Ultracold collisions of fermionic OD radicals

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We discuss consequences of Fermi exchange symmetry on collisions of polar molecules at low temperatures (below 1 K), considering the OD radical as a prototype. At low fields and low temperatures, Fermi statistics can stabilize a gas of OD molecules against state-changing collisions. We find, however, that this stability does not extend to temperatures high enough to assist with evaporative cooling. In addition, we establish that a novel "field-linked" resonance state of OD dimers exists, in analogy with the similar states predicted for bosonic OH.

I. INTRODUCTION

Cooling and trapping molecules in their rovibrational ground states has proven to be a daunting experimental task, yet now it has been achieved [1–3]. Currently, the samples produced by Stark slowing are limited to temperatures around 1 mK, which is cold enough to trap, but not yet cold enough for interesting applications to novel dilute quantum gases of fermions [4–8] or bosons [9–14]. To produce colder, denser samples for these applications, an attractive approach may be to use sympathetic cooling with an easily cooled species (such as Rb) [15] or else evaporative cooling. Knowledge of collision cross sections is therefore essential in understanding prospects for the success of either approach.

We have previously considered electrostatic trapping of polar Π-state molecules from the point of view of stability with respect to collisions [16]. The main bottleneck here is that *electrostatic* trapping requires the molecules to be in a weak-field-seeking state, in which case the molecules of necessity have an even lower-energy strong-field seeking state. Collisions involving the strong and anisotropic dipole-dipole interaction between molecules appear more than adequate to drive the molecules into these unfavorable states, leading to unacceptably high trap loss and heating. For this reason, it may be necessary to seek alternative methods that can confine the strong-field-seeking, absolute lowest-energy ground state of the molecule using time-varying electric fields [17–20]. A recent proposal for an *electrodynamic* trap is based on the microwave analog of the familiar far-off-resonant optical dipole trap—only the microwave version does not have to be far off resonance, making the trap very deep [20].

Polar fermions may have an important advantage for electrostatic trapping: namely, low inelastic rates at cold temperatures. Kajita has discussed state-changing collisions of dipolar fermionic molecules [21], based on the well-known Wigner threshold laws for dipolar interactions. Namely, elastic scattering cross sections are essentially independent of collision energy E at low energies, but state-changing cross sections scale as $E^{1/2}$. Therefore, at "sufficiently low" tem-

peratures, elastic scattering always wins, and evaporative cooling should be possible. Using the Born approximation, Kajita concludes that this is the case for the molecules OCS and CH_3Cl, at reasonable experimental temperatures [21]. This analysis may yet prove too optimistic, since the results include regions where the Born approximation may not be strictly applicable [22]. Still, the idea is a sound one that deserves further exploration.

A complete theoretical description of molecule-molecule scattering is complicated by the complexity of the short-range interaction between molecules. Indeed, for open-shell molecules the potential energy surface is difficult to compute by *ab initio* methods and remains inadequately known. It is therefore worthwhile to seek situations in which the influence of short-range physics is minimal. It appears that for weak-field-seeking states the influence of the short-range potential is weak, owing to avoided crossings in the long-range interaction [16]. For collisions of identical fermionic molecules, the influence of short-range physics may be even smaller, since only partial waves with $l \geq 1$ are present, and there is centrifugal repulsion in all scattering channels.

A main aim of the present paper is thus to explore the suppression of inelastic collisions in fermionic Π-state molecules, using the OD radical as an example. This is an illustrative choice of molecule, since we have studied its bosonic counterpart (OH) extensively in the past [16,23,24]. It is also a species at the center of current experimental interest [3,25]. To this end we employ full close-coupling calculations to a model of the OD-OD interaction that includes only the dipolar part. We find, as we must, that the fermionic threshold laws ultimately favor elastic over inelastic scattering at low temperatures. For OD, however, we find that the energy scales for this to happen remain quite low, on the order of microkelvins or below, so that the usefulness of this result to evaporative cooling remains questionable.

On the bright side, the suppression of inelastic collisions does mean that a gas that is already cold will be stable under collisions, even in an electrostatic trap. This is a similar conclusion to one we have drawn in the past for magnetostatic trapping of spin-polarized paramagnetic (nonpolar) species [26,27]. This is useful for cold collisions studies, since it is

believed that collisions of weak-electric-field seekers are dominated by, and can be understood in terms of, purely long-range dipolar forces [23]. In particular, such collisions are predicted for bosons to have long-range resonant states, termed field-linked resonances, which may be useful in understanding cold collisions. A second goal of this paper is to verify that the fermionic OD molecules also possess these resonances.

