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Ultracold spin-polarized mixtures of $^2\Sigma$ molecules with *S*-state atoms: Collisional stability and implications for sympathetic cooling

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The prospects of sympathetic cooling of polar molecules with magnetically cotrapped alkali-metal atoms are generally considered poor due to strongly anisotropic atom-molecule interactions leading to large spin relaxation rates. Using rigorous quantum scattering calculations based on *ab initio* interaction potentials, we show that inelastic spin relaxation in low-temperature collisions of CaH($^2\Sigma$) molecules with Li and Mg atoms occurs at a slow rate despite the strongly anisotropic interactions. This unexpected result, which we rationalize using multichannel quantum-defect theory, opens up the possibility of sympathetic cooling of polar $^2\Sigma$ molecules with alkali-metal atoms in a magnetic trap and with alkaline-earth-metal atoms in an optical dipole trap.

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The experimental realization of molecular ensembles cooled to temperatures below 1 K has opened up a multitude of fascinating research directions in physics and chemistry [1]. The unique properties of ultracold molecular gases such as their long-range, anisotropic interactions may be used to implement quantum logic gates [2], create and explore novel phases of quantum matter [3], and study nonequilibrium dynamics and many-body localization phenomena [4]. Recent experimental and theoretical work has demonstrated the possibility of controlling chemical reactions in an ultracold gas of KRb molecules by applying external electric fields and confining the reactants in low dimensions [5,6].

A variety of experimental techniques have been developed for cooling and trapping polar molecules [1]. Indirect cooling techniques such as photo- and magnetoassociation are capable of producing high phase-space density gases of ground-state polar molecules, but are currently limited in scope to Σ-state alkali-metal dimers [1]. Alternatively, molecules can be cooled directly via thermal contact with a cryogenic He buffer gas (buffer-gas cooling [1,7]) or molecular beam deceleration [8]. The direct cooling techniques are ideally suited for the production of diverse classes of polar molecules required for specific applications in quantum simulation [1] and precision measurements [9,10]. However, these techniques tend to produce cold ($T > 10$ mK) rather than ultracold molecules, necessitating an additional stage of cooling to reach ultralow temperatures.

Sympathetic cooling is arguably the most straightforward way of cooling molecules below 10 mK by bringing them in thermal contact with a reservoir of ultracold atoms such as the alkali-metal atoms [11–13]. Molecular cooling and trapping experiments typically employ static electromagnetic traps, which confine molecules in their low-field-seeking Zeeman states [7]. Inelastic collisions with buffer-gas atoms lead to spin relaxation and trap loss [14–20], and a large ratio of elastic to inelastic collision rates ($\gamma_c > 100$) is required for efficient cooling [1]. Previous theoretical work has shown that spin relaxation in collisions of alkali-metal atoms with OH [11], NH [12], and NH_3 [13] is extremely fast ($\gamma_c \sim 1$). Based on these results, the alkali-metal atoms were judged unsuitable as collision partners for sympathetic cooling of molecules, and attention turned to the alkaline-earth-metal [15] and rare-gas [16] atoms, which lack magnetic moments and are thus challenging to accumulate in a magnetic trap. Spin-polarized nitrogen atoms have favorable collisional properties with NH molecules [19,20] but their use in sympathetic cooling experiments is limited by a lack of efficient detection techniques.

In this Rapid Communication, we explore the possibility of sympathetic cooling of diatomic molecules with one unpaired electron ($^2\Sigma$ molecules) by collisions with spin-polarized alkali-metal atoms in a magnetic trap and with alkaline-earth-metal atoms in an optical dipole trap. Low-temperature collisional properties of $^2\Sigma$ molecules have received much less attention than those of $^3\Sigma$ molecules [12,15,17–21]. Using converged quantum scattering calculations based on *ab initio* interaction potentials, we demonstrate that spin relaxation in collisions of CaH($^2\Sigma$) molecules with Li and Mg atoms occurs slowly despite the fact that Li-CaH and Mg-CaH interactions are extremely strong and anisotropic. To explain this surprising result, we map out the dependence of the inelastic cross section on the potential anisotropy using multichannel quantum defect theory. Our analysis shows that spin relaxation in low-temperature collisions of $^2\Sigma$ molecules can be slow even in systems with strongly anisotropic interactions such as Li-CaH, suggesting that sympathetic cooling of $^2\Sigma$ molecules with alkali-metal atoms in a magnetic trap is likely to be successful. While evidence for low inelastic collision rates has been reported recently for Li-NH [21], our work demonstrates favorable prospects for sympathetic cooling in a strongly anisotropic atom-molecule collision system based on a fully converged quantum scattering calculation.

