Magnetic field and pressure effects on charge density wave, superconducting, and magnetic states in Lu$_5$Ir$_4$Si$_{10}$ and Er$_5$Ir$_4$Si$_{10}$

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Abstract

We have studied the charge-density-wave (CDW) state for the superconducting Lu$_5$Ir$_4$Si$_{10}$ and the antiferromagnetic Er$_5$Ir$_4$Si$_{10}$ as variables of temperature, magnetic field, and hydrostatic pressure. For Lu$_5$Ir$_4$Si$_{10}$, the application of pressure strongly suppresses the CDW phase but weakly enhances the superconducting phase. For Er$_5$Ir$_4$Si$_{10}$, the incommensurate CDW state is pressure independent and the commensurate CDW state strongly depends on the pressure, whereas the antiferromagnetic ordering is slightly depressed by applying pressure. In addition, Er$_5$Ir$_4$Si$_{10}$ shows negative magnetoresistance at low temperatures, compared with the positive magnetoresistance of Lu$_5$Ir$_4$Si$_{10}$.

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The charge density wave (CDW) ground state develops in low-dimensional materials as a consequence of electron-phonon interactions in that the electronic instabilities lead to structural modulations. The periodic charge density modulation accompanied by a periodic lattice distortion has a tendency to achieve nesting of the Fermi surface and to open an energy gap at the Fermi level. There have been many studies on a phase diagram where CDW coexists with superconductivity (SC) [1]. There have been a couple of previous studies to look for possible interplay between CDW and magnetism [2, 3]. However, the precise role of CDW with respect to SC or magnetism is unclear so far. It is worthwhile to study strongly coupled CDW systems such as Lu$_5$Ir$_4$Si$_{10}$ and Er$_5$Ir$_4$Si$_{10}$ with SC and magnetism, respectively.

Both Lu$_5$Ir$_4$Si$_{10}$ and Er$_5$Ir$_4$Si$_{10}$ compounds crystallize in the Sc$_5$Co$_4$Si$_{10}$-type (space group $P\bar{4}/mbm$) tegragonal structure [4]. The chainlike structure of Lu1/Er1 atoms along the c axis may not only form a quasi-one-dimensional electronic band but also achieve a CDW ground state. Lu$_5$Ir$_4$Si$_{10}$ shows a strongly coupled commensurate CDW state at $T_{CDW} = 83$ K, which is followed by a weakly coupled BCS-type superconducting state below $T_C = 3.9$ K [5]. It has been found that this structural transition is accompanied with a partial gapping of the Fermi surface [6] and a periodic lattice distortion with the wave vector $\vec{q} = (0, 0, 3/7)$ [5], leading to a metallic nature even in the CDW state. The superconducting transition temperature is slightly depressed by an applied pressure up to 20 kbar at a rate of $dT_C/dP \sim -1 \times 10^2$ K/kbar and suddenly jumps up into $T_C = 9.1$ K at 21.4 kbar [7]. On the other hand, Er$_5$Ir$_4$Si$_{10}$ has a transition on cooling to a 1D incommensurate CDW state at $T_{ICDW} = 155$ K and a lock-in transition at $T_{CCDW} = 55$ K [2]. Then, the well-localized Er$^{3+}$ moments are antiferromagnetically ordered below $T_N = 2.8$ K [8]. It has been suggested that the incommensurate CDW state is favored in the doubled unit cell with $\vec{q} = (0, 0, 1/4 \pm \delta)$ and then locks into a purely commensurate CDW phase at $T_{CCDW}$, whereby $\delta$ jumps to zero. The magnetic moments of the Er ions may play a definite role in these transitions. This paper reports the first observation of magnetic field and pressure effects on the CDW states in the superconducting Lu$_5$Ir$_4$Si$_{10}$ and the local-moment magnet Er$_5$Ir$_4$Si$_{10}$ single crystals. The present results suggest that there is an interplay between CDW and magnetism, which becomes important near $T_N$.

