Radio-frequency spectroscopy and Clock shifts of optical transitions

In atomic gases

YU Zhenhua

IASTU, Tsinghua University

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RF & BEC-BCS crossover

In a two component fermion gas



RF spectroscopy

J. Kinnunen, M. Rodriguez, and P. Torma, Science 305, 1131 (2004)



Noninteracting, flipping hyperfine spin pays Zeeman energies; With pairing, breaking pairing pays extra energy.



However, interactions affect atomic levels



Hartree-Fock mean field ~ n, ...

Detection of gap by breaking pairs via rf excitation



Chin et al., Science 305, 1128 (2004)

Initially $n_1 = n_2$, $n_3 = 0$, Pairing between |1> and |2> Excite |2> to |3> by rf.

Naively expect that to excite $|2\rangle$ to $|3\rangle$ by rf, breaking pair, requires twice gap energy, 2Δ

$$E_p = \sqrt{\xi_p^2 + \Delta^2}$$



First attempt of theoretical calculation

The rf field couples to the electronic dipole moment:

$$H_{rf} = \langle \sigma' | \vec{E} \cdot \vec{d}_{e} | \sigma \rangle \int d^{3} \vec{r} \psi_{\sigma'}^{+}(\vec{r}) \psi_{\sigma}(\vec{r})$$

Since $\lambda_{rf} \sim 1m \gg 100 \,\mu m$, take $k_{rf} \approx 0$

Fermi golden rule \implies RF spectroscopy

$$\chi''(\omega) \sim \sum_{f,i} \rho_i |\langle f | H_{rf} | i \rangle^2 |\delta(\omega - E_f + E_i)$$

Time ordered correlation function

J. Kinnunen, M. Rodriguez, and P. Torma, Science **305**, 1131 (2004)



= hyperfine (spin) vertex

$$\chi(t-t') \sim \int d^{3}\vec{r} \int d^{3}\vec{r} \, \langle T \psi^{+}{}_{\sigma}(\vec{r},t) \psi_{\sigma'}(\vec{r},t) \psi^{+}{}_{\sigma'}(\vec{r}\,',t\,') \psi_{\sigma}(\vec{r}\,',t\,') \rangle$$

inducing particle-hole excitations
$$\approx \int d^{3}\vec{r} \int d^{3}\vec{r}\,' \langle T \psi^{+}{}_{\sigma}(\vec{r},t) \psi_{\sigma}(\vec{r}\,',t\,') \rangle \langle \psi_{\sigma'}(\vec{r},t) \psi^{+}{}_{\sigma'}(\vec{r}\,',t\,') \rangle$$

Not good enough

Hartree-Fock approximation: $\Sigma_{\sigma} = \sum_{\sigma' \neq \sigma} g_{\sigma,\sigma'} n_{\sigma'}$



$$\chi \sim G_1 G_2 \sim \frac{n_1 - n_2}{\omega + g_{12}(n_2 - n_1)}$$

Predicts frequency shift:

$$\Delta \omega = g_{12}(n_1 - n_2)$$

$$g_{\sigma\sigma'} = 4\pi \hbar^2 a_{\sigma\sigma'}/m$$

$$But, \qquad A = 0$$

M. W. Zwierlein, Z. Hadzibabic, S. Gupta, and W. Ketterle, PRL 91, 250404 (2003)

Triple fermions



 $\Delta \omega = 4 \pi \hbar^2 n_1 (a_{13} - a_{12}) / m$

A quick save:



Constraint by symmetry

G. Baym and L.P. Kadanoff, PR 124, 287 (1961) G. Baym, PR 127, 1391 (1962)

Continuity equation:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \vec{j} = 0$$

Equation of motion:

$$i \frac{\partial \rho}{\partial t} = [\rho, H]$$

$$H = T + U$$
, $T \sim \psi^+ \psi$, $U \sim \psi^+ \psi^+ \psi \psi$

 $[\rho, T] + [\rho, U] - i \nabla \cdot \vec{j} = 0$ Ward identity

 $\langle [\rho, T] \rangle \sim \langle \psi^+ \psi \rangle, \langle [\rho, U] \rangle \sim \langle \psi^+ \psi^+ \psi \psi \rangle, \langle j \rangle \sim \langle \psi^+ \psi \rangle$

SU(2) matters for RF spectrum!

