Pairing of Fermions with Arbitrary Spin

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Motivated by the recent success of optical trapping of alkali Bose condensate, we have studied the superfluid state of optically trapped alkali fermions, which can have Cooper pairs with total spin $J \geq 2$. In this paper, we shall discuss the general structure of these large spin Cooper pairs and their close relation with singlet Cooper pairs with nonzero orbital angular momentum. We also present the exact solution for the $J = 2$ pairing which shows a surprising change of ground state as the spin f of the constituent fermion increases. [S0031-9007(98)08168-X]

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The discovery of Bose-Einstein condensation [1] in atomic gases has stimulated many new research directions. Among these is the search of the superfluid phases of alkali fermions. This search has become even more exciting in view of the recent success of confining Bose condensates in optical traps [2]. Since optical traps are nonmagnetic, the spin of the trapped atoms is no longer frozen as it was in magnetic traps. This leads to a new class of superfluid phenomena. In the case of spin-1 Bose gas like ^{23}Na and ^{87}Rb, one of us [3] has recently pointed out ^{23}Na and ^{87}Rb should have a nonmagnetic and ferromagnetic *spinor* condensate, respectively, according to the current estimates of their scattering lengths [4]. Very recently, experiments at MIT [5] have verified the basically nonmagnetic spinor nature of ^{23}Na and found that its magnetic interaction is indeed antiferromagnetic [3].

The physics of alkali fermions in optical traps is equally rich. The fact that all alkali fermions (except ^{6}Li) have hyperfine spins (or simply "spins") $f > 1/2$ in their lowest hyperfine manifold implies that their Cooper pairs can have total spins $J > 1$. Fermions like ^{22}Na and ^{134}Cs which have $f = 5/2$ and $7/2$ can have Cooper pairs with total spin as high as 4 and 6. From the example of superfluid ^{3}He, one can be sure that the internal structure of these large spin Cooper pairs will generate a multitude of macroscopic quantum phenomena. The purpose of this paper is to point out the structure of these large spin Cooper pairs, and a surprising change in behavior of a spin-J Cooper pair as a function of fermion spin f.

As a first step, we shall focus on *homogenous* dilute Fermi gases in zero magnetic fields. It is important to understand the homogeneous situation before studying the trapped cases [6]. Moreover, the physics of homogeneous systems are important in their own right. At first sight, the weak field limit seems difficult to achieve, for even the Earth's magnetic field amounts to 10^{-4} K, enough to polarize the whole gas. Despite this "strong" background field, which can be shielded off to a large extent, one can reduce it effectively to the weak field limit by specifying the total spin S of the system. Since the dynamics of these systems is spin conserving [3], a prepared spin S out of equilibrium with an external field cannot relax to its equilibrium value. The system therefore sees an effective field which would have been in equilibrium with the prepared S. By choosing S appropriately, the effective field can be made much smaller than the external one. This method has very recently been used by Ketterle's group to study the spinor nature of the ^{23}Na condensate [5].

Of course, for a pairing state to be observable, its pairing interaction has to be sufficiently negative to produce an observable T_c. While the scattering lengths of some alkali fermions have been calculated, they remain unknown for many alkalis. (See later discussion.) In view of the lack of information, we have performed a general study of the large spin Cooper pairs. In particular, we shall discuss the $J = 2$ pairing in detail. This is the simplest among all large spin pairing which also has an exact solution. The phenomena contained in this case reveal the rich physics of large spin Fermi systems, which turns out to be remarkable indeed. For simplicity, we shall call the S-wave spin-J Cooper pairs (made up of two spin f fermions) "spin" Cooper pairs, and singlet Cooper pairs with *orbital* angular momentum J (made up of two spin-$1/2$ fermions) "orbital" Cooper pairs. Let us first summarize our findings:

(A) The structure of spin Cooper pairs is analogous to that of orbital Cooper pairs with the same angular momentum. This allows one to obtain information of the former from the latter, for which an exact solution already exists for $J = 2$ [7].

