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Nonlinear Pressure Shifts of ^{133}Cs Hyperfine Frequencies FEI GONG, YUAN-YU JAU, WILLIAM HAPPER, Princeton University — The hyperfine (microwave) magnetic-resonance frequencies of optically pumped alkali-metal atoms in buffer-gas have long been used in compact, portable frequency standards. The buffer gas is needed to slow down the diffusion of optically pumped atoms to the cell walls, and to eliminate Doppler broadening of the microwave resonances. Van der Waals molecules, consisting of an alkali-metal atom loosely bound to a buffer gas atom, can form in such vapor cells. The molecules strongly affect the spin relaxation of alkali metal atoms in Ar, Kr and Xe gases at pressures of a few Torr. The hyperfine-shift interaction, $\delta A I \cdot S$, the modification a nearby buffer-gas atom makes to the Fermi contact interaction between S and the nuclear spin I of the alkali atom, can contribute to the width of the microwave resonance line, and it is responsible for the pressure shifts of the hyperfine resonance frequencies that are so important for clocks. Our experiments show that Van der Waals molecules also modify the effects of the hyperfine-shift interaction $\delta A I \cdot S$. For Ar pressures of a few tens of Torr or less, the shift of the microwave resonance frequency of ^{133}Cs in Ar buffer gas is not linear in the buffer gas pressure. This occurs because the contribution to the pressure shift from molecules is suppressed when $\tau \delta A I > h$.

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