Highly anisotropic magnon dispersion in Ca$_2$RuO$_4$: evidence for strong spin orbit coupling

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The magnon dispersion in Ca$_2$RuO$_4$ has been determined by inelastic neutron scattering on single crystals containing 1% of Ti. The dispersion is well described by a conventional Heisenberg model suggesting a local moment model with nearest neighbor interaction of $J$=8 meV. Nearest and next-nearest neighbor interaction as well as inter-layer coupling parameters are required to properly describe the entire dispersion. Spin-orbit coupling induces a very large anisotropy gap in the magnetic excitations in apparent contrast with a simple planar magnetic model. Orbital ordering breaking tetragonal symmetry, and strong spin-orbit coupling can thus be identified as important factors in this system.

The properties of strongly correlated systems with significant spin-orbit coupling (SOC) present a challenging problem. The intensively studied example is the reduction of the magnetic state of Ir$^{4+}$ (electronic structure $5d^5$ or $t^2_g$, $L_{eff} = 1$, $S=1/2$) to an effective Kramers doublet with $J=1/2$ [1]. But even more drastic effects can be expected for heavy ions with $4d$ occupation ($t^2_g$, $L_{eff}=1$, $S=1$), e.g. in Ir$^{5+}$, Ru$^{4+}$, Os$^{4+}$ etc. [2]. According to Hund’s rules (generalized for ions sensing crystal electric fields) the ground state should be a nonmagnetic singlet with $J=0$, see e.g. reference [3,4]. And indeed isolated Ir$^{5+}$ ions and also most of concentrated Ir$^{5+}$ compounds are nonmagnetic, although a few magnetic Ir$^{5+}$ cases are known [5]. In a solid magnetic order can occur even if the ground state of an isolated ion is a singlet, see chapter 5.5 in reference [5], but it requires a strong exchange interaction, so that the exchange splitting of excited magnetic states (in the Ru$^{4+}$ case a $J=1$ triplet) is larger than the energy difference between the ground-state singlet and the excited triplet, which is given by the SOC parameter $\lambda$. The SOC can also be at least partially suppressed by a non-cubic crystal field (CF), $\Delta_{noncub}$, which splits the $t^2_g$ ($L_{eff}=1$) triplet and stabilizes real orbitals. Both these factors, CF and magnetic interaction, can combine to suppress the $J=0$ state and to eventually induce the magnetically ordered ground state. In terms of energy scales, one should expect such magnetic ordering for $\Delta_{noncub} + \mu \cdot H_{exch} > \lambda$, which seems quite unlikely for Ir$^{5+}$ where $\lambda = 2 \zeta / D = \zeta / 2$ amounts to 0.2 to 0.25 eV ($\zeta$ is the atomic spin-orbit parameter). But for $4d$ compounds this relation can easily be reached, as for Ru$^{4+}$ $\lambda \sim 0.075\text{eV}$ [2,6]. Indeed, practically all Ru$^{4+}$ compounds order magnetically from the metallic ones - and even some metallic ruthenates are magnetic, such as the ferromagnetic metal SrRuO$_3$. The persisting role of SOC in these magnetic Ru$^{4+}$ compounds is an intriguing open issue.

Ca$_2$RuO$_4$ (CRO) is such a Ru$^{4+}$ case, which has been intensively studied as the Mott-insulating analogue of the unconventional superconductor Sr$_2$RuO$_4$ [7,10]. CRO exhibits a metal-insulator (MI) transition at 357 K which is accompanied by a flattening of the RuO$_6$ octahedra [8,12]. This flattening continues upon further cooling until it saturates near the onset of magnetic order at $T_N \sim 110$ K. The magnetic structure is antiferromagnetic (AFM) with moments aligned parallel to the layers [8,12]. The electronic structure has been stud-

