Repulsively-bound exciton-biexciton states in high-spin fermions in optical lattices

A. Argüelles and L. Santos
Institut für Theoretische Physik, Leibniz Universität Hannover, Appelstr. 2 D-30167, Hannover, Germany

We show that the interplay between spin-changing collisions and quadratic Zeeman coupling provides a novel mechanism for the formation of repulsively bound composites in high-spin fermions, which we illustrate by considering spin flips in an initially polarized hard-core 1D Mott insulator of spin-3/2 fermions. We show that after the flips the dynamics is characterized by the creation of two types of exciton-biexciton composites. We analyze the conditions for the existence of these bound states, and discuss their intriguing properties. In particular we show that the effective mass and stability of the composites depends non-trivially on spin-changing collisions, on the quadratic Zeeman effect and on the initial exciton localization. Finally, we show that the composites may remain stable against inelastic collisions, opening the possibility of novel quantum composite phases.

PACS numbers:

I. INTRODUCTION

Ultra cold gases in optical lattices offer an extraordinary controllable environment for the analysis of many-body phenomena, as highlighted by the realization of the Mott insulator to superfluid phase transition in Bose gases [1], and more recently by the observation of the Mott insulator to metal transition in two component Fermi systems [2, 3]. In addition to a wealth of possible quantum phases [4, 5], and due to its discrete nature, optical lattices offer as well novel possibilities for composite formation, including local on-site dimer (or n-mer) formation in attractive gases [6] and fermionic composites in Bose-Fermi mixtures [7]. An even more striking effect of the lattice discreteness is given by the recent observation of repulsively bound atomic pairs (doublons) at a given lattice site [8, 9], which are dynamically stable in the absence of dissipation. Note that contrary to standard condensed-matter systems, where e.g. interactions with phonons would lead to a rapid dissipation of the dimers, optical lattices provide to a large extent a dissipation-free environment. Indeed experiments on optical lattices provide an extraordinary scenario for the study of the intriguing physics of metastable bound states and other far-of-equilibrium phenomena [3].

Spinor gases, formed by atoms with various available Zeeman substates, present a very rich physics due to the interplay between internal and external degrees of freedom [10]. Interestingly, interatomic interactions lead to spin-changing processes in which population is transferred between Zeeman sublevels. The corresponding spinor dynamics has attracted a large interest, mostly in the realm of spinor Bose-Einstein condensates [11, 12]. Spinor lattice gases offer fascinating novel physics, most relevantly in what concerns quantum magnetism, including antiferromagnetic order in spin-1/2 fermions [13, 14] and even more intriguing phases for higher spins [15–20].

Repulsively on-site bound pairs in spin-1/2 fermions are formed since the large interaction energy of the doublon cannot be accommodated by the maximal kinetic energy for two atoms in the lowest band (proportional to the hopping energy) [8, 21, 22]. In this paper we show that a completely different mechanism, based on the interplay between spin-changing and quadratic Zeeman effect (QZE), may sustain novel types of composites for repulsive high-spin lattice fermions. We illustrate this physics for the case of hard core 1D spin-3/2 fermions in a Mott insulator initially polarized in \( m = −3/2 \). We show that spin-flips into \( m = 3/2 \) may lead to two types of composites formed by an exciton (\( m = 3/2 \) particle and \( m = −3/2 \) hole) and an antisymmetric biexciton (\( m = ±1/2 \) particles and two \( m = −3/2 \) holes) (Fig. 1).

We show that the dynamics and stability of the exciton-biexciton composites exhibits a non-trivial dependence with spin-changing, QZE and center-of-mass momentum. Finally, we show that inelastic composite-composite interactions may be very inefficient, opening exciting possibilities for many-body composite gases.

The structure of the paper is as follows. In Sec. III we introduce the spin-3/2 system under consideration as well as the possible exciton, and biexciton excitations occurring after an individual spin flip. Sec. III introduces the idea of repulsively bound exciton-biexciton pairs, with a particular emphasis in their band-like dispersion and the conditions for their existence. Sec. IV is devoted to the intriguing dynamics of the repulsively bound pairs, and its dependence on the spatial delocalization of the initial spin-flip excitations. In Sec. V we analyze the case of multiple spin-flip excitations. Finally, in Sec. VI we...
summarize our conclusions.