Calculation of RF spectrum

ZY & G.Baym, Phys. Rev. A 73, 063601 (2006)

RF probe couples to atomic magnetic moment (essentially valence e):

$$H_{probe} = \gamma_e \vec{B}_{rf} \cdot \vec{S}_e$$

Thus measure long-wavelength transverse spin-spin correlation function:

$$\chi''(\omega) \sim \langle \vec{S}_e \vec{S}_e \rangle(\omega)$$

Calculate $\chi''(\omega)$ in manifold of three lowest hyperfine states (|1>, |2>, |3>) :

$$H = \sum_{i=1}^{3} \int d^{3}r \left(\epsilon_{i} \psi_{i}^{\dagger}(\mathbf{r}) \psi_{i}(\mathbf{r}) + \frac{\hbar^{2}}{2m} \nabla \psi_{i}^{\dagger}(\mathbf{r}) \cdot \nabla \psi_{i}(\mathbf{r}) \right)$$
$$+ \sum_{i < j} \int d^{3}r d^{3}r' v_{ij}(\mathbf{r} - \mathbf{r}') \psi_{i}^{\dagger}(\mathbf{r}) \psi_{j}^{\dagger}(\mathbf{r}') \psi_{j}(\mathbf{r}') \psi_{i}(\mathbf{r})$$

Construct L=1 pseudospin space:

$$Y = \Psi^+ \sigma_x \Psi, \quad \Psi = (\Psi_{1,} \Psi_{2,} \Psi_3)$$

Then the measured corresponds to $\langle YY \rangle(\omega)$ (B_r along x direction)



Constraints by SU(2) symmetry

Interaction SU(2) invariant: long wavelength response of χ " must be at the Larmor frequency (cf. ³He, Leggett), no mean field or pairing corrections.

$$\chi(t) = -i \langle TY(t)Y(0) \rangle \qquad \chi(\Omega) = \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\Omega - \omega}$$

Equation of operator $i \frac{\partial}{\partial t} Y(t) = [Y, T + H_z + U]$
 $[Y, T] = 0$
 $H_z \sim Y_z, [Y, H_z] \sim Y$

If interaction is SU(2) invariant [Y, U]=0

The equation of motion for Y is closed and energy scales Associated with U drop out. Zeeman energies are the only Energy scales left.

Experimentally, U is not SU(2) invariant.

Self-consistent Hartree-Fock-BCS calculation





$$v_{ij}(r) = \overline{g}_{ij}\delta(\vec{r})$$



Self-consistent Hartree-Fock-BCS calculation

Shift of "pairing" peak from the normal peak:





Mean frequency shift by sum rules

$$\bar{\omega} = \frac{\int_{-\infty}^{\infty} d\omega \omega \chi''(\omega)}{\int_{-\infty}^{\infty} d\omega \chi''(\omega)} \equiv \omega_0 + \Omega_c$$

 $\omega_0 = \epsilon_3 - \epsilon_2$ Ω_c = mean clock shift

For atoms initially in |1> and |2>

Mean frequency shift by sum rules

Weak coupling HF-BCS:

$$\frac{1}{N} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \omega \chi \,'\,'(\omega) = \epsilon_{32} - \frac{1}{n} (g_{12} - g_{13})(n^2 + \Delta^2/g_{12}^2) \quad (n_1 = n_2 = n)$$

correction vanishes if $g_{12} = g_{13}$ (SU(2) symmetric)

microscopic calculation requires summing bubles

$$\Delta \omega_{H} = (g_{13} - g_{12})n$$
$$\Delta \omega_{BCS} = (g_{13} - g_{12})\Delta^{2} / n g_{12}^{2}$$