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**FIG. 1:** Intensity distribution in energy versus scattering vector, $Q$, planes taken at 2K around the $(1.5 0.5 0)$ magnetic zone center. a) and c) show the symmetrically equivalent dispersion along $(0,\xi,0)$ and $(\xi,0,0)$ direction, subplot b) and d) along $(\xi,\xi,0)$ and $(\xi,-\xi,0)$. The color coding corresponds to the raw data. Open symbols indicate the dispersion obtained by fitting single scans. Data were taken with final energies of 35 meV for constant $Q$ scans at high energy transfer and 14.7 meV elsewhere.
ied by various approaches [13–17]. From the spectroscopic study of CRO it was concluded that SOC indeed plays an important role but is not sufficiently strong to stabilize the $J=0$ state [13]. Density functional theory calculations indicate a pronounced shift in orbital polarization leading to almost full occupation of the $d_{xy}$ levels at low temperature [14–18]. More recently the $J=0$ state was explicitly proposed for CRO [2, 6]. Starting from the scenario of strong SOC and including noncubic CF and intersite exchange, the magnetically ordered state in CRO is reproduced and several unusual features of the magnetic excitation spectrum of CRO are predicted, such as a peculiar shape and large width. The alternative, more conventional picture is to attribute the magnetism of CRO to the conventional $S=1$ state of Ru$^{4+}$ ions, with SOC playing a less significant but still prominent role. In this case one can describe the magnetic state, including spin waves, by the usual exchange Hamiltonian.

Here we present an inelastic neutron scattering (INS) study and spin-wave calculations of the magnetic excitations in CRO. We find that a conventional model can well describe the obtained dispersion, while there are considerable differences with the proposed $J=0$ model [6]. Most interestingly there is a sizable spin-gap which indicates that rotating the magnetic moment within the layers costs large energy. The breaking of the local tetragonal symmetry and the associated orbital polarization, which has been neglected in theory so far [14–18], are important parameters to understand the magnetism in CRO.

CRO single crystals of of several 100mm$^3$ volume containing 1% of Ti were obtained by the travelling solvent floating zone method in a mirror furnace (Canon SC1-MDH11020-CE). We added 1% of Ti as this seems to avoid bursting of the crystal upon cooling below the MI transition. Characterization by magnetic susceptibility and by neutron diffraction experiments indicate a magnetic transition at $T_N=112$ K and no significant impact of the very small amount of Ti. INS experiments were performed with the IN8 triple-axis spectrometer. Most experiments were performed with fixed final momentum of $k_f=2.662$ Å$^{-1}$ ($E_f=14.7$ meV); some scans at high energy transfer or aiming at better resolution were performed with $k_f=4.1$ and $k_f=1.97$ Å$^{-1}$, respectively. We studied the magnon dispersion in the two scattering geometries $(100)/(010)$ and $(110)/(001)$ in reduced units of the tetragonal lattice [12]. For both setups two crystals were coaligned. The sample of the second scattering plane was essentially untwinned as determined on the IN3 spectrometer.

Fig. 1 shows color mappings of the measured intensity distribution. Due to the weakness of scattering in CRO (small moment and rapidly decreasing form factor) contaminations by various phonon branches are highly visible. By analyzing and comparing results taken in different Brillouin zones and geometries the dispersion can be unambiguously determined. Magnon excitations start at the AFM Bragg points $(\tfrac{2n_k+1}{2} \tfrac{2n_l+1}{2} n_l)$ with integer $n_k$, $n_k$ and $n_l$. However, there is a sizeable spin gap of 13.04(5) meV. For a square planar antiferromagnet the magnon dispersion extends from $Q=(0.5 0)$ to $(0.75 0.75)$ in [1 1] direction, as (1 1) is a Bragg point, and to $(0.5 0.5)$ in [1 0] direction. $Q=(0.25 0.25)$ and $(0.5 0.5)$ are AFM Brillouin zone boundaries. In CRO there is, however, a severe structural distortion [12]. Some characteristic scans performed to determine the magnon dispersion in CRO are shown in Fig. 2. Constant energy scans at intermediate energy cut through the magnon cones at two positions. Constant $Q$ scans taken just at the AFM zone center show a characteristic asymmetric shape, see Fig. 2 (c) and (d): Intensity rapidly increases when crossing the spin gap and slowly diminishes with further energy increase. We have calculated the folding of the spin-wave dispersion including its expected signal strength with the experimental resolution using the RESLIB [20] package and verified that scans across transversal acoustic phonons are well reproduced, see Fig. 2 (b). The steep spin wave dispersion perfectly describes the asymmetric shape of the spectra taken at the zone center, see Fig. 2 (c). The total width of the dispersion is low, as maximum energies of 37.8(3) and 41.2(5) meV are reached at the magnetic zone boundaries, $(0.5 0.0)$ and $(0.25 0.25)$.

