Magnetism induced by interlayer electrons in the quasi-two-dimensional electride Y_2C: Inelastic neutron scattering study

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Magnetic excitations in layered electride Y_2C have been found by inelastic neutron scattering. We have observed weak but clear magnetic scattering around the wave number $Q = 0$, but no magnetic order down to the lowest temperature measured (7 K). The imaginary part of the dynamical susceptibility deduced is well described by the Lorentz function of energy E for each momentum Q. The width $\Gamma(Q)$ of the Lorentzian is proportional to $Q(Q^2 + \kappa^2)$ with $\kappa^{-1} \sim 4$ Å at $T = 7$ K. We have also found that with increasing Q the magnetic form factor decays faster than that of a $4d$ electron in a single Y atom, which indicates a more extended magnetic moment in Y_2C. These results provide experimental evidence that the itinerant magnetism in Y_2C originates from the anionic electrons that reside in the interlayers. The Curie-Weiss-like behavior of the magnetic susceptibility reported in Y_2C is ascribed to the mode coupling effects of spin fluctuations.

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I. INTRODUCTION

The electride is a special type of ionic crystal in which electrons serve as anions [1,2]. In contrast to nearly free electrons in a metal, the anionic electrons in electrides are loosely bound at the interstitial spaces, which leads to the unique physical properties of electrides. For example, electron-doped $12CaO \cdot 7Al_2O_3$, the first inorganic electride, has a significantly low work function comparable with that of alkali metals, while it is stable in air [3,4]. The low work function means that electrides provide a fertile field of applications, such as electron injection layers in organic light-emitting diodes [5] and catalysts for ammonia synthesis [6,7].

Subsequently, Ca_2N and Y_2C with layered structures are reported to be quasi-two-dimensional electrides [8–11]. As shown in Fig. 1(a), Y_2C as well as Ca_2N exhibit the anti-$CdCl_2$ type structure with the space group $R\bar{3}m$. The standard valence states of Y and C are Y^{3+} and C^{4-}, respectively; therefore, there are two excess electrons per formula unit that reside between the cationic layers of $[Y_2C]^{2+}$ with the charge neutrality preserved. Density functional theory (DFT) calculations have predicted that these excess electrons in Y_2C are confined to certain interstitial sites as anionic electrons, and that the anionic s-like orbitals hybridize with the Y $4d$ orbitals to form "electride bands" at the Fermi level [9–12]. This prediction has partly been confirmed by transport measurements of a single crystal [13]. Angle-resolved photoemission spectroscopy (ARPES) has recently provided more direct evidence of the electride bands in Y_2C [14]. These studies have established that Y_2C is a semimetal with multiple sheets of quasi-two-dimensional Fermi surfaces, and the anionic electrons are mostly confined in the interlayer space. It has been argued that hybridization between anionic electrons and Y $4d$ electrons may be one of the key ingredients to realizing topologically nontrivial phases in quasi-two-dimensional electrides [12,15,16].

Magnetism arising from the anionic electrons has been a fundamental issue since the discovery of organic electrides in the 1980s [2]. Among the reported electrides, Y_2C exhibits unusual magnetic features. According to DFT band calculations [12,17], Y_2C is a weak itinerant ferromagnet and the magnetic moment resides mostly at interstitial sites. The ferromagnetic moment is estimated to be 0.3–0.4 μ_B per site. However, the magnetic susceptibility and effective magnetic moment of Y_2C are highly sample dependent, although all measurements agree that magnetic order in Y_2C does not occur down to 2 K [10,13,18,19]. For example, the magnetic susceptibility $\chi(T)$ of polycrystalline Y_2C has Curie-Weiss-like temperature dependence, and the effective moment is estimated to be 0.564–0.604 μ_B/Y [10,18]. In single crystals, the estimated moments vary from 0.124 μ_B/Y [19] to 0.0427 μ_B/Y [18]. Moreover, the magnetization and resistivity in certain single crystals have highly anisotropic behavior

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[13]; magnetic moments are estimated to be 2.82 μ_B ($H \parallel c$) and 1.08 μ_B ($H \perp c$). All of the estimated Weiss temperatures in both polycrystalline and single crystal Y_2C are negative, as is the case with antiferromagnets [10,13,18,19].

