PHYSICAL REVIEW A **66**, 063404 (2002)

Resonant oscillation modes of sympathetically cooled ions in a radio-frequency trap

Taro Hasegawa
Department of Materials Science, Faculty of Science, Himeji Institute of Technology, 3-2-1, Koto, Kamigori, Ako-gun, Hyogo 678-1297, Japan

Tadao Shimizu
Department of Electronics and Computer Science, Science University of Tokyo in Yamaguchi, 1-1-1, Daigakudori, Onoda, Yamaguchi 756-0884, Japan

(Received 1 October 2001; published 12 December 2002)

Sympathetic cooling of Ca^+, Zn^+, Sr^+, Ba^+, and Yb^+ as guest ions with laser-cooled $^{24}\mathrm{Mg}^+$ as host ions in a rf ion trap is carried out, and resonant frequencies of their motion in the trap potential are measured. Various oscillation modes of the sympathetically cooled ions are observed. The resonant frequency of the oscillation mode is different from the frequency of either the collective oscillation frequency of the trapped ions or the oscillation frequency of each ion without host ions. This difference is well explained by a theoretical model in which coupled equations of motion of the host ion cloud with a single guest ion are considered.

DOI: 10.1103/PhysRevA.66.063404 PACS number(s): 32.80.Pj, 52.27.Jt, 52.58.Qv

I. INTRODUCTION

A rf trap is a promising device for application to frequency standards [1,2], high-resolution spectroscopy [3–5], and fundamental experiments such as observation of quantum jumps [6–9], because it provides an ideally isolated ion system. For these applications, cooling of ions, especially laser cooling [10,11], is an important technique. For the purpose of cooling ion species that cannot be laser-cooled directly, sympathetic cooling [12–19] by which guest ions are cooled indirectly through collision with laser-cooled coolant host ions which are simultaneously trapped, is proposed and demonstrated in a Penning trap. Sympathetic cooling of molecular ions in a Penning trap [15] and crystallization of sympathetically cooled atomic ions in a linear rf trap [16] have been observed. Application of the sympathetically cooled ions to quantum logic is also proposed [20]. The sympathetic cooling in a rf trap in which no magnetic field is used is preferred to that in a Penning trap for frequency standards and high-resolution spectroscopy. In contrast to a number of studies in a Penning trap, demonstration of sympathetic cooling in a rf trap is difficult to carry out owing to the difficulty of simultaneous ion trapping of hosts and guests and to the higher ion temperature caused by rf heating [21]. Moreover, the sympathetically cooled ion species in a rf trap have been limited to those whose mass is similar to the host ions (for example, MgH^+ and MgD^+ with laser-cooled $^{24}\mathrm{Mg}^+$ [17], or $^{112}\mathrm{Cd}^+$ with laser-cooled $^{114}\mathrm{Cd}^+$ [19]). However, sympathetic cooling in a rf trap has recently been carried out with an ionization technique using charge exchange [22].

In this paper we study the dynamics of sympathetically cooled ions characterized by a resonant frequency in the time-averaged pseudopotential of the rf trap. The important properties such as the temperature and the density of the trapped ions depend strongly on the resonant frequency of the ion motion, and, furthermore, the resonant frequency of the ion motion is practically used to identify the trapped ion mass in mass spectroscopy [23]. Therefore, it is important to measure the resonant frequency of the trapped ions, and several studies have been carried out as follows. Collective oscillation of trapped ions is observed when the same number of two ion species (Ho^+ and Er^+) with a close mass [164.93 and 167.26 in atomic mass units (amu), respectively] are simultaneously trapped without laser cooling [24], and it has been found that the resonant frequency of the collective oscillation is an average of the resonant frequencies of the two trapped ions. In the case of Xe^+ and Ho^+, however, the mass difference is large (131.30 amu and 164.93 amu, respectively), and two resonant frequencies are observed corresponding to each ion species. Different points of our study compared to Ref. [24] are that the ions are sympathetically cooled and therefore cold, that the number of sympathetically cooled ions is much less than that of the coolant ions, and that the mass difference is much larger (for example, $^{24}\mathrm{Mg}^+$ and $^{138}\mathrm{Ba}^+$). A new theoretical model is necessary to describe such a different experimental condition.