II. THRESHOLD LAWS IN THE BORN APPROXIMATION

Although we will compute numerically exact cross sections within our model, we are especially interested in the behavior of cross sections at low collision energies. To this end, in this section we summarize the main results relevant to the energy dependence of cross sections, using the first Born approximation to make the mathematics transparent. Similar arguments are presented in Refs. [9,21], but we note that the general subject of threshold laws for various power-law long-range potentials has a long and rich history [28–41].

A first point to be considered is why the Born approximation should be of any use at all, since it is ordinarily associated with collisions of "fast" particles. Strictly speaking, however, the Born approximation is valid when the potential responsible for scattering is suitably "weak," meaning that the true scattering wave function is well approximated by the unperturbed wave function. For dipolar scattering, the argument is as follows. Consider elastic scattering in a single channel whose long-range potential varies as $1/R^s$. Then, in partial wave l, the elastic scattering phase shift will vary with wave number k as [40]

$$\delta_l \sim \alpha k^{2l+1} + \beta k^{(s-1)}, \qquad (1)$$

where α and β are constants depending on details of the potential.

Thus for a dipolar potential with $s=3$, the second term in Eq. (1) is the dominant contribution to the phase shift for any partial wave $l \geq 1$, yielding $\delta_l \sim k$. (Moreover, the $l=0$ contribution to a realistic dipole-dipole interaction rigorously vanishes by symmetry.) It can be shown (for example, using the JWKB approximation [40]) that the second contribution in Eq. (1) arises from purely long-range physics—i.e., for intermolecular separations outside the centrifugal barrier imposed by the partial wave. As the collision energy approaches threshold, this distance gets ever larger, and the influence of the $1/R^3$ perturbing potential gets ever weaker. Thus, near threshold, the wave function is well approximated by unperturbed spherical Bessel functions in each partial wave and the Born approximation can be used.

We adopt this view in the multichannel case. Because the dipole-dipole interaction is anisotropic, different partial waves are coupled together. Nevertheless, the diagonal pieces of the Hamiltonian matrix have the general form

$$\frac{\hbar^2 l(l+1)}{2\mu^2 R^2} + \frac{C_3^{\text{eff}}}{R^3}, \qquad (2)$$

where R is the distance between molecules, μ is their reduced mass, and the effective C_3 coefficient depends, in general, on the channel as well as on the degree of electric field polarization (see Ref. [16]). When C_3^{eff} is negative, the potential (2) presents a finite barrier of height

$$E_b = \frac{4}{27} \left[\frac{\hbar^2 l(l+1)}{2\mu} \right]^3 \frac{1}{(C_3^{\text{eff}})^2}. \qquad (3)$$

For energies E considerably less than E_b, scattering only occurs from outside the barrier (barring resonances [9]), thus setting an energy scale for the utility of the Born approximation. To make an estimate of this energy scale, consider the strong-field limit, where polarized molecules have $C_3 \sim d^2$, the square of the dipole moment. For OD, this sets the relevant p-wave centrifugal barrier height at ~ 10 nK. At higher energies, the incoming wave spills over the barrier, samples smaller-R interactions, and is no longer well described as a plane wave.

Assuming that the Born approximation holds, we proceed as follows. The partial scattering cross section for a collision process entering on channel i and exiting on channel f is given in terms of the transition matrix T by

$$\sigma_{if} = \frac{\pi}{k_i^2} |\langle i|T|f \rangle|^2, \qquad (4)$$

where the channel indices i and f include partial-wave contributions l_i and l_f, which need not be the same. In the first Born approximation, the T-matrix elements are given by the matrix elements of the potential (Chap. 7 of Ref. [42], where we have reinserted the dimensionful factors)

$$\langle i|T|f \rangle = 2 \left(\frac{2\mu}{\hbar^2} \right) (k_i k_f)^{1/2} \int_0^\infty R^2 dR j_{l_i}(k_i R) \frac{C_3(l_i l_f)}{R^3} j_{l_f}(k_f R). \qquad (5)$$

Here $C_3(l_i l_f)$ represents the appropriate off-diagonal coupling matrix element, which, again, depends on field.