The muon spin rotation/relaxation (μSR) technique was used by Hiraishi *et al*. recently to show that the Curie-Weiss-like behavior of $\chi(T)$ is not due to magnetic impurities, but is intrinsic to the electronic state of the polycrystalline sample [18]. This study motivated us to observe the magnetic fluctuations directly by neutron scattering, and identify the origin of the magnetic moments.

We first remark that the Curie-Weiss-like behavior of $\chi(T)$ does not necessarily imply the presence of localized magnetic moments. It is known that the mode coupling of spin fluctuations in itinerant electrons also causes Curie-Weiss-like behavior [20–22]. However, the apparent effective moment in the latter case, which is derived from the Curie constant, can be much larger than the ordered moment. The spin fluctuation theory has successfully explained characteristic behaviors of itinerant weak ferromagnets, such as $ZrZn_2$ and Sc_3In [23,24].

In this paper, we report results of inelastic neutron scattering for Y_2C using a polycrystalline sample. We provide clear experimental evidence for ferromagnetic spin fluctuations that are well fitted by the conventional form to describe nearly ferromagnetic metals. Inspection of the magnetic form factor in Y_2C indicates that the magnetism in Y_2C does not originate from Y 4*d* electrons, but from the anionic electrons. On the other hand, no signal that could imply the presence of antiferromagnetic correlations has been identified. Therefore, we suggest that the negative Weiss temperature at high temperature has its origin in the mode-coupling effect, rather than in antiferromagnetic correlations.

II. EXPERIMENTS

A polycrystalline sample of Y_2C was synthesized by an arc melting method. Details of the crystal growth and characterization of the sample are described in Refs. [10,18]. A 2.2 g polycrystalline ingot that came from the identical batch of that used in the previous μSR study [18] was clamped to an Al plate and sealed in an Al sample cell with He gas. The inelastic neutron scattering measurement was performed by the time-of-flight method using a near-backscattering spectrometer with high energy resolution (DNA, installed at BL02 in the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) [25]. To survey the range of energy transfer up to 1 meV while maintaining the high energy resolution ($\Delta E \sim 3.6$ μeV) and high signal-to-noise ratio (ca. 10^5) of DNA, five phases of a pulse shaping chopper were employed [25]. The time-of-flight data were converted into a $S(Q, E)$ map and analyzed using the Utsusemi software [26]. The intensities were normalized to absolute units with incoherent scatterings after background subtractions [27].

III. RESULTS AND DISCUSSION

Figure 1(b) shows the neutron scattering intensities of polycrystalline Y_2C against momentum (Q) and energy (E) transfers. Weak but clear scattering around $Q \sim 0$ is observed

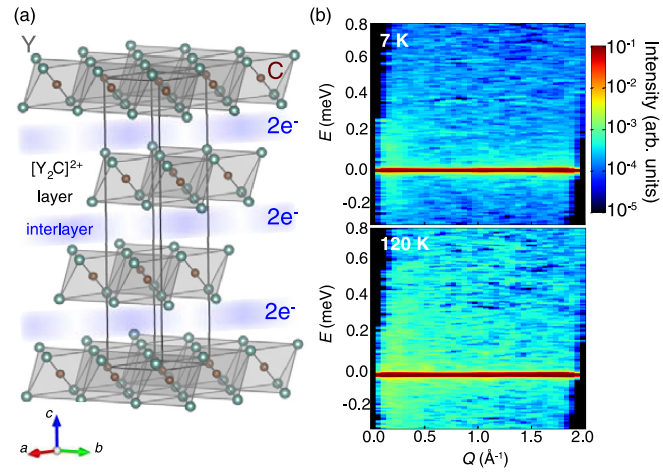

FIG. 1. (a) Crystal structure of Y_2C and schematics of an anionic electron layer. Solid lines indicate the conventional hexagonal unit cell. (b) Neutron scattering spectra of polycrystalline Y_2C at 7 and 120 K against momentum (Q) and energy (E) transfers. The color gauge shows the strength of intensity in a logarithmic scale.