In order to describe the magnon dispersion we use FIG. 2: Several characteristic scans: a) Constant energy scans at $(1.5,k,0)$ taken at 2 K fitted with gaussians and background. b) Phonon scans taken at $Q=(\xi,0,0)$; the lines correspond to the folding of the resolution function with a simple linear phonon dispersion. No additional parameter is needed to describe the shape of the intensity profile. c) and d) show the energy scans at the zone center $(1.5 0.5 0)$ and at $(0.5 0.5 0)$. 

a conventional Heisenberg model with a single-site anisotropy term arising from SOC: $H = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \delta \sum_i (S_i^z)^2$. We set $\delta = 0.67$ following the neutron diffraction study \cite{20}. The sum runs over pairs of magnetic ions, so that each pair or bond appears twice. Spin waves were calculated with the Holstein-Primakoff transformation as described in references \cite{21, 22}. We include the nearest-neighbor magnetic exchange of $J = 8$ meV, next-nearest neighbor interaction along the orthorhombic $a$ and $b$ directions of $J_{anb} = J_{nb} = 0.7$ meV and an AFM coupling between neighboring layers. The next-nearest neighbor interaction is chosen isotropic, as the twinned crystal used in the $(100)/(010)$ geometry prohibits distinguishing these directions. The need for the additional parameter can be seen when comparing the magnon energies at $Q = (0.25 0.25 0)$ and $(0.5 0 0)$, which are identical in the model with only nearest-neighbor interaction. The interlayer coupling, $J_z = 0.03$ meV is the only parameter that breaks the tetragonal symmetry in our model aside from the single-ion anisotropy. Note, however, that the crystal structure is orthorhombic lifting the degeneracy of magnetic interaction parameters. We chose the AFM interaction between the Ru at $(0,0,0)$ and that at $(0,0.5,0.5)$ (in the orthorhombic cell \cite{19}), which stabilizes an $A$ centered magnetic structure with magnetic space group $Pbca$ \cite{12}.

The magnetic moment in CRO points along the orthorhombic $b$ direction. Therefore, one might expect a large gap for the magnetic excitations involving rotations of the moment out of the RuO$_2$ layers, and much softer in-plane modes. The latter are described by the crystal structure is orthorhombic lifting the degeneracy of magnetic interaction parameters. We chose the AFM interaction between the Ru at $(0,0,0)$ and that at $(0,0.5,0.5)$ (in the orthorhombic cell \cite{19}), which stabilizes an $A$ centered magnetic structure with magnetic space group $Pbca$ \cite{12}.