For *elastic* scattering, where the initial and final wave numbers are equal, $k_f = k_i$, we can rewrite Eq. (5) in terms of the dimensionless variable $x = k_i R$:

$$\langle i|T|f \rangle = \frac{4\mu C_3(l_i l_f)}{\hbar^2} k_i \int_0^\infty dx \frac{j_{l_i}(x) j_{l_f}(x)}{x}. \qquad (6)$$

The integral in Eq. (6) converges whenever $l_i + l_f > 0$ and is moreover independent of k_i. Therefore, for any elastic scattering process by dipolar forces that changes l by at most 2 units, $T \sim k_i$ at low energies, and by Eq. (4), the associated cross section is independent of collision energy. In particular, the elastic scattering cross section of identical fermions does not vanish, if they interact via dipolar forces.

For completeness, we give the value of the integral. This is found by substituting ordinary Bessel functions for the spherical Bessel functions, $j_n(x) = \sqrt{\pi/2x} J_{n+1/2}(x)$, and using standard formulas for integrals [43]:

$$\int_0^{\infty} dx \frac{j_{l_i}(x) j_{l_f}(x)}{x}$$

$$= \frac{\pi \Gamma(\frac{l_i + l_f}{2})}{8 \Gamma(\frac{-l_i + l_f + 3}{2}) \Gamma(\frac{l_i + l_f + 4}{2}) \Gamma(\frac{l_i - l_f + 3}{2})}. \quad (7)$$

For an *exothermic* process, with $k_f > k_i$, a similar argument yields, for the transition amplitude,

$$\langle i|T|f\rangle = \pi \frac{2\mu C_3(l_i l_f)}{\hbar^2} \int_0^{\infty} dR J_{l_i+1/2}(k_i R) J_{l_f+1/2}(k_f R) R^{-2}. \quad (8)$$

This integral, too, can be done as long as $l_i + l_f > 0$ [43]:

$$\int_0^{\infty} dR J_{l_i+1/2}(k_i R) J_{l_f+1/2}(k_f R) R^{-2}$$

$$= \frac{k_i^{l_i+1/2} \Gamma(\frac{l_i+l_f}{2})}{4 k_f^{l_i-1/2} \Gamma(\frac{-l_i+l_f+3}{2}) \Gamma(l_i+3/2)}$$

$$\times F\left(\frac{l_i+l_f}{2}, \frac{l_i-l_f-1}{2}, l_i+3/2; \left(\frac{k_i}{k_f}\right)^2\right), \quad (9)$$

where F stands for a hypergeometric function.

Near threshold in an exothermic process, we have $k_f \gg k_i$. In this case the leading-order term of the hypergoemetric function F is a constant, and the only remaining dependence of Eq. (9) on k_i is in its prefactor. Thus $T \sim k_i^{l_i+1/2}$, and $\sigma \sim k_i^{2l_i-1} \sim E^{l_i-1/2}$. When the incident partial wave is $l_i=0$, as would be the case for identical bosons, the inelastic scattering cross section diverges at threshold. For any higher partial wave—say, the $l_i=1$ partial wave that dominates scattering of identical fermions—the inelastic cross section instead vanishes in the threshold limit.

III. COLLISION CROSS SECTIONS FOR OD

The OD radical differs from OH in two significant ways, for our present purposes: first, its λ-doubling constant is somewhat smaller [44]. Second, its hyperfine structure depends on the nuclear spin of deuterium being 1 instead of 1/2 for hydrogen, meaning that total spin states $f=1/2$, 3/2, and 5/2 are possible in the $^2\Pi_{3/2}$ electronic ground state of OD (as in our OH work, we consider exclusively the electronic ground state and neglect excited vibrational and rotational levels). Figure 1 presents the Stark effect for OD, which can be compared to the similar figure for OH, Fig. 1 of Ref. [16]. Note that, due to the smaller λ-doublet in OD, this radical enters the linear Stark regime at applied electric fields of ~ 200 V/cm, as opposed to ~ 1000 V/cm in OH.

