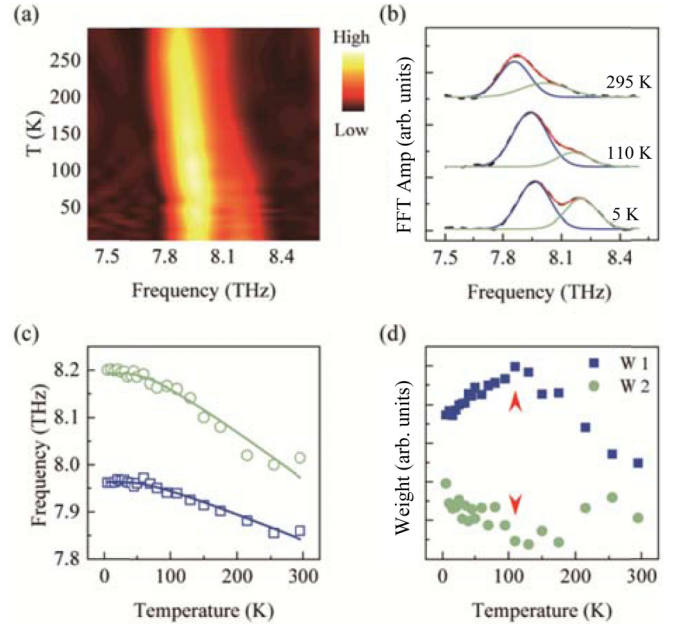


FIG. 4. The splitting behavior of the  $\sim 7.8$  THz coherent phonon mode. (a) A zoomed-in pseudocolor photograph around the frequency of 8.0 THz. (b) Gaussian line shaping fitting to the FT spectra at 295, 110, and 5 K. (c) Absence of frequency anticrossing for these two splitting modes. The solid lines represent the fit using the anharmonic phonon model. (d) The temperature-dependent amplitude of these two splitting modes.

in CeNiGe<sub>3</sub> since the temperature dependence of a coherent phonon amplitude in absorbing media is often dominated by the photoexcited carrier density and a gap opening will change the population of excited carriers [20].

Now we discuss the peculiar amplitude behavior of the coherent phonon mode at  $\sim 7.9$  THz which displays a kink at  $T^+ = \sim 105$  K. As shown in the zoomed-in pseudocolor map of temperature-dependent FT spectra [see Fig. 4(a)], a “side lobe” is gradually developed with decreasing temperature and leads to the amplitude kink observed in Fig. 3(b). Here, we exclude the Fano effect [45–47] to account for the occurrence of the coherent phonon mode splitting since two symmetric Gaussian line shapes can describe the measured phonon profile reasonably well at all temperatures, as shown in Fig. 4(b). Moreover, although  $T^+$  is close to the first excitation crystalline electric field (CEF) level in CeNiGe<sub>3</sub> (the measured energy scale is at around 100 K) [9,10], it is unlikely that the mode splitting is coming from a strong coupling between the coherent phonon mode and the crystal field excitations of the Ce<sup>3+</sup> 4*f* electrons [48–50] because such coupling should result in the frequency anticrossing (i.e., when the temperature increases, the higher mode shifts to lower frequencies while the lower peak shifts to higher frequencies), which is inconsistent with the similar frequency shifting trend observed in our measurements [Fig. 4(c)]. Therefore, we propose that the mode splitting may originate from an orbital crossover [24,25]: With increased temperature, the conduction band hybridizes with the first excitation CEF level in CeNiGe<sub>3</sub> and the

electron occupancy will be changed. As suggested by a series of experimental and theoretical works [51,52], once the CEF states become thermally occupied, electron-lattice scattering and the energy scale of lattice vibrations are affected, leading to the phonon mode splitting at  $T^+ \sim 105$  K observed in Fig. 4(d).

Please note that the magnetic ground state and a large magnetic anisotropy in CeNiGe<sub>3</sub> are assumed to be associated with the CEF splitting of the Ce Hund's rule ground state multiplet [10]. Splitting of the sixfold degeneracy of the  $J = 5/2$  multiplet of Ce<sup>3+</sup> is expected to develop a long-range magnetic order, which results in a fairly weak Kondo coupling strength between conduction carriers and the local moments of the  $f$  electrons (i.e., CeNiGe<sub>3</sub> is expected to locate at the left side of the Doniach diagram with large Ruderman Kittel Kasuya Yosida (RKKY) interactions) [25]. This may be the main reason that explains why the expected Kondo behavior is absent in our coherent phonon dynamics. More importantly, our experimental results have established that thermal energy can change the energy hybridization between the conduction band in CeNiGe<sub>3</sub> and the first excitation CEF level to form an orbital crossover and change the electronic occupation, which manifests as the phonon energy modulation. This observation suggests that future studies can take the crystal field splitting into consideration as a tuning knob for the Kondo effect in CeNiGe<sub>3</sub> since it has been demonstrated that the Kondo energy scale is highly dependent on the orientations and occupations of different CEF orbitals [25,51].

### III. CONCLUSION

In summary, by using femtosecond-resolved coherent phonon spectroscopy, we show that unlike other cerium-based intermetallics, the nonequilibrium dynamics of bosonic quasiparticles in CeNiGe<sub>3</sub> does not exhibit any features which can be associated with the emergence of collective hybridization induced by the Kondo effect at low temperature. Instead, the temperature-dependent coherent phonon behavior can be well described by the common anharmonic effect. Moreover, we have also indicated that the appearance of the crystal field effect can affect the lattice vibrations in CeNiGe<sub>3</sub>. Our results may provide a supplementary perspective on the understanding of heavy-fermion physics in cerium-based materials.

The data that support the plots of this paper are available from the corresponding authors upon reasonable request.

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The authors declare no competing financial interests.

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