II. EXPERIMENT

The resonant frequency of oscillating motion of sympathetically cooled Ca^+, Zn^+, Sr^+, Ba^+, or Yb^+ (guest ions) by laser-cooled $^{24}\mathrm{Mg}^+$ (host ions) in a rf trap is measured using a technique similar to laser-cooled mass spectroscopy [23]. The procedure of simultaneous trapping of guest and host ions is described in Ref. [22]. The neutral guest atoms introduced into the trap are ionized by charge exchange with the trapped host ions. The guest ion number is much less than the host ion number (typically 5 barium ions for 100 magnesium ions [22]).

The ion trap consists of one ring and two endcap electrodes of hyperboloid of revolution. A rf voltage of amplitude $V_{\mathrm{ac}} = 290$ V is applied between the ring and cap electrodes at a frequency of $\Omega/2\pi = 2.06$ MHz. No dc voltage is applied. The secular frequency along the symmetry axis of revolution $\omega/2\pi$ is approximately given by

$$\omega/2\pi = \frac{\sqrt{2}QV_{\mathrm{ac}}}{\pi\mu\Omega(r_0^2+2z_0^2)}. \tag{1}$$

©2002 The American Physical Society

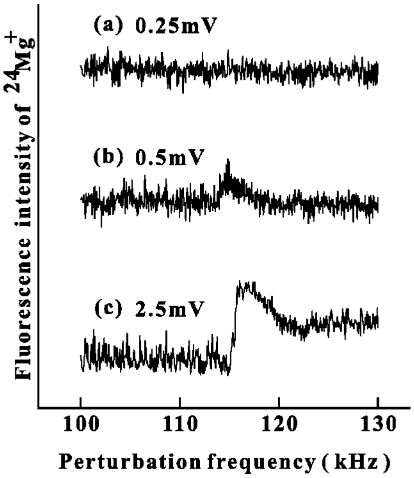

FIG. 1. Fluorescence intensity of laser-cooled $^{24}\mathrm{Mg}^{+}$ trapped with Ba^{+} as a function of perturbation rf frequency. Perturbation amplitude is (a) 0.25 mV, (b) 0.5 mV, or (c) 2.5 mV as described in the figure. Sweep time is 80 s.

Here, Q and μ are the charge and mass of the ion, r_0 the radius of the ring electrode, and $2z_0$ the spacing between two cap electrodes. In our experimental setup, $\omega/2\pi = 352$ kHz for $^{24}\mathrm{Mg}^{+}$.

Additionally, a rf voltage of a few millivolts is applied between the cap electrodes for excitation of the resonant oscillation in the trap potential [23,25]. The resonant frequency is measured by observing the $^{24}\mathrm{Mg}^{+}$ fluorescence intensity as a function of the frequency of the additional perturbation rf voltage. If the perturbation frequency coincides with the resonant frequency of guest ion motion, the guest ions are heated and the $^{24}\mathrm{Mg}^{+}$ ions are also heated through collision with the heated guest ions [15,23]. The amplitude of the perturbation rf voltage should be large enough to obtain a signal, but too large an amplitude ejects ions from the trap. Thus it is necessary to choose a suitable amplitude in the experiment.

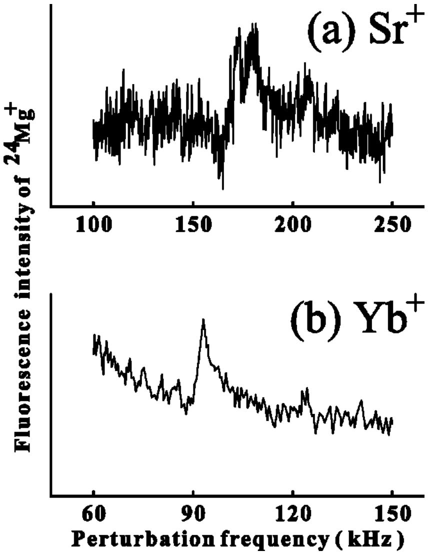

FIG. 2. Fluorescence intensity of laser-cooled $^{24}\mathrm{Mg}^{+}$ trapped with (a) Sr^{+} and (b) Yb^{+} as a function of perturbation rf frequency. The secular frequency for Sr^{+} without $^{24}\mathrm{Mg}^{+}$ is calculated as 96 kHz, and for Yb^{+} 49 kHz.