We consider collisions of the highest-energy weak-field-seeking state in Fig. 1, with quantum numbers $|f, m_f, \text{parity}\rangle = |5/2, 5/2, f\rangle$. The details of our scattering
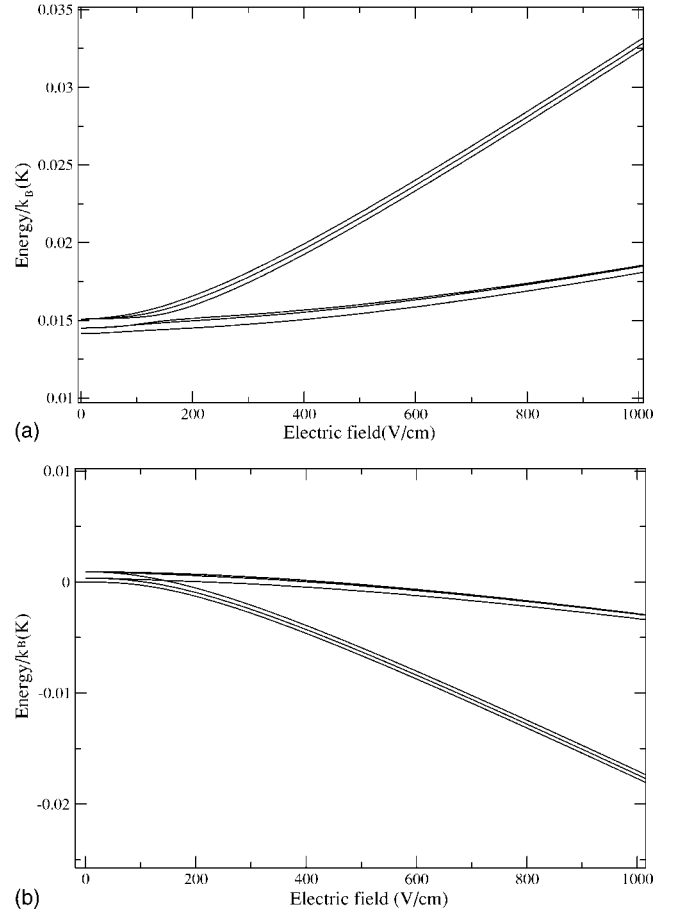

FIG. 1. Stark effect for the $^2\Pi_{3/2}|f, m_f, \text{parity}\rangle$ ground states of OD. Shown are state of f parity (a) and e parity (b).

theory have been presented elsewhere, for OH [16]. The main difference in handling OD is to incorporate Fermi exchange symmetry, which amounts to changing plus signs to minus signs in Eq. (17) of Ref. [16]. Otherwise, we treat the scattering in the same way, by including only the Stark and dipole-dipole interactions, along with the hyperfine structure. We then numerically solve the coupled-channel Schrödinger equations for scattering, to yield cross sections. We find that cross sections are relatively insensitive to the details of the short-range potential, as was also the case for OH in weak-electric-field seeking states [16]. This insensitivity arises from a set of avoided crossings in the long-range potentials; see below.

Figure 2 shows the main scattering results, as collision cross sections versus collision energy at different electric fields. Three different applied electric fields are indicated by color coding: $\mathcal{E}=0$ (black), $\mathcal{E}=100$ V/cm (red), and $\mathcal{E}=1000$ V/cm (blue). In each case, the solid line denotes elastic scattering, while the dashed line represents total inelastic scattering to channels where one or both molecules loses internal energy. In zero field, the molecules are completely unpolarized, and the dipole-dipole interaction vanishes. Thus the cross sections obey the familiar Wigner threshold laws for short-ranged interactions between fermions: the elastic cross section $\sigma_{el} \propto E^2$, whereas the (exothermic) inelastic cross section $\sigma_{inel} \propto E^{1/2}$ [40]. Thus in zero field elastic scat-
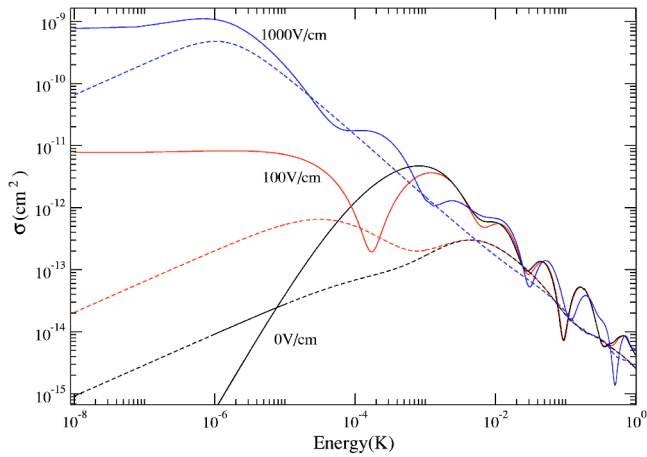
FIG. 2. Elastic (solid lines) and total inelastic (dashed lines) cross sections for several values of electric field: $\mathcal{E}=0$, 100, 1000 V/cm.

tering is actually *less* likely than inelastic scattering at lower energies (below about 10 μK in this example). Above this energy elastic scattering appears somewhat more favorable than inelastic scattering, at least until several mK, where both cross sections start to hit the unitarity limit.