at both 7 and 120 K. The intensities of these scatterings are three orders of magnitude smaller than those of elastic scatterings, and are dependent on the temperature. The signals from acoustic phonons around the 003 Bragg point ($Q = 1.05$ Å^{-1}) are not observed; the intensity from phonon scatterings is proportional to Q^2, and may be negligible in the region of $Q \sim 1$ Å^{-1}. Therefore, these weak scatterings are considered to have a magnetic origin. On the other hand, no magnetic order was observed down to 7 K, which is consistent with the previous studies [10,18]. These results indicate that the observed scattering is from spin fluctuations rather than spin waves in the ferromagnet.

The scattering function $S(Q, E)$ is related to the imaginary part of the dynamical susceptibility $\chi''(Q, E)$ by the fluctuation-dissipation theorem: $S(Q, E) = [2/(\pi g^2 \mu_B^2)]$ $\chi''(Q, E)[n(E) + 1]$, where $n(E)$ is the Bose-Einstein distribution function and g is the g factor, which is taken to be 2 because of the small spin-orbit coupling [27]. For the paramagnetic state of itinerant electrons, $\chi''(Q, E)$ with small Q and E can be described by [23,24,28]

$$\chi''(Q, E) = \chi(Q, 0)\frac{E\Gamma_Q}{\Gamma_Q^2 + E^2}, \tag{1}$$

where Γ_Q is the relaxation rate, or the inverse lifetime of spin fluctuation with Q, and $\chi(Q, 0)$ is the Q-dependent static susceptibility. The latter is parametrized as

$$\chi(Q, 0) = \chi\frac{\kappa^2}{\kappa^2 + Q^2}, \tag{2}$$

where χ and κ are the (homogeneous) static susceptibility and the inverse correlation length, respectively.

If the magnetic moment arises mostly from spin under the weak spin-orbit interaction, then the orbital anisotropy of the anionic electrons may not appear in the magnetic susceptibility. It has indeed been reported that $\chi(T)$ in a single crystal shows only a weak anisotropy [19,29]. We add that an opposite result has also been reported [13] that contradict these