II. MODEL

A. Hamiltonian

We consider spin-3/2 fermions \((m = \pm 3/2, \pm 1/2)\) in a deep 1D lattice, such that at low filling only the lowest band is relevant. In this regime, the physics is given by the interplay between inter-site hopping (characterized by the constant \(t\)), QZE (given by a constant \(q\), which may be externally controllable), and s-wave collisions. The latter may occur in two different channels with total spin \(F = 0\) and 2, characterized by the coupling constants \(g_F = 4\pi\hbar^2a_F/M\), with \(a_F\) the scattering length and \(M\) the atomic mass. Although typically \(a_0\) and \(c_2\) are similar, they may be varied by microwave dressing \([24]\) or optical Feshbach resonances \([25]\). Below we use \(G = (g_0 + g_2)/2\), and \(g = (g_2 - g_0)/(g_2 + g_0)\). The s-wave interactions preserve magnetization (and hence the linear Zeeman effect is irrelevant), but may induce spin-changing collisions (characterized by \(g\)), which redistribute the populations at different components. For \(G \gg t\) we may consider maximally one fermion per site (hard-core case). For a sufficiently large chemical potential the system enters into the Mott insulator with one fermion per site. The spin physics within the Mott insulator is given by an effective Hamiltonian:

\[
\hat{H} = \sum_{i,m} qm^2 \hat{n}_{m,i} + \sum_{(i,j)} \hat{H}_{i,j},
\]

where \(\langle \cdots \rangle\) denotes nearest neighbors and

\[
\hat{H}_{i,j} = c_2 \sum_{|m|=1/2} \left( \hat{n}_{m,i} \hat{n}_{m',j} - \psi_{m,i}^\dagger \psi_{m',j} \right) + c_{3/2} \sum_{|m|=3/2} \left( \hat{n}_{m,i} \hat{n}_{m',j} - \psi_{m,i}^\dagger \psi_{m',j} \right) + c_{3/2} \left( \psi_{3/2,i}^\dagger \psi_{3/2,j} - \psi_{3/2,i} \psi_{3/2,j} \right),
\]

describes the effects of the interatomic interactions, with

\[
c_{3/2} = -2t^2 \left( \frac{\cos^2 \phi}{9q/2 + \lambda_+} \right),
\]

\[
c_{1/2} = -2t^2 \left( \frac{\cos^2 \phi}{g/2 + \lambda_-} \right),
\]

\[
c_{sc} = t^2 \sin 2\phi \sum_{\beta = \pm} \eta_{\beta / 2} \Gamma_{3/2, m = 1/2} (2q\bar{m}^2 + \lambda_\beta),
\]

\[
c_2 = -t^2 / (g + G),
\]

where \(\eta_{\pm} = \pm 1, \lambda_\pm \equiv G - 5q/2 \pm [4q^2 + g^2G^2]^{1/2}\), and \(\tan \phi = ([4q^2 + g^2G^2]^{1/2} + 2q) / gG\). The constant \(c_{3/2} / (c_{1/2})\) characterizes the super-exchange between neighboring sites with \(\pm 3/2 / \pm 1/2\), whereas \(c_2\) does the same for sites with \(m\) and \(m'\) such that \(|m| \neq |m'|\). Finally, \(c_{sc}\) characterizes the super-exchange leading to spin-changing from \(\pm 1/2\) to \(\pm 3/2\) or viceversa.

B. Single-flip excitations

We consider a system initially polarized into \(m = -3/2\), \(|\psi_{BG}\rangle = \prod_i \psi_{-3/2,i}^\dagger \langle \text{vac}\rangle\), where \(\langle \text{vac}\rangle\) is the vacuum. \(|\psi_{BG}\rangle\) is stable since collisions preserve magnetization. We are interested in excitations of the form \(|m,j\rangle = \psi_{m\neq-3/2,j}^\dagger \psi_{-3/2,j} \psi_{BG}\), created by spin flips. Note that these excitations have actually an exciton-like character, since they are formed by a particle (in \(m \neq -3/2\)) and a hole (in \(m = -3/2\)) (Fig. 1b).

