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An ab initio study of the magnetocrystalline anisotropy and magnetoelastic coupling of half-metallic CrO_2

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Abstract

First-principles density functional calculations of the total energy, magnetic moments and magnetocrystalline anisotropy energy (MAE) of CrO_2 as a function of both volume and uniaxial strain along the *c*-axis have been performed. The highly accurate all-electron full-potential linearized augmented plane wave method and the generalized gradient approximation to the exchange–correlation potential are used. The calculated structural properties (lattice constants and unit cell volume) are in excellent agreement with experiments (with 0.5%). The calculated bulk and Young's modulii are 2.56 and 2.02 Mbar, respectively. The calculated MAE increases almost linearly with the uniaxial strain and remains positive in the strain range of −4–4%. Thus, the calculations predict that the easy magnetization axis is along the *c*-axis, in agreement with experiments. However, the calculated anisotropy constant is about six times larger than the measured value. The calculated magnetoelastic coupling constant is 1.2×10^7 erg/cm^3 and the magnetostriction coefficient λ_{001} is -2.59×10^{-5}.
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1. Introduction

CrO_2 has been attracting considerable interest in recent years, in part due to the fundamental interest in its half-metallic electronic band structure [1] and its applications of this, and in part because of its practical importance in magnetic recording. Being a half-metallic ferromagnet, with only one spin band at the Fermi level, CrO_2 has the highest measured spin polarization of all materials to date [2,3]. Together with a high Curie temperature of 390 K [4], CrO_2 provides excellent prospects for magnetoelectronic devices at room temperature.

Because of its uniaxial crystalline structure, it is expected to have a large magnetic anisotropy which makes it the favoured material for magneto-optical

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recording applications. Despite a large number of published theoretical calculations (see Refs. [1,5–8] and references therein), no ab initio calculation of the magnetic anisotropy energy has been reported yet. Here in this paper, we present the results of our systematic theoretical calculations of the magneto-crystalline anisotropy energy (MAE) and also its strain dependence for CrO_2.

CrO_2 crystallizes in the rutile structure (space group D_{4h}^{14}: $P4_2/mnm$) with a tetragonal symmetry. The primitive unit cell contains two molecular formula. The two chromium atoms are located at the positions $(0,0,0)$ and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, and the four oxygen atoms are at $(u,u,0)$, $(1-u,1-u,0)$, $(\frac{1}{2}+u,\frac{1}{2}-u,\frac{1}{2})$, and $(\frac{1}{2}-u,\frac{1}{2}+u,\frac{1}{2})$. The measured lattice constants a and c are 4.419 and 2.912 Å, respectively [9]. The internal coordinate of the oxygen atoms u is 0.303 [9]. The electronic structure and total energy of CrO_2 were calculated by using the highly accurate full-potential linearized augmented plane wave (FLAPW) method, as encoded in WIEN97 [10]. The calculations were based on the first-principles density functional theory (DFT) with the generalized gradient approximation (GGA) [11].

In the present calculations, the muffin-tin sphere radii R_{mt} of 1.9 a.u. for Cr and 1.5 a.u. for O were used. The parameter $R_{mt}K_{max}$ was set to 7.0, i.e., a large number of augmented plane waves (PW), about 110 PWs/atom, were used. In the total energy calculations, the number of k-points in the irreducible Brillouin zone wedge (IBZW) used is 290. However, because the MAE is generally very small, a much finer grid for the Brillouin zone integration is needed. Thus, a large number of the k-points in the IBZW (2896) was used.

