Magnetic Feshbach resonances and Zeeman relaxation in bosonic chromium gas with anisotropic interaction

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Rigorous calculations of magnetic Feshbach resonances and Zeeman relaxation cross sections in an ultracold and cold chromium gas are reported. This work presents the calculation of such inelastic processes in collisions of atoms with large magnetic moments. Using our recently constructed accurate interaction potentials for the chromium dimer, we compute the positions and widths of magnetically tuned resonances in an optically trapped ultracold Cr gas. We find good agreement with recent experimental data and report that spin-change collision cross sections quickly increase with decreasing collision energy and magnetic-field strength at energies below 1 K and magnetic fields larger than 40 G.

In the past decade, ground-breaking experiments on cooling alkali-metal atoms to ultracold temperatures have led to the creation of atomic Bose-Einstein condensates (BEC) [1] and degenerate Fermi gases [2], Bose condensation of Fermionic atoms [3], formation of ultracold molecules and the creation of molecular BEC [3–5], as well as the demonstration of the reversible superfluid-to-insulator phase transition in optical lattices [6]. A major thrust of recent research has been to cool and trap other atoms [7,8]. Trapping ultracold alkaline-earth-metal atoms is pursued in an effort to develop an improved optical time standard [9–11] and the possibility of the creation of ultracold Rydberg atoms and molecules is enticing [12]. Cooling and trapping atoms with large magnetic moments such as dysprosium ($10 \mu_B$) or chromium ($6 \mu_B$) is of particular interest. It was predicted that polar species may form unique phases of matter at very low temperatures [13] and highly magnetic atoms may be used in schemes for quantum computation [14].

The possibility of cooling Cr atoms to ultracold temperatures was explored in two experiments. Initially, Doyle and co-workers [15] found that inelastic Zeeman relaxation in Cr-Cr collisions induced by the anisotropy of the magnetic dipole-dipole interaction precludes the evaporative cooling of $6 \mu_B$ atoms in a purely magnetic trap. An alternative approach of Pfau and co-workers [16–18] based on evaporative cooling in a magneto-optical trap (MOT) was able to circumvent the problem, and the creation of Cr BEC has been reported [19]. The stability and properties of the Cr BEC depend on details of Cr - Cr interactions. Pfau and coworkers found that Cr-Cr collisions at a temperature of $6 \mu K$ are influenced by a number of magnetic Feshbach resonances [20]. A simple interaction potential model was used in a nonlinear least-squares-fitting procedure to identify the observed resonances [20]. These resonances can be used to tune the Cr-Cr scattering length or to form ultracold Cr_2 molecules by varying magnetic field, as was realized with alkali-metal atoms [3–5].

In this work, we simultaneously address the two distinct experiments on Cr. In the experiment with the optical dipole trap [20], there are no two-body losses because the initial channel is the lowest-energy high-field-seeking state. In the other experiment with the magnetic trap [15], the dominant losses are due to two-body spin-changing collisions because the initial channel is the the highest-energy low-field-seeking state. We have recently computed accurate interaction potentials for the Cr_2 molecule in the lowest seven electronic states [21]. The dynamics of Cr - Cr collisions in the MOT is determined by the manifold of the interaction potentials and the magnetic dipole-dipole interaction that induces coupling between different adiabatic states. Here, we employ the interaction potentials and the spin-spin coupling to compute Feshbach resonances occurring in collisions of trapped Cr atoms. The comparison between the calculation and the experimental measurement provides a sensitive probe of the accuracy of our interaction potentials that can be used for understanding the prospects for Cr_2 molecule formation in the Cr BEC, or for studies of higher-energy collisions. We explore the sensitivity of the Feshbach resonance parameters to details of the interaction potentials. To discuss the stability of Cr samples in a magnetic trap, we present calculations of Zeeman relaxation in Cr - Cr collisions.

