Unusual Nature of Ferromagnetism Coexisting with Superconductivity in UGe_2

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We report the discovery of a jump in the magnetization of a macroscopic single crystalline sample of UGe_2 that shows coexistence of ferromagnetism and superconductivity. In particular, we observe that the jump occurs at regular intervals of field and only at very low temperatures. This novel feature implies that the magnetic field induces a sudden change of the direction of the magnetization between two equivalent easy axes of magnetization even in a macroscopic sample. We ascribe it to a field-tuned resonant tunneling between quantum spin states, and we propose that the size of a magnetic domain is smaller than a superconducting coherence length.

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UGe_2 is a ferromagnet with a Curie temperature T_{Curie} of about 53 K at ambient pressure. When a pressure P is applied, T_{Curie} shows monotonous decrease with increasing P and seems to vanish at around 16 kbar. Recently Saxena *et al.* discovered superconductivity (SC) that emerges under high pressures between about 10 and 16 kbar [1,2]; therefore it seems that the superconductivity coexists with the ferromagnetism (FM) [3]. In the case of a heavy fermion superconductor coexisting with antiferromagnetism, such as UPd_2Al_3 [4], internal fields due to the antiferromagnetic ordering that superconducting Cooper paired electrons may observe are probably canceled out, because the period of its static antiferromagnetic ordered structure is much shorter than a superconducting coherence length of the compound. However, in ferromagnetism we expect that superconducting electrons detect a nonvanishing internal field. Thus, it is quite surprising that the FM with a local moment of the order of $1\mu_{\mathrm{B}}$ per U atom coexists with the SC in UGe_2. This interesting feature raises a question how FM is compatible with SC. We believe that it is helpful for a deeper understanding of the coexistence to investigate the nature of the FM. In this Letter we report the observation of quantum-mechanical effects in the magnetization of a macroscopic sample; jumps at regular intervals of magnetic field in the hysteresis loop of a single crystalline sample of UGe_2. We ascribe these jumps to the field-tuned resonant tunneling, which is a kind of the macroscopic quantum tunneling (MQT) effect, and propose that the magnetic domain size of UGe_2 is smaller than its superconducting coherence length.

In general, a microscopic object that consists of one or a few numbers of elementary particles, such as an electron and α particle, can pass through a potential barrier by virtue of tunneling, which is the passage of the quantum-mechanical particles by way of a classically forbidden path, while a macroscopic object does not pass harmlessly. This is also the case for spins; the probability that macroscopic number ($\sim 10^{23}$) of spins in a bulk ferromagnet would change simultaneously their direction is extremely small. However, it was theoretically suggested that dynamics of a sufficiently tiny magnet consisting of a lot of spins can be represented by a single parameter of the magnetization that is a sum over all these spins [5,6]. Experimentally a magnetization jump in a hysteresis loop was observed not only in an ensemble of chemically identical molecular magnets such as $Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4$ (referred to as Mn_{12} hereafter) [7], in which each molecule contains spins of order of 10, but also in a small (i.e., nanometer-scale) magnetic entity, which includes many more spins [8,9]. This implies that a lot of spins change their direction simultaneously from one easy axis of magnetization to another by virtue of tunneling, which we call MQT.

Single crystalline ingots of UGe_2, which crystallizes in the orthorhombic crystal structure, were grown by the Czochralski pulling method using a tetra-arc furnace. Two small pieces (named sample No. 1 and No. 3 in the present paper) with approximate size of $3 \times 2 \times 2$ mm^3 were cut from different ingots for the present measurements. As may be seen from Ref. [10], sample No. 3 showed superconductivity at pressures above about 11 kbar, with a superconducting onset temperature T_c of about 0.7 K at 11.8 kbar [10]. Sample No. 1 also showed the superconducting transition (not shown here); $T_c \sim 0.6$ K at 12.1 kbar. The dc magnetization was measured by means of a laboratory-made vibrating-sample magnetometer, and the specimen was immersed in liquid ^{3}He; the accessible lowest temperature was about 0.4 K. The measurements at high pressures were made with a beryllium-copper piston-cylinder clamp device using Fluorinert as a pressure-transmitting medium. The maximum pressure in the present investigation was about 12 kbar due to pressure cell limitations. Detailed description of the sample preparation and the measurements is given elsewhere [10].