The calculated total energy of CrO_2 as a function of volume is shown in Fig. 1. Here c/a ratio and u are fixed to the experimental values [9], respectively. By fitting this total energy vs. volume curve to the equation of state, we obtain the theoretical unit cell volume V_0 of 57.7 Å^3 or 389.2 a.u.^{3} and bulk modulus B of 2.56 Mbar. The calculated total and local Cr spin-magnetic moments in CrO_2 as a function of volume are also shown in Fig. 1. As mentioned before, CrO_2 is a half-metallic ferromagnet at ambient pressure. Its spin-magnetic moment is therefore quantized,

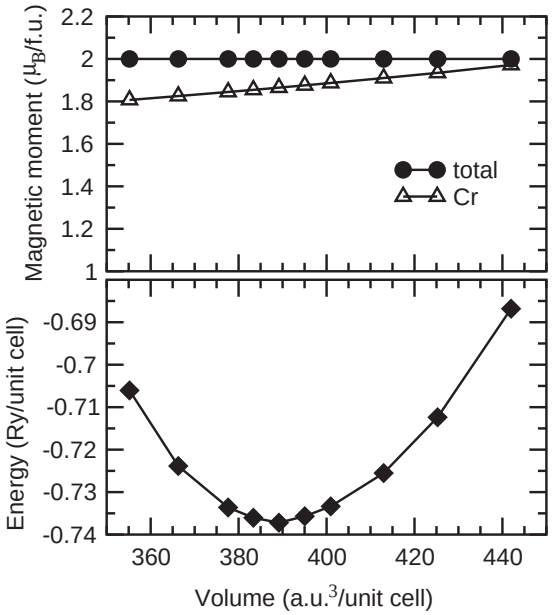

Fig 1. Total energy (lower), total and local Cr spin magnetic moments (upper) of CrO_2 as a function of volume, c/a is fixed at 0.664. The total energy is relative to −4805.0 Ry/unit cell. The experimental unit cell volume is 385 a.u.^{3}.

being equal to 2.0 μ_B/f.u. Fig. 1 shows that the total magnetic moment of CrO_2 remains constant (2.0 μ_B/f.u.), indicating that it is half-metallic throughout the volume range studied. Nevertheless, the local magnetic moment in the Cr atom does increase monotonically with volume (Fig. 1).

Now, we calculate the total energy as a function of c/a ratio with the volume fixed to 57.7 Å^3. This is equivalent to applying a uniaxial elastic strain ε to CrO_2 along the c-axis. The strain ε is defined as $a = a_0(1+\varepsilon)$ and is given by $[(c_0/a_0)/(c/a)]^{1/3}-1$. The results are displayed in Fig. 2a. Clearly, the total energy minimum is fairly close to the zero strain. By fitting a polynomial to the total energy vs. strain curve, we get the theoretical c/a ratio of 0.659 and young modulus Y_{001} of 2.02 Mbar. Thus, the theoretical lattice constants a and c are 4.439 and 2.926 Å, respectively. Clearly, the theoretical lattice constants and unit cell volume are in good agreement with the experimental ones (within 0.5%), suggesting that the present GGA calculations describe the structural properties of CrO_2 very well. It should be noted that in the strain range

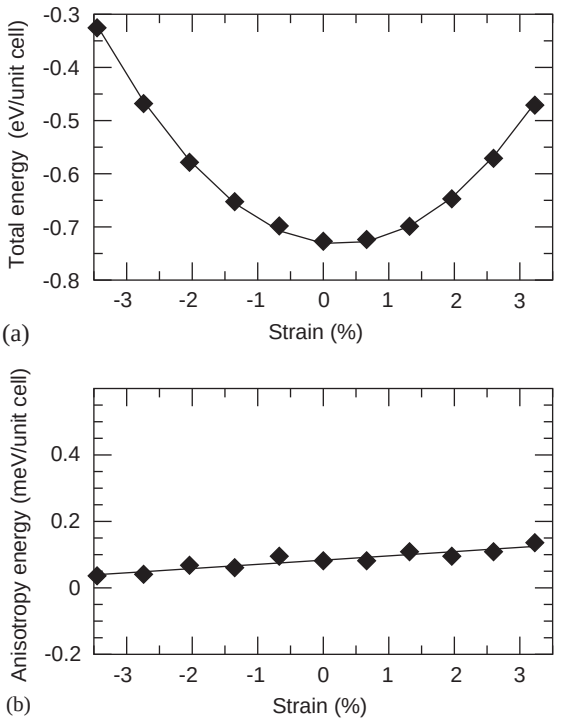

Fig 2. Total energy (a) and magnetocrystalline anisotropy energy of CrO_2 as a function of uniaxial strain along *c-axis*. In (a), the total energy is relative to −65385.727 eV/unit cell.