Single crystals of Lu$_5$Ir$_4$Si$_{10}$ and Er$_5$Ir$_4$Si$_{10}$ have been grown with a tri-arc furnace using a modified Czochralski technique (see Ref. [2, 3] for details). The electrical resistivity was
measured by a standard four-probe dc method using conventional 10 T and 20 T superconducting magnets at the Korea Basic Science Institute in South Korea. Magnetization measurements were performed with a Quantum Design superconducting quantum interference device (SQUID) magnetometer. Data of the specific heat were taken by a thermal relaxation method utilizing a Quantum Design physical property measurement system (PPMS). The pressure cells for the transport and magnetic measurements are of the piston-cylinder type constructed out of BeCu alloy.

Figure 1 demonstrates the presence of both CDW and SC transitions in Lu$_5$Ir$_4$Si$_{10}$. At $T_{\text{CDW}} = 83$ K, the specific heat $C(T)$ has a sharp spike, of which the size is about 160 J/mol K. This indicates the first-order nature of the CDW phase transition [5]. In the inset of Fig. 1, a peak structure observed at $T_C = 3.9$ K corresponds to the SC transition. The application of magnetic field does not change the CDW structure at all but suppresses the SC state. On the other hand, Er$_5$Ir$_4$Si$_{10}$ displays multiple CDW phases and magnetic ordering. Figure 2 shows the drastic changes in $C(T)$ at these transitions. With decreasing temperature, the normal state changes into an incommensurate CDW (ICDW) state at $T_{\text{ICDW}} = 155$ K, where $C(T)$ has a sharp peak. It has been reported to be characteristic of the second-order nature of the ICDW transition [2]. This ICDW state develops into a commensurate CDW (CCDW) state at lower temperature $T_{\text{CCDW}} = 55$ K, where $C(T)$ has a broad shoulder. With further cooling around $T_N = 2.9$ K, there is a peak in the inset of Fig. 2 because the antiferromagnetic ground state occurs. When the magnetic field is applied, both $T_{\text{ICDW}}$ and $T_{\text{CCDW}}$ are unchanged, while $T_N$ is smeared out with magnetic field. These data provide that the magnetic field effect is driven with no change in the electronic instabilities but strong change in the superconductivity and the magnetic ordering.

The anisotropic magnetism of Er$_5$Ir$_4$Si$_{10}$ is apparent in Fig. 3. The magnetic susceptibility $\chi(T)$ has been measured in a field of 100 G along the $a$ and $c$ axes. At high temperatures above 50 K, both $\chi_a$ and $\chi_c$ are well fitted by the Curie-Weiss law, $\chi = C/(T - \theta_P)$. From this fit, we obtain the effective magnetic moments of $\mu_a = 9.58(3)\mu_B$ and $\mu_c = 9.59(3)\mu_B$ and the paramagnetic Curie temperatures of $\theta_a = -6.76(0)$ K and $\theta_c = 3.50(5)$ K for $H \parallel a$ and $H \parallel c$, respectively. The values of $\mu_{\text{eff}}$ indicate that the Er ion is in the normal trivalent state in Er$_5$Ir$_4$Si$_{10}$. The value of $(2\theta_a - \theta_c)/3 = -3.34$ K is in qualitative agreement with the previous value of $\theta_P = -1.98$ K for a polycrystalline sample [6], implying a weak antiferromagnetic-type correlation between the Er$^{3+}$ moments. At low temperatures below
50 K, the small deviation from the Curie-Weiss law is attributed to the crystal-field effect. Since the difference between $\theta_a$ and $\theta_c$ is proportional to the tetragonal crystal-field parameter $B_2^0$, we obtain $B_2^0 = -0.13(6)$ K using $\theta_a - \theta_c = -10.26(5)$ K. With further cooling, there appears a significant difference that $\chi_a$ are much smaller than $\chi_c$, which reveals only the antiferromagnetic phase transition at $T_N = 2.9$ K.