Unitary regime

в

$$\Omega_{c} = \frac{1}{n_{2}} \int d^{3}r [v_{13}(r) - v_{12}(r)] \langle \psi_{1}^{\dagger}(\mathbf{r})\psi_{2}^{\dagger}(0)\psi_{2}(0)\psi_{1}(\mathbf{r})\rangle$$
$$\Omega_{c} = \frac{1}{n_{2}} (\bar{g}_{13} - \bar{g}_{12}) \langle \psi_{1}^{\dagger}(0)\psi_{2}^{\dagger}(0)\psi_{2}(0)\psi_{1}(0)\rangle$$

Calculate correlation function in terms of coupling dependence of the free energy:

$$\frac{\partial (F/V)}{\partial \bar{g}_{12}} = \frac{1}{V} \langle \partial H/\partial \bar{g}_{12} \rangle = \langle \psi_1^{\dagger}(0)\psi_2^{\dagger}(0)\psi_2(0)\psi_1(0) \rangle$$
In this case: $\bar{a}_{13}/\bar{a}_{12} \simeq 1$

$$\Omega_c = (\frac{\bar{a}_{13}}{\bar{a}_{12}})(\frac{1}{g_{13}} - \frac{1}{g_{12}})\frac{1}{n_2}\frac{\partial F/V}{\partial g_{12}^{-1}}$$
mean clock shift in terms of renormalized quantities

Physical meaning of the mean frequency shift

Interaction with rf field (long wavelength)

 $H_{\text{int}} = \mathcal{B}(t)Y \quad \text{with} \quad Y = i \int d^3r \left(\psi_3^{\dagger}(\mathbf{r})\psi_2(\mathbf{r}) - \psi_2^{\dagger}(\mathbf{r})\psi_3(\mathbf{r}) \right)$ H_int rotates spin states from $|2\rangle \rightarrow |\beta\rangle = \cos\theta |2\rangle + \sin\theta |3\rangle$

with spatial wavefunction unchanged

Rotated many particle state: $|1\beta\rangle = e^{-i\theta Y}|12\rangle$

If on average only one single atom promoted from |2> to |3>:

 $heta=1/\sqrt{N_2}$ (realized by short rf pulse)

Energy difference of two states:

$$\begin{split} \delta E &= \langle 1\beta |H|1\beta \rangle - \langle 12|H|12 \rangle \\ _{,} &= \frac{1}{2N_2} \langle 12|[[Y,H],Y]|12 \rangle = \omega_0 + \Omega_c \end{split}$$

Mean clock shift is the difference in interaction energy of coherently rotated state $|1\beta\rangle$ and initial state $|12\rangle$.

Further development and application:





Spin-orbit coupled gases



The period of any periodic physical process can be used as a Standard for time. Any physical system that undergoes such a Periodic process can be used as a clock.





Atomic clocks

Hydrogen atoms

 $\omega_0 \sim e^2 / a_0 \sim 10^{15} Hz$ optical

Fine-structure splitting

$$\omega_0 \sim \alpha^2 e^2 / a_0 \sim 10^{11} Hz$$

Hyperfine-structure splitting

 $\omega_0 \sim 100 MHz$

microwave

Combination of fundamental physical constants.

The problem is to measure the intrinsic frequency accurately.

Atomic clocks



Clock shifts

$$\omega_{measure} - \omega_0$$



NIST-F1 Cesium (铯) fountain atomic clock of microwave transition, serving as the US time and frequency standard, with an uncertainty of 5.10×10-16 (as of 2005).