(B) The structure of a spin-J Cooper changes as the spin f of the constituent fermions increases beyond a critical value. For Cooper pairs with spin $J = 2$, they are "ferromagnetic" (or "axial") if $f \geq 7/2$, but nonmagnetic (or "real") if $f \leq 5/2$. This change of character as a function of f is a result of maximizing the phase space for pairing and is *independent* of interaction parameters, as long as they favor $J = 2$ pairing.

Free energy.—The low energy effective Hamiltonian of a spin-f dilute Fermi gas with s-wave interactions has been derived in Ref. [3]. It is rotationally invariant in spin

space, and is of the form $H - \mu N = \int d\mathbf{x}\, \psi_\alpha^+(\mathbf{x}) \times \mathcal{H}^o_{\alpha\beta}(\mathbf{x})\psi_\beta(\mathbf{x}) + \frac{1}{2}\int d\mathbf{x}\, \psi_\alpha^+(\mathbf{x})\psi_\beta^+(\mathbf{x})\Gamma_{\alpha\beta;\mu\nu}\psi_\mu(\mathbf{x})\psi_\nu(\mathbf{x})$, $\mathcal{H}^o_{\alpha\beta}(\mathbf{x}) = -\frac{\hbar^2}{2M}\nabla^2\delta_{\alpha\beta} - \gamma\mathbf{B}\cdot\mathbf{F}_{\alpha\beta}$,

$$\Gamma_{\alpha\beta;\mu\nu} = \sum_{F=0}^{2f-1} g_F \times \sum_{m=-F}^{F} \langle ff\alpha\beta|ff;Fm\rangle\langle ff;Fm|ff\mu\nu\rangle , \tag{1}$$

where M is the mass of the fermion, $\langle ff;Fm|ff\mu\nu\rangle$ is the Clebsch-Gordan coefficient for forming a total spin F from two spin-f particles, $g_F = 4\pi\hbar^2 a_F/M$, and a_F is the s-wave scattering length of two spin-f fermions in the scattering channel with total spin F. Because of antisymmetry of the fermions, only even F's appear in Eq. (1).

The order parameter of an S-wave superfluid is $\Psi_{\alpha\beta}(\mathbf{x}) = \langle\psi_\alpha(\mathbf{x})\psi_\beta(\mathbf{x})\rangle$, which is a $(2f+1)\times(2f+1)$ antisymmetric matrix in spin space. For homogeneous systems, $\Psi_{\alpha\beta}$ is independent of $\mathbf{x}$. It is convenient to define the gap function

$$\Delta_{\alpha\beta} = \Gamma_{\alpha\beta;\mu\nu}\Psi_{\mu\nu} . \tag{2}$$

Applying the standard BCS theory [8], we obtain the free energy as

$$\mathcal{F} = \frac{1}{2}\mathrm{Tr}\,\Delta^+\Gamma^{-1}\Delta - \frac{k_BT}{2}\sum_{\mathbf{k}\omega_n}\sum_{\ell=1}^{\infty}\frac{1}{\ell}\mathrm{Tr}[\Delta\tilde{G}(\mathbf{k}\omega_n)\Delta^+G(\mathbf{k}\omega_n)]^\ell , \tag{3}$$

where Γ^{-1} is given by Eq. (1) with g_F replaced by g_F^{-1}, $\omega_n = (2n+1)\pi k_BT$ are the Matsubara frequencies, $G_{\alpha\beta}(\mathbf{k},\omega_n)$ and $\tilde{G}_{\alpha\beta}(\mathbf{k},\omega_n)$ are normal Greens functions satisfying matrix equation $[i\omega_n - \mathcal{H}_o(\mathbf{k})]G(\mathbf{k}\omega_n) = 1$, $[i\omega_n + \mathcal{H}_o^T(\mathbf{k})]\tilde{G}(\mathbf{k}\omega_n) = 1$.