In order to examine the pressure effect on the magnetism of Er$_5$Ir$_4$Si$_{10}$, we have measured $\chi(T)$ for $H \parallel c$ at constant pressure. As is seen in Fig. 4, the high-temperature data at 9 kbar satisfy the Curie-Weiss law with $\mu_{\text{eff}} = 9.52(3) \mu_B$ and $\theta_P = 1.33(9)$ K. This yields that the valence of Er ion is pressure independent and the magnetic correlation slightly depends on pressure. The most remarkable effect of pressure on the magnetism is illustrated in the inset of Fig. 4. The application of pressure strongly depresses the low-temperature moments and thus the magnetic transition becomes very sluggish. One could anticipate that at higher pressure beyond our experimental range here the magnetic ordering is suppressed completely, resulting in free spin paramagnetism of Er ions.

We now present the pressure and magnetic field effects on the transport of Lu$_5$Ir$_4$Si$_{10}$. The pressure was fixed at room temperature and the data of electrical resistivity $\rho(T)$ have been taken at constant magnetic field as the sample was slowly warming up. The pressure dependence of $\rho(T)$ for Lu$_5$Ir$_4$Si$_{10}$ as a function of temperature at different magnetic fields is shown in Fig. 5. At ambient pressure, $\rho(T)$ shows a sharp upward jump at $T_{\text{CDW}} = 81$ K. This is well understood by considering the decrease in area of the Fermi surface as a result of the opening of an energy gap. The CDW transition occurs near 68 K at $P = 9$ kbar. As the pressure is increased, one can expect a band broadening due to pressure-promoted intralayer/interlayer coupling and hence the CDW state becomes unstable and $T_{\text{CDW}}$ is lowered with pressure. The monotonic depression of $T_{\text{CDW}}$ is consistent with the general trend in conventional CDW materials such as layered transition-metal dichalcogenides and NbSe$_3$. Since the Fermi surface is sharper at low temperature, the CDW transition is rather sensitive under pressure. The striking feature in $\rho(T)$ is the metallic behavior even in the CDW state, indicating that the energy gap is not completely opened, i.e., there is a definite electronic density of states within the gap. We also observe a slight increase of $T_C$ with pressure. This pressure effect can be ascribed to the enhancement of the density of states at the Fermi level resulting from the decrease of the gap. In addition, $\rho(T)$ has been studied in the nonmagnetic BeCu cell at various magnetic fields. In a series of temperature
cycles at different pressures, the CDW transition is not changed, while the SC transition is strongly suppressed. We note that the application of magnetic field slightly enhances the resistance at low temperatures. This positive magnetoresistance is more pronounced under pressure than at ambient pressure. These results could be attributable to the semimetallic CDW character of Lu$_5$Ir$_4$Si$_{10}$.

In contrast to the monotonic depression of $T_{CDW}$ for Lu$_5$Ir$_4$Si$_{10}$ with pressure, the application of pressure in Er$_5$Ir$_4$Si$_{10}$ has complex effects on the CDW phase transitions. In Fig. 6, $\rho(T)$ at ambient pressure displays an abrupt increase at $T_{ICDW} = 150$ K and a sharp drop at $T_{CCDW} = 55$ K in zero field. The former anomaly at $T_{ICDW}$ can be understood in a way similar to the sharp upward jump at $T_{CDW}$ in Lu$_5$Ir$_4$Si$_{10}$. The latter anomaly at $T_{CCDW}$ may reflect another mechanism that the commensurate modulation destroys the perfect nesting of the Fermi surface, leading to the gain of a portion of the Fermi surface. Consequently, the density of states at the Fermi level is enhanced and thus $\rho(T)$ drops sharply at $T_{CCDW}$. The influence of pressure on the phase transitions is evident in Fig. 6. The ICDW phase transition is independent of the pressure within our experimental error in temperature of $\pm 0.1$ K. This result is contrary to the CDW state for Lu$_5$Ir$_4$Si$_{10}$, whose transition temperature strongly depends on pressure. The anomaly associated with the CCDW phase is smeared out under pressure. These pressure effects on the ICDW and CCDW transitions may reflect that the incommensurate-commensurate transition at $T_{CCDW}$ depends more critically on the band structure of Er$_5$Ir$_4$Si$_{10}$ than the normal-incommensurate transition at $T_{ICDW}$ and there is possible interplay between CDW and magnetism in this compound. Further crucial experiments such as low-temperature single-crystal electron or neutron diffraction measurements should be performed to provide the evidence for the strongly coupled CDW interplaying with the magnetic ordering of Er$_5$Ir$_4$Si$_{10}$. By an applied magnetic field, both $T_{ICDW}$ and $T_{CCDW}$ remain essentially unchanged, while $T_N$ is strongly depressed. In addition, the magnetoresistance of Er$_5$Ir$_4$Si$_{10}$ is negative at low temperatures. This is compared with the positive magnetoresistance observed in Lu$_5$Ir$_4$Si$_{10}$.