New generation: optical atomic clocks



Narrow line width and higher frequency good for high Q=f/df Charge neutral good for high signal-noise ratio

Sr (锶)-87, 1S0->3P0 transition, df~1mHz

Two obstacles to high accuracy:

Translational motions and interatomic interactions



Solutions:

Freeze the translational motion by loading atoms in optical lattices



Suppress interatomic interactions by using identical fermions



Atomic clock of ⁸⁷Sr @ JILA

Ye's group, Science 324,



1.0 +

Coherent Evolution



Confusion?!

Construct S=1/2 pseudospin space:

 $\hat{\boldsymbol{S}} = \frac{1}{2} \int d\vec{r} [\psi_e^+(\vec{r}), \psi_g^+(\vec{r})] \boldsymbol{\sigma} [\psi_e(\vec{r}), \psi_g(\vec{r})]^T$

$$H_0 = \int d\vec{r} \sum_{\sigma=e,g} \psi_{\sigma}^{+}(\vec{r}) \left[\frac{-\nabla^2}{2m} + E_{\sigma} + V_{\sigma}(\vec{r})\right] \psi_{\sigma}(\vec{r})$$

 $\omega_0 = E_e - E_g$

 $H = H_0 + H_1$

external potential independent of $\boldsymbol{\sigma}$

$$H_{1} = \int d\vec{r}_{1} d\vec{r}_{2} U(|\vec{r}_{1} - \vec{r}_{2}|) \psi_{e}^{+}(\vec{r}_{1}) \psi_{g}^{+}(\vec{r}_{2}) \psi_{g}(\vec{r}_{2}) \psi_{e}(\vec{r}_{1})$$
$$H_{1} = \sum_{i < j} U(|\vec{r}_{i} - \vec{r}_{j}|) (1/2 - 2\hat{s}_{i} \cdot \hat{s}_{j}) \qquad \hat{S} = \sum_{i} \hat{s}_{i}$$

Interaction SU(2) invariant $[H_{1}, \hat{S}] = 0$



Equations of pseudospin operators

To calculate $N_g(t)$ or $N_e(t)$, note $S_z=(N_g-N_e)/2$ and $N=N_g+N_e$

Homogeneous external probe field couples to the system as

$$H_{probe} = B_{+}(t)\hat{S}_{-} + B_{-}(t)\hat{S}_{+}$$

The coherent evolution of the pseudospin, governed by $K=H_0+H_1+H_{probe}$, is determined as

$$i\frac{d}{dt}\hat{\boldsymbol{S}} = [\hat{\boldsymbol{S}}, H_0 + H_1 + H_{probe}] = \omega_0[\hat{\boldsymbol{S}}, \hat{\boldsymbol{S}}_z] + [\hat{\boldsymbol{S}}, H_{probe}]$$

S_z independent of U(r) since $[H_1, \hat{S}] = 0$ and $[\hat{S}, H_{probe}]$ only depends on S

For a two-component Fermi gas, there should be no frequency shift for a homogenous probe field.

Inhomogeneity of probe fields





Frequency shift

The symmetry of the Bloch equations

$$\frac{d}{dt}\vec{S}_i = \vec{\Omega}_i \times \vec{S}_i + 2\sum_j g_{ij}\vec{S}_i \times \vec{S}_j$$

For simplicity, assume Ω_{ix} nonzero

Noninteracting (g=0), invariant under the transformation

$$\begin{aligned} \{\Delta, S_{kx}, S_{ky}, S_{kz}\} &\to \{-\Delta, -S_{kx}, S_{ky}, S_{kz}\} \\ S_z(\Delta) = S_z(-\Delta), \delta \, \omega = 0 \end{aligned}$$

Interacting, invariant under the transformation

$$\begin{split} \{\Delta, g, S_{kx}, S_{ky}, S_{kz}\} &\to \{-\Delta, -g, -S_{kx}, S_{ky}, S_{kz}\} \end{split}$$

For small shift:
$$\delta \omega = \frac{2\delta S_z(t, \Delta)}{dS_z^0(t, \Delta)/d\Delta} \end{split}$$