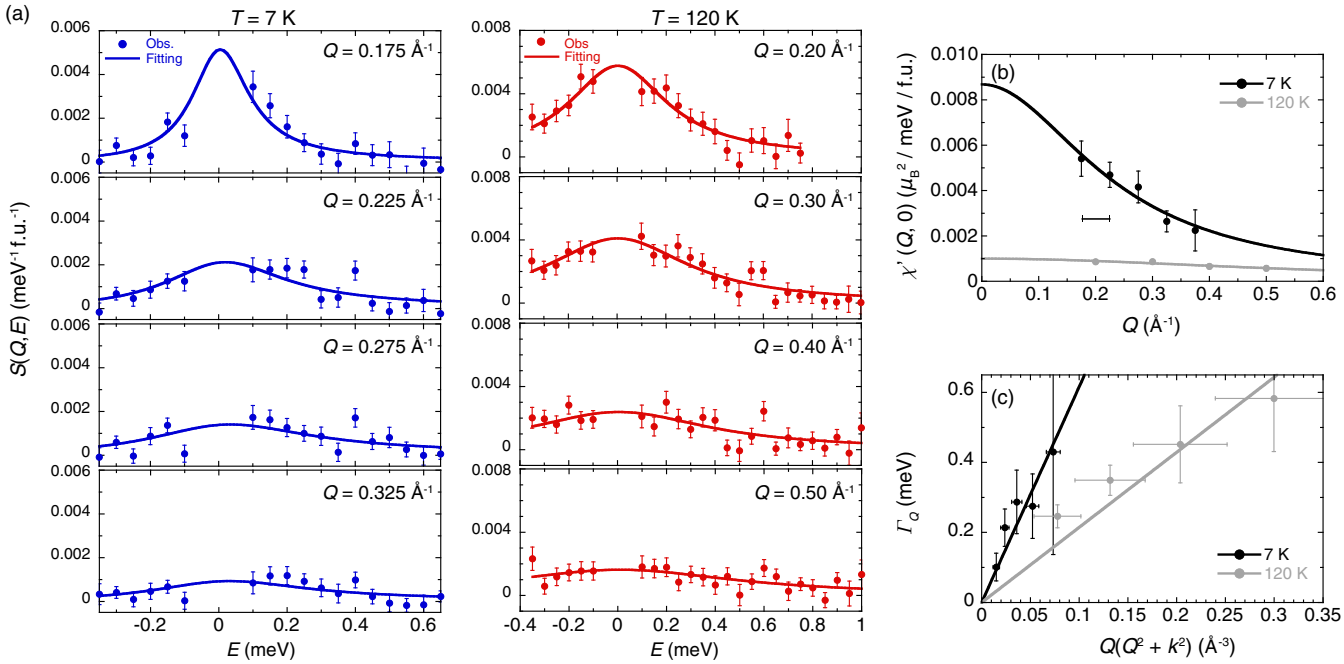

FIG. 2. (a) E dependence of $S(Q, E)$ at 7 and 120 K. The data shown for 7 (120) K are the average of those over ± 0.025 (0.05) Å^{-1} around each value of Q indicated. The elastic components between $E \pm 0.05$ meV are masked. Note that the instrumental resolution does not affect the observed excitation because of the high energy resolution ($\Delta E \sim 3.6$ μeV). The data of $Q = 1.525(1.70)$ Å^{-1} at 7 (120) K are used for the respective backgrounds. (b) Q dependence of $\chi(Q, 0)$ at 7 and 120 K. The horizontal bar indicates the Q resolution ($\Delta Q \sim 0.05$ Å^{-1}). (c) Γ_Q as a function of $Q(Q^2 + \kappa^2)$. The solid curves in (a), (b), and (c) are the results of fitting to Eqs. (1), (2), and (3), respectively.

results. In the present analysis for a polycrystalline sample, we do not include the possible effects of anisotropy in the magnetic response.

Figure 2(a) shows the E dependence of $S(Q, E)$ for different values of Q at 7 and 120 K. The data are well described by Eq. (1), and $\chi(Q, 0)$ and Γ_Q are deduced. As shown in Fig. 2(b), the $\chi(Q, 0)$ so obtained are well reproduced by Eq. (2). This fitting procedure gives a spatial correlation length $l = 2\pi/\kappa \sim 26(\sim 10)$ Å at 7 (120) K. It is reasonable that the spatial correlation length at 7 K is longer than that of 120 K. It should be noted that the presence of spatial magnetic correlation demonstrates the bulk response intrinsic to Y_2C, rather than being due to magnetic impurities. This conclusion is consistent with the μSR study that claims a bulk response [18].

In nearly ferromagnetic metal, where spin fluctuations with small $|Q|$ and E are dominant, Γ_Q is fitted by

$$\Gamma_Q = \Gamma_0 |Q|(Q^2 + \kappa^2). \tag{3}$$

The values of κ derived by the fitting are used to plot Γ_Q against $Q(Q^2 + \kappa^2)$ [Fig. 2(c)]. The linearity is satisfactory, especially at $T = 7$ K. On the other hand, in the case of localized moment (Heisenberg) systems, Γ_Q in the paramagnetic phase is often proportional to $Q^2(Q^2 + \kappa^2)$ [30], which represents the spin diffusion. As shown in the Supplemental Material [31], the proportionality of Γ_Q to $Q(Q^2 + \kappa^2)$ is determined to be more plausible than that to $Q^2(Q^2 + \kappa^2)$.