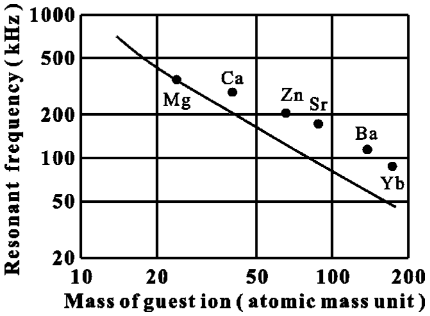

FIG. 3. Dependence of observed resonant frequencies of sympathetically cooled ions on the guest ion mass. A solid line implies the calculated secular frequency of pure ions. Observed ion species are Ca^{+} (40.1 amu), Zn^{+} (65.4 amu), Sr^{+} (87.6 amu), Ba^{+} (137.3 amu), and Yb^{+} (173.0 amu), and the host ion is $^{24}\mathrm{Mg}^{+}$.

III. EXPERIMENTAL RESULTS

Figure 1 shows the fluorescence intensity of $^{24}\mathrm{Mg}^{+}$ induced by the cooling laser at 280 nm as a function of the perturbation rf frequency at several amplitudes V_{rf} when barium ions are simultaneously trapped and sympathetically cooled. In this case the temperatures of Mg^{+} and Ba^{+} are estimated to be 100 K and 500 K, respectively, by observing the Doppler broadened optical spectral line of each ion species [22]. When $V_{\mathrm{rf}} = 0.25$ mV [Fig.1(a)], a change in the fluorescence intensity is not observed. This implies that the perturbation field cannot excite the ion motion enough. On the other hand, when $V_{\mathrm{rf}} = 2.5$ mV [Fig.1(c)], the perturbation field is too large for the ions to be kept in the potential. The irreversible increase of the fluorescence intensity is shown in Fig. 1(c), because barium ions are removed from the trap and the temperature of Mg^{+} decreases [26]. This result implies that $V_{\mathrm{rf}} = 0.5$ mV [Fig. 1(b)] is the most suitable for this observation.

The observed resonant frequency of sympathetically cooled Ba^{+} is 115kHz in Fig. 1. However, the secular frequency of pure barium ions without $^{24}\mathrm{Mg}^{+}$ is calculated to be 61 kHz from Eq. (1). Similarly, the higher resonant frequencies of 170 kHz for Sr^{+} [Fig. 2(a)] and 90 kHz for Yb^{+}

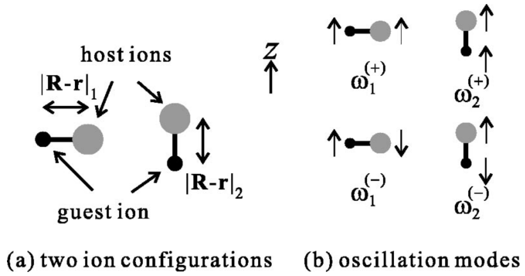

FIG. 4. Schematic representations of (a) ion configurations and (b) oscillation modes. There are two oscillation modes for each configuration.

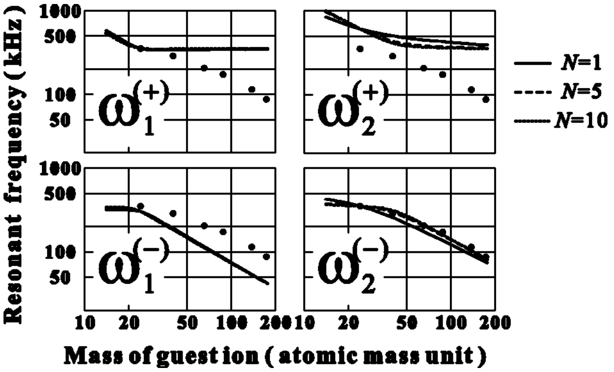

FIG. 5. Calculated resonant frequencies of ion motion when the number of the host ions N is 1 (solid line), 5 (dashed line), or 10 (dotted line), with the experimental results (solid circles).