The Hamiltonian of the collision complex formed by a $^2\Sigma$ molecule A and an *S*-state atom B is ($\hbar = 1$) [22,23]

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$$\hat{H} = -\frac{1}{2\mu R}\frac{\partial^2}{\partial R^2}R + \frac{\hat{l}^2}{2\mu R^2} + \sum_{S,\Sigma} V_{S\Sigma}(R,r,\theta)|S\Sigma\rangle\langle S\Sigma| + \hat{H}_{\mathrm{A}} + \hat{H}_{\mathrm{B}} - \sqrt{\frac{24\pi}{5}}\frac{\alpha^2}{R^3}\sum_q Y^\star_{2q}(\hat{R})[\hat{S}_{\mathrm{A}}\otimes\hat{S}_{\mathrm{B}}]^{(2)}_q, \quad (1)$$

where μ is the reduced mass, R is the atom-molecule separation, r is the internuclear distance, θ is the angle between the Jacobi vectors $\mathbf{R}$ and $\mathbf{r}$, $\hat{l}$ is the orbital angular momentum for the collision, $V_{S\Sigma}(R,r,\theta)$ is the potential energy surface (PES) for the atom-molecule interaction, and $\hat{S} = \hat{S}_{\mathrm{A}} + \hat{S}_{\mathrm{B}}$ is the total spin. The Hamiltonian of the $^2\Sigma$ molecule is given by $\hat{H}_{\mathrm{A}} = B_e\hat{N}^2 + \gamma\hat{N}\cdot\hat{S}_{\mathrm{A}} + 2\mu_0 B\hat{S}_{\mathrm{A}z}$, where $\hat{N}$ is the rotational angular momentum of the molecule, B is the magnetic field strength, $\hat{S}_{\mathrm{A}z}$ is the projection of molecular spin $\hat{S}_{\mathrm{A}}$ on the field axis, B_e is the rotational constant, γ is the spin-rotation constant [24], and μ_0 is the Bohr magneton. The atomic Hamiltonian is given by $\hat{H}_{\mathrm{B}} = 2\mu_0 B\hat{S}_{\mathrm{B}z}$ and the term proportional to R^{-3} represents the magnetic dipole interaction. Both of these terms are absent for the Mg atom, which bears no magnetic moment. In this work, we are interested in collisions of CaH molecules with Li atoms initially in their fully spin-polarized Zeeman states $M_{S_{\mathrm{A}}} = M_{S_{\mathrm{B}}} = 1/2$, where $M_{S_{\mathrm{A}}}$ and $M_{S_{\mathrm{B}}}$ are the projections of $\hat{S}_{\mathrm{A}}$ and $\hat{S}_{\mathrm{B}}$ on the magnetic field axis. We can therefore neglect the weak magnetic dipole and spin-rotation couplings between basis states of different S [19] and include only the $S = 1$ (triplet) Li-CaH PES in scattering calculations.

The interaction PESs for Li-CaH ($S = 1$) and Mg-CaH ($S = 1/2$) were evaluated *ab initio* [25] using a highly correlated open-shell coupled cluster CCSD(T) method and large quadruple-ζ correlation-consistent basis sets augmented by atomic-centered diffuse and R-centered bond functions. To study the dependence of the Li-CaH interaction energy on the CaH stretching coordinate r, we calculated the PES over the range of $r = 3.0a_0$ to $5.7a_0$. We found that the interaction potential becomes slightly deeper with increasing r, but retains its repulsive wall. This demonstrates that the chemical reaction Li + CaH → LiH + Ca, which is exothermic by ∼0.67 eV, is forbidden when both the reactants are spin polarized. We note that the reaction has been observed to occur rapidly at cryogenic temperatures with unpolarized reactants [26].