General structure of Cooper pairs with spin angular momentum J.—To obtain the general form of Δ, we consider its transformation properties. Under a spin rotation $U = \exp(-i\boldsymbol{\theta}\cdot\mathbf{F})$, $\psi_\alpha \to (U\psi)_\alpha$. This implies $\Psi \to \Psi' = U\Delta U^T$, and hence $\Delta \to \Delta' = U\Delta U^T$. For gap functions that transform like an angular momentum state $|Jm\rangle$, they must satisfy

$$[U\Delta_m^{(J)}U^T]_{\alpha\beta} = D^{(J)}_{mm'}(\boldsymbol{\theta})[\Delta^{(J)}_{m'}]_{\alpha\beta} . \tag{4}$$

It is easy to see that the solution of Eq. (4) is $(\Delta_m^{(J)})_{\alpha\beta} \propto \langle ff\alpha\beta|ff;Jm\rangle$. The general structure of the spin-J gap function is therefore

$$(\Delta^{(J)})_{\alpha\beta} \propto \sum_{m=-J}^{J} c_m\langle ff\alpha\beta|ff;Jm\rangle ,$$

$$\text{or}\quad |\Delta^{(J)}\rangle \propto \sum_{m=-J}^{J} c_m|Jm\rangle , \tag{5}$$

where the second expression in Eq. (5) is simply the first written in abstract form.

To find the $\Delta^{(J)}$ that minimizes the energy, and to illustrate the relation of spin and orbital Cooper pairs, it is useful to consider a different representation of $\Delta^{(J)}$. First, we note that the singlet state $(\Delta^{(0)})_{\alpha\beta} \propto \eta \equiv \langle ff;00|ff\alpha\beta\rangle\sqrt{2f+1}$ satisfies $U\eta U^T = \eta$, and has the properties $\eta^+\eta = 1$, $U^+\eta = \eta U^T$, and $F_i\eta = -\eta F_i^T$. Defining $\Delta \equiv \Xi\eta$, Eq. (4) then becomes $U\Xi_m^{(J)}U^+ = D^{(J)}_{mm'}\Xi^{(J)}_{m'}$, which has the solution $[\Xi_m^{(J)}]_{\alpha\beta} \propto [Y_{Jm}(\mathbf{F})]_{\alpha\beta}$, where $Y_{Jm}(\mathbf{F})$ is a matrix obtained by first writing the spherical harmonic $k^JY_m^{(J)}(\hat{\mathbf{k}})$ in a symmetric rectangular form, and then by replacing k_i by the matrix F_i [9]. For example, since $k^2Y_{21}(\hat{\mathbf{k}}) \propto k_z(k_x + ik_y)$, we have $Y_{21}(\hat{\mathbf{F}}) \propto F_z(F_x + iF_y) + (F_x + iF_y)F_z$. The general form of the order parameter within the angular momentum J subspace is then

$$\Delta^{(J)}_{\alpha\beta} = \sum_{m=-J}^{J} c_m[Y_{Jm}(\mathbf{F})\eta]_{\alpha\beta} . \tag{6}$$

Using the Wigner-Eckart theorem, it is easily seen that the two representations, Eqs. (5) and (6), are identical.

Next, we note that $r^JY_{Jm}(\hat{\mathbf{r}})$ is a homogenous polynomial of $\mathbf{r}$ satisfying Laplace's equation. It can therefore be written as $r^JY_{Jm}(\hat{\mathbf{r}}) = A_{i_1i_2\cdots i_J}r_{i_1}r_{i_2}\cdots r_{i_J}$, where $A_{i_1i_2\cdots i_J}$ is *symmetric in all its indices and vanishes whenever any two indices contract*. We can then write $\Delta^{(J)}$ as

$$\Delta^{(J)} = \sum_{i_1\cdots i_J} A_{i_1i_2\cdots i_J}F_{i_1}F_{i_2}\cdots F_{i_J}\eta . \tag{7}$$

