Comment on "Investigation of Ultrafast Nuclear Spin Polarization Induced by Short Laser Pulses"

In a recent Letter [1], Nakajima proposed a pulsed optical method for transiently inducing high levels of nuclear spin polarization in neutral atoms. This polarization is expected to be periodically resurgent at rates much faster than intrinsic hyperfine precession frequencies. If these predictions were correct, successful realizations of the proposal might prove useful in the study of short-lived radioactive isotopes. Unfortunately, the analysis upon which these claims are based is flawed; it fails to properly identify the eigenstates of an atom with nuclear spin angular momentum $\mathbf{I}$ and total electronic angular momentum $\mathbf{J}$ in a uniform static electric field [2,3]. As a result, the primary conclusions drawn from the study—namely the exceptional rapidity and simultaneous efficiency of the polarization process—are not valid.

In brief, the proposed polarization scheme amounts to the preparation of an atom in a coherent superposition of excited states that evolves in time under the influence of an externally applied static electric field $\mathbf{E}$. In the zero-field limit, where the Stark splitting of these states is much smaller than the corresponding hyperfine splitting, the total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$ of each eigenstate is a constant of the motion. Thus, even though the optical radiation field does not couple directly to the nuclear spin, the magnetic hyperfine interaction ensures that large amplitude periodic variations in nuclear spin polarization P can occur if an appropriate superposition of eigenstates is populated [3,4]. In effect, the Hamiltonian for this interaction $\mathcal{H}_{\mathrm{hf}} = A\mathbf{I}\cdot\mathbf{J}$ enables a coherent internal exchange of angular momenta. Variations in the expectation value of the nuclear spin angular momentum $\langle\mathbf{I}\rangle$ are directly compensated by corresponding variations in $\langle\mathbf{J}\rangle$ so as to conserve orientation. The time scale for this exchange is set by the energy interval(s) $\Delta E = \mathcal{O}(A/\hbar^2)$ between interfering hyperfine states. Starting from an initial value of zero, P reaches an extremum on a time scale of order $\tau = \pi\hbar/\Delta E$. An appropriately timed interruption of $\mathcal{H}_{\mathrm{hf}}$ (e.g., by photoionization of the atom, eliminating the electrons responsible for $\mathbf{J}$) could then be used to halt further evolution of $\langle\mathbf{I}\rangle$. To this extent, the proposed experiment is a variation on methods that have long been used for quantum beat spectroscopy [4–8].

Nakajima argues that the application of a large electric field is an essential element of the proposed polarization scheme; its intended role is to increase the Stark splitting—and hence the quantum beat frequency—to the point where the time scale τ for the transfer of angular momentum to the nucleus becomes substantially shorter than the spontaneous emission lifetime of the coherently excited states. To accomplish this, he advocates the use of Stark splittings that are 2 to 3 orders of magnitude larger than the hyperfine splitting under consideration.

Herein lies a fundamental problem that is not addressed by Nakajima's analysis. The conditions for the proposed polarization scheme are such that the atom is unambiguously in the strong-field limit, wherein the coupling between $\mathbf{I}$ and $\mathbf{J}$ is broken and $\mathbf{F}$ is no longer a constant of the motion. The coherence induced by the laser is therefore almost entirely electronic in character, and to large extent, angular momentum is not exchanged with the nucleus—either on the time scale of the Stark splitting *or* on that of the hyperfine splitting. Viewed from a semiclassical perspective, the applied field $\mathbf{E}$ causes $\mathbf{J}$ to precess so rapidly that $\mathbf{I}$ cannot track its motion. More formally, for integer electronic angular momentum quantum numbers J (cf. the examples in [1]) the matrix representation of $\mathcal{H}_{\mathrm{hf}}$ is reduced to its diagonal elements $Am_I m_J$ in the basis of states labeled by m_I and m_J, the quantum numbers for the projections of $\mathbf{I}$ and $\mathbf{J}$ along $\mathbf{E}$. Hence, $d\langle\mathbf{I}\rangle/dt = -i\langle[\mathbf{I}, \mathcal{H}_{\mathrm{hf}}]\rangle/\hbar = 0$. For half-integral values of J, angular momentum transfer is still possible via the nonvanishing off-diagonal matrix elements $\langle m_I \pm 1, m_J = \mp\frac{1}{2}|\mathcal{H}_{\mathrm{hf}}|m_I, m_J = \pm\frac{1}{2}\rangle$, although this exchange takes place on the time scale of the hyperfine splitting, not the Stark splitting. Either way, if $P = 0$ when the excitation is applied, it will necessarily remain nearly so thereafter on the time scale of the Stark splitting.

Ultimately, Nakajima's analysis fails because it incorrectly views the admixture of electronic and nuclear spin states that comprise the induced atomic coherence as being independent of $|\mathbf{E}|$. A proper treatment of this problem requires identification of the relevant eigenstates through diagonalization of the full Hamiltonian.

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Received 21 May 2008; published 30 October 2008
DOI: 10.1103/PhysRevLett.101.189501
PACS numbers: 29.27.Hj, 29.25.−t, 42.50.Md

[1] Takashi Nakajima, Phys. Rev. Lett. **99**, 024801 (2007).
[2] J. R. P. Angel and P. G. H. Sandars, Proc. R. Soc. A **305**, 125 (1968).
[3] U. Fano and J. H. Macek, Rev. Mod. Phys. **45**, 553 (1973).
[4] S. Haroche, J. A. Paisner, and A. L. Schawlow, Phys. Rev. Lett. **30**, 948 (1973).
[5] E. B. Aleksandrov, Opt. Spectrosc. **17**, 522 (1964).
[6] J. N. Dodd, R. D. Kaul, and D. M. Warrington, Proc. Phys. Soc. London **84**, 176 (1964).
[7] S. Haroche, in *High-Resolution Laser Spectroscopy*, edited by K. Shimoda, Topics in Applied Physics Vol. 13 (Springer-Verlag, Berlin, 1976).
[8] T. Aoto *et al.*, J. Electon Spectros. Relat. Phenomena **144–147**, 47 (2005).