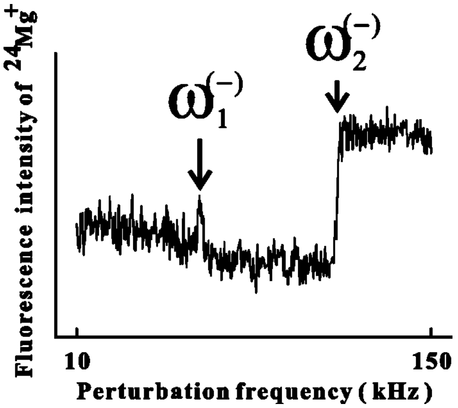

FIG. 6. Observation of the oscillation mode of $\omega_2^{(-)}$ for Ba$^+$ in Mg$^+$. In this trace, V_{rf} is 100 mV.

[Fig. 2(b)] are observed, while the secular frequencies of the pure ions are 96 kHz and 49 kHz, respectively. The observed resonant frequency of motion for each ion species is summarized in Fig. 3 as a function of guest ion mass. It is found that the resonant frequency of sympathetically cooled ions is definitely larger than that without ^{24}Mg$^+$ but much lower than the secular frequency of ^{24}Mg$^+$ (352 kHz). It is considered that a sympathetically cooled ion oscillates rather independently but is perturbed appreciably by the cloud of host ions, and the resonant frequency is shifted as a result. The collective motion with other ions is not expected (in the case of the collective motion, the resonant frequency should be close to the secular frequency of ^{24}Mg$^+$ because few guest ions are trapped in the present experiment and the charge-to-mass ratio of the whole ion cloud is close to that of Mg$^+$). The observed frequency is rather independent of the numbers of host and guest ions (in the case of the longer loading time of the guest ions, which means a smaller number of the host ions and a larger number of the guest ions, the observed resonant frequency is not different from that in Fig. 1).

IV. THEORETICAL MODEL AND DISCUSSIONS

There are two characteristic results in the experiment. First, the resonant frequency of sympathetically cooled ions is shifted from that without host ions. Second, the significant dependence of the resonant frequency on the numbers of both host and guest ions is not observed.

The weak dependence of the resonant frequency on the guest ion number is considered to be caused by the weaker Coulomb interaction between the guest ions themselves and between the guest and the host ions owing to the smaller guest ion number. Thus we consider the theoretical model in which a single guest ion is confined and interacting with a cloud of the host ions. The guest ion is likely located outside the host ion cloud because a heavier guest ion is in a shallower trap potential. The host ions are regarded as a single ion with the mass $N\times m$ and the charge $N\times e$ (N is the host ion number). The effect of micromotion is eliminated by using the time-averaged pseudopotential.

Dynamics of a guest ion (with mass M and charge e) and the host ion cloud ($N\times m$ and $N\times e$) is described by the equations of motion,

$$Nm\frac{d^2\mathbf{r}}{dt^2}+Nm\omega_s^2\begin{pmatrix} x/4 \\ y/4 \\ z \end{pmatrix}+\frac{Ne^2}{4\pi\varepsilon_0}\frac{\mathbf{R}-\mathbf{r}}{|\mathbf{R}-\mathbf{r}|^3}=0, \tag{2}$$

and

$$M\frac{d^2\mathbf{R}}{dt^2}+M\nu^2\omega_s^2\begin{pmatrix} X/4 \\ Y/4 \\ Z \end{pmatrix}-\frac{Ne^2}{4\pi\varepsilon_0}\frac{\mathbf{R}-\mathbf{r}}{|\mathbf{R}-\mathbf{r}|^3}=0. \tag{3}$$

Here, $\mathbf{r}=(x,y,z)$ and $\mathbf{R}=(X,Y,Z)$ are position vectors of the center of mass of the host ion cloud and the guest ion, ε_0 the electric permittivity of vacuum, $\omega_s/2\pi$ the secular frequency of the host ion along the symmetry axis of revolution (z direction), and ν is m/M. The secular frequency of the guest ion is equal to $\nu\omega_s/2\pi$. The perturbation force and the damping force by laser cooling and sympathetic cooling are not included since these do not affect the resonant frequency.