studied here, the total spin-magnetic moment of CrO_2 remains constant (2.0 μ_B/f.u.), again suggesting that the strained CrO_2 is still half-metallic.

The calculated MAE ΔE of CrO_2 as a function of the strain ε is shown in Fig. 2b. Here the MAE is defined as the difference in the total energy between the magnetization parallel to the *c*-axis and that parallel to one of the two *a*-axes. Fig. 2b shows that the MAE increases almost linearly with the strain. Furthermore, the MAE remain positive in the entire strain range studied here, i.e., the *c*-axis remains to be the easy axis and there is no spin-reorientation transition in this strain range. By fitting $\Delta E = K_{MA} + 3B^*(\varepsilon - \varepsilon_0)$ [12–14] to the curve, we obtain the magneto-elastic coupling constant B of 0.425 meV/unit cell or 1.18×10^7 erg/m^{3}and the uniaxial anisotropy constant K_{MA} of 23.2×10^5 erg/cm^3. From these numbers, we also obtain the magnetostriction coefficient $\lambda_{001} = -2.59 \times 10^{-5}$ by making use of magnetoelastic theory [15,16].

Experimental investigations of the magnetic anisotropy of bulk and thin film CrO_2 have been carried out by several research groups [17–21]. Because of its tetragonal symmetry, the leading terms of the magnetization orientation dependence energy of the bulk CrO_2 are $\Delta E = K_1 \sin^2\theta + K_2 \sin^4\theta + K'_2 \sin^4\theta \cos 4\phi$. The K'_2 is much smaller than K_1 and K_2 and is thus ignored for the bulk samples. Clearly, the above theoretical K_{MA} is equal to $K_1 + K_2$. Ferromagnetic resonance (FMR) experiments show that both K_1 and K_2 of bulk CrO_2 fine grains are positive from just below the Curie temperature down to low termperatures of a few Ks [17,18]. The measured K_1 and K_2 at about 120 K are 33×10^4 and 6×10^4 erg/cm^3, respectively [18]. Therefore, both the experiments and the present calculations show that the *c*-axis is the easy magnetization axis for CrO_2. Nonetheless, the theoretical anisotropy constant is nearly six times larger than the measured value. The large discrepancy between experiment and theory could be attributed in some extent to the fact that the measurements were made on fine grains. No measurements on single crystalline CrO_2 have been reported so far.

In summary, we have performed first-principles density functional calculations of the total energy, magnetic moments and MAE of tetragonal CrO_2 as a function of both volume and uniaxial strain along the *c*-axis. We used the highly accurate all-electron full-potential linearized augmented plane wave method and the GGA to the exchange-correlation potential. The calculated lattice constants are in excellent agreement with the experiments (with 0.5%). The calculated bulk and Young's modulii are 2.56 and 2.02 Mbar, respectively. The calculated MAE increases almost linearly with the uniaxial strain and remains positive in the strain range of −4–4%. Thus, the calculations predict that the easy magnetization axis is along the *c*-axis, in agreement with experiments. However, the calculated uniaxial anisotropy constant is about six times larger than the measured value. The calculated magnetoelastic coupling constant is 1.2×10^7 erg/cm^3 and the magnetostriction coefficient is -2.59×10^{-5}. We hope that these results will stimulate further experimental investigations of the MAE, magnetoelastic coupling and magnetostriction of CrO_2.

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