Turning on the electric field partially polarizes the molecules, so that the dipole-dipole interaction is "activated." Then the dipole-dipole threshold laws take effect, $\sigma_{el} \propto$ const, whereas we still have $\sigma_{inel} \propto E^{1/2}$. Figure 2 illustrates where this threshold behavior kicks in for different electric field values. Notice that the higher the electric field, the lower is the energy where the threshold behavior is attained. This is because the effective C_3 coefficient that determines the barrier height E_b is an increasing function of electric field, at least until it saturates [16]. In particular, for $\mathcal{E}=1000$ V/cm, the molecules are strongly polarized. According to the estimate in Sec. II, we would then expect the threshold laws to hold only up to energies $\sim$10 nK. Figure 2 shows that this is only approximately correct and that deviation from threshold behavior actually begins at several tens of nK.
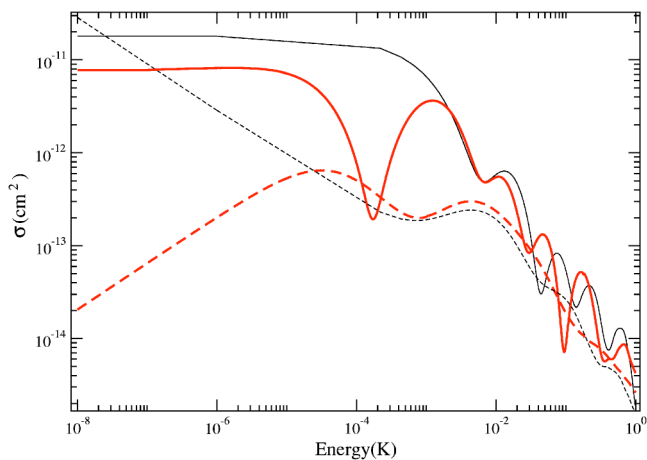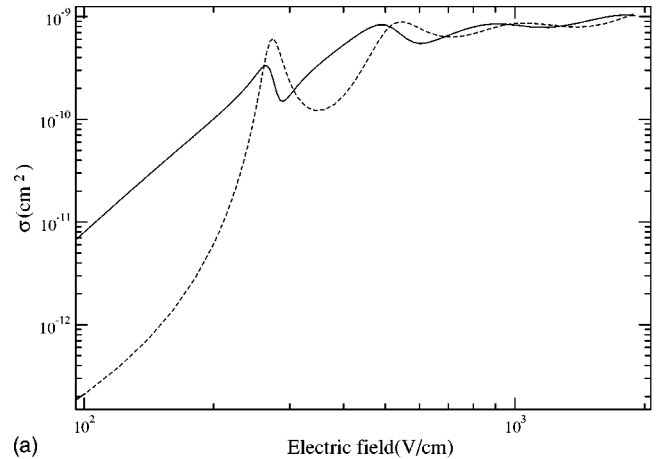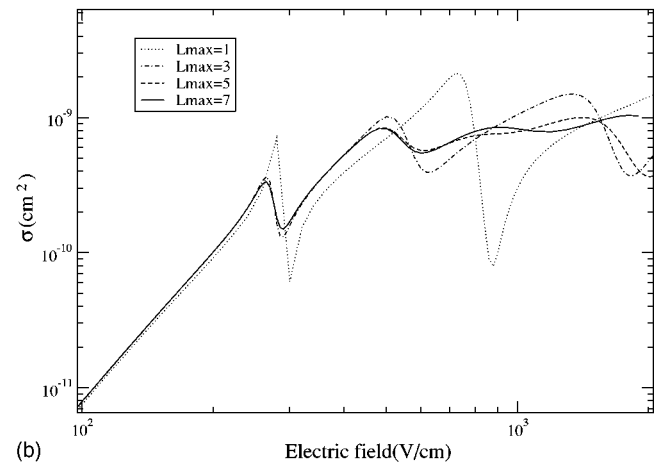

FIG. 3. Comparison of cross sections for OD (red) and OH (black) molecules. Solid and dashed lines refer to elastic and inelastic cross sections, respectively.

FIG. 4. (a) Elastic (solid lines) and inelastic (dashed lines) cross sections for OD scattering as a function of applied electric field. The collision energy is $E=1$ μK. (b) Convergence of the elastic cross section upon increasing the number of partial waves included in the calculation.