The total Hamiltonian for two Cr atoms in a homogeneous magnetic field can be written in atomic units as

$$\hat{H} = -\frac{1}{2\mu R}\frac{\partial^2}{\partial R^2}R + \frac{\boldsymbol{l}^2}{2\mu R^2} + \hat{V}_{es} + \hat{V}_{dip}$$
$$+ 2\mu_0 \boldsymbol{S}_A \cdot \boldsymbol{B} + 2\mu_0 \boldsymbol{S}_B \cdot \boldsymbol{B}, \qquad (1)$$

where μ is the reduced mass of the chromium dimer molecule, R is the interatomic separation, $\boldsymbol{l}$ is the rotational angular momentum of the nuclei, $\hat{V}_{es}$ is the operator of the electrostatic nonrelativistic interaction, $\hat{V}_{dip}$ is the operator of the magnetic dipolar interaction discussed below, $\boldsymbol{S}_A$ and $\boldsymbol{S}_B$ are the electronic-spin momenta of the two Cr atoms (labeled A and B), $\boldsymbol{B}$ is the vector of the magnetic field, and μ_0 is the Bohr magneton. The vector sum of $\boldsymbol{S}_A$ and $\boldsymbol{S}_B$ gives the total spin S of the Cr dimer. We assume that the magnetic field

does not vary substantially on the scale of the collision length. If the quantization axis z is chosen in the direction of the magnetic field vector, the last two terms in the Hamiltonian (1) can be rewritten in terms of the projection M_S of the total spin on the magnetic-field vector as $2\mu_0 B M_S$. The interaction with the magnetic field thus splits the energy levels of the Cr_2 molecule with different values of M_S. The total angular momentum defined by the vector sum of S and l is not conserved, but its projection, $M = M_S + m_l$, where m_l is the projection of l on the magnetic-field axis, remains a good quantum number. The $\hat{V}_{es}$ operator is defined in terms of the Born-Oppenheimer interaction potentials $V_S(R)$ of the Cr_2 molecule as follows:

$$\hat{V}_{es} = \sum_S \sum_{M_S} |SM_S\rangle V_S(R) \langle SM_S|. \qquad (2)$$

We expand the total wave function in terms of products of τ_{SM_S}, the eigenfunctions of S^2 and S_z, and Y_{lm_l}, the eigenfunctions of l^2 and l_z,

$$\Psi = \sum_S \sum_{M_S} \sum_l \sum_{m_l} F_{SM_S l m_l}(R) \tau_{SM_S} Y_{lm_l}(\hat{R}), \qquad (3)$$

and write the dipolar interaction operator as [22]

$$\hat{V}_{dip} = -\sqrt{\frac{24\pi}{5}} \frac{\alpha^2}{R^3} \sum_{q=-2}^{2} (-1)^q Y_{-q}^{(2)} [S_A \otimes S_B]_q^{(2)}, \qquad (4)$$

where α is the fine-structure constant. Substitution of the expansion (3) in the Schrödinger equation leads to a system of coupled differential equations for the expansion coefficients $F_{SM_S l m_l}(R)$ parametrized by the coupling matrix that comprises the electrostatic $\hat{V}_{es}$ and magnetic dipolar $\hat{V}_{dip}$ interactions. The matrix of the $\hat{V}_{es}$ operator is diagonal in the basis (3) and the matrix of the dipolar interaction (4) can be evaluated analytically as demonstrated, for example, in Refs. [22,23]. The solutions of the coupled differential equations with the appropriate scattering boundary conditions [23] yield the scattering matrix.

The elastic and inelastic cross sections are computed from the S matrix as shown in Refs. [22,23] and earlier in the work of Bohn [24]. We consider collisions of bosonic ^{52}Cr atoms with zero nuclear spin so the basis sets in Eq. (3) are constrained by the symmetry requirement $S + l =$ even. Also, the spin-spin operator in Eq. (4) is a tensor of rank 2, which further restricts the states to even S and l as the initial collision channel has quantum numbers $S = 6$, $M_S = 6$, or $M_S = -6$, depending on whether the initial channel is the lowest- or the highest-energy state. In addition, we found that neglecting the basis states with $M_S < 4$ in the close-coupling expansion (3) did not change the results for Zeeman relaxation of maximally stretched Cr atoms. The resulting basis set included only states that preserve the atomic spin or allow spin to change by one.