The magnon dispersion in CRO points along the orthorhombic $b$ direction. Therefore, one might expect a large gap for the magnetic excitations involving rotations of the moment out of the RuO$_2$ layers, and much softer in-plane modes. The latter are described by the expectedly small in-plane anisotropy. Following reference \cite{22, 23} both branches can be described simultaneously with two anisotropy parameters. Surprisingly, in CRO the in-plane anisotropy turned out to be extremely strong. The magnon dispersion starts at 13.04(5) meV which we may identify with the in-plane gap. There is no magnon branch at lower energy as is clearly shown in the intensity maps. We find some weak intensity at an energy transfer of 5 meV appearing near $Q = (1.5 0.5 0)$, but this signal is flat in energy and restricted to the AFM zone-center. Furthermore, this signal is much weaker than the magnon modes at higher energies suggesting a possible origin associated with the Ti impurities and some local effect. As shown in Fig. 2 (d) and 3 there is a finite inter-layer dispersion visible in the scans taken at $Q = (0.5 0.5 q_j)$ with the second untwinned crystal. The tetragonal $[110]$ direction corresponds to orthorhombic $b$ in the used mounting and thus to the direction of the magnetic moment; therefore, the transverse magnon with in-plane polarization (thus parallel to orthorhombic $a$) fully contributes. Also in the other configuration there is a clear difference in spectra taken at $Q = (0.5 0.5 0)$ and $= (1.5 0.5 0)$. For the twinned sample we superpose AFM zone-centers and zone boundaries. For a twinned crystal c polarized magnons will always contribute, while for the in-plane magnon the geometry condition that only magnetic components perpendicular to $Q$ contribute, suppresses some modes. The fact that we see a clear difference at various $(\frac{2n_2+1}{2}, \frac{2n_1+1}{2}, n_1)$ unambiguously shows that the modes dispersing between 13.04(5) and 14.2(1) meV posses an in-plane polarization. This furthermore agrees with the $Q_f$ dependence of the signal. We may thus conclude that the lowest magnon branch in CRO possesses an in-plane character and that it disperses between 13.04(5) and 14.2(1) meV along the $c$ direction and up to 41.2 and 37.8 meV along $(\xi, 0, 0)$ and $(\xi, 0, 0)$ paths, respectively. We cannot identify the $c$ polarized modes as they may remain hidden in the asymmetric shape. There is some evidence for a nearly flat branch around 36 meV, but we cannot fully rule out that these modes are purely nuclear or that they carry longitudinal polarization. For simplicity, the experimental dispersion is described by an easy-axis anisotropy \cite{23}, see Fig. 3.

The magnon dispersion including its large gap can be very well described within the spin-wave theory suggesting a conventional local moment $S = 1$ magnetism with a strong - but not decisive - impact of SOC. Starting from the other scenario, a spin-orbit driven $J = 0$ singlet nature which is rendered magnetic by noncubic CF and intersite exchange, Akbari and Khalilullin \cite{6} predicted several unusual features of the magnetic excitation spectrum, such as the energy continuously softening from the value $\lambda$ at $\Gamma$, and the presence of extra modes in some part of the spectrum. Our results, however, do not support this model \cite{6}. First, the observed dispersion is much flatter than this prediction, as it does not reach energies of the order of the expectedly large values of $\lambda$, and as there is a strong gap. Second, the singlet picture predicts a continuously increasing dispersion near the AFM zone boundaries, while our experiments find the saturation predicted by the Heisenberg model, see Fig. 1 and 2. The Heisenberg scenario also implies several branches: two transversal branches arise from the orthorhombic anisotropy (in-
plane and c polarized), and longitudinal modes can exist in CRO being on the border to itinerancy.

Using the standard description, with the hopping parameters $t \sim 100\text{meV}$, obtained by ab-initio calculations [19, 24], and using the Hubbard’s $U \sim 2\text{eV}$, we would obtain for the exchange constant $J=2t^2/U \sim 10\text{meV}$, in good agreement with our experimental finding. However, CRO is not a strong Mott insulator with completely localized electrons as it is already indicated by the low-lying MI transition. In this case the basic $J=0$ Ansatz may be not a good starting point, as the $J=0$ state can be suppressed by electron hopping. Also for Ir$^{4+}$ (specifically for Na$_2$IrO$_3$) the sizable hopping modifies the whole picture [25, 26], leading to novel quasi-molecular orbital states with reduced impact of SOC. The conspicuous but typical absence of $J=0$ physics in most of the Ru$^{4+}$ materials seems largely connected with the hopping.