Note that the different behaviors of Γ_Q in itinerant and localized systems do not always apply, i.e., in the limit of small $|Q|$, the Γ_Q in itinerant systems should cross over to the diffusive relaxation $\Gamma_Q \propto Q^2$ [23], whereas in spin systems with strong quantum fluctuations, $\chi''(Q, E)$ may deviate substantially from the Lorentzian. Even with these restrictions, the linear Q-dependence in the dominant region can be a convenient characterization of itinerant magnetism. According to the form factor inspected, as shown next, the magnetization density in Y_2C should be highly delocalized. Therefore, the dominance of $|Q|(Q^2 + \kappa^2)$-type relaxation is in accordance with the spatially extended magnetic moment in Y_2C.

Thus far, we have approximated the magnetic form factor $f(Q)$ by unity in obtaining $S(Q, E)$ and $\chi''(Q, E)$ from the scattering intensity measured. The approximation is justified, provided that the relevant range of Q^2 ($\sim \kappa^2$) for the ferromagnetic scattering is much smaller than the characteristic value Q_c^2 in $|f(Q)|^2$. As discussed later, $|Q_c|^2 \sim 9\kappa^2$ is obtained at $T = 7$ K. Therefore, the approximation is reasonable in view of the fairly scattered data points.

In addition to these dominant contributions around $Q = 0$, extremely small signals of ferromagnetic fluctuation have also been observed around the 003 Bragg point ($Q = \tau_{003} \sim 1.05$ Å^{-1}), as detailed in the Supplemental Material [31]. The lattice periodicity demands that the magnetic scattering around each Bragg point $\boldsymbol{\tau}$, give the same scattering function, $S(\boldsymbol{Q} - \boldsymbol{\tau}, E)$. By comparing the magnitudes of raw scattering intensities around $\boldsymbol{Q} = \boldsymbol{\tau}$ and $\boldsymbol{Q} = 0$, $|f(\boldsymbol{\tau})|^2$ can be estimated within the approximation $|f(\boldsymbol{Q})|^2 \sim |f(\boldsymbol{\tau})|^2$ for $\boldsymbol{Q} \sim \boldsymbol{\tau}$. Here we assume that other contributions, such as those from phonons, are negligible.

Figure 3 illustrates $|f(Q)|^2$ around $Q = \tau_{003} \simeq 1.05$ Å^{-1} at 7 K, deduced with use of the parameters listed in Table I. Shown together are the $|f(Q)|^2$ values expected for $4d$ electrons in the Y atom [32], and for anionic electrons predicted

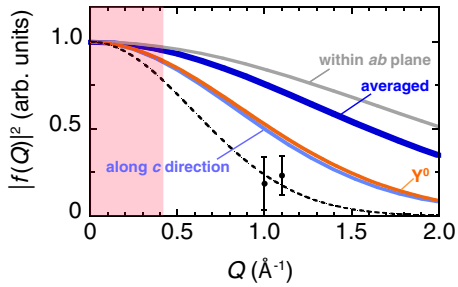

FIG. 3. Estimate of the squared magnetic form factor $|f(Q)|^2$, determined from the relative magnitude of the magnetic scattering intensity around $Q = \tau_{003} \simeq 1.05$ Å^{-1} to that around $Q \simeq 0$. For comparison, squared form factors in the following four models are shown; the orange curve represents the magnetization distribution of a 4*d* electron in a single Y atom, gray and light blue curves represent the calculated magnetization distribution of Y_2C within the *ab* plane and along the *c* direction, respectively, and the blue curve represents the angle average of magnetization distribution [17]. The dotted line is given as a guide.

by band calculations [17]. The red hatched region in Fig. 3 corresponds to those Q for the dominant magnetic scattering. It is concluded that the magnetic form factor in Y_2C decays faster than that of a 4*d* electron in a single Y atom, and that derived by band calculation. Therefore, the magnetization density in Y_2C is highly delocalized. Note that the half width at half maximum, Q_c, in the $|f(Q)|^2$ determined is $\sim 0.7 \sim 3\kappa$. Therefore, the approximation used for the scatterings around $Q = 0$ is justified. The electride bands in Y_2C consist of hybridized anionic *s*-like orbitals and Y 4*d* orbitals that have been derived by DFT calculations [10,12,17], and have been observed by the ARPES experiment [14]. Furthermore, band calculations have suggested that the magnetic moments of the Y 4*d* electrons contribute to only 7% of the total magnetization in Y_2C [17]. The results shown in Fig. 3 would provide direct experimental evidence that the magnetism in Y_2C does not originate from Y 4*d* electrons, but the anionic electrons in the interlayers.