The positions and widths of Feshbach resonances can be obtained from an analysis of the S matrix as described by Ashton *et al.* [25]. In the vicinity of a Feshbach resonance, the magnetic-field dependence of the scattering length is given by $a(B) = a_S[1 - \Gamma/(B - B_{res})]$, where Γ is the width of

TABLE I. Calculated resonance positions (B_{res}) and widths (Γ) in comparison with the experimental findings [20]. The closed channels are labeled by $|S, M_S, l, m_l\rangle$. The molecular potential parameters are: $C_6 = 745$, $a_6 = 102.8$, $a_4 = 53.8$, $a_2 = -18.5$ a.u. The entrance channel corresponds to $S = 6$, $M_S = -6$, $l = 0$, $m_l = 0$; $\Delta B_{res} = B_{res}^{(calc)} - B_{res}^{(exp)}$, $\Delta\Gamma = \Gamma^{(calc)} - \Gamma^{(exp)}$.

$|S,M_S,l,m_l\rangle$	$B_{res}^{(calc)}$(G)	ΔB_{res}(G)	$\Gamma^{(calc)}$(mG)	$\Delta\Gamma$(mG)
$|6,-2,4,-4\rangle$	50.1	0.0	0.00024	
$|6,-3,4,-3\rangle$	64.9	-0.2	0.0025	-0.0035
$|6,-4,4,-2\rangle$	98.6	-0.3	0.16	-0.14
$|4,-2,4,-4\rangle$	143.2	-0.7	0.026	-0.094
$|4,-3,4,-3\rangle$	187.8	-0.5	0.11	-0.11
$|6,-5,4,-1\rangle$	206.0	0.2	12	0
$|4,-4,4,-2\rangle$	288.7	2.1	0.54	-11.46
$|6,-4,2,-2\rangle$	295.6	5.3	56	5
$|2,-2,4,-4\rangle$	379.2	0.0	0.31	-0.11
$|4,-4,2,-2\rangle$	506.9	7.0	83	2
$|6,-5,2,-1\rangle$	600.5	11.4	1590	-110

the resonance and a_S is the zero-field scattering length corresponding to the potential with total spin S [26].

Pfau and co-workers trapped Cr atoms in a MOT in the lowest Zeeman level $M_S = -6$ [20] and measured a number of Feshbach resonances, arising from the coupling of the $M_S = -6$ state to higher Zeeman levels with larger M_S values. This coupling is mediated by the magnetic dipolar interaction. We have computed several of the resonances that may occur in ultracold collisions of Cr atoms initially in the $M_S = -6$ state in a weak magnetic field (see Tables I–III). Our results for the positions and widths of the resonances are in good agreement with the experimental findings [20]. These resonances are determined by the positions of the least bound energy levels in the $S = 6$, $S = 4$, and $S = 2$ molecular potentials, which also sensitively affect the corresponding values of the scattering lengths. The scattering length is determined by the parameters of the long-range interatomic interaction and the

TABLE II. Same as in Table I, but for the potential parameters: $C_6 = 770$, $a_6 = 102.5$, $a_4 = 53.6$, $a_2 = -22.4$ a.u.

$|S,M_S,l,m_l\rangle$	$B_{res}^{(calc)}$(G)	ΔB_{res}(G)	$\Gamma^{(calc)}$(mG)	$\Delta\Gamma$ (mG)
$|6,-2,4,-4\rangle$	50.1	0.0	0.00024	
$|6,-3,4,-3\rangle$	64.9	-0.2	0.0024	-0.0036
$|6,-4,4,-2\rangle$	98.5	-0.4	0.16	-0.14
$|4,-2,4,-4\rangle$	143.0	-0.9	0.025	-0.095
$|4,-3,4,-3\rangle$	187.6	-0.7	0.11	-0.11
$|6,-5,4,-1\rangle$	205.6	-0.2	12	0
$|4,-4,4,-2\rangle$	288.0	1.4	1.53	-10.47
$|6,-4,2,-2\rangle$	293.0	2.7	53	2
$|2,-2,4,-4\rangle$	379.2	0.0	0.29	-0.13
$|4,-4,2,-2\rangle$	503.8	3.9	81	0
$|6,-5,2,-1\rangle$	595.0	5.9	1530	-170

TABLE III. Predicted resonance positions (B_{res}), with $l=4$ and $l=6$ nuclei rotational angular momenta, see caption of Table II for the potential parameters. The resonances are listed in the order of increasing magnetic field.