In conclusion, we have determined the transition temperatures from the normal to incommensurate phase at $T_{ICDW}$ and from the incommensurate to commensurate phase at $T_{CCDW}$ as functions of hydrostatic pressure and magnetic field. We find that for Er$_5$Ir$_4$Si$_{10}$ $T_{CCDW}$ is suppressed rapidly with pressure, while $T_{ICDW}$ remains constant. It is worthwhile to mention that we cannot rule out the complex transport mechanism, in which the CDW order
parameters have different pressure dependence. In addition, the application of magnetic field depresses the resistivity, leading to a negative magnetoresistance at low temperatures. These observations imply that conduction electrons couples strongly with phonons involved in the CDW transition and f electrons with the local-moment magnetism. Comparison of these results with Lu$_5$Ir$_4$Si$_{10}$ suggests that the suppression of $T_{\text{CDW}}$ by pressure is not a necessary condition for the strongly coupled CDW systems with magnetic ordering. It would not be so surprising for one to see the CDW phase interplaying with the magnetism, because of its electronic instability due to magnetic correlation. More information is needed concerning the exact mechanism in terms of the strong electron-phonon interaction as well as the critical coupling between the $f$ and conduction electrons.

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FIG. 1: Specific heat $C(T)$ of Lu$_5$Ir$_4$Si$_{10}$, which displays a CDW transition at $T_{CDW} = 83$ K. The inset shows a SC transition at $T_C = 3.9$ K. The solid circles indicate zero-field data and the open circles indicate in-field (9 T) data.

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FIG. 2: Specific heat $C(T)$ of $\text{Er}_5\text{Ir}_4\text{Si}_{10}$, which undergoes two CDW transitions at $T_{\text{ICDW}} = 155$ K and $T_{\text{CCDW}} = 55$ K. The low temperature data is shown in the inset, where an antiferromagnetic transition occurs at $T_N = 2.9$ K. The solid circles indicate zero-field data and the open circles indicate in-field (9 T) data.
FIG. 3: Magnetic susceptibility $M/B$ and its inverse $B/M$ for the single crystals of $\text{Er}_5\text{Ir}_4\text{Si}_{10}$ at ambient pressure in a field of 100 G applied magnetic field along the $a$ and $c$ axis. The insets show the low temperature parts.
FIG. 4: Magnetic susceptibility $M/B$ and its inverse $B/M$ for the single crystals of $\text{Er}_5\text{Ir}_4\text{Si}_{10}$ measured at 9 kbar for a field along the $c$ axis. The low temperature data at ambient pressure are compared with those at 9 kbar in the inset.
FIG. 5: Temperature dependence of electrical resistivity $\rho(T)$ of Lu$_5$Ir$_4$Si$_{10}$ in various magnetic fields 0, 10, and 18 T. The data are taken in constant pressures 1 bar and 9 kbar at room temperature.
FIG. 6: Temperature dependence of electrical resistivity $\rho(T)$ of $\text{Er}_5\text{Ir}_4\text{Si}_{10}$ at several magnetic fields 0 and 8 T. The data are taken in constant pressures 1 bar and 8 kbar at room temperature.