Figure 1 shows the magnetization M for single crystalline sample No. 1 as a function of an external magnetic field H. We note in Fig. 1(a) that M for H parallel to the crystallographic a axis, which is the easy axis of the magnetization [11], is easily saturated by an external field of $\sim$1.5 kOe, while an anisotropy field needed to saturate M for hard axes (i.e., b and c axes) is in excess of 10^3 kOe. This large anisotropy is ascribed to the magnetocrystalline effect, as is usual for actinide magnets; spins in UGe_2 are

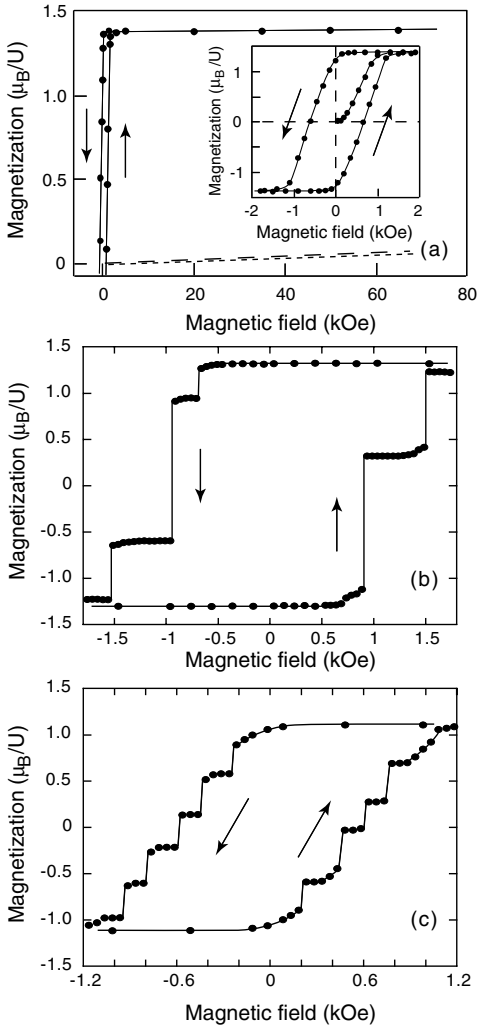

FIG. 1. Magnetization M as a function of applied external magnetic field H. Arrows indicate the sweep direction of the field. (a) Measurements were done at atmospheric pressure and 4.2 K. Closed circles correspond to H applied parallel to the a axis, and the dotted (broken) line to the field parallel to the b axis (c axis), the latter being taken from Ref. [11]. Measurements were also carried out (b) at atmospheric pressure and 0.52 K and (c) at 11.5 kbar and 0.44 K. The ambient pressure data were taken after pressurization. Note the jumps in the hysteresis loop that were observed only at low temperatures.

strongly coupled to the electronic charge density via relativistic spin-orbit interaction, and their energy is therefore dependent on their absolute orientation with respect to the crystal axes. As is shown in the inset of Fig. 1(a), a continuous hysteresis loop was observed at 4.2 K and ambient pressure, which is usual for a macroscopic size ferromagnet. When the temperature is reduced down to 0.52 K, the magnetization curve changes to a steplike or staircaselike loop, as seen from Fig. 1(b). This unusual feature was also detected at a high pressure of 11.5 kbar [Fig. 1(c)], at which T_{Curie} is about 2/3 of that at ambient pressure. Since these steps were observed only at low temperatures below about 1 K, the novel phenomenon is likely due to a quantum-mechanical effect. Indeed, it is known that a single domain particle of ferromagnets such as Ni shows a similar jump in the magnetization near a coercive field H_c [9]. However, the magnetization jumps for the present case occur at a field that is 3 orders of magnitude smaller than a calculated value of $H_c = 2K_1/M_0 \sim 7 \times 10^2$ kOe, where K_1 and M_0 will be defined below. Furthermore, we observe similar behavior for sample No. 3 with differing quality [12], which suggests that the steplike curve of the magnetization is inherent to UGe_2.