A single spin-flip into \(m = \pm 1/2\) is not accompanied by spin-changing, and hence lead to isolated \(\pm 1/2\) excitons which hop with an effective hopping \(c_2\). Spin-changing collisions lead to a much less trivial dynamics when spins are flipped into \(m = 3/2\). After the spin-flip the 3/2 exciton may hop to the nearest neighbor via \(c_{3/2}\) super-exchange (second line in Eq. (2)). In absence of spin-changing \((g = 0)\) the 3/2 exciton propagates freely with an energy \(E_{3/2}(k) = 2c_{3/2}(1 - \cos k)\), where \(k\) is the center-of-mass momentum of the 3/2 exciton.

Spin-changing modifies this simple picture, since the \(m = 3/2\) spin may interact via \(c_{sc}\), super-exchange with a neighboring \(-3/2\) spin leading to two neighboring spins with \(m = \pm 1/2\), i.e. a biexciton of the form \(|m,j; -m,j + 1\rangle \equiv \psi_{m,j}^\dagger \psi_{-m,j+1} \psi_{-3/2,j+1} \psi_{-3/2,j} \psi_{BG}\) (Fig. 1). After spin-changing, one of the newly created excitons, say \(m = 1/2\), may move apart from the neighboring \(m = -1/2\) one by a \(c_2\) superexchange (Fig. 1f). Further \(c_2\) processes bring the pair even further away from each other. Since an isolated \(\pm 1/2\) spin surrounded by \(-3/2\) neighbors remains stable against spin-changing, one naively expects all 3/2 excitons to eventually dissolve into independent \(\pm 1/2\) ones, which would freely move away with a hopping \(c_2\). As shown below, contrary to the naive expectation, the 3/2 excitons may become very robust.

III. EXCITON-BIEXCITON REPULSIVELY BOUND STATES

A. Band structure

We introduce the Fourier transformed states \([27]\)

\[
|\phi^\pm_j(k)\rangle = \frac{1}{\sqrt{L}} \sum_{l=1}^L e^{ilk(\pm j/2)} \mathbf{1} \pm \frac{1}{2}, \pm 1/2, \pm j\rangle\]

and

\[
|\psi_{3/2,j}(k)\rangle = \frac{1}{\sqrt{L}} \sum_{l=1}^L e^{ilk(3/2,j)}, \]

where \(L\) is the number of sites and \(k\) the center of mass quasi-momentum either of the 3/2 exciton or of the \(\pm 1/2\) biexciton. Note that the biexciton states \(|\phi^\pm_j(k)\rangle\) contain not only biexcitons
of neighboring ±1/2 (i.e. \( j = 1 \), as in Fig. 1), but also broken biexcitons (i.e. \( j > 1 \), as in Fig. 1). This is true even for the bound solutions discussed before, which may contain as well a contribution of broken biexciton pairs.

It is particularly useful to introduce the symmetric and antisymmetric states \( |\phi_{j,\pm}^S(k)\rangle = (|\phi_j^+(k)\rangle \pm |\phi_j^-(k)\rangle)/\sqrt{2} \). Note that the Hamiltonian (2) do not couple symmetric and antisymmetric states, and hence the Hamiltonian in \( k \)-space splits into \( H = \sum_k (H_S(k) + H_A(k)) \). The physics of the symmetric biexcitons is given by

\[
H_S(k) = E_{1S} |\phi_{1}^S(k)\rangle \langle \phi_{1}^S(k)| + E_B \sum_{j>1} |\phi_j^S(k)\rangle \langle \phi_{j+1}^S(k)| - \Omega_2(k) \sum_{j>1} (|\phi_j^S(k)\rangle \langle \phi_{j+1}^S(k)| + H.c.),
\]

with \( E_{1S} = 2c_2 - 4q \), \( E_B = 4c_2 - 4q \) and \( \Omega_2(k) = 2c_2 \cos(k/2) \), whereas the coupling between 3/2 excitons and antisymmetric \( \pm 1/2 \) biexcitons is given by

\[
H_A(k) = E_{1A} |\phi_{1}^A(k)\rangle \langle \phi_{1}^A(k)| + E_B \sum_{j>1} |\phi_j^A(k)\rangle \langle \phi_{j+1}^A(k)| + E_{3/2}(k) |\psi_{3/2}(k)\rangle \langle \psi_{3/2}(k)| + \Omega_{sc}(k) (i |\psi_{3/2}(k)\rangle \langle \phi_{1}^A(k)| + H.c.) - \Omega_2(k) \sum_{j>1} (|\phi_j^A(k)\rangle \langle \phi_{j+1}^A(k)| + H.c.),
\]

where \( E_{1A} = 2c_2 + 2c_{1/2} - 4q \), \( E_{3/2}(k) = 2c_{3/2}(1 - \cos k) \), and \( \Omega_{sc}(k) = \sqrt{8c_{sc}} \sin(k/2) \).