It is also useful to compare the *spin* structure of $\Delta^{(J)}$ in Eq. (7) with the *orbital* structure of the singlet Cooper pairs of spin-1/2 fermions. The order parameter of the latter is $\Delta(\mathbf{k}) = \langle c_\uparrow(\mathbf{k})c_\downarrow(-\mathbf{k})\rangle$, where $c_\uparrow^+(\mathbf{k})$ creates a spin $+1/2$ fermion with momentum $\mathbf{k}$ at the Fermi surface. For pairing with *even* orbital angular momentum J, $\Delta^{(J)}(\mathbf{k}) = \sum_m c_mY_{Jm}(\hat{\mathbf{k}})$, or

$$\Delta^{(J)}(\mathbf{k}) = \sum_{i_1\cdots i_J} A_{i_1i_2\cdots i_J}k_{i_1}k_{i_2}\cdots k_{i_J} . \tag{8}$$

Comparing Eqs. (7) and (8), one finds that they are almost identical except that the F_i's are noncommuting matrices, whereas the k_is are c numbers. On the other hand, this means that these two structures approach each other as f increases, since the spin operator $\mathbf{F}$ behaves more like a classical vector.

The general scheme for determining Δ^J and the $J = 2$ pairing: As temperature is lowered, superfluid condensation first takes place at the (even) J channel with most negative coupling g_J since it has the highest transition temperature (T_c). The free energy Eq. (3) to the quartic order in $\Delta^{(J)}$ is

$$\mathcal{F} = -\frac{1}{2}\alpha\,\mathrm{Tr}\,\Delta^{(J)+}\Delta^{(J)} + \frac{1}{4}\beta\,\mathrm{Tr}(\Delta^{(J)}\Delta^{(J)+})^2 , \tag{9}$$

where $\alpha = N(0)\ln(T_c/T)$, $T_c = 1.14\epsilon_Fe^{-1/(|g_J|N(0))} = 1.14\epsilon_Fe^{-\pi/(2k_F|a_J|)}$, $N(0)$ is the density of state at the

Fermi surface per spin, $\beta = 7\zeta(3)/(8\pi^2 T_c^2)$, and ϵ_F and k_F are the Fermi energy and momentum. To determine $\Delta^{(J)}$, we substitute Eq. (7) into Eq. (9) and find the matrix A that minimizes the energy. In the following, we shall present the exact solution for S-wave $J = 2$ Cooper pairs formed by spin-f fermions. The solutions of $J > 2$ Cooper pairs will be studied elsewhere, for they require much lengthier calculations than the $J = 2$ case, which is already lengthy. Our method, however, applies to all $J \geq 2$ pairs.

From Eq. (9), one can see that $\mathcal{F}$ is of the form

$$\mathcal{F} = -\frac{\alpha}{2} A_{ij} A^*_{pq} \operatorname{Tr}(F_i F_j F_p F_q) + \frac{\beta}{4} A_{ij} A^*_{k\ell} A_{pq} A^*_{st} \operatorname{Tr}(F_i F_j F_k F_\ell F_p F_q F_s F_t). \quad (10)$$

After evaluating the traces (see Appendix), we find

$$\mathcal{F} = -\tilde{\alpha} \operatorname{Tr} A A^+ + \beta_1 |\operatorname{Tr} A^2|^2 + \beta_2 (\operatorname{Tr} A^* A)^2 + \beta_3 \operatorname{Tr}(A^2 A^{*2}), \quad (11)$$

$$\beta_1 = \frac{\beta}{4}\left[-\frac{29}{70} I_2 + \frac{121}{60} I_4 - \frac{22}{15} I_6 + \frac{4}{35} I_8\right], \quad (12)$$

$$\beta_2 = \frac{\beta}{4}\left[-\frac{2}{70} I_2 + \frac{1}{30} I_4 + \frac{4}{15} I_6 + \frac{8}{35} I_8\right], \quad (13)$$

$$\beta_3 = \frac{\beta}{4}\left[\frac{3}{5} I_2 - \frac{8}{3} I_4 + \frac{16}{15} I_6\right], \quad (14)$$

where $I_n \equiv \sum_{m=-f}^{f} m^n$, and $\tilde{\alpha} = \frac{\alpha}{12}[4I_4 - I_2]$.