A contour plot of our *ab initio* PES for Li-CaH is shown in Fig. 1(a). The global minimum is located at $R = 5.6a_0$, $\theta = 35^\circ$ and is 7063 cm^{-1} deep. Two saddle points occur in the linear Li-CaH (Li-HCa) configurations at $R = 10.7a_0$ ($7.0a_0$) with the corresponding well depths of -158.5 and -4384 cm^{-1}, respectively. This demonstrates that the Li-CaH interaction is not only strong, but also strikingly anisotropic. For Mg-CaH, in addition to the global minimum of 2110 cm^{-1} at $R = 6.08a_0$, $\theta = 42^\circ$, we find a local minimum at the linear Mg-CaH arrangement at $R = 7.0a_0$ with the well depth of 1855 cm^{-1}. As shown in Fig. 1, both the Li-CaH and Mg-CaH PESs have similar topology at long range, but differ at small R, with the Li-CaH interaction being significantly more anisotropic.

In order to assess the prospects for sympathetic cooling of CaH molecules with Li and Mg atoms, we solve the quantum collision problem specified by the Hamiltonian (1) numerically using a close-coupling approach in the body-fixed coordinate frame [22] and the *ab initio* PESs for Li-CaH and Mg-CaH calculated with the CaH fragment kept frozen at equilibrium ($r_e = 3.803a_0$). Due to the large anisotropy of the Li-CaH and Mg-CaH interactions, a large number of rotational channels must be included in the basis set to obtain converged results. As shown in the inset of Fig. 2(a), the cross sections oscillate dramatically with increasing size of the rotational basis set until convergence is reached at $N_{\max} = 55$. Five total angular momentum states ($J = 0.5$ to 4.5) were included in the basis to produce the cross sections converged to better than 10%. The maximum number of channels was 3250 for the total angular momentum projection $M = 1/2$. We note that the same problem reformulated in the space-fixed representation

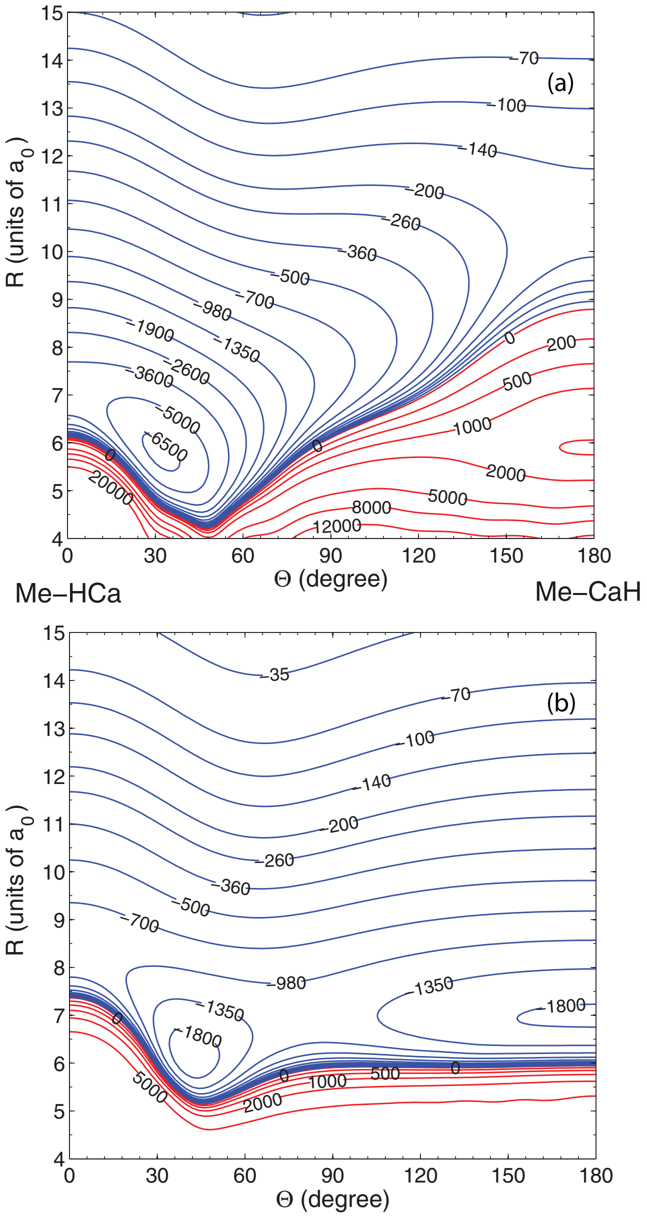

FIG. 1. (Color online) Contour plots of *ab initio* PESs for Li-CaH ($S = 1$) (a) and Mg-CaH ($S = 1/2$) (b). Energies are in units of cm^{-1}. Collinear arrangements of atoms for $\theta = 0^\circ$ and 180° are indicated between panels, where Me = Li, Mg.