$|S,M_s,l,m_l\rangle$	$B_{res}^{(calc)}$(G)
2,0,6,−6	116.0
2,−1,6,−5	133.1
2,−2,6,−4	167.4
6,0,6,−6	340.7
6,−1,6,−5	402.0
6,−2,6,−4	496.4
4,0,6,−6	543.4
0,0,6,−6	556.4
4,−1,6,−5	645.5
6,−3,6,−3	659.7
6,−2,4,−4	786.9
4,−2,6,−4	801.9
2,0,6,−6	964.9
6,−4,6,−2	992.4

shape of the molecular potential such as the position of the classical inner turning point. It is possible to tune the scattering length by varying the strength of either the short-range repulsion or the long-range attraction.

To obtain a better agreement with the experimental measurements, we modified slightly the position of the classical inner turning point and the long-range interaction C_6 coefficient of the original $S=6$, $S=4$, and $S=2$ molecular potentials. For each curve, the position of the *ab initio* data points at separations less than the equilibrium distance R_e was shifted according to $R_{shifted}=R+s(R-R_e)/(R_c-R_e)$, where s corresponds to the shift of the zero-energy classical inner turning point R_c. The shifted data points were joined smoothly to the rest of the data and the modified long-range tail. Table II shows that the results obtained with $C_6=770$ a.u. are in better agreement with the measured data. This is consistent with our earlier suggestion that the previously determined value of $C_6=745$ a.u. is a lower limit on the C_6 coefficient computed with an estimated accuracy of 50 a.u. [21]. Based on the results of Table II, we recommend a new value of $C_6=770$ a.u. for the Cr-Cr interaction potentials. The scattering lengths we have obtained with our potentials are in harmony with the experimental measurements [20]. We note that the calculated resonances in Tables I and II correspond to $l=0$ in the collision channel. Reference [20] reported two narrow resonances which we have also identified as belonging to the $l=2$ collision channels and one other resonance that we, as well as Ref. [20], have not yet identified.

In Table III, we give our predicted positions for a number of resonances that form in the $l=4$ and $l=6$ energetically closed channels whose widths are expected to be extremely narrow. These resonances result from bound states in the gerade molecular potentials with spin multiplicities 1, 5, 9, and 13.
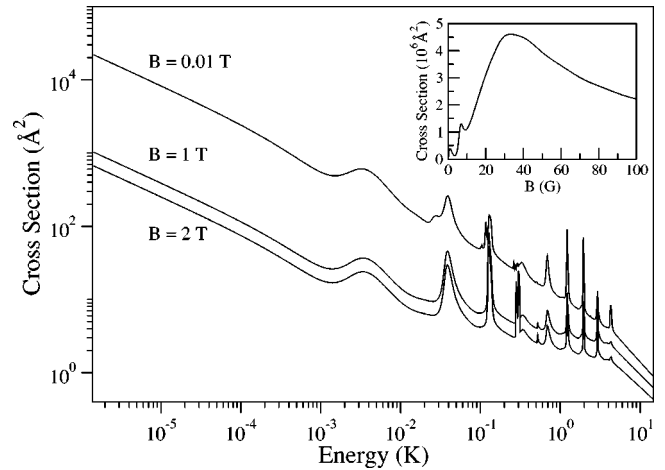

FIG. 1. Zeeman relaxation cross sections in collisions of two Cr atoms initially in the $M_S=+6$ state. Inset: Zeeman relaxation at the collision energy 1.4×10^{-10} K, 1 K=0.695 cm^{-1}.