Equation (11) is identical to the free energy of a general *d-wave* singlet superfluid. The minimization problem of Eq. (11) was solved by Mermin [7]. Only three equilibrium phases are possible [10]: (I) Axial state: when $\beta_3 > -\beta_1 + |\beta_1|$, $\Delta \propto Y_{22}(\mathbf{F})\eta$; (II) Cyclic state: when $0 > \beta_3 > -6\beta_1$, $\Delta \propto (F_x^2 + e^{2\pi i/3} F_y^2 + e^{4\pi i/3} F_z^2)\eta$; (III) Real state: when $\beta_3 < -4\beta_1 - 2|\beta_1|$, $\Delta \propto \{\zeta_1 Y_{20}(\mathbf{F}) + \zeta_2[Y_{22}(\mathbf{F}) + Y_{2,-2}(\mathbf{F})]\}\eta$, where ζ_1 and ζ_2 are real.

The portion of the phase diagram in β_1–β_3 space relevant for our discussion is shown in Fig. 1. Using Eqs. (12) to (14), we note that (β_1, β_3) is in region III for $f = \frac{3}{2}$ and $\frac{5}{2}$, and in region I for $f \geq \frac{7}{2}$. The superfluid is therefore a real state for $f = \frac{3}{2}$ and $\frac{5}{2}$, but changes to the axial state for $f \geq \frac{7}{2}$.

This change of pairing behavior can be understood as follows. As mentioned before, as $f \to \infty$, the order parameters in Eqs. (7) and (8) become identical, and the energy Eq. (9) becomes the weak coupling d-wave superfluid, which has an optimum order parameter $Y_{22}(\hat{\mathbf{k}})$ [7]. (This state has "more pairing" than Y_{20} and $Y_{2\pm1}$ in the sense that its absolute square only has point nodes whereas both $|Y_{20}(\hat{\mathbf{k}})|^2$ and $|Y_{2,\pm1}(\hat{\mathbf{k}})|^2$ have line nodes.) On the other hand, in the most quantum case $f = \frac{3}{2}$, there are four degenerate Fermi surfaces, labeled by

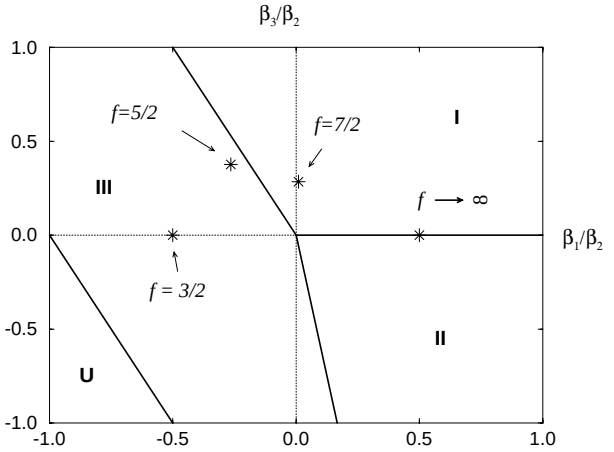

FIG. 1. Distribution of stable phases in the $(\beta_1/\beta_2, \beta_3/\beta_2)$ space for $J = 2$ pairing [7]. Regions I, II, and III are the stable regions of the "axial," "cyclic," and "real" states, respectively. **U** corresponds to an unstable region. The stars are the ratios $(\beta_1/\beta_2, \beta_3/\beta_2)$ calculated from Eqs. (12) to (14) for the J value indicated.