Because the perturbation voltage is applied along the z direction, we consider resonant frequencies of the z components in Eqs. (2) and (3). Since V_{rf} is small, Eqs. (2) and (3) can be approximated as a set of linear equations by replacing $|\mathbf{R}-\mathbf{r}|$ with that in equilibrium. There are two possible equilibriums as shown in Fig. 4(a). In the configuration perpendicular to the z axis, the distance is written as

$$|\mathbf{R}-\mathbf{r}|_1=\left[\frac{4e^2(\nu+N)}{4\pi\varepsilon_0 m\omega_s^2\nu}\right]^{1/3}, \tag{4}$$

and along the z axis,

$$|\mathbf{R}-\mathbf{r}|_2=\left[\frac{e^2(\nu+N)}{4\pi\varepsilon_0 m\omega_s^2\nu}\right]^{1/3}. \tag{5}$$

The resonant frequency of a set of the linearized equations is calculated as

$$\omega_1^{(\pm)}=\omega_s\sqrt{\frac{3\nu+4\nu^3+N(4+3\nu^2)\pm\sqrt{(4N+3\nu+3N\nu^2+4\nu^3)^2-48\nu^2(N+\nu)^2}}{8(N+\nu)}}, \tag{6}$$

for the configuration of Eq. (4) and

$$\omega_2^{(\pm)}=\omega_s\sqrt{\frac{3\nu+\nu^3+N(1+3\nu^2)\pm\sqrt{(N+3\nu+3N\nu^2+\nu^3)^2-12\nu^2(N+\nu)^2}}{2(N+\nu)}}, \tag{7}$$

for Eq. (5). The oscillation mode of each frequency is schematically shown in Fig. 4(b). For $N=1$, 5, and 10, the dependence of the calculated resonant frequency of Eqs. (6) and (7) on the guest ion mass is shown in Fig. 5 along with the experimental results. It is found that the observed resonant motion completely agrees with the $\omega_2^{(-)}$ mode. Furthermore, the predicted weak dependence of the resonant frequency on the host ion number N in Fig. 5 agrees with the experimental result. The curves for N larger than 10 remain almost unchanged.

The frequencies of $\omega_1^{(+)}$ and $\omega_2^{(+)}$ modes are very close to the frequency of pure Mg^+ ion, and it is hard to resolve them from each other. The mode $\omega_1^{(-)}$, however, can be observed together with $\omega_2^{(-)}$ as shown in Fig. 6 when $V_{\mathrm{rf}}=100$ mV, which is a much larger value of V_{rf} than those in Fig. 1. The sudden increase of the fluorescence signal at $\omega_2^{(-)}$ is due to escape of Ba^+ from the trap. The observed frequency $\omega_1^{(-)}$ of $\sim$60 kHz agrees well with the predicted value of 54 kHz (Fig. 5).

The reason why the larger perturbation amplitude is required for detection of $\omega_1^{(-)}$ may be explained as follows. In the experiment we do not observe the ion motion directly, but rather the intensity change of the fluorescence of $^{24}\mathrm{Mg}^+$ induced by the excited ion motion which includes micromotion. The amplitude of micromotion is proportional to the distance between the ion and the trap origin. The mode $\omega_2^{(-)}$ readily changes the distance between the ion and the trap origin, while the mode $\omega_1^{(-)}$ does not change this distance nearly as much as the mode $\omega_2^{(-)}$. The mode $\omega_2^{(-)}$ easily excites the ion motion and increases the temperature of $^{24}\mathrm{Mg}^+$. This causes a change in the fluorescence intensity (signal amplitude). The excitation of the ion motion through the mode $\omega_1^{(-)}$ requires a much stronger perturbation.

V. CONCLUSION

We have observed resonant modes of motion of ions sympathetically cooled in a rf trap. It is found that the resonant frequency of the sympathetically cooled heavier ion species is larger than that of the pure ion without $^{24}\mathrm{Mg}^+$ present and is almost independent of the trapped ion numbers of both host and guest ions. The observed results are well explained by the theoretical model, in which the ion system is treated with the composition of a host ion cloud and a single guest ion, and interaction between them is taken into account. It is found that the ions behave independently rather than collectively in the present condition of a much smaller number of heavy guest ions than the host ions, although the oscillation frequency of the guest ions is affected by the host ion cloud. The present theoretical model explains well the dynamics of the sympathetically cooled ion system. In future, we will investigate the sympathetically cooled ions in the crystal state, which may be preferred for application of the sympathetically cooled ions to frequency standards, high-resolution spectroscopy, and so on, because of the much lower temperature compared to the cloud state.

ACKNOWLEDGMENTS

This work was financially supported by the Ministry of Education, Culture, Sports, Science and Technology of Japan.