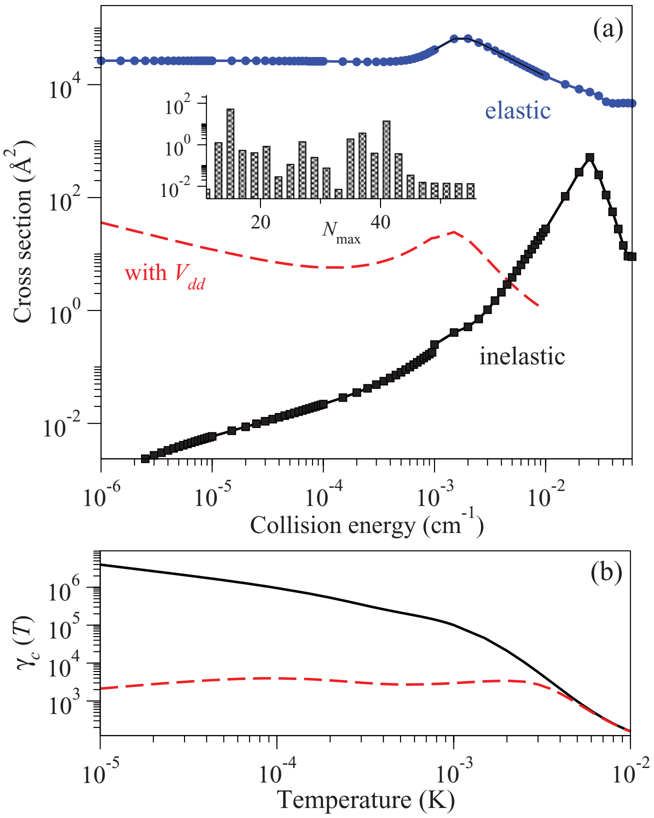

FIG. 2. (Color online) (a) Cross sections for spin relaxation in Li-CaH collisions calculated as functions of collision energy for $B = 0.1$ T. Full lines and symbols indicate calculations without the magnetic dipole interaction and the dashed line indicates calculations including the magnetic dipole interaction. (b) Thermally averaged ratio of elastic to inelastic collision rates for Li-CaH as a function of temperature. The inset shows the convergence of the inelastic cross section at $\epsilon = 0.01$ cm^{-1} with the maximum number of rotational channels included in the basis set ($N_{\max}$).