Another argument in favor of the applicability of the usual picture of Ru$^{4+}$ ions (S$\sim$1) is the strong flattening of RuO$_6$ octahedra [9, 11] occurring below the MI transition. Such distortion is typical for the usual Jahn-Teller effect: it stabilizes the doubly-occupied $d_{xy}$ orbital, leaving two electrons on $d_{xz}$ and $d_{yz}$. In such state the orbital moment and spin-orbit interaction are partially quenched. The sign of this distortion proves that in this system the Jahn-Teller effect is stronger than the SOC which would have caused the opposite distortion and CF splitting [4]. Recent spectroscopy data [17] confirm this significant splitting of $t_{2g}$ orbitals.

On the other hand the observation of the strong in-plane magnetic gap is remarkable for a layered system. It underlines the relevance of the SOC in CRO even in the conventional scenario. Several Raman scattering experiments observed an additional signal in $B_{1g}$ symmetry appearing in the AFM phase [27, 29]. This feature was interpreted as a two-magnon excitation, but our results clearly rule out such explanation. The Raman feature appears at 102 cm$^{-1}$=12.6 meV at 10 K which is much below the energies for two magnon excitations and the expected peak in the two-magnon density of states (near 80 meV). Instead this energy agrees with that of the in-plane gap mode. The single magnon mode, however, is not Raman active in first approximation, but SOC can induce a finite signal. The temperature dependence and the extreme broadening of the Raman signal at higher temperature agree reasonably well with the corresponding behavior of the magnon gap, see inset in Fig. 4 (b).

The magnetic in-plane anisotropy in CRO must originate from SOC and from an orbital arrangement breaking tetragonal symmetry. There have been many experimental and theoretical analyzes [13–18] elucidating the change of the orbital polarization upon cooling and the increasing occupation of the $d_{xy}$ versus the $d_{xz}/d_{yz}$ orbitals following the flattening of the RuO$_6$ octahedron. This distortion possesses $E_g$ symmetry, which is the most frequently analyzed in Jahn-Teller models [30]. The $t_{2g}$ orbitals, however, also couple to the $T_{2g}$ octahedron distortions [30] which break tetragonal symmetry in the case of CRO but which were neglected so far. The temperature dependence of the crystal structure of CRO in the insulating phase reveals an ongoing elongation of the RuO$_6$ octahedra [9, 11] along the orthorhombic $b$ direction along which moments align. This distortion corresponds to the $T_{2g}$ ”scissor” mode of the free octahedron [30] lifting the $d_{xz}/d_{yz}$ degeneracy. Similar to a tetragonal distortion, e.g. along $z$-axis, which would stabilize orbitals with $t^z = \pm 1, (d_{xz} \pm id_{yz})$, and which, by SOC would orient spins along $z$-direction, (or a trigonal elongation along [111] (in cubic setting), which would make [111] an easy axis, see e.g. [4]), such $T_{2g}$ distortion (elongation along $b$ axis) makes the orthorhombic $b$-direction the easy axis.

In conclusion we have studied the magnon dispersion in CRO which considerably differs from recent predictions for a $J=0$ singlet ground state. Instead, the dispersion is well described in a local moment Heisenberg model with strong anisotropy terms yielding a nearest-neighbor exchange interaction of $J=8\text{meV}$ which agrees with the large calculated hopping integrals. Large hopping seems to be the main cause for the suppression of the $J=0$ state in Ru$^{4+}$ compounds. On the other hand, the remarkably strong in-plane anisotropy clearly shows that considering tetragonal crystal fields is insufficient. There is important orbital polarization breaking tetragonal symmetry, which is related to the prominent elongation of RuO$_6$ octahedra along the orthorhombic $b$ direction and which renders spin-orbit coupling still active in this system.

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