Figure 2 shows a typical spectrum (which resembles that of self-bound repulsive pairs of spin-1/2 fermions [8]). Unbound \( \pm 1/2 \) biexcitons form a continuous band, due to their free relative motion with an effective hopping \( \Omega_2(k) \), which leads to a dispersion \( E(k, k_r) = E_B + 2\Omega_2(k) \cos k_r \), where \( k_r \) is the relative momentum of the pair. We observe also the appearance of up to three bound states (resembling the case of repulsive spin-1/2 fermions with nearest neighbor interactions [27]). One of the bound states (upper state in Fig. 2, with energy \( E(k) = E_B - 2c_2(1 + \cos^2(k/2)) \), belongs to the symmetric manifold, being hence decoupled from the 3/2 exciton, and thus we do not consider it further.

### B. Pairing of excitons and antisymmetric biexcitons

The other two bound states result from the coupling of antisymmetric ±1/2 biexcitons and 3/2 excitons. In order to understand these states, we introduce the ansatz

\[
|\psi(k)\rangle = \cos \varphi \sum_{j \geq 1} (-1)^{j-1} e^{-(j-1)/\gamma} |\phi_j^A(k)\rangle + i \sin \varphi |\psi_{3/2}(k)\rangle,
\]

where \( \gamma \) is the localization length. Imposing \( E|\psi(k)\rangle = H_A(|\psi(k)\rangle) \) results in a transcendental equation for \( \gamma \):

\[
E_B + 2\Omega_2 \cosh(1/\gamma) = \frac{1}{2} \left[ E_{1A} + \Omega_{2} e^{1/\gamma} + E_{3/2} \right] + \frac{1}{2} \sqrt{[E_{1A} + \Omega_{2} e^{1/\gamma} - E_{3/2}]^2 + 4\Omega_{sc}^2},
\]

where the relation between energy and \( \gamma \) is given by

\[
E(k) - E_B = 2\Omega_2(k) \cosh(1/\gamma(k)).
\]

In Fig. 2 we indicate the results obtained from Eqs. (10) and (11), which are in excellent agreement to those directly obtained from the diagonalization of (8). Eq. (11) shows that the separation between the bound state and the lowest boundary \( E_B + 2\Omega_2(k) \) of the sea of unbound ±1/2 pairs is crucial for its localization, and hence while the lower bound state is tightly bound, the upper one is much more loose. In particular, the upper bound state unbinds (\( \gamma \) diverges) if its energy approaches \( E_B + 2\Omega_2 \). Hence the existence of the bound exciton-biexciton states depends non-trivially on QZE, spin-changing collisions, and exciton momentum. Fig. 3 shows those values of \( g \) and \( q \) for which one or two bound states are expected for \( k = 0 \).

### IV. DYNAMICAL PROPERTIES OF THE EXCITON-BIEXCITON BOUND PAIRS

The mixing angle \( \varphi \) fulfills

\[
\tan \varphi(k) = -\frac{1}{\Omega_{sc}(k)} \left( E(k) - E_{1A} - \Omega_2(k) e^{-1/\gamma(k)} \right).
\]

The contribution of the 3/2 exciton to the bound states (\( \sin^2 \varphi \)) is particularly important to understand the dynamics following a spin-flip into \( m = 3/2 \). In particular, at \( k = 0 \) (when the center of mass of the spin-flip excitation is maximally delocalized), \( \Omega_{sc} = 0 \) and hence the
Note that if the created initial exciton has a sharply
upper bound state is a pure antisymmetric ±1/2 biexciton
with $E = E_{1A} + 4c_3^2/E_{1A}$ and $\gamma = 1/\ln(E_{1A}/2c_2)$,
wheras the lower bound state is a pure $m = 3/2$ exciton
with energy $E_{3/2}(0)$. On the contrary, when the center of
mass of the spin-flip excitation is strongly localized there
is a strong coupling between the exciton and the anti-
symmetric biexciton which leads to a strongly distorted
dynamics, as shown below.