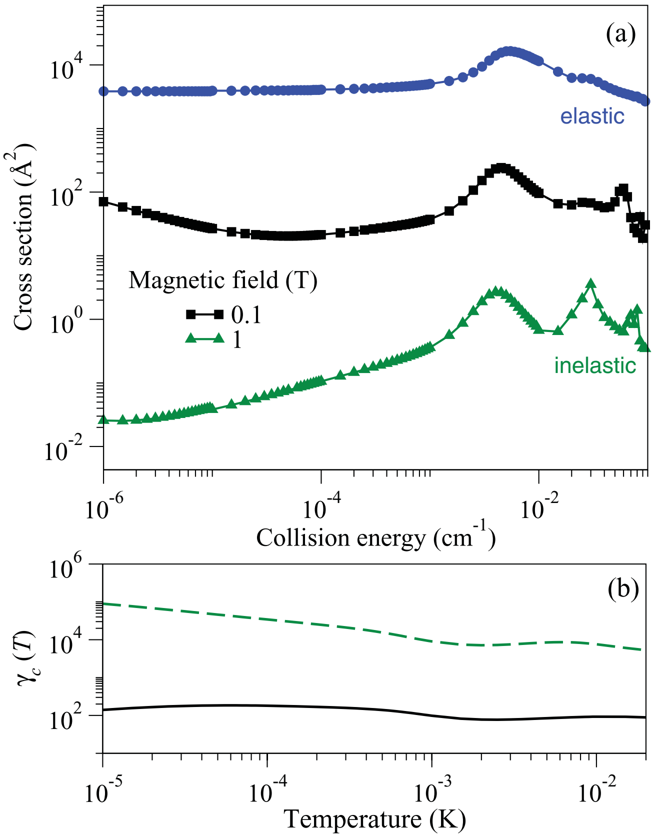

FIG. 3. (Color online) (a) Cross sections for spin relaxation in Mg-CaH collisions calculated as functions of collision energy for $B = 0.1$ T (squares) and 1 T (triangles). (b) Thermally averaged ratio of elastic to inelastic collision rates for Mg-CaH vs temperature for $B = 0.1$ T (full line) and 1 T (dashed line).

[23] with $N_{\max} = 55$, $l_{\max} = 55$ would amount to 234 136 channels, making it computationally intractable.

Figure 2(a) shows the cross sections for spin relaxation and elastic scattering in Li-CaH collisions as functions of collision energy. The inelastic cross sections remain small over the entire range of collision energies considered except in the vicinity of a shape resonance at $\epsilon \approx 0.025$ cm^{-1} (1 cm^{-1} ≈ 1.439 K). The magnetic dipole interaction has a dramatic enhancing effect on inelastic cross sections at collision energies below 0.01 cm^{-1}. The cross sections for spin relaxation in Mg-CaH collisions shown in Fig. 3(a) are comparable to those calculated for Li-CaH. The ratios of elastic-to-inelastic collision rates for Mg-CaH displayed in Fig. 3(b) are large at all temperatures, suggesting favorable prospects for sympathetic cooling of CaH molecules with Mg atoms in an optical dipole trap.

Given the extremely large anisotropy of the Li-CaH and Mg-CaH interactions, it is remarkable that the inelastic cross sections shown in Figs. 2 and 3 are so small in absolute magnitude. In order to gain insight into the mechanism of spin relaxation in strongly anisotropic collision systems, we employ the powerful formalism of multichannel quantum-defect theory (MQDT) [27]. We begin by partitioning the full K matrix into closed- and open-channel blocks [27] using a minimal basis set consisting of two open ($|N = 0, M_{S_{\rm A}} = \pm 1/2\rangle$) and two closed ($|N = 1, M_{S_{\rm A}} = \pm 1/2\rangle$) channels. In the absence of the magnetic dipole interaction, there is no direct coupling between the open channels [14], and we find $K^{\rm oo} = 0$ and $K^{\rm oc} = K^{\rm co} = {\rm diag}(K_1, K_1)$, where K_1 quantifies the strength of the anisotropic coupling between the $N = 0$ and $N = 1$ rotational states. The closed-closed block of the K-matrix is given by

$$K^{\rm cc} = \begin{pmatrix} K_{bb} & K_\gamma \\ K_\gamma & K_{bb} \end{pmatrix},$$

where K_{bb} is the closed-channel level shift [27] and K_γ is the weak spin-rotation coupling between the closed channels. The purpose of our MQDT analysis is to obtain scaling laws for the inelastic cross sections and examine their variation with the spin-rotation and anisotropy parameters K_γ and K_1. A full numerical calculation of these parameters [28] will be presented in future work [29].

Having defined our model 4×4 K matrix, we obtain the reduced K matrix using $K^{\rm red} = K^{\rm oo} + K^{\rm oc}[\tan \nu_b + K^{\rm cc}]^{-1} K^{\rm co}$ [27]:

$$K^{\rm red} = \frac{-1}{D} \begin{pmatrix} K_1^2(\tan \nu_b + K_{bb}) & -K_\gamma K_1^2 \\ -K_\gamma K_1^2 & K_1^2(\tan \nu_b + K_{bb}) \end{pmatrix}, \qquad (2)$$

where $D = (\tan \nu_b + K_{bb})^2 - K_\gamma^2$. Using this result, we obtain for the spin relaxation probability

$$|S_{1/2\to -1/2}|^2 = \frac{K_\gamma^2}{(\tan \nu_b + K_{bb})^2 + \frac{1}{K_1^4}\left(D - K_1^4\right)^2}. \tag{3}$$