On the other hand, a Fermi gas of molecules that is *already cold* will enjoy the benefits of Wigner-law suppression of inelastic collisions. Suppose a quantum degenerate gas of OD could be produced at nK temperatures, as is the case for current experiments in ^{40}K and ^{6}Li. Then Fig. 2 suggests that a small bias field of $\sim$100 V/cm reduces inelastic cross sections to an acceptable level of $\sim 2 \times 10^{-14}$ cm^2, corresponding to a rate constant $\sim 10^{-16}$ cm^3/s.

To emphasize the difference between bosons and fermions, we reproduce the $\mathcal{E}=100$ V/cm cross sections in Fig. 3 (red) along with the corresponding cross sections for OH in the same field (black). It is clear that in both cases elastic scattering (solid lines) is quite similar, whereas the behavior of inelastic scattering is utterly different at low energies for the two species. Equally importantly, at collision energies above about 1 mK, all the cross sections have the same general behavior. This is a manifestation of the strength of the dipolar interactions and the fact that in this energy range all processes are essentially unitarity limited.

IV. QUESTION OF FIELD-LINKED RESONANCES

Finally, we comment on the occurrence of field-linked (FL) resonance states in this system. Figure 4(a) shows the
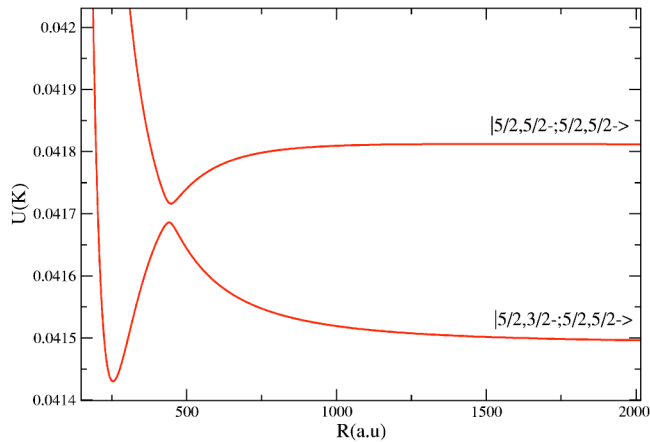
FIG. 5. A set of avoided crossings that generate FL states for OD.

elastic and inelastic cross sections versus electric field, at a fixed collision energy of 1 μK. This figure exhibits the characteristic peaks indicative of field-linked resonances; compare Fig. 2 of Ref. [16]. To converge these results at higher field demands an increasing number of partial waves. Figure 4(b) illustrates the convergence of the resonant scattering cross section for various numbers of partial waves included. For partial waves $L=1, 3, 5, 7$, the cross section is well converged up to several hundred V/cm. This is sufficient to compute the first two resonance states, which are the only well-resolved ones anyway.

As discussed in Ref. [16] and elaborated on in Ref. [24], the FL resonances live in adiabatic potential energy surfaces generated by avoided crossings in the long-range dipole-dipole interaction. The surfaces have a somewhat different character for fermions than for bosons, however. Figure 5 shows a set of adiabatic curves for a single partial wave L $=1$; this is the analogue of Fig. 1 in Ref. [23].

For identical bosons, the long-range attractive part of the relevant curve is predominantly s wave in character, hence has a $1/R^4$ behavior in an electric field. This reflects the fact that the direct dipolar interaction vanishes for s waves and makes an effect only at second order [16]. For fermions in identical spin states, however, the attractive part involves the p-wave interaction, where the dipole is already nonzero, so that the long-range interaction scales as $1/R^3$. The net effect is that the inner turning point of the s-wave FL states approaches smaller R as the field is increased for bosons, but that this inner turning point is relatively fixed for fermions. (The outer turning point is set by the energy of the resonant state relative to the threshold and is thus arbitrarily large.)

These resonant states, if sufficiently stable, may form a novel kind of pair of fermions, which may ultimately lead to an exotic Fermi superfluid state. Unfortunately, as seen in Fig. 4, these resonances are quite readily susceptible to pre-dissociation, indicated by the large inelastic cross sections near resonance. In this they resemble their bosonic counterparts. However, stabilization of cold dipolar gases using magnetic fields has been recently discussed [45]. It is yet conceivable that these resonances could be tamed long enough to put them to use.

In summary, we have computed scattering cross sections for cold collisions of the fermionic free radical OD, as functions of both collision energy and electric field. We find that, similar to the case of bosonic OH, these molecules are unlikely to be stable against collisions in traps warmer than about 10 μK. Unlike OH, however, they will be collisionally stable at lower temperatures, owing to the unique Wigner threshold laws for fermionic polar particles in such a gas.

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