The magnetic moment of Cr is six times larger than that of alkali-metal atoms, making it easier to trap Cr in a magnetic trap [15]. Magnetic traps confine atoms in low-field-seeking states or the Zeeman levels with a positive gradient of energy with respect to the magnetic field. The stability of trapped atomic samples is determined by the efficiency of the Zeeman relaxation in atom-atom collisions. The potentials of the Cr$_2$ molecule with the different spin values are significantly split at finite interatomic separation (see Fig. 1 in Ref. [21]). This suggests that there is substantial spin anisotropy [27] in Cr - Cr collisions and the Zeeman relaxation of Cr atoms in all but maximally stretched states must occur rapidly. The Zeeman relaxation of Cr atoms in the maximally stretched $M_S=+6$ state is driven by the magnetic dipolar interaction. It was found in the experimental measurements that, because of the large magnetic moment of Cr, the dipolar Zeeman relaxation is also quite efficient in collisions of maximally stretched Cr atoms at temperatures between 0.01 and 1 K [15]. The experiment could not be extended to lower temperatures due to technical difficulties. In order to understand the prospects for evaporative cooling of large-Bohr-magneton atoms such as Cr at ultralow temperatures in a magnetic trap, it is necessary to analyze the efficiency of the Zeeman relaxation as a function of the magnetic-field strength and temperature.

Figure 1 presents the energy dependence of cross sections for the Zeeman relaxation of Cr atoms in the $M_S=+6$ state at three experimental magnetic-field strengths. Several observations are in order: the cross sections increase with decreasing collision energy, increase with decreasing magnetic field, and display several shape resonances whose positions are independent of the magnetic-field strength. The latter indicates that the elastic scattering is not affected by the magnetic field.

The magnetic-field dependence of the cross sections at ultracold collision energies is surprising. The cross section for the Zeeman (or projection-changing) transitions vanishes at ultracold collision energies in the absence of a magnetic field [28]. In a finite magnetic field, the cross sections for the Zeeman relaxation tend to infinity as the collision energy

vanishes and they must therefore increase with the field strength at low magnetic fields [29]. Zeeman transitions are accompanied by orbital angular momentum transfer and the centrifugal barrier in the final channel impedes the outgoing collision wave packet at low magnetic fields, thereby suppressing the transition probability. Increasing the magnetic-field strength leads to larger energy separation between the Zeeman levels, and the role of the centrifugal barrier in the outgoing channel is mitigated at high magnetic fields. Increasing the magnetic-field strength at high magnetic fields should somewhat decrease the Zeeman relaxation cross section due to the energy-gap law [30]. It should thus be expected that the Zeeman relaxation cross sections quickly increase with the magnetic-field strength at low fields, pass through a maximum, and decrease at high magnetic fields. The maximum in the magnetic-field dependence of the Zeeman relaxation cross sections in typical atomic and molecular systems occurs at magnetic fields on the order of 1–2 T [30].

The results in Fig. 1 show that the Zeeman relaxation cross section in Cr - Cr collisions decreases monotonously in the magnetic-field interval 0.01–2 T. Calculations with lower magnetic fields (see the inset of Fig. 1) demonstrate that the Zeeman relaxation cross section does follow the anticipated trend; however, the maximum in the cross section occurs at about 40 G. Apparently, the large mass of Cr reduces the magnitude of the centrifugal barrier in the outgoing channel

and the field strength of as little as 40 G is enough to mitigate the role of the orbital angular momentum in the final Zeeman level. We conclude that this should be characteristic of most heavy atoms with zero hyperfine interaction.

In summary, we have presented accurate calculations of Feshbach resonances occurring in ultracold collisions of optically trapped Cr atoms in the lowest-energy Zeeman level and cross sections for Zeeman relaxation of magnetically trapped Cr atoms in the highest-energy Zeeman level. It was found that calculations with the revised value of the long-range interaction coefficient $C_6 = 770$ a.u. are in better agreement with the experimental measurements of the Feshbach resonances. The Zeeman relaxation cross sections were found to increase as both the collision energy and the magnetic-field strength decrease to very low values. This trend may inhibit the evaporative cooling of heavy atoms with large magnetic moments in a magnetic trap.

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