This expression immediately yields the scaling law $\sigma_{1/2\to -1/2} \sim \gamma^2$ [14] and illustrates several important features of the mechanism of collisional spin relaxation in $^2\Sigma$ molecules. First, the inelastic probability passes through a maximum at $K_1 = [(\tan \nu_b + K_{bb})^2 - K_\gamma^2]^{1/4}$ and tends to zero as K_1^{-4} when $K_1 \to \infty$. The maximum possible value of $|S_{1/2\to -1/2}|^2$ is given by $K_\gamma^2/(\tan \nu_b + K_{bb})^2$. This implies that spin relaxation can be slow even when anisotropic interactions are extremely strong ($K_1 \to \infty$). The physical meaning of this result is as follows: In a strongly anisotropic collision system, the interaction potential mixes the incident $N = 0$ collision channel with many closed $N > 0$ channels. However, spin relaxation cannot occur without the spin-rotation interaction flipping the electron spin within each $N > 0$ manifold. Because the spin-rotation interaction is weak, this process is inefficient, acting as a "dynamical bottleneck" which is qualitatively similar to the role of the transition state in abstraction chemical reactions [30]. In the opposite limit of weakly anisotropic interaction ($K_1 \to 0$), Eq. (3) yields $|S_{1/2\to -1/2}|^2 \sim K_1^4$.

In summary, we have presented a theoretical analysis of low-temperature collisions of CaH($^2\Sigma$) molecules with Li and Mg atoms based on accurate *ab initio* interaction potentials and a rigorous quantum mechanical approach [22]. We have found that the interactions between Li and CaH are extremely strong and anisotropic, but the chemical exchange $\mathrm{Li} + \mathrm{CaH} \to \mathrm{LiH} + \mathrm{Ca}$ is not allowed when both collision partners are fully spin polarized. Our calculations show that inelastic spin relaxation in collisions of fully spin-polarized CaH molecules with Li and Mg atoms occurs slowly, with elastic-to-inelastic ratios in excess of 100 over a wide range of temperatures from 10 μK to 10 mK.

Our results demonstrate that ultracold spin-polarized mixtures of $^2\Sigma$ molecules with alkali-metal atoms are stable against collisional spin relaxation, strongly suggesting that sympathetic cooling of CaH($^2\Sigma$) molecules with Li atoms will be successful. Furthermore, as shown in Fig. 3, spin relaxation in Mg-CaH collisions occurs at a comparably slow rate, suggesting good prospects for sympathetic cooling of CaH molecules with ground-state Mg atoms in an optical dipole trap. The sympathetic cooling experiments with spin-polarized Li-CaH mixtures should be particularly straightforward, as Li atoms can be held in a magnetic trap for hundreds of seconds [31] and cotrapped with CaH molecules loaded from an ablation source [7] or a buffer-gas cooled molecular beam [32]. Sympathetic cooling with Mg atoms would require an optical dipole trap to confine the nonmagnetic atoms. While cold Mg atoms have not yet been produced in copious quantities, confined in an optical dipole trap, or evaporatively cooled, laser cooling of up to 10^8 Mg atoms to sub-Doppler temperatures (0.5 mK) has been achieved experimentally [33].

We note that, in $^3\Sigma$ and $^2\Pi$ molecules, collisional spin relaxation occurs via direct couplings between different Zeeman states induced by intramolecular interactions and the anisotropy of the interaction potential [17–21,23,34]. This mechanism is more efficient than the indirect mechanism of spin relaxation in $^2\Sigma$ molecules considered here. We therefore expect that $^2\Sigma$ molecules (such as CaH [7], YbF [10], and SrF [35]) will generally be more stable against collisional spin relaxation than $^3\Sigma$ and $^2\Pi$ molecules.

Our analysis indicates that collisions of $^2\Sigma$ molecules with each other are likely to be predominantly elastic, and hence evaporative cooling of $^2\Sigma$ molecules in a magnetic trap may be feasible. Finally, our work opens up the possibility of sympathetic cooling of polyatomic spin-1/2 radicals, which, according to recent *ab initio* and quantum scattering calculations [36], have favorable collisional properties for buffer-gas cooling and magnetic trapping at milli-Kelvin temperatures.

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