Figure 2(a) shows a plot of a derivative of M with respect to H that was measured at 0.43 K and 9.5 kbar. Reflecting the sudden jump in the M-H curve, the derivative yields a very sharp peak. We stress that the peak appears regularly as a function of the field. This is more clearly seen in Fig. 2(b); all of the data mentioned above are lying on the same straight line in a plot of step number n versus H/H_0, independent of the pressure (equivalent to the Curie temperature) and the sample quality, where H_0

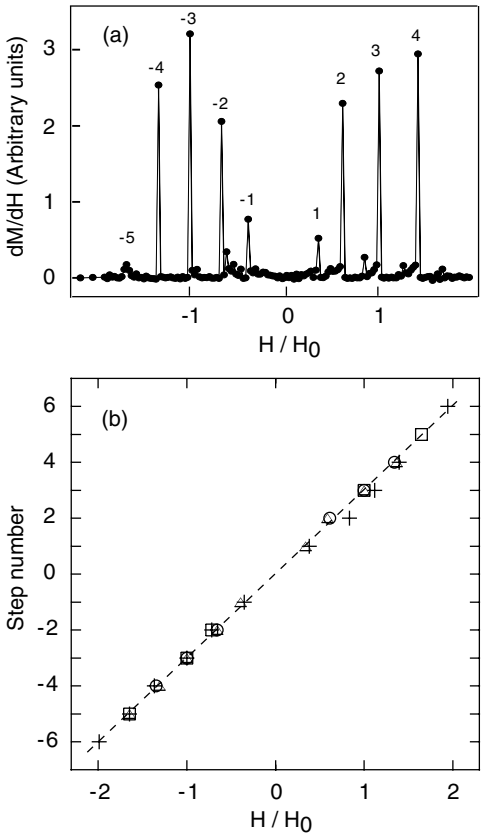

FIG. 2. Relationship between step number and magnetic field at which steps occur. (a) Derivative of magnetization with respect to field is plotted as a function of H/H_0, where H_0 denotes a field at which $M = 0$. Measurements were done at 0.43 K and 9.5 kbar. Each peak is labeled by step number n as illustrated. (b) Step number is plotted as a function of H/H_0. Symbols are defined as follows; (square) ambient pressure for sample No. 1, (triangle) 9.5 kbar for sample No. 1, (cross) 11.5 kbar for sample No. 1, and (circle) 0.4 kbar for sample No. 3. Note that all data lie on the same straight line that demonstrates a universal quantization relation $H/H_0 = (1/3)n$, independent of the Curie temperature and sample quality.

is defined as a magnetic field at which $M = 0$. H_0 depends on both temperature and pressure, and thus the above universal relationship indicates that this effect on the steplike behavior of magnetization is renormalized in the parameter H_0. This regularity, showing the relation $H = n(H_0/3)$, where n is an integer and H is a field at which the step occurs, strongly suggests that the steplike curve is not due to extrinsic effects such as the motion of domain walls pinned at imperfections as revealed in the disordered alloy $SmCo_{3.5}Cu_{1.5}$ [13].

These observations are our primary results in the present investigation and are to be distinguished from others: The jumps observed in the single domain particle of Ni and in the bulk sample of $SmCo_{3.5}Cu_{1.5}$ do not exhibit such a quantization relation. The similar staircaselike magnetization curve was observed in the oriented crystal of Mn_{12}; however, it is to be noted that the crystal is simply a collection of molecules containing 12 magnetic Mn ions, which is essentially distinguished from the bulk sample of UGe_2 used in the present study.

In order to understand an origin of the novel feature, let us first consider an effect of the anisotropy on the magnetization of a single domain. The anisotropy and Zeeman energies can be expressed as follows [5]:

$$E = (K_1 + K_2 \sin^2\phi)\sin^2\theta - M_0 H(1 - \cos\theta), \quad (1)$$

where K_1 and K_2 are anisotropy constants corresponding to unit volume, θ and ϕ denote angles for the direction of $\vec{M}$ in a spherical coordinate system, and M_0 indicates a magnitude of $\vec{M}$. The longitudinal fields K_1 and H create an effective potential with a barrier of a height U (see Fig. 3). Equilibrium easy directions of the magnetization correspond to the local minima of the energy at $\theta = 0$ and π for a positive K_1. Next we consider the following Hamiltonian that is equivalent to Eq. (1) up to a constant and corresponds to the energy per domain with volume V [5,7], to obtain eigenvalues of the system,

$$\mathcal{H} = -d_1 J_\parallel^2 + d_2 J_\perp^2 + g_{\text{eff}}\mu_B J_\parallel H . \quad (2)$$