Let us now turn to the origin of the Curie-Weiss-like behavior of the magnetic susceptibility. As we have discussed, the magnetic moments are carried by anionic electrons with substantial spacial extension; therefore, it is natural to invoke the mode-coupling effect of spin fluctuations as the origin, as described by the self-consistent-renormalization (SCR) theory [20–22]. The Curie-Weiss-like behavior comes from the temperature dependence of the variance $\langle \boldsymbol{M}_L(T)^2 \rangle$ in the local magnetization at each site. Through the fluctuation-dissipation theorem, $\langle \boldsymbol{M}_L(T)^2 \rangle$ is given by weighted integrals of $\chi''(Q, E)$ over Q and E. On the other hand, the homogeneous susceptibility $\chi(T)$ is related to the Helmholtz free energy, $F(M, T)$ via $1/\chi \propto \partial^2 F/\partial M^2$. It is important that $F(M, T)$ depends on $\langle \boldsymbol{M}_L(T)^2 \rangle$ with inclusion of spin fluctuations. Therefore, $\chi(T)$ is determined self-consistently together with $\chi''(Q, E)$, which is dominated by a small Q and E, so that the self-consistency takes into account the mode-coupling effect.

TABLE I. Parameters obtained by fitting.

T (K)	χ (μ_B^2/meV/f.u.)	κ (Å^{-1})	Γ_0 (meV Å^3)
7	0.0087(15)	0.24(4)	6.2(5)
120	0.0010(1)	0.6(1)	2.1(2)

In the case where the parameters of the system do not reach the so-called Stoner condition for ferromagnetism, the ground state remains paramagnetic. In such a case, being close to the ferromagnetic ground state, $1/\chi(T)$ in the SCR theory increases almost linearly with T at high temperature, which appears like the Curie-Weiss law with a negative value for the apparent Weiss temperature [21]. This theoretical consequence is in approximate agreement with the reported property of Y_2C, especially in polycrystalline samples that include the negative Weiss temperature.

However, the observed $1/\chi(T)$ with a decrease in temperature shows a downward deviation from the linear behavior [10,13] in contrast with a upward deviation predicted by the SCR theory [21,23]. We tentatively ascribe this peculiar feature to the singular structure in the density of states around the Fermi level, which comes from the quasi-two-dimensional semimetallic band structure of Y_2C [12,17]. With such a singular density of states, the magnetic properties should be sensitively dependent on the location of the Fermi level, which may also explain the conspicuous sample dependence in Y_2C.

Anionic electrons and their magnetic order have been extensively investigated in other systems, such as zeolites incorporated with alkali clusters [33,34] or alkali metals under applied pressure [35]. However, to the best of our knowledge, magnetic fluctuations or excitations due to these anionic electrons have not been reported. This may be due to the small scattering intensities from the anionic electrons. The present results demonstrate that state-of-the-art equipment for neutron scattering measurements has enabled observation of such low intensity signals of magnetic excitation from anionic electrons.

IV. SUMMARY

In summary, inelastic neutron scattering experiments were performed to investigate the unusual magnetism in polycrystalline Y_2C. Ferromagnetic spin fluctuations were successfully observed over a wide temperature range. Analysis of the dynamical susceptibility and the magnetic form factor demonstrated that Y_2C is a nearly ferromagnetic metal, and the magnetism in Y_2C originates from anionic electrons in the interstitial sites, rather than from Y 4*d* electrons. The Curie-Weiss-like behavior of the magnetic susceptibility is ascribed to the mode coupling effect as described by the SCR theory.

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[31] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.102.224406, where we show (i) fitting results using a model function of a critical scattering in the Heisenberg system, (ii) spectrum around the 003 Bragg point and an estimation of magnetic form factor, and (iii) strength of local fluctuations in Y_2C.
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