[1] G. Werth, IEEE Trans. Instrum. Meas. **IM-34**, 238 (1985).

[2] P.T.H. Fisk, Rep. Prog. Phys. **60**, 761 (1997).

[3] R.E. Drullinger, D.J. Wineland, and J.C. Bergquist, Appl. Phys. **22**, 365 (1980).

[4] R.J. Rafac, B.C. Young, J.A. Beall, W.M. Itano, D.J. Wineland, and J.C. Bergquist, Phys. Rev. Lett. **85**, 2462 (2000).

[5] Th. Becker, J.v. Zanthier, A.Yu. Nevsky, Ch. Schwedes, M.N. Skvortsov, H. Walther, and E. Peik, Phys. Rev. A **63**, 051802(R) (2001).

[6] J.C. Bergquist, R.G. Hulet, W.M. Itano, and D.J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).

[7] A.A. Madej and J.D. Sankey, Phys. Rev. A **41**, 2621 (1990).

[8] A.A. Madej, J.D. Sankey, G.R. Hanes, K.J. Siemsen, and A.R.W. McKellar, Phys. Rev. A **45**, 1742 (1992).

[9] E. Peik, G. Hollemann, and H. Walther, Phys. Rev. A **49**, 402 (1994).

[10] D.J. Wineland and W.M. Itano, Phys. Rev. A **20**, 1521 (1979).

[11] J.I. Cirac, L.J. Garay, R. Blatt, A.S. Parkins, and P. Zoller, Phys. Rev. A **49**, 421 (1994).

[12] D.J. Larson, J.C. Bergquist, J.J. Bollinger, W.M. Itano, and D.J. Wineland, Phys. Rev. Lett. **57**, 70 (1986).

[13] H. Imajo, K. Hayasaka, R. Ohmukai, U. Tanaka, M. Watanabe, and S. Urabe, Phys. Rev. A **53**, 122 (1996).

[14] H. Imajo, K. Hayasaka, R. Ohmukai, U. Tanaka, M. Watanabe, and S. Urabe, Phys. Rev. A **55**, 1276 (1997).

[15] M.A. van Eijkelenborg, M.E.M. Storkey, D.M. Segal, and R.C. Thompson, Phys. Rev. A **60**, 3903 (1999).

[16] P. Bowe, L. Horneœr, C. Brodersen, M. Drewsen, J.S. Hangst, and J.P. Schiffer, Phys. Rev. Lett. **82**, 2071 (1999).

[17] K. Mølhave and M. Drewsen, Phys. Rev. A **62**, 011401(R) (2000).

[18] A.S. Newbury, B.M. Jelenkovic, J.J. Bollinger, and D.J. Wine-

land, Phys. Rev. A **62**, 023405 (2000).
[19] B.B. Blinov, L. Deslauriers, P. Lee, M.J. Madsen, R. Miller, and C. Monroe, Phys. Rev. A **65**, 040304 (2002).
[20] D. Kielpinski, B.E. King, C.J. Myatt, C.A. Sackett, Q.A. Turchette, W.M. Itano, C. Monroe, D.J. Wineland, and W.H. Zurek, Phys. Rev. A **61**, 032310 (2000).
[21] F.G. Major and H.G. Dehmelt, Phys. Rev. **170**, 91 (1968).
[22] T. Hasegawa and T. Shimizu, IEEE Trans. Instrum. Meas. **50**, 556 (2001).
[23] T. Baba and I. Waki, Jpn. J. Appl. Phys. **35**, L1134 (1996).
[24] K. Jungmann, J. Hoffnagle, R.G. DeVoe, and R.G. Brewer, Phys. Rev. A **36**, 3451 (1987).
[25] R.F. Wuerker, H. Shelton, and R.V. Langmuir, J. Appl. Phys. **30**, 342 (1959); R. Alheit, X.Z. Chu, M. Hoefer, M. Holzki, G. Werth, and R. Blümel, Phys. Rev. A **56**, 4023 (1997); M.A.N. Razvi, X.Z. Chu, R. Alheit, G. Werth, and R. Blümel, *ibid.* **58**, R34 (1998).
[26] T. Hasegawa and T. Shimizu, Appl. Phys. B: Lasers Opt. **70**, 867 (2000).