Here g_{eff} and $J_\parallel$ ($J_\perp$) are an effective g factor and a parallel (perpendicular) component of a total angular momentum operator $\vec{J}$ to the a axis, respectively. $\vec{J}$ is equal to $N\vec{j}$, where N is the number of spins contained in the domain and $\vec{j}$ is a single ion total angular momentum. Simple calculation leads to the following relation: $K_1 = d_1 J(J + 1)/V$ and $K_2 = d_2 J(J + 1)/V$. Since the projection of $\vec{J}$ onto the a axis does not commute $\mathcal{H}$, eigenvalues of $J_\parallel$ are not conserved quantum numbers even at $H = 0$. Consequently, the transverse anisotropy d_2 plays a crucial role to generate quantum transitions of M between the two energy minima. However, d_2 can be neglected to calculate eigenvalues because the anisotropy within the plane perpendicular to the a axis is very small (see Fig. 1). The results are schematically displayed by a horizontal bar in Fig. 3, and an energy separation between the ground and first excited state is denoted by ΔE.

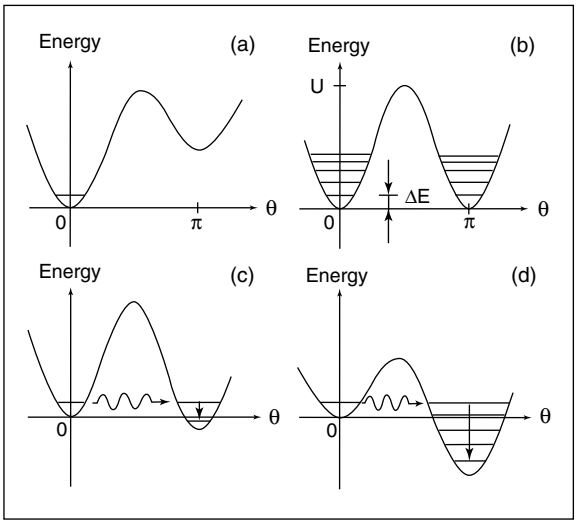

FIG. 3. Schematic illustration of effective potential and discrete energy levels. θ denotes an angle of the magnetization measured from the easy axis. (a) At an initial state, every domain is assumed to be in the ground state of the left-hand minimum at $\theta = 0$. (b) When $H = 0$, two energy minima are degenerate at $\theta = 0$ and π. (c) When $H = d_1/(g_{\text{eff}}\mu_B)$, the resonating tunneling from the metastable state in the left to the (first) excited state in the right occurs, followed by a rapid spontaneous decay into the ground state. (d) When $H = nd_1/(g_{\text{eff}}\mu_B)$ with $n \sim 5$, most of the domains will finish to move from the left to the right, and thus there will be no more transition.

Then let us consider the magnetism of an ensemble of domains at low temperatures. (Since we do not identify "domain" at present, we call a unit carrying the moment defined by $-g_{\text{eff}}\mu_B\vec{J}$ as the magnetic domain throughout this paper.) We assume that at an initial state all domains are in the ground state of the left-hand well [Fig. 3(a)]. When the field is reduced to zero [Fig. 3(b)], the two minima are degenerate, but almost all domains may remain in the left due to the energy barrier. When the field is reversed [Fig. 3(c)], the left-hand minimum becomes metastable, which probably induces tunneling across the barrier. In particular, when the energy level of the ground state of the left coincides with the nth excited level of the right, it seems that a lot of domains resonantly tunnel from the left to the right, which probably explains the observed sharp and large jumps in the magnetization. (This resonant tunneling is likely followed by rapid spontaneous decay from the excited to the ground state [7].) A simple calculation shows that this resonance is possible when the condition $H = n(d_1/g_{\text{eff}}\mu_B)$ is satisfied. This is compatible with the quantization relation deduced from Fig. 2. When the field strength reaches a value corresponding to $n \sim 5$ [Fig. 3(d)], a significant fraction of domains will finish to tunnel into the right, leaving only a small number of domains in the left. Therefore, we will observe no more noticeable jumps.