If the initial spin-flip is delocalized in a spatial region
much larger than the inter-site spacing the momentum
distribution is strongly peaked at $k = 0$. Hence after
the spin-flip there is no significant transfer into ±1/2
biexcitons, since the lower bound state is basically a 3/2
exciton. Without spin-changing the exciton tunnels with
hopping $c_{3/2}$, and hence at $k = 0$ the effective exciton
mass is $m_{s0} = 1/2c_{3/2}$. In spite of the absence of any
significant biexciton admixture, spin-changing strongly
modifies the effective exciton mass into:

$$\frac{1}{m_s} \simeq 2c_{3/2} - \frac{4c_{3c}^2}{E_{1A}}.$$  \hspace{1cm} (13)

Note that $m_s$ differs significantly from $m_{s0}$ (it may
even invert its sign), radically modifying the exciton
wavepacket dynamics, as illustrated in Fig. 4.

Note that if the created initial exciton has a sharply
defined but finite center-of-mass momentum $k_0$, then the
3/2 exciton is not any more a single bound state but
a linear superposition of both bound states (and possibly
a slight contribution of broken biexcitons). Since the
bound states have different group velocities of opposite
sign, the dynamics after the spin-flip is hence charac-
terized by the appearance of two wavepackets moving
in opposite directions formed by the two different types
of exciton-biexciton composites (Fig. 5, middle, which
should be compared with the single wavepacket spreading
for $k_0 = 0$ in Fig. 4, top).

The situation is markedly different if the initial spin-
flip is strongly localized, where all $k$ in the Brillouin zone
contribute. As a consequence, the initial 3/2 excitation
results in an admixture of both bound states, which re-
sult, as shown in Fig. 5 (bottom), to a completely differ-
en wavepacket dynamics as that for initially extended
spin-flips (Fig. 5 top). Note that, in addition, a localized
initial spin-flip may become considerably more unstable
against unbinding, especially if the upper antisymmetric
3/2 bound state unbinds for a given range of $k$ values. In
that case a significant part of the initial exciton excita-
tion would dissolve into unbound biexciton pairs.

V. MULTIPLE SPIN-FLIP EXCITATIONS

Up to this point we have considered the dynamics af-
after single spin-flips. Although single spin flips may be
created using state of the art techniques for single-site
addressing \cite{28,29}, in typical experiments more than
one spin-flip will be induced. As a result more than one
composite will be simultaneously created, hence opening
the possibility of inelastic composite-composite collisions
which may induce a decay channel for the bound com-
posites into broken biexciton pairs. The various initial
spin-flip excitations result in a complicated many-body
non-equilibrium dynamics for the created bound com-
posites, which we have studied by means of time-dependent
Matrix-Product-State calculations \cite{30} for up to 5 spin-
flips delocalized in an initial extension of 10 sites (for a
total of 30 sites).

Figure 5 shows our results for the case of $qG/t^2 = -4$
and $g = 0.6$. For this particular case the bound states are
well below the sea of broken pairs. Hence, as discussed
above, it is expected that single spin-flip excitations do
not lead to a significant population of unbound solutions.
Two main features can be observed in Fig. 5. On one side,
there are marked Rabi like oscillations of the 3/2-exciton
population due to the presence of the two bound states.
The oscillation amplitude is small since, as discussed in
the previous section, the lowest bound state is to a large
extent the 3/2-exciton due to the initial delocalization of
the spin-flip. Note that the Rabi oscillations are how-
ever damped due to dephasing, since different momentum
components close to $k_0 = 0$ have a slightly different
Rabi frequency. Note that a more localized initial spin-
flip would result in a stronger frequency beating, due to
the participation of momenta over all the Brillouin zone.
Note also that the presence of additional spin-flips leads
to an even stronger damping of the Rabi oscillations.