$m_z = \pm\frac{3}{2}, \pm\frac{1}{2}$. The structure of the axial and the real state are given by $|\Delta_{\text{axial}}\rangle = |2;2\rangle = \frac{1}{\sqrt{2}}(|\frac{3}{2},\frac{1}{2}\rangle - |\frac{3}{2},\frac{1}{2}\rangle)$, $|\Delta_{\text{real}}\rangle = \zeta_1|2;0\rangle + \zeta_2(|2;2\rangle + |2,-2\rangle) = \frac{\zeta_1}{2}[(|\frac{3}{2},\frac{-3}{2}\rangle - |\frac{-3}{2},\frac{3}{2}\rangle) + (|\frac{1}{2},\frac{-1}{2}\rangle - |\frac{-1}{2},\frac{1}{2}\rangle)] + \frac{\zeta_2}{2}[(|\frac{3}{2},\frac{1}{2}\rangle - |\frac{1}{2},\frac{3}{2}\rangle) + (|\frac{-1}{2},\frac{-3}{2}\rangle - |\frac{-3}{2},\frac{-1}{2}\rangle)]$, where the state vectors with integer entries such as $|2;0\rangle$ means $|\frac{3}{2}\frac{3}{2}; J = 2, m = 0\rangle$, those with half integer entries such as $|\frac{1}{2},\frac{-1}{2}\rangle$ mean $|f = \frac{3}{2}, m = \frac{1}{2}\rangle\,|f = \frac{3}{2}; m = \frac{-1}{2}\rangle$. One can see that the only two Fermi surfaces ($m = \frac{3}{2}$ and $\frac{1}{2}$) are involved in the pairing in axial state, whereas all four Fermi surfaces are involved in the pairing of the real state. Since the real state maximizes the amount of pairing, it is favored in this extreme quantum case. As f increases, the number of Fermi surfaces appearing in the axial state (i.e., the spin state $|J = 2, m = 2\rangle$) quickly increases. By the time f reaches $\frac{7}{2}$, the real state no longer has the advantage of involving most Fermi surfaces, and the system switches to the axial state, where the spin operator **F** begins to resemble a classical vector. We have thus established statements (A) and (B).

Observability.—The long lived alkali fermions which have $f > \frac{1}{2}$ in their lowest hyperfine manifold are ^{22}Na, ^{40}K, ^{86}Rb, ^{132}Cs, ^{134}Cs, and ^{136}Cs, which have $f =$ 5/2, 9/2, 5/2, 3/2, 7/2, and 9/2 and lifetimes 2.5 yr, 10^9 yr, 18 days, 6 days, 2 yr, and 13 days, respectively [11]. According to the recent calculation of Greene, Burke, and Bohn [4], the scattering lengths of ^{40}K are positive, hence unfavorable for pairing. At present, there is no information about the scattering lengths of the Cs fermions. On the other hand, $a_4 = -65(+40, -20)a_B$, $a_2 = -130(+40, -70)a_B$, $a_0 = -145(+40, -65)a_B$ for ^{86}Rb; and $a_4 = -108(+27, -40)a_B$, $a_2 = -115(+32, -50)a_B$, $a_0 = -117(+34, -55)a_B$ for ^{22}Na, where a_B is the Bohr radius and the numbers in the bracket are error bars.

To estimate T_c, we use the value of k_F and ϵ_F *at the center of the trap*. For an anisotropic trap with frequencies $\omega_\perp$ and ω_z in the xy plane and along z, it is easy to show that $k_F a_2 = (\frac{R}{a_\perp})(\frac{a_2}{a_\perp})$, $\epsilon_F = \frac{1}{2}\hbar\omega_\perp(\frac{R}{a_\perp})^2$, where $a_\perp = \sqrt{\hbar/M\omega_\perp}$, where R is the radius of the cloud in the xy plane related to the total number of particles N as $\frac{R}{a_\perp} = (\frac{48N\lambda}{(2f+1)})^{1/6}$, with $\lambda \equiv \omega_z/\omega_\perp$. For an isotropic trap ($\lambda = 1$) with $\omega_\perp/2\pi = 2000$ Hz, the expression $T_c(J = 2) = 1.14\epsilon_F e^{-\pi/2k_F|a_2|}$ gives $T_c(J = 2) \sim 1.9 \times 10^{-8}K$ for ^{22}Na with $N = 4 \times 10^6$ atoms and $T_c(J = 2) \sim 2.3 \times 10^{-7}K$ for ^{86}Rb with $N = 10^6$ atoms. Since the lowest temperature reached in current BEC experiments is 10^{-9} K, these transition temperatures of fermions (which can be made higher by increasing the trap frequency or the anisotropy λ) appear to be feasible. Since g_0 is most negative, singlet instead of $J = 2$ pairing will first occur in zero field. This, however, does not mean that all higher spin pairing states are nonobservable. The singlet spin states can be efficiently suppressed in a magnetic field (obtained by specifying the total spin of the system as mentioned in the introduction), thereby revealing all other higher spin pairing states [12]. For length reasons, magnetic field effects will be discussed elsewhere.