Pseudospins & Bloch equations

The Bloch equations can be converted into integral ones

$$\vec{S}_{i}(t) = G_{i}(t)\vec{S}_{i}(0) + 2\sum_{j}g_{ij}\int_{0}^{t}dt'G_{i}(t-t')\vec{S}_{i}(t') \times \vec{S}_{j}(t')$$





The change of the total pseudospin due to interaction is

$$\delta \vec{S}(t) = \sum_{j} g_{ij} \int_{0}^{t} dt \,' [G_{i}(t-t') - G_{j}(t-t')] [\vec{S}_{i}(t') \times \vec{S}_{j}(t')]$$

Zero if probe field is homogeneous, i.e., $\boldsymbol{\Omega}_{_{i}}$ is independent of i

Small dispersion limit

In experiment, small dispersion in driving frequencies:

$$|\vec{\Omega}| = |\vec{\Omega}_i| \sim 6 Hz \qquad |\delta \vec{\Omega}_{ij}| = |\vec{\Omega}_i - \vec{\Omega}_j| \sim 0.1 Hz$$

Since
$$P_g = N/2 - \sum_i S_{iz}$$

 $\delta P_g(t) = C_1 \Xi_1 + C_3 \Xi_3 + C_3 \Xi_3,$

where

$$\begin{aligned} \Xi_1 &= \sum_{i,j} \frac{4g_{ij} S_i(0) S_j(0)}{\Omega_{xy}^3} (\delta \mathbf{\Omega}_{ij\parallel})^2 \\ \Xi_2 &= \sum_{i,j} \frac{4g_{ij} S_i(0) S_j(0)}{\Omega_{xy}^3} (\delta \mathbf{\Omega}_{ij\perp})^2 \\ \Xi_3 &= \sum_{i,j} \frac{4g_{ij} S_i(0) S_j(0)}{\Omega_{xy}^3} \mathbf{e}_z \cdot (\delta \mathbf{\Omega}_{ij\parallel} \times \delta \mathbf{\Omega}_{ij\perp}) \end{aligned}$$

with $\delta = \Delta/\Omega_{xy}, \tau = \Omega_{xy}t, \ \Omega_{xy} = \overline{\Omega}_i - \Delta \mathbf{e}_z, \ \delta \Omega_{ij\parallel} = (\delta \Omega_{ij} \cdot \Omega_{xy})\Omega_{xy}/\Omega_{xy}^2$, and $\delta \Omega_{ij\perp} = \delta \Omega_{ij} - \delta \Omega_{ij\parallel}$

For fixed pulse duration t
pulse duration t
ncies:

$$C_{1} = \frac{\delta}{6(1+\delta^{2})^{4}} \left\{ 3(2+\delta^{2}) \sin^{2}(\tau\sqrt{1+\delta^{2}}) - 3[\tau^{2} + (3\tau^{2} - 4)\delta^{2} + 2(2+\tau^{2})\delta^{4}] + \cos(\tau\sqrt{1+\delta^{2}}) + 3[4\delta^{4} - 4\delta^{2} - \tau^{2}(1+\delta^{2})] + \tau\sqrt{1+\delta^{2}} [\tau^{2} + (\tau^{2} + 9)\delta^{2} - 6\delta^{4}] + \sin(\tau\sqrt{1+\delta^{2}}) \right\},$$

$$C_{2} = \frac{\delta}{2(1+\delta^{2})^{7/2}} \left\{ 4\delta^{2}\sqrt{1+\delta^{2}} - 4\delta^{2}\sqrt{1+\delta^{2}} + \sin(\tau\sqrt{1+\delta^{2}}) \right\},$$

$$C_{3} = \frac{2\sin^{2}(\tau\sqrt{1+\delta^{2}})}{(1+\delta^{2})^{5/2}} + \sin(\tau\sqrt{1+\delta^{2}}) \right\},$$

Frequency shift

S-wave scattering length a unknown



Ongoing story

Ye's group, Science 331, 1043 - 1046 (2011)



Thank you