In the following we numerically estimate the abovementioned quantities. Given that the step occurs every

300 Oe, we find the anisotropy constant (corresponding to the anisotropy energy per domain) to be $d_1 \sim 2 \times 10^{-18}$ erg $= 1 \times 10^{-2}$ K, assuming $g_{\text{eff}} = 4/5$ for $5f^2$ configuration, as suggested for the superconducting antiferromagnet UPd_2Al_3 [4]. The anisotropy field of 10^3 kOe may yield $K_1 \sim 2 \times 10^8$ erg/cm^3. From these values and the aforementioned relation $K_1 \simeq d_1 J^2/V = d_1 N j^2/\upsilon$, where $\upsilon \sim 6 \times 10^{-23}$ cm^3 is the volume occupied by one uranium atom, we estimate $N \sim J \sim 10^3$, assuming $j = 4$ for the $5f^2$ configuration. This is equivalent to a domain size of $l \sim (N\upsilon)^{1/3} \sim 40$ Å. This extraordinarily small number of spins contained in the domain, or equivalently small volume of the domain, explains why UGe_2 exhibits the field-tuned macroscopic quantum resonance; for a conventional ferromagnet whose domain size is typically of the order of 10^4 Å, the tunneling probability is very small, because the energy barrier U is proportional to N. Furthermore, the energy barrier U and the energy separation ΔE are estimated as $U \simeq d_1 J^2 \sim 10^4$ K and $\Delta E \simeq d_1 J \sim 10$ K, respectively. This value of ΔE may be consistent with the observation that the steps were observed only below $\sim$1 K, because the quantum-mechanical phenomenon can be observed only when $\Delta E \gg k_B T$; otherwise statistical averaging over states masks all evidence of discrete energy levels.

Finally, let us make a brief discussion about the relationship between the unusual nature of FM and SC. Remembering that the superconducting coherence length of UGe_2, $\xi \sim 130$–200 Å [14], we note that the domain size l is several times smaller than ξ. If UGe_2 were like the conventional ferromagnet, the pair wave function would be as in an infinite bulk specimen; thus the domain boundary may give only a secondary effect. In the present case, however, it is likely that the domain structure affects the nature of superconductivity, as discussed by Leggett for superfluid ^{3}He [15]; a boundary such as a wall may in general strongly distort the bulk Cooper pair wave function. Provided there is a perfect energy splitting of spin-up ($\uparrow$) and spin-down ($\downarrow$) bands due to a ferromagnetic exchange interaction, the spin state of Cooper pairs in the form of (say) $\uparrow\uparrow$ may be preferred in a given magnetic domain. However, the pair will not survive in neighboring domains with opposite polarization. Indeed, if there is some interaction such as Josephson coupling between the $\uparrow\uparrow$ and $\downarrow\downarrow$ pair wave functions in adjacent domains with opposite polarization, a distorted pair wave function that varies in the real space may be formed. If so, the SC can be inhomogeneous in the real space. This seems to be consistent with a heat capacity experiment that possibly suggests that only a part of a sample becomes superconducting [16]. Instead of such a pair breaking effect, it is possible that such a domain structure favors the coexistence of FM and SC; if the domains are adversely oriented, then internal molecular fields due to the FM can be canceled out in the scale of coherence length.

In conclusion, we report quantum-mechanical effects on the magnetization of macroscopic single crystalline samples of UGe_2. We attribute this to the field-tuned resonant tunneling of magnetization between different quantum spin states, and suggest that the observation is ascribed to a very small size of the magnetic domain. Many years ago Anderson and Suhl argued theoretically that purely ferromagnetic alignment of ion-core spins in superconductors should not be observed in preference to a domainlike alignment [17]. In UGe_2 there is no experimental evidence that the long-wavelength paramagnetic susceptibility is reduced below the superconducting transition temperature; therefore it is not clear if the above "cryptoferromagnetic" alignment is relevant to the present results. Finally we hope that the present discovery will stimulate such theoretical investigations that take into account the magnetic domain effect on the spin-triplet superconductor.

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