On the other side, note that composite-composite collisions basically leave unaffected the total population in the bound states, which for this particular case basically reduces to the population of \(3/2\) excitons (Fig. 1k) and unbroken \(\pm 1/2\) biexcitons (Fig. 1b). Hence, deeply

bound states may be created, which do not undergo significant inelastic unbinding due to inelastic collisions. Thus, multiple spin-flips lead to an out-of-equilibrium (but highly metastable) many-body state, which may understood as a basically stable gas of bound composites, opening interesting perspectives for quantum composite gases. For the particular case considered in Fig. 6 we expect a quantum gas of almost pure \(3/2\)-excitons with "biexciton dressed" dynamics. Note that although the \(3/2\)-excitons present an effective attractive nearest neighbor interaction \((-2c_{3/2})\), this interaction becomes irrelevant at low momenta \(k \rightarrow 0\) compared to the infinite on-site repulsion due to Pauli exclusion. Hence the low \(k\) properties of 1D \(3/2\) excitons will be as those of a Tonks-Girardeau gas.

VI. CONCLUSIONS

In summary, spin flips in a polarized repulsive high-spin Fermi gas may lead to the formation of novel types of bound composites, which we illustrated for the case of 1D spin-\(3/2\) hard-core fermions in the Mott phase. In that case the composites are formed by a \(3/2\) exciton-like excitation and an antisymmetric \(\pm 1/2\) biexciton-like one. Intriguing dynamics and stability properties of the composites result from a non trivial interplay between spin-changing, QZE and exciton momentum. The stability of the exciton gas against inelastic interactions opens exciting possibilities for the creation of intricate novel quantum composite phases.

Acknowledgments

We thank the DFG for support (Center of Excellence QUEST).
[1] M. Greiner et al., Nature (London) 415, 39 (2002).
[2] R. Jördens et al., Nature (London) 455, 204 (2008).
[3] U. Schneider et al., Science 322, 1520 (2008).
[4] M. Lewenstein et al., Adv. Phys. 56, 243 (2006).
[5] J. Bloch, J. Dalibard, and W. Zwerger, Rev. Mod. Phys. 80, 885 (2008).
[6] A. Rapp, G. Zarand, C. Honerkamp, and W. Hofstetter, Phys. Rev. Lett. 98, 160405 (2007).
[7] M. Lewenstein et al., Phys. Rev. Lett. 92, 050401 (2004).
[8] K. Winkler et al., Nature (London) 441, 653 (2006).
[9] N. Strohmaier et al., Phys. Rev. Lett. 104, 080401 (2010).
[10] T.-L. Ho, Phys. Rev. Lett. 81, 742 (1998).
[11] H. Schmaljohann et al., Phys. Rev. Lett. 92, 043402 (2004).
[12] L. E. Sadler et al., Nature 443, 312 (2006).
[13] C. Klempt et al., Phys. Rev. Lett. 104, 195303 (2010).
[14] F. Werner et al., Phys. Rev. Lett. 95, 056401 (2005).
[15] C. Honerkamp and W. Hofstetter, Phys. Rev. Lett. 92, 170403 (2004).
[16] C. Wu, Phys. Rev. Lett. 95, 266404 (2005).
[17] P. Lecheminant et al., Phys. Rev. Lett 95, 240402 (2005); S. Capponi et al., Phys. Rev. B 75, 100503(R) (2007); G. Roux et al., Eur. Phys. J. B 68, 293 (2009).
[18] Gorshkov et al, Nature Physics 6, 289, (2010).
[19] M. A. Cazalilla et al, New J. Phys. 11, 103033, (2009).
[20] R. Jördens et al., Phys. Rev. Lett. 104, 180401 (2010).
[21] D. Petrogyan et al., Phys. Rev. A 76, 033606 (2007).
[22] F. Alzetto and X. Leyronas, Phys. Rev. A 81, 043604 (2010).
[23] Y.-M. Wang and J.-Q. Liang, Phys. Rev. A 81, 045601 (2010).
[24] D. J. Papoular, G. V. Shlyapnikov, and J. Dalibard, Phys. Rev. A 81, 041603(R) (2010).
[25] P. O. Fedichev et al., Phys. Rev. Lett. 77, 2913 (1996).
[26] A divergence occurs when \( q = \pm G(g^2 - 1)/4 \), where perturbation theory breaks down, since the Mott insulator energy equals that with a particle-hole excitation. We avoid this divergent case.
[27] J.-P. Nguyen and S. Flach et al., Phys. Rev. A 80, 015601 (2009).
[28] W. S. Bakr et al., Nature 462, 74 (2009).
[29] J. F. Sherson et al., Nature 467, 68 (2010).
[30] F. Verstraete et al., PRL 93, 227205 (2004).