We have shown that the superfluid phenomena of alkali fermions become amazingly rich once the spin degrees of freedom are released. Should the current efforts of cooling alkali fermions to degenerate limit be successful, transferring the degenerate gas into an optical trap [2] will help one to uncover the superfluid phases discussed here. Since ^{132}Cs, ^{134}Cs, and ^{136}Cs have $f = 3/2$, $7/2$, and $9/2$ in their lowest hyperfine multiplet, respectively, if their scattering lengths turned out to be negative, our result predicts that like ^{22}Na and ^{86}Rb, the ground state of ^{132}Cs will be a real state, whereas ^{134}Cs, and ^{136}Cs will be an "axial" state.

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Appendix.—Evaluation of the quartic term in Eq. (10): Denoting $\mathcal{I}_4 = A_{ij}A^*_{k\ell}A_{pq}A^*_{st}\,\mathrm{Tr}(F_iF_jF_kF_\ell F_pF_qF_sF_t)$, we note that $\mathcal{I}_4 = \mathcal{D}[\mathrm{Tr}\,U]_0$, where $\mathcal{D} \equiv A_{ij}A^*_{k\ell}A_{pq} \times A^*_{st}\frac{\partial^2}{\partial a_i\partial a_j}\frac{\partial^2}{\partial b_k\partial b_\ell}\frac{\partial^2}{\partial c_p\partial c_q}\frac{\partial^2}{\partial d_s\partial d_t}$, $U \equiv e^{-i\mathbf{a}\cdot\mathbf{F}}e^{-i\mathbf{b}\cdot\mathbf{F}}e^{-i\mathbf{c}\cdot\mathbf{F}} \times e^{-i\mathbf{d}\cdot\mathbf{F}} \equiv e^{-i\boldsymbol{\theta}\cdot\mathbf{F}}$, and the subscript "0" means $\mathbf{a} = \mathbf{b} = \mathbf{c} = \mathbf{d} = 0$. Expanding U in powers of $\boldsymbol{\theta}$, it is easy to see that $\mathcal{I}_4 = \sum_{n=1}^4 \frac{(-1)^n}{(2n)!}I_{2n}(\mathcal{D}\,\theta^{2n})_0$, where $I_{2n} = \sum_{m=-f}^{f} m^{2n}$. Next, we note that the relation between θ and $\mathbf{a}, \ldots, \mathbf{d}$ is independent of f. For spin $1/2$ systems, the quantity $Q \equiv \mathrm{Tr}[e^{-i\mathbf{a}\cdot\boldsymbol{\sigma}/2}e^{-i\mathbf{b}\cdot\boldsymbol{\sigma}/2}e^{-i\mathbf{c}\cdot\boldsymbol{\sigma}/2} \times e^{-i\mathbf{d}\cdot\boldsymbol{\sigma}/2}]/\mathrm{Tr}(1) = \langle e^{-i\boldsymbol{\theta}\cdot\boldsymbol{\sigma}/2}\rangle$ can be written as $\xi = Q - 1 = \sum_{n=1,2,\ldots}\frac{(-1)^n}{2^{2n}(2n)!}\theta^{2n}$. Inverting this relation, we obtained θ^2 as a power series of ξ, or Q. From this expression, we can calculate $(\mathcal{D}\,\theta^{2n})_0$ for $n = 2$ to 4 by calculating $\mathcal{D}\,Q^p$ for $p = 1, 2, 3, 4$. The latter can be easily calculated because they involve only spin $1/2$ quantities. Evaluating $\mathcal{I}_4$ this way gives Eq. (12) to Eq. (14).

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